HYPERUNIFORMITY OF POINT PATTERNS AND
TWO-PHASE COMPOSITE MEDIA

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Abstract

This dissertation describes the theoretical investigation of hyperuniform many-body systems, including point patterns and two-phase composite media. Hyperuniform systems encompass all perfect crystals, perfect quasicrystals, and some exotic amorphous states of matter. Hyperuniform systems arise in a broad spectrum of contexts in the physical, mathematical, and materials sciences. Using both analytical and computational methods of statistical mechanics, we devise methods to construct disordered hyperuniform systems. We also characterize the structure and physical properties of both ordered and disordered hyperuniform systems.

The first part of this dissertation (Chapters 2-5) concerns hyperuniform point patterns. In Chapter 2, we prove that the use of aspherical windows can affect the detection of hyperuniformity and propose how to resolve such problems. In Chapter 3, we study the degree to which the introduction of various types of imperfections in perfectly hyperuniform systems degrades or destroys their original perfect hyperuniformity. In Chapter 4, we study how random, uncorrelated displacements of particles on a lattice can lead to “cloaking” of the Bragg peaks in the diffraction pattern of the original lattice. We study sufficient conditions for inverting the collective coordinates of particle distributions in Chapter 5.

In the second part of the dissertation (Chapters 6-10), we study hyperuniform two-phase systems. In Chapter 6, we rank order the degree of hyperuniformity of certain two-dimensional models of Class I hyperuniform two-phase media. In Chapter 7, we devise a tessellation-based procedure to construct perfectly hyperuniform disordered packings. Chapters 8-10 concern the prediction of effective physical properties of hyperuniform and nonhyperuniform two-phase composite media. The strong-contrast expansion formalism is extended to derive exact expansions for the effective dynamic properties of a two-phase medium that account for complete microstructural information (infinite set of $n$-point correlation functions) and hence multiple scattering to all
orders. Using these expansions, we derive accurate approximations for the effective
dynamic dielectric constant and elastic moduli that apply beyond the long-wavelength
regime in Chapters 8 and 9, respectively. In Chapter 10, we establish cross-property
relations linking these two effective wave properties of a given composite by utilizing
the results in Chapters 8 and 9.
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3.1 (Color online) Left panel: Configurations of (a) an initially perfectly hyperuniform integer lattice and (b)-(e) imperfect lattices. The different types of imperfections include the following: (b) a single vacancy, (c) a single interstitial defect [denoted by a solid cyan five-pointed star], (d) uncorrelated stochastic particle displacements $u$ via a uniform distribution with the variance $\langle u^2 \rangle = 0.05$, and (e) thermalized excitations, i.e., elastic waves at $\overline{T} = 0.05$, where $\overline{T}$ is a dimensionless temperature (3.54). Right panel: Corresponding deviations in the number of particles $N(x_0; L)$ inside a window centered at $x_0$, i.e., $\delta N(x_0; L) \equiv N(x_0; L) - \langle N(x_0; L) \rangle$ at two different window sizes $L$. The local number variance $\sigma_N^2(L)$, or equivalently the volume average of $\delta N(x_0; L)^2$, measures the degree of density fluctuations at a given length scale $L$. Roughly speaking, a system is nonhyperuniform if $\sigma_N^2(L)$ grows as the window size increases, as in cases (b), (c) and (d). By contrast, the perturbed system (d) is hyperuniform.
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4.2 Structure factors of URL models in \( \mathbb{R}^2 \), where the perturbation strength \( a \) increases from left to right. Samples of point patterns are shown on top, structure factors \( S(\mathbf{k}) \) of single configurations (including the forward scattering) are shown below, represented by the color code, as a function of the two-dimensional wave vector \( \mathbf{k} := (k_x, k_y) \). The Bragg peaks vanish when the perturbations cover the entire space without overlap (\( a = 1.0 \)) but reappear when the perturbations become stronger (\( a = 1.2 \)). Clearly visible are only peaks with \( k_x = 0 \) or \( k_y = 0 \), other peaks have small weights.

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4.8 Three-point correlation function $g_3(0, x_2, x_3)$ of the 1D cloaked URL with $a = 1$. The lattice remains cloaked at the three-point level, in the sense, that there are no features that exhibit the periodicity of the underlying lattice.

4.9 Four-point correlation function $g_4(0, x_2, x_3, x_4)$ of the 1D cloaked URL with $a = 1$ (a) as a function of $x_3$ choosing a specific path in configuration space, where $x_1$ and $x_2$ are constant and $x_4 = x_2 + x_3$. The curves represent four different values of $x_2$ (assuming without loss of generality that $x_1 = 0$). In contrast to the two-point and three-point correlation functions, the periodicity of the original lattice can be identified for $0 < x_2 < 1$. (b) The schematic explains the occurrence of this periodicity. There cannot be two particles within a single unit cell of the lattice. Therefore, the contribution of cases 1, 3, and 5 (counted from the top to bottom, colored red) to $g_4$ is identically zero. Moreover, the probability density of simultaneously finding a pair of points at $(x_1, x_2)$ and another one at $(x_3, x_4)$ depends on the distance $|x_2 - x_3|$ modulo the lattice spacing.
5.1 Schematics of typical arrangements of collective-coordinate constraints in Fourier space for a periodic $d$-dimensional square fundamental cell of side length $L$. Here, upper and lower panels represent cases for $d = 1$ and 2, respectively. Constraints are taken from $\tilde{n}(\mathbf{k})$’s at wavevectors between two concentric circles centered at the origin: there are $2M$ wavevectors (black dots) within the blue circle, except for $2N_k + 1$ wavevectors inside the red-shaded region. In Refs. [74, 321, 317, 352, 353], a spherical region with $N_k = 0$ was considered; see a list of available $M$ values for two-dimensional cases in Table II in Ref. [321]. For our present purposes, the number of constraints is denoted by $M = 2M$ because the real and/or the imaginary parts of collective coordinates are considered independently.

5.2 Illustrations for solutions of the inversion problem for a single-particle target configuration. (a) Cases with $N_k = 0$ and $M = 2$. When $\tilde{n}_T(k_1)$ is given as constraints (left), both its real and imaginary parts are required for a unique solution; see the cross ($\times$) mark in the right panel. Red and blue lines represent the real and the imaginary parts of $\tilde{n}(k_1)$ of a solution, respectively. (b) Cases with $N_k = 1$ and $M = 2$. When $\tilde{n}_T(k_2)$ is given, we have two solutions.
5.3 Graphical solutions of (5.8) for given respective target configurations. In each panel, black solid lines and dashed ones represent solutions of the “real” and the “imaginary” parts of (5.8), respectively. Contour plots depict potential energy landscape [i.e., \( \log_{10}(\Phi(r^2; R^2)) \)] for each target configuration. Solutions (intersections of solid and dashed lines) are unique and identical to the target configuration (red dots), unless it is the integer lattice (i.e., \(|R_1 - R_2| = L/2\)) as shown in (c). Otherwise, there are infinitely many solutions, and one needs additional constraint \( \tilde{n}_T(k_2) \) for unique solutions.

5.4 Numerical results of the inversion procedure for three-particle perturbed lattices in cases with \( N_k = 0 \) and \( M = 3 \). (a) The average number of distinct solutions per target configuration. Two different optimization algorithms (BFGS+MINOP and the steepest descent) and two constraint conditions [the real (5.4) and the imaginary (5.5) ones] are used for comparison with the energy tolerance \( \epsilon_E = 10^{-29} \). For any target configuration, the number of distinct solutions is at most two, but the average can vary with the target configurations. (b) Examples of nontrivial solutions for a given target perturbed lattice with various displacements \( \delta \). Nontrivial solutions by the real (5.4) or the imaginary (5.5) conditions, respectively, are different from each other, and are not translations of the target.
5.5 Log-log plots of histograms for energy distributions of numerically distinct solutions \{r^3\} of a three-particle target configuration \(R^3\) for parameters \(N_k = 0, M = 3,\) and \(\epsilon_E = 10^{-20}\). Given a target configuration, there are at most two distinct solutions; a trivial solution and a nontrivial one. (a-b) Results from two constraint types [i.e., the real condition (5.4) and the imaginary condition (5.5)] are compared for two different types of target configurations: (a) perturbed lattices with \(\delta = 0.1\) and (b) Poisson configurations. Here, BFGS+MINOP (B.M.) algorithms are used. (c) For Poissonian target configurations, we compare results from two different optimization algorithms: B.M., and steepest descent (S.D.). Here, the real condition (5.4) is considered.

5.6 Log-log plots of histograms for energy distribution of numerically distinct solutions \(\{r^N\}\) for odd-number system sizes: \(N = 9\) (a, d), 19 (b, e), and 29 (c, f). Using the real condition (5.4) condition and parameters \(N_k = 0\) and \(\epsilon_E = 10^{-20}\), two types of target configurations are considered: (a-c) perturbed lattices with \(\delta = 0.1\) and (d-f) Poisson configurations. When \(M = N\), while a target perturbed lattice has a single nontrivial solution \(r^N \neq R^N\), whose occurrence rate is similar to that of trivial ones, a Poissonian target mainly has the trivial solution but occasionally can have multiple nontrivial solutions. When \(M = N + 1\) is an even number, while there is a unique solution for perturbed lattices, there can be more than one solution for a Poisson target configuration in relatively lower occurrence rates. Even in the latter case, however, the nontrivial solutions can be eliminated by lowering the tolerance \(\epsilon_E\) around \(10^{-25}\).
5.7 Log-log plots of histograms for energy distribution of numerically distinct solutions \( \{ r^N \} \) for \( N_k > 0 \) and odd-number system sizes: \( N = 9 \) (a, d), 19 (b, e), and 29 (c, f). Considering perturbed lattices with \( \delta = 0.1 \) as the target configurations, we search solution configurations under the real condition (5.4) and the tolerance \( \epsilon_E = 10^{-20} \), and via the BFGS+MINOP algorithms. We note that there is no nontrivial solution with \( \Phi(r^N; R^N) < 10^{-20} \) if \( N_k > 0 \) and \( M = N + 1 \).

5.8 Numerical results for the average number of numerically distinct solutions per a target configuration of particle number \( N \) with various values of \( N_k \). Using the real condition (5.4) and BFGS+MINOP algorithms, we consider two types of target configurations: (a-c) perturbed lattices with \( \delta = 0.1 \) and the tolerance \( \epsilon_E = 10^{-20} \), and (d-f) Poisson configurations with \( \epsilon_E = 10^{-25} \). When \( N_k = 0 \), both types of target configurations require \( M = N \) constraints for an even-number \( N \), and \( M = N + 1 \) is the minimal for an odd-number \( N \): The minimal number of \( M \) is \( 2\lceil N/2 \rceil \). If \( N_k > 0 \), for both types of target configurations, the minimal number of constraints becomes \( M = N + 1 \).

5.9 The minimal number of successive collective-coordinate constraints \( \min M \) as a function of particle number \( N \) for various \( N_k \).

5.10 Schematics of some possible ways to select collective-coordinate constraints in the two-dimensional Fourier space. Collective coordinates are specified at wavevectors inside (a) an annular region of outer radius \( K \) and inner radius \( K_0 \) (see Fig. 5.1), (b) a rectangular region of width \( K_x \) and height \( K_y \), and (c) \( n \) mutually non-parallel strips which lengths are \( K_i, i = 1, \ldots, n \). We note that the red-shaded region is excluded.
6.1 Illustrations of parameters used to compute the form factors of polygonal figures in $\mathbb{R}^3$. (a) A pentagon has a face $\Gamma$ surrounded by five vertices $V_1, \ldots, V_5$. The vertex indices increase counter-clockwise when the normal vector $\hat{n}$ is towards the reader. (b) A polyhedron with five faces, for each of which the ordering of vertices fulfills the right-hand-rule with the normal vector $\hat{n}_j$. Note that all normal vectors point towards the outside of the polyhedron.

6.2 Illustrations of the six different periodic cellular networks considered in this chapter: from top to bottom, the square, rhombic, honeycomb, square-octagon, triangular, and kagomé networks. We show each of them at three solid-phase volume fractions: $\phi = 0.10$, $\phi = 0.50$, and $\phi = 0.95$. Note that these networks can be regarded as periodic point patterns in the limit of $\phi \to 1$.

6.3 Unit cells of two-dimensional periodic networks: (a) square, (b) rhombic, (c) honeycomb, (d) square-octagon, (e) triangular, and (f) kagomé networks. While (a) and (d) have square fundamental cells, the rest of networks have rhombic fundamental cells. The length parameters $L_1$ and $L_2$ (shown in black and blue arrows, respectively) determine the volume fraction of the solid phase (red regions) $\phi = 1 - (L_2/L_1)^2$. These periodic networks can be treated as packings of polygons defined by the white regions: (a)-(c) can be expressed by a single polygon in the fundamental cells, whereas (d)-(f) needs multiple polygons.

6.4 Illustrations of the four different models of the two-dimensional periodic dispersions of nonoverlapping identical disks considered in this chapter with different solid-phase volume fractions: $\phi = 0.50$ and $\phi = 0.95$. From top to bottom, we present dispersions associated with the square and triangular lattices and honeycomb and kagomé crystals.
6.5 Representative images of the three different models of the two-dimensional disordered/irregular packings of identical circular disks considered in this chapter at different volume fractions: (a) stealthy hyperuniform packing of \( \chi = 0.49 \) and \( \phi = 0.63 \), (b) stealthy hyperuniform packing of \( \chi = 0.40 \) and \( \phi = 0.85 \), and (c) perturbed-lattice packing of \( \phi = 0.79 \). 148

6.6 Log-log plots of the local volume-fraction variances \( \sigma^2_V(R) \) of two-dimensional ordered and disordered cellular solids at a selected solid-phase volume fraction \( \phi = 0.85 \): (a) honeycomb network, (b) triangular-lattice disk packing, and (c) stealthy hyperuniform packings of \( \chi = 0.4 \). The first two models are periodic structures, whereas the last is a disordered one. Here we take the inverse of the specific surface \( 1/s \) to be unity, i.e., \( D = 1/s = 1 \). . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 149

6.7 Surface-area coefficient \( B_V(R) \) as a function of window radius \( R \) of two-dimensional ordered and disordered cellular solids at a selected solid-phase volume fraction \( \phi = 0.85 \), as per Fig. 6.6. . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 150

6.8 Asymptotic values of the surface-area coefficient \( B_V \) as a function of the solid-phase volume fraction \( \phi \) for (a) two-dimensional periodic networks and (b) ordered and disordered disk packings. We take the length scale as \( D = 1/s = 1 \), where \( s \) is the specific surface. In (b), SHU and PLP stand for the stealthy hyperuniform packing and perturbed-lattice packing, respectively. The inset in (b) is a magnification of the larger panel. . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 151
6.9 Numerical results for simple-cubic lattice packings of nonoverlapping spheres and cubes. (a) Surface-area coefficient $B_V(R)$ as a function of window radius $R$ at a selected solid-phase volume fraction $\phi = 0.9$. (b) Asymptotic values of the surface-area coefficient $\overline{B}_V$ as a function of the solid-phase volume fraction $\phi$. In both panels, we take the inverse of the specific surface $1/s$ to be unity, i.e., $D = 1/s = 1$.

7.1 (a-b) Portions of initial disordered tessellations: (a) Voronoi tessellation (black lines) of a nonhyperuniform packing (Sec. 7.5) and (b) a multiscale-disk tessellation (Sec. 7.6). A progenitor disk packing in (a) is illustrated by white dashed circles. The tessellation-based procedure can be applied to higher dimensions, but we illustrate two-dimensional cases here for simplicity. (c-d) Portions of disordered hyperuniform packings (dispersions) constructed from the initial tessellations (a-b) via the tessellation-based procedure, i.e., local-cell packing fraction $\phi$ of a particle (blue disks) within each cell is identical to the global packing fraction. Importantly, (d) multiscale coated-disks model corresponds to optimal Hashin-Shtrikman structures.
7.2 Schematics illustrating the tessellation-based procedure to construct disordered hyperuniform dispersions. (a) One first tessellates the space with disjoint cells whose maximal lengths are shorter than a certain length scale $\ell_{\text{max}}(\ll L)$, where $|C_j|$ represents the volume of cell $j$. (b) For a specified packing fraction $0 < \phi < 1$, within each cell $C_j$, one places a single particle of general shape of volume $\phi|C_j|$. (c) A special case of constructed dispersions with circular disks and a circular observation window of large radius $R(\gg \ell_{\text{max}})$. The volume-fraction fluctuations arise only inside a narrow yellow-shaded region whose effective thickness is thinner than $\ell_{\text{max}}$. (d) The corresponding hyperuniform dispersion of randomly oriented squares. The volume-fraction fluctuations arise only inside a narrow yellow-shaded region like (c).

7.3 Numerical simulations of probability distributions of the largest hole volume ($2r_{\text{max}}$) in each sample of 1D Poisson point patterns: (a) on a semilog scale and (b) on a log-log scale. The $x$-axis in (b) represents the relative size $\delta = v_1(r_{\text{max}})/V$ of the largest holes to the box volume $V(= L)$. For each particle number $N$, the distribution is obtained from $10^4$ independent samples. (c) Log-log plot of hole probability $E_V(r)$ of finite-size Poisson point patterns as a function of system size $N$. Values are computed from Eq. (7.30).
7.4 Schematic for three types of the maximal packing fractions $\phi_{\text{max}}^{(2)} \leq \phi_{\text{max}}^{(1)} \leq \phi_{\text{max}}$. For a Voronoi cell of an initial particle (white circle), the black circle and the green one represent the largest inscribed circles when the circle center is fixed or free to move, respectively. These three circles (white, black, and green ones) illustrate the largest particles $|P_j|$ of this cell for three distinct maximal packing fractions $\phi_{\text{max}}^{(2)}$, $\phi_{\text{max}}^{(1)}$, and $\phi_{\text{max}}$, respectively. We note that the initial particle size is exaggerated for a clear visualization.

7.5 (a) A portion of a hyperuniform disk packing that was converted from a 2D RSA packing with the packing fraction $\phi_{\text{init}} = 0.41025$. (b) A portion of a hyperuniform sphere packing that was converted from a 3D saturated RSA packing.

7.6 Log-log plot of the scaled local volume-fraction variance $v_1(R) \sigma^2_v(R)$ of the progenitor and constructed packings. The progenitor packings are 2D saturated RSA packings, and $\sigma^2_v(R)$ are estimated by the Monte Carlo method; see Chapter 6. Three vertical grids represent a quarter of side length of the simulation boxes (i.e., $R = L/4$) for $N = 10^4$, $10^5$, and $10^6$, respectively.
7.7 Simulation results for sphere packings from Voronoi tessellations of RSA packings in (a)-(c) $\mathbb{R}^2$ and (d)-(f) $\mathbb{R}^3$. (a) and (d) Probability density $p(|C_j|)$ of Voronoi cell volumes versus the scaled cell volume $|C_j|/|C|$ plotted on a semi-log scale (larger panel) and a linear scale (inset). Here, $|C|$ represents the expected cell volume. (b) and (e) The spectral densities versus wavenumber $k$ for small wavenumbers plotted on a log-log scale. Inset in (b) is on a linear scale. Here, we note that all packings are rescaled to the common packing fraction $\phi = 0.01$. (c) and (f) Log-log plots of the spectral densities for progenitor packings (saturated RSA) and associated constructed packings according to system size $N$.

7.8 Simulation results for disk packings converted from 2D equilibrium hard-disk liquids of $N = 10^5$. (a) Probability density $p(|C_j|)$ of Voronoi cell volumes versus the scaled cell volumes $|C_j|/|C|$ plotted on a semi-log scale. (b) The spectral densities versus wavenumber $k$ for small wavenumbers plotted on a log-log scale. Theoretical values of $\tilde{\chi}_V(0)$ are obtained from Eq. (7.31). (c) The spectral densities versus wavenumber $k$ for intermediate and large wavenumbers plotted on a semi-log (upper) and a linear (lower) scales. Here, the sample size is $N = 10^3$, and $\bar{\sigma}$ is the mean particle radius of the constructed packings.

7.9 Simulation results for constructed disk packings from 2D lattice packing for various values of vacancy concentration $c$. (a) Semi-log plots of probability density functions of Voronoi cell volumes ($N_s = 10^6$). (b) Log-log plots of the spectral densities for progenitor packings of various vacancy concentrations and the constructed packings ($N_s = 10^6$). (c) Log-log plots of the spectral densities of the constructed packings of various sample sizes ($c = 0.4$).
7.10 Simulation results for $\tilde{\chi}_V(k)$ of the constructed packings depending on particle shapes. Here, the progenitor point patterns are the centers of saturated RSA packings with $N = 10^6$ in (a) two and (b) three dimensions.

7.11 A representative of disordered coated-disks model with the local-cell packing fraction $\phi = 0.25$ derived from a sphere tessellation for a power-law scaling with $p = 1.5$ and $m = 400$. Its magnification is presented in Fig. 7.18.

7.12 Simulation results for multiscale-disk tessellations. For the power-law scaling (7.36), (a) the semi-log plot of fraction of uncovered space $(1 - \eta_m)$ versus the number of stages $m$, and (b) the log-log plot of total cell number $N_m$ versus $(1 - \eta_m)$. For the exponential scaling (7.39), (c) the semi-log plot of $(1 - \eta_m)$ in the $m$th stage, and (d) the log-log plot of total cell number $N_m$ as functions of $(1 - \eta_m)$. Here, we note that dashed lines represent prescribed covering fractions, given by Eqs. (7.38) and (7.41), and $\delta \equiv v_{\text{max}}/|V_F|$.

7.13 Simulation results for the coated-disks models for (a-b) power-law and (c-d) exponential scalings of cell volumes. (a) and (c) Log-log plots of the spectral densities $\tilde{\chi}_V^{(m)}(k)$ versus wavenumber $k$ for our coated-spheres model at various values of stages. (b) and (d) Log-log plots of the associated spectral densities $\tilde{\chi}_V^{(m)}(k = k_{\text{min}})$ at the minimal wavenumber as functions of the fraction of uncovered space $(1 - \eta_m)$. The dashed lines represent the values calculated from Eqs. (7.47) and (7.49), respectively.
7.14 Fabrication of designed hyperuniform dispersions and matrices in three dimensions. (a) A portion of the designed dispersion of spherical particles (left) and the corresponding matrix (right) at $\phi = 0.23$, which are constructed from a 3D saturated RSA packing. (b) A portion of the designed dispersion of cubical particles (left) and the corresponding matrix (right). The hyperuniform matrices with spherical or cubical pores can be fabricated using 3D printing techniques.

7.15 Schematic of implementation of VORO++ library [254]. A point pattern in a periodic simulation box is sampled by domains of equal size (squares bounded by dashed lines). Each domain consists of a subdomain (a shaded region in the center) and a marginal region surrounding the subdomain. Subdomains are mutually disjoint and fully cover a simulation box. The thickness of the marginal region is denoted by $W$.

7.16 Simulation results for the spectral densities $\tilde{\chi}_v(k)$ of 3D equilibrium hard-sphere liquids with various values of packing fraction $\phi_{\text{init}}$ and sample size $N$. “Prediction” represents $\tilde{\chi}_v(0)$, obtained by Eq. (7.50).

7.17 (a) The number $N_m$ of spheres inserted in the $m$th stage in our multiscale-disk tessellations. (b) The inserted number $N_m$ as a function of cell-volumes $v^{(m)}$. Error bars represent sample standard deviations.

7.18 An enlarged portion ($\times 8$ magnification) of Fig. 7.11. This packing is generated via a power-law scaling with $p = 1.5$ up to the 400th stage.
8.1 Representative images of configurations of the four models of 2D disordered particulate media described in this section. These include (a) overlapping spheres, (b) equilibrium packings, (c) class I hyperuniform polydisperse packings, and (d) stealthy hyperuniform packings. For all models, the volume fraction of the dispersed phase (shown in black) is \( \phi_2 = 0.25 \). Note that (a) and (b) are not hyperuniform.

8.2 Plots of the spectral density \( \tilde{\chi}_V(Q) \) for the four models of 3D disordered media: overlapping spheres, equilibrium packings, class I hyperuniform polydisperse packings, and stealthy hyperuniform packings. In all cases, the volume fraction of the dispersed phase is \( \phi_2 = 0.25 \). For hyperuniform polydisperse packings, \( a \) is the mean sphere radius. The other three models consist of identical spheres of radius \( a \). Corresponding graphs of the spectral densities for the 2D models are provided in Appendix 8.20.

8.3 (a) Schematic of a large \( d \)-dimensional ellipsoidal, macroscopically anisotropic two-phase composite medium embedded in an infinite reference phase of dielectric constant tensor \( \varepsilon_I \) (gray regions) under an applied electric field \( \mathbf{E}_0(\mathbf{x}) = \tilde{\mathbf{E}}_0 \exp(i(\mathbf{k}_I \cdot \mathbf{x} - \omega t)) \) of a frequency \( \omega \) and a wavevector \( \mathbf{k}_I \) at infinity. The wavelength \( \lambda \) associated with the applied field can span from the quasistatic regime (\( 2\pi \ell/\lambda \ll 1 \)) down to the intermediate-wavelength regime (\( 2\pi \ell/\lambda \lesssim 1 \)), where \( \ell \) is the inhomogeneity length scale. (b) After homogenization, the same ellipsoid can be regarded to be a homogeneous specimen with an effective dielectric constant \( \varepsilon_e(\mathbf{k}_I, \omega) \), which depends on \( \omega \) and \( \mathbf{k}_I \). As noted in the main text, we omit the \( \omega \) dependence of \( \varepsilon_e \) because (without loss of generally) we assume a linear dispersion relation between \( |\mathbf{k}_I| \) and \( \omega \).
8.4 Schematic of the optimal multiscale "coated-spheres" model that realizes the isotropic Hashin-Shtrikman bounds on $\varepsilon_e$ [115]. Each composite sphere is composed of a spherical inclusion of one phase (dispersed phase) that is surrounded by a concentric spherical shell of the other phase such that the fraction of space occupied by the dispersed phase is equal to its overall phase volume fraction. The composite spheres fill all space, implying that their sizes range down to the infinitesimally small. When phase 2 is the disconnected inclusion (dispersed) phase, this two-phase medium minimizes and maximizes the effective static dielectric constant $\varepsilon_e$ for prescribed volume fraction and contrast ratio, when $\varepsilon_2/\varepsilon_1 > 1$ and $\varepsilon_2/\varepsilon_1 < 1$, respectively. It has recently been proved that these highly degenerate optimal Hashin-Shtrikman multiscale distributions of spheres are hyperuniform [157, 156].

8.5 The negatives of (a) the real and (b) imaginary parts of the nonlocal attenuation function $F(Q)$, defined in Eq. (8.71), for the four models of 3D disordered composite media considered in this chapter. The inset in (b) is the log-log plot of the larger panel. The volume fraction of the dispersed phase for each model is $\phi_2 = 0.25$. The first three models consist of identical spheres of radius $a$. For class I hyperuniform polydisperse particulate media, $a$ is the mean sphere radius.
8.6 Schematic of the general simulation setup for either (a) periodic or (b) non-periodic consisting of \( N \) spheres of radius \( a \) in a matrix. In both cases, Gaussian pulses of electric fields propagate from the planar sources (shown in red lines) to the packings (shown in black circles). The wavenumber (spectrum) of the pulses spans between \( \min[k_1] \) and \( \max[k_1] \). Periodic boundary conditions are applied along all directions, except for the propagation direction \( \hat{x} \). The perfectly matched layers (PML, shown in blue) of thickness \( L_{\text{pml}} \) are placed at the both ends of the simulation box to absorb any reflected and transmitted waves.

8.7 Comparison of the predictions of the strong-contrast formulas, Eqs. (8.70) and (8.76), to the Maxwell-Garnett [Eqs. (8.16) and (8.15)] and QCA (8.17) approximations for the effective dynamic dielectric constant \( \varepsilon_e(k_1) \) of periodic packings to our corresponding computer simulation results. We consider (a) 3D simple cubic lattice and (b) 2D square lattice of packing fraction \( \phi_2 = 0.25 \) and contrast ratio \( \varepsilon_2/\varepsilon_1 = 4 \). Here \( k_1 \) is the wavenumber in the reference (matrix) phase along the \( \Gamma-X \) direction, and \( L \) is the side length of a unit cell.

8.8 Comparison of the predictions of the strong-contrast formulas, Eqs. (8.70) and (8.76), to the MGA(8.15) and QCA (8.17) approximations for the effective dynamic dielectric constant \( \varepsilon_e(k_1) \) of 3D disordered sphere packings to our corresponding computer simulation results. We consider (a) equilibrium packings and (b) stealthy hyperuniform packings \( \tilde{\chi}_v(Q) = 0 \) for \( 0 \leq Qa < 1.5 \) of sphere radius \( a \), packing fraction \( \phi_2 = 0.25 \), and phase contrast ratio \( \varepsilon_2/\varepsilon_1 = 4 \). Here \( k_1 \) is the wavenumber in the reference (matrix) phase, and the error bars in the FDTD simulations represent the standard errors over independent configurations.
8.9 Predictions of the scaled strong-contrast approximation (8.76) for the effective dynamic dielectric constant $\varepsilon_e(k_1)$ of the four models of disordered media at volume fraction $\phi_2 = 0.25$ and contrast ratio $\varepsilon_2/\varepsilon_1 = 10$: (a) three dimensions and (b) two dimensions. The inset in the lower panel is the log-log plot of the larger panel.

8.10 Predictions of the strong-contrast approximation (8.76) for the effective dynamic dielectric constant $\varepsilon_e(k_1)$ of the four disordered models, as per Fig. 8.9, as function of dielectric-contrast ratio $\varepsilon_2/\varepsilon_1$ at packing fraction $\phi_2 = 0.25$ and wavenumber (a) $\sqrt{\varepsilon_{HS}/\varepsilon_1}k_1a = 0.7$ in three dimensions and (b) $\sqrt{\varepsilon_{HS}/\varepsilon_1}k_1a = 0.6$ in two dimensions.

8.11 Predictions of the scaled strong-contrast approximation (8.76) for the effective wave speed $c_e$ and the negative of the attenuation coefficient $\gamma_e$ as a function of $k_1$ for 3D stealthy hyperuniform sphere packings of contrast ratio $\varepsilon_2/\varepsilon_1 = 4$ at two different packing fractions: $\phi_2 = 0.4$ with $Q_Ua = 1.5$ and $\phi_2 = 0.25$ with $Q_Ua \approx 1.33$. The inset is a magnification of the lower panel.

8.12 Numerical verification of the Kramers-Kronig relations for the (unscaled) strong-contrast approximation. We consider 3D equilibrium packing of packing fraction $\phi_2 = 0.25$ and contrast ratio $\varepsilon_2/\varepsilon_1 = 4$. In the upper panel, we compare the real part $\text{Re}[\varepsilon_e(k_1)]$ of the approximation to that evaluated from the Kramers-Kronig relation (8.88) and the imaginary part of the strong-contrast approximation. In the lower panel, we compare the imaginary part $\text{Im}[\varepsilon_e(k_1)]$ of the approximation to that evaluated from (8.89) and the real part of the approximation. The predictions from the approximation and its transform via Kramers-Kronig relations show excellent agreement.
8.13 Evaluation of negatives of (a) the real and (b) negative imaginary parts of the local attenuation function $F(Q)$, defined in Eq. (8.99), for the four models of three-dimensional disordered particulate media. Each model has the same volume fraction for the dispersed phase $\phi_2 = 0.25$. The inset in (b) is the log-log plot of the larger panel. The first three models consist of spheres of radius $a$. For class I hyperuniform packings via tessellation-based procedure, $a$ is the mean sphere radius, i.e., $\phi_2 = \rho v_1(a)$, where $\rho$ is number density of particle centers, and $v_1(a)$ is the volume of a $d$ dimensional sphere of radius $a$.

8.14 Comparison of negatives of (a) the real and (b) imaginary parts of the nonlocal attenuation function $F(Q)$ [see Eq. (8.170)] and its local counterpart $F(Q)$ [see Eq. (8.99)] for 3D disordered models. We consider (left panel) stealthy hyperuniform packings and (right panel) stealthy nonhyperuniform packings of packing fraction $\phi_2 = 0.25$ and identical spheres of radius $a$.

8.15 Comparison of $\varepsilon_e(k_1)$ for periodic packings of $\phi_2 = 0.25$ estimated from the band-structure (via MPB) and FDTD simulations along the $\Gamma$-$X$ direction. Three systems are considered: (a) square lattice packing of $\varepsilon_2/\varepsilon_1 = 1/4$, (b) square lattice packing of $\varepsilon_2/\varepsilon_1 = 4$, and (c) simple cubic lattice packing of $\varepsilon_2/\varepsilon_1 = 4$. Note that the band-structure calculations are omitted in the photonic bandgaps. Both types of simulations show excellent agreement, implying that our homogenization estimates from FDTD simulations are valid down to intermediate wavelengths ($k_1L < 4$ and $k_1a \lesssim 1.1$).
8.16 Plots of (a) spectral density, and (b) the real and (c) imaginary parts of the nonlocal attenuation function for the four models of two-dimensional disordered particulate composite media. All models have the same volume fraction of dispersed phase $\phi_2 = 0.25$. Here, the first three models consist of identical spheres of radius $a$. For class I hyperuniform packings via tessellation-based procedure, $a$ is the mean sphere radius, i.e., $\phi_2 = \rho v_1(a)$. 268

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\[
\begin{align*}
\text{Contour plots, evaluated from Eq. (14) in the main text, represent the surface on which } \epsilon_e \text{ and the real (the upper panel) and the imaginary (the lower panel) parts of } K_e \text{ at a prescribed wavenumber } k_{Lq} \text{ should lie. We then overlap cross-property relations for four models of 3D disordered dispersions with } \phi_2 = 0.25: \text{ stealthy hyperuniform (SHU), stealthy nonhyperuniform (SNHU), overlapping spheres (OVS), and equilibrium hard spheres (EHS). These curves start from the Hashin-Shtrikman bounds (blue stars) at } k_{Lq} a = 0 \text{ to } k_{Lq} a = 5. 
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Chapter 1

Introduction

The hyperuniformity concept generalizes the traditional notion of long-range order in many-particle systems to include all perfect crystals, perfect quasicrystals, and certain exotic amorphous states of matter [313, 346, 299, 181, 300]. A hyperuniform point patterns in $d$-dimensional Euclidean space $\mathbb{R}^d$ is characterized by an anomalous suppression of large-scale density fluctuations relative to those in typical disordered systems, such as liquids and structural glasses. The hyperuniformity notion was generalized to the case of two-phase (heterogeneous) composite media [346, 299], i.e., materials consisting of two phases, such as composites, porous media, foams, cellular solids, colloidal suspensions, and polymer blends.

While disordered hyperuniform systems were largely unnoticed about a decade and a half ago, there has been an increasing realization that such systems arise in a wide range of topics across the physical [317, 354, 121, 245, 225, 186, 343, 324, 300, 177, 104, 85, 155, 160, 300, 78, 110, 180, 48, 185], chemical [252, 112, 194], mathematical [276, 231, 97, 30, 316], biological [137, 203, 171, 300, 362], and materials sciences [193, 339, 303, 46, 157]. Furthermore, evidence is beginning to emerge that such materials have important practical and technological implications, such as photonic band gap materials [80], efficient light extraction [41, 104], and transparent...
amorphous materials [179]. Material microstructures with such unusual correlated
disorder also can provide advantages over periodic ones with high crystallographic
symmetries [186, 341], which include perfect isotropy and robustness against defects
[79, 196]. Remarkably, such unusual amorphous systems can exist as equilibrium or
nonequilibrium states of matter. However, our theoretical understanding of disor-
dered hyperuniform systems is still in its infancy.

This dissertation reports results on the structure and properties of hyperuniform
point patterns (Chapters 2-5) and hyperuniform two-phase composite media (Chap-
ters 6-10), including both ordered and disordered varieties. These results were ob-
tained by using both theoretical and computational methods of statistical mechanics.
In what follows, we summarize the individual chapters of the dissertation.

In Chapter 2, we study how aspherical window shapes affect the characterization
of hyperuniformity of point patterns. Hyperuniform point patterns in $d$-dimensional
space $\mathbb{R}^d$ are characterized by an anomalous suppression of infinite-wavelength density
fluctuations, i.e., the local number variance within a “spherical” observation window
grows slower than the window volume. This direct-space condition is equivalent to
the Fourier-space hyperuniformity condition that the structure factor vanishes as
the wavenumber goes to zero. However, we have demonstrated that for certain sta-
tistically anisotropic hyperuniform systems, including lattices, the use of aspherical
windows of a linear characteristic size $L$ can result in the variance growth rate de-
pending on the window shape and orientation. In some cases, the growth rate can be
even faster than the window volume (i.e., $L^d$), which may lead one to falsely conclude
that the system is nonhyperuniform solely according to the direct-space condition.
Specifically, to understand the window-shape dependence, we numerically investigate
the variance of two-dimensional lattices using “superdisk” windows with a fixed ori-
tentation defined as $|x|^{2p} + |y|^{2p} = L^{2p}$, whose convex shapes continuously interpolate
between circles ($p = 1$) and squares ($p = \infty$), as prescribed by a deformation parame-
ter $p$. Subsequently, using square windows (superdisk when $p = \infty$), we demonstrate that the large-$L$ asymptotic behaviors of variance for 2D lattices depend on window orientations, which can grow faster than the window area or even slower than the window perimeter [e.g., like $\ln(L)$]. We then generalize the window-orientation condition, under which the variance can grow as fast as or faster than $L^d$ (window volume), to the case of Bravais lattices and parallelepiped windows in $\mathbb{R}^d$. In the case of statistically isotropic systems, however, we prove that the large-$L$ asymptotic behavior of the variance is independent of the window shape for convex windows. Based on this observation, unlike the conventional variance using windows with a fixed orientation, we show that the orientation-average of variance $\langle \sigma^2_N(R) \rangle_O$ exhibits a common large-window asymptotic behavior for an arbitrarily-shaped convex window. Finally, when aspherical windows are employed, we suggest a new direct-space hyperuniformity condition based on $\langle \sigma^2_N(R) \rangle_O$. The work in this chapter has been published previously as the following refereed journal article:


This work has also been presented in short talks at the 115th Statistical Mechanics Conference at Rutgers University in May 2016, and at the 116th Statistical Mechanics Conference at Rutgers University in December 2016.

In Chapter 3, we study the degree to which the introduction of imperfections in perfectly hyperuniform systems degrades or destroys their original perfect hyperuniformity. A quantitative understanding of such imperfection effects is practically and theoretically important because, in practice, there are no "perfect" ordered or disordered hyperuniform systems due to the inevitable presence of imperfections. We first derive explicit formulas for $S(k)$ in the small wavenumber regime for three types of imperfections: (1) uncorrelated point defects, including vacancies and interstitials,
stochastic particle displacements, and (3) thermal excitations in the classical harmonic regime. We then verify these theoretical predictions via numerical simulations. We have found that introducing the first and third types of imperfections destroy hyperuniformity immediately. By contrast, stochastic displacements (i.e., the second type) destroy hyperuniformity only when they are strongly correlated. It is noteworthy that imperfections in disordered hyperuniform systems can be unambiguously detected. These results provide the theoretical underpinnings to systematically study the effect of imperfections on the physical properties of hyperuniform materials. The work in this chapter has been published previously as the following refereed journal article:


This work has also been presented in short talks at the 117th Statistical Mechanics Conference at Rutgers University in May 2017, and at the 118th Statistical Mechanics Conference at Rutgers University in December 2017.

In Chapter 4, we investigate special examples of stochastic particle displacements of a lattice, discussed in Chapter 3. Random, uncorrelated displacements of particles on a lattice preserve the hyperuniformity of the original lattice, that is, normalized density fluctuations vanish in the limit of infinite wavelengths. Typically, the resulting point pattern inherits the Bragg peaks (long-range order) of the original lattice. We demonstrate how these Bragg peaks can be hidden in the effective diffraction pattern of independent and identically distributed perturbations. All Bragg peaks vanish if and only if the sum of all probability densities of the positions of the shifted lattice points is a constant at all positions. The underlying long-range order is then “cloaked” in the sense that it cannot be reconstructed from the pair-correlation function alone. On the one hand, density fluctuations increase monotonically with the strength of perturbations $a$, as measured by the hyperuniformity order metric $\Lambda$. On the other
hand, the disappearance and re-emergence of long-range order, depending on whether
the system is cloaked or not as the perturbation strength increases, is manifestly
captured by the $\tau$ order metric. Therefore, while the perturbation strength $a$ may
seem to be a natural choice for an order metric of perturbed lattices, the $\tau$ order
metric is a superior choice. It is noteworthy that cloaked perturbed lattices allow
one to easily simulate very large samples (with at least $10^6$ particles) of disordered
hyperuniform point patterns without Bragg peaks. The work in this chapter has been
published previously as the following refereed journal article:

- M. A. Klatt, J. Kim, and S. Torquato, Cloaking the Underlying Long-Range

In Chapter 5, we study the sufficient conditions for inverting the collective coordi-
nates in a many-particle system that are complex Fourier components of the particle
density $n(x) \equiv \sum_{j=1}^{N} \delta(x - r_j)$. While collective coordinates often provide useful
physical insights, it is desirable to infer the particle coordinates via inverse trans-
formations for given collective coordinates. In principle, a sufficiently large set of
collective coordinates are equivalent to particle coordinates, but the nonlinear rela-
tion between collective and particle coordinates makes the inversion procedure highly
nontrivial. Given a “target” configuration in one-dimensional Euclidean space, we
investigate the minimal set of its collective coordinates that can be uniquely inverted
into particle coordinates. For this purpose, we treat a finite number $M$ of the real
and/or the imaginary parts of collective coordinates of the target configuration as
constraints, and then reconstruct “solution” configurations whose collective coordi-
nates satisfy these constraints. Both theoretical and numerical investigations reveal
that the number of numerically distinct solutions depends sensitively on the chosen
collective-coordinate constraints and target configurations. We conclude that collec-
tive coordinates at the $\left\lceil \frac{N}{2} \right\rceil$ smallest wavevectors are the minimal set of constraints for
unique inversion, where $\left\lceil \cdot \right\rceil$ represents the ceiling function. This result provides useful
groundwork for the inverse transform of collective coordinates in higher-dimensional systems. The work in this chapter has been published previously as the following refereed journal article:


In Chapter 6, we change our focus to study hyperuniform two-phase media. The hyperuniformity concept provides a unified means to classify all perfect crystals, perfect quasicrystals, and exotic amorphous states of matter according to their capacity to suppress large-scale density fluctuations. While the classification of hyperuniform point configurations has received considerable attention, much less is known about the classification of hyperuniform heterogeneous two-phase media, including composites, porous media, foams, cellular solids, colloidal suspensions, and polymer blends. This chapter aims to begin such a program for certain two-dimensional models of hyperuniform two-phase media by ascertaining their local volume-fraction variances $\sigma_V^2(R)$ and the associated hyperuniformity order metrics $B_V$. This is a highly challenging task because the geometries and topologies of the phases are generally much richer and more complex than point-configuration arrangements, and one must ascertain a broadly applicable length scale to make key quantities dimensionless. Therefore, we purposely restrict ourselves to a certain class of two-dimensional periodic cellular networks as well as periodic and disordered/irregular packings, some of which maximize their effective transport and elastic properties. Among the cellular networks considered, the honeycomb networks have the minimal value of the hyperuniformity order metrics $\mathcal{B}_V$ across all volume fractions. On the other hand, among all packings considered, the triangular-lattice packings have the smallest values of $\mathcal{B}_V$ for the possible range of volume fractions. Among all structures studied in this chapter, the triangular-lattice packing has the minimal order metric for almost all volume frac-
tions. Our study provides a theoretical foundation for establishing hyperuniform order metrics for general two-phase media and a basis to discover new hyperuniform two-phase systems by inverse design procedures. The work in this chapter has been submitted to Physical Review E as:


In Chapter 7, we develop a geometric principle to construct disordered hyperuniform two-phase media. While such materials have received considerable attention, a stumbling block has been an inability to create large samples that are truly hyperuniform due to current computational and experimental limitations. By contrast, our procedure makes it possible for the first time to construct perfectly hyperuniform systems of unprecedentedly large system sizes, e.g., up to ten million particles. This procedure involves tessellating space into cells and then inserting a particle into each cell such that the local-cell particle packing fractions are identical to the global packing fraction. We prove that the resulting dispersions are perfectly hyperuniform in the infinite-system-size limit. Then we numerically demonstrate in two and three dimensions that this methodology can efficiently convert a very large nonhyperuniform disordered dispersion into a *perfectly* hyperuniform one. Using a similar analysis, we also establish the hyperuniformity of the famous *Hashin-Shtrikman multiscale dispersions*, which possess optimal transport and elastic properties. We discuss the feasibility of these hyperuniform designs via modern photolithographic and 3D printing technologies. Our tessellation-based procedure enables one to design an enormous class of hyperuniform dispersions by tuning particle shapes, positions, and numbers within each cell while preserving hyperuniformity. Exploration of the resulting dispersions paves the way for accelerating the discovery of novel disordered hyperuniform materials. The work in this chapter has been published previously as the following refereed journal articles:


They have also been presented in short talks at the 119th Statistical Mechanics Conference at Rutgers University in May 2018; at the 120th Statistical Mechanics Conference at Rutgers University in December 2018; at APS March Meeting at Boston in March 2019; at the 121st Statistical Mechanics Conference at Rutgers University in May 2019; and at CMO workshop at Oaxaca, Mexico in October 2019.

In Chapters 8-10, we derive approximate formulas to estimate the effective electric and elastic wave characteristics of two-phase composite materials, including both hyperuniform and nonhyperuniform systems. The strong-contrast expansion formalism is extended to derive the exact series expansions accounting for complete microstructural information (infinite set of $n$-point correlation functions) and hence multiple scattering to all orders for the wider range of wavelengths. By utilizing the fast-convergence properties of these resulting expansions, we then obtain accurate approximations for the effective dynamic dielectric constant and elastic moduli that apply beyond the long-wavelength regime in Chapters 8 and 9, respectively. Using these results, we show that disordered hyperuniform media are generally less lossy than their nonhyperuniform counterparts. We also show that certain disordered hyperuniform particulate composites exhibit novel wave characteristics, including the capacity to act as low-pass filters that transmit electric and elastic waves “isotropically” up to a selected wavenumber.

In Chapter 8, we derive exact nonlocal homogenized constitutive relations for the effective electromagnetic wave properties of disordered two-phase composites and metamaterials from first principles. This exact formalism enables us to extend the
long-wavelength limitations of conventional homogenization estimates of the effective dynamic dielectric constant tensor $\varepsilon_e(k,\omega)$ for arbitrary microstructures so that it can capture spatial dispersion well beyond the quasistatic regime (where $\omega$ and $k$ are frequency and wavevector of the incident radiation). We accomplish this by deriving nonlocal strong-contrast expansions that exactly account for complete microstructural information (infinite set of $n$-point correlation functions) and hence multiple scattering to all orders for the range of wavenumbers for which our extended homogenization theory applies, i.e., $0 \leq |k| \ell \lesssim 1$ (where $\ell$ is a characteristic heterogeneity length scale). Due to the fast-convergence properties of such expansions, their lower-order truncations yield accurate closed-form approximate formulas for $\varepsilon_e(k,\omega)$ that apply for a wide class of microstructures. These nonlocal formulas are resummed representations of the strong-contrast expansions that still accurately capture multiple scattering to all orders via the microstructural information embodied in the spectral density, which is easy to compute for any composite. The accuracy of these microstructure-dependent approximations is validated by comparison to full-waveform simulation methods for both 2D and 3D ordered and disordered models of composite media. Thus, our closed-form formulas enable one to predict accurately and efficiently the effective wave characteristics well beyond the quasistatic regime for a wide class of composite microstructures without preforming full-waveform simulations. We also show that certain disordered hyperuniform two-phase composites exhibit novel wave characteristics, including the capacity to act as low-pass filters and refractive indices that abruptly change over a narrow range of wavenumbers. Our results can facilitate the design of disordered composites with prescribed effective wave characteristics by engineering spatial correlations of microstructures at desired length scales. The work in this chapter has been submitted to Physical Review X:

- S. Torquato and J. Kim, Nonlocal Effective Electromagnetic Wave Characteristics of Composite Media: Beyond the Quasistatic Regime, (2020).
In Chapter 9, we derive exact expressions for effective elastodynamic properties of two-phase composites in the long-wavelength (quasistatic) regime via homogenized constitutive relations that are local in space. This is accomplished by extending the “strong-contrast” expansion formalism that was previously applied to the static problem. These strong-contrast expansions explicitly incorporate complete microstructural information of the composite via an infinite set of \( n \)-point correlation functions. Utilizing the rapid-convergence properties of these series expansions (even for extreme contrast ratios), we extract accurate approximations that depend on the microstructure via the spectral density, which is easy to compute or measure for any composite. We also investigate the predictive power of modifications of such approximation formulas postulated elsewhere [J. Kim and S. Torquato, Proc. Nat. Acad. Sci. 117, 8764 (2020)] to extend their applicability beyond the quasistatic regime. The accuracy of these nonlocal microstructure-dependent approximations is validated by comparison to full-waveform simulation results for certain models of dispersions. We apply our formulas to a variety of models of nonhyperuniform and hyperuniform disordered composites. Finally, we discuss how to utilize our approximations for engineering composites with prescribed elastic wave characteristics. The work in this chapter has been submitted to the New Journal of Physics:


In Chapter 10, we establish cross-property relations for two-phase composite media that link effective elastic and electromagnetic wave characteristics to one another, including the respective effective wave speeds and attenuation coefficients, which facilitate multifunctional material designs. This is achieved by utilizing the accurate approximations at the two-point level for the effective electromagnetic and elastodynamic properties that are derived in Chapters 8 and 9, respectively. Our formulas enable us to explore the wave characteristics of a broad class of disordered
microstructures because they apply, unlike conventional formulas, for a wide range of incident wavelengths, i.e., well beyond the long-wavelength regime. This capability enables us to study the dynamic properties of exotic disordered “hyperuniform” composites that can have advantages over crystalline ones, such as nearly optimal, direction-independent properties and robustness against defects. Our cross-property relations for the effective wave characteristics can be applied to design multifunctional composites via inverse techniques. Design examples include structural components that require high stiffness and electromagnetic absorption, heat-sinks for CPUs, and sound-absorbing housings for motors that have to emit thermal radiation and suppress mechanical vibrations efficiently, and nondestructive evaluation of the elastic moduli of materials from the effective dielectric response. The work in this chapter has been published previously as:


This work has also been presented in a short talk at APS DSOFT Virtual Sessions in March 2020.
Chapter 2

Effect of Window Shape on the Detection of Hyperuniformity via the Local Number Variance

2.1 Introduction

A hyperuniform state matter is characterized by an anomalous suppression of density fluctuations at large length scales [313, 346, 299]. The hyperuniformity concept provides a unified way to categorize crystals, quasicrystals, and certain exotic disordered systems [313, 346, 345]. Disordered hyperuniform states lie between a crystal and liquid: they behave like perfect crystals in the manner in which they suppress large-scale density fluctuations and yet, like liquids and glasses, are statistically isotropic without Bragg peaks. In this sense, disordered hyperuniform systems have a hidden order on large length scales, which endows them with novel physical properties [137, 79, 80, 196, 179]. During the last decade, it has been discovered that these systems play a vital role in a number of problems across the physical, mathematical, and biological sciences. Specifically, we now know that disordered hyperuniform materials
can exist as both equilibrium and nonequilibrium phases, including maximally random jammed packings [138, 23, 62], Coulomb gas [176, 135, 66], certain fermionic and bosonic systems [259, 311, 77], liquids that freeze into degenerate disordered ground states [197], novel disordered photonic materials [80, 79, 196], spatial patterns of photoreceptors in avian retina [137], structure of bird feathers [220], highly excited states of ultracold gases [178], terahertz quantum cascade lasers [57], driven nonequilibrium systems [122, 261, 133], transparent dense disordered materials [179], and number theory [66, 216].

Figure 2.1: Schematics indicating an observation window $\Omega$ and its centroid $x_0$ for a disordered point pattern (left) and a periodic one (right), as adapted from [313].

Consider a point process in $d$-dimensional Euclidean space $\mathbb{R}^d$ and let $N(R, x_0)$ denote the number of points contained in a $d$-dimensional window $\Omega$, whose shape and centroid position are characterized by $R$ and $x_0$, respectively; see Fig. 2.1. Density fluctuations can be quantified by $\sigma^2_N(R)$, i.e., the variance in $N(R, x_0)$ over either the ensemble of the point process or the window centroid position $x_0$ for a realization of the point process. The quantity $\sigma^2_N(R)$ is directly related to the pair statistics of the point process and the geometry of the window $\Omega$ in the following way [313, 346, 299]: see Sec. 2.2.2.

For a Poisson point process, $h(r) = 0$ for all $r$, and the number variance grows as fast as the window volume $v_1(R)$; see Eq. (2.14). This volume-like growth of
is typical of most disordered systems, including liquids and structural glasses [313, 197, 125]. A hyperuniform [313] (also known as “superhomogeneous” [86]) point process is defined by the following infinite-wavelength behavior of the structure factor:

\[
\lim_{{|k| \to 0}} S(k) = 0, \tag{2.1}
\]

which we call the Fourier-space hyperuniformity condition. The use of definition (2.1) for spherical windows (and some aspherical windows with sufficiently smooth boundaries) implies that hyperuniform point processes have vanishing normalized density fluctuations at large length scales as specified by [313, 299]:

\[
\lim_{{v_1(R) \to \infty}} \frac{\sigma_N^2(R)}{v_1(R)} = 0, \tag{2.2}
\]

which is the usual direct-space hyperuniformity condition. Henceforth, we will call this spherical-window as before. For spherical (and many apherical) windows, \(\sigma_N^2(R)\) for a hyperuniform point process has a growth rate that varies between the window surface area \(s_1(R)\) and the window volume \(v_1(R)\) in the large-window limit. The variance for perfect crystals and a large class of quasicrystals grows asymptotically like \(s_1(R)\).

The hyperuniformity concept has been extended to two-phase heterogeneous media [346, 299]. Here, one needs to use the spectral density \(\tilde{\chi}_V(k)\) associated with the appropriate two-point probability function and the local volume-fraction variance \(\sigma_V^2(R)\). Then, the Fourier-space hyperuniformity condition is

\[
\lim_{{|k| \to 0}} \tilde{\chi}_V(k) = 0, \tag{2.3}
\]
and equivalently, the corresponding spherical-window condition is

\[
\lim_{v_1(R) \to \infty} v_1(R) \sigma^2_V(R) = 0. \quad (2.4)
\]

Hyperuniform systems can be identified through small-angle scattering experiments [116, 338], yielding either the structure factor \(S(k)\) or the spectral density \(\chi_V(k)\). However, for some systems, e.g., colloidal suspensions [64, 329], nano-copolymers [364] and simulations [122], scattering experiments may not be available. In such instances, one can measure the local number variance \(\sigma^2_N(R)\) in direct space via microscopy to ascertain hyperuniformity of these systems [64]. In this chapter, we focus on hyperuniform point processes.

Previous theoretical investigations on the number variance of hyperuniform systems have primarily focused on spherical windows. By contrast, little attention has been paid to the analysis of aspherical windows, except for a few investigations [150, 19, 239, 349], including Beck’s study of the use of rectangular windows that have a fixed height and a special orientation to analyze the square lattice [19]. He showed that in this case, the variance could grow slower than the window surface area. Zachary et al. [349] studied the two-dimensional checkerboard model and square lattice decorated by identical squares, and showed that their volume-fraction variances decrease as slow as the inverse of the window volume, i.e., \(v_1(R)^{-1}\), despite the fact that these systems are hyperuniform [see Eq. (2.4)]. In summary, we see that for certain window shapes, the spherical-window hyperuniformity criterion alone may lead one to falsely conclude that a hyperuniform system (as identified via the Fourier-space condition) is non-hyperuniform.

Thus, the overall objective of this chapter is to understand the effect of aspherical window shapes on the asymptotic growth rate of the variance quantitatively and to resolve the possible inconsistencies that may arise with respect to the spherical-
window condition (2.2). It is noteworthy that the study of $\sigma^2_N(R)$ for aspherical windows is an interesting problem in physics and mathematics in its own right. For example, it has been shown that finding a window-shape that minimizes $\sigma^2_N(R)$ of a point process is equivalent to designing a finite-ranged repulsive pair potential that leads the point process to be the ground state [313]. For periodic systems and regular polyhedral windows, we will show that the window orientation with respect to the system significantly affects the large-window asymptotic behavior of $\sigma^2_N(R)$ (Sec. 2.4). This is also one of many physical examples [11, 37, 124, 335, 52, 235], in which (in)commensurability of a certain parameter plays a crucial role in the physical properties, e.g., friction coefficient [37] and Hall conductivity [52]. It is interesting to note that our analysis of the window-orientation-dependence of the variance under the incommensurate condition (Sec. 2.4.2) is closely related to Diophantine analysis in number theory [20].

We begin by studying the variance for the lattices using aspherical windows that have a fixed orientation with respect to the lattices. For this purpose, we numerically investigate the variance for the square lattice using the “superdisk” windows. A superdisk is the two-dimensional version of the versatile superball in $d$-dimensional Euclidean space $\mathbb{R}^d$, whose shape is defined by

$$|x_1|^{2p} + |x_2|^{2p} + \cdots + |x_d|^{2p} = L^{2p},$$

where a positive real number $p$ is called deformation parameter and $L$ is called the

Figure 2.2: Illustration of superdisk shapes for several values of the deformation parameters $p$. From the left to the right $p = 0, 0.25, 0.5, 0.75, 1, 1.25, \infty$. 
characteristic length scale. If the parameter $p$ is smaller than 0.5, a superdisk is concave, and it interpolates smoothly between across ($p = 0$) and a perfect square ($p = 0.5$). On the other hand, a superdisk with $p \geq 0.5$ is convex and is continuously transformed from a square ($p = 0.5$) to the circle ($p = 1$) and to a square of side length $2L$ ($p = \infty$), as shown in Fig. 2.2. Considering the case of $p \geq 1$, we show that the asymptotic behavior of the cumulative moving average of the variance has the power-law form, i.e., $\sigma^2_N(L) \sim L^\gamma$ as $L \to \infty$. We numerically demonstrate that the exponent $\gamma$ increases continuously from 1 to 2 as the window shape becomes closer to the perfect square, i.e., $p$ tends to $\infty$. When the window is a perfect square ($p = \infty$), $\gamma = 2$, the value of which might lead one to falsely conclude that the square lattice is not hyperuniform, since it conflicts with the spherical-window condition (2.2). We say that for a $d$-dimensional hyperuniform system, the growth rate of the variance is “anomalously” large whenever the exponent $\gamma \geq d$ because it is larger than what we expect from the “spherical-window” condition (2.2).

Subsequently, we investigate the variance for the $d$-dimensional cubic (or hypercubic) lattice using hypercubic windows (superball when $p = \infty$) of side length $2L$ to understand the mathematical conditions under which the variance is anomalously large at large length scales. When the windows are perfectly aligned with the lattice, we show that the variance grows like the square of the window surface area, i.e., $L^{2(d-1)}$ (see Sec. 2.9). Surprisingly, this growth rate can be even faster than the window volume, $L^d$, and such a large growth rate is typical of super-Poissonian point processes, such as systems at thermal critical points [127]. For more detailed analysis, we numerically and analytically investigate the two-dimensional square windows partly because the square is amenable to exact analysis and partly because it is the most frequently used aspherical window to detect the hyperuniformity of a two-dimensional system in direct space.

We also demonstrate that the asymptotic growth rate of $\sigma^2_N(L, \theta)$ depends on the
angle \( \theta \) between the symmetry axes of the window and the lattice. Importantly, we identify two classes of angles, at one of which, so-called rational angles, the variance grows "anomalously largely", i.e., \( \sigma_N^2(L, \theta) \sim L^2 \) as \( L \to \infty \). We explain the origin of such an orientational dependence from two different points of view: the correlation of density fluctuations concentrated in the vicinity of the window surface and conditional convergence of the second moment (\( d \)th moment in \( \mathbb{R}^d \)) of the total correlation function. Based on the analysis, we generalize the concept of "rational angles" for the square lattice and square window to Bravis lattices and parallelepiped windows in \( \mathbb{R}^d \) (see Sec. 2.8). To discuss the conditional convergence of the second moment of the total correlation function, we investigate this integral for the circular and square boundaries with Abelian summability method (see 2.10). In the case of statistically isotropic disordered hyperuniform point processes in \( \mathbb{R}^d \), we prove that the asymptotic behavior of \( \sigma_N^2(R) \) is independent of the window shape if it is convex.

In addition, we suggest a new direct-space hyperuniformity condition (2.73) using the orientationally-averaged local number variance \( \langle \sigma_N^2(R) \rangle_O \). We prove that for a \( d \)-dimensional anisotropic hyperuniform point process, corresponding to either a crystal or disordered one, \( \langle \sigma_N^2(R) \rangle_O \) always exhibits the same asymptotic behavior for any convex window shape. Then, we show that in the case of the square lattice and square windows, \( \langle \sigma_N^2(L) \rangle_O \sim L \), which is consistent with the case for circular windows.

In Sec. 2.2, we describe basic definitions and mathematical equations to compute the variance \( \sigma_N^2(R) \) for any window shape. We numerically compute the variance for the square lattice using superdisk windows of various shapes and show the relation between its asymptotic behavior and the deformation parameter \( p \) in Sec. 2.3. In Sec. 2.4, we investigate the case of the two-dimensional square lattice with a square window. In Sec. 2.5, we consider disordered hyperuniform point processes and study the asymptotic behavior of their variance for convex windows, including square windows. Orientationally-averaged variance is explained, and its asymptotic behavior is derived...
in Sec. 2.6. Finally, we provide concluding remarks in Sec. 2.7. A generalization of “rational angles” to \(d\)-dimensional Bravais lattices and parallelepiped windows is presented in Sec. 2.8. Then, we carry out some example calculations for the cases of the square lattice and rectangular windows with a fixed aspect ratio, and the triangular lattice and square windows. In Sec. 2.9, we show \(\sigma_N^2(L)\) for \(d\)-dimensional hypercubic lattice with aligned hypercubic windows of side length \(2L\).

### 2.2 Background and Definitions

#### 2.2.1 Point processes

Roughly speaking, a point process in \(d\)-dimensional Euclidean space \(\mathbb{R}^d\) is a distribution of infinitely many points \(r_1, r_2, \ldots\). For statistically homogeneous point processes in \(\mathbb{R}^d\) at a given number density \(\rho\), \(\rho^n g_n(r^n) \, dr^n\) represents the probability density for finding \(n\) points at \(r^n = r_1, r_2, \ldots, r_n\), and \(g_n(r^n)\) is called the \(n\)-particle correlation function. The statistical homogeneity of a point process implies that \(g_n\) is determined by only relative positions of \(n\) particles, i.e., \(g_n(r^n) = g_n(r_{21}, r_{31}, \ldots, r_{n1})\) with \(r_{ij} \equiv r_j - r_i\) for \(1 \leq i \neq j \leq n\).

The pair correlation function \(g_2(r)\) has a significant importance [179, 43, 75]. In systems without long-range order, \(g_2(r) \to 1\) as \(|r| \to \infty\). Therefore, it is useful to introduce the total correlation function \(h(r)\) defined as

\[
h(r) \equiv g_2(r) - 1, \quad (2.6)
\]

which decays to zero for large \(|r|\) in the absence of long-range order. The structure factor \(S(k)\) is related by Fourier transform of \(h(r)\):

\[
S(k) = 1 + \rho \tilde{h}(k). \quad (2.7)
\]
A (Bravais) lattice $\mathcal{L}$ in $\mathbb{R}^d$ belongs to a special subgroup of point processes, which can be expressed as integer linear combinations of $d$ linearly independent vectors $a_i$ for $i = 1, 2, \ldots, d$, i.e.,

$$\mathcal{L} = \left\{ \mathbf{x} = \sum_{i=1}^{d} n_i a_i \in \mathbb{R}^d \bigg| \mathbf{n} = (n_1, n_2, \ldots, n_d) \in \mathbb{Z}^d \right\}.$$ (2.8)

Every lattice $\mathcal{L}$ has a reciprocal lattice $\mathcal{L}^*$, which is a set of all reciprocal vectors $\mathbf{q}$ satisfying $\exp\{(i\mathbf{q} \cdot \mathbf{x})\} = 1$ for every $\mathbf{x} \in \mathcal{L}$. The structure factor $S(\mathbf{k}; \mathcal{L})$ of the lattice $\mathcal{L}$ is a sum of delta functions centered at each point in $\mathcal{L}^*$ except for the origin:

$$S(\mathbf{k}; \mathcal{L}) = \frac{(2\pi)^d}{\nu_c} \sum_{\mathbf{q} \in \mathcal{L}^* \setminus \mathbf{0}} \delta(\mathbf{k} - \mathbf{q}),$$ (2.9)

where $1/\nu_c$ is the number density of lattice $\mathcal{L}$, and $\delta(\mathbf{k})$ is the $d$-dimensional Dirac delta function. We will use the following definition of Fourier transform $\tilde{f}(\mathbf{k})$ and the inverse transform $f(\mathbf{r})$ (assuming their existence):

$$\tilde{f}(\mathbf{k}) = \int_{\mathbb{R}^d} f(\mathbf{r}) e^{-i \mathbf{k} \cdot \mathbf{r}} d\mathbf{r},$$ (2.10)

$$f(\mathbf{r}) = \frac{1}{(2\pi)^d} \int_{\mathbb{R}^d} \tilde{f}(\mathbf{k}) e^{i \mathbf{k} \cdot \mathbf{r}} d\mathbf{k}.$$ (2.11)

For radially symmetric functions, i.e., $f(\mathbf{r}) = f(|\mathbf{r}|)$ and $\tilde{f}(\mathbf{k}) = \tilde{f}(|\mathbf{k}|)$, the Fourier and inverse Fourier transform can be expressed as

$$\tilde{f}(k) = (2\pi)^{d/2} \int_0^\infty r^{d-1} f(r) \frac{J_{d/2-1}(kr)}{(kr)^{d/2-1}} dr,$$ (2.12)

$$f(k) = \frac{1}{(2\pi)^{d/2}} \int_0^\infty k^{d-1} \tilde{f}(k) \frac{J_{d/2-1}(kr)}{(kr)^{d/2-1}} dk,$$ (2.13)

where $J_\nu(x)$ is the Bessel function of order $\nu$. 

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2.2.2 Number variance and hyperuniformity

Consider a statistically homogeneous point process at the number density $\rho$ in $d$-dimensional Euclidean space. Using an observation window $\Omega$ whose shape and orientation are characterized by a set of parameters $R$, one can obtain the exact expression for $\sigma^2_N(R)$ in both the direct- and Fourier-space representations [313]:

$$\sigma^2_N(R) \equiv \langle N^2(R) \rangle - \langle N(R) \rangle^2 = \rho v_1(R) \left[ 1 + \rho \int_{\mathbb{R}^d} h(r) \alpha_2(r; R) \, dr \right] \quad (2.14)$$

$$= \frac{\rho v_1(R)}{(2\pi)^d} \int S(k) \tilde{\alpha}_2(k; R) \, dk, \quad (2.15)$$

where $S(k)$ is the structure factor, defined by (2.7), and $\alpha_2(r; R)$ is the scaled intersection volume of two identical windows but separated by $r$, i.e., $\Omega$ and $\Omega + r$. Note that both (2.14) and (2.15) represent the variance when windows have a fixed orientation with respect to the point process. Here, $\alpha_2(r; R)$ is expressed as the convolution of the window indicator functions $w(x; R)$:

$$\alpha_2(r; R) \equiv \frac{v_2^{\text{int}}(r; R)}{v_1(R)} = \frac{1}{v_1(R)} \int_{\mathbb{R}^d} w(x; R) w(x - r; R) \, dx, \quad (2.16)$$

where $v_1(R)$ is the window volume of $\Omega$. Clearly, the volume integral of $\alpha_2(r; R)$ over $\mathbb{R}^d$ is equal to the window volume:

$$\tilde{\alpha}_2(0; R) = \int_{\mathbb{R}^d} \alpha_2(r; R) \, dr = \int_{\mathbb{R}^d} w(r; R) \, dr = v_1(R). \quad (2.17)$$

For a $d$-dimensional sphere of radius $R$, the analytical expression for $\alpha_2(r; R)$ is well-known in any spatial dimension $d$ [296, 314]. The explicit expression for the Fourier transform of $\alpha_2(r; R)$ is given by

$$\tilde{\alpha}_2(k; R) = \frac{\tilde{w}(k; R)^2}{v_1(R)} = 2^d \pi^{d/2} \Gamma(1 + d/2) \left[ J_{d/2}(kR) \right]^2 \frac{1}{k^d}. \quad (2.18)$$
Denoting \( x \equiv r/2R \), \( \alpha_2(r; 2R) \) can be expressed in the series representation in terms of \( x \) for \( x < 1 \) [314]:

\[
\alpha_2(r; R) = 1 - c(d) x + c(d) \sum_{n=2}^{\infty} \frac{(-1)^n \Gamma((d+1)/2)}{(2n-1) \Gamma(n) \Gamma((d+3)/2-n)} x^{2n-1},
\]

(2.19)

where \( c(d) = 2 \frac{\Gamma(d/2 + 1)}{[\Gamma((d+1)/2) \Gamma(1/2)]} \) and \( \Gamma(x) \) is the Gamma function.

For a given window \( \Omega \) and a realization \( X \) of a point process, \( \langle N(R) \rangle \) represents the ensemble average of the number of particles of \( X \) within the window \( \Omega \), \( N(R; X) \), over every realization \( X \) of the point process. The ergodic hypothesis enables us to equate an ensemble average to a volume average in a single infinite realization \( X \) [296], and trivially \( \langle N(R) \rangle = \rho v_1(R) \). In practice, we implement both definitions to compute \( \langle N(R) \rangle \). The scaled intersection volume \( \alpha_2(r; R) \) that appears in (2.14) can be interpreted as a monotonic repulsive pair potential energy with compact support, where \( R \) defines the interaction range. Thus, the point configurations that globally minimize \( \sigma_N^2(R) \) in a fixed dimension \( d \) correspond to the classical ground states associated with this repulsive pair potential [313]. Due to the integrability requirement of (2.14), the variance cannot increase faster than the square of window volume, \( [v_1(R)]^2 \) [346].

The hyperuniformity concept has been recently generalized to treat systems with directionally-dependent structure factors, so-called “directionally-hyperuniform” systems [299]. In this chapter, unless otherwise stated, hyperuniform systems refer solely to direction-independent hyperuniform point processes, defined by (2.1).

For spherical windows of radius \( R \), substituting (2.19) into (2.14), one can obtain [313]:

\[
\sigma_N^2(R) = \rho v_1(1) \left[ A_N(R) R^d + B_N(R) R^{d-1} + o(R^{d-1}) \right],
\]

(2.20)

where \( o(x^a) \) represents all terms of order less than \( x^a \), and coefficients \( A_N(R) \) and
\[ A_N(R) = 1 + \rho \int_{|r|<R} h(r) \, dr \]  
\[ B_N(R) = -\frac{\rho d \Gamma(d/2)}{2 \Gamma(1/2) \Gamma((1 + d)/2)} \int_{|r|<R} h(r) |r| \, dr , \]  

where \( B_N(R) \) is essentially the \( d \)-th moment of the total correlation function \( h(r) \).

In the limit of \( R \to \infty \), coefficients \( A_N \) and \( B_N \) are convergent for all periodic point patterns, a large class of quasicrystals, and some disordered point patterns whose total correlation functions \( h(r) \) decay to zero faster than \( 1/r^{d+1} \) [313, 346, 347].

Since \( \lim_{R \to \infty} A_N(R) = \lim_{|k| \to 0} S(k) \), a hyperuniform system has the vanishing scaled long-wavelength density fluctuations, i.e., \( \lim_{R \to \infty} \sigma_N^2(R) / v_1(R) = 0 \). When the structure factor goes to zero with power-law form \( S(k) \sim |k|^\alpha \) and \( 0 < \alpha \leq 1 \), the coefficient term \( B_N(R) \), defined by (2.22), asymptotically converges to a function of \( R \) and hence, [313, 346, 311]

\[ \sigma_N^2(R) \sim \begin{cases} R^{d-\alpha}, & 0 < \alpha < 1 \\ R^{d-1} \ln R, & \alpha = 1 \\ R^{d-1}, & \alpha > 1 \end{cases} \]  

(2.23)

For some systems, e.g., lattices, the associated variances oscillate around some global average behavior so that it can be difficult to obtain smooth asymptotic behaviors. In such cases, it is advantageous to use the cumulative moving average of the variance \( \overline{\sigma_N^2}(R) \) [313, 150], defined as

\[ \overline{\sigma_N^2}(R) \equiv \frac{1}{R} \int_0^R \sigma_N^2(x) \, dx , \]  

(2.24)

to observe the asymptotic behavior.
2.2.3 Basic quantities for a square window and square lattice

Consider a square lattice in \( \mathbb{R}^2 \) of unit lattice constant. Its total correlation function can be expressed as

\[
h(r) = \prod_{i=1}^{2} \left[ \sum_{n=-\infty}^{\infty} \delta(r_i - n) \right] - \delta(r) - 1, \tag{2.25}
\]

where \( r_i \) is the \( i \)-th component of position vector \( r \). Its isotropic form is

\[
h(r) = \sum_{k=1}^{\infty} \frac{Z_k}{2\pi r_k} \delta(r - r_k) - 1, \tag{2.26}
\]

where \( r_k \) is the radius of the \( k \)-th shell, and \( Z_k \) is the corresponding coordination number. Both expressions immediately follow from the definition of the square lattice and pair correlation function \( g_2(r) \), given in Section (2.2.1). The structure factor forms another square lattice, excluding the lattice point at the origin:

\[
S(k) = (2\pi)^2 \sum_{n \in \mathbb{Z}^2 \setminus 0} \delta(k - 2\pi n) = (2\pi)^2 \left( \prod_{i=1}^{2} \left[ \sum_{n=-\infty}^{\infty} \delta(k_i - 2\pi n) \right] - \delta(k) \right). \tag{2.27}
\]

The scaled intersection volume of two identical, aligned square windows of side length \( 2L \) \([349]\), whose centers are separated by \( r \), is given by

\[
\alpha_2(r; L) = \prod_{i=1}^{2} \left( 1 - \frac{|r_i|}{2L} \right) \Theta(2L - |r_i|), \tag{2.28}
\]

where \( \Theta(x) \) is the Heaviside step function, defined as

\[
\Theta(x) = \begin{cases} 
1, & x \geq 0 \\
0, & x < 0
\end{cases} \tag{2.29}
\]
The Fourier transform of $\alpha_2(r; L)$ is

$$\tilde{\alpha}_2(\mathbf{k}; L) = \left(\frac{2}{L}\right)^2 \prod_{i=1}^{2} \left[ \frac{\sin(k_i L)}{k_i} \right]^2.$$ (2.30)

We note in passing that $\tilde{\alpha}_2(\mathbf{k}; L)$ is the same as the intensity profile of Fraunhofer diffraction pattern through a square aperture of side length $2L$. From this analogy, we can know that “brightest” spots of (2.30) lie on principal axes on which either $k_x$ or $k_y$ is zero. On those principal axes, $\tilde{\alpha}_2(\mathbf{k}; L)$ is expressed as

$$\tilde{\alpha}_2((0, k_y); L) = 4 \left( \frac{\sin (k_y L)}{k_y} \right)^2,$$ (2.31)

and the magnitudes of peaks are proportional to $L^2$.

### 2.3 Variance for Square Lattice with Superdisk Windows

The asymptotic expression for $\sigma_N^2$ for non-circular windows has been intensively studied in $\mathbb{Z}^2$ (the square lattice) in various contexts, including the “lattice-point counting problem” in number theory [150, 151, 29, 132], discrete math [19, 20], and stereology [200]. It has been known that the asymptotic behavior of $\sigma_N^2$ can sensitively depend on the window shape [150, 152] as well as the orientation of the window [19, 250]. For instance, $\sigma_N^2(a) \sim a^{3/2}$ when the window is $\Omega = \{(x, y) \in \mathbb{R}^2 \mid (xa)^2 + y^4 \leq a^4\}$ [150], and $\sigma_N^2(W, H) \sim W^2 + H^2$ when the window is a $W \times H$ rectangle whose sides are parallel to the principal axes of the lattice [152]. All of these asymptotic behaviors are different from the linear growth rate in $R$, which one expects to use circular windows [313, 150].

In this section, to observe how the window shape can affect the growth rate of the
Figure 2.3: The variance $\sigma_N^2(L)$ for the square lattice using superdisk windows. (a) A plot of $\sigma_N^2(L)$ vs $L$ for the cases of $p = 1, 16$. Note that the variance is divided by the characteristic length $L$. The inset is a magnification of the larger panel to show the case of $p = 1$, which is otherwise not visible in the larger panel. (b) A semi-log plot of power exponent $\gamma$ of $\sigma_N^2(L) \sim L^\gamma (L \to \infty)$ vs the deformation parameter $p$. We compute the variances via the Monte Carlo method up to $L = 70$ and then estimate its power exponent by using linear regression of the log-log plot of $\sigma_N^2(L)$. The three inset figures show superdisks for certain deformation parameters.

Figure 2.3(a) demonstrates the variances for square lattice using two different superdisk windows with a fixed orientation. Superdisks are two-dimensional figures whose shapes are described by the equation

$$|x|^{2p} + |y|^{2p} = L^{2p},$$

(2.32)

where $L$ is called the characteristic length scale, and $p$, also known as deformation parameter, is a positive real number. Superdisks are ideal for our purpose to probe the effect of the window shape on the variance because one can generate a family of superdisks just by changing the parameter $p$. When $p = \infty$, a superdisk is just a square of side length $2L$. As $p$ decreases from $\infty$ to 1, superdisks smoothly interpolates between the square ($p = \infty$) to the circle ($p = 1$). When $p < 0.5$, the superdisk is concave and becomes a cross in the limit $p \to 0$. 

Figure 2.3(a) demonstrates the variances for square lattice using two different
superdisks: one is a circular window \((p = 1)\) and another is a virtually square window \((p \sim 56)\). As one can expect, \(\sigma_N^2(L) \sim L\) when \(p = 1\). Importantly, when the window shape is a perfect square \((p = \infty)\), one can obtain the closed expression for the variance by substituting (2.25) and (2.28) into (2.14):

\[
\sigma_N^2(L) = g(2L) \left(2(2L)^2 + g(2L)\right),
\]

(2.33)

where the function \(g(x)\) is defined as

\[
g(x) \equiv \{x\} (1 - \{x\}),
\]

(2.34)

and \(\{x\}\) represents the fractional part of a positive real number \(x\). The cumulative moving average of (2.33) is

\[
\bar{\sigma}_N^2(L) \approx \frac{4}{9} L^2 + O(L) \quad (L \to \infty).
\]

(2.35)

Thus, as the parameter \(p\) continuously increases from 1 to \(\infty\), the growth rate of the variance will vary from \(L\) to \(L^2\), i.e.,

\[
\bar{\sigma}_N^2(L) \sim L^\gamma \quad (L \to \infty),
\]

(2.36)

where \(\alpha\) increases from 1 to 2 (see Fig. 2.3(b)). This observation demonstrates that hyperuniform systems may exhibit anomalously large long-wavelength density fluctuations for aspherical windows, which is inconsistent with the “spherical-window” hyperuniformity condition (2.2).
2.4 Square-Window Variance for Periodic Point Configurations

In this section, we investigate the effect of the window orientations on the growth rate of the variance for the square lattice, $\mathbb{Z}^2$. We use square windows because this shape is amenable to exact analysis for some quantities and is one limit of superdisk ($p = \infty$).

For this purpose, we denote by $\sigma^2_N(L; \theta)$ the variance for $\mathbb{Z}^2$ using square windows that have side length $2L$ and is rotated counterclockwise by an angle $\theta$ with respect to the lattice. Here, it is sufficient to consider $\theta \in [0, \pi/4)$ due to 4-fold rotational symmetry and parity inversion symmetry of both the window and the lattice. We identify two classes of angles, rational and irrational angles, according to the asymptotic behavior of $\sigma^2_N(L; \theta)$. For rational angles, we derive the exact and asymptotic expressions for $\sigma^2_N(L; \theta)$, and then compare these expressions with the Monte Carlo calculations. For irrational angles, we compute the exact values for $\sigma^2_N(L; \theta)$ and conjecture its asymptotic behavior at a special set of irrational angles.

We obtain expressions of $\sigma^2_N(L; \theta)$ in both direct and Fourier space representations. Using (2.25) and (2.28), the direct-space formula (2.14) yields

$$
\sigma^2_N(L; \theta) = (2L)^2 \left[ 1 - 4L^2 + \sum_{\mathbf{n} \in \mathbb{Z}^2 \setminus 0} \alpha_2(R^T \mathbf{n}; L) \right],
$$

(2.37)

where $\alpha_2(r; L)$ is given in (2.28) and the rotation matrix $R$ is

$$
R = \begin{bmatrix}
\cos \theta & -\sin \theta \\
\sin \theta & \cos \theta
\end{bmatrix}.
$$

(2.38)
The summation in (2.37) can be written as

\[
\sum_{n \in \mathbb{Z}^d \setminus 0} \alpha_2(R^T n; L) = 4 \sum_{j=0}^{M(L, \theta)} (n_2 - n_1 + 1) \left[ 1 - \frac{(n_2 + n_1) \sin \left( \frac{\pi}{4} - \theta \right) + 2j \sin \left( \frac{\pi}{4} + \theta \right)}{2\sqrt{2}L} \right.
\]

\[
+ \left( j (n_1 + n_2) \frac{\cos (2\theta)}{8L^2} + \left( n_1 n_2 + \frac{1}{6} (n_2 - n_1) (2(n_2 - n_1) + 1) - j^2 \right) \frac{\sin (2\theta)}{8L^2} \right),
\]

(2.39)

where \( M(L, \theta) = \lfloor 2\sqrt{2}L \cos (\pi/4 - \theta) \rfloor \), \( n_1 \) and \( n_2 \) are abbreviations of \( n_1(j) \) and \( n_2(j) \), which are defined as

\[
n_1(j) = \max \{ \lfloor j \tan \theta \rfloor + 1, \lfloor \cot \theta (j - 2L \sec \theta) \rfloor \},
\]

(2.40)

\[
n_2(j) = \min \{ \lfloor j \cot \theta \rfloor, \lfloor \tan \theta (2L \csc \theta - j) \rfloor \},
\]

(2.41)

where \( \lfloor x \rfloor \) means the largest integer less than or equal to \( x \), and \( \lceil x \rceil \) is the smallest integer larger than or equal to \( x \).

The Fourier-space formula (2.15) yields

\[
\sigma_N^2(L; \theta) = \left( \frac{L}{\pi} \right)^d \int S(k) \tilde{\alpha}_2(k; L) \, dk
\]

\[
= \frac{1}{\pi^d} \sum_{n \in \mathbb{Z}^d \setminus 0} \left[ \prod_{i=1}^2 \sin^2 \left( 2\pi \frac{[R^T n]_i}{L} \right) \right],
\]

(2.43)

where \( L^* \) is a set of Bragg peaks of square lattice rotated by \(-\theta\) with respect to the window, \( R \) is the rotation matrix given by (2.38), and \([x]_i\) denotes the component of a vector \( x \) along the \( i \)-th principal axes of the square window.
2.4.1 Rational angles

An angle $\theta$ is called a rational angle if its tangent is a rational number, i.e.,

$$\tan \theta \in \mathbb{Q}. \quad (2.44)$$

In fact, the definition of rational angles can vary with the lattice and window (see 2.9). For a rational angle $\theta = \tan^{-1} n/m$, where $n$ and $m$ are coprime integers, we can express the positions of Bragg peaks $\mathcal{L}^*$ in (2.43) as those of the square lattice of lattice constant $L_0$ with $L_0$ basis points:

$$\mathcal{L}^* = \{(iL_0 + k\cos \theta, jL_0 + k\sin \theta) | (i, j) \in \mathbb{Z}^2, k = 1, 2, \ldots, L_0\} \setminus \mathbf{0}, \quad (2.45)$$

where $L_0 = \sqrt{n^2 + m^2}$. Substituting (2.45) into (2.43), we obtain the expression for the variance

$$\sigma_N^2(L; \theta) = \sum_{k=1}^{L_0^2} \left[ \sum_{i=-\infty}^{\infty} \frac{\sin^2(2\pi(i + x^{(k)})x)}{\pi^2(i + x^{(k)})^2L_0^2} \right] \left[ \sum_{j=-\infty}^{\infty} \frac{\sin^2(2\pi(j + y^{(k)})x)}{\pi^2(j + y^{(k)})^2L_0^2} \right] - 16x^4 \frac{L_0^4}{L_0^4} \quad (2.46)$$

$$= \frac{g(2x)}{L_0^4} (8x^2 + g(2x)) + \frac{1}{L_0^4} \sum_{k=1}^{L_0^2-1} B(x, x^{(k)})B(x, y^{(k)}), \quad (2.47)$$

where $x = L_0L$, $x^{(k)} = \{nk/L_0^2\}$, $y^{(k)} = \{mk/L_0^2\}$, $\{x\}$ is the fractional part of a real number $x$, $g(x)$ is defined by (2.34), and

$$B(x, y) = \sum_{i=-\infty}^{\infty} \frac{\sin^2(2\pi(i + y)x)}{\pi^2(i + y)^2} \quad (2.48)$$

$$= \csc^2(\pi y) \left[ \sin^2(\pi y[2x]) + \sin(\pi y)\sin(\pi y(2[2x] + 1)) \right] \{2x\}. \quad (2.49)$$

Note that the closed-form expression for $B(x, y)$ in (2.49) is valid when $y \in \mathbb{Q}$, and this expression is equivalent to piecewise linear interpolation of $\csc^2(\pi y) \sin^2(2\pi xy)$.
Figure 2.4: A plot of $\sigma_N^2(L; \text{atan}(1/100))$ vs $L$. It shows a typical behavior of variance at rational angles. Top panel shows the comparison between $\bar{\sigma}_N^2$ and its asymptote (2.50). The bottom panel is a magnification of the yellow-boxed region in the top panel. It demonstrates that the exact result by (2.47) is consistent with the Monte Carlo calculations.

whose data points are at $x = i/2$ for every integer $i$. A proof of (2.49) is given in Sec. 2.11. At $\theta = 0$, (2.47) is recovered into (2.33).
Now, let us examine properties of $\sigma_N^2(L; \theta)$ at rational angles. At first, $\sigma_N^2(L; \theta)$ vanishes whenever side length of a square window is an integer multiple of $L_0$, i.e., $2L = nL_0$. This arises because under this condition, the lengths of the square along the principal axes of the lattice are integers, and thus the number of lattice points inside the window does not change while translating the window. Figure 2.4 clearly demonstrates that $\sigma_N^2(L; \atan 1/100)$ vanishes at every $L = n\sqrt{10001}/2 \approx 50n$ for an integer $n$. Secondly, $\sigma_N^2(L; \theta)$ grows like the window area, $L^2$. More precisely, using the fact that $B(x, y)$ is a periodic function of $x$, $\sigma_N^2(L; \theta)$ is computed as

$$\sigma_N^2(L; \theta) \approx \frac{4L^2}{9L_0^2} - \frac{2L}{3L_0^3} + \frac{16}{45} + \frac{1}{4} H(n, m) \sim \frac{4L^2}{9L_0^2} \quad (L \to \infty), \quad (2.50)$$

where $H(n, m)$ is the constant term of the summation in (2.47):

$$H(p, q) = \frac{1}{L_0^4} \sum_{k=1}^{L_0-1} \csc^2 \left( \pi x^{(k)} \right) \csc^2 \left( \pi y^{(k)} \right). \quad (2.51)$$
Figure 2.5 depicts $\sigma^2_N(L; \theta)$ computed via the Monte Carlo method at various rational angles, and they are in a good agreement with the corresponding asymptotic expression (2.50).

We note that relation (2.33) was presented in [152], and relation (2.47) was derived by Rosen [250] for rectangular windows using a different approach. Their derivations, however, highly rely on the geometry of the lattice and the window, and thus they are difficult to generalize to other lattices and other window shapes. On the other hand, it is straightforward to generalize the formula (2.42) to other lattices and other window shapes; see 2.8. Furthermore, relation (2.46) provides a useful insight to explain the origin of the anomalously large density fluctuations; see Fig. 2.8(a).

Note that asymptotic result (2.50) is inconsistent with the spherical-window condition (2.2), which may lead one to falsely conclude that the square lattice is non-hyperuniform. Similar phenomena also have been observed in models of two-phase heterogeneous media, e.g., the checkerboard pattern and the square lattice decorated by identical squares [349, 239]. Specifically, even though these periodic heterogeneous media are hyperuniform by the Fourier-space condition (2.3), the resulting volume-fraction variance decays in an anomalous fashion, i.e., $\lim_{L \to \infty} (2L)^2 \sigma_v^2(L) \neq 0$. Such anomalously large density fluctuations for hyperuniform systems were not predicted or noticed in previous theoretical works concerning hyperuniformity.

How do such anomalously large density fluctuations arise in what are hyperuniform systems? We can provide two answers to this question: the first is geometrically based, and the second is analytically based. For the sake of simplicity, we will assume $\theta = 0$. Generally speaking, anomalously large density fluctuations arise when density fluctuations on the boundary of a window are correlated. Specifically, for a square window, a single line of lattice points near the boundary of the window can fall alternately in and out of the window as the window moves around the lattice with a fixed orientation [see Fig. 2.6(a) and (b)]. Thus, the resulting number variance is
Figure 2.6: (color online) Correlation of density fluctuations for $\mathbb{Z}^2$ lattice points on the perimeter of the square window. (a) $\theta = 0$ and $2L = 8.5$, (b) $\tan \theta = 3/4$ and $2L = 40.5/ L_\theta(n, m)$, and (c) $\tan \theta = 1/\sqrt{2}$ and $L = 7.5$. When the leftmost window moves toward the right upper side, the blue points fall in the window, but the red points fall out of it. As shown in (a) and (b), at rational angles, lines of blue points can fall in the window at the same time. However, such a correlation of the fluctuations does not happen in the case of irrational angles.

proportional to the square of the window perimeter in large-$L$ limit, i.e.,

$$\frac{\sigma_N^2(L; 0)}{\sigma_N^2(a; 0)} \sim \left( \frac{L}{a} \right)^2 (L \to \infty). \quad (2.52)$$

Roughly speaking, if the window surface (perimeter if $d = 2$) has higher curvature on average or is closer to the spherical (circular) shape, then density fluctuations on the window surface are less correlated so that the growth rate of the variance becomes slower, as shown in Fig. 2.3(b). Essentially, such correlations of density fluctuations on the window surface can be demonstrated in the form of “resonance” between $\tilde{\alpha}_2(k; L)$ and $S(k)$ in the Fourier space, as shown in Fig. 2.8 (a). For the same reason, in $d$-dimensional space, the variance for the hypercubic lattice $\mathbb{Z}^d$ via a hypercubic window of side length $2L$ asymptotically grow like square of the window surface area in the large-$L$ limit (see 2.9):

$$\overline{\sigma_N^2(L; 0)} \approx \frac{d}{6(2d-1)}(2L)^{2(d-1)}. \quad (2.53)$$
Here, the coefficient $d$ comes from the number of faces of a $d$-dimensional hypercube.

Another way to explain anomalously large density fluctuations involves noting the conditional convergence of the second moment of total correlation function, $\int |x| h(r) \, dr$ (it becomes $d$-th moment in $d$-dimensional space). Using the analysis in (2.20) which was done by Torquato et al.[313], one can asymptotically expand $\alpha_2(r; L)$ in (2.37) in terms of $L$:

$$\sigma_N^2(L; 0) \approx (2L)^2 \left[ A_{\text{square}}(L) + \frac{B_{\text{square}}(L)}{L} \right], \quad (2.54)$$

where

$$A_{\text{square}}(L) = 1 + \int_{|x|,|y|<2L} h(r) \, dr, \quad (2.55)$$

$$B_{\text{square}}(L) = -\int_{|x|,|y|<2L} |x| h(r) \, dr. \quad (2.56)$$

Note that the integrand $|x| h(r)$ in (2.56) is different from that in (2.22), $r h(r)$. The area integral in (2.55) becomes an infinite sum, and its Abelian sum converges to $-1$, i.e.,

$$\int_{\mathbb{R}^2} h(r) \, dr = \lim_{\beta \to 0^+} \left[ \sum_{k=1}^{\infty} Z_k e^{-\beta r_k^2} - \frac{\pi}{\beta} \right] = -1, \quad (2.57)$$

where $Z_k$ stands for the coordination number of $k$-th shell of the square lattice, and $r_k$ is the radius of $k$-th shell. Thus, $A_{\text{square}}(L)$ converges to 0 as $L$ tends to infinity in the sense of Abelian mean. On the other hand, the second moment of the total correlation function, given by (2.56), does not converge even in Abelian sum, but asymptotic behavior of its Abelian sum is $\beta^{-0.5}$ (see Fig. 2.15). This implies that $B_{\text{square}}(L) \sim L$ by the dimensional analysis of (2.115). On the other hand, the Abelian sum of the counterpart of (2.56) for circular windows of radius $R$ converges in the large-$R$ limit [313]. More details are provided in 2.10.
2.4.2 Irrational angles

For a square window and the square lattice, we define an irrational angle \( \theta \) to be one that satisfies the condition \( \tan \theta \in \mathbb{R} \setminus \mathbb{Q} \), where \( \mathbb{R} \setminus \mathbb{Q} \) stands for the set of all irrational numbers. Estimating the local number variance at irrational angles is intimately related to the concept of the Diophantine approximation in number theory and discrepancy theory in discrete mathematics [20, 19]. For a given window shape (usually a rectangular box of arbitrary aspect ratios) and a finite point configuration in the unit hypercube in \( \mathbb{R}^d \), the discrepancy is the largest difference between the window volume and the number points within the window. Generating configurations of \( N \) points with the lowest discrepancy is a problem of central concern.

Currently, a general theory to analytically deal with number variances for square or rectangular windows at all irrational angles has yet to be developed. Thus, previous works have been mainly restricted to certain types of irrational angles whose slopes have bounded partial quotients, so-called “badly approximable numbers” (see Appendix 2.12), e.g., Fibonacci lattice [25]. Beck [19] studied the variance for the square lattice with rectangular strips that have a fixed width and is tilted by an irrational angle whose slope belongs to the badly approximable numbers. Interestingly, one can generate a large class of one-dimensional quasicrystals by projecting the square lattice points within an infinitely long rectangular strip tilted by an irrational angle onto the long axis of the strip [71, 275]. Thus, the variance for the square lattice via a long rectangular strip tilted by some irrational angles is closely related to the variance for one-dimensional quasicrystals.
Figure 2.7: Semi-log plot of $\sigma_{N}^{2}(L; \tan^{-1}(1/\sqrt{2}))$ vs $L$. Top panel shows the exact values of $\sigma_{N}^{2}$ from (2.37), and the cumulative moving average $\overline{\sigma_{N}^{2}}(L)$, which was computed by the trapezoidal rule. The red dashed line is a curve fit of the natural logarithm function. The bottom panel is a magnification of a part of the top panel. Note that the bottom panel shows good agreement between the Monte Carlo calculations (red squares) and exact calculations (black line).

Figure 2.8: Visualization of the superposition of two functions $S(k)$ and $\tilde{\alpha}_{2}(k; L)$ that appear in the Fourier-space representation of $\sigma_{N}^{2}(L; \theta)$ as given by (2.42), when $L = 1$. $x$ and $y$ axes of both panels are $k_{x}/2\pi$ and $k_{y}/2\pi$, respectively. The left and right panels show the cases of rational and irrational angles, respectively. The white dots represent the Bragg peaks that are located on the sites of a square lattice, i.e., the structure factor $S(k)$. The function $\tilde{\alpha}_{2}(k; L)$, given by the formula (2.30), is depicted as a contour plot in log scale for vivid visualization purposes. The variance via (2.43) is equivalent to the summation of $\tilde{\alpha}_{2}(k; L)$ at each Bragg peak.
We use (2.37) to compute \( \sigma_N^2(L; \theta) \) at irrational angles. Comparing Fig. 2.4 with Fig. 2.7, one can see that the variance at irrational angles generally has a much smaller magnitude than the variance at rational angles. Furthermore, \( \sigma_N^2(L; \atan 1/\sqrt{2}) \) exhibits a logarithmic asymptotic behavior, which is similar to that for rectangular strips at certain types of irrational angles, i.e., quadratic irrational numbers [19]. This asymptotic behavior of the variance is anomalously small in the sense that it is slower than the window perimeter, \( L \).

Figure 2.9: (color online) Cumulative moving average \( \sigma_N^2(L) \), vs log-scaled \( L \) at various irrational angles. Exact results are computed by (2.37). The top panel shows the cases in which the slopes \( \tan(\theta) \) are badly approximable numbers. The bottom panel shows the cases for \( \tan(\theta) \) whose partial quotients are unbounded.

Fourier space also provides a clear way to understand the anomalously small density fluctuations for irrational angles. Figure 2.8 illustrates two different terms, \( S(k) \) and \( \tilde{\alpha}_2(k; L) \), in the Fourier representation (2.15) of \( \sigma_N^2 \). For simplicity, we choose \( L = 1 \). If \( L \) increases (not shown in the figure), the peaks of \( \tilde{\alpha}_2(k; L) \) that lie along the principal axes become narrower in their widths, and larger in their intensities. As can be seen in Fig. 2.8 (a), at rational angles, some Bragg peaks always lie along the
principal axes of $\tilde{\alpha}_2(k; L = 1)$. Then, at certain values of $L$, the peaks of $\tilde{\alpha}_2(k; L)$ on the principal axes are coincident with those Bragg peaks, resulting in $L^2$ growth of $\sigma_N^2$. At irrational angles, on the other hand, there are no Bragg peaks on the principal axes of $\tilde{\alpha}_2(k; L)$, as shown in Fig. 2.8 (b). Instead, the major contribution to the variance comes from Bragg peaks, which are close to the principal axes of $\tilde{\alpha}_2(k; L)$. For those Bragg peaks, corresponding indices $(i, j)$ are the denominator and the numerator $(n_k, m_k)$ of convergents of $\tan(\theta)$ (see Sec. 2.12). For this reason, it is expected that the asymptotic behavior of $\sigma_N^2(L; \theta)$ for irrational angles largely depends on the distribution of partial quotients $a_k$, which are mainly concerned in the Diophantine approximation in number theory. Furthermore, the variance at irrational angles is unusually small.

In terms of rational approximation, irrational numbers can be classified into two sets. One is called “badly approximable numbers”, including $\sqrt{n}$ and the golden ratio $\phi$. Roughly speaking, a badly approximable number $x$ cannot have an excellent rational representation in the sense that any rational approximation $n/m$ of $x$ approaches to $x$ at most in the order of $1/m^2$. Thus, if $\tan(\theta)$ belongs to this set, then the Bragg peaks of the square lattice cannot be closer to the principal axes of $\tilde{\alpha}_2(k; L)$ than a certain amount, leading to a smaller variance compared to those at irrational angles, which do not belong to badly approximable numbers. The top panel in Fig. 2.9 shows the cumulative moving averages of the variance at some badly approximable slopes. Note that $\sigma_N^2(L; \tan(\phi))$ is smallest, where $\phi$ is the golden ratio, an extreme example of a badly approximable number. The logarithmic growth rate of $\sigma_N^2(L)$ was also predicted in the case of rectangular strips at angles whose slopes are quadratic irrational numbers [19], which are also “badly approximable numbers”. The bottom panel in Fig. 2.9 shows the asymptotic behaviors of $\sigma_N^2(L; \theta)$ at some irrational slopes which are not badly approximable numbers. For these angles, their asymptotic behaviors are rather unclear.
Figure 2.10: (color online) The variance for square windows with fixed orientations in various ranges of $L$. Window orientations are expressed in the form of slopes, $\tan(\theta)$. Here, $c_k$ stands for $k$-th convergent of the golden ratio $\phi$, defined in Appendix 2.12. $c_{37}$ and $c_{75}$ correspond to rational approximations of $\phi$ up to the double and quadruple-precisions, respectively; see table 2.2.

In addition, the difference in the asymptotic behaviors of $\sigma_N^2(L; \theta)$ at rational and irrational angles is one of many physical examples [11, 37, 124, 335, 52, 235] at which the (in)commensurability of a certain parameter plays an critical role in physical properties.

**Remarks**

1. There are two numerical issues in computing $\sigma_N^2(L; \theta)$ at irrational angles with (2.37). One is a huge round-off error. Relation (2.37) involves the subtraction of two terms on the order of $(2L)^4$ to obtain a value on the order of unity. Thus, a round-off error in $\sigma_N^2(L; \theta)$ for $L \gtrsim 2000$ is estimated as 10% even if we fully exploit double-precision. For this reason, we employ quadruple-precision to compute $\sigma_N^2(L; \theta)$.

2. Another numerical issue is the inevitable rational approximation in an irrational angle while computing the variance at the irrational angle. This implies that
the numerically computed variances inevitably grow like $L^2$. Then, it is a natural question to ask: To what extent are the numerical calculations of (2.37) sufficiently reliable to obtain the exact variance at irrational angles? A qualitative answer is that for two distinct angles, the difference in variance at these angles is negligible up to a certain window size $L_{\text{max}}(\Delta \tan \theta)$, which is certainly a decreasing function of $\Delta \tan \theta$, as shown in Fig. 2.10. This is because (2.37) is a finite summation of continuous functions of $\theta$ within a range between $n_1(i)$ and $n_2(i)$, given by (2.40) and (2.41), also determined by continuous functions of $\theta$.

### 2.5 Variance for Disordered Hyperuniform Point Processes

Throughout this section, we solely consider a $d$-dimensional convex unit window $\omega$ of a general shape in $\mathbb{R}^d$ and its scalar multiplication $a\omega$, where $a$ is a positive real number. Here, the largest distance from the centroid of $\omega$ to the boundary is the unity: $\max_{x \in \omega} |x| = 1$. The window indicator function can be written as

$$w(x; R) = w(x; a, \omega) = \begin{cases} 1, & x/a \in \omega \\ 0, & \text{otherwise} \end{cases}$$

For brevity, we abbreviate the parameter set $R$, which characterizes the window shape and orientation, to a single length-scale parameter $a$, e.g., $\sigma_N^2(R)$ to $\sigma_N^2(a)$, $v_1(R)$ to $v_1(a)$. Then, we obtain a general expression for the asymptotic behavior of $\sigma_N^2(a)$ for disordered hyperuniform point processes. In what follows, we prove that for any convex window, the variance for a disordered hyperuniform point process has the common scaling relation, which is identical to (2.23). Then, we present some
example calculations for two isotropic disordered hyperuniform systems.

### 2.5.1 Analysis

Consider statistically homogeneous and isotropic hyperuniform point processes at number density $\rho$ in $\mathbb{R}^d$. Then, the vector-dependent total correlation function becomes a radial function $h(r)$, where $r = |r|$. Taking advantage of the rotational symmetry, we can rewrite (2.14) for a general window shape, using the orientationally-averaged scaled intersection volume function $\langle \alpha_2(|r|; a) \rangle_O$:

$$
\sigma_N^2(a) = \rho v_1(a) \left[ 1 + \rho \int_{\mathbb{R}^d} h(r) \langle \alpha_2(r; a) \rangle_O \, dr \right], \quad (2.59)
$$

where $\langle \alpha_2(r; a) \rangle_O$ is the average of $\alpha_2(r; a)$ over all possible orientations of $r$ with fixed $|r|$.

To compute the large-$a$ asymptotic behavior of $\sigma_N^2(a)$, we need to find an expression for $\overline{\alpha}(|r|; a)$ for a window $a\omega$. Generally, it is extremely difficult to find a closed expression for $\alpha_2(r; a)$ of an arbitrarily shaped window. For small displacements $|r| \ll a$, however, one can approximate it up to the first order in $r$. Since $\alpha_2(r; a)$ is not differentiable at $r = 0$, we cannot apply the multivariable Taylor theorem to $\alpha_2(r; a)$ immediately. Instead, we apply the Taylor theorem to $\alpha_2(r; a)$ around $r = \epsilon \hat{r}$ and obtain a one-sided limit of the expansion as $\epsilon \to 0^+$ in the following way:

$$
v_2^{\text{int}}(r) = \lim_{\epsilon \to 0^+} \left[ v_2^{\text{int}}(\epsilon \hat{r}) + \sum_{\alpha=1}^d \frac{\partial v_2^{\text{int}}(r')}{\partial r'_\alpha} \right|_{r' = \epsilon \hat{r}} \left( 1 - \frac{\epsilon}{|r|} \right)^\alpha r_\alpha + \ldots \right] = v_1(a) - A_\perp(\hat{r}; a) |r| + O\left(|r|^2\right), \quad (2.60)
$$

where $v_2^{\text{int}}(r)$ is an abbreviation for $v_2^{\text{int}}(r; a)$, defined in (2.16), and $\hat{r}$ is the unit vector.
of \( \mathbf{r} \). Here, the first order coefficient \( A_\perp (\hat{\mathbf{r}}; a) \) is defined as

\[
A_\perp (\hat{\mathbf{r}}; a) \equiv \lim_{\epsilon \to 0^+} \oint_{\partial (a \omega)} w(\mathbf{x} - \epsilon \hat{\mathbf{r}}; a) \, d\mathbf{s} \cdot \hat{\mathbf{r}},
\]

(2.61)

where \( d\mathbf{s} \) represents the infinitesimal surface area element whose direction is normal to the surface and \( \partial (a \omega) \) stands for the boundary of the window \( a \omega \). Geometrically, \( A_\perp (\hat{\mathbf{r}}; a) \) is the projected area of the window on a hyperplane normal to the displacement vector \( \mathbf{r} \). The second-order Taylor coefficient of (2.60) is written as

\[
\lim_{\epsilon \to 0^+} \int_{\partial (a \omega) \cap (\partial (a \omega) - \epsilon \hat{\mathbf{r}})} \mathbf{x} - \epsilon \hat{\mathbf{r}} \, d\mathbf{s}.
\]

(2.62)

Note that since \( d\mathbf{s} \) in (2.62) is normal to \( \mathbf{r} \), the second-order term in (2.60) is identically zero. Then,

\[
\alpha_2 (\mathbf{r}; a) \equiv \frac{v_2^\text{int} (\mathbf{r}; a)}{v_1 (a)} \approx 1 - \frac{A_\perp (\hat{\mathbf{r}}; a)}{v_1 (a)} |\mathbf{r}|.
\]

(2.63)

Using the well-known average-projected-area theorem for convex bodies (see [273]), we obtain the expression for the orientational-average of the scaled intersection volume:

\[
\langle \alpha_2 (\mathbf{r}; a) \rangle_\mathcal{O} \approx 1 - \frac{\langle A_\perp (\hat{\mathbf{r}}; a) \rangle_\mathcal{O}}{v_1 (a)} |\mathbf{r}| = 1 - \kappa (d) \frac{s_1 (1)}{v_1 (1)} \frac{|\mathbf{r}|}{a},
\]

(2.64)

where \( r = |\mathbf{r}| \), \( s_1 (a) \) is the surface area of a window \( a \omega \) and \( \langle A_\perp (\hat{\mathbf{r}}; a) \rangle_\mathcal{O} \) stands for the orientational-average of \( A_\perp (\hat{\mathbf{r}}; a) \):

\[
\langle A_\perp (\hat{\mathbf{r}}; a) \rangle_\mathcal{O} = \frac{1}{\Omega_d} \oint A_\perp (\hat{\mathbf{r}}; a) \, d\hat{\mathbf{r}},
\]

(2.65)

where \( \Omega_d = \frac{2 \pi^{d/2}}{\Gamma (d/2)} \) stands for the surface area of the \( d \)-dimensional unit sphere. Here, \( \kappa (d) \) is the constant depending only on spatial dimension \( d \) [273]:

\[
\kappa (d) = \frac{\Gamma (d/2)}{2 \Gamma (1/2) \Gamma ((1 + d)/2)}.
\]

(2.66)
Using approximation (2.64) and the analogous analysis in (2.20), we obtain

\[
\frac{\sigma_N^2(a)}{s_1(a)} \approx -\rho^2 \kappa(d) \int_{|r| < a} h(r) |r| \, dr \quad (a \to \infty).
\] (2.67)

Note that the right-hand side of (2.67) is independent of the window shape, thus implying that the asymptotic behavior of \( \sigma_N^2(a) \) is independent of the window shape.

### 2.5.2 Example calculations

![Graphs of \( \frac{\sigma_N^2(a)}{s_1(a)} \) for circular and square windows](image)

Figure 2.11: The scaled variance \( \frac{\sigma_N^2(a)}{s_1(a)} \) for 2D disordered hyperuniform point processes for both circular and square windows as a function of \( a \). Here, \( a = R \) and \( a = L \) for circular and square windows, respectively, and \( s_1(a) \) denotes the window perimeter. (a) \( g_2 \)-step-function point process at \( \rho = 1 \) and \( D = 1/\sqrt{\pi} \) and (b) one-component plasma at \( \rho = 1 \) are presented. We can see that \( \sigma_N^2(R)/s_1 \) converges to the same value as the perimeter of a window increases for each case, as shown by (2.67).

To demonstrate the implications of (2.67), we study two disorderd hyperuniform point processes in \( \mathbb{R}^2 \): one is one-component plasma (OCP) [313, 176] and the other is two-dimensional \( g_2 \)-step-function point process [313]. OCP is a system consisting of point particles of charge \( e \) and uniform background charge satisfying overall charge neutrality. In the thermodynamic limit, when the coupling constant is \( \Gamma \equiv e^2/(kT) = \ldots \)
2, the total correlation function is given by [135]

\[ h(r) = -e^{-\rho \pi r^2}, \]  
(2.68)

and its associated structure factor [313] is \( S(k) \sim k^2 \ (k \to 0) \). We also consider a \( g_2 \)-invariant point process defined by the following pair correlation function

\[ g_2(r) = \Theta(r - D), \]  
(2.69)

where \( D \) is diameter of hard spheres. A \( g_2 \)-invariant process is one in which a chosen non-negative \( g_2(r) \) function remains invariant over a non-vanishing density range without changing all other macroscopic variables [278, 257]. This system, so called \( g_2 \)-step-function point process, turns out to be hyperuniform at the “terminal” density \( \rho = (\pi D^2)^{-1} \) [313].

Since the exact expressions for \( g_2(r) \) for both aforementioned point processes are given, one can compute their variance for both square and circular windows. Figure 2.11 clearly shows that both OCP and hyperuniform \( g_2 \)-step-function point process at the unit density have common scaling relations for circular and square windows. It is a noteworthy fact that spherical windows measure the minimal asymptotic variance among all convex windows of the same volume. This is because the variance is proportional to the window surface area due to (2.67) and spheres have the smallest surface-to-volume ratio among convex bodies (see Isoperimetric problems).

### 2.6 Orientationally-Averaged Number Variance

In the previous section, we proved that for statistically homogeneous and isotropic point processes, the asymptotic behavior of the scaled variance \( \sigma_N^2(a) / s_1(a) \) is independent of the window shape (see (2.67)), if windows are convex. On the other
hand, for anisotropic hyperuniform systems, including disordered ones and lattices, the growth rate of variance depends on both the shape and the orientation of windows, as shown in Sec. 2.3 and Sec. 2.4. Thus, statistical isotropy plays an important role in making such a difference in asymptotic behavior. In this section, we define orientationally-averaged local number variance and study its asymptotic behavior.

Consider statistically homogeneous but anisotropic hyperuniform point processes in $\mathbb{R}^d$. Then, we can re-interpret the variance formula (2.59) for aspherical windows as the orientationally-averaged one. This is because there are implicitly two different orientations in the scaled intersection volume $\alpha_2(r; a)$ of a window $a\omega$: one is the orientation of the displacement vector $r$ and the other is that of windows. Thus, $\langle \alpha_2(r; a) \rangle_O$ also implies the average of $\alpha_2(r; a)$ over the orientations of windows with the displacement vector $r$ fixed. For the purposes of illustration, consider the explicit expression (2.70) for square windows of side length $2L$ and its scaled intersection volume $\alpha_2(r; L)$. The rightmost side of the first line of (2.70) schematically represents the definition of $\langle \alpha_2(r; a) \rangle_O$ as the average of $\alpha_2(r; L)$ (shaded regions) over all orientations of $r$ (red arrows) with the orientations of windows fixed. Rotating pairs of windows in the first line in the manner that the orientations of displacement vectors (red arrows) are identical, as shown in the second line of (2.70), yields the average of $\alpha_2(r; L)$ over all orientations of square windows with a common displacement vector.
\[ \langle \sigma_N^2(a) \rangle_O \equiv \frac{1}{2\pi} \int_{|r|=r} \alpha_2(r; a) \, dr = \frac{1}{2\pi} \left[ \int \alpha(r; L) \, d\theta = \frac{1}{2\pi} \left[ \int \alpha(r; L) \, d\theta \right] \right] \]

\[ = \frac{1}{2\pi} \int_{\text{window orientations}} \alpha(r; L) \, d\theta = \frac{1}{2\pi} \left[ \int \alpha(r; L) \, d\theta \right] \]

\[ = \begin{cases} 
1 - \frac{4}{\pi}x + \frac{1}{\pi}x^2, & x < 1 \\
1 - \frac{2+2x^2}{\pi} + \frac{4}{\pi} \left( \sqrt{x^2 - 1} - \cos^{-1} \left( \frac{1}{x} \right) \right), & 1 \leq x < \sqrt{2} \\
0, & x > \sqrt{2} 
\end{cases} \]  

(2.70)

where \( x = \frac{r}{2L} \) [296]. Note that the argument made in (2.70) is valid for any \( d \)-dimensional aspherical windows. Thus, for anisotropic point processes, the expression

\[ \langle \sigma_N^2(a) \rangle_O \equiv \rho v_1(a) \left[ 1 + \int_{\mathbb{R}^d} h(r) \langle \alpha_2(r; a) \rangle_O \, dr \right] \]  

(2.71)

represents the orientationally-averaged variance for windows \( a\omega \). Since (2.71) is the same as (2.59), one can conclude that any hyperuniform point process will satisfy the following relation

\[ \frac{\langle \sigma_N^2(a) \rangle_O}{s_1(a)} \approx -\rho^2 \kappa(d) \int_{|r|<a} h(r) \, |r| \, dr \quad (a \to \infty). \]  

(2.72)

This implies that orientationally-averaged variance \( \langle \sigma_N^2(a) \rangle_O \) of any hyperuniform point process will give the same scaling relation (2.23) for any convex window shape. Therefore, we conclude that for any convex window shape, the following hyperuniformity conditions applies:

\[ \lim_{a \to \infty} \frac{\langle \sigma_N^2(a) \rangle_O}{v_1(a)} = 0, \]  

(2.73)

which is consistent with the spherical-window condition (2.2).

Now, we consider the orientationally-averaged variance, \( \langle \sigma_N^2(L) \rangle_O \) for the square
lattice using square windows. In this case, (2.72) implies that

$$\lim_{L \to \infty} \frac{\langle \sigma_N^2(L) \rangle}{8L} = \lim_{R \to \infty} \frac{\overline{\sigma_N^2}(R)}{2\pi R}.$$  \hspace{1cm} (2.74)

Kendall [150] proved the same result for square lattice and randomly oriented planar convex windows with non-vanishing curvatures, and derived the following:

$$\lim_{R \to \infty} \frac{\overline{\sigma_N^2}(R)}{2\pi R} = \frac{1}{2\pi^3} \zeta\left(\frac{3}{2}\right) L\left(\frac{3}{2}, \chi\right) \approx 3.6419 \times 10^{-2},$$  \hspace{1cm} (2.75)

where $\zeta(x)$ is the Riemann zeta function and $L(x, \chi)$ is the Dirichlet $L$-function, which is defined as $L(x, \chi) \equiv \sum_{n=0}^{\infty} (-1)^n / (2n + 1)^x$. Figure 2.12 clearly shows that $\langle \sigma_N^2(L) \rangle \sim L$, which also is consistent with Matérn’s observation [200], and from Fig. 2.12 we obtain

$$\lim_{L \to \infty} \frac{\langle \sigma_N^2(L) \rangle}{8L} = (3.642 \pm 0.0001) \times 10^{-2},$$  \hspace{1cm} (2.76)

which is in good agreement with (2.75).

This result gives a clue to the distribution of the asymptotic behaviors of $\sigma_N^2(L; \theta)$ with respect to the angle $\theta$. The orientationally-averaged variance is

$$\langle \sigma_N^2(L) \rangle = \frac{1}{\pi} \int_{-\pi/2}^{\pi/2} \sigma_N^2(L; \theta) \, d\theta.$$  \hspace{1cm} (2.77)

Here (2.77) may not be well-defined as a Riemann integral, because for a fixed $L$, the continuity of $\sigma_N^2(L; \theta)$ with respect to $\theta$ is unclear. Therefore, it is better to introduce the probabilistic integral to compute (2.77). Then, since the set of rational angles, $\tan^{-1}(Q)$, have zero measure among all angles, rational angles do not make
any contribution to the orientational-average of the variance, and thus

\[
\frac{1}{\pi} \int_{-\pi/2}^{\pi/2} \sigma_N^2(L; \theta) \, d\theta = \frac{1}{\pi} \int_{[-\pi/2, \pi/2] \setminus \tan^{-1}(\mathbb{Q})} \sigma_N^2(L; \theta) \, d\theta.
\]

In order for (2.78) to be consistent with the result (2.76), it is expected that \(L\)-growth rate for \(\sigma_N^2(L; \theta)\) should exist for \(\theta\) of non-zero measure subset of \([-\pi/2, \pi/2] \setminus \tan^{-1}(\mathbb{Q})\). However, we have yet to observe linear growth rate and so it is an interesting problem in mathematics to identify an irrational angle at which the variance for the square windows asymptotically grows like the window perimeter.
2.7 Conclusions and Discussion

We have studied the window-shape dependence on the large-window asymptotic behavior of the local number variance of hyperuniform point processes to understand conditions under which the growth rate of the variance is not slower than the window volume, conflicting with the spherical-window hyperuniformity condition (2.2). For this purpose, we compute the variance for several hyperuniform systems using aspherical windows with a fixed orientation with respect to the systems.

We demonstrate that for hyperuniform systems, the growth rate of the variance can depend on not only the window shape but also the window orientation. We begin with the numerical computation of the variance for the square lattice with superdisk windows, and demonstrate that its asymptotic behavior varies with the window shape, i.e., the deformation parameter $p$. Importantly, as the window shape is closer to perfect squares ($p = \infty$), the asymptotic behavior of the variance approaches to $L^2$ (from $L$), which is inconsistent with “spherical-window” condition (2.2).

Then, to better understand the conditions under which hyperuniform systems can have anomalously large variance growth in conflict with the spherical-window condition (2.2), we investigate the case of the square lattice and square windows (superdisk of $p = \infty$). We identify two classes of angles of the square window with respect to the lattice, at which the asymptotic behavior is different. At the rational angles, defined by (2.44), the variance for square lattice increases like the window volume. However, at the irrational angles, the variance is significantly smaller than the variance via the spherical windows.

Based on the analysis for the square window and square lattice, we explain the origin of the inconsistency in the direct-space hyperuniformity conditions for spherical and aspherical windows in two aspects. One is the resonance between the structure factor $S(k)$ and the Fourier transform of the window $\tilde{\alpha}_2(k; L)$ in the Fourier space (see Fig. 2.8). For the square lattice and square windows, “rational angles” are the
angles at which the resonance occurs to cause the anomalously large variance growth. Subsequently, we extend the concept of rational angles to the case of $d$-dimensional Bravais lattice and parallelepiped windows (see 2.8). We explicitly compute rational angles corresponding to square lattice and rectangular windows with a fixed aspect ratio, and the case of triangular lattice and square windows. Another explanation is the conditional convergence of the second moment of the total correlation function, denoted by $\lim_{R \to \infty} B_N(R)$ in (2.22). Using Abelian summability method (2.10), we demonstrate that the improper integral that involves $\lim_{R \to \infty} B_N(R)$ is divergent for square boundaries while it is convergent for the circular one.

We proved that for statistically isotropic disordered hyperuniform systems, the variance associated with aspherical convex windows exhibits the same asymptotic behavior as the variance for spherical windows. We verified this result for two isotropic disordered hyperuniform point processes, i.e., one-component plasma and $g_2$-step-function point process at the critical density.

We also suggest a new direct-space hyperuniformity condition that is independent of the window shape, i.e.,

$$\lim_{a \to \infty} \frac{\langle \sigma_N^2(a) \rangle_O}{v_1(a)} = 0,$$

(2.79)

where $\langle \sigma_N^2(a) \rangle_O$ represents the local number variance averaged over window orientations. This is consistent with the fact that for a planar convex window, $\langle \sigma_N^2(L) \rangle_O$ of the square lattice is asymptotically bounded by the perimeter of the window [150, 29, 132]. We note that the same analysis and general conclusions directly extend to two-phase media because the formulas for $\sigma_N^2(R)$ and $\sigma_V^2(R)$ are essentially the same.

We have studied how to reduce the dependence of the window shape on the variance at large length scales. For future study, it will be interesting to investigate how to design the window shape and its orientation to maximize or minimize the variance for a given system at short length scales. Minimizing the variance corresponds to
finding the ground state of the repulsive pair potential defined by the scaled window intersection volume $\alpha_2(r;R)$ [313]. The implication of this problem may be used in the field of self-assembly. For instance, in the presence of depletants, the contact attraction is exerted between two cubic nano-shells due to the osmotic pressure [251]. Here, the attractive pair potential is proportional to the scaled intersection volume $\alpha_2(r;L')$, given by (2.104) in $d = 3$, where $L' = L + R_g$, $2L$ is the side length of the cubic particle, and $R_g$ is the gyration radius of a depletant.

### 2.8 Appendix A: Generalizations of rational angles to other Bravais lattices and parallelepiped windows

In the case of the square lattice and square windows, we identify rational angles (Sec. 2.4.1) at which the growth rate of the variance is not slower than the window volume. The concept of rational angles (orientations in higher dimensions) can be extended to general Bravais lattices and parallelepiped windows in $d$-dimension. For this purpose, we will derive a Fourier space representation of the variance for Bravais lattices using parallelogram observation windows in two-dimensions, and then generalize the expression to higher dimensions. Denote by $\Omega(L)$ a parallelogram window with a single length scale $L$, which is defined as

$$
\Omega(L) \equiv \left\{ \mathbf{x} = \sum_{i=1}^{2} y_i \mathbf{A}_i \in \mathbb{R}^2 \left| |y_i| < L, \text{ for } i = 1, 2 \right. \right\},
$$

(2.80)
where $\mathbf{A}_1$ and $\mathbf{A}_2$ are linearly independent vectors in $\mathbb{R}^2$. The window indicator function $w(\mathbf{r}; L)$ of this parallelogram window is given by

$$w(\mathbf{r}; L) \equiv \begin{cases} 1, & \mathbf{r} \in \Omega(L) = \prod_{i=1}^{2} \Theta(L - |y_i|) = w_0(M\mathbf{r}; L), \\ 0, & \text{otherwise} \end{cases}$$

(2.81)

where $w_0(\mathbf{r}, L)$ is the window indicator function of a two-dimensional square window that has side length $2L$ and is centered at the origin, and $M$ is a linear operator that transforms the unit parallelogram $\Omega(1)$ into the unit square. In a matrix representation,

$$M = \begin{bmatrix} \mathbf{B}_1^T \\ \mathbf{B}_2^T \end{bmatrix},$$

(2.82)

satisfying $\mathbf{B}_i \cdot \mathbf{A}_j = \mathbf{B}_i^T \mathbf{A}_j = \delta_{ij}$ for $i, j = 1, 2$, where $\delta_{ij}$ is the Kronecker delta symbol. The Fourier transform of the indicator function $w(\mathbf{r}, L)$ can be written as:

$$\hat{w}(\mathbf{k}; L) \equiv \int_{\mathbb{R}^2} d\mathbf{r} e^{i\mathbf{k} \cdot \mathbf{r}} w(\mathbf{r}; L) = \int_{\mathbb{R}^2} d\mathbf{r} \exp \left( i\mathbf{k} \cdot \sum_{i=1}^{2} y_i \mathbf{A}_i \right) \prod_{i=1}^{2} \Theta(L - |y_i|)
$$

$$= \left| \frac{\partial (r_1, r_2)}{\partial (y_1, y_2)} \right| \int_{\mathbb{R}^2} d\mathbf{y} e^{i\mathbf{q} \cdot \mathbf{y}} \prod_{i=1}^{2} \Theta(L - |y_i|),$$

(2.83)

where $\mathbf{r} \equiv M^{-1}\mathbf{y}$, and thus the Jacobian of the transformation from $\mathbf{r}$ to $\mathbf{y}$, $\left| \frac{\partial (r_1, r_2)}{\partial (y_1, y_2)} \right|$ is identical to the determinant of $M^{-1}$. Then, (2.83) becomes

$$\hat{w}(\mathbf{k}; L) = |\det(M^{-1})| \ \hat{w}_0(\mathbf{q}; L) = |\det(M^{-1})| \ \hat{w}_0\left( (M^{-1})^T \mathbf{k}; L \right),$$

(2.84)

where $\hat{w}_0(\mathbf{q}; L)$ is the Fourier transform of the square window of side length $2L$, and $\mathbf{q} = (M^{-1})^T \mathbf{k}$. Using the convolution theorem, the Fourier transform of the scaled
intersection volume function, $\tilde{\alpha}_2(\mathbf{r}; L)$, can be expressed as

$$
\tilde{\alpha}_2(\mathbf{k}; L) = |\tilde{\omega}(\mathbf{k}; L)|^2 / |\Omega(L)| = |\det(M^{-1})| \left| \tilde{\omega}_0\left( (M^{-1})^T \mathbf{k}; L \right) \right|^2 / (2L)^2,
\quad (2.85)
$$

where $|\Omega(L)| = (2L)^2 |\det(M^{-1})|$. In terms of reciprocal vectors $\mathbf{b}_i$ of lattice vectors $\mathbf{a}_i$, the structure factor is

$$
S(\mathbf{k}) = (2\pi)^2 / v_c \sum_{\mathbf{n} = (n_1, n_2) \in \mathbb{Z}^2 \setminus \mathbf{0}} \delta(\mathbf{k} - 2\pi \mathbf{Bn}),
\quad (2.86)
$$

where $\mathbf{B} = \begin{bmatrix} \mathbf{b}_1 \\ \mathbf{b}_2 \end{bmatrix}$ in a matrix representation, and the volume of the fundamental (unit) cell $v_c$ is equal to the inverse of the number density, i.e., $v_c = |\det(\mathbf{B}^{-1})| = 1 / \rho$.

Substituting (2.86) and (2.85) into (2.15), the variance can be written as

$$
\sigma_N^2(L; \theta) = \frac{\rho|\Omega(L)|}{(2\pi)^2} \int_{\mathbb{R}^2} d\mathbf{k} S(\mathbf{k}) \tilde{\alpha}_2(\mathbf{k}; L)
= 16 \frac{|\det(M^{-1})|^2}{|\det(\mathbf{B}^{-1})|^2} \sum_{\mathbf{n} \in \mathbb{Z}^2 \setminus \mathbf{0}} \prod_{i=1}^2 \left( \frac{\sin(2\pi (M^{-1})^T \mathbf{Bn}_i)}{2\pi ((M^{-1})^T \mathbf{Bn}_i)} \right)^2
= \left| \frac{|\det(P)|^2}{\pi^4} \sum_{\mathbf{n} \in \mathbb{Z}^2 \setminus \mathbf{0}} \prod_{i=1}^2 \left( \frac{\sin(2\pi [P\mathbf{n}]_i)}{[P\mathbf{n}]_i} \right)^2 \right|,
\quad (2.87)
$$

where $\theta$ stands for the angle between two vectors $\mathbf{A}_1$ and $\mathbf{a}_1$, $P = (M^{-1})^T \mathbf{B}$ whose matrix element is

$$
P_{ij} = \mathbf{A}_i^T \mathbf{b}_j,
\quad (2.88)
$$

and $[P\mathbf{n}]_i = \mathbf{A}_i^T (n_1 \mathbf{b}_1 + n_2 \mathbf{b}_2)$ for $i = 1, 2$. We define $\theta$ as a “rational angle” for any two-dimensional Bravais lattice and parallelogram windows if only one of $[P\mathbf{n}]_i = 0$ has a non-trivial integral solution $\mathbf{n}$. Note that $\sigma_N^2(L; \theta)$ grows like $L^2$ at such an angle $\theta$, to be contrasted with the spherical-window condition (2.2). Straightforwardly, one
generalizes (2.87) to $d$-dimension as

$$\sigma^2_N(L; P) = \frac{|\det(P)|^2}{\pi^{2d}} \sum_{n \in \mathbb{Z}^d \setminus \{0\}} \prod_{i=1}^{d} \left( \frac{\sin(2\pi [Pn]_i L)}{[Pn]_i} \right)^2,$$  \hspace{1cm} (2.89)

and the window orientations with respect to the lattice is characterized by the $d \times d$ matrix $P$, defined in (2.88). We say that the window is at the rational orientation if $P$ has a non-trivial integral solution $n \in \mathbb{Z}^d \setminus \{0\}$ satisfying that a vector $Pn$ has at least $d/2$ vanishing elements.

**Remarks**

1. For the square lattice and for the square windows is rotated by $\theta$ with respect to the lattice, $M$ is the identity matrix, and $B = R^T$, where $R$ is given by (2.38). Then, we can recover (2.43).

2. If both window and lattice are spanned by the same basis vectors and are aligned, i.e., $\theta = 0$ and $P_{ij} = \delta_{ij}$, then (2.87) becomes identical to $\sigma^2_N(L; \theta)$ of the square lattice using a square window of side length $2L$, given by (2.33), up to a proportional constant. However, if the lattice is rotated with respect to the window, then operator $P$, defined in (2.87), can be written as

$$P = (M^T)^{-1} R M^T,$$  \hspace{1cm} (2.90)

where $R$ is a rotation operator. Thus, $P$ is a similarity transform of the rotation operator $R$ in this case.
2.8.1 Square lattice and rectangular windows

The generalization to a rectangular window and the square lattice is straightforward. For a rectangular window, rotated by an angle $\theta$, of two side lengths $aL$ and $bL$,

$$P = \begin{bmatrix} a & 0 \\ 0 & b \end{bmatrix} R, \quad (2.91)$$

and thus $[P \mathbf{n}]_1 = a [R \mathbf{n}]_1$ and $[P \mathbf{n}]_2 = b [R \mathbf{n}]_2$, where $R$ is the rotation matrix, defined by (2.38). Therefore, (2.87) becomes

$$\sigma_N^2(L; \theta) = \frac{1}{\pi^4} \sum_{\mathbf{n} \in \mathbb{Z}^2 \setminus \mathbf{0}} \left( \frac{\sin (2\pi [R \mathbf{n}]_1 aL)}{[R \mathbf{n}]_1} \right)^2 \left( \frac{\sin (2\pi [R \mathbf{n}]_2 bL)}{[R \mathbf{n}]_2} \right)^2. \quad (2.92)$$

Note that (2.92) is similar to its counterpart to square windows (2.42). Thus, one can immediately notice that for the square lattice, rational angles for both rectangular and square windows are the same, and the variance at the rational angle is

$$\sigma_N^2(L; \theta) = \frac{(2ax)^2}{L_0^4} g(2bx) + \frac{(2bx)^2}{L_0^4} g(2ax) + \frac{g(2ax) g(2bx)}{L_0^4} + \frac{1}{L_0^4} \sum_{k=1}^{L_0^2-1} B\left(ax, x^{(k)}\right) B\left(bx, y^{(k)}\right), \quad (2.93)$$

where we use the same notations that we defined in (2.47). We note that (2.93) asymptotically increases like $L^2$. The same result was derived by Rosen [250] using a different approach.
2.8.2 Triangular lattice and square windows

Consider a triangular lattice of lattice constant $a$ with whose number density $\rho = \frac{2\sqrt{3}}{a^2}$ and square windows of side length $2L$. Let the lattice vectors be specified by

$$\mathbf{a}_1 = a \begin{bmatrix} 1 & 0 \end{bmatrix}^T, \quad \mathbf{a}_2 = a \begin{bmatrix} 1/2 & \sqrt{3}/2 \end{bmatrix}^T,$$

thus the corresponding reciprocal vectors are given by

$$\mathbf{b}_1 = a^{-1} \begin{bmatrix} 1 & -1/\sqrt{3} \end{bmatrix}^T, \quad \mathbf{b}_2 = a^{-1} \begin{bmatrix} 0 & 2/\sqrt{3} \end{bmatrix}^T. \tag{2.95}$$

For simplicity, consider that the principal axes of square windows are aligned along axes of Cartesian coordinates, and the lattice is rotated counterclockwise by $-\theta$ with respect to the square window. Then, the matrix $P$, defined by (2.88), will be

$$P = IB = R^T \begin{bmatrix} \mathbf{b}_1 & \mathbf{b}_2 \end{bmatrix} = \frac{1}{a} \begin{bmatrix} \cos \theta - \frac{1}{\sqrt{3}} \sin \theta & \frac{2}{\sqrt{3}} \sin \theta \\ -\sin \theta - \frac{1}{\sqrt{3}} \cos \theta & \frac{2}{\sqrt{3}} \cos \theta \end{bmatrix}. \tag{2.96}$$

Here, we can obtain two types of “rational angles” in this case:

$$\tan(\theta) = \frac{\sqrt{3}n}{n - 2m}, \frac{2m - n}{\sqrt{3}n} \tag{2.97}$$

for integers $n$ and $m$. For a given rational angle $\theta$, defined by (2.97), there are at most two pairs of coprime integers $n_i = (n_i, m_i)$ for $i = 1, 2$:

$$m_1/n_1 = \left(1 + \sqrt{3}\tan(\theta)\right)/2, \quad m_2/n_2 = \left(1 - \sqrt{3}\cot(\theta)\right)/2. \tag{2.98}$$

Then, the relation between $n_1$ and $n_2$ can be obtained:

$$m_2/n_2 = (m_1 - 2n_1) / (2m_1 - n_1). \tag{2.99}$$
Thus, we can define two length scales $L_1$, $L_2$ at a given rational angle $\theta$:

$$L_i \equiv |[P \mathbf{n}_i]| = \frac{2}{\sqrt{3a}} \sqrt{n_i^2 - n_i m_i + m_i^2}, \quad (2.100)$$

for $i = 1, 2$, and these two length scales are related in the following way:

$$L_2 = \sqrt{3} \left| \frac{n_2}{2m_1 - n_1} \right| L_1 = \sqrt{3}L_1. \quad (2.101)$$

If neither $m_2$ and $n_2$ is zero, $|m_1 - 2n_1|$ and $|2m_1 - n_1|$ in (2.98) are coprime due to the working principle of Euclidean algorithm (see [277]). Because of the uniqueness of the irreducible fraction, $|n_2| = |2m_1 - n_1|$ and $|m_2| = |m_1 - 2n_1|$, which leads the last equality in (2.101) to be valid. Table 2.1 lists some $n_1$ and $n_2$ pairs.

<table>
<thead>
<tr>
<th>$\tan(\theta)$</th>
<th>$m_1$</th>
<th>$n_1$</th>
<th>$m_2$</th>
<th>$n_2$</th>
<th>$L_1$</th>
<th>$L_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$1/(3\sqrt{3})$</td>
<td>2</td>
<td>3</td>
<td>-4</td>
<td>1</td>
<td>$2\sqrt{7/3}$</td>
<td>$2\sqrt{7}$</td>
</tr>
<tr>
<td>$1/(7\sqrt{3})$</td>
<td>4</td>
<td>7</td>
<td>-10</td>
<td>1</td>
<td>$2\sqrt{37/3}$</td>
<td>$2\sqrt{37}$</td>
</tr>
<tr>
<td>$11/(9\sqrt{3})$</td>
<td>10</td>
<td>9</td>
<td>-11</td>
<td>8</td>
<td>$2\sqrt{91/3}$</td>
<td>$2\sqrt{91}$</td>
</tr>
<tr>
<td>$13/(7\sqrt{3})$</td>
<td>10</td>
<td>7</td>
<td>-4</td>
<td>13</td>
<td>$2\sqrt{79/3}$</td>
<td>$2\sqrt{79}$</td>
</tr>
<tr>
<td>$17/(3\sqrt{3})$</td>
<td>10</td>
<td>3</td>
<td>4</td>
<td>17</td>
<td>$2\sqrt{79/3}$</td>
<td>$2\sqrt{79}$</td>
</tr>
</tbody>
</table>

Table 2.1: $n_1$ and $n_2$ pairs for rational angles in the triangular lattice and the square windows.

Then, we can find an approximate (2.87) at rational angles:

$$\sigma_N^2(L; \theta) \approx \frac{\det(P)^2}{\pi^4} (2\pi L)^2 \sum_{k=1}^{\infty} \left[ \frac{2 \sin^2(2\pi [P(k\mathbf{n}_1)]_1 L)}{[P(k\mathbf{n}_1)]_1^2} + \frac{2 \sin^2(2\pi [P(k\mathbf{n}_2)]_2 L)}{[P(k\mathbf{n}_2)]_2^2} \right]$$

$$= \left( \frac{2}{\sqrt{3a^2}} \right)^2 (2L)^2 \left[ \frac{g(2L_1 L)}{L_1^2} + \frac{g(2L_2 L)}{L_2^2} \right] \quad (L \to \infty), \quad (2.102)$$

where an identity, $g(x) \equiv \{x\} (1 - \{x\}) = 2 \sum_{n=1}^{\infty} (\sin(n\pi x)/(n\pi))^2$, is used. Figure 2.13(a) shows that good agreement between the approximation (2.102) and the Monte Carlo calculations. The asymptotic behavior of $\sigma_N^2(L; \theta)$ of the approximation is given
Figure 2.13: The variance for the triangular lattice of the unit lattice constant using square windows. (a) The case of \( \tan(\theta) = 1/\sqrt{27} \), i.e., \((m_1, n_1) = (2, 3)\). This clearly demonstrates good agreement between the approximation (2.102) and the Monte Carlo calculations. (b) A log-log plot of cumulative moving average \( \sigma_N^2 \) vs \( L/L_1 \) for various window orientations. Here, the integer pairs in the legend represent \((m_1, n_1)\) in table 2.1. These values collapse onto a single scaling function \( 32x^2/81 \) (2.103) in the large-\( L \) limit. In the Monte Carlo calculations, 40 000 square windows were randomly thrown with a fixed orientation.

by (2.102):

\[
\overline{\sigma_N^2}(L) \approx \left( \frac{2}{\sqrt{3}a^2} \right)^2 \frac{2}{9} \left( \frac{1}{L_1^2} + \frac{1}{L_2^2} \right) L^2 = \frac{32}{81} \left( \frac{L}{aL_1} \right)^2 \quad (L \to \infty).
\] (2.103)

Figure 2.13(b) illustrates that the cumulative moving averages at five different angles collapse onto a single scaling function, as shown in Fig. 2.5.

2.9 Appendix B: Generalizations of the expression

\( \sigma_N^2(L; 0) \) to the higher dimensions

The generalizations of the variance for the square lattice using perfectly-aligned square windows, (2.33), to the higher dimensions are straightforward. In \( d \)-dimension, (2.28)
Figure 2.14: A semi-log plot of $\sigma_N^2(L; 0)$ vs $2L$, as obtained from (2.105).

becomes

$$
\alpha_2(r; L) \equiv v^\text{int}_2(r; L) = \prod_{i=1}^{d} \left( 1 - \frac{|r_i|}{2L} \right) \Theta(2L - |r_i|).
$$

(2.104)

Using (2.104), the variance (2.37) can be readily generalized to the $d$-dimensional case:

$$
\sigma_N^2(L; 0) = (2L)^d \left[ 1 - (2L)^d - \alpha_2(0; L) + \sum_{\mathbf{n} \in \mathbb{Z}^d} \alpha_2(\mathbf{n}; L) \right] \\
= (2L)^d \left[ -(2L)^d + \left( \sum_{n=-[2L]}^{[2L]} (1 - |n|/(2L)) \right)^d \right] \\
= ((2L)^2 + g(2L))^d - (2L)^{2d}.
$$

(2.105)

Its asymptotic expression, $\overline{\sigma_N^2}(L)$, is

$$
\overline{\sigma_N^2}(L; 0) \approx \frac{d}{6(2d - 1)} (2L)^{2(d-1)} (L \to \infty).
$$

(2.106)

Note that the variance increases like the square of the window surface area, and the coefficient $d$ in (2.106) can be interpreted as the maximum number of faces in which
density fluctuations can occur while the window moves. Figure 2.14 shows $\sigma_N^2(L; 0)$ for hypercubic lattice for the first four dimensions.

2.10 Appendix C: Convergence of the second moment of total correlation function in $\mathbb{R}^2$

There are several methods of summability methods to assign a finite value to an infinite sequence that is not convergent in the conventional sense. Here, we will briefly introduce two summability methods, Cesàro and Abelian means, to explain the anomalously large density fluctuations of $\sigma_N^2(L; 0)$ in (2.33).

2.10.1 Cesàro summability

For a given real-valued function $f(x)$ defined in $\mathbb{R}$, its improper integral $\int_0^\infty f(x) \, dx$ is called Cesàro summable or, equivalently, $(C, \alpha)$ summable to a real number $I$, if

$$\lim_{t \to \infty} \int_0^t \left(1 - \frac{x}{t}\right)^\alpha f(x) \, dx = I,$$

(2.107)

$\alpha > 0$. $(C, 0)$ summable is equivalent to the conventional convergence of the given improper integral $\int_0^\infty f(x) \, dx$. $(C, 1)$ summability of the improper integral is the same as the convergence of the cumulative moving average of the integral [286]:

$$\lim_{t \to \infty} \int_0^t \left(1 - \frac{x}{t}\right) f(x) \, dx = \lim_{t \to \infty} \frac{1}{t} \int_0^t \left(\int_0^{x_2} f(x_1) \, dx_1\right) \, dx_2.$$  (2.108)

Using mathematical induction, one can easily show that for a natural number $n$, an improper integral $\lim_{L \to \infty} \int_0^L f(x) \, dx$ is $(C, n)$ summable to $I$, if and only if its $n$-th
multiple cumulative moving average converges to $I/n!$ in the large-$L$ limit:

$$
\lim_{L \to \infty} \int_0^L \left(1 - \frac{x}{L}\right)^n f(x) \, dx = \lim_{L \to \infty} \frac{n!}{L^n} \int_0^L dx_n \int_0^{x_n} \cdots \int_0^{x_1} dx \, f(x).
$$

Note that for spherical windows of radius $R$, $\sigma_N^2(R)$ for the square lattice is asymptotically linear in $R$ in the large-$R$ limit in the sense that [150]

$$
\lim_{R \to \infty} \frac{1}{R} \int_1^R \frac{\sigma_N^2(r)}{r} \, dr
$$

converges to the constant. Using the analysis in (2.20), one can express (2.110) as

$$
\sim \lim_{R \to \infty} \frac{-1}{2 \Gamma(1/2) \Gamma(3/2) R} \int_1^R dr \int_0^r dr' \, r'^2 h(r'),
$$

which implies that the 2nd moment of the total correlation function of the square lattice is $(C,1)$ summable in the circular boundary.

## 2.10.2 Abelian summation

Suppose that $\lambda = \{\lambda_0, \lambda_1, \lambda_2, \cdots\}$ is a strictly increasing sequence approaching infinity, and $\lambda_0 > 0$. The Abelian mean $A_\lambda$ of a sequence $c = \{c_0, c_1, c_2, \cdots\}$ is defined as

$$
A_\lambda = \lim_{\beta \to 0^+} f(\beta),
$$

where $f(\beta) \equiv \sum_{n=0}^\infty c_n e^{-\beta \lambda_n}$, and $f(\beta)$ is assumed to be convergent for all real numbers $\beta > 0$. Abelian summation of a sequence $a$ is a special case of its Abelian mean in which $\lambda_n = n$, and thus $f(\beta) = \sum_{n=0}^\infty c_n e^{-\beta n}$. 

2.10.3 Conditional convergence of the integral involving the total correlation function.

For the square lattice, the integrals of total correlation function, (2.21) and (2.55), oscillate but do not converge in the conventional sense. These integrals are summable via an Abelian sum, which turns out to be equivalent to the “convergence trick” [313]. For both square and circular windows, the volume integral $\int h(r) \, dr$ can be written in the same form (see (2.57))

$$\int_{\mathbb{R}^2} h(r) \, dr = \lim_{\beta \to 0^+} \int_{\mathbb{R}^2} h(r) e^{-\beta r^2} \, dr = \lim_{\beta \to 0^+} \left[ \sum_{k=1}^{\infty} Z_k e^{-\beta r_k^2} - \frac{\pi}{\beta} \right] = -1. \quad (2.113)$$

In the analysis leading to (2.20), the convergence of the second moment of total correlation function $h(r)$ determines the asymptotic behavior of the variance $\sigma_N^2$ in

Figure 2.15: A log-log scale plot of Abelian sums of second moment of total correlation function for circular and square windows as functions of $\beta$. Black circles and blue squares represent the integrals for circular windows (2.114) and for square windows (2.115), respectively. To compute (2.114), we summed $Z_k r_k$ up to $r_k = \sqrt{10^7}$, leading the integral to diverge as $\beta \to 0$ in the figure. We compute the limiting value of the infinite sum (2.114) by a linear regression (red dotted line). For a given $\beta$, we compute the infinite sum (2.115) within arbitrarily accuracy, and observe that it diverges in the order of $\beta^{-0.5}$ (red solid line).
In order to assign a finite value to (2.22) for a circular window, we use the Abelian sum again, yielding

\[
- \int_{\mathbb{R}^2} r h(r) \, dr = - \lim_{\beta \to 0^+} \int_{\mathbb{R}^2} r h(r) e^{-\beta r^2} \, dr = \lim_{\beta \to 0^+} \left[ - \sum_{k=1}^{\infty} Z_k r_k e^{-\beta r_k^2} + \frac{1}{2} \left( \frac{\pi}{\beta} \right)^{3/2} \right].
\]

(2.114)

Similarly, the Abelian sum of the second moment of total correlation function for a square window is given by:

\[
- \int_{\mathbb{R}^2} |x| h(r) \, dr = - \lim_{\beta \to 0^+} \int_{\mathbb{R}^2} |x| h(r) e^{-\beta r^2} \, dr = \lim_{\beta \to 0^+} \left[ -2 \sum_{n=1}^{\infty} n e^{-\beta n^2} \left( 1 + 2 \sum_{m=1}^{\infty} e^{-\beta m^2} \right) + \frac{\sqrt{\pi}}{\beta^{3/2}} \right].
\]

(2.115)

As we can see in Fig. 2.15, the second moment for a circular window (2.114) converges to a finite value, while that for square window (2.115) diverges.

Figure 2.15 shows the second moments of the total correlation functions for both circular and square windows. The second moment of \( h(r) \) for circular windows (2.114) can be approximated by a straight line, and its \( y \)-interpolation gives the limit value (0.228821), which is consistent with the value given in [313]. The second moment of \( h(r) \) for square window (2.115) can be approximated by \( 0.3/\sqrt{\beta} \), yielding the result

\[
- \lim_{\beta \to 0^+} \int_{\mathbb{R}^2} |x| h(r) e^{-\beta r^2} \, dr \sim L.
\]

(2.116)

2.11 Appendix D: Proof of \( B(x, y) \)

Here we prove that the series representation of \( B(x, y) \) given in (2.48) is identical to its closed-form expression given in (2.49). Specifically, we prove this result by showing that the Fourier series of (2.49) is the same as the infinite sum because Eq. (2.49) is continuous and its derivative is piecewise continuous with respect to \( x \) [38]. Since

\[
\text{the limit of } R \to \infty.
\]

...
$B(x, y)$ is an even function with respect to $x$, we can express it as a cosine series:

$$B^{(\text{sum})}(x, y) = \frac{a_0}{2} + \sum_{k=1}^{\infty} a_k \cos \left( 2\pi \frac{k}{T_0} x \right),$$  \hspace{1cm} (2.117)

where

$$a_k = \frac{2}{T_0} \int_{0}^{T_0} B(x, y) \cos \left( 2\pi \frac{k}{T_0} x \right) \, dx,$$  \hspace{1cm} (2.118)

for non-negative integers $n$. Note that the closed expression for $B(x, y)$ is the linear interpolation of $\sin^2(2\pi yx)/\sin^2(\pi y)$ whose data points are at $x = k/2$. Using the expression for the linear interpolation of a function $f(x)$ given by

$$f^{(1)}(x) = \sum_{k=-\infty}^{\infty} f(kT) \Delta_T(x - kT),$$  \hspace{1cm} (2.119)

where $\Delta_T(x) = (1 - |x|/T) \Theta(T - |x|)$ and $T$ is the sampling interval, the closed expression $B^{(\text{closed})}(x, y)$ can be written as

$$B^{(\text{closed})}(x, y) = \sum_{k=-\infty}^{\infty} \frac{\sin^2(\pi yk)}{\sin^2(\pi y)} \Delta_{1/2}\left(x - \frac{k}{2}\right).$$  \hspace{1cm} (2.120)

Substituting (2.120) into (2.118), for a positive integer $m$, one obtains

$$a_i = \frac{2}{T_0} \sum_{k=-\infty}^{\infty} \frac{\sin^2\left(\frac{\pi yk}{T_0}\right)}{\sin^2\left(\frac{\pi y}{T_0}\right)} \int_{0}^{T_0} \Delta_{1/2}\left(x - \frac{k}{2}\right) \cos \left( 2\pi \frac{i}{T_0} x \right) \, dx,$$  \hspace{1cm} (2.121)

where $y = c/T_0$, $c$ is a natural number, $T_0 = L_0^2$, and $L_0 \equiv \sqrt{n^2 + m^2}$ in the text. Since

$$\int_{k/2-1/2}^{k/2+1/2} \Delta_{1/2}\left(x - \frac{k}{2}\right) \cos (2\pi Kx) \, dx = 2 \left( \frac{\sin (\pi K/2)}{\pi K} \right)^2 \cos (\pi Kk),$$  \hspace{1cm} (2.122)
for a real number \( K \), (2.121) can be written as

\[
a_i = 2 \left( \frac{\sin \left( \frac{\pi i}{T_0} \right)}{\pi \frac{i}{T_0} \sin \left( \frac{\pi c}{T_0} \right)} \right)^2 \frac{1}{2T_0} \sum_{k=1}^{2T_0} \left[ 2 \cos \left( \frac{\pi k i}{T_0} \right) - \cos \left( 2\pi k \frac{c+i/2}{T_0} \right) - \cos \left( 2\pi k \frac{c-i/2}{T_0} \right) \right]
\]

\[
= 2 \left( \frac{\sin \left( \frac{\pi i}{T_0} \right)}{\pi \frac{i}{T_0} \sin \left( \frac{\pi c}{T_0} \right)} \right)^2 \sum_{k=-\infty}^{\infty} \left[ 2\delta_{1,2kL_0} - \delta_{1,2kT_0-2c} - \delta_{1,2kT_0+2c} \right], \tag{2.123}
\]

where \( \delta_{n,m} \) is a Kronecker delta symbol. Therefore, non-trivial coefficients \( a_m \) are

\[
a_0 = \frac{1}{\sin^2 \left( \frac{\pi c}{T_0} \right)} = \frac{1}{\sin^2 \left( \pi y \right)} \tag{2.124}
\]

\[
a_{2nL_0 \pm 2c} = -2L_0^2 / \left( \pi^2 (2nL_0 \pm 2c)^2 \right) = -1 / \left( 2\pi^2 (n \pm y)^2 \right). \tag{2.125}
\]

Using the identity \( \sum_{k=-\infty}^{\infty} 1/[\pi^2 (k+y)^2] = 1/[\sin^2(\pi y)] \), and inserting (2.124) and (2.125) into (2.117), one can obtain

\[
B^{(\text{sum})}(x, y) = \frac{1}{2 \sin^2(\pi y)} - \sum_{n=-\infty}^{\infty} \frac{\cos (4\pi (n+y)x)}{2\pi^2 (n+y)^2} = \sum_{n=-\infty}^{\infty} \frac{\sin^2 (2\pi (n+y)x)}{\pi^2 (n+y)^2}. \tag{2.126}
\]

Note that (2.126) is identical to the summation form given in (2.48) in the main text, which completes the proof.

### 2.12 Appendix E: Continued fraction and irrational numbers

In Sec. 2.4.2, we heavily used terms in number theory (e.g., badly approximable numbers, \( k \)th partial quotients, \( \cdots \)), but we do not explain them in detail in the middle of the text for the readability. In this section, we provide definitions of the
terms relevant to the rational approximation. Consider the continued fraction

\[
a_0 + \frac{1}{a_1 + \frac{1}{a_2 + \frac{1}{\cdots + \frac{1}{a_k}}}}
\]

(2.127)

by \([a_0, a_1, a_2, \cdots, a_k]\) for non-negative integers \(a_0, a_1, \cdots, a_k\), a real number \(\alpha\) can be expressed as

\[
\alpha = \lim_{n \to \infty} [a_0, a_1, a_2, \cdots, a_n, \cdots],
\]

(2.128)

where an integer \(a_k\) is called \(k\)-th partial quotient, and is computed by the following recurrence relations:

\[
r_0 = \alpha, \quad a_0 = \lfloor r_0 \rfloor
\]

(2.129)

\[
r_k = \frac{1}{r_{k-1} - a_{k-1}}
\]

(2.130)

\[
a_k = \lfloor r_k \rfloor
\]

(2.131)

for positive integers \(n\). If \(\alpha\) is a rational number, then its partial quotient is terminated in finite terms. Otherwise, its partial quotients form an infinite sequence of non-vanishing integers. If we consider the finite number of partial quotients of an irrational number \(\alpha\), it gives a rational approximation of \(\alpha\), \(c_k = \frac{n_k}{m_k} = [a_0, a_1, \cdots, a_k]\) where \(c_k\) is called \(k\)-th convergent of \(\alpha\). Note that the \(k\)-th convergent is the best rational approximation of \(\alpha\) in the sense that for any rational number \(n/m\),

\[
|\alpha - \frac{n}{m}| > |\alpha - \frac{n_k}{m_k}|
\]

(2.132)

as long as \(m < m_k\) [277].

Table 2.2 shows partial quotients of several irrational numbers and their rational
approximations up to double and quadruple precision in the form of the irreducible fraction. Note that irrational numbers whose partial quotients are bounded, e.g., $\sqrt{N}$ and the golden ratio $\phi$, are “badly approximable numbers” (BAN) [36] in the sense that for every rational number $n/m$

$$\left| \alpha - \frac{n}{m} \right| > \frac{c(\alpha)}{m^2}. \quad (2.133)$$

In other words, the best error involving the rational approximation for a badly approximable number decreases like $1/m^2$, while the best error for a non-badly approximable number occasionally can be arbitrary smaller than $1/m^2$. 

<table>
<thead>
<tr>
<th>$\alpha$</th>
<th>$a_0$</th>
<th>$a_1$</th>
<th>$a_2$</th>
<th>$a_3$</th>
<th>$a_4$</th>
<th>$a_5$</th>
<th>$a_n$</th>
<th>$c_{\text{double}}$</th>
<th>$c_{\text{quadruple}}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\sqrt{2}$</td>
<td>1</td>
<td>2</td>
<td>2</td>
<td>2</td>
<td>2</td>
<td>2</td>
<td>2</td>
<td>54608393</td>
<td>5964153172084899</td>
</tr>
<tr>
<td>$\frac{1}{\sqrt{2}}$</td>
<td>0</td>
<td>1</td>
<td>2</td>
<td>2</td>
<td>2</td>
<td>2</td>
<td>2</td>
<td>38613095</td>
<td>4217285120164900</td>
</tr>
<tr>
<td>$\phi = \frac{1+\sqrt{5}}{2}$</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>39088169</td>
<td>3416454622906707</td>
</tr>
<tr>
<td>$e$</td>
<td>2</td>
<td>1</td>
<td>2</td>
<td>1</td>
<td>1</td>
<td>4</td>
<td>${ 2k, \ n = 3k - 1 }$</td>
<td>28245729</td>
<td>2124008553358849</td>
</tr>
<tr>
<td>$\pi$</td>
<td>3</td>
<td>7</td>
<td>15</td>
<td>1</td>
<td>292</td>
<td>1</td>
<td>Unknown$^1$</td>
<td>80143857</td>
<td>6134899525417045</td>
</tr>
<tr>
<td>tanh (1)</td>
<td>0</td>
<td>1</td>
<td>3</td>
<td>5</td>
<td>7</td>
<td>9</td>
<td>$3n$</td>
<td>25510582</td>
<td>1932739169684491</td>
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<td></td>
<td></td>
<td></td>
<td>3025576</td>
<td>323383850850476</td>
</tr>
</tbody>
</table>

Table 2.2: List of partial quotients of some irrational numbers. $c_{\text{double}}$ and $c_{\text{quadruple}}$ represent convergents of the values of an irrational number up to 16 and 32 significant figures. Upper three numbers are examples of irrational numbers belong to BAN, and the lower three are not in BAN.
Chapter 3

Effect of Imperfections on the Hyperuniformity of Many-Body Systems

3.1 Introduction

It is well-known that imperfections play significant roles in determining the physical and structural properties of crystals; see Refs. [7, 159, 42] and references therein. For instance, metallic crystals can be significantly hardened by increasing densities of point defects and dislocations, i.e., strain hardening [60, 7]. In semiconductors and insulators, impurities of certain elements can change their properties, such as electrical conductivities and colors [7, 159]. Another example is an anomalous phenomenon that the electrical resistivity of metal increases as the temperature drops below a certain value due to magnetic impurities, called the Kondo effect [165, 120].

Importantly, understanding how imperfections can affect the hyperuniformity of a system and its associated physical properties has been currently lacking for both ordered and disordered hyperuniform solid-phase systems. Our interest in this chapter
is to explore the degree to which the introduction of imperfections in perfectly ordered or disordered hyperuniform systems degrades or destroys their original perfect hyperuniformity. This is accomplished by quantifying the corresponding structure factors. Interestingly, the hyperuniformity concept provides a precise mathematical means of detecting imperfections in amorphous hyperuniform systems via either the violation of the hyperuniformity criterion, i.e., \( \lim_{|k| \to 0} S(k) = 0 \), or changes in the small-wavenumber behavior of \( S(k) \). It is important to note that in the same way that there is no perfect ordered hyperuniform system (i.e., crystals and quasicrystals) in practice due to the inevitable existence of imperfections, such as point defects [7, 159, 42], dislocations [7, 159, 42], and phasons [274], there is no perfect disordered hyperuniform system.

It is instructive to discuss briefly computational and experimental methods that have been formulated to construct disordered hyperuniform systems. Computational methods have been developed in both equilibrium and nonequilibrium systems. For example, the collective-coordinate optimization technique is a computational tool that generates classical ground states with a target structure factor \( S(k) \) for a set of wavevectors [321, 322, 317, 352, 353, 347]. Packing protocols such as Lubachevsky-Stillinger (LS) and Torquato-Jiao (TJ) algorithms are used to obtain (putative) strictly jammed MRJ hard-sphere packings, which are conjectured to be hyperuniform [313, 62, 10]. Random organization models [122, 287, 123, 121] can yield disordered hyperuniform point configurations at the critical point. Recently, Ma and Torquato demonstrated that the Cahn-Hilliard and Swift-Hohenberg equations could yield disordered hyperuniform scalar fields [194]. Experimental methods have been devised to produce (nearly) disordered hyperuniform systems, including periodically driven colloidal suspensions [329], jammed colloidal suspensions [168, 64, 245], annealed amorphous silicon [338], and self-assembling patterns of block-copolymers [364].

Such disordered hyperuniform systems, whether they are in thermal equilibrium
or not, inevitably include some fraction of imperfections. Such imperfections may degrade or destroy the hyperuniformity, albeit in some cases to a small degree. For instance, any compressible system in thermal equilibrium, which has a positive isothermal compressibility ($\kappa_T > 0$), cannot be hyperuniform at positive temperatures due to thermal excitations. This conclusion follows from the fluctuation-compressibility relation [317, 299, 300], i.e., $S(0) = \rho \kappa_T k_B T$, where $\rho$ is number density, $k_B$ is the Boltzmann constant, and $T$ is the temperature. By theoretically estimating the isothermal compressibility of excited states at sufficiently low $T$ associated with certain disordered hyperuniform ground states, Torquato et al. [317] utilized the aforementioned compressibility relation to quantify how hyperuniformity is destroyed in such cases. However, an understanding of the underlying mechanisms that result in the destruction of hyperuniformity for positive temperatures as well as the behavior of the structure factor away from the origin, whether the ground states are ordered or not, has heretofore been lacking.

It can be also difficult to generate perfect realizations of nonequilibrium hyperuniform systems, partly due to a type of critical slowing down phenomenon [10, 300]. This refers to the fact that such nonequilibrium systems are at critical points, which require significantly longer and longer computational times to achieve as the critical states are approached [62, 10, 122, 287, 288]. For instance, MRJ hard-sphere packings correspond to the hyperuniform critical states that occur at the jamming transition [62, 10]. Due to a critical slowing down as well as the presence of “rattlers” imperfections, numerically generated MRJ packings deviate from being perfectly strictly jammed and hence are not perfectly hyperuniform, e.g., $S(0) \sim 10^{-4}$ [62, 10]. It has been conjectured that the ideal MRJ state is free of any rattlers and hence would be exactly hyperuniform [300, 313].

Since imperfections, as well as finite-size effects and numerical errors, affect hyperuniformity of systems in computer simulations and experiments, it is desirable to
develop a rough criterion to determine whether a system is “nearly” or “effectively” hyperuniform. A useful empirical and operational criterion that has been proposed [10, 300] for this purpose is the hyperuniform metric $H$, which is defined by

$$H \equiv \frac{S(k \to 0)}{S(k_{\text{peak}})},$$

(3.1)

where $S(k_{\text{peak}})$ is the structure factor at the first dominant peak. A given disordered system can be regarded as effectively hyperuniform if the ratio $H$ is of the order of $10^{-4}$ or smaller.

Figure 3.1: (Color online) Left panel: Configurations of (a) an initially perfectly hyperuniform integer lattice and (b)-(e) imperfect lattices. The different types of imperfections include the following: (b) a single vacancy, (c) a single interstitial defect [denoted by a solid cyan five-pointed star], (d) uncorrelated stochastic particle displacements $u$ via a uniform distribution with the variance $\langle u^2 \rangle = 0.05$, and (e) thermalized excitations, i.e., elastic waves at $T = 0.05$, where $T$ is a dimensionless temperature (3.54). Right panel: Corresponding deviations in the number of particles $N(x_0; L)$ inside a window centered at $x_0$, i.e., $\delta N(x_0; L) \equiv N(x_0; L) - \langle N(x_0; L) \rangle$ at two different window sizes $L$. The local number variance $\sigma^2_{\delta N}(L)$, or equivalently the volume average of $\delta N(x_0; L)^2$, measures the degree of density fluctuations at a given length scale $L$. Roughly speaking, a system is nonhyperuniform if $\sigma^2_{\delta N}(L)$ grows as the window size increases, as in cases (b), (c) and (d). By contrast, the perturbed system (d) is hyperuniform.

The overall objective of this chapter is to understand quantitatively the extent to which hyperuniformity is degraded or destroyed when one introduces the following
three types of imperfections into perfectly hyperuniform many-body systems: (1) uncorrelated point defects, including vacancies and interstitials, (2) stochastic particle displacements, and (3) thermal excitations. The left panel of Fig. 3.1 illustrates configurations of (a) a perfect integer lattice and (b)-(e) four imperfect variants. While the integer lattice (a) is obviously hyperuniform, it is difficult to determine whether the others are hyperuniform with the naked eye.

Note that the right panel of Fig. 3.1 enables one to gauge qualitatively whether they are hyperuniform by looking at the deviation $\delta N(x_0; L)$ or the volume average of $\delta N(x_0; L)^2$, i.e., the local number variance $\sigma^2_N(L)$. We show that while in three systems (b), (c) and (e), density fluctuations are larger [i.e., the deviation $\delta N(x_0; L)$ becomes non-zero more frequently] at the larger length scales ($L = 10$) than at the smaller ones ($L = 1$), the density fluctuations in (d) barely change with length scales. This observation qualitatively shows that the imperfect system described in (d) is hyperuniform, but the others in (b), (c), and (e) are not, although counterintuitively the particles in (d) look more clustered than those in (e). The nonhyperuniformity of the example in (e) can be qualitatively understood by noting that thermalized excitations in a crystal can be decomposed into a sum of sinusoidal functions with different wavelengths. A single longitudinal lattice wave of the longest wavelength induces global inhomogeneity with density modulation. One half of the system becomes denser than the other half of the system, resulting in nonhyperuniformity.

In the main text, we will quantitatively validate all of these qualitative explanations by deriving explicit formulas for $S(k)$ in the small-$|k|$ regime for each type of imperfection. We then demonstrate that our theoretical results are in excellent agreement with corresponding numerical simulations.

We begin by showing that uncorrelated point defects (both vacancies and interstitials) destroy the hyperuniformity in proportion to the defect concentration $p$ for small $p$. While we focus in this context on two types of point defects, these results
can be easily generalized to other types of point defects. Subsequently, we quantitatively study *perturbed point processes* [331, 90, 85], which are generated from an initial point process by stochastically displacing each point in either an uncorrelated or correlated manner. When displacements are uncorrelated, although the hyperuniformity is degraded to some extent, it is never destroyed, i.e., still \( \lim_{|k| \to 0} S(k) = 0 \). Using this property, we present a simple method that transforms class I systems (e.g., lattices and disordered stealthy hyperuniform systems) to class III systems [300]; see Eq. (3.12). In addition, we ascertain conditions under which stochastic displacements can destroy the hyperuniformity of initial hyperuniform point processes, both ordered and disordered.

This study is followed by an investigation of *thermalized crystals*, i.e., classical crystals in thermal equilibrium in the harmonic regime. Due to thermal motions, particles in a thermalized crystal are displaced from their ideal arrangement, i.e., a ground-state crystal. At first glance, one might think that at a sufficiently low temperature, a thermalized crystal is the same as an uncorrelated perturbed lattice, and thus it is hyperuniform. However, this picture is wrong because excited particles move collectively and form long-wavelength lattice waves. We prove that thermalized crystals can be mapped to special correlated perturbed lattices that are “nonhyperuniform.” We show that our expression for \( S(k) \) are in excellent agreement with numerical simulations at temperatures much lower than the melting point \(^1\). Our expression for \( S(k) \) in the zero-wavenumber limit also yields the correct isothermal compressibility \( \kappa_T \) in the low-temperature limit.

We present basic mathematical definitions and concepts in Sec. 3.2. In Sec. 3.3, we theoretically and numerically investigate the effects of uncorrelated point defects on the hyperuniformity of otherwise hyperuniform systems. In Sec. 3.4, we study the hyperuniformity of perturbed point processes. Here, we introduce and examine a

\(^1\)As the temperature increases, anharmonic effects would also contribute to the degradation of hyperuniformity in crystalline ground-states.
family of singlet displacement probability densities to generate class III systems [300].

In Sec. 3.5, we investigate the effects of thermal excitations in classical harmonic lattices on their hyperuniformity. Finally, we provide concluding remarks in Sec. 3.6.

3.2 Background and Definitions

3.2.1 Point processes

Roughly speaking, a point process in $d$-dimensional Euclidean space $\mathbb{R}^d$ is a spatial distribution of infinitely many points $x_1, x_2, \ldots$, which can be described by a microscopic density function $n(r)$:

$$n(r) = \sum_{i=1}^{\infty} \delta(r - x_i),$$  \hspace{1cm} (3.2)

where $\delta(x)$ denotes the Dirac delta function in $\mathbb{R}^d$. The $n$-point density correlation function $\rho_n(r^n)$ is defined by $\rho_n(r^n) \equiv \langle n(r_1) n(r_2) \cdots n(r_n) \rangle$, where $r^n = r_1, r_2, \ldots, r_n$ and $\langle \cdot \rangle$ represents an ensemble average. This function is proportional to the probability density associated with finding $n$ different points at $r_1, r_2, \ldots, r_n$. For statistically homogeneous point processes at a given number density $\rho$ (number of particles per unit volume), the $n$-point correlation function depends on relative positions of points, i.e., $\rho_n(r^n) = \rho^n g_n(r_{21}, \ldots, r_{n1})$ with $r_{ij} \equiv r_j - r_i$ for $1 \leq i \neq j \leq n$ and $\rho_1(r) = \rho$.

The pair correlation function $g_2(r)$ and total correlation function $h(r)$, defined as $h(r) \equiv g_2(r) - 1$, are of special importance in statistical mechanics [108]. For systems without long-range order, $g_2(r) \to 1$ and $h(r) \to 0$ as $|r| \to \infty$. The autocovariance function $\chi(r) \equiv \langle [n(r + r_0) - \rho][n(r_0) - \rho] \rangle$ is related to $h(r)$ via

$$\chi(r) = \rho \left[ \delta(r) + \rho h(r) \right].$$  \hspace{1cm} (3.3)
3.2.2 Structure factor

The **static structure factor** is a mathematical description of scattering intensities. For a finite point configuration \( \{ \mathbf{x}_i \}^{N}_{i=1} \) of \( N \) particles in a unit cell of volume \( V \), the static structure factor is defined as

\[
S(k) \equiv \frac{1}{N} |\tilde{n}(k)|^2 = 1 + \frac{1}{N} \sum_{i \neq j=1}^{N} \exp(-i \mathbf{k} \cdot (\mathbf{x}_i - \mathbf{x}_j)),
\]

(3.4)

where the **collective coordinates** \( \tilde{n}(k) \) is the Fourier transform of the microscopic density \( n(r) \). Under periodic boundary conditions, wavevectors \( k \) are constrained to lie on reciprocal lattice vectors of the unit cell, satisfying \( \exp(i \mathbf{k} \cdot \mathbf{a}_j) = 1 \) for all basis vectors \( \mathbf{a}_j \) of the unit cell. Thus, in the thermodynamic limit \( (N \to \infty \text{ with } \rho \equiv N/V \text{ fixed}) \), a wavevector \( k \) becomes a continuous parameter.

In the thermodynamic limit, the static structure factor of a point process is defined by an ensemble average \( \langle S(k) \rangle \) of (3.4) with the forward scattering excluded:

\[
S(k) \equiv \langle S(k) \rangle - (2\pi)^d \rho \delta(k).
\]

(3.5)

The static structure factor (3.5) is related to the Fourier transform of two-point functions, \( h(r) \) and \( \chi(r) \), defined in the expression (3.3): \( S(k) = 1 + \rho \tilde{h}(k) = \tilde{\chi}(k) / \rho \).

In the rest of this chapter, unless otherwise stated, \( S(k) \) denotes the static structure factors defined in (3.5).

We will use the following definition of Fourier transform \( \tilde{f}(k) \) and the inverse transform \( f(r) \) (assuming their existence):

\[
\tilde{f}(k) = \int_{\mathbb{R}^d} f(r) e^{-i \mathbf{k} \cdot \mathbf{r}} \, d\mathbf{r},
\]

(3.6)

\[
f(r) = \left( \frac{1}{2\pi} \right)^d \int_{\mathbb{R}^d} \tilde{f}(k) e^{i \mathbf{k} \cdot \mathbf{r}} \, d\mathbf{k}.
\]

(3.7)
3.2.3 Hyperuniformity

Consider a statistically homogeneous point process at number density $\rho$ in $d$-dimensional Euclidean space $\mathbb{R}^d$. Hyperuniform [313, 299] (also known as ‘superhomogeneous’ [233]) point processes are ones in which long-wavelength density fluctuations are suppressed. Quantitatively, hyperuniformity can be defined in Fourier space via

$$\lim_{|k| \to 0} S(k) = 0,$$  \hspace{1cm} (3.8)

or, alternatively, in direct-space via the local number variance $\sigma_N^2(R)$:

$$\lim_{v_1(R) \to \infty} \frac{\sigma_N^2(R)}{v_1(R)} = 0,$$  \hspace{1cm} (3.9)

where $v_1(R)$ is the volume of a $d$-dimensional hypersphere of radius $R$. Here, the local number variance $\sigma_N^2(R)$ represents the variance in the number of points sampled by randomly placed spherical windows of radius $R$ (see Fig. 3.1), which can be calculated from the following relations [313, 300]:

$$\sigma_N^2(R) = \rho v_1(R) \left[ 1 + \rho \int_{\mathbb{R}^d} \mathrm{d}r h(r) \alpha_2(r; R) \right]$$  \hspace{1cm} (3.10)

$$= \frac{\rho v_1(R)}{(2\pi)^d} \int_{\mathbb{R}^d} \mathrm{d}k S(k) \tilde{\alpha}_2(k; R),$$  \hspace{1cm} (3.11)

where $\alpha_2(r; R)$ represents the scaled intersection volume of two spherical windows of radius $R$ that are separated by $r$, and $\tilde{\alpha}_2(k; R)$ is its Fourier transform.

Consider hyperuniform systems that are characterized by structure factors with power-law form for small wavenumbers; $S(k) \sim |k|^\alpha$. The exponent $\alpha$ determines the large-$R$ asymptotic behavior of $\sigma_N^2(R)$ [313, 346, 299]. Using this asymptotic behavior, Torquato recently has categorized hyperuniform point processes into three


\[ \sigma_N^2(R) \sim \begin{cases} 
R^{d-1}, & \alpha > 1 \text{ (class I)} \\
R^{d-1} \ln(R), & \alpha = 1 \text{ (class II)} \\
R^{d-\alpha}, & 0 < \alpha < 1 \text{ (class III)}.
\end{cases} \tag{3.12} \]

Class I systems include crystals, some quasicrystals, and disordered stealthy hyperuniform systems [101, 313, 346, 352, 317, 181]. A variety of examples of class II and III systems are given in Ref. [300].

Stealthy hyperuniform ground-state systems are defined by the condition that \( S(k) = 0 \) for \( 0 \leq |k| < K \) for some positive number \( K \). The parameter \( \chi \) provides a measure of the relative fraction of the number of wavevectors at which \( S(k) \) is constrained to be zero to the total number of degrees of freedom [317, 300]. For \( 0 < \chi < 1/2 \), the entropically favored stealthy hyperuniform ground states are highly degenerate and disordered, while they crystallize for \( 1/2 < \chi \leq 1 \).

For single-component systems in thermal equilibrium, the fluctuation-compressibility relation is given by [108]

\[ \lim_{|k| \to 0} S(k) = \rho \kappa_T k_B T, \tag{3.13} \]

where \( \kappa_T \) is the isothermal compressibility, and \( T \) is the temperature of the system. Since the left-hand side of (3.13) is directly related to the long-wavelength density fluctuations, i.e., \( S(0) = \lim_{R \to \infty} \sigma_N^2(R)/\rho v_1(R) \), any compressible system in thermal equilibrium (\( \kappa_T > 0 \)) cannot be hyperuniform at a positive \( T \); see Refs. [317] and [300] for more discussion on this subject.

### 3.3 Effect of Spatially Uncorrelated Defects

The dimensionality of the spatial distribution of imperfections enables a classification into four types: point, line, surface, and volume defects. Roughly speaking, three types of point defects can be considered; vacancies (missing atoms), interstitial
impurities (excess atoms), and substitutional impurities (different kinds of atoms). Dislocations and stacking faults are examples of line and surface defects, respectively. Volume defects include pores and cracks.

Some theories have been devised to identify types and amount of point defects in crystalline solids via existing experimental techniques such as EPR and X-ray scattering experiments [98, 229]. For instance of scattering experiments, one can determine types and sizes of point defects, and whether they are isolated or aggregated, by analyzing the shifts of Bragg peaks and asymmetric diffuse scattering around the Bragg peaks called Huang diffuse scattering [56, 126].

In this section, we investigate how the introduction of uncorrelated point defects influences the hyperuniformity of an original hyperuniform point process at number density $\rho$. Here, we consider a $d$-dimensional hyperuniform point configuration $\{r_i\}_{i=1}^{N_s}$ in a periodic unit cell of volume $V$, and two types of point defects; (1) vacancies and (2) interstitials. The structure factor of this original configuration is denoted by $S_0(k)$.

For simplicity, we do not consider elastic deformations in imperfect configurations due to defects, which can arise in Huang diffuse scattering [56, 126], as well as steric repulsion that can restrict the interstitial positions.

### 3.3.1 Point vacancies

Here, we consider spatially uncorrelated point vacancies by independently removing particles in original configurations. Let us define a stochastic function $f(r)$ to describe point defects in $\mathbb{R}^d$. In general, $f(r)$ is complex-valued, but for uncorrelated point vacancies, it becomes real-valued such that

$$
\Pr(f(r) = a) = \begin{cases} 
1 - p, & \text{if } a = 1 \\
p, & \text{if } a = 0,
\end{cases}
$$

(3.14)
Figure 3.2: (Color online) From (a)-(c): Semi-log plots of simulated structure factors of two-dimensional defective point configurations with point vacancies of the fraction $p$. Three types of original configurations include (a) the square lattice, (b) stealthy disordered hyperuniform configurations with $\chi = 0.1$, and (c) perturbed square lattices via a distribution $f_1(u; \delta = 10^{-4}, \alpha = 0.8)$ given by (3.35). In (c), theoretical values of $S_0(k)$ for perturbed lattices (magenta dashed line) are calculated from (3.36). Insets of each panel zoom into the small-wavenumber regime in linear scale, but $y$-axes are rescaled as $(S(k) - p)/(1 - p)$. Note that peaks observed in (a) and (c) correspond to the first two Bragg peaks of the square lattice. Subfigure (d) is comparison of the prediction (3.20) to corresponding computer simulations of $S(0)$ of defective systems as functions of the vacancy concentration $p$ for three original systems.

where $p$ is the concentration of vacancies. After introducing uncorrelated point vacancies, one can express the structure factor of a defective configuration in terms of the function $f(r)$;

$S(k) = 1 + \left\langle \frac{1}{N} \sum_{i \neq j = 1}^{N_v} f_i f_j^* e^{-i k \cdot (r_i - r_j)} \right\rangle_f$,

(3.15)
where $f_i$ is an abbreviation of $f(r_i)$, $f_i^*$ is its complex conjugate, $\langle \cdot \rangle_f$ represents the expectation value over $f(r)$. Here, $N \equiv \sum_{i=1}^{N_s} f_i$ is the number of remaining particles in the configuration, which is a random variable that follows the binomial distribution:

$$\Pr(N = N) = \binom{N_s}{N}(1-p)^N p^{N_s-N}, \quad (3.16)$$

and its expectation value is $\langle N \rangle_f = (1-p)N_s$.

One can alternatively express the expectation in (3.15) as

$$\left\langle \frac{1}{N} \sum_{i \neq j=1}^{N_s} f_if_j e^{-ik(r_i-r_j)} \right\rangle_f = \sum_{N=0}^{N_s} \Pr(N = N) \frac{1}{N} \sum_{i \neq j=1}^{N_s} \langle f_if_j \rangle_N e^{-ik(r_i-r_j)} \quad (3.17)$$

$$= (1-p) \frac{1}{N_s} \sum_{i \neq j=1}^{N_s} e^{-ik(r_i-r_j)}, \quad (3.18)$$

where the conditional expectation value of $f_if_j$ when $N = N$ is given by

$$\langle f_if_j \rangle_N = \frac{N(N-1)}{N_s^2} = \left( \frac{N}{N_s} \right)^2 + O(N_s^{-1}), \quad (3.19)$$

where $O(f(x))$ means that its value is smaller than $a f(x)$ for some positive constant $a$ as $x$ increases. Substituting Eqs. (3.16) and (3.19) into Eq. (3.17) yields the result (3.18), and thus, the structure factor (3.15) of the defective point process in any space dimension is given by

$$S(k) = 1 + (1-p) \frac{1}{N_s} \left\langle \sum_{i \neq j=1}^{N_s} e^{ik(r_i-r_j)} \right\rangle = 1 + (1-p)(S_0(k) - 1)$$

$$= p + (1-p) S_0(k). \quad (3.20)$$

This result was previously known for defective crystals [56, 229].

We note that Eq. (3.20) is valid for any original point configuration, whether it is hyperuniform or not. Formula (3.20) implies that the rescaled the structure factor as
\[ S(k) - p / (1 - p) \] yields the structure factor \( S_0(k) \) of the original system, regardless of the vacancy concentration \( p \). Using this rescaling idea in insets in Fig. 3.2, we show that numerical simulations are in excellent agreement with our theoretical prediction (3.20). Application of the effective hyperuniformity criterion (3.1) to the imperfect disordered systems described in Fig. 3.2 (b) shows that their hyperuniformity metric \( H \) is of the same order of \( p \) (i.e., \( H_{\text{vacancies}} \sim p \)), and thus they cannot be regarded as effectively hyperuniform whenever \( p > 10^{-4} \).

Using Eqs. (3.20) and (3.11), the local number variance \( \sigma^2_N(R; p) \) of a point process in \( \mathbb{R}^d \) with the vacancy concentration \( p \) is straightforwardly obtained as

\[
\sigma^2_N(R; p) = (1 - p)p \rho v_1(R) + (1 - p)^2 \sigma^2_N(R; 0) ,
\]

(3.21)

where \( \rho \) is number density of the initial point process. Note that the first term in Eq. (3.21) corresponds to variance in the number of vacancies inside a spherical window. Thus, the nonhyperuniformity due to uncorrelated point vacancies is attributed to the tendency of vacancies to cluster, as in Poisson point processes. This result is consistent with an expression derived by Chieco et al. [47] for the related volume-fraction variance of two-dimensional square lattices with uncorrelated vacancies.

Remarks:

1. In the limit of \( p \to 1 \), \( S(k) \to 1 \), which implies that the system behaves like the ideal gas regardless of the initial configuration.

2. To treat the case of substitutional impurities, one need to replace the random function \( f(r) \) in Eq. (3.15) with the following expression;

\[
[1 - f(r)] a_0(k) + f(r) a_s(k) ,
\]

(3.22)

where \( a_0(k) \) and \( a_s(k) \) are the scattering amplitude of a dominant atom and
an impurity atom, respectively. Here, \( f(r) \) is identical to the random function (3.14), but now its expectation value \( p \) stands for the concentration of the substitutional impurities.

3. In Fig. 3.2, we generate 100 independent initial configurations of \( N_s = 10^4 \) for each type of two-dimensional systems, which correspond to the case \( p = 0 \). In order to generate stealthy hyperuniform systems, we use the collective coordinate optimization technique described in Refs. [321] and [352]. From these initial configurations, for a given \( p(\neq 0) \), we generate 10000 defective configurations by randomly removing \( n \) particles, where \( n \) is a random variable following the Poisson distribution with a mean \( pN_s \). In Fig. 3.2(d), \( S(0) \) represents an average of \( S(k) \) at the three lowest wavenumbers.

### 3.3.2 Point interstitials

![Graphs showing comparison of predictions and numerical simulations](image)

Figure 3.3: Left panel: Comparison of the predictions of Eq. (3.25) to the corresponding computer simulations of \( S(0) \) of the defective configurations with the fraction \( p \) of interstitials. Right panel: Semi-log plot of numerical results for the structure factor of defective point configurations with uncorrelated interstitials. The original perfectly hyperuniform systems are taken to be one-dimensional stealthy hyperuniform ground states with \( \chi = 0.3 \) and \( K = 1 \).

Here, we consider spatially uncorrelated interstitials by independently adding particles in original perfectly hyperuniform configurations in \( \mathbb{R}^d \). Suppose that the orig-
inal configuration has $N_s$ particles at number density $\rho$, and $pN_s$ interstitials are introduced into the configuration. Thus, number density of a defective system becomes $(1 + p)\rho$. Separating the collective coordinate $\tilde{n}(k)$ of a defective system into those of the original system and the interstitials, $\tilde{n}_0(k)$ and $\tilde{n}_I(k)$, the structure factor of the defective system can be written as

\begin{align}
S(k) &= \frac{1}{(1 + p)N_s} \left\langle |\tilde{n}_0(k) + \tilde{n}_I(k)|^2 \right\rangle \approx \frac{1}{(1 + p)N_s} \left[ \left\langle |\tilde{n}_0(k)|^2 \right\rangle + \left\langle |\tilde{n}_I(k)|^2 \right\rangle \right] \\
&= \frac{1}{1 + p} S_0(k) + \frac{p}{1 + p} S_I(k),
\end{align}

where an approximation $\left\langle \tilde{n}_0(k) \tilde{n}_I(-k) \right\rangle \approx 0$ is used under the assumption that interstitials are uncorrelated with respect to the original systems. Here, the structure factor of interstitials is denoted by $S_I(k) \equiv \left\langle |\tilde{n}_I(k)|^2 \right\rangle / (pN_s)$.

When the interstitial positions are completely uncorrelated (i.e., $S_I(k) \to 1$), Eq. (3.24) can be simplified as

\begin{equation}
S(k) = \frac{p}{1 + p} + \frac{1}{1 + p} S_0(k).
\end{equation}

We note that as like Eq. (3.20), Eq. (3.25) is valid for any original configuration whether it is hyperuniform or not, and in any space dimension $d$. For instance, if an original configuration is a Poisson point configuration [$S_0(k) = 1$], then the defective configuration obviously becomes another Poisson point configuration at a different number density, i.e., $S(k) = S_0(k)$.

For computer simulations, we consider only in one-dimension for simplicity; corresponding results for higher dimensions will not change qualitatively. Figure 3.3 compares the predictions of Eq. (3.25) to numerical simulations of one-dimensional (entropically-favored) stealthy hyperuniform ground states [352, 317] with $\chi = 0.3$, $K = 1$, and $N_s = 10^3$. For each of 100 original configurations, we generated 100 de-
fective configurations by randomly adding \( m \) particles, where \( m \) is a random number chosen from the Poisson distribution with a mean \( pN_s \). According to the effective hyperuniformity criterion (3.1), the imperfect disordered systems described in Fig. 3.3 are definitively not hyperuniform \( (H_{\text{interstitials}} \sim p > 10^{-4}) \). Figure 3.3 shows that Eq. (3.25) can provide a good approximation, even for large values of \( p \).

Using Eqs. (3.11) and (3.25), one obtains an expression for the local number variance in the presence of uncorrelated interstitials as follows:

\[
\sigma_N^2(R; p) = p\rho v_1(R) + \sigma_N^2(R; 0),
\]

where \( p \) is the fraction of interstitials and \( \rho \) is the number density of the original system. Since the first term in Eq. (3.26) corresponds to the number variance of a Poisson point process at number density \( p\rho \), the nonhyperuniformity of the defective systems is attributed to the tendency of the defects to be clustered, which also occurs in a Poisson point process.

### 3.4 Effect of Stochastic Particle Displacements

We consider a perturbed point process, in which the position of \( i \)th particle in an initial point process is stochastically displaced from \( r_i \) to \( r_i + u(r_i) \), where \( i = 1, 2, \ldots \). When the initial point process is a lattice, the perturbed system is referred to a perturbed lattice [330, 85] (also known as ‘shuffled lattice’ [86, 313]). Perturbed lattices have been studied or used in various contexts, including models of lattice disorders [330, 279], and subjects in probability theory, such as the distribution of zeros of random entire functions [276] and number rigidity [231, 95]. Moreover, perturbed lattices are used to generate disordered initial configurations for numerical simulations [68], configurations of sampling points [244], and disordered hyperuniform point configurations [222, 175]. Here, we start with a summary of the previous studies.
3.4.1 General properties of Perturbed Point Processes

In the rest of this section, we consider a hyperuniform point process \( \{ r_i \}_{i=1}^{\infty} \) at number density \( \rho \) in \( \mathbb{R}^d \), and its structure factor and pair-correlation function are denoted by \( S_0(k) \) and \( g_2^{(0)}(r) \), respectively. For the simplicity, in the rest of this section, we assume that the stochastic displacement vectors \( u(r) \) follow an identical and isotropic singlet probability density function \( f_1(u) \), and thus \( \langle u \rangle = 0 \).

The structure factor of a perturbed point process depends on the initial point process and statistical properties of displacements [85, 331]:

\[
S(k) = 1 + \rho \int dr \, e^{-ik \cdot r} g_2^{(0)}(r) \hat{\phi}(k; r),
\]

(3.27)

where \( \hat{\phi}(k; r) \equiv \int du \, dv \exp(-ik \cdot (u - v)) f_2(u, v; r) \), and \( f_2(u, v; r) \) stands for a conditional joint probability density function that two particles, separated by \( r \) in the initial point process, are displaced by \( u \) and \( v \), respectively.

If displacements of distinct particles are uncorrelated, then the joint probability density function can be reduced into a product of two singlet probability densities \( f_1(u) \) and \( f_1(v) \), i.e., \( f_2(u, v; r) = f_1(u) f_1(v) \), where \( f_1(u) \) is the singlet probability density function of a displacement vector \( u \). Thus, the structure factor (3.27) of a perturbed point process with uncorrelated displacements is simply expressed by [85, 90]

\[
S(k) = \left( 1 - |\tilde{f}_1(k)|^2 \right) + \left| \hat{f}_1(k) \right|^2 S_0(k),
\]

(3.28)

where \( \hat{\phi}(k; r) = |\tilde{f}_1(k)|^2 \) and \( \tilde{f}_1(k) \) is the characteristic function, or equivalently the Fourier transform, of \( f_1(u) \). In contrast to the effect of point defects described in Sec. 3.3, “uncorrelated” displacements cannot destroy the hyperuniformity of an original hyperuniform point process because \( \tilde{f}_1(k = 0) = 1 \) by definition.

Suppose that the singlet probability density can be approximated by \( f_1(u) \approx A|u|^{-(d+\gamma)} \) for large \( |u| \), and thus the small-wavenumber behavior of \( \tilde{f}_1(k) \) depends...
on the exponent $\gamma$ \cite{85};

\[ \tilde{f}_1(k) \approx 1 - B_\gamma |k|^{\min\{2, \gamma\}} \quad (|k| \ll 1), \tag{3.29} \]

where $\langle u \rangle = 0$, $\min C$ represents the smallest element of a set $C$, and the coefficient $B_\gamma$ is written as

\[ B_\gamma = \begin{cases} 
\frac{1}{2d} \langle |u|^2 \rangle, & \gamma > 2 \\
A(2\pi)^{d/2} \int_0^\infty dx \frac{J_{d/2-1}(x)}{x^{d/2+\gamma}}, & 0 < \gamma \leq 2.
\end{cases} \tag{3.30} \]

If $S_0(k) \sim |k|^\alpha$ for small $|k|$, then the structure factor (3.28) of a perturbed point process is approximately given as

\[ S(k) \sim |k|^{\min\{2, \gamma, \alpha\}} \quad (|k| \ll 1). \tag{3.31} \]

Thus, an uncorrelated perturbed point process can belong to any class of hyperuniformity, i.e., classes I, II, and III \cite{300}, as long as the growth rate of its local number variance $\sigma_N^2(R)$ is faster than or equal to that of the original hyperuniform system.

For instance of one-dimensional perturbed lattice, if $f_1(u)$ has a finite variance, e.g., Gaussian distribution, the system always belongs to class I. Using the Cauchy distribution ($\gamma = 1$) \footnote{The Cauchy probability distribution function is given by $f_1(x; \delta, \mu) = \frac{1}{\pi \delta(1 + ((x - \mu)/\delta)^2)}$, where $\delta \in (0, \infty)$ is a length scale and $\mu \in \mathbb{R}$ is the mean. For more details, see A. Papoulis, \textit{Probability, Random Variables, and Stochastic Processes}, 2nd ed. (McGraw-Hill, 1984).} and the Pareto distribution ($\gamma < 1$) \footnote{The Pareto probability distribution function has semi-infinite support, i.e., $x \in [0, \infty)$ and is given by $f_1(x; \alpha, \alpha) = \Theta(x - x_m) \alpha(x_m/x)^{\alpha+1}/x_m$, where $\Theta(x)$ is the Heaviside step function and $\alpha \in (0, \infty)$ is an exponent. For more details, see B.C. Arnold, \textit{Pareto distribution}, 2nd ed. (CRC Press, 2015).} as $f_1(u)$, one obtains class II \footnote{Gabrielli demonstrated class II perturbed lattices for $d = 1$ via the Cauchy distribution; see Ref. \cite{85}.} and class III perturbed lattices, respectively. However, it is impossible to change a class II system ($\alpha = 1$) to a class I system ($\alpha > 1$) via the uncorrelated
stochastic displacements.

Now we consider cases of correlated displacements. Assuming that a displacement vector $\mathbf{u}$ is isotropically distributed and its variance $\langle |\mathbf{u}|^2 \rangle$ exists, $\hat{\phi}(\mathbf{k}; \mathbf{r})$ in Eq. (3.27) can be expanded as a Taylor series of $\mathbf{k}$ for small $|\mathbf{k}|$ [85]:

$$
\hat{\phi}(\mathbf{k}; \mathbf{r}) = 1 + \sum_{\mu, \nu=1}^d k_\mu k_\nu [G_{\mu\nu}(\mathbf{r}) - G_{\mu\nu}(0)] + O(|\mathbf{k}|^4),
$$

(3.32)

where the displacement-displacement correlation function is defined as $G_{\mu\nu}(\mathbf{r}) \equiv \langle u_\mu(\mathbf{r} + \mathbf{r}_0) u_\nu(\mathbf{r}_0) \rangle$. For the simplicity, if we assume that two orthogonal components of displacements are uncorrelated when $d \geq 2$, which implies that $G_{\mu\nu}(\mathbf{r}) = \delta_{\mu\nu} G(\mathbf{r})$, then the general expression for the structure factor (3.27) can be approximated by for small wavenumbers [85]:

$$
S(\mathbf{k}) \approx \left[ |\mathbf{k}|^2 G(\mathbf{0}) + (1 - |\mathbf{k}|^2 G(\mathbf{0})) S_0(\mathbf{k}) \right] + \rho |\mathbf{k}|^2 \left( \tilde{G}(\mathbf{k}) + \int d\mathbf{r} h_0(\mathbf{r}) G(\mathbf{r}) e^{-i\mathbf{k}\cdot\mathbf{r}} \right),
$$

(3.33)

where $\tilde{G}(\mathbf{k})$ is the Fourier transform of $G(\mathbf{r})$. Note that the terms in the square brackets in Eq. (3.33) are contributions from the individual displacements (i.e., $G(\mathbf{0})$), and these terms are identical to the right-hand side of Eq. (3.28) for small $|\mathbf{k}|$ because $|\mathbf{k}|^2 G(\mathbf{0}) = |\mathbf{k}|^2 \langle |\mathbf{u}|^2 \rangle / d \approx 1 - |\tilde{f}_1(\mathbf{k})|^2$ in the same regime. The rest terms in Eq. (3.33) are contributions from “correlations” in displacements.

**Remarks:**

1. Correlated perturbed lattices are different from uncorrelated ones in two respects: (a) Bragg peaks and (b) existence of nonhyperuniform states. Both types of perturbed lattices have Bragg peaks centered at the same positions as their progenitor lattices. While correlated perturbed lattices have broadened peaks as thermalized crystals do, uncorrelated ones have peaks that are not
broadened but weakened compared to those in the initial lattice.

2. In contrast to uncorrelated perturbed lattices that are always hyperuniform, correlated ones can be nonhyperuniform. Suppose that for a perturbed lattice in $\mathbb{R}^d$, the function $\tilde{G}(k)$, which is given in Eq. (3.33), exhibits the power-law behavior $\tilde{G}(k) \sim |k|^{\beta}$ for small $|k|$. It follows that the perturbed lattice is no longer hyperuniform when $\beta = -2$, i.e.,

$$\tilde{G}(k) \sim |k|^{-2} \quad (|k| \ll 1),$$

(3.34)

because the term $|k|^2 \tilde{G}(k)$ in Eq. (3.33) converges to a positive constant as $|k| \to 0$. For low dimensional perturbed lattices ($d \leq 2$), this condition implies that the variance $\langle |u|^2 \rangle$ becomes infinite.

3.4.2 Class III hyperuniformity

Class III (hyperuniform) point processes are characterized by the exponent $0 < \alpha < 1$ in the large-$R$ behavior of the number variance $\sigma_N^2(R)$, or equivalently, in the small-wavenumber behavior of $S(k)$; see Eq. (3.12). A few class III systems have been reported, e.g., critical absorbing states of random organization models ($\alpha \approx 0.45$ ($d = 1$) and $0.425$ ($d = 2$)) [122] and some classical ground states generated by the collective coordinates optimization techniques [347, 321]. However, as noted in Sec. 3.4.1, one can generate class III perturbed lattices with $0 < \alpha < 1$. Moreover, when constructing class III systems, use of uncorrelated perturbed lattices is computationally advantageous over other methods, e.g., the collective coordinate optimization technique [321, 347] and random organization models [122, 288], for two reasons: (a) parallelization is straightforward, and (b) the computational cost is in the order of particle number $N$.

To generate $d$-dimensional class III perturbed lattices, the singlet probability den-
Figure 3.4: Log-log plots of approximation and numerical results for structure factors of perturbed point configurations, which are generated by stochastically displacing each particle by the singlet distribution $f_1(u; \delta, \alpha)$ defined in Eq. (3.35). Initial configurations include the following: (a) the integer lattice for $d = 1$, (b) the square lattice for $d = 2$, and (c)-(d) stealthy disordered hyperuniform point configurations with $\chi = 0.1$ ($d = 1, 2$ for (c) and (d), respectively). A black solid line in each panel shows $S_0(k)$ of the initial configurations.

The density should have a power-law tail, i.e., $f_1(u) \sim |u|^{-(d+\alpha)}$ as $|u| \to \infty$ and $0 < \alpha < 1$ [85, 88]. The $\alpha$-stable distributions [221] are one-dimensional examples of such singlet densities. However, it is difficult to implement them since they can only be analytically expressed in terms of their characteristic functions.

Here, we will present one of the simplest singlet density functions to generate class
III perturbed lattices in $\mathbb{R}^d$:

$$f_1(\mathbf{r}; \delta, \alpha) \equiv \begin{cases} K(d, \alpha, \delta), & |\mathbf{r}| \leq \delta \\ K(d, \alpha, \delta) \left(|\mathbf{r}|/\delta\right)^{-d-\alpha}, & |\mathbf{r}| > \delta, \end{cases} \quad (3.35)$$

where the normalization constant is given by $K(d, \alpha, \delta) = \frac{\Gamma(1 + d/2)\alpha}{\pi^{d/2}(d + \alpha)\delta^{d}},$ and two parameters: an exponent $\alpha \in (0, 1)$ and a characteristic length scale $\delta \in (0, \infty)$. Expressions for the cumulative distribution, its inverse and the characteristic function of $f_1(\mathbf{r}; \delta, \alpha)$ are provided in Appendix 3.7. We note that this method can be applied to any class I system to obtain class III systems; see Fig. 3.4(c) and (d) for examples.

Substituting the characteristic function (3.63) into Eq. (3.28), we obtain the small-$|\mathbf{k}|$ asymptotic behavior of $S(\mathbf{k})$ for an uncorrelated perturbed point process via the singlet density (3.35):

$$S(\mathbf{k}) = 2 \, A(d, \alpha) \left(\frac{k\delta}{2}\right)^{\alpha} + \mathcal{O}(k^{\min(2\alpha, 2)}) \quad (|k\delta| \ll 1), \quad (3.36)$$

where $k \equiv |\mathbf{k}|$ and $A(d, \alpha)$ is defined as

$$A(d, \alpha) \equiv \frac{\Gamma(1 + d/2)\Gamma(1 - \alpha/2)}{\Gamma(1 + (d + \alpha)/2)}. \quad (3.37)$$

As shown in Fig. 3.4, our approximation formula (3.36) is in excellent agreement with numerical simulations of $S(\mathbf{k})$.

Large system size is necessary to observe the class III behavior in $S(\mathbf{k})$ because this behavior results from large displacements. Suppose that an initial point configuration lay in a $d$-dimensional periodic hypercubic box of side length $L$. Then, one can estimate the relative error $\epsilon$ in the approximation (3.36) at the lowest wavenumber $k = 2\pi/L$ by comparing terms in series expansion of $S(\mathbf{k})$ about $k = 0$. We find the
lower bound \( L_{\text{min}} \) of system size as follows:

\[
L > L_{\text{min}} \equiv \frac{2\pi}{k} = \pi \delta \left( \frac{A(d, \alpha)}{2\epsilon} \right)^{1/\alpha},
\]  

(3.38)

ignoring statistical uncertainties that also increase as wavenumber decreases.

**Remarks:**

1. The singlet function (3.35) can be used to generate class II perturbed lattices by setting \( \alpha = 1 \).

2. We generate 100 initial configurations at unit number density for each system. Initial point configurations have particles of \( N = 2 \times 10^3 (d = 1) \) or \( N = 10^4 (d = 2) \). Subsequently, we generate \( 10^2 \) perturbed configurations from each initial configuration via a singlet function \( f_1(u; \delta, \alpha) \), given in Eq. (3.35), using parameters in Fig. 3.4. The computed structure factors are presented in Fig. 3.4.

3. Taking the relative error to be \( \epsilon = 10^{-3} \), some lower bounds of system size \( L_{\text{min}} \), calculated from Eq. (3.38), are listed in Table 3.1.

<table>
<thead>
<tr>
<th>( d = 1, \delta = 10^{-3}, L = 2 \times 10^3 )</th>
<th>( \alpha = 0.8 )</th>
<th>( \alpha = 0.6 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( d = 1, \delta = 10^{-3}, L = 2 \times 10^3 )</td>
<td>( 1.1 \times 10 )</td>
<td>( 1.4 \times 10^2 )</td>
</tr>
<tr>
<td>( d = 2, \delta = 10^{-4}, L = 10^2 )</td>
<td>( 9.3 \times 10^{-1} )</td>
<td>( 1.2 \times 10 )</td>
</tr>
</tbody>
</table>

### 3.5 Effect of Phonon Modes

Previously, Torquato *et al.* [317] theoretically and numerically showed that disordered stealthy hyperuniform ground states become nonhyperuniform in proportion to the temperature (i.e., \( S(0) \propto T \)) for sufficiently low \( T \). In this study, however, the
small-wavenumber behavior of \( S(\mathbf{k}) \) was not obtained, and the theoretical prediction was derived from the compressibility relation (3.13) without considering the underlying mechanisms. In this section, we investigate the mechanisms associated with thermal excitations that destroy the hyperuniformity of ground states, whether they are disordered or not, and obtain a corresponding predictive formula for the structure factor \( S(\mathbf{k}) \) for small wavenumbers. For simplicity, we focus on thermal excitations (phonon modes) in classical crystalline solids for sufficiently low temperatures.

Consider a ground-state crystal at \( T = 0 \), which is necessarily hyperuniform and indeed stealthy [313, 317], meaning that \( S(\mathbf{k}) = 0 \) up to the first Bragg-peak wavenumber. Now, imagine gradually increasing the temperature. One might surmise that each particle symmetrically moves around its equilibrium position, and thus the averaged positions of the particles over a long period of time would be identical to the ideal crystalline structure. This scenario would lead one to falsely conclude that thermalized crystals (crystals in thermal equilibrium) are hyperuniform.

![Figure 3.5: Schematics illustrating particle displacements in Einstein (left) and Debye solids (right) at a positive temperature. For illustrative purposes, displacements are exaggerated. The particles (large blue dots) in an Einstein solid ‘independently’ experience harmonic restoring interactions (blue line) toward their equilibrium positions. By contrast, the particles in a Debye solid interact with their nearest neighbors. In summary, displacements in Einstein and Debye solids are uncorrelated and correlated, respectively, leading to different behaviors in long-wavelength density fluctuations.](image-url)
Thermalized crystals have been extensively investigated in fields of solid-state physics and crystallography. In solid-state physics, there are two important (quantum mechanical) models for harmonic crystals, i.e., Einstein and Debye solids; see Fig. 3.5. Roughly speaking, in an Einstein solid, constituent particles behave as independent harmonic oscillators. In a Debye solid, collective motions of the particles arise as elastic waves, each mode of which behaves like an independent oscillator, called a phonon. It is noteworthy that Einstein and Debye solids can be mapped to uncorrelated and correlated perturbed lattices, respectively. Thus, an Einstein solid, which corresponds to the aforementioned scenario, cannot be nonhyperuniform at a positive temperature, and we focus on Debye solids.

In crystallography, it has been known that thermally excited elastic waves (phonons) in crystals cause background scattering, called thermal diffuse scattering. In the past, this subject had been extensively studied by utilizing approximations for \( \hat{\phi}(k; r) \), which is given in Eq. (3.27). For instance, the well-known Debye-Waller factor [7], \( \exp(-\langle (q \cdot u)^2 \rangle) \) is essentially a higher-order expression of the quantity \( 1 - \sum_{\mu,\nu=1}^{d} k_{\mu} k_{\nu} G_{\mu\nu}(0) \) in Eq. (3.32). However, heretofore, a quantitative description of the small-wavenumber behavior of the structure factor of a thermalized crystal in the harmonic regime has been lacking. This is partly because previous studies have mainly focused on diffuse scattering near Bragg peaks rather than small-wavenumber behavior. Furthermore, many previous theoretical predictions become invalid for low-dimensional crystals \( (d \leq 2) \) because they contain the Debye-Waller factor that vanishes at any positive temperature for these cases [131, 7].

Our objective here is to derive an explicit expression for the structure factor of classical Debye solids within the harmonic regime, especially in the vicinity of \( k = 0 \). In order to formulate a predictive theory for low dimensions, we avoid starting from standard formulas with the accompanying Debye-Waller factor.
3.5.1 Simple harmonic lattices

In the harmonic approximation, the potential energy $\Phi(\{R\})$ of crystalline solids can be described as [7, 143, 42]:

$$\Phi(\{R\}) \approx \frac{1}{2} \sum_{R,R'} u_\mu(R) D_{\mu\nu}(R-R') u_\nu(R'),$$

where $u_\nu(R)$ represents the $\nu$-component of displacement vector $u(R)$ of a particle whose equilibrium position is $R$, and $D_{\mu\nu}(R)$ is called the dynamical matrix.

Suppose a finite subset $\Lambda(L)$ of an infinitely large Bravais lattice in $\mathbb{R}^d$, which contains $N = L^d$ particles. Under periodic boundary conditions, motion of a particle at $R$ can be described by a superposition of normal modes $\tilde{u}_s(q,t)e^{iq\cdot R}$, i.e.,

$$u(R,t) = \frac{1}{\sqrt{N}} \sum_{q,s} \tilde{u}_s(q,t) e^{iq\cdot R},$$

(3.39)

where a normal coordinate $\tilde{u}_s(q,t) \equiv \tilde{u}_s(q) \tilde{\epsilon}_s(q)e^{i\omega_s(q)t}$ represents elastic wave characterized by a wavevector $q$, polarization $\tilde{\epsilon}_s(q)$, and angular frequency $\omega_s(q)$. We use the shorthand notation $\sum_{q,s} \equiv \sum_{q \in \Lambda_1^\ast(L)} \sum_{s=1}^d$, where $\Lambda_1^\ast(L)$ denotes the first Brillouin zone of $\Lambda(L)$. Note that a normal coordinate is essentially a spatial Fourier component of $u(R,t)$.

The quantities $\tilde{\epsilon}_s(q)$ and $\omega_s(q)$ can be determined by solving the following eigenvalue problem:

$$m \omega_s^2(q) \tilde{\epsilon}_s^\nu(q) = \sum_{\nu=1}^d \tilde{D}_{\mu\nu}(q) \tilde{\epsilon}_s^\mu(q),$$

(3.40)

where $\tilde{\epsilon}_s^\nu(q)$ is the $\nu$-component of $\tilde{\epsilon}_s(q)$, $m$ is the mass of a single particle and $\tilde{D}_{\mu\nu}(q)$ is the Fourier transform of the dynamical matrix $D_{\mu\nu}(R)$. Thus, for each wavevector $q$, there are $d$ independent normal modes, and their polarization vectors $\tilde{\epsilon}_s(q)$ satisfy the orthogonality (3.41) and closure (3.42) relations [143]:

$$\tilde{\epsilon}_s(q) \cdot \tilde{\epsilon}_{s'}(q) = \delta_{s,s'},$$

(3.41)

$$\sum_{s=1}^d \tilde{\epsilon}_s^\mu(q) \tilde{\epsilon}_s^\nu(q) = \delta_{\mu,\nu},$$

(3.42)
where $\delta_{s,s'}$ is the Kronecker delta symbol. Using the normal coordinates (3.39) and the relation (3.40), total energy $E$ of a harmonic crystal can be decomposed into a sum of elastic and kinetic energy of each normal mode:

$$E = \sum_{q,s} \left[ \frac{1}{2} m \omega_s^2(q) \left| \tilde{u}_s(q,t) \right|^2 + \frac{1}{2} m \left| \frac{\partial \tilde{u}_s(q,t)}{\partial t} \right|^2 \right]. \quad (3.43)$$

In Sec. 3.5, we will consider a $d$-dimensional simple cubic lattice where each particle is connected to its nearest neighbors by springs of spring constant $K$. The potential energy of this system is approximately given as

$$\Phi_{\text{cubic}} \approx \frac{K}{2} \sum_{\langle \mathbf{R}, \mathbf{R}' \rangle} |\mathbf{u}(\mathbf{R}) - \mathbf{u}(\mathbf{R}')|^2, \quad (3.44)$$

where $\langle \mathbf{R}, \mathbf{R}' \rangle$ indicates that two sites $\mathbf{R}$ and $\mathbf{R}'$ are nearest neighbors. Its dynamical matrix is given by $D_{\mu\nu}(\mathbf{R}) = \delta_{\mu,\nu} K \sum_{\langle \mathbf{R}',0 \rangle} (\delta_{\mathbf{R},0} - \delta_{\mathbf{R},\mathbf{R}'})$, and its Fourier transform is

$$\tilde{D}_{\mu\nu}(\mathbf{k}) = \delta_{\mu,\nu} 4K \sum_{i=1}^{d} \sin^2(k_i a/2), \quad (3.45)$$

where $a$ is the lattice constant. Using (3.40) and (3.45), one can obtain the degenerate dispersion relations:

$$\omega^2(\mathbf{k}) = \frac{4K}{m} \sum_{i=1}^{d} \sin^2(k_i a/2), \quad (3.46)$$

regardless of polarization $s$. For small $|\mathbf{k}|$, one obtains a linear dispersion relation:

$$\omega(\mathbf{k}) = c|\mathbf{k}| + \mathcal{O}\left(|\mathbf{k}|^2\right), \quad (3.47)$$

where $c$ is the sound speed in the continuum limit ($|\mathbf{k}| \to 0$), given by $c \equiv \sqrt{K a^2/m}$. Here, the speed sound $c$ is independent of the polarization, but in general, it depends on the polarization. In Appendix 3.9, we derive an isotropic expression of (3.46) as
follows:
\[ \omega^2(k) \approx (ck)^2 \left( 1 - \frac{(ka)^2}{4(d + 2)} \right). \] (3.48)

### 3.5.2 Static structure factor of thermalized crystals

We will consider a finite subset \( \Lambda(L) \) of a \( d \)-dimensional Bravais lattice of unit lattice constant, as described in Sec. 3.5.1, and assume the classical harmonic interactions. We first present a heuristic derivation of the small-wavenumber behavior of the structure factor at low temperatures \(^5\). The rigorous derivation that leads to the same result is provided in Appendix 3.8.

The collective coordinates can be approximated for small displacements by

\[ \tilde{n}(k) = \sum_U \exp(-i k \cdot R) \exp(-i k \cdot u(R, t)) \approx \sum_U \exp(-i k \cdot R) \left[ 1 - i k \cdot u(R, t) \right] \]

\[ = \left[ \sum_U \exp(-i k \cdot R) \right] - i \sqrt{N} k \cdot \tilde{u}(k, t), \] (3.49)

where we use the fact that normal coordinates \( \tilde{u}(k, t) \) are the Fourier components of displacement vectors \( u(R, t) \). Here, we note that for small \( |k| \), the quantity \( \sum_U \exp(-i k \cdot R) \) is zero in this regime because an ideal lattice is stealthy hyperuniform. Thus, the structure factor of harmonic crystals can be approximately given by

\[ S(k) = \lim_{N \to \infty} \frac{1}{N} \left\langle |\tilde{n}(k)|^2 \right\rangle \approx \left\langle |k \cdot \tilde{u}(k, t)|^2 \right\rangle, \] (3.50)

for \( 0 < |k| \ll 1 \). In fact, for low-dimensional crystals \( (d \leq 2) \), the approximation used in Eq. (3.49) is not justifiable because the quantity \( \left\langle |u|^2 \right\rangle \) diverges in the thermodynamic limit [131]. Nonetheless, it is noteworthy that the result (3.50) is identical to the one rigorously derived in Appendix 3.8.

According to the equipartition theorem, the ensemble average of potential energy

\(^5\)This derivation is similar to that used to derive Huang diffuse scattering in Ref. [16]
Figure 3.6: Semi-log plots of approximate and numerical results for structure factors for thermalized hypercubic lattices calculated from the Monte Carlo technique; (a) $d = 1$, (b) $d = 2$, and (c) $d = 3$. Note that structure factors are normalized by the dimensionless temperature $T$ defined by the relation (3.54). The approximate results are calculated from the formula (3.53). Insets in each of panels are magnifications in the small-wavenumber regime.

(3.43) of a normal mode with a polarization index $s$ is expressed as

$$\frac{1}{2} m \omega_s^2(k) \left\langle |\tilde{u}_s(k,t)|^2 \right\rangle = \frac{1}{2} k_B T, \quad s = \|, 2, \ldots, d, \quad (3.51)$$

where the index $\|$ indicates the longitudinal polarization at a given wavevector $k$ and $m$ is the mass of a single particle. Thus, the expression (3.50) can be simplified as

$$S(k) = \frac{|k|^2}{m \omega_{\|}^2(k)} k_B T. \quad (3.52)$$

For the hypercubic model described in Sec. 3.5.1, we obtain the small-wavenumber expression for the isotropic structure factor $S(|k|)$ by substituting the dispersion relation (3.48) into Eq. (3.52):

$$S(|k|) = T \left[ 1 + \frac{(|k|a)^2}{4(d+2)} \right] + O(|k|^4), \quad (3.53)$$

where $a$ is the lattice constant and $T$ is a dimensionless temperature, defined as

$$T \equiv k_B T/(me^2). \quad (3.54)$$
Our approximate result (3.53) is consistent with numerical simulations for spatial dimensions \(d = 1, 2, 3\): see Fig. 3.6. Taking the limit \(|\mathbf{k}| \to 0\) in Eq. (3.53) and comparing to the compressibility relation (3.13) enables us to determine the isothermal compressibility explicitly:

\[
\kappa_T = (Ka^{2-d})^{-1}, \tag{3.55}
\]

which is identical to the inverse of the bulk modulus for the corresponding spring networks.

Expression (3.50) implies that only sound waves (longitudinal elastic waves) contribute to long-wavelength density fluctuations or nonhyperuniformity of thermalized crystals. It is reasonable because while sound waves cause density modulations, transverse waves result in volume-preserving shear deformations. In addition, the predicted hyperuniformity of an “incompressible” system in thermal equilibrium would result from a non-relativistic (infinite) speed of sound. The reader is referred to a recent study on a perfect glass model that is hyperuniform and has the same attribute of a non-relativistic (infinite) speed of sound [354].

For a non-Bravais crystal of \(N_b\) basis atoms, there are \(N_b\) independent “longitudinal” normal modes at each wavevector \(\mathbf{k}\) [7], and thus Eq. (3.52) will be modified as

\[
S(\mathbf{k}) = N_b \sum_{i=1}^{N_b} \frac{|\mathbf{k}|^2}{\mu_i \omega_{\parallel(i)}^2(\mathbf{k})} k_B T, \tag{3.56}
\]

where \(\mu_i\) are some finite constants of mass unit, and \(\omega_{\parallel(i)}(\mathbf{k})\) is the angular frequency of \(i\)th longitudinal normal mode. In the acoustic mode \((i = 1)\), all basis atoms in the same unit cell move in phase, while in optical modes \((i = 2, \cdots, N_b)\), the basis atoms move out of phase. For small wavenumbers, only acoustic modes have linear dispersion relations, as in (3.47), while optical modes have non-linear ones, i.e., \(\lim_{|\mathbf{k}| \to 0} \omega_{\parallel(i)}(\mathbf{k}) \neq 0\). Therefore, in the limit of \(|\mathbf{k}| \to 0\), it is only the “longitudinal acoustic modes” that can contribute to the long-wavelength density fluctuations, i.e.,
\[ S(0) = \frac{K_B T}{\mu c_1^2}, \text{ where } c_1 \equiv \lim_{|k| \to 0} \omega_{\|k\|=1}(k) / |k|. \]

Since thermalized crystals at a positive temperature can be mapped to “nonhyperuniform” perturbed lattices, their displacement-displacement correlation function \( G_{\mu\nu}(r) \) satisfies the condition (3.34). Indeed, the simple harmonic crystal model in \( \mathbb{R}^d \) satisfies consistently the condition as follows:

\[ \tilde{G}_{\mu\nu}(k) \approx \delta_{\mu\nu} \frac{T}{|k|^2} \quad (|k| a \ll 1), \quad (3.57) \]

where \( T \) is defined by Eq. (3.54) and a detailed derivation is provided in Appendix 3.10. Here, we note that since \( \langle |u|^2 \rangle / d = G(0) \), the asymptotic relation (3.57) implies that the variance in displacements \( \langle |u|^2 \rangle \) diverge, or equivalently, the Debye-Waller factor [131] vanishes for low dimensional crystals.

**Remarks:**

1. Equilibrium hard-sphere systems in \( \mathbb{R}^3 \) exhibits the same structure factor scaling as in (3.53) as they approach to the FCC jamming point along the stable crystal branch [9]. This behavior is attributed to collective “vibrational” motions due to collisions, which are the hard-sphere analogs of phonons in systems of particles interacting with continuous pair potentials.

2. The selection rule \( \sum_\mathbf{Q} \delta(-\mathbf{k} + \sum_{\mathbf{q},s} \mathbf{q}(z(\mathbf{q}, s) + z'(\mathbf{q}, s)), \mathbf{Q}) \) in Eq. (3.68) can be interpreted as the crystal momentum conservation [7]. Then, the result (3.50) corresponds to the single-phonon scattering.

3. To gain some physical idea of the dimensionless temperature \( T \), we provide estimations of a melting point \( T_M \) and Debye temperature \( T_D \) [7] in the unit of \( T \). To estimate the order of magnitude of \( T_M \), we use the Lindemann criterion [361, 7] that \( \langle |u|^2 \rangle \approx (c_l a)^2 \) near \( T_M \), where \( a \) is the lattice constant and \( c_l \approx 0.1 \). In \( d = 3 \), \( T_M \sim 10^{-2} \) and \( T_D \sim 10^{-3} \), which are consistent with
experimental data for many solids; see Table 3.3. In numerical simulations, temperatures are much lower than the melting point, i.e., $T \leq 0.01 T_M$. We note that for the illustrative purposes, a thermalized lattice in Fig. 3.1 is set to be at an exceedingly high temperature $\bar{T} = 0.05$.

4. We use the simulated-annealing technique to simulate thermalized crystals with the potential energy (3.44). Each simulation starts from the melting point ($\bar{T} = 0.01$) and then employs an exponential cooling schedule to achieve a target temperature. At each temperature in the cooling schedule, we adjust the maximum displacement of trial moves such that the acceptance rate is around a half. We let the systems evolve for $10 \tau_s$ MC cycles before sampling the configurations, and then check whether the acceptance ratio is around half for the next $\tau_{\text{test}}$ MC cycles. Table 3.2 lists the sampling parameters for $d = 1, 2,$ and 3, respectively.

<table>
<thead>
<tr>
<th>$d$</th>
<th>1</th>
<th>2</th>
<th>3</th>
</tr>
</thead>
<tbody>
<tr>
<td>$N$</td>
<td>1000</td>
<td>100$^2$</td>
<td>30$^3$</td>
</tr>
<tr>
<td>$N_{\text{samp}}$</td>
<td>$5 \times 10^4$</td>
<td>$10^4$</td>
<td>$10^3$</td>
</tr>
<tr>
<td>$\tau_s$</td>
<td>$5 \times 10^3$</td>
<td>$10^4$</td>
<td>10</td>
</tr>
<tr>
<td>$\tau_{\text{test}}$</td>
<td>$10^3$</td>
<td>$5 \times 10^2$</td>
<td>$10^2$</td>
</tr>
</tbody>
</table>

5. For a $N \times N$ symmetric positive-definite matrix $A$, its Gaussian integral has the following property [42]:

$$
\langle x_i x_j \rangle \equiv \frac{\int_{\mathbb{R}^N} x_i x_j \exp\left(-\frac{1}{2} \sum_{i,j=1}^N A_{ij} x_i x_j\right) \, dx}{\int_{\mathbb{R}^N} \exp\left(-\frac{1}{2} \sum_{i,j=1}^N A_{ij} x_i x_j\right) \, dx} = (A^{-1})_{ij},
$$

where $A^{-1}$ is the inverse of $A$. Thus, one immediately obtains an expression
equivalent to Eq. (3.57):

\[ \tilde{G}_{\mu\nu}(k) \equiv \langle \tilde{u}_\mu(k, t) \tilde{u}_\nu^*(k, t) \rangle_{th} = (k_B T) \tilde{D}_{\mu\nu}^{-1}(k), \]

where \( \tilde{D}^{-1}(k) \) is the inverse of the Fourier transform \( \tilde{D}(k) \) of the dynamical matrix.

### 3.6 Conclusions and Discussion

In this chapter, we have theoretically and numerically investigated the degree to which hyperuniformity is degraded or destroyed due to imperfections in otherwise perfect hyperuniform point processes. We focused on three types of imperfections, including (1) uncorrelated point defects, (2) stochastic particle displacements, and (3) thermal excitations. We derived explicit formulas for the small-wavenumber behaviors (3.20), (3.25), (3.36), and (3.52) for the structure factors \( S(k) \) and showed that these expressions are consistent with numerical simulations in Figs. 3.2, 3.3, 3.4, and 3.6. These results show that either the violation of the infinite-wavelength criterion (3.8) or changes in the small-\(|k|\) behavior of \( S(k) \) without violating the condition provides an unambiguous means to detect imperfections in otherwise amorphous hyperuniform systems.

Our results show that uncorrelated point defects (vacancies and interstitials) and thermal excitations destroy the hyperuniformity of initial systems. However, stochastic displacements can only destroy it when displacements are strongly correlated as noted in (3.34). Importantly, our result (3.52) also directly demonstrates that thermal excitations can destroy the hyperuniformity of harmonic crystals, which has been indirectly predicted [317, 300] by the compressibility relation (3.13).

Note that these results are consistent with the qualitative arguments that we made concerning the several example configurations shown in Fig. 3.1; specifically, the
seemingly more disordered configuration (d) is hyperuniform, but the others (b), (c), and (d) are not. To explain these counterintuitive results, it was helpful to examine the local number variance $\sigma^2_N(R)$. In the presence of point defects, as we show in Eqs. (3.21) and (3.26), the major contributions in $\sigma^2_N(R)$ come from the variances in the number of point defects contained within a large window. In other words, the tendency of point defects to cluster, as in the Poisson point process, destroys hyperuniformity.

Interestingly, as illustrated in Fig. 3.1(d), uncorrelated stochastic displacements degrade but cannot destroy the hyperuniformity because particles only near a window boundary can fall in and out of the window in an independent manner, i.e., $\sigma^2_N(R) < R^d$ for large $R$. Using this property, we presented a simple method to transform class I systems, such as lattices and disordered stealthy hyperuniform systems, to class III systems, defined in Eq. (3.12), by application of relation (3.35).

By contrast, correlated stochastic displacements can destroy hyperuniformity in the way that particles near the window boundary move in and out of the window simultaneously.⁶ Our results (3.50) and (3.57) show that for thermalized crystals, long-wavelength “longitudinal acoustic waves” arise such correlated displacements; see Fig. 3.1(e).

We have studied the effect of each type of imperfection on the small-wavenumber behavior of structure factors for otherwise perfectly hyperuniform systems. It would be interesting to study how multiple types of imperfections simultaneously affect hyperuniformity. In the reverse direction, it would also be interesting to know the general conditions under which correlated particle displacements in a nonhyperuniform system can lead to a hyperuniform system.

The results in this chapter provides the theoretical underpinnings to study the ef-

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⁶For the same reason, the local number variance for a cubic lattice in $\mathbb{R}^3$, which is clearly a hyperuniform system, can increase faster than window volume when one uses a cubic window that is aligned with the lattice; see J. Kim and S. Torquato, J. Stat. Mech: Theory Exp. 2017, 013402 (2017)
fect of imperfections on the physical properties that depend sensitively on the degree of hyperuniformity of materials. For instance, according to the type of imperfection that is introduced in a “stealthy hyperuniform” system, the system becomes merely “hyperuniform” or even “nonhyperuniform.” Thus, imperfections may influence some exotic physical properties associated with the stealthy hyperuniformity, e.g., complete isotropic photonic bandgaps [80, 79, 82], transparency at high densities [179], negative compressibilities [18] and nearly optimal transport properties [355]. For future study, it would be interesting to investigate the degree to which the presence of imperfections in otherwise hyperuniform systems affect their physical properties. Furthermore, one could explore whether it is possible to continuously modulate the degree of imperfections in a hyperuniform material to achieve desired properties.

3.7 Appendix A: Mathematical Details of the Singlet Density (3.35)

In this appendix, we provide explicit formulas for the cumulative distribution function $C(R)$, its inverse $C^{-1}(y)$, and the characteristic function of the singlet density function (3.35). The cumulative distribution function $C(R; \delta, \alpha)$ is

$$C(R) \equiv \int_{|r|<R} dr \, f_1(r; \delta, \alpha) = \begin{cases} \dfrac{\alpha}{d+\alpha} \left( \dfrac{R}{\delta} \right)^d, & R \leq \delta \\ 1 - \dfrac{d}{d+\alpha} \left( \dfrac{R}{\delta} \right)^{-\alpha}, & R > \delta \end{cases} \quad (3.59)$$

and its inverse function $C^{-1}(y)$ is

$$C^{-1}(y) = \begin{cases} \delta \left( y(1 + d/\alpha) \right)^{1/d}, & 0 < y \leq \alpha/(d + \alpha) \\ \delta \left( (1 - y)(1 + \alpha/d) \right)^{-1/\alpha}, & \alpha/(d + \alpha) \leq y < 1 \end{cases} \quad (3.60)$$
One can generate a random radius followed by the probability density function (3.35) by substituting \( y \) in (3.60) with a uniformly-distributed random number between 0 and 1.

The explicit expression for the characteristic function of Eq. (3.35) is

\[
\tilde{f}_1(k; \delta, \alpha) = -A(d, \alpha) (k\delta/2)^\alpha + \alpha/(d + \alpha) \, _0F_1\left( (1 + d)/2; - (k\delta/2)^2 \right) + d/(d + \alpha) \, _1F_2\left( -\alpha/2; d/2, 1 - \alpha/2; - (k\delta/2)^2 \right),
\]

where \( k = |k| \), \( A(d, \alpha) \) is given in Eq. (3.37), and the generalized hypergeometric function \( _pF_q(\{a\}; \{b\}; x) \) is defined as

\[
_pF_q(\{a\}; \{b\}; x) = \sum_{P=0}^{\infty} \frac{(a_1)_P \cdots (a_p)_P \, x^P}{(b_1)_P \cdots (b_q)_P \, P!},
\]

where \( (a)_P \equiv \Gamma(a + P) / \Gamma(a) \). For small \( k \), Taylor series expansion of Eq. (3.61) is

\[
\tilde{f}_1(k; \delta, \alpha) = 1 - A(d, \alpha) (k\delta/2)^\alpha + O(k^2).
\]

### 3.8 Appendix B: Rigorous Derivation of Eq. (3.50)

Suppose a finite subset \( \Lambda(L) \) of a \( d \)-dimensional infinite large Bravais lattice, as defined in Sec. 3.5.1. Using the Jacobi-Anger expansions

\[
\exp(i a \cos \theta) = \sum_{z=-\infty}^{\infty} J_z(a) \, e^{iz(\theta + \pi/2)}
\]

\[
\exp(i a \sin \theta) = \sum_{z=-\infty}^{\infty} J_z(a) \, e^{iz\theta},
\]
and normal coordinates (3.39), the collective coordinates \( \tilde{n}(k) \) of a thermalized crystal can be written in a Fourier series:

\[
\tilde{n}(k) = \sum_{R} \exp(-ik \cdot (R + u(R, t)))
\]

\[
= \sum_{R} e^{-i k \cdot R} \prod_{q_s} \exp(i k_{q_s} \cos(q \cdot R + \omega_s(q)t)) \exp(i^2 k_{q_s} \sin(q \cdot R + \omega_s(q)t))
\]

\[
= \sum_{R} e^{-i k \cdot R} \prod_{q_s} \sum_{z_{q_s}, z'_{q_s} = -\infty}^{\infty} J_{z_{q_s}}(k_{q_s}) J_{z'_{q_s}}(i k_{q_s}) \exp(i(z_{q_s} + z'_{q_s}) (q \cdot R + \omega_s(q)t) + i z_{q_s} \pi/2)
\]

\[
= \sum_{R} e^{-i k \cdot R} \sum_{\{z(q_s), z'(q_s)\}} \prod_{q_s} \left[ J_{z(q_s)}(k_{q_s}) J_{z'(q_s)}(i k_{q_s}) \right. \\
\left. \exp(i(z(q_s) + z'(q_s)) (q \cdot R + \omega_s(q)t) + i z(q_s) \pi/2) \right],
\]

where \( J_n(x) \) is the Bessel function of order \( n \) and we use the shorthand notation \( k_{q,s} \equiv -k \cdot u_s(q)/\sqrt{N} \). Here, \( \sum_{\{z(q_s), z'(q_s)\}} \) represents a summation over all possible functions \( z(q_s) \) and \( z'(q_s) \) from wavevectors in the first Brillouin zone \( \Lambda_1^* (L) \) and polarization indices to integers. Separating the products of exponential functions in Eq. (3.66), one obtains

\[
\tilde{n}(k) = \left\{ \sum_{\{z(q_s), z'(q_s)\}} \prod_{q_s} J_{z(q_s)}(k_{q_s}) J_{z'(q_s)}(i k_{q_s}) \right. \\
\left. \times \exp\left( i \sum_{q_s} \left\{ [z(q_s) + z'(q_s)] \omega_s(q) t + \frac{\pi}{2} z(q_s) \right\} \right) \right\} \\
\times \sum_{R} \exp\left( i \left\{ -k + \sum_{q_s} q [z(q_s) + z'(q_s)] \right\} \cdot R \right),
\]

\[
= \sum_{\{z(q_s), z'(q_s)\}} \prod_{q_s} J_{z(q_s)}(k_{q_s}) J_{z'(q_s)}(i k_{q_s}) \times N \sum_{Q} \delta \left( -k + \sum_{q_s} q [z(q_s) + z'(q_s)], Q \right) \\
\times \exp\left( i \sum_{q_s} \left\{ [z(q_s) + z'(q_s)] \omega_s(q) t + \frac{\pi}{2} z(q_s) \right\} \right),
\]
where we used an identity \( \sum_{\mathbf{R}} e^{i\mathbf{k} \cdot \mathbf{R}} = N \sum_{\mathbf{Q}} \delta(\mathbf{k}, \mathbf{Q}) \), and \( \sum_{\mathbf{R}} \) and \( \sum_{\mathbf{Q}} \) represent a summation over all lattice vectors \( \mathbf{R} \) and over all reciprocal lattice vectors \( \mathbf{Q} \) of the lattice \( \{ \mathbf{R} \} \), respectively. The Kronecker delta symbol is denoted here by \( \delta(\mathbf{q}, \mathbf{q}') \).

Note that for small \(|\mathbf{k}|\) and \(|\mathbf{q}|\), the arguments in Bessel functions are small, i.e., \( k_{qs} = \mathcal{O}(T/\sqrt{N}) \) so that we can use a power series expansion of the Bessel functions (i.e., \( |J_n(x)| = (x/2)^n/n! + \mathcal{O}(x^{n+2}) \) for small \( x \)) to approximate the product of Bessel functions in Eq. (3.68):

\[
\prod_{\mathbf{q},s} J_{z(q,s)}(k_{qs}) J_{z'(q,s)}(ik_{qs}) \propto |\mathbf{k}|^{\sum_{\mathbf{q},s}(|z(q,s)| + |z'(q,s)|)} (|\mathbf{k}| \to 0). \tag{3.69}
\]

Combining the selection rule \( \sum_{\mathbf{Q}} \delta(-\mathbf{k} + \sum_{\mathbf{q},s} \mathbf{q}(\mathbf{q},s) + z'(\mathbf{q},s), \mathbf{Q}) \) in Eqs. (3.68) and (3.69) implies that the leading order in Eq. (3.69) should be the unity, i.e., \( \sum_{\mathbf{q},s} (|z(\mathbf{q},s)| + |z'(\mathbf{q},s)|) = 1. \) Thus, the collective coordinates can be approximated as

\[
\tilde{n}(\mathbf{k}) \approx N \sum_{s=1}^{d} \left[ (J_1(k_{ks}) i + J_1(ik_{ks})) e^{i\omega_s(\mathbf{k})t} + (J_{-1}(k_{-ks}) (-i) + J_{-1}(ik_{-ks})) e^{-i\omega_s(-\mathbf{k})t} \right]
\approx 2N \sum_{s=1}^{d} \frac{ik_{ks}}{2} e^{i\omega_s(\mathbf{k})t} = -i\sqrt{N} \mathbf{k} \cdot \sum_{s=1}^{d} \tilde{u}_s(\mathbf{k}) e^{i\omega_s(\mathbf{k})t} = -i\sqrt{N} \mathbf{k} \cdot \tilde{\mathbf{u}}(\mathbf{k}, t).
\]

Therefore, the leading-order behavior of the structure factor of a thermalized crystal is written as

\[
S(\mathbf{k}) \equiv \langle S(\mathbf{k}) \rangle = \lim_{N \to \infty} \frac{1}{N} \langle \tilde{n}(\mathbf{k}) \rangle \approx \langle |\mathbf{k} \cdot \tilde{\mathbf{u}}(\mathbf{k}, t)|^2 \rangle \quad (0 < |\mathbf{k}| a \ll 1). \tag{3.70}
\]
In this appendix, we derive an isotropic dispersion relation (3.48). Starting from the second-order power series of the exact dispersion relation (3.46):

\[
\omega^2(k) = c^2 |k|^2 \left( 1 - \frac{(|k|a)^2}{12} \sum_{i=1}^{d} \left( \frac{k_i}{|k|} \right)^4 \right) + O\left(|k|^6\right),
\]

(3.71)

we will obtain its average over the orientations of \( k \). Its orientational average simplifies as

\[
\omega^2(k) = (ck)^2 \left( 1 - d/12 \langle x_1^4 \rangle_{\text{ang}} \right) + O(k^6),
\]

(3.72)

where \( k \equiv |k| \), \( x_i \) is the first component of a unit vector \( x \), \( \langle x_i^4 \rangle_{\text{ang}} \equiv \oint_{|x|=1} dx x_i^4 / S_d(1) \), and \( S_d(1) = 2\pi^{d/2}/\Gamma(d/2) \) is the surface area of \( d \)-dimensional sphere of unit radius.

Here, we use the fact that \( \langle x_i^4 \rangle_{\text{ang}} \) are identical for \( i = 1, \cdots, d \) due to the rotational symmetry.

In \( d \)-dimensional spherical coordinates \( \phi \in [0, 2\pi] \) and \( \theta_j \in [0, \pi] \) for \( j = 1, \cdots, d-2 \), the Cartesian coordinates of a unit vector \( x \) are expressed as

\[
\begin{align*}
x_i &= \begin{cases} 
\cos \phi \prod_{j=1}^{d-2} \sin \theta_j, & i = 1 \\
\sin \phi \prod_{j=1}^{d-2} \sin \theta_j, & i = 2 \\
\cos \theta_{i-2} \prod_{j=1}^{i-3} \sin \theta_j, & i = 3, \cdots, d.
\end{cases}
\end{align*}
\]

(3.73)

The infinitesimal area \( dS \) of the spherical shell of unit radius is written as

\[
dS = d\phi \prod_{i=1}^{d-2} \left[ d\theta_i \sin(\theta_i) \right]^{d-i-1}.
\]

(3.74)

Using the following identity \( \int_0^\pi dx \sin^m x = \sqrt{\pi} \Gamma((m+1)/2) / \Gamma((m+2)/2) \), one
can calculate

\[
\langle x_1^4 \rangle_{\text{ang}} = \frac{1}{S_d(1)} \int_0^{2\pi} d\phi \cos^4 \phi \prod_{i=1}^{d-2} \int_0^\pi d\theta_i \sin(\theta_i)^{d+3-i} = \frac{\Gamma(d/2)}{2\pi^{d/2}} \frac{3\pi}{4} \prod_{i=1}^{d-2} \left[ \sqrt{\pi} \Gamma\left(\frac{d+4-1}{2}\right) \Gamma\left(\frac{d+5-1}{2}\right) \right] = \frac{\Gamma(d/2) 3 \left(\frac{d+2}{2}\right)}{4 \Gamma(2 + d/2)} = \frac{3}{d(d+2)}. \quad (3.75)
\]

Thus, Eq. (3.72) becomes

\[
\omega^2(k) \approx (ck)^2 \left(1 - \frac{(ka)^2}{4(d+2)}\right). \quad (3.76)
\]

3.10 Appendix D: Displacement-Displacement Correlation Functions of Thermalized Crystals

In this appendix, we derive Eq. (3.57) in Sec. 3.5.2 for a classical Debye solid. Since \(G_{\mu\nu}(r)\) is defined for the complex variables as

\[
G_{\mu\nu}(r) \equiv \langle u_\mu(r + R, t) u_\nu^*(R, t) \rangle = \langle \langle u_\mu(r + R, t) u_\nu(R, -t) \rangle_R \rangle,\]

we obtain its Fourier transform by using properties of the autocovariance:

\[
\tilde{G}_{\mu\nu}(k) = \langle \tilde{u}_\mu(k, t) \tilde{u}_\nu(-k, -t) \rangle_t = \langle \tilde{u}_\mu(k, t) \tilde{u}_\nu^*(k, t) \rangle_t, \quad (3.77)
\]

where \(\langle \cdot \rangle_R\) means the average over the positions \(R\) and, \(\langle \cdot \rangle_t\) means a canonical ensemble average.

Using normal coordinates (3.39), Eq. (3.77) simplifies as

\[
\tilde{G}_{\mu\nu}(k) = \sum_{s,s'=1}^{d} \hat{e}_s^\mu(k) \hat{e}^\nu_{s'}(k) \langle \tilde{u}_s(k) \tilde{u}_{s'}^*(k) e^{i(\omega_s(k) - \omega_{s'}(k))t} \rangle_t = \sum_{s=1}^{d} \hat{e}_s^\mu(k) \hat{e}^\nu_s(k) \langle \tilde{u}_s(k) \tilde{u}_{s}^*(k) \rangle_t, \quad (3.78)
\]
where $\hat{\epsilon}_s^\mu(k)$ is the $\mu$-component of a unit polarization vector $\hat{\epsilon}_s(k)$ and we used the fact that normal coordinates with different polarizations are independent of one another.

Applying the equipartition theorem into Eq. (3.78), we obtain the small-\(|k|\) behavior of $\tilde{G}_{\mu\nu}(k)$:

$$
\tilde{G}_{\mu\nu}(k) = \sum_{s=1}^d \frac{\hat{\epsilon}_s^\mu(k)\hat{\epsilon}_s^\nu(k)}{m \omega_s^2(k)} k_B T \approx \sum_{s=1}^d \frac{\hat{\epsilon}_s^\mu(k)\hat{\epsilon}_s^\nu(k)}{mc^2 |k|^2} k_B T = \delta_{\mu\nu} \frac{T}{|k|^2},
$$

(3.79)

where the degenerate dispersion relations (3.47) for a simple harmonic crystal in $\mathbb{R}^d$ and the closure relation (3.42) are used in Eq. (3.79). Here, $T$ is defined in Eq. (3.54).

### 3.11 Appendix E: Debye Temperatures and Melting Points of Some Solids

We tabulate dimensionless Debye temperatures $T_D \equiv k_B T_D/mc^2$ and melting points $T_M \equiv k_B T_M/mc^2$ of some solids in Table 3.3. These quantities are estimated from the experimental data taken from Refs. [7] and [51]; the sound speeds $c$ are taken from Ref. [51], and the rest of them are taken from Ref. [7].
Table 3.3: Estimations of Debye temperatures $T_D$ and melting points $T_M$ of solids of some elements in the unit of the dimensionless temperature $\overline{T}$ up to two significant figures. The mass and sound speed are denoted by $m$ and $c$, respectively.

<table>
<thead>
<tr>
<th></th>
<th>$m$ ($10^{-23}$g)</th>
<th>$c$ ($10^3$m/s)</th>
<th>$T_D$ (K)</th>
<th>$T_M$ (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Be</td>
<td>1.50</td>
<td>12.9</td>
<td>1000</td>
<td>1550</td>
</tr>
<tr>
<td>Al</td>
<td>4.48</td>
<td>6.42</td>
<td>394</td>
<td>933</td>
</tr>
<tr>
<td>Cu</td>
<td>10.6</td>
<td>4.76</td>
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<td>1356</td>
</tr>
<tr>
<td>Au</td>
<td>32.7</td>
<td>3.24</td>
<td>170</td>
<td>1337</td>
</tr>
<tr>
<td>Mo</td>
<td>15.9</td>
<td>6.25</td>
<td>380</td>
<td>2890</td>
</tr>
<tr>
<td>Fe</td>
<td>9.44</td>
<td>5.00</td>
<td>420</td>
<td>1808</td>
</tr>
<tr>
<td>Pd</td>
<td>17.7</td>
<td>2.16</td>
<td>275</td>
<td>1825</td>
</tr>
<tr>
<td>Mg</td>
<td>4.04</td>
<td>5.77</td>
<td>318</td>
<td>922</td>
</tr>
<tr>
<td>Sn(white)</td>
<td>19.7</td>
<td>3.32</td>
<td>170</td>
<td>505</td>
</tr>
<tr>
<td>Ag</td>
<td>17.9</td>
<td>3.65</td>
<td>215</td>
<td>1234</td>
</tr>
<tr>
<td>Zn</td>
<td>10.9</td>
<td>4.21</td>
<td>234</td>
<td>693</td>
</tr>
<tr>
<td>W</td>
<td>30.5</td>
<td>5.22</td>
<td>310</td>
<td>3683</td>
</tr>
<tr>
<td>Ni</td>
<td>9.75</td>
<td>5.04</td>
<td>375</td>
<td>1726</td>
</tr>
<tr>
<td>Pt</td>
<td>32.4</td>
<td>3.26</td>
<td>230</td>
<td>2045</td>
</tr>
</tbody>
</table>
Chapter 4

Cloaking the Underlying
Long-Range Order of Randomly Perturbed Lattices

4.1 Introduction

A common way to introduce disorder into an otherwise ordered system such as a perfect crystal or quasicrystal is to randomly perturb the particle positions of that system [331, 279, 117, 2]. A perturbed lattice is a point pattern (process) in $d$-dimensional Euclidean space $\mathbb{R}^d$ obtained by displacing each point in a Bravais lattice $^1$ according to some stochastic rule [331, 85, 96, 155]. Perturbed lattices have been intensively studied in a broad range of contexts, from statistical physics and cosmology [86, 15], models of distorted lattices [331, 279], to probability theory, including distribution of zeros of random entire functions [276] and number rigidity [231, 95, 161]. They are related to queueing problems [8], in particular, G processes [103], and to stable matchings in any dimension [161]. Moreover, perturbed lattices are used to generate

$^1$This extends to any periodic point pattern with high crystallographic symmetries.
disordered initial configurations for numerical simulations [68] or configurations of sampling points [244].

The simplest stochastic rule involves independent and identically distributed (i.i.d.) perturbations. This model is also known as a shuffled lattice [86, 313]. The choice of the distribution of perturbations then specifies the model. A typical stochastic rule is the Gaussian distribution [231], in which case the model is also called an Einstein pattern [47]. Alternatively, the distributions can have heavy tails like the Cauchy or the Pareto distributions [155].

Another stochastic rule of special interest in the present study is where each point in a Bravais lattice \( \mathcal{L}^2 \) is displaced by a random vector that is uniformly distributed on a rescaled unit cell \( aC := \{x \in \mathbb{R}^d : x/a \in C\} \), where \( a > 0 \) is a scalar factor, and \( C \) is a unit cell of the lattice. We henceforth refer to this case as the uniformly randomized lattice (URL) model. We will use it as the main example for our more general results on the cloaking of Bragg peaks. The constant \( a \) controls the strength of perturbations. Counterintuitively, the long-range order suddenly disappears at certain discrete values of \( a \) and reemerges for stronger perturbations, as we will show.

For simplicity, we here use the simple cubic lattice \( \mathcal{L} = \mathbb{Z}^d \) with \( aC := [-a/2, a/2)^d \), see Fig. 4.1. It is a popular model studied in the optics community, among others, where it is used to understand how the introduction of disorder in lattices influences the resultant optical properties of the materials [14, 118, 227, 272, 117, 247, 215, 3, 13, 2, 342].

Perturbed lattices are special cases of hyperuniform systems. A hyperuniform point pattern is one in which the structure factor \( S(k) := 1 + \rho \tilde{h}(k) \) tends to zero as

\(^2\)Typical examples of Bravais lattices are the hexagonal and square lattice in two dimensions and the face-centered, body-centered, and simple cubic lattices in three dimensions.
the wavenumber \( k := \| \mathbf{k} \| \) tends to zero \([313, 300]\):

\[
\lim_{\| \mathbf{k} \| \to 0} S(\mathbf{k}) = 0,
\]

where \( \tilde{h}(\mathbf{k}) \) is the Fourier transform of the total correlation function \( h(\mathbf{r}) = g_2(\mathbf{r}) - 1 \) and \( g_2(\mathbf{r}) \) is the standard pair-correlation function. This implies that infinite-wavelength density fluctuations are anomalously suppressed.

An equivalent definition of hyperuniformity is based on the local number variance \( \sigma^2(R) \), which is associated with the number \( N(R) \) of points within a spherical observation window \( B_R \) of radius \( R \). A point pattern in \( \mathbb{R}^d \) is hyperuniform if its local number variance \( \sigma^2(R) := \text{Var}[N(R)] \) grows in the large-\( R \) limit slower than \( R^d \). This is in contrast to typical disordered systems, such as Poisson point patterns and liquids where the number variance scales like the volume \( v_1(R) \) of the observation window, for example, see Ref. [300].

If the structure factor vanishes at the origin continuously, then its asymptotic
behavior

\[ S(k) \sim |k|^\alpha \text{ for } |k| \to 0 \]  

(4.2)

with \( \alpha > 0 \) determines the large-\( R \) asymptotic scaling of the number variance [313] for \( R \to \infty \):

\[
\sigma^2(R) \sim \begin{cases} 
R^{d-1}, & \alpha > 1 \text{ (class I)} \\
R^{d-1} \ln R, & \alpha = 1 \text{ (class II)} \\
R^{d-\alpha}, & \alpha < 1 \text{ (class III)}
\end{cases}
\]  

(4.3)

These scalings of \( \sigma^2(R) \) define three classes of hyperuniformity [300], with class I and III describing the strongest and weakest forms of hyperuniformity, respectively.

Perturbed lattices with i.i.d. displacements are always hyperuniform, but the hyperuniformity class depends on whether the first and second moments of the perturbations exist [85, 155]. If both exist, then the perturbed lattice is class I hyperuniform with \( \sigma^2(R) \sim R^{d-1} \), that is, the number variance grows like the surface area of the observation window.

In hyperuniform systems, the suppression of large-scale density fluctuations can be quantitatively characterized by the hyperuniformity order metric [313, 300]. For class I systems, it is defined as

\[
\kappa := \lim_{L \to \infty} \frac{1}{L} \int_0^L \frac{\sigma^2(R)}{(R/D)^{d-1}} dR,
\]  

(4.4)

where \( D \) is a characteristic length scale in the system, e.g., the lattice constant.

A different measure of order in general statistically homogeneous point patterns is the \( \tau \) order metric [317]. It measures deviations of two-point statistics (i.e., structure
factor and pair-correlation function) from that of the ideal gas (Poisson point process):

$$
\tau := \frac{1}{D^d} \int_{\mathbb{R}^d} [g_2(r) - 1]^2 dr = \frac{1}{(2\pi)^d D^d \rho^2} \int_{\mathbb{R}^d} [S(k) - 1]^2 dk. \tag{4.5}
$$

By definition, $\tau = 0$ for the homogeneous Poisson point process with $g_2(r) = S(k) = 1$. By contrast, $\tau = \infty$ if there is a Bragg peak contribution to $S(k)$ (because of the squared difference).

In what follows, we will compute both $\Lambda$ and $\tau$ to thoroughly characterize the degree of order and disorder in hyperuniform perturbed lattices. Currently, perturbed lattices with weak or no correlations are among the rare examples of amorphous hyperuniform point patterns that can be easily simulated with a million particles per sample [222, 175, 155, 161]. However, in general, the resulting point patterns are not fully amorphous in the sense that their structure factor exhibits Bragg peaks, which are ‘inherited’ from the original lattice.

We demonstrate how a fine-tuned distribution of perturbations can hide or ‘cloak’ all or a portion of these Bragg peaks. A phenomenon that has been largely unnoticed in the community.\footnote{That is except for short side remarks by Gabrielli and Torquato [85, 90, 89] about the complete cancellation of the Bragg peak contribution being only possible for a very peculiar case fixing the zeros of the characteristic function of perturbations.}

Here, we provide an explicit real-space condition, present and discuss examples, and comprehensively structurally characterize the URL models using two different order metrics. First, we provide an intuitive necessary and sufficient criterion in Sec. 4.2 and discuss examples in Sec. 4.3. We also prove that perturbed lattices with i.i.d. displacements cannot be stealthy, which would require that $S(k) = 0$ for all $k$ in a neighborhood around the origin. In Sec. 4.4, we show that while the density fluctuations measured by $\Lambda$ increase for stronger perturbations, the degree of order measured by $\tau$ reveals a dramatic difference between the cloaked cases (no long-range order) and uncloaked cases (long-range order). While for the former, $\tau$ is finite, it
Perturbation strength $a$ increases from left to right

Figure 4.2: Structure factors of URL models in $\mathbb{R}^2$, where the perturbation strength $a$ increases from left to right. Samples of point patterns are shown on top, structure factors $S(k)$ of single configurations (including the forward scattering) are shown below, represented by the color code, as a function of the two-dimensional wave vector $k := (k_x, k_y)$. The Bragg peaks vanish when the perturbations cover the entire space without overlap ($a = 1.0$) but reappear when the perturbations become stronger ($a = 1.2$). Clearly visible are only peaks with $k_x = 0$ or $k_y = 0$, other peaks have small weights.

diverges for the latter. In that case, the rate by which $\tau$ increases with the system size still characterizes the degree of order in the system [316]. An outlook on related and open problems is given in the concluding Sec. 4.5.

4.2 Necessary and Sufficient Condition for Cloaking

We here consider uncorrelated displacements $u_x$ that follow the same probability density function $f(u_x)$ for each point $x$ in a lattice $\mathcal{L}$, see Fig. 4.1. The structure
Figure 4.3: Pair-correlation functions $h(x)$ of the URL model in 1D, cf. Eqs. (4.7) and (4.24), where the random displacement of each point in the lattice $\mathbb{Z}$ is uniformly distributed in $[-a/2, a/2)$. For $a = 1.0$, the total correlation function lacks any periodicity, see Eq. (4.25), and hence, the Bragg peaks are cloaked; for the angular-averaged pair-correlation function in the first three dimensions, see Figs. 4.5.

factor $S(k)$ is then given by [85]:

$$S(k) = 1 - \left| \tilde{f}(k) \right|^2 + \left| \tilde{f}(k) \right|^2 S_L(k),$$  

(4.6)

where $S_L(k)$ is the structure factor of the unperturbed lattice $L$ and $\tilde{f}$ is the characteristic function of the perturbations, that is, the Fourier transform of $f$. For convenience, the formula, which holds for more general point patterns, is rederived in Appendix 4.6.

Since the characteristic function is uniformly continuous at the origin, and since $\tilde{f}(0) = 1$, the perturbed point pattern is hyperuniform if and only if the original point pattern is hyperuniform. Hyperuniformity is preserved even if the moments of the perturbations do not exist, but in that case, the class of hyperuniformity changes (that is, the asymptotic behavior of the structure factor at the origin) [155, 300]; see Chapter 3. Stealthy hyperuniformity can never be preserved by independent random perturbations, as we prove in Appendix 4.7.

Equation (4.6) shows that a perturbed lattice generally will exhibit the same Bragg peaks as the original lattice. We can, however, choose the distribution of perturbations such that the characteristic function $\tilde{f}$ vanishes at these positions [85]. Intuitively speaking, the effective diffraction pattern of the perturbations cloaks the Bragg peaks.
The pair-correlation function offers an equivalent, intuitive criterion for the vanishing of all Bragg peaks. To obtain a statistically homogeneous point pattern, called *stationarized lattice*, we simultaneously shift all lattice points by a random vector that is uniformly distributed within a primitive unit cell of the lattice. The pair-correlation function of the perturbed lattice is then given by:

\[ g_2(r) = \frac{1}{\rho} \sum_{x \in L} f(r - x) - \frac{1}{\rho} f \ast f(r) \]  \hspace{1cm} (4.7)

where \( \rho \) is the number density and \( \ast \) denotes the convolution operator. The proof is given in Appendix 4.8.

All Bragg peaks vanish if and only if the series in Eq. (4.7) is constant, that is, independent of position \( r \):

\[ \sum_{x \in L} f(r - x) = \rho, \]  \hspace{1cm} (4.8)

which means that summing the probability density functions for all shifted lattice points add up to a constant function. By normalization, the constant has to be the number density. If this condition (4.8) is met, the resulting cloaked perturbed lattices have the following structure factor and pair correlation function, respectively:

\[ S(k) = 1 - |\tilde{f}(k)|^2 \]  \hspace{1cm} \text{and}  \hspace{1cm} g_2(r) = 1 - \frac{1}{\rho} f \ast f(r). \]

### 4.3 Examples of Cloaked and Uncloaked Perturbed Lattices

A straightforward example of how a lattice can be cloaked by perturbations is the uniform distribution of each lattice point within its unit cell. Our simulation study, shown in Fig. 4.2, demonstrates the appearance and cloaking of Bragg peaks for URL
Weights of Bragg peaks, $|\tilde{f}(k)|^2$

Perturbation strength, $a$

$k = (2\pi, 0)$

$k = (2\pi, 2\pi)$

$k = (4\pi, 0)$

Figure 4.4: The weights of the first three Bragg peaks of the 2D URL (cf. Fig. 4.2) as a function of the perturbation strength $a$.

Figure 4.5: Angular average of (a) the structure factor $S(k)$ and (b) the pair-correlation function (PCF) $g_2(r)$ for the cloaked URL with $a = 1$ in the first three dimensions. It is apparent that there are no Bragg peaks in $S(k)$, and that $g_2(r)$ lacks any periodicity.

models in $\mathbb{R}^2$, see Fig. 4.1.

We simulate three samples for different values $a$, each containing 10,000 points subject to periodic boundary conditions. Figure 4.2 shows portions of these point patterns in the upper panels and 2D plots of their structure factor in the lower panels. If the perturbation strength $a$ is an integer multiple of the lattice constant $4^4$For a single configuration with $N$ points at positions $r_1, r_2, \ldots$ under periodic boundary conditions, the structure factor including the forward scattering peak at the origin is equivalent to the scattering intensity $S(k) := \| \sum_{j=1}^N e^{-i k \cdot r_j} \|^2 / N$, where $k$ is a reciprocal lattice vector of the periodic simulation box.
$D$, Eq. (4.8) is fulfilled, and the Bragg peaks are cloaked.

Figure 4.3 shows the pair-correlation functions for the same parameters, but in 1D for better visualization. Only in the cloaked models with $a \in \mathbb{N} \setminus \{0\}$, $g_2(x)$ is not periodic for $\|x\| > a$. For the 1D model with $a = 1$, $g_2(x)$ was previously derived by [313].

We see that increasing the strength of the perturbations does generally not lead to a monotonic decay of the weights of Bragg peaks. Instead, these weights oscillate, as shown in Fig. 4.4. Interestingly, Bragg peaks can vanish for specific distributions of the random shifts, but they reappear as the perturbations become stronger. Fine-tuned perturbations at which the system appears to be without long-range order according to the two-point functions allow for the simulation of million-particle samples of hyperuniform systems without Bragg peaks. For these cloaked URLs, Fig. 4.5 shows for 1D, 2D, and 3D, the angular average of the structure factor $S(k)$ as a function of the wavenumber $k$ and of the pair-correlation function $g_2(r)$ as a function of the radial distance.

One could ask, to what extent are the higher-order functions cloaked with respect to the underlying lattice? Interestingly, for a cloaked URL with $a = 1$, we can actually express all of the $n$-point correlation functions explicitly by certain intersection volumes. Toward this end, we define $C_{ij} := (C + x_i) \cap (C + x_j)$ and $C^*_{ij} := C \cap \bigcup_{x \in \mathcal{L}} (C_{ij} + x)$, where $C + x$ denotes the translation of $C$ by $x$. Then, in case of a statistically homogeneous model (using a stationarized lattice), the multipoint correlation function is given by

$$g_n(x_1, \ldots, x_n) = 1 - \frac{1}{|C|} \left| \bigcup_{i,j=1,\ldots,n \atop i \neq j} C^*_{ij} \right|, \quad (4.9)$$

where here $|.|$ denotes the volume of a set and $C$ is a unit cell of the lattice $\mathcal{L}$. For a proof, see Appendix 4.9. For the 1D case, Figs. 4.8 and 4.9 display the three-point
and four-point functions, respectively. While $g_3$ does not exhibit explicit features of the underlying long-range order, there are specific paths in the parameter space of $g_4$ that reveal the periodicity of the original lattice.

A less obvious example of cloaked Bragg peaks is derived from i.i.d. perturbations with a probability density function $f(x) = (2\sin^2(x/2))/(\pi x^2)$. Due to its heavy tail, its characteristic function has bounded support: $\tilde{f}(k) = (1 - |k|)1_{[0,1]}(|k|)$, where $1_A(x)$ is the indicator function. The resulting structure factor is not analytic at the origin: $S(k) \sim k$ for $k \to 0$. The model is class II hyperuniform [300], and thus its hyperuniformity order metric $\Lambda$ is undefined.

4.4 Density Fluctuations and Order Metrics

Next, we focus on class I hyperuniform perturbed lattices, that is, for perturbations with finite first and second moments. In particular, we study the URL with $\mathcal{L} = \mathbb{Z}^d$. To quantify density fluctuations and the degree of order in the system, we compute both the hyperuniformity order metric $\Lambda$ and the $\tau$ order metric.

4.4.1 Hyperuniformity order metric $\Lambda$

The local number variance $\sigma^2(R)$ can be expressed in terms of a weighted integral over the structure factor [313]:

$$\sigma^2(R) = \frac{\rho v_1(R)}{(2\pi)^d} \int_{\mathbb{R}^d} S(k)\tilde{\alpha}_2(k; R)dk$$

(4.10)

with $\tilde{\alpha}_2(k; R) := 2^d\pi^{d/2}\Gamma(1 + d/2)[J_{d/2}(kR)]^2/k^d$, which is the square of the Fourier transform of the indicator function of $B_R$ divided by $v_1(R)$, $J_{\nu}(x)$ is the Bessel function of the first kind of order $\nu$.

We compute the hyperuniformity order metric $\Lambda$ of class I systems by substi-
tuting Eq. (4.6) into Eqs. (4.10) and (4.4). Using \( \lim_{L \to \infty} \frac{1}{L} \int_0^L \tilde{\alpha}_2(q; R) R dR = (2\pi)^d/[\pi v_1(1)|q|^{d+1}] \), we obtain:

\[
\bar{\Lambda} = \frac{(2\pi D)^d \rho}{\pi D} \left( \int_{\mathbb{R}^d} \frac{1 - |\tilde{f}(\mathbf{k})|^2}{(2\pi)^d |\mathbf{k}|^{d+1}} d\mathbf{k} + \rho \sum_{\mathbf{q} \in \mathcal{L}^* \setminus \{0\}} \left| \frac{\tilde{f}(\mathbf{q})}{|\mathbf{q}|^{d+1}} \right|^2 \right) ,
\]

(4.11)

where \( \mathcal{L}^* \) is the reciprocal lattice of \( \mathcal{L} \). The first term originates from the continuous contribution to \( S(\mathbf{k}) \) in Eq. (4.6), and the second term from the Bragg peak contribution. Both terms are non-negative. If \( f(\mathbf{r}) \) is a uniform distribution on a compact domain \( K \) and if the domains of different lattice points do not overlap, the second term equals the hyperuniformity order metric of a crystal, where each site in \( \mathcal{L} \) is decorated with \( K \).

For the URL, \( \bar{\Lambda} \) is a function of the perturbation strength \( a \). In 1D for \( \mathcal{L} = \mathbb{Z} \), we obtain the explicit expression

\[
\bar{\Lambda}(a) = \frac{a}{3} + \frac{\text{frac}(a)^2(1 - \text{frac}(a))^2}{6a^2} ,
\]

(4.12)

where \( \text{frac}(a) \) denotes the fractional part of \( a \). For \( a = 1 \), \( \bar{\Lambda} = 1/3 \) was first derived by [313]. While the second term in Eq. (4.12), that is, the Bragg contribution, vanishes for large values of \( a \), the first term grows linearly with \( a \). This behavior holds in any

Table 4.1: For the 2D URL, we report both the hyperuniformity order metric \( \bar{\Lambda} \), which quantifies large-scale density fluctuations, and the \( \tau \) order metric integrated over the entire system, which quantifies deviations from the ideal gas. If \( \tau(\infty) = \infty \), systems can still be distinguished by the growth rate of \( \tau \). The values for the unperturbed lattice are in agreement with those in Ref. [313].

<table>
<thead>
<tr>
<th>( a )</th>
<th>( Z^2 )</th>
<th>Perturbed lattices</th>
<th>Ideal gas</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>1/2</td>
<td>1</td>
<td>3/2</td>
</tr>
<tr>
<td>( \Lambda )</td>
<td>0.4576</td>
<td>0.63148</td>
<td>1.0428</td>
</tr>
<tr>
<td>( \tau(\infty) )</td>
<td>( \infty )</td>
<td>( \infty )</td>
<td>2/3</td>
</tr>
</tbody>
</table>

164
dimension in the sense that

\[ \overline{\Lambda}(a) = C a + \mathcal{O}(a^{-2d}), \text{ for } a \to \infty \]  

(4.13)

where \( C \) is a constant independent of \( a \) \(^5\) and \( \mathcal{O}(a^{-2d}) \) represents a vanishing bound on the Bragg contribution in Eq. (4.11) \(^6\).

Figure 4.6 shows explicit values for 2D obtained from Eq. (4.11) by numerical integration and by truncating the series at \( |q| < 2\pi \times 5000 \). Table 4.1 lists some of the values from Fig. 4.6.

The hyperuniformity order metric \( \overline{\Lambda} \) is a monotonically increasing function of the perturbation strength \( a \). Stronger perturbations imply strong density fluctuations.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{figure4_6.png}
\caption{The hyperuniformity order metric \( \overline{\Lambda} \) of the 2D URL as a function of the perturbation strength \( a \). Stronger perturbations imply stronger density fluctuations.}
\end{figure}

4.4.2 The \( \tau \) order metrics

There is, however, a dramatic difference in the degree of order as quantified by the \( \tau \) order metric \([317]\), see Eq. (4.5) and Table 4.1. The concept of \( \tau \) can be used to

\(^5\)The constant can easily be obtained from Eq. (4.11) by substituting \( k \) by \( x/a \): \( C := \frac{1}{\pi^d} \int_{\mathbb{R}^d} \left[ 1 - \prod_{i=1}^{d} \sin^2(x_i/2)/(x_i/2)^2 \right]/\|x\|^{d+1} \, dx. \)

\(^6\)It uses \( \sin^2(x) < 1 \) for all \( x \in \mathbb{R} \).
Figure 4.7: The $\tau$ order metric as a function of system size $L$ of a 2D URL. For almost all values of $a$, $\tau(L)$ diverges. Since the growth rate is small for $a > 1$, there is a range of values of $L$ where $\tau(L)$ is larger for $a = 1$ (cloaking) than, e.g., at $a = 1.4$ (non-cloaking). However, the curves cross at intermediate values of $L$. While $\tau(L)$ has converged to a constant at $L = 1$ for $a = 1$, it diverges for $a = 1.4$. Convergence to a constant for $a = 1$ and $a = 2$ is indicated by two points.

distinguish the degree of order in perturbed lattices even in the presence of Bragg peaks. To that end, $\tau(L)$ has been defined as a function of system size [9, 316]:

$$
\tau(L) := \frac{1}{D^d} \int_{[-L,L]^d} \left[ g_2(\mathbf{r}) - 1 \right]^2 d\mathbf{r}
$$

and consider its growth rate in the large-$L$ limit. Linear growth in the order metric was first identified in the integer lattice, prime numbers, and limit-periodic systems [316].

Figure 4.7 shows $\tau(L)$ for a 2D URL. For all non-integer values of $a$, $\tau(L)$ detects the long-range order and diverges for $L \to \infty$. While for small values of $L$, the degree of order seems to decrease monotonically with increasing perturbation strength $a$, the curves of $\tau(L)$ cross at intermediate values of $L$. This non-trivial degree of long-order as a function of $a$ can be quantified by the growth rate of $\tau(L)$. This growth rate vanishes for $a \to \infty$, but it does not decrease monotonically. Instead, it oscillates as a function of $a$, vanishes for integer values of $a$ and reemerges in between. In that sense, $a$ does unexpectedly not directly quantify the degree of order in a URL.
If $a \in \mathbb{N}$ (excluding zero), the Bragg peaks are cloaked, in which case the order metric converges to a constant:

$$\tau(L) = \left( \frac{2}{3a} \right)^d, \text{ for } L \geq a.$$ 

This constant decreases monotonically with increasing integer values of $a$.

### 4.5 Conclusions and Discussion

Often times for general perturbed lattices, pair-statistics are sufficient to detect the underlying long-range order via Bragg peaks. However, the latter are hidden by i.i.d. perturbations if and only if the characteristic function of the perturbations vanishes at the wave vectors of all reciprocal lattice points.

An equivalent real-space condition is that the probability density functions of the positions of all perturbed lattice points add up to a constant, see Eq. (4.8). This condition can be easily met for any Bravais lattice by uniformly distributing the lattice points inside their unit cells, that is, for any URL model with $a = 1$. In fact, this holds for any integer value of $a > 0$.

Specifically, for the URL, the perturbation strength $a$ at first glance may seem to be a natural metric of order in the system. Counterintuitively, we have shown in the present chapter that although the degree of long-range is damped for large perturbations, it oscillates as a function of $a$. Long-range correlations in two-point statistics can vanish at specific values of $a$ and reemerge for stronger perturbations; see Fig. 4.2. Our investigation has revealed that the $\tau$ order metric is a superior descriptor that quantifies both short- and long-range orders in the system.

Interestingly, the 1D perturbed lattice with uniform perturbations in the unit cell can be seen as a “two-point dual” of a Fermi-sphere point process [311], which means that the functional form of the structure factor of the former coincides with the pair-
correlation function of the latter and vice versa (up to a rescaling of the coordinates). It easily follows from Ref. [311] that the duality holds in any dimension for our URL with \( a = 1 \) and a “Fermi-cube” point process, that is, a determinantal point process whose Fourier transform of the kernel is an indicator function of the unit cube (instead of a sphere). The same duality does not hold for higher-order correlation functions.

The two-point function of the URL with \( a = 1 \) is perfectly cloaked, in the sense that it is impossible to reconstruct the underlying long-range order from the pair-correlation function alone. Higher-point correlation functions, however, can exhibit the periodicity of the original lattice. For cloaked URLs in \( \mathbb{R}^d \) with \( a = 1 \), we have derived the \( n \)-point correlation functions of arbitrary order. In 1D, we explicitly demonstrate in Fig. 4.9 how \( g_4 \) can reveal the periodicity of the underlying lattice.

An interesting open question for future research is how to construct isotropic amorphous hyperuniform point patterns or packings, for which samples with a million particles can easily be simulated (without any underlying lattice structure). For heterogeneous materials, the large-scale simulations of hyperuniform two-phase media that are fully amorphous has recently been made possible by a tessellation-based procedure [157], which locally enforces a global packing constraint in each cell.

### 4.6 Appendix A: Derivation of the Structure Factor of the Perturbed Lattice

Given a \( d \)-dimensional Bravais lattice \( \mathcal{L} \), the points of the perturbed lattice can be represented by \( \mathbf{x} + \mathbf{u}_x \), where \( \mathbf{x} \in \mathcal{L} \). Here, the displacements \( \mathbf{u}_x \) are i.i.d. with a probability density function \( f(\mathbf{u}_x) \).

For a finite ball \( B_r \) with radius \( r \) (centered at the origin), we denote by \( N_r \) the number of points of \( \mathcal{L} \) that fall into \( B_r \). Then, conditional on \( N_r = n \), we define the
scattering intensity within the finite ball by

\[ S_{n,r}(k) := \frac{1}{n} \left\langle \left| \sum_{x \in \mathcal{L} \cap B_r} e^{-ik \cdot (x + u_x)} \right|^2 : N_r = n \right\rangle, \tag{4.15} \]

where \( \langle \cdot \rangle \) denotes an ensemble average. In the following, we use the shorthand notation \( \mathbb{E}_n[\cdot] := \langle \cdot : N_r = n \rangle \).

In the thermodynamic limit, the structure factor \( S(k) \) is then given by [109]

\[ S(k) := \lim_{r \to \infty} \langle S_{n,r}(k) \rangle. \tag{4.16} \]

Using the mutual independence of the displacements, Eq. (4.15) can be simplified to

\[
S_{n,r}(k) = \frac{1}{n} \mathbb{E}_n \sum_{x,y \in \mathcal{L} \cap B_r} e^{-ik \cdot (x-y)} e^{-ik \cdot (u_x - u_y)} \\
= 1 + \left| \mathbb{E}_n [e^{-ik \cdot u}] \right|^2 \frac{1}{n} \mathbb{E}_n \sum_{x,y \in \mathcal{L} \cap B_r \atop x \neq y} e^{-ik \cdot (x-y)},
\]

where we denote by \( \tilde{f}(k) \) the characteristic function, that is, the Fourier transformation of the probability density function \( f \):

\[
\tilde{f}(k) := \mathcal{F}[f](k) = \int_{\mathbb{R}^d} f(r) e^{-ik \cdot r} dr. \tag{4.17}
\]

Note that \( \tilde{f}(-k) \) is the complex conjugate of \( \tilde{f}(k) \).

In the thermodynamic limit, the scattering intensity converges to Eq. (4.6):

\[ S(k) = 1 + \left| \tilde{f}(k) \right|^2 (S_L(k) - 1), \tag{4.18} \]

where \( S_L(k) \) is the structure factor of the lattice \( \mathcal{L} \). In fact, the derivation is valid for more general point patterns.
4.7 Appendix B: Proof of the Non-Stealthy Hyperuniformity of Perturbed Lattices

Stealthy hyperuniform point patterns are ones satisfying that $S(k) = 0$ if $|k| < K$ for some positive value of $K$ [317]. We note that a perturbed lattice is stealthy hyperuniform if and only if the displacements are deterministic, that is, $f(u_x) = \delta(u_x - c)$ for some $c \in \mathbb{R}^d$. This implies that perturbed lattices cannot be stealthy hyperuniform for any truly random perturbation.

From Eq. (4.6), the sufficient and necessary condition for a perturbed lattice to be stealthy hyperuniform is $\left| \tilde{f}(k) \right| = 1$ for all $|k| < K$ for some positive value of $K$. Straightforwardly, any deterministic displacement meets this condition. We now show that only such deterministic shifts with vanishing variance fulfill this condition.

Let $U$ and $V$ be two i.i.d. random variables with a characteristic function $\varphi(k)$ such that $|\varphi(k)|^2 = 1$ in a neighborhood around the origin. We define the random variable $D = U - V$. Its characteristic function is given by $\varphi_D(k) := \varphi(k)\varphi^*(k) = |\varphi(k)|^2$. So it is by construction infinitely differentiable at the origin. Therefore, all moments of $D$ exist, from which follows in turn that $\varphi_D(k)$ is an analytic function. Hence, $\varphi_D(k) := 1$ and $\text{Var}[D] = 0$. Since $U$ and $V$ are i.i.d., $2\text{Var}[U] = \text{Var}[U - V] = \text{Var}[D] = 0$.

4.8 Appendix C: Derivation of the Pair Correlation Function of Perturbed Lattices

To obtain a stationary point pattern with a pair-correlation function that does only depend on the relative position of two particles, we now consider a stationarized lattice. We shift the entire lattice $\mathcal{L}$ by a random vector that is uniformly distributed within a primitive unit cell. Then, we perturb each point independently following the probability density function $f$. Note that this stationarized model has the same
structure factor given by Eq. (4.6).

For a point pattern in the thermodynamic limit, its structure factor is directly related to its pair correlation function:

\[
S(k) = 1 + \rho \tilde{h}(k),
\]

where \( \tilde{h}(k) \) is the Fourier transform of the total correlation function \( h(r) := g_2(r) - 1 \), and \( \rho \) is the number density.

So, the pair-correlation function of perturbed lattices is given by

\[
g_2(r) = 1 + \mathcal{F}^{-1} \left[ \frac{S(k) - 1}{\rho} \right] (r) = 1 + \frac{1}{\rho} \mathcal{F}^{-1} \left[ \tilde{f}(k) \tilde{f}(-k) [S_L(k) - 1] \right] (r), \quad (4.19)
\]

where \( \mathcal{F}^{-1} [\cdot] (r) \) denotes the inverse Fourier transform. Note that the structure factor of a Bravais lattice \( L \) is

\[
S_L(k) = (2\pi)^d \rho \sum_{q \in L^* \setminus \{0\}} \delta(k - q),
\]

(4.20)

where \( L^* \) represents the reciprocal lattice of \( L \). Using Eq. (4.20) and the convolution theorem, one can rewrite Eq. (4.19) as

\[
g_2(r) = 1 - \frac{1}{\rho} \mathcal{F}^{-1} \left[ \tilde{f}(k) \tilde{f}(-k) \right] (r) + \sum_{q \in L^* \setminus \{0\}} \left| \tilde{f}(q) \right|^2 \cos(q \cdot r) \\
= 1 - \frac{1}{\rho} f * f(r) + \sum_{q \in L^* \setminus \{0\}} \left| \tilde{f}(q) \right|^2 \cos(q \cdot r),
\]

where \( f * g(r) := \int_{\mathbb{R}^d} f(x) g(r - x) \, dx \) represents the convolution operation.

Evaluating the Fourier series with the Poisson summation formula, we obtain the

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pair-correlation function of the perturbed lattice as

\[ g_2(\mathbf{r}) = f * f * g_\mathcal{L}(\mathbf{r}) - \frac{1}{\rho} f * f(\mathbf{r}) \quad (4.21) \]

where

\[ g_\mathcal{L}(\mathbf{r}) = \frac{1}{\rho} \sum_{\mathbf{x} \in \mathcal{L}} \delta(\mathbf{r} - \mathbf{x}) \quad (4.22) \]

is the pair-correlation function of the stationarized lattice. Inserting Eq. (4.22) into Eq. (4.21), we obtain Eq. (4.7), which can also be written as:

\[ g_2(\mathbf{r}) = \frac{1}{\rho} \sum_{\mathbf{x} \in \mathcal{L}\backslash\{0\}} f * f(\mathbf{r} - \mathbf{x}). \quad (4.23) \]

For the URL in \(d\)-dimensional Euclidean space, the probability density functions of different coordinates are independent of each other. So, the convolution in Eq. (4.23) factorizes:

\[ f * f(\mathbf{r}) = \frac{1}{a^d} \prod_{i=1}^{d} \left( 1 - \frac{|x_i|}{a} \right) 1_{[-a,a]}(x_i), \quad (4.24) \]

where \(\mathbf{r} = (x_1, x_2, \ldots)\). In the case of cloaking, i.e., \(a \in \mathbb{N} \backslash \{0\}\), the total correlation function \(h(\mathbf{r})\) also factorizes:

\[ h(\mathbf{r}) := \frac{-1}{a^d} \prod_{i=1}^{d} \left( 1 - \frac{|x_i|}{a} \right) 1_{[-a,a]}(x_i). \quad (4.25) \]
4.9 Appendix D: Derivation of the \( n \)-Point Correlation Functions of Cloaked URLs

For URLs with \( a = 1 \), we derive here the \( n \)-point correlation functions in arbitrary dimension \( d \). First, we state the \( n \)-point correlation function \( g_n^{(0)}(x_1, \ldots, x_n) \) for a statistically inhomogeneous model that uses a fixed lattice \( \mathcal{L} \). Since each lattice point \( y_i \in \mathcal{L} \) is uniformly distributed within its unit cell \( C + y_i \), \( g_n \) is 0 if a pair of distinct points \( x_i \) and \( x_j \) in the same unit cell, or 1 otherwise.

![Cloaked URL 1D](image)

Figure 4.8: Three-point correlation function \( g_3(0, x_2, x_3) \) of the 1D cloaked URL with \( a = 1 \). The lattice remains cloaked at the three-point level, in the sense, that there are no features that exhibit the periodicity of the underlying lattice.

For a statistically homogeneous URL \(^7\), the \( n \)-point correlation function is given by

\[
g_n(x_1, \ldots, x_n) = \frac{1}{|C|} \int_C g_n^{(0)}(x_1 - u, \ldots, x_n - u) \, du = 1 - \frac{1}{|C|} \left| \bigcup_{i,j=1,\ldots,n \atop i \neq j} S(x_i, x_j) \right|,
\]

where \( S(x_i, x_j) \) denotes the set of all points \( u \in C \), for which \( x_i - u \) and \( x_j - u \) are in the same unit cell, and thus the last term represents the probability for finding at least

\(^7\)The statistically homogeneous URL is based on a stationarized lattice \( \mathcal{L} + U \), where the random vector \( U \) is uniformly distributed on the unit cell \( C \).
one pair of points inside the same unit cell. Without loss of generality, we assume that $C = -C$. To prove Eq. (4.9), it remains to be shown that $S(x_i, x_j) = C_{ij}^*$. Assume that $u \in S(x_i, x_j)$. Then there exists $l \in L$ so that $x_i - u \in C + l$ and $x_j - u \in C + l$. Therefore $(-u) \in (C - x_i) \cap (C - x_j) + l$. Using $C = -C$, $(-l) \in L$, and $S(x_i, x_j) \subset C$, it follows that $u \in C_{ij}^*$, and thus $S(x_i, x_j) \subset C_{ij}^*$.

Assume that $u \in C_{ij}^*$. Then there exists $l \in L$ so that $u \in (C + x_i) \cap (C + x_j) + l$ and therefore $x_i - u \in C - l$ and $x_j - u \in C - l$. Hence, $u \in S(x_i, x_j)$, and thus $C_{ij}^* \subset S(x_i, x_j)$.

---

**Figure 4.9:** Four-point correlation function $g_4(0, x_2, x_3, x_4)$ of the 1D cloaked URL with $a = 1$ (a) as a function of $x_3$ choosing a specific path in configuration space, where $x_1$ and $x_2$ are constant and $x_4 = x_2 + x_3$. The curves represent four different values of $x_2$ (assuming without loss of generality that $x_1 = 0$). In contrast to the two-point and three-point correlation functions, the periodicity of the original lattice can be identified for $0 < x_2 < 1$. (b) The schematic explains the occurrence of this periodicity. There cannot be two particles within a single unit cell of the lattice. Therefore, the contribution of cases 1, 3, and 5 (counted from the top to bottom, colored red) to $g_4$ is identically zero. Moreover, the probability density of simultaneously finding a pair of points at $(x_1, x_2)$ and another one at $(x_3, x_4)$ depends on the distance $|x_2 - x_3|$ modulo the lattice spacing.

For the cloaked URL in 1D, Fig. 4.8 displays the three-point correlation function. It has no features with the periodicity of the underlying lattice. However, this periodicity can be extracted from the four-point function shown in Fig. 4.9.
Chapter 5

Inversion Problems for Fourier Transforms of Particle Distributions

5.1 Introduction

For $N$ identical point particles at positions of $r_1, \cdots, r_N$ in a periodic fundamental cell $\mathcal{F}$, the particle distribution can be described by a particle density $n(x) \equiv \sum_{j=1}^{N} \delta(x - r_j)$. Equivalently, this function can be represented by the (complex) Fourier components at wavevectors $k$'s, associated with the geometry of $\mathcal{F}$, i.e.,

$$\tilde{n}(k) \equiv \sum_{j=1}^{N} e^{-ikr_j},$$

called \textit{collective coordinates}. These quantities are often found to be a natural way to describe the distribution of particles, and thereby provide useful insights into many physical problems, e.g., excited states of liquid helium [77], conduction electrons in metals [234], general theory of simple liquids [230], and characterization of density...
fluctuations [313, 317]. Furthermore, using functional Fourier transformation, governing equations of many-body systems, such as the Fokker-Planck equation, can be expressed in terms of collective coordinates [67].

It is often desirable to infer the particle coordinates from given collective coordinates via inverse transformations. Importantly, amplitudes of collective coordinates or, equivalently, structure factors $S(k)$’s have long been used to probe the particle distributions since $S(k)$ can be measured by scattering experiments [7]. However, unless the particle distribution is a perfect crystal, the structure factor alone cannot uniquely determine the particle distribution. To solve this problem in X-ray crystallography, one should acquire additional information from other physical properties, such as the interference pattern with known molecules (specific site labeling) [145], anomalous dispersion relations [232, 119], or sequential projections onto constrained hyperplanes [72]. Such inversion tasks are called the phase-retrieval problems [72, 265, 111] because the tasks are essentially equivalent to retrieving the “phase” information contained in collective coordinates, the complete set of which are in principle invertible into particle coordinates. Even if the phase information is incorporated, however, this inversion task is still highly nontrivial, due to the nonlinear relation between collective and particle coordinates.

Given a target point configuration in one-dimensional Euclidean space $\mathbb{R}$, our primary objective of this chapter is to find the minimal set of its collective coordinates that uniquely determine particle coordinates under exchange of particle indices. This minimal set, therefore, uniquely determines collective coordinates at other wavevectors. To carry out this search, we treat the number $M$ of the real and/or the imaginary parts of collective coordinates of a target configuration as constraints, and find all configurations, called solutions, whose collective coordinates satisfy these constraints. The number of constraints $M$ is increased one-by-one until we have a unique solution that is identical to the target pattern.
Previous studies on this inversion task [317, 74, 352, 353] focused on some special types of constraints in collective coordinates for a given set of wavevectors, such as the stealthy constraints, where \( \tilde{n}(k) = 0 \), and amplitude-constraints for a prescribed radial function \( f(r) \), i.e., \( |\tilde{n}(k)| = f(|k|) \). This inversion task is often carried out via the collective-coordinate optimization technique [352, 353, 321, 322, 357] that is designed to find ground-state configurations of the potential associated with those constraints. Here, it is useful to define a new parameter \( \chi \equiv M/(dN) \) [352, 321] that represents the relative fraction of the number of constrained collective coordinates \( M \) to the total number of degrees of freedom; see Fig. 5.1 for typical arrangements of the constraints in \( d = 1, 2 \). These studies analytically or numerically showed that when the stealthy constraints are imposed for \( \chi < 1/2 \), the associated ground states, called stealthy disordered hyperuniform systems [317, 352, 353, 321], are disordered, highly degenerate, and statistically isotropic. Importantly, it has been shown that systems, derived from these special disordered point configurations by decorating the points with particles of certain shapes, are endowed with some novel photonic and transport properties [80, 355, 356, 46, 179, 82, 102, 336]; see also Ref. [300] and references therein. Under the stealthy constraints with \( \chi \geq 1/2 \), on the other hand, (virtually all) available configurations are crystalline in the first three spatial dimensions [317, 74, 321]. From the uniqueness of the solution at \( \chi = 1/2 \) in \( d = 1 \) [74] as well as the importance of phase information of collective coordinates, one can argue that each constrained collective coordinate \( \tilde{n}(k) \) removes two degrees of freedom in the accessible configurational space. Thus, it is natural to surmise that the minimum value of \( M \) for the unique inversion would be \( M = dN \).

In this chapter, we consider more general types of constraints, in which the real and/or the imaginary part of each collective coordinate are independently prescribed. For simplicity, we focus on one-dimensional systems. For such systems, we show that the minimal set of collective-coordinate constraints consists of collective coordinates...
at the \([N/2]\) smallest wavevectors, i.e., \(M = 2\lceil N/2 \rceil\) rather than \(N\). This result also implies that both real and imaginary parts of a collective coordinate (at a wavevector \(k\)) must be specified to determine particle coordinates uniquely. We analytically show this result for small systems of \(N \leq 3\). However, this result is invalid if the target configurations are the integer lattice because one cannot determine its center of mass without a collective coordinate at the first Bragg peak. In our numerical studies for larger systems, we exclude the pathological case (i.e., the integer lattice), and consider two distinct ensembles of target configurations: perturbed lattices \([85]\) via uniformly distributed displacements, and Poisson point distribution configurations. For each of these target configurations, we find solutions numerically via the collective-coordinate optimization technique. Our numerical results show that these two types of ensembles occupy qualitatively different energy landscapes: those in perturbed lattices are relatively simpler than those in Poisson ones.

In section 5.2, we present basic definitions and background. In section 5.3, we describe the numerical method that we employ to find solutions. In section 5.4, we theoretically and numerically determine the minimal sets of collective coordinates for small systems. Larger systems are numerically investigated in section 5.5. Finally, we provide concluding remarks in section 5.6.

5.2 Background and Definitions

5.2.1 General Properties of Collective Coordinates

For a \(N\)-particle point configuration within a periodic fundamental cell \(\mathcal{F}\), collective coordinates (5.1), which are also known as collective density variables, are complex-valued quantities that are defined at certain real-valued discrete wavevectors \(k\)'s. Here, the available wavevectors correspond to the reciprocal lattice vectors of the cell \(\mathcal{F}\). For instance, if \(\mathcal{F}\) is a \(L_1 \times \cdots \times L_d\) rectangular box, then \(k\)'s can be described
Figure 5.1: Schematics of typical arrangements of collective-coordinate constraints in Fourier space for a periodic $d$-dimensional square fundamental cell of side length $L$. Here, upper and lower panels represent cases for $d = 1$ and $2$, respectively. Constraints are taken from $\tilde{n}(k)$’s at wavevectors between two concentric circles centered at the origin: there are $2\mathcal{M}$ wavevectors (black dots) within the blue circle, except for $2N_k + 1$ wavevectors inside the red-shaded region. In Refs. [74, 321, 317, 352, 353], a spherical region with $N_k = 0$ was considered; see a list of available $\mathcal{M}$ values for two-dimensional cases in Table II in Ref. [321]. For our present purposes, the number of constraints is denoted by $M = 2\mathcal{M}$ because the real and/or the imaginary parts of collective coordinates are considered independently.

as follows: $k = 2\pi (\frac{m_1}{L_1}, \cdots, \frac{m_d}{L_d})$ for $(m_1, \cdots, m_d) \in \mathbb{Z}^d$. For the simplicity, we focus on one-dimensional systems in the rest of this chapter, and thus use the following short-hand notation:

$$k_m = 2\pi m/L. \quad (5.2)$$

At two different wavevectors, the collective coordinates are not always independent. For instance, the complex conjugate of a collective coordinate by definition is equal to its parity inversion, i.e., $\tilde{n}^*(k) = \tilde{n}(-k)$. Thus, if we constrain such a pair of collective coordinates, only one of them is considered independent. For this reason, the relative fraction $\chi$ of constrained degrees of freedom is defined as not $2\mathcal{M}/(dN)$,
but $\mathcal{M}/(dN)$; see Fig. 5.1.

Only certain sets of complex numbers can be collective coordinates of a "realizable" point configuration. For example, there are some trivial necessary conditions of realizable collective coordinates, such as $|\tilde{n}(\mathbf{k})| \leq N$ for any wavevector $\mathbf{k}$, and $\tilde{n}(0) = N$. However, it is highly nontrivial to find sufficient and necessary conditions of realizable collective coordinates. To avoid such realizability problems [167], we take constraints from the collective coordinates of a target configuration.

The value of a collective coordinate is independent of the choice of particle permutations: When we invert collective coordinates, the resulting particle coordinates also should be equivalent under the exchange of particle indices.

5.2.2 Definitions

In the rest of this chapter, we clearly distinguish a target and a solution configurations by using separate notations $\mathbf{R}^N = \{R_1, R_2, \cdots, R_N\}$ and $\mathbf{r}^N = \{r_1, r_2, \cdots, r_N\}$, respectively. The corresponding collective coordinates are denoted by $\tilde{n}_T(k)$ and $\tilde{n}(k)$, respectively.

In numerical studies, two types of target configurations at unit number density are considered:

1. Perturbed lattices [85, 331], generated from the integer lattice by independently displacing each particle via a uniform distribution in $[-\delta, \delta]$, and

2. Poisson point distribution configurations.

We note that the perturbed lattices become identical to the Poisson point distribution configurations if $\delta = N/2$ under the periodic boundary condition.

We denote $M$ constraints, used in the inversion task, by $E_i = 0$ for $i = 1, 2, \cdots, M$. Starting from the origin in the Fourier space, we skip the first $N_k$ wavenumbers and
constrain the collective coordinates at the next $\lfloor M/2 \rfloor$ wavenumbers:

$$E_i \equiv \begin{cases} 
\text{Re}[\tilde{n}_T(k_{N_k+m}) - \tilde{n}(k_{N_k+m})], & i = 2m - 1 \ (i < M) \\
\text{Im}[\tilde{n}_T(k_{N_k+m}) - \tilde{n}(k_{N_k+m})], & i = 2m, \ (i \leq M)
\end{cases} \quad (5.3)$$

where $\lfloor x \rfloor$ is the floor function, $m \in \mathbb{N}$, and $\text{Re}\{x\}$ and $\text{Im}\{x\}$ represent the real and the imaginary parts of a complex number $x$, respectively. Thus, if $M$ is an even number, both the real and the imaginary parts of collective coordinates at $M/2$ consecutive wavenumbers are constrained. If $M$ is an odd number, we prescribe the last term $E_M$ via two conditions, each of which is concerning either the real or the imaginary parts of a target collective coordinate as follows:

$$E_M = \begin{cases} 
\text{Re}[\tilde{n}_T(k_{N_k+[M/2]}) - \tilde{n}(k_{N_k+[M/2]})], & \text{condition (5.4)} \\
\text{Im}[\tilde{n}_T(k_{N_k+[M/2]}) - \tilde{n}(k_{N_k+[M/2]})], & \text{condition (5.5)}
\end{cases} \quad (5.4)$$

where $\lceil x \rceil$ is the ceiling function. Table 5.1 lists some examples of constraints.

Table 5.1: Examples of constraints $E_i$ for corresponding shorthand notations. We note that when $M$ is an even number, the real condition (5.4) and the imaginary condition (5.5) give the identical collective-coordinate constraints.

<table>
<thead>
<tr>
<th>$N_k$ and $M$ conditions</th>
<th>$E_1$</th>
<th>$E_2$</th>
<th>$E_3$</th>
<th>$E_4$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$N_k = 0$ and $M = 4$</td>
<td>$\text{Re}[\tilde{n}_T(k_1) - \tilde{n}(k_1)]$</td>
<td>$\text{Im}[\tilde{n}_T(k_1) - \tilde{n}(k_1)]$</td>
<td>$\text{Re}[\tilde{n}_T(k_2) - \tilde{n}(k_2)]$</td>
<td>$\text{Im}[\tilde{n}_T(k_2) - \tilde{n}(k_2)]$</td>
</tr>
<tr>
<td>$N_k = 1$ and $M = 4$</td>
<td>$\text{Re}[\tilde{n}_T(k_2) - \tilde{n}(k_2)]$</td>
<td>$\text{Im}[\tilde{n}_T(k_2) - \tilde{n}(k_2)]$</td>
<td>$\text{Re}[\tilde{n}_T(k_3) - \tilde{n}(k_3)]$</td>
<td>$\text{Im}[\tilde{n}_T(k_3) - \tilde{n}(k_3)]$</td>
</tr>
<tr>
<td>$N_k = 0$, $M = 3$, and condition (5.4)</td>
<td>$\text{Re}[\tilde{n}_T(k_1) - \tilde{n}(k_1)]$</td>
<td>$\text{Im}[\tilde{n}_T(k_1) - \tilde{n}(k_1)]$</td>
<td>$\text{Re}[\tilde{n}_T(k_2) - \tilde{n}(k_2)]$</td>
<td>$\cdot$</td>
</tr>
<tr>
<td>$N_k = 0$, $M = 3$, and condition (5.5)</td>
<td>$\text{Re}[\tilde{n}_T(k_1) - \tilde{n}(k_1)]$</td>
<td>$\text{Im}[\tilde{n}_T(k_1) - \tilde{n}(k_1)]$</td>
<td>$\text{Im}[\tilde{n}_T(k_2) - \tilde{n}(k_2)]$</td>
<td>$\cdot$</td>
</tr>
</tbody>
</table>

5.3 Numerical Method

Given a target configuration $\mathbf{R}^N$ of $N \geq 3$, we take $M$ constraints from its collective coordinates, and numerically find solution configurations $\mathbf{r}^N$ via a modified “collective-coordinate optimization technique” [352, 353, 321, 322, 357] that was ini-
tially designed to generate disordered classical point configurations, such as stealthy ground states [317, 352, 17], and the perfect-glass model [354]. The detailed procedure is described as follows:

1. Starting from a random initial configuration \( \{ r_i^{(0)} \}_{i=1}^N \) of \( N \) particles, numerically search for an energy-minimizing configuration \( r^N \equiv \{ r_i \}_{i=1}^N \) for the following potential energy,

\[
\Phi(r^N; R^N) \equiv \sum_{l=1}^{M} |E_l(r^N; R^N)|^2
\]

\[
= \begin{cases} 
\sum_{l=N_{k}+1}^{M/2+N_{k}} |\tilde{n}_T(k_l) - \tilde{n}(k_l)|^2, & M \text{ is even} \\
\sum_{l=N_{k}+1}^{[M/2]+N_{k}} |\tilde{n}_T(k_l) - \tilde{n}(k_l)|^2 + |E_M(r^N; R^N)|^2, & M \text{ is odd.}
\end{cases}
\]

The \( j \)th component of its gradient is given by

\[
F_j(r^N; R^N) \equiv -\frac{\partial \Phi}{\partial r_j} (r^N; R^N)
\]

\[
= \begin{cases} 
\sum_{l=N_{k}+1}^{M/2+N_{k}} 2k_l \text{Im}[\tilde{n}(k_l) - \tilde{n}_T(k_l)] e^{i k_l r_j}, & M \text{ is even} \\
\sum_{l=N_{k}+1}^{[M/2]+N_{k}} 2k_l \text{Im}[\tilde{n}(k_l) - \tilde{n}_T(k_l)] e^{i k_l r_j} - 2E_M \frac{\partial E_M}{\partial r_j}, & M \text{ is odd,}
\end{cases}
\]

where \( E_l \) is defined by (5.3), and for an odd number \( M \), \( E_M \) is defined by one of two conditions (5.4) and (5.5). This configuration is called a “solution” if \( \Phi(r^N; R^N) < \epsilon_E \) for a specified small tolerance \( \epsilon_E \).

2. Test if this solution \( r^N \) agrees with the target configuration \( R^N \) or other solutions found previously within another small tolerance \( \epsilon_X \), i.e.,

\[
\max_{i=1}^{N} \{ \min_{j=1}^{N} \{|r_i - R_j|\}\} < \epsilon_X. \]

If they agree, then \( r^N \) is deemed to be identical to one of the previous solutions, and we increase the solution’s count.
Otherwise, we record $r^N$ as a new solution.

3. Repeat the steps 1-2 for $N_I$ random initial configurations.

4. Repeat the steps 1-3 for $N_T$ different target configurations.

Roughly speaking, the potential (5.6) represents a “deviation” or numerical error of a solution configuration from the target configuration in terms of given collective-coordinate constraints. In step 1, we mainly use two different optimization algorithms: the low-storage BFGS (L-BFGS) algorithm [219, 141] with the MINOP algorithm [58, 352], and the steepest descent algorithm [91]. We repeat this inversion task for many distinct initial configurations $\{r_i^{(0)}\}_{i=1}^N$ and target configurations $R^N$s. Unless stated otherwise, we use parameters as follows: $N_I = 1\,000$, $N_T = 1\,000$, and $\epsilon_X = 10^{-6}$.

For all numerically distinct solutions $\{r^N\}$ of a target configuration $R^N$, the trivial solution refers to the one that is identical to the target ($r^N = R^N$), while nontrivial solutions refer to the others ($r^N \neq R^N$).

## 5.4 Results for $N \leq 3$

Here, we theoretically and numerically investigate solutions for small target configurations.

### 5.4.1 $N = 1$

For a single-particle configuration, $\tilde{n}(k_1) = e^{-i2\pi r_1/L}$ is a one-to-one function from $r_1 \in [0, L]$ onto the unit circle on the complex plane, i.e., $\{z \in \mathbb{R} : |z| = 1\}$. Thus, it is straightforward to show that there is a unique solution, given constraints $\tilde{n}_T(k_1)$ that correspond to the cases of $N_k = 0$, and $M = 2$. Equivalently, collective coordinates at larger wavenumbers can be expressed in terms of $\tilde{n}_T(k_1)$, i.e., $\tilde{n}_T(k_m) = \tilde{n}_T(k_1)^m$. 
On the other hand, cases of \( N_k = 0 \) and \( M = 1 \), i.e., a single constraint of either \( \text{Re}\{\tilde{n}_T(k_1)\} \) or \( \text{Im}\{\tilde{n}_T(k_1)\} \), give two solutions; see Fig. 5.2(a). Thus, we need at least two constraints \( (M = 2) \) for the unique inversion of a single-particle configuration.

We note that \( \tilde{n}_T(k_1) \) is the minimal set of constraints for single-particle systems. This is because when \( m > 1 \), \( \tilde{n}_T(k_m) \) is no longer a one-to-one function from \( r_1 \in [0, L) \) onto the unit circle on \( \mathbb{R} \), and thus cases with \( N_k = m \) and \( M = 2 \) for \( m > 1 \) give \( m \) distinct solutions; see Fig. 5.2(b).

Figure 5.2: Illustrations for solutions of the inversion problem for a single-particle target configuration. (a) Cases with \( N_k = 0 \) and \( M = 2 \). When \( \tilde{n}_T(k_1) \) is given as constraints (left), both its real and imaginary parts are required for a unique solution; see the cross (×) mark in the right panel. Red and blue lines represent the real and the imaginary parts of \( \tilde{n}(k_1) \) of a solution, respectively. (b) Cases with \( N_k = 1 \) and \( M = 2 \). When \( \tilde{n}_T(k_2) \) is given, we have two solutions.

5.4.2 \( N = 2 \)

Using graphical solutions, one can straightforwardly show a single constraint \( (N_k = 0 \) and \( M = 1) \) gives infinitely many solutions; see one of the solid or dashed lines in Fig. 5.3. However, Fig. 5.3 also immediately shows that the following equation \( (N_k = 0 \) and \( M = 2) \)

\[
\tilde{n}_T(k_1) = e^{-i2\pi r_1/L} + e^{-i2\pi r_2/L},
\]

(5.8)
and it yields a unique solution under exchanges of particle indices, as follows:

\[ e^{-i2\pi r_1/L} = \frac{\tilde{n}_T(k_1)}{2} \left( 1 \pm i \sqrt{\frac{4}{|\tilde{n}_T(k_1)|^2 - 1}} \right) \]  
(5.9)

\[ e^{-i2\pi r_2/L} = \frac{\tilde{n}_T(k_1)}{2} \left( 1 \mp i \sqrt{\frac{4}{|\tilde{n}_T(k_1)|^2 - 1}} \right) \]  
(5.10)

if \( \tilde{n}_T(k_1) \neq 0 \) or, equivalently, \(|R_1 - R_2| \neq 0.5L\). Otherwise, the periodic image of the target configuration becomes the integer lattice, and all of translated lattices are solutions of (5.8), i.e., there are infinitely many solutions, as shown in Fig. 5.3(c).

If the target configuration is the integer lattice, in order to obtain a unique solution, the collective coordinate at the first Bragg peak [i.e., \( \tilde{n}_T(k_2) \)] should be addi-
tionally specified, which corresponds to the cases with \( N_k = 0 \) and \( M = 4 \). Then, the unique solution is

\[
e^{-i2\pi r_1/L} = \frac{1}{2} \left( \tilde{n}_T(k_1) \pm \sqrt{2\tilde{n}_T(k_2)^2 - \tilde{n}_T(k_1)^2} \right),
\]

\[
e^{-i2\pi r_2/L} = \frac{1}{2} \left( \tilde{n}_T(k_1) \mp \sqrt{2\tilde{n}_T(k_2)^2 - \tilde{n}_T(k_1)^2} \right).
\]

This is because the collective coordinate at the first Bragg peak provides the center of mass of this lattice configuration.

We note that the constraint \( \tilde{n}_T(k_2) \) alone (i.e., \( N_k = 1 \) and \( M = 2 \)) cannot be uniquely inverted into particle coordinates. It can be straightforwardly shown that there exist at least four distinct solutions, i.e., \((r_1, r_2) = a + (R_1, R_2)\), where \( a/L = (0, 0), (0, 1/2), (1/2, 0) \), and \((1/2, 1/2)\). By the same analysis, one can identify there are at least \( m^2 \) distinct solutions if only \( \tilde{n}_T(2\pi m/L) \) is given. Therefore, we can conclude that for a two-particle configuration that is not the integer lattice, the minimal set of constraints for a unique solution is \( \{\tilde{n}_T(k_1)\} \).

**Remarks**

1. For a configuration of particle number \( N > 1 \), Fan, et al. [74] proved that \( \tilde{n}(k_m) = 0 \) for \( m = 1, \ldots, \lfloor N/2 \rfloor \) is a sufficient and necessary condition for the configuration to be the integer lattice or its translations. Thus, if one inverts collective coordinates at the \( \lfloor N/2 \rfloor \) smallest wavenumbers of the integer lattice, its solutions are inevitably degenerate with a translational degree of freedom; see Fig. 5.3 (c) for example.

**5.4.3 \( N = 3 \)**

In the previous sections, we show that there is a unique solution in the inversion procedure with parameters \( N_k = 0 \) and \( M = \lfloor N/2 \rfloor \), unless the target configuration is a pathological case (i.e., either the integer lattice or its translations). Otherwise,
there are infinitely many solutions. It implies that there would be a sudden transition in the number of distinct solutions varying with the type of target configurations. For this reason and simplicity in analysis, our target configurations are restricted here to perturbed lattices that can continuously interpolate between the integer lattice to Poisson configurations via the displacement parameter $\delta$; see section 5.2.2.

For a perturbed lattice, its particle coordinates are described as $r_i = (i-1) + N\delta_i$ for $i = 1, \cdots, N$. Assuming weak perturbations (i.e., $|\delta_i| \ll 1$) for $N = 3$, collective-coordinate constraints can be approximated up to the second order of displacements;

\[
\text{Re}[\tilde{n}(k_m)] \approx \begin{cases} 
3 - 2(m\pi)^2(\delta_1^2 + \delta_2^2 + \delta_3^2), & m = 3i \\
\sqrt{3}m\pi(\delta_2 - \delta_3) + (m\pi)^2(-2\delta_1^2 + \delta_2^2 + \delta_3^2), & m = 3i + 1,
\end{cases} \quad (5.13)
\]

\[
\text{Im}[\tilde{n}(k_m)] \approx \begin{cases} 
2m\pi(\delta_1 + \delta_2 + \delta_3), & m = 3i \\
m\pi(2\delta_1 - \delta_2 - \delta_3) + \sqrt{3}(m\pi)^2(\delta_2^2 - \delta_3^2), & m = 3i + 1,
\end{cases} \quad (5.14)
\]

where $i$ represents non-negative integers.

For parameters $N_k = 0$ and $M = 3$ with the real condition (5.4) [or the imaginary one (5.5)], the quadratic approximations (5.13) and (5.14) yield at most two distinct solutions (5.18): the trivial solution ($r^3 = R^3$), and a nontrivial one ($r^3 \neq R^3$). This prediction is consistently observed in numerical results; see Fig. 5.4(a). Thus, the set of numerically distinct solutions abruptly changes from an uncountably many set into a finite one, as $\delta$ becomes nonzero. Figure 5.4(a) also shows that if $\delta$ increases, while the maximal number of numerically distinct solutions remains two, its occurrence decreases regardless of constraint conditions (5.4) and (5.5).

In numerical studies, it is important to know how results depend on the optimiza-
The number of distinct solutions
condition (4): BFGS+MINOP
condition (5): BFGS+MINOP
condition (4): Steepest Descent
condition (5): Steepest Descent

Figure 5.4: Numerical results of the inversion procedure for three-particle perturbed lattices in cases with $N_k = 0$ and $M = 3$. (a) The average number of distinct solutions per target configuration. Two different optimization algorithms (BFGS+MINOP and the steepest descent) and two constraint conditions [the real (5.4) and the imaginary (5.5) ones] are used for comparison with the energy tolerance $\epsilon_E = 10^{-29}$. For any target configuration, the number of distinct solutions is at most two, but the average can vary with the target configurations. (b) Examples of nontrivial solutions for a given target perturbed lattice with various displacements $\delta$. Nontrivial solutions by the real (5.4) or the imaginary (5.5) conditions, respectively, are different from each other, and are not translations of the target.

...tion algorithms and values of parameters, such as $\epsilon_E$ and $\epsilon_X$. For this purpose, we investigate the energy distributions of numerical solutions obtained in the parameters of $N_k = 0$ and $M = 3$, and various conditions, as shown in Fig. 5.5. From Fig. 5.5 (a) and (b), we see that given a target configuration, both trivial and nontrivial
solutions have qualitatively similar energy profiles, regardless of the real (5.4) and
the imaginary (5.5) conditions. Figure 5.5(c) demonstrates that the energy profiles
of numerical solutions vary with optimization algorithms, but for a given algorithm,
both trivial and nontrivial solutions still have qualitatively similar energy profiles.
Thus, a nontrivial solution cannot be eliminated by lowering the energy tolerance $\epsilon_E$ when $N = M = 3$. In the rest of this chapter, we mainly use the BFGS and MINOP algorithms because the solutions obtained via these algorithms tend to have lower numerical errors than those via the steepest descent method.

For parameters $N_k = 0$ and $M = 4$, a unique solution can be obtained. This also can be deduced from the observation in the cases with $N_k = 0$ and $M = 3$ that given a target configuration, nontrivial solutions, respectively obtained by the real (5.4) and the imaginary (5.5) conditions, are numerically distinct; see Fig. 5.4(b). Thus, the common solution from two conditions (5.4) and (5.5) should be identical to the target. The unique solution also can be obtained from the quadratic approximations (5.13) and (5.14) as follows:

$$\delta_1 = \frac{1}{12\pi} \left[ \frac{6 \text{Im}[2\tilde{n}_T(k_1) - \tilde{n}_T(k_2)]}{\text{Re}[4\tilde{n}_T(k_1) - \tilde{n}_T(k_2)]} + \text{Im}[4\tilde{n}_T(k_1) + \tilde{n}_T(k_2)] \right],$$

$$\delta_2 = \frac{1}{12\pi} \left[ \frac{6 \text{Im}[2\tilde{n}_T(k_1) - \tilde{n}_T(k_2)]}{\text{Re}[4\tilde{n}_T(k_1) - \tilde{n}_T(k_2)]} - \frac{\text{Re}[4\tilde{n}_T(k_1) - \tilde{n}_T(k_2)]}{\sqrt{3}} \right],$$

$$\delta_3 = \frac{1}{12\pi} \left[ \frac{6 \text{Im}[2\tilde{n}_T(k_1) - \tilde{n}_T(k_2)]}{\text{Re}[4\tilde{n}_T(k_1) - \tilde{n}_T(k_2)]} + \frac{\text{Re}[4\tilde{n}_T(k_1) - \tilde{n}_T(k_2)]}{\sqrt{3}} \right],$$

and thus the minimal set for three-particle systems is (both real and imaginary parts of) collective coordinates at the two smallest wavenumbers.

**Remarks:**

1. For parameters $N_k = 0$, $M = 3$, and the real condition (5.4), the quadratic approximations (5.13) and (5.14) give two exact solutions (5.18). While one of the solutions is the same as the target configuration up to some numerical errors, another solution cannot precisely predict the nontrivial solution partly because the nontrivial one is not a perturbed lattice with small displacements.

2. For parameters $N_k = 1$ and $M = 4$, a unique solution is obtained; see (5.20), (5.21), and (5.22).
5.5 Results for $N > 3$

Figure 5.6: Log-log plots of histograms for energy distribution of numerically distinct solutions \( \{r^N\} \) for odd-number system sizes: \( N = 9 \) (a, d), 19 (b, e), and 29 (c, f). Using the real condition (5.4) condition and parameters \( N_k = 0 \) and \( \epsilon_E = 10^{-20} \), two types of target configurations are considered: (a-c) perturbed lattices with \( \delta = 0.1 \) and (d-f) Poisson configurations. When \( M = N \), while a target perturbed lattice has a single nontrivial solution \( (r^N \neq R^N) \), whose occurrence rate is similar to that of trivial ones, a Poissonian target mainly has the trivial solution but occasionally can have multiple nontrivial solutions. When \( M = N + 1 \) is an even number, while there is a unique solution for perturbed lattices, there can be more than one solution for a Poisson target configuration in relatively lower occurrence rates. Even in the latter case, however, the nontrivial solutions can be eliminated by lowering the tolerance \( \epsilon_E \) around \( 10^{-25} \).

Here, we numerically investigate the properties of the inversion procedure from collective coordinates, such as proper values of the tolerances \( \epsilon_E \) and \( \epsilon_X \). For this purpose, we obtain distributions of energy \( \Phi(r^N; R^N) \) for numerically distinct solutions, as we did in Fig. 5.5. Our results, shown in figures 5.6 and 5.7, demonstrate that the energy distributions sensitively depend on the number of skipped collective-
coordinate constraints $N_k$ as well as target configurations and the particle number $N$.

At first, we consider the cases with $N_k = 0$ (Fig. 5.6). When there are even-number $N$ of particles, $M(\geq N)$ constraints can give unique solutions for both types of target configurations: perturbed lattices and Poisson point distribution configurations. If $N$ is an odd number, however, $M = N$ constraints no longer ensure unique solutions. When perturbed lattices are the target configurations (Fig. 5.6(a-c)) and $M = N$ constraints are considered, the energy $\Phi(r^N; R^N)$ always has two global minima, which correspond to the trivial solution ($r^N = R^N$) and a nontrivial one ($r^N \neq R^N$), respectively. On the other hand, the energy $\Phi(r^N; R^N)$ of a Poissonian
Figure 5.8: Numerical results for the average number of numerically distinct solutions per a target configuration of particle number $N$ with various values of $N_k$. Using the real condition (5.4) and BFGS-MINOP algorithms, we consider two types of target configurations: (a-c) perturbed lattices with $\delta = 0.1$ and the tolerance $\epsilon_E = 10^{-20}$, and (d-f) Poisson configurations with $\epsilon_E = 10^{-25}$. When $N_k = 0$, both types of target configurations require $M = N$ constraints for an even-number $N$, and $M = N + 1$ is the minimal for an odd-number $N$: The minimal number of $M$ is $2 \lceil N/2 \rceil$. If $N_k > 0$, for both types of target configurations, the minimal number of constraints becomes $M = N + 1$.

target configuration (Fig. 5.6(d-f)) mostly has a single minimum that is identical to the target ($r^N = R^N$) but occasionally has more than two nontrivial solutions. Given parameters $N_k = 0$ and $M = N + 1$, when the target is a perturbed lattice, the inversion procedure gives a unique solution. By contrast, when the target is a Poisson configuration, this procedure may give multiple solutions. We note that since the nontrivial solutions in the latter case have qualitatively different energy profiles from the trivial solution (see Fig. 5.6(d-f)), the nontrivial solutions can be eliminated by lowering the tolerance $\epsilon_E$ to a proper level. Thus, when $N$ is an odd number, $M = N + 1$ constraints are required for the unique determination.

When the first few collective coordinates are skipped ($N_k > 0$), there is no advantage of even-number particles, i.e., one cannot determine unique solutions with
$M = N$ successive collective-coordinate constraints when $N$ is an even number. Figure 5.7 shows the histograms for energies of numerical solutions obtained in the inversion procedure with odd-number particles and $N_k > 0$. In Fig. 5.7, we note that for $M = N$ constraints, there can be multiple nontrivial solutions whose energy profiles are similar to that of the trivial solutions. However, $M = N + 1$ constraints allow us to find the trivial solutions without any nontrivial one.

In general, as the system size $N$ increases, both trivial and nontrivial solutions tend to have higher energies, i.e., larger numerical errors. Moreover, for parameters $N_k = 0$ and $M = N$, although for smaller systems, the distribution of trivial and nontrivial solutions have tails in the low-energy regime [Fig. 5.6 (a, d)], for larger systems the tails are shifted to the high-energy regime [Fig. 5.6 (c, f)]; see also Fig. 5.7 for cases with $N_k > 0$. This observation implies that it becomes less probable to obtain numerical solutions, whether they are trivial or not, as the particle number $N$ increases, or the energy tolerance $\epsilon_E$ is lowered.

![Figure 5.9: The minimal number of successive collective-coordinate constraints $\text{min } M$ as a function of particle number $N$ for various $N_k$.](image)

The average number of numerically distinct solutions, obtained in the inversion procedure, is shown in Fig. 5.8. This figure clearly demonstrates that for Poissonian targets (Fig. 5.8(d-f)) the two curves ($M = N$ and $N + 1$) collapses into a single
line as $N$ increases, and thus $\min M \to N$ as $N$ increases. On the other hand, these two curves are separated for perturbed lattices (Fig. 5.8(a-c)), and thus $\min M$ is determined by the cases where perturbed lattices are the target configurations. Figure 5.9 summarizes the results from analytic investigation into small systems (section 5.4) and numerical studies on larger systems (section 5.5). One can uniquely determine particle coordinates from collective coordinates at the $\left\lceil \frac{N}{2} \right\rceil$ smallest wavenumbers, i.e., parameters of $N_k = 0$ and $M = 2\left\lceil \frac{N}{2} \right\rceil$, by properly selecting $\epsilon_E$. On the other hand, if $N_k > 0$, one requires $M = N + 1$ successive collective-coordinate constraints to uniquely determine particle coordinates. Therefore, when both cases are considered, the minimal set of collective-coordinate constraints are collective coordinates at the $\left\lceil \frac{N}{2} \right\rceil$ smallest wavenumbers.

5.6 Conclusions and Discussion

In this chapter, we have investigated the minimal set of collective-coordinate constraints as a function of the particle number $N$ to determine the progenitor particle coordinates in one dimension uniquely. We also considered how the minimal collective-coordinate constraints depend on constraint types (the real (5.4) and imaginary (5.5) conditions) and types of target configurations, i.e., perturbed lattices and Poisson point distribution configurations. As shown in Fig. 5.9, the minimal set of constraints are collective coordinates at the $\left\lceil \frac{N}{2} \right\rceil$ smallest wavenumbers: It corresponds to the parameters of $N_k = 0$ and $M = 2\left\lceil N/2 \right\rceil$. In other words, the removed number of degrees of freedom in the solution space will vary with each collective-coordinate constraint. Thus the real and the imaginary parts of a collective coordinate are not completely independent.

For this result to accommodate the pathological case, i.e., the integer lattice, one needs to regard all of its translations to be equivalent. As we noted in section
5.4.2, this is because translations of the integer lattices cannot be distinguished in terms of \( \tilde{n}_T(k_m) \) for \( m = 1, \cdots, \lceil N/2 \rceil \), since their collective coordinates are identically zero, except at the Bragg peaks, i.e., \( k = 2\pi, 4\pi, \cdots \). An additional constraint \( \tilde{n}_T(k_N) \equiv \tilde{n}_T(2\pi) \) at the first Bragg peak is necessary to remove the translational degree of freedom. However, we note that non-Bravais lattices are not pathological cases because their lattice constants are larger than one, and thus their first Bragg peaks should appear within the range of \( |k| \leq \pi \).

It is worthwhile to compare this conclusion with the result of Fan et al. [74]. These authors proved that for a one-dimensional system, one needs its collective coordinates at the \( \lfloor N/2 \rfloor \) smallest wavenumbers as well as the center of mass in order to determine all of its collective coordinates; see 5.8 for the detailed summary. In the same context, our investigation shows that if the center of mass is unknown, one needs collective coordinates at the \( \lceil N/2 \rceil \) smallest wavenumbers. Moreover, when there are even-number particles, the knowledge of the center of mass does not reduce the necessary information.

While this chapter focused on one-dimensional systems for simplicity, it is useful to discuss the implications of our results for the inversion problem in higher-dimensional systems. Unlike one-dimensional systems, higher-dimensional systems can have many different ways to select collective-coordinate constraints; see Fig. 5.10. Consider the case (c) where selected wavevectors form \( n \) nonparallel strips orienting toward the origin. Based on our present results, if the \( i \)th strip has a slope \( s_i = n_i/m_i \), where \( n_i \) and \( m_i \) are integers and coprime, and includes the smallest \( \lceil N/2 \rceil \) wavevectors, then one can uniquely determine values of the coordinates on a line, i.e., \( m_i x_j + n_i y_j \) for \( j = 1, \cdots, N \). Thus, by using two perpendicular strips that include a total of \( 2\lceil N/2 \rceil \) collective-coordinate constraints, one can “separately” determine the \( x \) and \( y \) coordinates of particle positions. In order to determine the pairing between the \( x \) and \( y \) coordinates, one needs collective-coordinate constraints along additional strips.
in the Fourier space, as shown in Fig. 5.10(c). Therefore, in this scheme, at least $3 \lceil N/2 \rceil$ collective-coordinate constraints are required.

Figure 5.10: Schematics of some possible ways to select collective-coordinate constraints in the two-dimensional Fourier space. Collective coordinates are specified at wavevectors inside (a) an annular region of outer radius $K$ and inner radius $K_0$ (see Fig. 5.1), (b) a rectangular region of width $K_x$ and height $K_y$, and (c) $n$ mutually non-parallel strips which lengths are $K_i$, $i = 1, \cdots, n$. We note that the red-shaded region is excluded.

It is interesting to compare collective coordinates with Fourier components in the discrete Fourier transform (DFT). While a Fourier component $X_k$ in DFT is a linear function of a complex sequence $\{x_n\}_{n=0}^{N-1}$, a collective coordinate $\tilde{n}(k_m)$ is a nonlinear function of particle coordinates $\{R_j\}_{j=1}^{N}$. In both cases, wavenumbers must be equally spaced due to the periodic boundary conditions in direct spaces. On the contrary, the direct spaces are different in the two cases in that while the direct spaces in DFT are digitized into $N$ pixels, those in collective coordinates are continuous. If one discretizes the space of a point configuration with $N$ pixels of width $\Delta x$, the configuration can be described by a real-valued sequence $\{x_n\}$, where $x_n$ represents the number of particles in the $n$th pixel. Then, this conversion can be
straightforwardly written as follows:

\begin{align*}
\text{Particlecoordinates}: & \{R_i\}_{i=1}^{N} \subset \mathbb{R} \Rightarrow \{x_n\}_{n=0}^{N-1} \subset \mathbb{N} \cup \{0\} \\
\text{Collectivecoordinates}: & \tilde{n}(k_m) = \sum_{i=1}^{N} \exp(-ik_m x_i) \Rightarrow X_m = \sum_{j=0}^{N-1} x_j \exp\left[-i \frac{2\pi m}{N\Delta x} (j\Delta x)\right].
\end{align*}

Thus, the \(m\)th collective coordinate \(\tilde{n}(k_m)\) of a point configuration corresponds to the \(m\)th Fourier component \(X_m\) of its digitized version. From this relationship, one can surmise that the inverse DFT with the first \(N/2\) collective coordinates will give a discretized point configuration with a position precision \(\Delta x\). In other words, one needs around \(10^7\) Fourier components to achieve \(\Delta x \sim \mathcal{O}(10^{-7})\), which is a typical error in our solution configurations.

In this chapter, we focused on the search for the minimal set of constraints, rather than computational costs. Our inversion procedure is intuitive and provides easy-to-estimate numerical errors in solutions [i.e., energy \(\Phi(r^N; R^N)\)], but this method is inefficient for large systems. For instance, as system size \(N\) increases, the computation cost grows at least in the order of \(N^2\). Furthermore, since this method tends to have larger numerical errors in solution configurations as \(N\) increases (see figures 5.6 and 5.7), it becomes more likely to fail to find any solution with a given value of the energy tolerance \(\epsilon_E\). The failure rate becomes especially much higher when a target is more complicated. Therefore, it would be important for future studies to develop more efficient procedures to invert collective coordinates into particle coordinates.
5.7 Appendix A: Approximate Solutions of Equations (5.13) and (5.14)

For parameters $N_k = 0$ and $M = 3$, and the real condition (5.4), from (5.13) and (5.14), one can find two solutions as follows:

\[
\begin{align*}
\delta_1 & \approx \frac{-18 \Im[\tilde{n}_T(k_1)] \pm D}{\pi (\Re[4 \tilde{n}_T(k_1) - \tilde{n}_T(k_2)]) (\Re[4 \tilde{n}_T(k_1) - \tilde{n}_T(k_2)] - 12)} , \\
\delta_2 & \approx -\frac{\Re[4 \tilde{n}_T(k_1) - \tilde{n}_T(k_2)]}{12 \sqrt{3} \pi} + \frac{6}{\Re[4 \tilde{n}_T(k_1) - \tilde{n}_T(k_2)] - 6} \left( \frac{\Im[\tilde{n}_T(k_1)]}{2 \pi} - \delta_1 \right) , \\
\delta_3 & \approx -\frac{\Re[4 \tilde{n}_T(k_1) - \tilde{n}_T(k_2)]}{12 \sqrt{3} \pi} + \frac{6}{\Re[4 \tilde{n}_T(k_1) - \tilde{n}_T(k_2)] - 6} \left( \frac{\Im[\tilde{n}_T(k_1)]}{2 \pi} - \delta_1 \right) ,
\end{align*}
\]

(5.18)

where the discriminant $D$ is written as

\[
D \equiv \frac{1}{12 \sqrt{3}} \{ \Re[4 \tilde{n}_T(k_1) - \tilde{n}_T(k_2)] - 6 \} \left[ \left( \Re[4 \tilde{n}_T(k_1) - \tilde{n}_T(k_2)] - 6 \right)^2 - 36 \right] \times \left( \Re[4 \tilde{n}_T(k_1) - \tilde{n}_T(k_2)]^2 - 36 \Re[2 \tilde{n}_T(k_1) + \tilde{n}_T(k_2)] + 3888 \Im[\tilde{n}_T(k_1)]^2 \right)^{1/2} .
\]

(5.19)

Here, a trivial solution is obtained from (5.18) when a minus sign is taken in $\delta_1$. Otherwise, (5.18) become a nontrivial solution.

For parameters $N_k = 1$ and $M = 4$, (5.13) and (5.14) give a single solution:

\[
\begin{align*}
\delta_2 & = -\frac{\delta_1}{2} + \frac{\Im[\tilde{n}_T(k_3)]}{12 \pi} + \left[ \sqrt{3} \pi \delta_1^2 + \frac{1}{4 \sqrt{3} \pi} \left( \Re[\tilde{n}_T(k_2)] + \frac{2}{9} \tilde{n}_T(k_3) \right) - \frac{2}{3} \right] , \\
\delta_3 & = -\frac{\delta_1}{2} + \frac{\Im[\tilde{n}_T(k_3)]}{12 \pi} - \left[ \sqrt{3} \pi \delta_1^2 + \frac{1}{4 \sqrt{3} \pi} \left( \Re[\tilde{n}_T(k_2)] + \frac{2}{9} \tilde{n}_T(k_3) \right) - \frac{2}{3} \right] ,
\end{align*}
\]

(5.20) (5.21)
where $\delta_1$ is determined by the following cubic equation:

$$
\delta_1^3 - \frac{\text{Im}[\tilde{n}_T(k_3)]}{6\pi} \delta_1^2 - \frac{1}{12\pi^2} \left( \text{Re}[\tilde{n}_T(k_2)] + \frac{2}{9} \tilde{n}_T(k_3) \right) - \frac{11}{3} \delta_1
$$

$$
+ \frac{1}{72\pi^3} \left[ \text{Im}[\tilde{n}_T(k_3)] \left( \text{Re}[\tilde{n}_T(k_2)] + \frac{2}{9} \tilde{n}_T(k_3) \right) - \frac{5}{3} - 3 \text{Im}[\tilde{n}_T(k_2)] \right] = 0,
$$

which has a single real root.

### 5.8 Appendix B: The Uniqueness of Solutions for the Inversion Problem

Using the generating function argument [74], one can prove that there is a unique configuration to satisfy $N$ prescribed collective coordinates. Let us define a generating function as

$$
f(z) \equiv \sum_{m=1}^{\infty} \frac{\tilde{n}(k_m)}{m} z^m,
$$

which is well-defined for $|z| < 1$ because $|\tilde{n}(k_m)|$ is bounded. Using the definition (5.1) and power series expansion of the log function $[\ln(1 - z) = \sum_{n=1}^{\infty} z^n/n$ for $|z| < 1$],

$$
f(z) = \sum_{n=1}^{\infty} \left( \sum_{j=1}^{N} e^{-ix_j} \right) \frac{z^n}{n} = \sum_{j=1}^{N} \sum_{n=1}^{\infty} (ze^{-ix_j})^n = \sum_{j=1}^{N} -\ln(1 - ze^{-ix_j})
$$

$$
= -\ln \left[ \prod_{j=1}^{N} (1 - ze^{-ix_j}) \right].
$$

Since the term inside square brackets of logarithm is a polynomial of order $N$, $\exp\{f(z)\}$ also should be a polynomial of order $N$.

$$
\prod_{j=1}^{N} (1 - ze^{-ix_j}) = \exp(-f(z)) = \mathbb{P}_N \exp(-f(z)) = \mathbb{P}_N \exp(-\mathbb{P}_N f(z))
$$

$$
= \mathbb{P}_N \exp \left( - \sum_{m=1}^{N} \frac{\tilde{n}(k_m)}{m} z^m \right),
$$

(5.25)
where $\mathbb{P}_N$ represents a projection to a degree $N$ polynomial of $z$.

By substituting (5.25) into (5.24) and doing further analysis, Fan, et al. [74] derived the following identity:

\[
\sum_{m=1}^{N} \frac{\tilde{n}(k_m)}{m} z^m = -\ln \left[ \mathbb{P}_{\lfloor N/2 \rfloor} \exp \left( -\sum_{m=1}^{\lfloor N/2 \rfloor} \frac{\tilde{n}(k_m)}{m} z^m \right) - \mathcal{F} z^N \mathbb{P}_{-\lfloor N/2 \rfloor} \exp \left( \sum_{m=1}^{\lfloor N/2 \rfloor} \frac{\tilde{n}(-k_m)}{m} z^m \right) \right],
\]

(5.26)

where $\mathcal{F} \equiv \exp(-2\pi \sum_{n=1}^{N} x_n)$, and $\lfloor x \rfloor$ is the floor function of $x$. Since $\tilde{n}(k_m) = \tilde{n}(-k_m)^*$, if collective coordinates at the $\lfloor N/2 \rfloor$ smallest wavenumbers and the center of mass are known, in principle one can determine collective coordinates at other wavenumbers. In other words, there is a unique point configuration that satisfies these conditions.
Chapter 6

Characterizing the Hyperuniformity of Ordered and Disordered Two-Phase Media

6.1 Introduction

The hyperuniformity concept generalizes the traditional notion of long-range order in many-particle systems to include all perfect crystals, perfect quasicrystals, and exotic amorphous states of matter [313, 300]. A hyperuniform point configuration in $d$-dimensional Euclidean space $\mathbb{R}^d$ is characterized by an anomalous suppression of large-scale density fluctuations relative to those in typical disordered systems, such as liquids and structural glasses. The hyperuniformity notion was generalized to the case of heterogeneous (multiphase) materials [346, 299, 195], i.e., materials consisting of two or more phases [296, 255], such as composites, porous media, foams, cellular solids, colloidal suspensions and polymer blends. Subsequently, the concept was extended to quantify hyperuniformity in a variety of different systems, including random scalar fields, divergence-free random vector fields, and statistically anisotropic many-particle
systems [299]. Hyperuniformity has been attracting great attention across many fields, including physics [317, 354, 121, 245, 225, 194, 186, 343, 324, 300, 177, 104, 85, 155, 160, 300, 78, 110, 180, 48, 185], materials science [193, 339, 303, 46, 157], mathematics [276, 231, 97, 30, 316] and biology [137, 203, 171, 300, 362].

In the case of point configurations, one can rank order crystals, quasicrystals, and special disordered systems within a hyperuniformity class according to the degree to which they suppress density fluctuations, as measured by the hyperuniformity parameter $B_N$ [313, 346]. Much less is known about the analogous rank ordering of hyperuniform two-phase media via the appropriate hyperuniformity parameter $B_V$, as defined below. However, it is much more challenging to do so for two-phase media for two reasons. First, the geometries and topologies of the phases are generally much richer and more complex than point-configuration arrangements. Second, one must determine length scales that are broadly applicable for the multitude of possible two-phase media microstructures to make $B_V$ dimensionless. The purpose of this article is to begin such a program for certain two-dimensional periodic and disordered models of two-phase media.

For two-phase heterogeneous media in $d$-dimensional Euclidean space $\mathbb{R}^d$, which include cellular solids, composites, and porous media, hyperuniformity is defined by the following infinite-wavelength condition on the spectral density $\tilde{\chi}_V(k)$[346, 300], i.e.,

$$\lim_{|k| \to 0} \tilde{\chi}_V(k) = 0,$$  \hspace{1cm} (6.1)

where $k$ is the wavevector. The spectral density $\tilde{\chi}_V(k)$ is the Fourier transform of the autocovariance function $\chi_V(r) \equiv S^{(i)}_2(r) - \phi_i^2$, where $\phi_i$ is the volume fraction of phase $i$, and $S^{(i)}_2(r)$ gives the probability of finding two points separated by $r$ in phase $i$ at the same time [255, 296]. It can be easily obtained for general microstructures either theoretically, computationally, or via scattering experiments [54]. Hyperuniformity can equivalently be defined in terms of the local volume-fraction variance $\sigma^2_V(R)$ as-
associated with a spherical window of radius $R$. Specifically, a hyperuniform two-phase system is one in which $\sigma^2_v(R)$ decays faster than $R^{-d}$ in the large-$R$ regime [346, 300], i.e.,

$$\lim_{R \to \infty} R^d \sigma^2_v(R) = 0.$$  \hfill (6.2)

The local variance $\sigma^2_v(R)$ is directly determined by $\tilde{\chi}_v(k)$ via the following integral [300, 346]:

$$\sigma^2_v(R) = \frac{1}{v_1(R)(2\pi)^d} \int_{\mathbb{R}^d} \tilde{\chi}_v(k) \tilde{\alpha}_2(|k|; R) \, dk,$$  \hfill (6.3)

where $v_1(R) = \pi^{d/2}[\Gamma(d/2 + 1)]^{-1} R^d$ is the volume of a $d$-dimensional sphere of radius $R$, $\Gamma(x)$ is the gamma function, and

$$\tilde{\alpha}_2(|k|; R) \equiv 2^d \pi^{d/2} \Gamma(d/2 + 1) \left(\frac{|J_d/2(|k|R)|}{|k|^d}\right)^2,$$  \hfill (6.4)

is the Fourier transform of the scaled intersection volume of two spheres of radius $R$ that are separated by $r$.

As in the case of hyperuniform point configurations [313, 346, 347, 300], there are three different scaling regimes (classes) that describe the associated large-$R$ behaviors of the volume-fraction variance when the spectral density goes to zero as a power-law scaling $\tilde{\chi}_v(Q) \sim |Q|^\alpha$ as $Q$ tends to zero:

$$\sigma^2_v(R) \sim \begin{cases} 
    R^{-(d+1)}, & \alpha > 1 \quad \text{(Class I)} \\
    R^{-(d+1)} \ln R, & \alpha = 1 \quad \text{(Class II)}, \\
    R^{-(d+\alpha)}, & 0 < \alpha < 1 \quad \text{(Class III)}
\end{cases}$$  \hfill (6.5)

where the exponent $\alpha$ is a positive constant. Classes I and III are the strongest and weakest forms of hyperuniformity, respectively. One aim of this chapter is to compute the implied coefficient hyperuniformity order metric $\overline{B}_V$ (defined in Sec.
6.2.1) multiplying $R^{-(d+1)}$ for certain class I structures, which is a measure of the degree to which large-scale volume-fraction fluctuations are suppressed within that class.

An overarching goal of this chapter is to characterize the hyperuniformity of models of two-phase media that belong to class I. Due to the infinite variety of possible two-phase microstructures (geometries and topologies), we purposely restrict ourselves to a certain class of periodic cellular networks as well as periodic and disordered/irregular packings. Even this restrictive set of models of two-phase media presents challenges, since one must ascertain relevant length scales that are broadly applicable to make key quantities dimensionless, as discussed in Sec. 6.6. In particular, we evaluate the volume-fraction variance as a function of the window radius $R$ for all models. We also compute the aforementioned hyperuniformity order metric $B_V$ for each model to rank order them.

In Sec. 6.4, we present relevant theoretical background to characterize hyperuniform two-phase media and describe the computational methods employed in this study. In Sec. 6.3, we provide exact closed-form formulas of the form factors of general polyhedra in $\mathbb{R}^2$ and $\mathbb{R}^3$, which are important to characterize periodic networks. We then describe the two-dimensional models of two-phase media of class I hyperuniformity considered in this investigation: periodic cellular networks (Sec. 6.4), periodic disk packings, and disordered/irregular disk packings (Sec. 6.5). In Sec. 6.6, we provide the rationale for choosing the inverse of the specific surface as the characteristic length scale $D$ in the hyperuniformity order metric $B_V$. In Sec. 6.7, we investigate the microstructure-dependence of the volume-fraction variance and rank order all of our class I models according to $B_V$. Finally, we present concluding remarks and outlooks for future research in Sec. 6.8.
6.2 Background and Methods

6.2.1 Asymptotic Analysis of Hyperuniform Two-Phase Media

For a statistically homogeneous and isotropic medium in $\mathbb{R}^d$, the local volume-fraction variance $\sigma^2_V(R)$ can be written as the following large-$R$ asymptotic expansion [346, 300]:

$$\sigma^2_V(R) = A_V(R) \left( \frac{D}{R} \right)^d + B_V(R) \left( \frac{D}{R} \right)^{d+1} + o\left( \frac{D}{R} \right)^{d+1}, \quad (6.6)$$

where $A_V(R)$ and $B_V(R)$ are dimensionless asymptotic coefficients of powers $R^{-d}$ and $R^{-(d+1)}$, respectively, and they are defined by

$$A_V(R) = \frac{1}{v(D)} \int_{|r| \leq 2R} \chi_V(r) \, dr \quad (6.7)$$
$$B_V(R) = - \frac{c(d)}{2Dv_1(D)} \int_{|r| \leq 2R} \chi_V(r) \, |r| \, dr, \quad (6.8)$$

where $c(d) \equiv 2 \Gamma(d/2 + 1) / \left[ \pi^{d/2} \Gamma((d + 1)/2) \right]$, and $D$ is a characteristic length scale of the medium. In the large-$R$ limit, the coefficient $A_V(R)$ is proportional to the spectral density at the origin, i.e.,

$$\overline{A}_V \equiv \lim_{R \to \infty} A_V(R) \propto \lim_{|k| \to 0} \tilde{\chi}_V(k), \quad (6.9)$$

and thus for any hyperuniform medium, $\overline{A}_V = 0$, and hence the expansion (6.6) reduces to

$$\sigma^2_V(R) = B_V(R) \left( \frac{D}{R} \right)^{d+1} + o\left( \frac{D}{R} \right)^{d+1}. \quad (6.10)$$

It is noteworthy that, unlike $\sigma^2_V(R)$, the coefficient $B_V(R)$ depends on the choice of the length scale $D$.

In the case of class I hyperuniform systems, $\sigma^2_V(R)$ decays like $R^{-(d+1)}$ for large
\( R \), as specified by
\[
\sigma^2_V(R) \sim \mathcal{B}_V \left( \frac{D}{R} \right)^{d+1}, \quad R \to \infty.
\] (6.11)

As \( R \) increases, the coefficient \( B_V(R) \) converges to the hyperuniformity order metric \( \mathcal{B}_V \) for typical disordered systems. For some infinite media, such as periodic and aperiodic structures, the associated coefficient \( B_V(R) \) oscillates around some running average value. In such cases, it is advantageous to estimate \( \mathcal{B}_V \) by using the cumulative moving average, as defined by [300]
\[
\mathcal{B}_V \equiv \lim_{L \to \infty} \frac{1}{L} \int_0^L B_V(R) \, dR. \quad (6.12)
\]

### 6.2.2 Spectral Density and the Local Volume-Fraction Variance

Here we present explicit formulas for the spectral density \( \tilde{\chi}_V(k) \) of general packings in \( \mathbb{R}^d \), ordered or not [349, 298, 300]. We also describe the formula for the local volume-fraction variance \( \sigma^2_V(R) \) for class I hyperuniform two-phase media and the associated hyperuniformity order metric \( \mathcal{B}_V \). Importantly, these formulas also can be applied to characterize periodic cellular networks, as we will discuss later.

In the case of packings of identical particles \( \mathbf{P} \) of arbitrary shape, it is known that
\[
\tilde{\chi}_V(k) = \rho |\tilde{m}(k; \mathbf{P})|^2 S(k), \quad (6.13)
\]

where \( \rho \) is the number density of particle centers, \( \tilde{m}(k; \mathbf{P}) \) is the Fourier transform of (also called form factor) of the particle indicator function \( m(x; \mathbf{P}) \) defined by
\[
m(x; \mathbf{P}) = \begin{cases} 1, & \text{x is inside } \mathbf{P} \\ 0, & \text{otherwise} \end{cases}, \quad (6.14)
\]
where $\mathbf{x}$ is the position vector with respect to the centroid of $\mathbf{P}$, and $S(\mathbf{k})$ is the structure factor of particle centers; see Refs. [349, 298, 300] for the definition of $S(\mathbf{k})$ and its computation. One can immediately obtain from (6.13) the specific formulas for a $d$-dimensional (Bravais) lattice packing in which a single particle $\mathbf{P}$ is placed in a fundamental cell $\mathcal{F}$ of the Bravais lattice $\mathcal{L}$ as follows:

$$
\tilde{\chi}_V(\mathbf{k}) = |V_\mathcal{F}|^{-1} |\tilde{m}(\mathbf{k}; \mathbf{P})|^2 S_\mathcal{L}(\mathbf{k}),
$$

(6.15)

where $|V_\mathcal{F}|$ is the volume of the fundamental cell $\mathcal{F}$ of $\mathcal{L}$, $S_\mathcal{L}(\mathbf{k})$ is the structure factor of $\mathcal{L}$ given by [300]

$$
S_\mathcal{L}(\mathbf{k}) = \frac{(2\pi)^d}{|V_\mathcal{F}|} \sum_{\mathbf{q} \in \mathcal{L}^* \setminus \{0\}} \delta(\mathbf{k} - \mathbf{q}),
$$

(6.16)

$\mathcal{L}^*$ denotes the reciprocal lattice of $\mathcal{L}$, and $\delta(\mathbf{x})$ is the Dirac delta function. For a periodic packing in which a fundamental cell contains $M$ distinct particles $(\mathbf{P}_1, \cdots, \mathbf{P}_M)$ whose centroids are at $\mathbf{r}_1, \cdots, \mathbf{r}_M$, formula (6.15) can be easily extended as

$$
\tilde{\chi}_V(\mathbf{k}) = |V_\mathcal{F}|^{-1} |\tilde{m}(\mathbf{k}; \left\{\mathbf{P}_j\right\})|^2 S_\mathcal{L}(\mathbf{k}),
$$

(6.17)

where

$$
\tilde{m}(\mathbf{k}; \left\{\mathbf{P}_j\right\}) \equiv \sum_{j=1}^{M} \tilde{m}(\mathbf{k}; \mathbf{P}_j) e^{-i\mathbf{k} \cdot \mathbf{r}_j}.
$$

(6.18)

Equation (6.17) is a special case of the multicomponent packing formula given in Ref. [298]. Thus, given the form factors and structure factors for a particulate two-phase structure, one can immediately compute the corresponding spectral density via either (6.13), (6.15), or (6.17). It is crucial to note that these formulas also can be applied to any periodic cellular network by treating it as a periodic packing of polygons (polyhedra for $d = 3$) defined by the void regions (shown in white in Fig. 6.3). In such cases, the set $\left\{\mathbf{P}_j\right\}$ represents the regions of void phase (shown in white regions in Fig. 6.3).
Given the spectral density of a general packing, one can compute the local volume-fraction variance by computing Eq. (6.3) either numerically or analytically. The associated hyperuniformity order metric $B_V$ is obtained from the running average associated with (6.12). In the case of periodic packings or periodic networks, it immediately follows from Eqs. (6.3) and (6.17) that the associated local volume-fraction variance $\sigma^2_V(R)$ and the surface-area coefficient $B_V(R)$ are written as

$$\sigma^2_V(R) = \frac{2^d \Gamma(d/2 + 1)^2}{R^d} \frac{1}{|V_F|^2} \sum_{k \in \mathcal{L}^d \setminus \{0\}} |\tilde{m}(k; \{P_j\})|^2 \frac{[J_{d/2}(kR)]^2}{k^d}$$  \hspace{1cm} (6.19)

$$\sim B_V(R) \left( \frac{D}{R} \right)^{d+1}, \quad (R \rightarrow \infty).$$  \hspace{1cm} (6.20)

Thus, we see that periodic packings fall in class I. The hyperuniformity order metric $B_V$ is obtained by substituting (6.20) into (6.12):

$$\overline{B}_V = \frac{2^d \Gamma(d/2 + 1)^2}{\pi D^{d+1}} \frac{1}{|V_F|^2} \sum_{k \in \mathcal{L}^d \setminus \{0\}} \frac{|\tilde{m}(k; \{P_j\})|^2}{q^{d+1}},$$  \hspace{1cm} (6.21)

where we have used the identity $\lim_{x \rightarrow \infty} x^{-1} \int^x_0 x' \left[ J_{d/2}(x') \right]^2 = 1/\pi$.

### 6.2.3 Computation of $\sigma^2_V(R)$ and $\overline{B}_V$

Here we describe two methods that we employ to estimate the local volume-fraction variance $\sigma^2_V(R)$ and the associated asymptotic value $\overline{B}_V$: numerical integration of Eq. (6.3) and the Monte Carlo (MC) method. For periodic media, we mainly use the former method because of its accuracy and efficiency for such structures [cf. Eq. (6.20)]. The key step of this method is to compute the spectral density $\tilde{\chi}_V(k)$ of a periodic structure via either (6.15) and (6.17). For periodic packings of identical circular disks, we use the exact formula for the form factor of a $d$-dimensional sphere.
of radius $a$ given by [296]

$$
\tilde{m}(k; a) = \left( \frac{2\pi a}{k} \right)^{d/2} J_{d/2}(ka).
$$

In the case of periodic networks, we employ the formulas for general polyhedra in two and three dimensions given in Sec. 6.3. Provided that $\tilde{\chi}_V(k)$ given in Eq. (6.17) can be computed, it is in practice sufficient to perform the summations in Eqs. (6.20) and (6.21) up to $|k||V_x|^{1/d} < 1000$ for $d = 2, 3$.

Because the numerical calculations of (6.3) can be computationally expensive, we employ the MC method to estimate $\sigma^2_V(R)$ for disordered disk packings. Specifically, $\sigma^2_V(R)$ is estimated by uniformly sampling the local volume fraction with a $d$-dimensional spherical observation window of radius $R$ a single packing or an ensemble of packings. Since this method involves computing the volume of domains in one phase intersected by a window, it is highly nontrivial and computationally expensive for general packings. In the case of disk packings (sphere packings for $d = 3$), however, such a calculation can be efficiently carried out by using an exact closed-form formula for the intersection volume of two spheres of radii $R_1$ and $R_2$ whose centers are separated by $r$, given in Ref. [296].

### 6.3 Form Factors of Polygons and Polyhedra

In order to compute the spectral density of a periodic cellular network using relation (6.17), one needs to compute the form factors of the relevant polyhedra. Here, we provide the exact closed-form formulas for general polyhedra in three dimensions and two dimensions (polygons) that were derived in Ref. [337].

We first consider a planar polygon $\Gamma$ that is placed in an arbitrary orientation in $\mathbb{R}^3$ [see Fig. 6.1(a)] and consists of $J$ vertices $V_1, \cdots, V_J$ in a cyclic order, implying that the adjacent vertices $V_{i-1}$ and $V_i$ are connected by a segment, and $V_{J+1} = V_1$. 

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Figure 6.1: Illustrations of parameters used to compute the form factors of polygonal figures in $\mathbb{R}^3$. (a) A pentagon has a face $\Gamma$ surrounded by five vertices $V_1, \cdots, V_5$. The vertex indices increase counter-clockwise when the normal vector $\hat{n}$ is towards the reader. (b) A polyhedron with five faces, for each of which the ordering of vertices fulfills the right-hand-rule with the normal vector $\hat{n}_j$. Note that all normal vectors point towards the outside of the polyhedron.

It is convenient to consider a planar polygon in three dimensions since such planar polygons will be employed to define a polyhedron in $\mathbb{R}^3$ later. For two adjacent vertices $V_{i-1}$ and $V_i$, we define

$$R_i \equiv \frac{1}{2}(V_i + V_{i-1}),$$

$$E_i \equiv \frac{1}{2}(V_i - V_{i-1}),$$

where $R_i$ is the center of the two vertices, and $E_i$ stands for the segment from $V_{i-1}$ to $R_i$. The form factor of $\Gamma$ at a wavevector $k$ is

$$\tilde{m}(k; \Gamma) = \frac{2}{-i k_\|^2}k_\times \cdot \sum_{j=1}^J E_j \text{sinc}(k \cdot E_j) e^{-i k R_j},$$

where $\hat{n}$ is the unit normal vector of the face $\Gamma$, $k_\| \equiv k - (k \cdot \hat{n})\hat{n}$, $k_\times = \hat{n} \times k$, and

$$\text{sinc}(x) \equiv \begin{cases} 1, & x = 0, \\ \frac{\sin x}{x}, & \text{otherwise} \end{cases}.$$
Importantly, the ordering of vertices should fulfill the right-hand-rule with respect to the normal vector $\hat{n}$ [see Fig. 6.1(a)], implying that the vertex index increases counterclockwise when $\hat{n}$ is towards the reader. In two-dimensional applications, one should take $k = k_\parallel$.

We now consider a polyhedron $\mathcal{P}$ with $K$ faces ($\Gamma_1, \cdots, \Gamma_K$) in which a face $\Gamma_j$ is a polygon with $J_j$ vertices. For each face $\Gamma_j$ ($j = 1, \cdots, K$), its unit normal vector $\hat{n}_j$ points towards the outside of $\mathcal{P}$, and the order of vertices fulfills the right-hand-rule with $\hat{n}_j$; see Fig. 6.1(b). Then, the form factor of $\mathcal{P}$ is

$$\tilde{m}(k; \mathcal{P}) = -\frac{1}{ik^2} k \cdot \sum_{j=1}^{K} \hat{n}_j \tilde{m}(k; \Gamma_j),$$

(6.27)

where $k \equiv |k|$. The reader is referred to Ref. [337] for derivations of Eqs. (6.25) and (6.27).

6.4 Periodic Network Models

In this chapter, we consider six different periodic networks with volume fractions that span the entire interval $[0,1]$: square, rhombic, honeycomb, square-octagon, triangular, and kagomé networks. Figure 6.2 shows each of these networks at small, intermediate, and large solid volume fractions ($\phi = 0.10, 0.5, \text{and} 0.95$, respectively). Figure 6.3 provides the dimensional parameters for the unit cells, which determine the corresponding solid-phase volume fractions. Except for the kagomé and square-octagon networks, all “wall” thicknesses are uniform across all volume fractions. In the cases of the former two structures, the wall thicknesses are uniform for each different polygon but are proportional to their area ratios in order to span all volume fractions in the interval $[0,1]$. Note that in the limit of $\phi \to 1$, these six network mod-

---

1Equations (6.25) and (6.27) are the complex conjugates of the formulas derived in Ref. [337] due to the sign convention of Fourier transform.
Figure 6.2: Illustrations of the six different periodic cellular networks considered in this chapter: from top to bottom, the square, rhombic, honeycomb, square-octagon, triangular, and kagomé networks. We show each of them at three solid-phase volume fractions: $\phi = 0.10$, $\phi = 0.50$, and $\phi = 0.95$. Note that these networks can be regarded as periodic point patterns in the limit of $\phi \to 1$.

These networks can be regarded as periodic point patterns. For example, the square, honeycomb, and triangular networks become the square-lattice, triangular-lattice, and honeycomb crystal, respectively.

It is noteworthy that these cellular solids can optimize certain effective physical properties. In the limit $\phi \to 0$, these network structures maximize certain effective
transport and elastic properties. Specifically, all networks maximize the effective conductivity $\sigma_e$ and effective bulk modulus $K_e$ \[305\]. The effective shear modulus $G_e$ is maximized for the triangular network \[305, 128\] as well as the kagomé network \[49\]. The triangular and kagomé networks are nearly optimal for $\sigma_e$, $K_e$ and $G_e$ over the possible range of volume fractions \[129\]. Due to the well-known mechanisms that lead to optimality in the aforementioned networks, we can report here that the rhombic and square-octagon networks maximize the effective conductivity and effective bulk moduli in the limit of $\phi \to 0$.

Many of the periodic networks considered in this chapter can be derived from the tessellations associated with certain underlying periodic point configurations. In order to make contact with the corresponding rank ordering of class I periodic point configurations previously obtained \[313, 346\] and the rank ordering of our two-phase networks, it is instructive here to briefly describe the relationships between the point configurations and their tessellations. For example, the Voronoi tessellation associated
with points arranged on a square lattice yields the square network. The Voronoi tessellation associated with points arranged on a triangular-lattice yields the honeycomb network. The Voronoi tessellation associated with points arranged on a honeycomb crystal yields the triangular network.

6.5 Periodic and Disordered Packing Models

\[
\phi = 0.50 \quad \phi = 0.95
\]

![Illustrations of the four different models of the two-dimensional periodic dispersions of nonoverlapping identical disks considered in this chapter with different solid-phase volume fractions: \( \phi = 0.50 \) and \( \phi = 0.95 \). From top to bottom, we present dispersions associated with the square and triangular lattices and honeycomb and kagomé crystals.]

Here we consider four different two-dimensional dispersions of identical nonoverlapping circular disks on the sites of the triangular and squares lattices as well as
Figure 6.5: Representative images of the three different models of the two-dimensional disordered/irregular packings of identical circular disks considered in this chapter at different volume fractions: (a) stealthy hyperuniform packing of $\chi = 0.49$ and $\phi = 0.63$, (b) stealthy hyperuniform packing of $\chi = 0.40$ and $\phi = 0.85$, and (c) perturbed-lattice packing of $\phi = 0.79$.

Stealthy hyperuniform packings of identical particles, which are also class I, are defined by the spectral density vanishing around the origin, i.e.,

$$\tilde{\chi}_V(k) = 0, \text{ for } 0 \leq |k| \leq K.$$  \hfill (6.28)

Specifically, we first generate stealthy hyperuniform point configurations that include $N$ particles in a periodic fundamental cell $\mathcal{F}$ via the collective-coordinate optimization technique [321, 17, 352]. We then circumscribe the points by identical nonoverlapping disks [355]. For stealthy hyperuniform packings (or point patterns), it is useful to define the $\chi$ parameter, which the ratio of constrained degrees of freedom to total number of degrees of freedom [317, 352], i.e.,

$$\chi \equiv \frac{M}{d(N - 1)}.$$ \hfill (6.29)

For $0 < \chi < 1/2$, the stealthy hyperuniform point patterns are highly degenerate and
disordered, whereas for $1/2 < \chi < 1$ they crystallize [352]. Remarkably, disordered stealthy hyperuniform nonoverlapping spherical obstacles (for sufficiently high $\chi$ below 1/2) in a liquid also have nearly maximal effective diffusion coefficients as well as maximal effective thermal/electrical conductivities for perfectly insulating inclusions [355].

In this work, we numerically generate 30 different point patterns of $10^4$ particles with $\chi = 0.4$ and $\chi = 0.49$. Then we determine their corresponding largest fractions of space covered by the disks, which is equivalent to smallest possible solid-phase volume fraction $\phi_{\text{min}}$, equal to about 0.153 (i.e., $\phi \geq \phi_{\text{min}} = 0.85$) and 0.377 (i.e., $\phi \geq \phi_{\text{min}} = 0.63$), respectively.

We also generate perturbed-lattice packings by independently displacing each point of a square lattice by a random vector that is uniformly distributed in a closed square $[-a/2, a/2]^2$. We then circumscribe the resulting points by identical nonoverlapping disks. The resulting point pattern (or packing) is class I hyperuniform; see Refs. [85, 155, 160] for details. In this work, we numerically generate 50 configurations of $10^4$ particles and $a = 0.48$. We find that their largest possible fraction of space covered by the disks, which is equivalent to the smallest possible solid-phase volume fraction $\phi_{\text{min}}$, is around 0.213 (i.e., $\phi \geq \phi_{\text{min}} = 0.79$).

### 6.6 Characteristic Length Scales

When ranking class I hyperuniform systems according to the hyperuniformity order metric $\overline{B}_V$ ($\overline{B}_N$ for the point-configuration counterparts [313, 346]), it is critical to choose an appropriate characteristic length scale $D$ because these order metrics depend on $D$, as we noted in Sec. 6.2.1. In the case of hyperuniform point patterns

---

2Such point configurations are characterized by a structure factor that contains a diffuse (disordered) contribution and Bragg-diffraction (long-range order) contribution [85, 155] and therefore cannot be considered to be truly disordered, as measured quantitatively by the $\tau$ order metric [160]. For this reason, we reserve the term *irregular* for such perturbed-lattice packings.
in $\mathbb{R}^d$, it is natural to choose $D = \rho^{-1/d}$, where $\rho$ is the number density of points. However, the choice of a length scale in the case of two-phase media is highly non-trivial because the geometries and topologies of the phases are generally much richer and more complex than point configurations. Indeed, there are an infinite number of ways of decorating a point configuration to produce two-phase media, all of which cannot be universally characterized.

In this chapter, we consider and evaluate several possible choices for the length scale $D$ according to the following three criteria: (i) $D$ must be defined for general two-phase media, (ii) $D$ must be independent of the choice of phase, and (iii) the associated order metric $B_V$ must be a finite number for any volume fraction. Seemingly obvious choices for $D$, including the size of a fundamental cell for periodic systems or the mean nearest-distance for disordered or irregular packings, fail to meet the criteria (i) and (ii). There are several candidates that satisfy the criterion (i), such as the mean chord length of one phase (i.e., the expected length of line segments in the phase between the intersections of an infinitely long line with the two-phase interface [323, 309, 296]). However, criteria (ii) and (iii) immediately eliminate the mean chord length of an individual phase. The resulting $B_V$ diverges at either $\phi = 0$ or $\phi = 1$. Averages based on the mean chord length for each phase, such as the arithmetic and geometric means, satisfy all criteria. One such example is the inverse of the specific surface (i.e., the mean interface area per volume) $1/s$, which turns out to be directly proportional to the arithmetic mean of the mean chord length $\ell^{(i)}_C$ of both phases, i.e.,

$$1/s = (\ell^{(1)}_C + \ell^{(2)}_C)/\pi \ [296].$$

Explicit formulas for the specific surface $s$ of all models considered in the chapter are provided in on Appendix 6.9. Henceforth, we employ $D = s^{-1}$. 

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Figure 6.6: Log-log plots of the local volume-fraction variances \( \sigma_V^2(R) \) of two-dimensional ordered and disordered cellular solids at a selected solid-phase volume fraction \( \phi = 0.85 \): (a) honeycomb network, (b) triangular-lattice disk packing, and (c) stealthy hyperuniform packings of \( \chi = 0.4 \). The first two models are periodic structures, whereas the last is a disordered one. Here we take the inverse of the specific surface \( 1/s \) to be unity, i.e., \( D = 1/s = 1 \).

### 6.7 Results

We consider two-dimensional ordered and disordered two-phase media, shown in Figs. 6.3, 6.4, and 6.5 with taking the inverse of the specific surface as the characteristic length scale, i.e., \( D = 1/s = 1 \). Figure 6.6 shows log-log plots of local volume-fraction variances \( \sigma_V^2(R) \) as a function of window radius \( R \) at a solid-phase volume fraction \( \phi = 0.85 \) for honeycomb network, triangular-lattice packing, and disordered stealthy hyperuniform packings (with \( \chi = 0.4 \)). The variances for the periodic models and the disordered example are obtained from Eq. (6.19) and the MC method, respectively. For all models considered here, \( \sigma_V^2(R) \) globally decays as fast as \( R^{-3} \) in the large-\( R \) regime, which are of class I hyperuniformity [cf. Eq. (6.5)], and fluctuates on “microscopic” length scales, which in the case of periodic structures, are associated with the spacing of the underlying Bravais lattice.

We plot the surface-area coefficients \( B_V(R) \) for the models considered in Fig. 6.6 to more closely investigate such local fluctuations of \( \sigma_V^2(R) \); see Fig. 6.7. As pointed earlier, \( B_V(R) \) oscillates around an average value \( \overline{B_V} \). For disordered systems [shown in Fig. 6.7(c)], such oscillations typically decay as \( R \) increases, whereas for periodic networks, the amplitude of the oscillations does not decrease, even in the limit of
Figure 6.7: Surface-area coefficient $B_V(R)$ as a function of window radius $R$ of two-dimensional ordered and disordered cellular solids at a selected solid-phase volume fraction $\phi = 0.85$, as per Fig. 6.6.

$R \to \infty$.

Figure 6.8 shows how the hyperuniformity order metric $\overline{B}_V$ for two-dimensional two-phase media varies with the volume fraction $\phi$. For periodic networks or disk packings, $\overline{B}_V$ is evaluated from Eq. (6.21), whereas the disordered/irregular counterparts are evaluated by applying the running average associated with (6.12) to the MC results. For periodic networks, where $\phi$ can span from 0 to 1, as shown in Fig. 6.8(a), $\overline{B}_V$ exhibits the following three common characteristics: (i) it vanishes trivially at $\phi = 0$ and $\phi = 1$, (ii) it is proportional to $\phi^2$ for small $\phi$, and (iii) it has a maximum at around $\phi = 0.4$. By contrast, for periodic or disordered disk packings, $\overline{B}_V$ trivially vanishes at $\phi = 1$, but the other characteristics are not observed; see Fig. 6.8(b). We first investigate the rankings of $\overline{B}_V$ for periodic networks shown in Fig. 6.8(a) and those for disk packings in Fig. 6.8(b) separately and then discuss the rankings for all models. Among the considered periodic networks, honeycomb and kagomé ones achieve the minimum and maximum values of $\overline{B}_V$, respectively, at a given value of volume fraction $\phi$. For the six network models, the values $\overline{B}_V$ increases from honeycomb, square, rhombic, square-octagon, triangular, to kagomé ones. Note that the ranking for the honeycomb, square, and triangular networks are consistent with the ranking of the corresponding metrics for the point counterparts of these three network models (triangular lattice, square lattice, and honeycomb crystals, respectively,
Figure 6.8: Asymptotic values of the surface-area coefficient $\mathcal{B}_V$ as a function of the solid-phase volume fraction $\phi$ for (a) two-dimensional periodic networks and (b) ordered and disordered disk packings. We take the length scale as $D = 1/s = 1$, where $s$ is the specific surface. In (b), SHU and PLP stand for the stealthy hyperuniform packing and perturbed-lattice packing, respectively. The inset in (b) is a magnification of the larger panel.

as discussed in Sec. 6.4) given in Ref. [313, 346]. Moreover, we note that periodic networks with a single void region in the fundamental cell (honeycomb, square, and rhombic) tend to be more ordered (i.e., smaller $\mathcal{B}_V$) than those with multiple void regions in the fundamental cell (square-octagon, triangular, and kagomé).

Figure 6.8(b) shows that among the periodic disk packings, the triangular and kagomé packings achieve the minimum and maximum values of $\mathcal{B}_V$, respectively. Considering all models of disk packings, the values of $\mathcal{B}_V$ at a given volume fraction $\phi$ increases from triangular, square, disordered stealthy ($\chi = 0.49$), disordered stealthy ($\chi = 0.4$), honeycomb, kagomé, to perturbed-lattice. Similar to the case of periodic networks, the Bravais-lattice packings (triangular and square) are more ordered than non-Bravais-lattice packings (honeycomb and kagomé). Importantly, such a ranking of disk packings is identical to the rankings of the point counterparts that were reported in Ref. [346, 300], i.e., triangular, square, disordered stealthy ($\chi = 0.496$), disordered stealthy ($\chi = 0.402$), honeycomb, and kagomé.

In the discussion above, we consider the rankings for periodic network models
Table 6.1: Hyperuniformity order metric $\mathcal{B}_V$ of the six models of two-dimensional periodic cellular networks at various values of volume fraction $\phi$; see Fig. 6.3. The quantities are computed by Eq. (6.21) and taking the characteristic length scale to be the inverse of the specific surface, i.e., $D = 1/s = 1$. Note that for a given value of $\phi$, $\mathcal{B}_V$ increases from top to bottom.

<table>
<thead>
<tr>
<th>Model</th>
<th>$\phi$ = 0.1</th>
<th>$\phi$ = 0.25</th>
<th>$\phi$ = 0.4</th>
<th>$\phi$ = 0.55</th>
<th>$\phi$ = 0.7</th>
<th>$\phi$ = 0.85</th>
</tr>
</thead>
<tbody>
<tr>
<td>Honeycomb</td>
<td>2.2570 × 10^{-2}</td>
<td>9.2268 × 10^{-3}</td>
<td>1.3279 × 10^{-2}</td>
<td>1.1351 × 10^{-2}</td>
<td>5.6546 × 10^{-3}</td>
<td>1.0020 × 10^{-3}</td>
</tr>
<tr>
<td>Square</td>
<td>3.1288 × 10^{-3}</td>
<td>1.2433 × 10^{-2}</td>
<td>1.7512 × 10^{-2}</td>
<td>1.4717 × 10^{-2}</td>
<td>7.2338 × 10^{-3}</td>
<td>1.2693 × 10^{-3}</td>
</tr>
<tr>
<td>Rhombic</td>
<td>3.1320 × 10^{-3}</td>
<td>1.2519 × 10^{-2}</td>
<td>1.7846 × 10^{-2}</td>
<td>1.5301 × 10^{-2}</td>
<td>7.7541 × 10^{-3}</td>
<td>1.4222 × 10^{-3}</td>
</tr>
<tr>
<td>Square-octagon</td>
<td>3.7286 × 10^{-3}</td>
<td>1.4953 × 10^{-2}</td>
<td>2.1225 × 10^{-2}</td>
<td>1.7943 × 10^{-2}</td>
<td>8.8514 × 10^{-3}</td>
<td>1.5538 × 10^{-3}</td>
</tr>
<tr>
<td>Triangular</td>
<td>6.7514 × 10^{-3}</td>
<td>2.5523 × 10^{-2}</td>
<td>3.4413 × 10^{-2}</td>
<td>2.7739 × 10^{-2}</td>
<td>1.3071 × 10^{-2}</td>
<td>2.1926 × 10^{-3}</td>
</tr>
<tr>
<td>Kagomé</td>
<td>6.9411 × 10^{-3}</td>
<td>2.7262 × 10^{-2}</td>
<td>3.8114 × 10^{-2}</td>
<td>3.1837 × 10^{-2}</td>
<td>1.5553 × 10^{-2}</td>
<td>2.7074 × 10^{-3}</td>
</tr>
</tbody>
</table>

[Fig. 6.8(a)] and those for disk packings [Fig. 6.8(b)] separately. There, the rankings for the models in each class does not change as the solid-phase volume fraction $\phi$ is varied. However, when rankings all models in both periodic networks and periodic and disordered disk packings, the resulting rankings can change with $\phi$, and hence the volume fraction $\phi$ should be specified. For this purpose, we tabulate $\mathcal{B}_V$ for the periodic networks, periodic disk packings, and disordered disk packings at selected values of the solid-phase volume fraction $\phi$ in Tables 6.1, 6.2, and 6.3, respectively. From Tables 6.1 and 6.2, we immediately see that while the triangular disk packing is generally more ordered than the honeycomb network, their rankings change for $\phi \lesssim 0.1$. As shown in Tables 6.1-6.3, among all considered models at $\phi = 0.85$, the triangular-lattice packing and perturbed-lattice packing have the smallest and highest values of $\mathcal{B}_V$, respectively. We also note that at $\phi = 0.85$, the triangular- and square-lattice packings have lower order metrics than their network counterparts (i.e., honeycomb and square networks, respectively). This implies that the length scale $D = 1/s = 1$ penalizes the order metric $\mathcal{B}_V$ of a packing of nonspherical particles compared to the corresponding sphere packing.
Table 6.2: Hyperuniformity order metric $\overline{B}_V$ of the four models of two-dimensional periodic disk packings at various values of volume fraction $\phi$; see Fig. 6.4. The quantities are computed by Eq. (6.21) and taking the characteristic length scale to be the inverse of the specific surface, i.e., $D = 1/s = 1$. Note that for a given value of $\phi$, $\overline{B}_V$ increases from top to bottom.

<table>
<thead>
<tr>
<th>Model</th>
<th>$\overline{B}_V$ at $\phi = 0.1$</th>
<th>$\overline{B}_V$ at $\phi = 0.25$</th>
<th>$\overline{B}_V$ at $\phi = 0.4$</th>
<th>$\overline{B}_V$ at $\phi = 0.55$</th>
<th>$\overline{B}_V$ at $\phi = 0.7$</th>
<th>$\overline{B}_V$ at $\phi = 0.85$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Triangular</td>
<td>2.7760 × 10^{-3}</td>
<td>8.5612 × 10^{-3}</td>
<td>1.1794 × 10^{-2}</td>
<td>9.9491 × 10^{-3}</td>
<td>4.9261 × 10^{-3}</td>
<td>8.6942 × 10^{-4}</td>
</tr>
<tr>
<td>Square</td>
<td>–</td>
<td>1.5130 × 10^{-2}</td>
<td>1.5402 × 10^{-2}</td>
<td>1.1455 × 10^{-2}</td>
<td>5.3311 × 10^{-3}</td>
<td>9.0849 × 10^{-4}</td>
</tr>
<tr>
<td>Honeycomb</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>2.1331 × 10^{-2}</td>
<td>7.9380 × 10^{-3}</td>
<td>1.1566 × 10^{-3}</td>
</tr>
<tr>
<td>Kagomé</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>5.2200 × 10^{-2}</td>
<td>2.5962 × 10^{-2}</td>
<td>9.0773 × 10^{-3}</td>
</tr>
</tbody>
</table>

Table 6.3: Hyperuniformity order metric $\overline{B}_V$ of the three models of two-dimensional disordered disk packings at a selected volume fraction $\phi = 0.85$ and their respective lowest volume fractions $\phi_{\text{min}}$; see Fig. 6.5. The values of $\phi_{\text{min}}$ for various models are provided in Fig. 6.5. The quantities are computed by the MC procedure and Eq. (6.12) and taking the characteristic length scale to be the inverse of the specific surface, i.e., $D = 1/s = 1$. The uncertainties are estimated from the statistical errors in the estimation of $\sigma^2_V(R)$.

<table>
<thead>
<tr>
<th>Model</th>
<th>$\overline{B}<em>V$ at $\phi</em>{\text{min}}$</th>
<th>$\overline{B}_V$ at $\phi = 0.85$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Stealthy hyperuniform packings ($\chi = 0.49$)</td>
<td>8.9655(6) × 10^{-3}</td>
<td>9.4349(6) × 10^{-4}</td>
</tr>
<tr>
<td>Stealthy hyperuniform packings ($\chi = 0.4$)</td>
<td>1.0313(1) × 10^{-3}</td>
<td>1.0313(1) × 10^{-3}</td>
</tr>
<tr>
<td>Perturbed-lattice packing</td>
<td>3.6034(3) × 10^{-3}</td>
<td>1.7298(1) × 10^{-3}</td>
</tr>
</tbody>
</table>

6.8 Conclusions and Discussion

In this chapter, we took initial steps to characterize a restricted subset of class I hyperuniform two-phase media in two dimensions by ascertaining their local volume-fraction variances $\sigma^2_V(R)$ and the associated hyperuniformity order metrics $\overline{B}_V$. These models include a variety of different periodic cellular networks, periodic packings, and disordered/irregular packings, some of which maximize their effective transport and elastic properties [305, 128, 49, 129, 355]. Using the estimated $\overline{B}_V$ and a judicious choice for a length scale to make it dimensionless (as discussed below), we ranked these class I models of two-phase media according to the degree to which they suppress large-scale volume-fraction fluctuations. Among the periodic networks, the honeycomb and kagomé networks always achieve the lowest and highest $\overline{B}_V$, respectively, and the rankings do not change as the solid-phase volume fraction $\phi$ varies. Similarly,
the rankings for disk packings also do not change with $\phi$. The triangular-lattice pack-
ings (whose Voronoi tessellations are honeycomb networks) and the perturbed-lattice pack-
ings have the minimum and maximum values of $\overline{B}_V$, respectively. Not surpris-
ingly, however, the overall rankings for both network and packing models with their
distinctly different geometries and topologies are difficult to unscramble because they
change with $\phi$. Nonetheless, we summarize these rankings by making two general
observations. First, the rankings for packings of identical disks are consistent with
those of the point-configuration order metric $\overline{B}_N$ corresponding to their underlying
point patterns [313, 346, 300] at any considered volume fraction $\phi$. Second, for both
periodic networks and periodic packings with the same underlying Bravais lattice, the
structures with smaller specific surfaces have lower values of $\overline{B}_V$. The same tendency
is also observed in two different three-dimensional models; see Appendix 6.10. We
note that the second observation is generally true, with a few notable exceptions in
which the volume fraction of the solid phase becomes so low that the disks are nearly
in contact with one another. Specifically, among all models considered in this chap-
ter, triangular-lattice packing has the minimal $\overline{B}_V$ for all solid-phase volume fraction
greater 0.1. Otherwise, the honeycomb networks record the smallest value.

When establishing these rankings according to $\overline{B}_V$, it is crucial to determine a
characteristic length scale $D$ to make $\overline{B}_V$ dimensionless, which is highly nontrivial
due to the need to account for a wide spectrum of two-phase structures. Among vari-
ous possibilities, we chose the inverse of the specific surface $1/s$ as the length scale $D$
by considering the three criteria of generality, phase-independence, and boundedness
of the associated $\overline{B}_V$. This choice is also reasonable in that $s$ is easy to compute.
Furthermore, it is one of the Minkowski functionals (i.e., volume, surface area, inte-
grated mean curvature, and Euler number), which are fundamental shape descriptors
that have been widely used in various applications [262, 162]. The integrated mean
curvature might also serve as a choice of the length scale $D$. Although we have made
a specific choice $D = s^{-1}$, we note that one can easily convert our results for $\overline{B}_V$ to the corresponding quantity for any another length scale $D = \ell$ by use of the relation

$$\overline{B}_V|_{D=\ell} = \overline{B}_V/(s\ell)^{d+1},$$

where $d$ is the space dimension.

Our study lays the theoretical foundation to establish the hyperuniformity order metrics of more general two-phase systems. Towards this end, one needs to develop methods to estimate $\sigma_V^2(R)$ for a wider class of hyperuniform two-phase media than what can be handled by the methods used in this chapter, such as labyrinth-like patterns associated with spinodal decomposition [194]. Such a development will also be beneficial in detecting (effective) hyperuniformity of relatively small systems, in which the asymptotic analysis of $\sigma^2_V(R)$ [cf. (6.2)] is more reliable than the spectral-density condition (6.1) [64]. Further studies in three and higher dimensions will be helpful in determining whether $\overline{B}_V$ scaled by $D = 1/s$ is a robust order metric. It would also be interesting to know whether the two-phase counterpart of the decorrelation principle [315, 350] for disordered two-phase media could be observed as the space dimension increases.

It will be of interest to determine whether hyperuniformity of fluctuations associated with the two-phase interface [299] leads to the same rank ordering as for volume-fraction fluctuations for the models considered in this investigation. Another promising avenue for future study is the construction of two-phase structures with a prescribed value of $\overline{B}_V$. This problem can be regarded as a type of Fourier-space based inverse design procedure [46], in which Eq. (6.21) is taken as the objective function. Such a procedure can be employed in discovering new types of periodic structures with a specified value of $\overline{B}_V$. The algorithm developed to solve this problem would also provide a tool for determining whether the triangular-lattice disk packing, which
we demonstrated minimizes $\mathcal{B}_V$ among the considered models, is a global minimizer for $\mathcal{B}_V$ among a larger class of models. An interesting question is what physical properties are optimized by the global minimizer of $\mathcal{B}_V$ under certain constraints.

### 6.9 Appendix A: Specific Surface for Various Models of Two-Phase Media

Table 6.4: Formulas for the specific surface $s$ for all models considered in this chapter. For periodic networks and disk packings, the specific surface can be expressed as $s = C\sqrt{1 - \phi/L_1}$, where $L_1$ is a length parameter of the unit cells; see Fig. 6.5. For any disordered/irregular disk packing of the number density $\rho$, the specific surface is written as $s = C\sqrt{1 - \phi\rho^{1/2}}$.

<table>
<thead>
<tr>
<th>Models</th>
<th>$C$</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Periodic networks</strong></td>
<td></td>
</tr>
<tr>
<td>Honeycomb</td>
<td>4</td>
</tr>
<tr>
<td>Square</td>
<td>4</td>
</tr>
<tr>
<td>Rhombic</td>
<td>$8/\sqrt{3}$</td>
</tr>
<tr>
<td>Square-octagon</td>
<td>$12/(1 + \sqrt{2})$</td>
</tr>
<tr>
<td>Triangular</td>
<td>$4\sqrt{3}$</td>
</tr>
<tr>
<td>Kagomé</td>
<td>$4\sqrt{3}$</td>
</tr>
<tr>
<td><strong>Periodic disk packings</strong></td>
<td></td>
</tr>
<tr>
<td>Triangular</td>
<td>$2\sqrt{2\pi/3^{1/4}}$</td>
</tr>
<tr>
<td>Square</td>
<td>$2\sqrt{\pi}$</td>
</tr>
<tr>
<td>Honeycomb</td>
<td>$4\sqrt{\pi/3^{1/4}}$</td>
</tr>
<tr>
<td>Kagomé</td>
<td>$2\sqrt{2\pi/3^{1/4}}$</td>
</tr>
<tr>
<td><strong>Disordered disk packing</strong></td>
<td>$2\sqrt{\pi}$</td>
</tr>
</tbody>
</table>

Table 6.4 provides the formulas for the specific surface $s$ for all two-dimensional models of class I hyperuniform two-phase media considered in this chapter. We note that all disk packings, ordered or not, have the same specific surface if they are at the same number density $\rho$. 

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6.10 Appendix B: Hyperuniformity Order Metrics of Three-Dimensional Simple-Cubic Lattice Packings

Here, we present some preliminary results of two models of three-dimensional periodic dispersions consisting of identical nonoverlapping void regions on the simple cubic lattice sites. One model is a packing of identical spheres (sphere packing), and the other one is a packing of identical cubes (cube packing). The solid-phase volume fractions of these sphere packings and cube packings span the closed intervals $[1 - \pi/6, 1]$ and $[0, 1]$, respectively. We obtain the surface-area coefficients $B_V(R)$ and their asymptotic values $\overline{B}_V$ for these two models by using Eqs. (6.20) and (6.21), respectively; see Fig. 6.9.

Figure 6.9(a) shows that $B_V(R)$ of each packing oscillates around an average value $\overline{B}_V$, and the amplitude of the oscillations does not decrease even in the limit of $R \to \infty$. Such behaviors are consistent with what we observed in Fig. 6.7(a-b) for two-dimensional periodic models. Figure 6.9(b) shows how $\overline{B}_V$ of these three-dimensional models varies with $\phi$. For the cube packing, similar to two-dimensional periodic networks, $\overline{B}_V$ vanishes at $\phi = 0$ and $\phi = 1$ and is proportional to $\phi^2$. For the sphere packing, $\overline{B}_V$ vanishes at $\phi = 1$. For both cube and sphere packings at a given value of $\phi$, the sphere packing always has a lower value of $\overline{B}_V$ than the cube one. Since a sphere has a smaller surface area than a cube of the same volume, this observation is consistent with the second general observation that we discussed in Sec. 6.8.
Figure 6.9: Numerical results for simple-cubic lattice packings of nonoverlapping spheres and cubes. (a) Surface-area coefficient $B_V(R)$ as a function of window radius $R$ at a selected solid-phase volume fraction $\phi = 0.9$. (b) Asymptotic values of the surface-area coefficient $\overline{B}_V$ as a function of the solid-phase volume fraction $\phi$. In both panels, we take the inverse of the specific surface $1/s$ to be unity, i.e., $D = 1/s = 1$. 

\begin{equation}
D = 1/s = 1
\end{equation}

\begin{equation}
\phi = 0.9
\end{equation}
Chapter 7

Tessellation-Based Procedure to Design Perfectly Hyperuniform Disordered Dispersions for Materials Discovery

7.1 Introduction

The notion of hyperuniformity was first defined in the context of point-particle systems [313] and then extended to two-phase heterogeneous systems [346] and random scalar/vector fields [299]. General two-phase systems abound in natural and artificial materials, including colloidal suspensions, particulate composites, and concrete [31, 328, 296, 228, 218, 255, 320, 365, 213, 205]. Packings (or dispersions), which are of central concern in this chapter, comprise a class of two-phase systems in which nonoverlapping particles are spatially distributed throughout a connected “void” (matrix) phase.

A hyperuniform two-phase system in $d$-dimensional Euclidean space $\mathbb{R}^d$ is one in
Figure 7.1: (a-b) Portions of initial disordered tessellations: (a) Voronoi tessellation (black lines) of a nonhyperuniform packing (Sec. 7.5) and (b) a multiscale-disk tessellation (Sec. 7.6). A progenitor disk packing in (a) is illustrated by white dashed circles. The tessellation-based procedure can be applied to higher dimensions, but we illustrate two-dimensional cases here for simplicity. (c-d) Portions of disordered hyperuniform packings (dispersions) constructed from the initial tessellations (a-b) via the tessellation-based procedure, i.e., local-cell packing fraction $\phi$ of a particle (blue disks) within each cell is identical to the global packing fraction. Importantly, (d) multiscale coated-disks model corresponds to optimal Hashin-Shtrikman structures.

which the local volume-fraction variance $\sigma_V^2(R)$ inside a spherical observation window of radius $R$ decays faster than $R^{-d}$ in the large-$R$ limit [346, 300]:

$$\lim_{R \to \infty} \nu_1(R) \sigma_V^2(R) = 0. \quad (7.1)$$

Equivalently, its associated spectral density $\tilde{\chi}_V(k)$ vanishes as the wavenumber $|k|$
tends to zero [346, 300]. For disordered hyperuniform two-phase systems, \( \tilde{\chi}_V(k) \) typically exhibits the power-law scaling:

\[
\tilde{\chi}_V(k) \sim |k|^{\alpha} \quad (\alpha > 0).
\]

This exponent \( \alpha \) is directly related to three distinct classes of hyperuniformity that are categorized based on the large-\( R \) scalings of \( \sigma_V^2(R) \) [300]:

\[
\sigma_V^2(R) \sim \begin{cases} 
R^{-(d+1)}, & \alpha > 1 \quad (\text{class I}) \\
R^{-(d+1)} \ln R, & \alpha = 1 \quad (\text{class II}) \\
R^{-d-\alpha}, & \alpha < 1 \quad (\text{class III})
\end{cases}
\]

where classes I and III represent the strongest and weakest forms of hyperuniformity, respectively. Class I systems include all crystals, some quasicrystals, and stealthy hyperuniform systems in which \( \tilde{\chi}_V(k) = 0 \) for \( 0 < |k| < K \) [17, 352, 353, 317, 101, 181].

Disordered hyperuniform two-phase systems [346, 355] are attracting considerable attention due to their unusual physical properties, such as complete isotropic photonic and phononic bandgaps [80, 82, 102, 186], nearly optimal transport properties [355, 46, 175], superior metamaterial designs [336], dense but transparent materials [179], and low-density materials of blackbody-like absorption [24]. Similar to ordinary two-phase systems, their effective properties are also tunable by engineering the phase properties and volume fractions as well as the spatial arrangements [80, 82, 242, 46, 304, 355, 336]. An important class of two-phase systems are Hashin-Shtrikman structures [113, 296, 310, 209, 213] that are optimal for effective thermal (electrical) conductivity, elastic moduli, and fluid permeability [310] for given phase volumes and phase properties. Remarkably, certain disordered hyperuniform systems possess nearly optimal transport and elastic properties [355, 46, 304, 303]. Although effective
hyperuniformity\(^1\) is sufficient to exhibit desirable physical properties, it has been known that driving systems into perfect hyperuniformity will further improve their performance \([116, 81]\).

Theoretical \([259, 122, 287, 184, 45]\), numerical \([348, 307, 189, 190, 10, 321, 355, 46, 61, 324]\), and experimental methods \([329, 169, 245, 364]\) have been developed to generate disordered hyperuniform packings (dispersions). In practice, however, these methods are system-size limited due to computational cost or imperfections. For instance, the collective-coordinate optimization technique \([321, 355, 46, 61]\) and equilibrium plasma \([135, 184, 45]\) can achieve perfect hyperuniformity, but their long-range interactions lead the computation cost to grow rapidly with system size. Random organization models \([122, 287]\) yield disordered hyperuniform packings at critical absorbing states but are limited in producing perfect hyperuniformity because of critical slowing-down phenomena \([10, 300]\). Determinantal point processes are perfectly hyperuniform in the thermodynamic limit, but the current numerical algorithm can hardly generate a realization of more than 100 particles due to accumulated numerical error \([259]\). Stealthy designs via the superposition procedure \([61]\) provide an efficient means to construct exactly stealthy hyperuniform digitized two-phase systems. However, this construction scheme requires one to prepare many different small systems as building blocks, which is computationally demanding as the system size increases.

Since hyperuniformity is a global property of an infinitely large system, limited sample sizes often make it difficult to ascertain whether such systems are truly hyperuniform or effectively hyperuniform. Furthermore, as discussed in Chapter 3, disordered hyperuniform systems often can include imperfections, such as point vacancies, stochastic displacements \([85]\), thermally excited phonon modes \([155]\), and rattlers \([10]\) in maximally random jammed (MRJ) packings. Such imperfections can either destroy or degrade the hyperuniformity (even if by a small amount) of other-

\(^1\)It implies that \(\tilde{\chi}_V(0)\) is not exactly zero but small compared to the peak value of the spectral density \([300, 10]\)
wise perfectly hyperuniform systems [155, 300]. Hence, there is a great need to devise exact and efficient procedures to construct extremely large realizations of disordered hyperuniform two-phase systems. Such capability could then be combined with state-of-the-art photolithographic and 3D printing techniques to fabricate large disordered hyperuniform systems.

In this chapter, we introduce a tessellation-based procedure that enables one to generate disordered packings in $\mathbb{R}^d$ that are perfectly hyperuniform in the infinite-system-size limit. Based on this theoretical methodology, we formulate an efficient algorithm to generate extremely large realizations of hyperuniform packings. Specifically, one first tessellates the space into certain disjoint cells that meet a bounded-cell condition, i.e., the maximal length of each cell should be much shorter than the side length of the entire tessellation. This mild restriction allows one to employ a wide class of tessellations, as discussed below and in Sec. 7.5. Then, one places hard particles in each cell such that the local packing fractions associated with the cells become identical to the global packing fraction $\phi$; see Fig. 7.1. Note that for “periodic” tessellations (e.g., Voronoi tessellations of Bravais lattices and Weaire-Phelan foam [327, 50]), our procedure yields periodic packings that are stealthy hyperuniform. However, our major concerns in this chapter are disordered tessellations whose cells have variability in sizes and shapes. Thus, the application of our procedure to such tessellations generates “disordered” hyperuniform packings in which the particles have a polydispersity in size. We present a detailed theoretical analysis of the small-wavenumber scalings of the spectral density $\tilde{\chi}(k)$ for the constructed packings in the case of arbitrary particle shapes. We thereby prove that whenever the initial tessellations meet the bounded-cell condition, these systems are strongly hyperuniform (class I) for any particle shape in the infinite-system-size limit. In this limit, the system size tends to be infinitely large with other intensive parameters (e.g., number density and packing fractions) held fixed. This procedure is a packing protocol to
generate packings of polydisperse particles that is uniquely different from previously known methods [301].

As a proof-of-concept, we verify our theoretical results by numerically constructing packings from certain initial tessellations, and by ascertaining their hyperuniformity from the spectral densities. Our procedure allows for the use of any initial tessellation, including Voronoi tessellations [296] and their generalizations [94, 130], sphere tessellations, disordered isoradial graphs [28], dissected tessellations [84], Delaunay triangulations, and “Delaunay-centroidal” tessellations [80, 303], as long as it meets the bounded-cell condition. For concreteness and simplicity, two types of initial tessellations are considered in this chapter: Voronoi tessellations (Sec. 7.5) and sphere tessellations, which are necessarily multi-scale divisions of space (Sec. 7.6). Employing Voronoi tessellations of general disordered point patterns in $\mathbb{R}^2$ and $\mathbb{R}^3$, we demonstrate that our methodology enables a remarkable mapping that converts extremely large nonhyperuniform packings (as large as $10^8$ particles) into hyperuniform ones. To carry out our simulations, we employ various statistically homogeneous progenitor systems. Based on the same idea, we establish the hyperuniformity of the aforementioned optimal Hashin-Shtrikman multiscale coated-spheres structures [113, 296, 310] (see Fig. 7.1(d)). Here, we provide a detailed derivation of $\tilde{\chi}_V^{(m)}(k)$ in the $m$th stage and numerical simulations for two distinct types of cell-volume distributions. It is noteworthy that our methodology only involves calculating cell volumes, which is exactly performed and easy to parallelize. Furthermore, we demonstrate that large samples of many of our designs can be readily fabricated via modern photolithographic and 3D printing techniques [334, 319, 269, 360] (Sec. 7.7). In this chapter, we treat a broader class of sphere packings as well as packings of nonspherical particles and provide detailed mathematical derivations. We also report associated simulation results that are not contained in Ref. [157].

We present basic mathematical definitions and concepts in Sec. 7.2. Then, we pre-
cisely describe the tessellation-based procedure in Sec. 7.3. In Sec. 7.4 we derive the small-$k$ scalings of the spectral densities for the constructed packings. Subsequently, we verify our theoretical results by numerical simulations using Voronoi tessellations and sphere tessellations in Secs. 7.5 and 7.6, respectively. Then, we discuss the feasibility of fabricating our designs in modern technologies in Sec. 7.7. Finally, we provide concluding remarks in Sec. 7.8.

### 7.2 Background and Definitions

The microstructure of a two-phase system can be described by the *phase indicator function* associated with phase $i = 1, 2$ [296]:

\[
I^{(i)}(r) = \begin{cases} 
1, & r \in \text{phase } i \\
0, & \text{otherwise}.
\end{cases}
\]  

(7.4)

If the system is statistically homogeneous, its one-point correlation function is independent of position $r$, and identical to the phase-volume fraction $\phi_i$, i.e., $\langle I^{(i)}(r) \rangle = \phi_i$, where $\langle \cdot \rangle$ represents an ensemble average. The *autocovariance* function can be defined in terms of the mean-zero fluctuating indicator function, $J^{(i)}(r) \equiv I^{(i)}(r) - \phi_i$, as follows:

\[
\chi_V(r) \equiv \left\langle J^{(i)}(r') J^{(i)}(r' + r) \right\rangle,
\]  

(7.5)

which is identical for each phase and tends to zero as $r$ increases if the system does not have long-range order. Its Fourier transform $\tilde{\chi}_V(k)$, called the *spectral density*, is a nonnegative real-valued function of a wavevector $k$. In experiments, the spectral densities are directly obtainable from elastic scattering intensities [54] when the wavelength of radiation is larger than atomic distance, but shorter than the length scale of domains. In numerical simulations, the spectral densities are
calculated from realizations of the media under the periodic boundary conditions as follows:
\[
\tilde{\chi}_V(k) = \frac{1}{|V_F|} \left\langle \left| \tilde{\mathcal{J}}^{(i)}(k) \right|^2 \right\rangle, \tag{7.6}
\]
where \(|V_F|\) is the volume of the simulation box, a wavevector \(k\) is a reciprocal lattice vector of the simulation box, and \(\langle \cdot \rangle\) represents an ensemble average, where \(\tilde{\mathcal{J}}^{(i)}(k)\) is the Fourier transform of \(\mathcal{J}^{(i)}(r)\) [295].

In the context of two-phase media, a packing can be regarded as domains of a “particle” phase (\(N\) generally shaped particles \(P_1, P_2, \ldots, P_N\)) that are dispersed throughout a continuous “matrix” (void) phase. For such a packing in a periodic fundamental cell \(V_F\), the random variable \(\tilde{\mathcal{J}}(k)\) associated with the particle phase (dropping the superscript) can be expressed as follows:
\[
\tilde{\mathcal{J}}(k) = N \sum_{j=1}^{N} \tilde{m}(k; P_j) e^{-ik \cdot r_j} - \phi \int_{V_F} dy e^{-ik \cdot y} \tag{7.7}
\]
\[
= \sum_{j=1}^{N} \tilde{m}(k; P_j) e^{-ik \cdot r_j} - \phi |V_F| \delta_{k,0}, \tag{7.8}
\]
where \(r_j\) is the centroid of \(P_j\), \(\tilde{m}(k; P_j)\) is its form factor (i.e., the Fourier transform of the “particle indicator function” \(m(r; P_j)\) with respect to \(r_j\)), and \(\delta_{k,0}\) represents a Kronecker delta symbol. Since the forward scattering term \(\phi |V_F| \delta_{k,0}\) in Eq. (7.8) always vanishes at nonzero reciprocal lattice vectors \(k\)’s, this term is often ignored in numerical calculations.

For some special particle shapes, closed-form expressions of \(\tilde{m}(k; P_j)\) are known. For a spherical particle of radius \(a\), the associated form factor is
\[
\tilde{m}(k; a) = \frac{(2\pi a/k)^{d/2}}{J_{d/2}(ka)}, \tag{7.9}
\]
where \(k \equiv |k|\) and \(J_n(x)\) is the Bessel function of order \(n\). For a cubic particle of side
length $L$, the corresponding function is

$$
\tilde{m}(\mathbf{k}; L) = L^d \prod_{l=1}^{d} \text{sinc}(k_l L/2),
$$

(7.10)

where $k_l$ represents $l$th component of a wavevector $\mathbf{k}$ and

$$
\text{sinc}(x) \equiv \begin{cases} 
\frac{\sin x}{x}, & x \neq 0 \\
1, & x = 0.
\end{cases}
$$

(7.11)

We note that Eqs. (7.9) and (7.10) have their global maxima at the origin, whose values are identical to their particle volumes.

From Eqs. (7.6) and (7.7), one can straightforwardly derive an expression of the spectral density for sphere packings of the identical particle radius $a$ [296, 299]:

$$
\tilde{\chi}_V(\mathbf{k}) = \rho |\tilde{m}(\mathbf{k}; a)|^2 S(\mathbf{k}),
$$

(7.12)

where $\rho$ is number density and $S(\mathbf{k})$ is the structure factor defined as

$$
S(\mathbf{k}) \equiv \frac{1}{N} \left\langle \left| \sum_{j=1}^{N} e^{-i\mathbf{k} \cdot \mathbf{r}_j} - N \delta_{\mathbf{k},0} \right|^2 \right\rangle.
$$

(7.13)

Equation (7.12) gives

$$
\tilde{\chi}_V(0) = \phi^2 S(0)/\rho,
$$

(7.14)

and thus, one obtains a hyperuniform packing by decorating a hyperuniform point pattern with spheres of an equal size [299]. In this chapter, however, we will not discuss such hyperuniform constructions.

In the case of a one-component many-particle system in thermal equilibrium in which the particles do not overlap, the fluctuation-compressibility relation $S(0) =$
\( \rho \kappa T k_B T \) [296, 108] and Eq. (7.14) yield

\[
\tilde{\chi}_V(0) = \phi^2 \kappa T k_B T,
\]

(7.15)

where \( \kappa T \) is the isothermal compressibility, \( k_B \) is the Boltzmann constant, and \( T \) is temperature. Equation (7.15) implies that any compressible (\( \kappa T > 0 \)) one-component system in thermal equilibrium cannot be hyperuniform at a positive temperature [317, 300].

The local volume-fraction variance \( \sigma^2_V(R) \) associated with spherical windows of radius \( R \) is defined as [300, 346]

\[
\sigma^2_V(R) \equiv \langle \tau^2(x; R) \rangle - \phi^2,
\]

(7.16)

where \( \tau(x; R) \) denotes the local volume fraction of the particle phase inside the spherical window of radius \( R \) centered at position \( x \).

From numerical simulations alone, it is difficult to ascertain whether a system is perfectly hyperuniform because the infinite-system-size limit is never achievable and \( \tilde{\chi}_V(k) \) usually has large relative statistical uncertainties at small wavenumbers. For these reasons, it is desirable to employ alternative criteria to determine whether a system is effectively hyperuniform. A useful empirical criterion to deem a system to be hyperuniform is that the hyperuniformity metric \( H \) is less than \( 10^{-2} \) or \( 10^{-3} \) [10, 300, 45], where \( H \) is defined by

\[
H \equiv \frac{\tilde{\chi}_V(\mathbf{k} \to 0)}{\tilde{\chi}_V(\mathbf{k}_{\text{peak}})},
\]

(7.17)

where \( \tilde{\chi}_V(\mathbf{k}_{\text{peak}}) \) is the spectral density at the first dominant (non-Bragg) peak. Note that this criterion is different from its counterpart for point patterns because of the presence of the form factor in the spectral density [45]; see Eq. (7.12).
7.3 Tessellation-Based Procedure

Figure 7.2: Schematics illustrating the tessellation-based procedure to construct disordered hyperuniform dispersions. (a) One first tessellates the space with disjoint cells whose maximal lengths are shorter than a certain length scale $\ell_{\text{max}} (\ll L)$, where $|C_j|$ represents the volume of cell $j$. (b) For a specified packing fraction $0 < \phi < 1$, within each cell $C_j$, one places a single particle of general shape of volume $\phi |C_j|$. (c) A special case of constructed dispersions with circular disks and a circular observation window of large radius $R (\gg \ell_{\text{max}})$. The volume-fraction fluctuations arise only inside a narrow yellow-shaded region whose effective thickness is thinner than $\ell_{\text{max}}$. (d) The corresponding hyperuniform dispersion of randomly oriented squares. The volume-fraction fluctuations arise only inside a narrow yellow-shaded region like (c).

Here, we precisely describe the tessellation-based procedure. Consider a hypercubic simulation box of side length $L$ in $d$-dimensional Euclidean space $\mathbb{R}^d$ under the periodic boundary conditions. As graphically illustrated in Fig. 7.2, the procedure consists of the following steps:

1. Divide the simulation box with $N$ disjoint cells $C_1, \cdots, C_N$ [Fig. 7.2(a)] in
which the maximal characteristic linear cell size \( \ell_{\text{max}} \equiv \max_{j=1}^N \{ \max_{\mathbf{r},\mathbf{r}' \in \mathcal{C}_j} \{ |\mathbf{r} - \mathbf{r}'| \} \} \) is much smaller than \( L \), i.e.,

\[
\ell_{\text{max}} \ll L. \tag{7.18}
\]

We call this the “bounded-cell” condition.

2. For a specified local-cell packing fraction \( 0 < \phi < 1 \), place hard particles of arbitrary shapes of total volume \( \phi |\mathcal{C}_j| \) within the \( j \)th cell \( \mathcal{C}_j \), and then repeat the same process over all cells; see Fig. 7.2(b) for illustrative example with a single particle.

Applying this procedure results in a packing in which the local-cell packing fraction \( \phi \) is identical to global packing fraction. Given an initial tessellation, this construction is realizable only when the local-cell packing fraction \( \phi \) in the step 2 is smaller than or equal to the maximal packing fraction \( \phi_{\text{max}} \), i.e.,

\[
\phi \leq \phi_{\text{max}} \equiv \min_{j=1}^N \left\{ \frac{|\mathcal{P}_j|_{\text{max}}}{|\mathcal{C}_j|} \right\}, \tag{7.19}
\]

where \( |\mathcal{C}_j| \) and \( |\mathcal{P}_j|_{\text{max}} \) represent volumes of the \( j \)th cell and the largest particle of a certain shape which is inscribed in this cell, respectively.

Roughly speaking, the maximal packing fraction \( \phi_{\text{max}} \) becomes larger when each cell tends to fully enclose a larger particle. Clearly, the largest particle that a cell can fully circumscribe should be congruent to the cell, and thus the maximal packing fraction will take its largest value \( (\phi_{\text{max}} = 1) \) only if the shape of each particle is similar to its circumscribing cell. For this reason, when only considering spherical particles, packing fraction of the multiscale coated-disks model (Fig. 7.1d) can span up to unity, i.e., \( \phi_{\text{max}} = 1 \).

The rationale behind our methodology can be intuitively understood by considering how the volume of the particle phase within a spherical window of radius \( R \) fluctuates around its global mean value \( \phi v_1(R) \). As shown in Fig. 7.2(c), our method-
ology ensures that the cells only in the yellow-shaded region can contribute to the fluctuations, which are proportional to the volume of this boundary region. Since the bounded-cell condition ensures that the thickness of this boundary region is smaller than $\ell_{\text{max}}$, the resulting variance in local phase-volume grows on the order of $R^{d-1}$ for sufficiently large windows ($R \gg \ell_{\text{max}}$). This observation implies that $\sigma_R^2(R) \sim R^{d-1}$, meaning that this packing is strongly hyperuniform (class I). This rationale also explains the hyperuniformity of the constructed packings, regardless of shapes [as shown in Fig. 7.2(d)] and number of particles inside each cell. However, for simplicity, we henceforth focus on the cases in which each cell contains exactly one particle.

### 7.4 General Theoretical Analyses

Here, we derive an asymptotic expression for the spectral density for the constructed packings in the small-wavenumber limit. Consider an initial tessellation $\{C_j\}_{j=1}^N$ of a cubic periodic fundamental cell $V_F$ of side length $L$ in $\mathbb{R}^d$. Since a fundamental cell is the union of all cells $C_1, \cdots, C_N$ of the tessellation, the Fourier transform of $V_F$ can be decomposed as follows:

$$
|V_F| \delta_{k,0} = \int_{V_F} \text{d}y \, e^{-i \mathbf{k} \cdot \mathbf{y}} = \int_{V_F} \text{d}y \, e^{-i \mathbf{k} \cdot \mathbf{y}} \sum_{j=1}^N m(y - \mathbf{x}_j; C_j)
$$

$$
= \sum_{j=1}^N e^{-i \mathbf{k} \cdot \mathbf{x}_j} \tilde{m}(\mathbf{k}; C_j),
$$

where $\delta_{k,0}$ represents the Kronecker delta symbol, $\mathbf{k}$ is a reciprocal lattice vector of $V_F$, and for the $j$th cell $C_j$, $\mathbf{x}_j$, $m(r; C_j)$, and $\tilde{m}(\mathbf{k}; C_j)$ represent its centroid, the indicator function with respect to $\mathbf{x}_j$, and the form factor, respectively.

The application of our procedure to this tessellation yields a particle packing that consists of $N$ particles $P_1, \cdots, P_N$ whose centroids are $\mathbf{r}_1, \cdots, \mathbf{r}_N$, respectively, with the identical local-cell packing fraction $\phi$. Using the decomposition of Eq. (7.20) and
the spectral density given in Eqs. (7.6) and (7.7), we obtain the following general expression:

$$\tilde{\chi}_V(k) = \frac{\left| \tilde{J}(k) \right|^2}{|V_F|^2} = \frac{1}{|V_F|^2} \left| \sum_{j=1}^{N} e^{-ik \cdot x_j} \left\{ \tilde{m}(k; P_j) e^{-ik \cdot \Delta X_j} - \phi \tilde{m}(k; C_j) \right\} \right|^2, \quad (7.21)$$

where $\Delta X_j \equiv r_j - x_j$, and $\tilde{m}(k; P_j)$ is the form factor of a particle $P_j$.

Due to the bounded-cell condition, $|r| < \ell_{\text{max}} \ll L$ for every $r$ in each cell $C_j$, or equivalently, one can consider small wavevectors satisfying that $|k \cdot r| \sim |r|/L \ll 1$.

Thus, the form factor of $V = (C_j$ or $P_j)$ can be well approximated by its Taylor series about $k = 0$:

$$\tilde{m}(k; V) = |V| \left[ 1 - \frac{k_\alpha k_\beta}{2} M_{\alpha\beta}(V) + \frac{ik_\alpha k_\beta k_\gamma}{6} M_{\alpha\beta\gamma}(V) \right] + \mathcal{O}(k^4), \quad (7.22)$$

where the Einstein summation convention is employed,

$$M_{\alpha_1\alpha_2\cdots\alpha_n}(V) \equiv \frac{1}{|V|} \int_V \text{d}r r_{\alpha_1} r_{\alpha_2} \cdots r_{\alpha_n}, \quad (7.23)$$

is the $m$th moment of the mass distribution of $V (= C_j$ or $P_j)$ that is normalized by its volume $|V|$, and $r_{\alpha_j}$ represents the $\alpha_j$th Cartesian component of a vector $r$. We note that since Eq. (7.23) refers to the moments with respect to the centroid of $V$, its first moment is identically zero.

Using Eq. (7.22), the term $\tilde{J}(k)$ given in Eq. (7.21) can be written as follows (see Appendix 7.12 for details):

$$\tilde{J}(k) = \tilde{J}_1(k) + \tilde{J}_2(k) + \tilde{J}_3(k) + \mathcal{O}(k^4), \quad (7.24)$$
where

\[ \tilde{J}_{(1)}(k) = \phi \sum_{j=1}^{N} (e^{-ik\Delta X_j} - 1) |C_j| e^{-ikx_j}, \quad (7.25) \]

and \( \tilde{J}_{(2)}(k) \) and \( \tilde{J}_{(3)}(k) \) are defined in Eq. (7.55). Since the leading order terms of \( \tilde{m}(k; P_j) \) and \( \tilde{m}(k; C_j) \) exactly cancel each other due to the local constraint \( |P_j| = \phi |C_j| \) for all \( j \), these three terms \( \tilde{J}_{(1)}(k) \), \( \tilde{J}_{(2)}(k) \), and \( \tilde{J}_{(3)}(k) \) exhibit the power-law scalings in the small-wavenumber limit. Assuming typical tessellations that exhibit \(|\tilde{J}_{(2)}(k)| \sim |k|^2\) and \(|\tilde{J}_{(3)}(k)| \sim |k|^3\), the first term can have two scalings (either \(|\tilde{J}_{(1)}(k)| \sim |k|\) or \(\tilde{J}_{(1)}(k) \sim O(|k|^2)\)), depending on the particle displacements \( \Delta X_j \) with respect to their cell centroids. Therefore, the scaling of \( \tilde{J}_{(1)}(k) \) determines that of the spectral density (7.21).

Specifically, whenever \(|\tilde{J}_{(1)}(k)| \sim |k|\), which can be achieved when particle displacements \( \Delta X_j \) are uncorrelated with one another \(^2\), the spectral density of the constructed packings tends to zero quadratically in \(|k|\); specifically,

\[ \tilde{\chi}_V(k) \sim \phi^2 |k|^2, \quad (7.26) \]

which corresponds to class I hyperuniformity. When \(|\tilde{J}_{(1)}(k)| \sim O(|k|^2)\), the packings are more strongly hyperuniform with a new scaling given by

\[ \tilde{\chi}_V(k) \sim \phi^2 |k|^4. \quad (7.27) \]

For example, this scaling can be achieved when \( \Delta X_j = 0 \) for all \( j \) (i.e., \( \tilde{J}_{(1)}(k) = 0 \)) or when \(|\tilde{J}_{(1)}(k)| \sim |k|^2\) due to certain spatial correlations in \( \Delta X_j \) (see Sec. 7.5). Therefore, the manipulation of particle displacements \( \Delta X_j \) enables us to engineer either quadratic or quartic scalings of the spectral density.

\(^2\)One such set of structures are perturbed crystal packing that result from stochastically displacing particles within a cell of a periodic Voronoi tessellation \([85, 155]\).
It is noteworthy that the appearance of \( \phi^2 \) factor in Eqs. (7.26) and (7.27) is common for small-\(|k|\) scalings of the spectral densities of all statistically homogeneous sphere packings: see Eq. (7.12). Here, we note that the theoretical results (7.26) and (7.27) can be straightforwardly generalized to the cases of multiple particles are added in each cell. This is achieved by dividing cells such that each subdivided cell should circumscribe a single particle with an identical local-cell packing fraction.

Now, we consider a special case where all particles are similar to the associated cells in the sense that the particles have identical shapes and orientations with their cells, but have different sizes \((P_j = \phi^{1/d}C_j\) for \(j = 1, \ldots, N\)). Then, the \(n\)th moments of particles and cells can be related as \(M_{\alpha_1 \cdots \alpha_n}(P_j) = \phi^{n/d}M_{\alpha_1 \cdots \alpha_n}(C_j)\). Substituting this expression into Eq. (7.24), we obtain

\[
\tilde{J}(k) = \tilde{J}(1)(k) + \phi(1 - \phi^{2/d}) \frac{k_\alpha k_\beta}{2} \sum_{j=1}^{N} M_{\alpha\beta}(C_j) \left| C_j \right| e^{-i k \cdot x_j} + \mathcal{O}(k^3). \tag{7.28}
\]

Again, whenever \(\left| \tilde{J}(1)(k) \right| \sim |k|\), the spectral density of the constructed packings shows a scaling \(\tilde{\chi}_V(k) \sim \phi^2|k|^2\). However, in the special case of \(\Delta X_j = \mathbf{0}\) for all \(j\) (i.e., \(\tilde{J}(1) = 0\)), the resulting scaling is

\[
\tilde{\chi}_V(k) \sim \phi^2(1 - \phi^{2/d})^2|k|^4. \tag{7.29}
\]

Both hyperuniform cases belong to the class I.

Here, we should note that all theoretically predicted scalings of \(\tilde{\chi}_V(k)\), given in Eqs. (7.26), (7.27), and (7.29), are analytic at the origin, i.e., the power exponents are even positive integers. This implies that autocovariance functions \(\chi_v(|r|)\) of the constructed packings must decay to zero exponentially fast (or faster) as \(|r| \to \infty\) [300].
7.5 Hyperuniform Packings from Voronoi Tessellations

In this section, we formulate an efficient numerical algorithm that is based on our tessellation-based methodology to generate very large disordered hyperuniform packings in $\mathbb{R}^2$ and $\mathbb{R}^3$. This can be accomplished from various types of tessellations [130, 94, 28, 80, 303, 84], but we focus on Voronoi tessellations of disordered nonhyperuniform point patterns in this section. For a given point pattern, the Voronoi cell of a point is defined as the region of space closer to this point than any other point, and the Voronoi tessellation is the collection of all Voronoi cells [296]. The computational time for the Voronoi tessellation of a point pattern of $N$ particles in $\mathbb{R}^d$ is at most of the order of $O(N \log N + N^{[(d-1)/2]})$ [12]. Due to such small computation cost, the implementation of our methodology in Voronoi tessellations enables a remarkably efficient mapping that converts very large nonhyperuniform point patterns or packings into very large disordered hyperuniform packings.

We choose the progenitor point patterns for the Voronoi tessellations because they easily meet the bounded-cell condition, and the resulting packings are easy to fabricate. From a practical viewpoint, it is useful to employ similar particle shapes and sizes to construct the packings. All of these conditions can be readily fulfilled by considering progenitor point patterns derived from dense hard-sphere packings. In what follows, we elaborate on the bounded-cell condition.

7.5.1 The bounded-cell condition

The bounded-cell condition in the initial tessellation is a central requirement to ensure hyperuniformity via the tessellation-based procedure. For Voronoi tessellations, the largest cell size $\ell_{\text{max}}$ is on the order of the largest nearest neighbor distance, which in turn is of the order of the largest radius $r_{\text{max}}$ of holes (i.e., spherical regions that
Figure 7.3: Numerical simulations of probability distributions of the largest hole volume ($2r_{\text{max}}$) in each sample of 1D Poisson point patterns: (a) on a semilog scale and (b) on a log-log scale. The $x$-axis in (b) represents the relative size $\delta = v_1(r_{\text{max}})/V$ of the largest holes to the box volume $V(=L)$. For each particle number $N$, the distribution is obtained from $10^4$ independent samples. (c) Log-log plot of hole probability $E_V(r)$ of finite-size Poisson point patterns as a function of system size $N$. Values are computed from Eq. (7.30).

are empty of particle centers); $\ell_{\text{max}} \sim r_{\text{max}}$. Therefore, the bounded-cell condition (7.18) can be satisfied if the relative size $r_{\text{max}}/L$ of the largest hole to the sample size [or equivalently, their volume ratio $\delta \equiv v_1(r_{\text{max}})/L^d$] is much smaller than a certain small value.

The information about largest hole size is contained in the void-exclusion or hole
probability function $E_V(r)$, which gives the probability for finding a hole of radius $r$ when it is randomly placed in a point-particle system in the thermodynamic limit [296]. Clearly, if the hole probability has compact support, i.e., $E_V(r) = 0$ at any $r > D$ for a certain length $D$, then Voronoi tessellations of the associated point patterns always meet the bounded-cell condition for relatively small sample sizes (say $10^{d+1}$ in space dimension $d$). Examples of such systems include all crystals, disordered stealthy hyperuniform point patterns [356, 97], and the saturated random sequential addition (RSA) packings [296, 358]. Specifically, RSA is a time-dependent process that irreversibly, randomly, and sequentially adds nonoverlapping spheres into space. In the infinite-time limit, the resulting packing does not have any available space to add further particles, called saturated.

On the other hand, many other disordered point patterns, including Poisson point patterns, equilibrium hard-sphere liquids, and unsaturated RSA packings, can possess arbitrarily large holes in the thermodynamic limit. Finite-size samples of these systems tend to have larger holes as the sample size grows [see Fig. 7.30(a) for 1D Poisson point patterns], but the hole size relative to the sample size decreases as the sample size grows [see Fig. 7.30(b)], implying that samples, in fact, tend to meet the bounded-cell condition (7.18).

We now rigorously show that for sufficiently large statistically homogeneous point patterns, Voronoi tessellations of almost every realization should obey the bounded-cell condition, as long as their hole probabilities are smaller than or equal to that of the Poissonian counterparts for large hole radii. For this purpose, we will show that Voronoi tessellations of almost every Poisson point pattern obey the bounded-cell condition for sufficiently large sample sizes. We begin by considering, for finite-size samples, the hole probability $E_V(r)$ which can be interpreted as the probability that a finite sample possesses at least a single hole of radius greater than $r$ (i.e., $\ell_{\max} \sim r_{\max} \geq r$). Thus, $E_V(L[\delta/\nu_1(1)]^{1/d})$ is, in turn, the probability that the
Voronoi tessellation of a single sample does not meet the bounded-cell condition (7.18). For Poisson point patterns of $N$ particles and volume $V = L^d$ in $\mathbb{R}^d$, its hole probability can be straightforwardly obtained as follows $^3$:

$$E_V(r) = \frac{[V - v_1(r)]^N}{V^N} = (1 - \delta)^N, \quad (7.30)$$

where $\delta \equiv v_1(r)/V$. This quantity converges to a well-known expression $\exp(-\rho v_1(r))$ in the thermodynamic limit [296], where $\rho$ is the number density. Equation (7.30) decays exponentially fast for a given $\delta < 1$ as particle number $N$ increases; see Fig. 7.3(c). This implies that for sufficiently large sample sizes, almost every realization of a Poisson point pattern should obey the bounded-cell condition. Thus, the constructed packings from Voronoi tessellations of Poisson point patterns should be hyperuniform, which is consistent with a recent study of random fields [146].

We will employ correlated and statistically homogeneous point patterns as the progenitor configurations, which are even more likely to obey the bounded-cell condition than Poisson point patterns at the same number density. In the ensuing discussion, we show that such nonhyperuniform point patterns can be converted into disordered hyperuniform packings.

### 7.5.2 Spherical particles

Here, we numerically implement our procedure by solely rescaling particle volumes without changing particle centers and shapes. It results in disordered sphere packings whose Voronoi tessellations are identical to those of their progenitor packings, and we show that they are exactly hyperuniform of class I.

Due to the fixed particle centers, we define two alternative maximal packing frac-

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$^3$In a Poisson point pattern, the probability that a single particle is placed in the space such that a hole of radius $r$ is empty of a particle is $(V - v_1(r))/V$ because all positions are equally probable. Since particle positions are mutually independent, the hole probability $E_V(r)$ that $N$ particles are placed in the space such that this hole is empty of particles is simply $[(V - v_1(r))/V]^N$. 

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Figure 7.4: Schematic for three types of the maximal packing fractions $\phi_{\text{max}}^{(2)} \leq \phi_{\text{max}}^{(1)} \leq \phi_{\text{max}}$. For a Voronoi cell of an initial particle (white circle), the black circle and the green one represent the largest inscribed circles when the circle center is fixed or free to move, respectively. These three circles (white, black, and green ones) illustrate the largest particles $|P_j|$ of this cell for three distinct maximal packing fractions $\phi_{\text{max}}^{(2)}$, $\phi_{\text{max}}^{(1)}$, and $\phi_{\text{max}}$, respectively. We note that the initial particle size is exaggerated for a clear visualization.

For $\phi_{\text{max}}^{(1)}$, the volume $|P_j|_{\text{max}}$ of the largest particle of a Voronoi cell in definition (7.19) is that of the smallest spherical particle inscribed in the cell. For $\phi_{\text{max}}^{(2)}$, the largest particle is the particle in the progenitor packing. By definition, we have the following inequalities: $\phi_{\text{max}}^{(1)} \geq \phi_{\text{max}} \geq \phi_{\text{max}}^{(2)}$, where $\phi_{\text{max}}^{(1)} = \phi_{\text{max}}^{(2)}$ occurs only when every particle in the progenitor has a neighbor in contact, and $\phi_{\text{max}} = \phi_{\text{max}}^{(1)}$ occurs only when every particle in the progenitor is inscribed in its Voronoi cell. If a constructed packing has the packing fraction $\phi \leq \phi_{\text{max}}^{(2)}$, none of its particles can be larger than those in the progenitor packing. For 2D and 3D saturated RSA packings, values of $\phi_{\text{max}}^{(1)}$ are around 0.35 or 0.25, respectively. These values tend to decrease as the packing fraction of the progenitor packings is smaller, and they become arbitrarly small for Poisson point patterns (i.e., RSA with the zero packing fraction). Additional values of $\phi_{\text{max}}^{(1)}$ and $\phi_{\text{max}}^{(2)}$ are summarized in Appendix 7.10.2.

We employ three different types of nonhyperuniform progenitor point patterns in both $\mathbb{R}^2$ and $\mathbb{R}^3$: RSA packing [296, 358], equilibrium hard-sphere liquids, and
lattice-packings with point vacancies. First, we employ RSA packings for various values of initial packing fractions $\phi_{\text{init}}$. These RSA packings are efficiently generated by the voxel-list algorithm [358]. Second, as progenitor packings, we use equilibrium hard-sphere liquids with a range of initial packing fraction $\phi_{\text{init}}$; see Appendix 7.10.1 for employed parameters. For $d = 2$, its $\tilde{\chi}_V(0)$ can be well approximated from Eq. (7.15) and the compressibility$^4$ as follows:

$$\tilde{\chi}_V(0) \approx \frac{\phi^2(1 - \phi_{\text{init}})^3}{\rho(1 + \phi_{\text{init}} + 0.38406\phi_{\text{init}}^2 - 0.12802\phi_{\text{init}}^3)},$$

(7.31)

where $\phi$ is the packing fraction of the decorated spheres.

The last type of progenitor packings is vacancy-riddled square and simple cubic lattices in two and three dimensions, respectively. These progenitor systems are characterized by the number $N_s$ of initial lattice sites and the fraction $c$ of point

$^4$The isothermal compressibility $\kappa_T \equiv -V^{-1}\left(\frac{\partial V}{\partial p}\right)_T$ of 2D equilibrium hard-sphere liquids with the packing fraction $\phi$ is computed from the equation of states $p/(\rho k_B T) = 1 + 2\phi g_2(D)$, where $g_2(D)$ is the contact value of the radial pair-correlation [296]. The contact value is $g_2(D) = G(D) = (1 - 0.436\phi)/(1 - \phi)^2$ for $0 \leq \phi \leq \phi_f$; see Ref. [291].
Figure 7.6: Log-log plot of the scaled local volume-fraction variance \( v_1(R) \sigma_v^2(R) \) of the progenitor and constructed packings. The progenitor packings are 2D saturated RSA packings, and \( \sigma_v^2(R) \) are estimated by the Monte Carlo method; see Chapter 6. Three vertical grids represent a quarter of side length of the simulation boxes (i.e., \( R = L/4 \)) for \( N = 10^4, 10^5, \) and \( 10^6 \), respectively.

vacancies to \( N_s \). While these imperfect lattices still have Bragg peaks, they are not hyperuniform [65, 155], and it is easy to generate extremely large samples (\( N \sim 10^8 \)). Furthermore, for high values of \( c \), their cell-volume distributions are similar to those for Poisson point patterns. Therefore, this investigation will immediately demonstrate our discussion in Sec. 7.5.1.

We compute the Voronoi tessellations of the three aforementioned types of progenitor packings via VORO++ library [254] (see Appendix 7.9 for the implementation), and then perform our methodology without changing particle centers. To get a visual sense of the resulting hyperuniform packings, we show representative images of hyperuniform packings for \( d = 2 \) and \( d = 3 \) derived from RSA initial conditions in Fig. 7.5.

We computed the local volume-fraction variances \( \sigma_v^2(R) \) of certain progenitor and constructed packings by Monte Carlo sampling of windows [296, 238] for many values of \( R \) up to \( L \) (i.e., the side length of the simulation box). For illustrative purposes, Fig. 7.6 plots such variances for progenitor packings that are 2D saturated RSA packings of various system sizes (\( N = 10^4, 10^5, \) and \( 10^6 \)). We note that these results for \( \sigma_v^2(R) \)
are reliable only up to a window radius \( R < L/4 \) \([64, 47]\) due to the finite-size effects. The constructed packings exhibit a common scaling \( \sigma^2(R) \sim R^{-(d+1)} \), as predicted from the heuristic rationale associated with Fig. 7.2(c) and (d). This rationale also predicts that such a scaling starts from window sizes \( R \gtrsim 5\ell_{\text{max}} \), where \( \ell_{\text{max}} \) is the maximal cell length defined in step 1, from which the region where volume-fraction fluctuations arise [the yellow-shaded regions in Fig. 7.2(c)-(d)] can be effectively regarded as window boundary. Since the Voronoi tessellations of 2D saturated RSA that have a small \( \ell_{\text{max}} \) (\( \lesssim 1.7 \rho^{-1/2} \)), the class I hyperuniform scaling is well-established in length scales over several orders of magnitude (even beyond the reliable regime) for even relatively small samples \( (N = 10^4) \) and the largest ones \( (N = 10^6) \); see Fig. 7.6.

![Figure 7.7: Simulation results for sphere packings from Voronoi tessellations of RSA packings in (a)-(c) \( \mathbb{R}^2 \) and (d)-(f) \( \mathbb{R}^3 \). (a) and (d) Probability density \( p(|C_j|) \) of Voronoi cell volumes versus the scaled cell volume \( |C_j|/|C| \) plotted on a semi-log scale (larger panel) and a linear scale (inset). Here, \( |C| \) represents the expected cell volume. (b) and (e) The spectral densities versus wavenumber \( k \) for small wavenumbers plotted on a log-log scale. Inset in (b) is on a linear scale. Here, we note that all packings are rescaled to the common packing fraction \( \phi = 0.01 \). (c) and (f) Log-log plots of the spectral densities for progenitor packings (saturated RSA) and associated constructed packings according to system size \( N \).](image-url)
Figure 7.8: Simulation results for disk packings converted from 2D equilibrium hard-disk liquids of $N = 10^5$. (a) Probability density $p(|C_j|)$ of Voronoi cell volumes versus the scaled cell volumes $|C_j|/|C|$ plotted on a semi-log scale. (b) The spectral densities versus wavenumber $k$ for small wavenumbers plotted on a log-log scale. Theoretical values of $\tilde{\chi}_V(0)$ are obtained from Eq. (7.31). (c) The spectral densities versus wavenumber $k$ for intermediate and large wavenumbers plotted on a semi-log (upper) and a linear (lower) scales. Here, the sample size is $N = 10^3$, and $\bar{a}$ is the mean particle radius of the constructed packings.

Figures 7.7, 7.8, and 7.9 summarize the simulation results for packings that were converted from RSA, equilibrium hard-sphere liquids, and vacancy-riddled lattices, respectively. By construction, these constructed packings have particle-volume distributions that are identical to those of Voronoi cell volumes in their progenitors; see Figs. 7.7(a), 7.7(d), 7.8(a), and 7.9(a). Furthermore, since particle centers are fixed in the procedure, the progenitor packings and their associated constructed packings have identical Voronoi tessellations and obviously have identical local statistics associated with Voronoi cells. This implies that local statistics alone generally may or may not determine hyperuniformity, which we elaborate in Sec. 7.8.

Isotropic spectral densities of progenitor and constructed packings are computed by using Eqs. (7.6) and (7.9). To compare them, we rescale all packings to a common packing fraction $\phi = 0.01$; see Figs. 7.7(b), 7.7(e), 7.8(b), and 7.9(b). For small wavenumbers (i.e., $0 < k\bar{a} \ll 1$, where $\bar{a} \sim 0.1r_{-1/d}$ is the mean of particle radii), as shown in these figures, the spectral densities of the progenitors are significantly different from those of the constructed packings. While progenitor packings are clearly nonhyperuniform (i.e., $H \sim 0.1 - 0.01$ for RSA packings and equilibrium hard-sphere
Figure 7.9: Simulation results for constructed disk packings from 2D lattice packing for various values of vacancy concentration $c$. (a) Semi-log plots of probability density functions of Voronoi cell volumes ($N_s = 10^6$). (b) Log-log plots of the spectral densities for progenitor packings of various vacancy concentrations and the constructed packings ($N_s = 10^6$). (c) Log-log plots of the spectral densities of the constructed packings of various sample sizes ($c = 0.4$).

liquids), the constructed packings are class I hyperuniform with a common power-law scaling $\bar{\chi}_V(|k|) \sim |k|^4$. Similar to the case of $\sigma_V^2(R)$ shown in Fig. 7.6, this power-law scaling is particularly well-established over several orders of magnitude, even for relatively small samples ($N = 10^3$ as shown in Figs. 7.7(c) and 7.7(f)). This $k^4$-scaling results from $|\tilde{J}^{(1)}(k)| \sim |k|^2$, which comes from special correlations in $\Delta X_j$; see Sec. 7.4. Values of $\bar{\chi}_V(0)$ and $H$-index are summarized in Tables 7.6, 7.7, and 7.8.

On the other hand, for intermediate wavenumbers (i.e., $1 < k\bar{\alpha} < 3$), the spectral densities of the constructed packings largely resemble those of their progenitors; see, for example, Fig. 7.8(c). This result reflects that local statistics of both packings...
are identical at corresponding length scales. At large wavenumbers (i.e., \( k\pi > 3 \)), however, the spectral densities of progenitor and constructed packings again become different from each other because the spectral density at this regime mainly depends on the particle-size distributions; see Fig. 7.8(c).

### 7.5.3 Nonspherical particles

![Figure 7.10: Simulation results for \( \tilde{\chi}_V(k) \) of the constructed packings depending on particle shapes. Here, the progenitor point patterns are the centers of saturated RSA packings with \( N = 10^6 \) in (a) two and (b) three dimensions.](image)

As shown in the derivation in Sec. 7.4, the tessellation-based procedure can generate class I hyperuniform packings even for nonspherical particle shapes, including regular and random polyhedrons. However, since it is generally nontrivial to compute the form factors of those particle shapes, we consider a simple geometric shape, i.e., a \( d \)-dimensional hypercubic particles; see Eq. (7.10).

For simplicity, we take advantage of the sphere packings constructed in Sec. 7.5.2 by replacing the spherical particles with the inscribed cubic particles. In this case, the corresponding maximal packing fraction for cubes will be related to that of spheres as follows: \( \phi_{\text{cube}}^{\text{max}} = \frac{\Gamma(1+d/2)2^{d-1}}{(d\pi)^{d}} \phi_{\text{sphere}}^{\text{max}} \). We consider two cases: (identically) oriented particles and randomly oriented particles. Figure 7.10 summarizes the simulation results for 2D and 3D packings converted from saturated RSA packings of \( N \approx 10^6 \).

We note that the results for squares or cubes are largely indistinguishable from those
for spheres because both particle shapes have comparable length scales and the Fourier transforms of both a cube (7.10) and a sphere (7.9) are isotropic near the origin and have similar profiles. However, these spectral densities will exhibit different behaviors at intermediate and large wavenumbers, depending on particle shapes and orientations.

### 7.6 Multiscale Coated-Spheres Model

The tessellation-based methodology can be applied to any type of tessellations of space beside the Voronoi tessellations, as long as they obey the bounded-cell condition (7.18). One interesting example is a tessellation of space that consists of nonoverlapping spherical cells. Since a monodisperse sphere packing in \( d > 1 \) cannot occupy all space, spherical cells in such tessellations should have a polydispersity in size down to the infinitesimally small; see Fig. 7.1(b). For these (multiscale) sphere tessellations, the bounded-cell condition can be guaranteed by making the ratio of the largest cell-volume to the sample volume, \( \delta = v_{max}/|V_F| \), sufficiently small. Implementing our methodology from these tessellations should result in hyperuniform packings regardless of particle shapes, positions, and numbers per cell.

A simple but important case of such disordered hyperuniform packings are ones in which each cell includes only a single particle to which the cell is concentric; see Fig. 7.1d. Since particle shapes are similar to those for cells, the local-cell packing fraction \( \phi \) of those packings can span up to unity, as mentioned in Sec. 7.3. In addition, we can use the expression of the second moment of a sphere of radius \( R \), \( \mathcal{M}_{\alpha\beta} = \frac{1}{(2\pi d)^2} R^2 \delta_{\alpha\beta}, \Delta X_j = 0 \), and Eq. (7.28) to obtain the following power-law
scaling for the spectral density:

\[
\tilde{\chi}_V(k) = \left[ \frac{\phi(1 - \phi^{2/d})}{2(2 + d)} \right]^2 \frac{1}{|V_F|} \left| \sum_{j=1}^{\infty} v_1(R_j) (kR_j)^2 e^{-ik \cdot x_j} \right|^2 + \mathcal{O}(k^5)
\]

\[
\sim \phi^2 (1 - \phi^{2/d})^2 k^4,
\] (7.32)

where radius of cell \(j\) is denoted by \(R_j\).

It is crucial to observe that cells and the associated particles form composite spheres that fill all space. Each composite sphere is comprised of a spherical core (particle) of one phase that is surrounded by a concentric spherical shell of the other phase such that the fraction of space occupied by the core phase is equal to its global phase volume fraction, which is guaranteed by our procedure. Thus, this structure is a packing (dispersion) in which spherical particles (blue regions) are “well-separated” from one another in a fully connected (continuous) void (matrix) phase (red region). This is traditionally understood as a key characteristic of optimal two-phase structures [303, 304].

Surprisingly, these hyperuniform multiscale structures are identical to the Hashin-Shtrikman multiscale coated-spheres models [113, 296, 310], which are perhaps the most famous results from the theory of heterogeneous materials. These special particle composites are optimal for the effective electric (thermal) conductivity and bulk elastic modulus for given phase fractions and phase properties. This observation will shed light on the origin of nearly optimal transport properties of disordered stealthy hyperuniform packings [355, 46] and cellular networks derived from hyperuniform systems [303].

The coated-spheres structures are infinitely degenerate with varying degrees of order/disorder. The most ordered structures would be ones, derived from certain deterministic sphere tessellations, such as Apollonian gaskets and initial lattice packings of identical spheres in which multiscale particles are added sequentially.
Figure 7.11: A representative of disordered coated-disks model with the local-cell packing fraction $\phi = 0.25$ derived from a sphere tessellation for a power-law scaling with $p = 1.5$ and $m = 400$. Its magnification is presented in Fig. 7.18.

7.6.1 Simulation model

In what follows, we describe our model to simulate the coated-spheres model and ascertain their hyperuniformity. We first construct very dense disordered sphere packings via a multistage version of the RSA procedure, in which the volume of the inserted spheres reduced in every stage. These dense “precursor” packings are later used to simulate ideal sphere tessellations. The coated-spheres model is then simulated by scaling spheres in the precursor packings at a certain ratio.

Specifically, these precursor packings are constructed from an empty simulation box $\mathcal{V}_F$ in $\mathbb{R}^d$ under the periodic boundary conditions. The construction procedure has two control parameters: the upper bound $v_{\text{max}}$ on cell volumes and a positive decreasing function $g(i)$ of positive integers $i$, which satisfies $g(1) = 1$ and $\sum_{j=1}^{\infty} g(j) <$
The infinite-system-size limit can be achieved as the upper bound $v_{\text{max}}$ tends to be infinitesimally small with the simulation box fixed. Subsequently, one determines the prescribed number $N$ of spheres that will be inserted in every stage and the maximum cell volume $v^{(1)}$ to fill all space:

$$ N \equiv \lceil |\mathcal{V}_F| / (v_{\text{max}} G) \rceil $$

$$ v^{(1)} \equiv |\mathcal{V}_F| / (NG) $$

where $G \equiv \sum_{j=1}^{\infty} g(j)$ and $\lceil x \rceil$ is the ceiling function. Every sphere in the $m$th stage has volume $v^{(m)} = v^{(1)} g(m)$ and the associated diameter $D_m$. Using these parameters, one can construct the precursor packings by the following steps:

1. In the first stage ($m = 1$), irreversibly, randomly, and sequentially add (i.e., via RSA procedure) nonoverlapping spheres of a diameter $D_1$. The insertion stops when $N$ spheres are added unless the packing becomes saturated.

2. In the $m$th stage ($m > 1$), nonoverlapping spheres of a diameter $D_m (< D_{m-1})$ are added via the RSA procedure. Make sure that newly inserted spheres do not overlap with the spheres in the previous stages $1, 2, \cdots, m - 1$. The insertion stops when the packing reaches to a prescribed covering fraction $N/|\mathcal{V}_F| \sum_{i=1}^{m} v^{(i)}$, unless the packing becomes saturated.

3. The procedure stops when it reaches a prescribed number of stages; see Fig. 7.1b.

Note that the aforementioned voxel-list algorithm [358] is implemented in steps 1 and 2. Using the $m$th stage precursor packings, we simulate the coated-spheres model of the inclusion volume fraction $\phi$ by reducing these spheres at a volume ratio $\phi$. Figure 7.11 shows a representative but small disordered multiscale coated-disks construction.
We note that due to saturation at each stage, the number $N_m$ of spheres inserted in the $m$th stage can be different from the prescribed number $N$. Let $N_m \equiv \sum_{i=1}^{m} N_i$ and $\eta_m = \sum_{i=1}^{m} N_i v^{(i)}/|V_F|$ denote the total number of spheres at the end of the $m$th stage and the fraction of space covered by these spheres, called the covering fraction, respectively. By construction, $\eta_m$ cannot exceed the prescribed covering fraction in the $m$th stage:

$$\eta_m \leq \frac{N}{|V_F|} \sum_{i=1}^{m} v^{(i)}.$$  \hfill (7.35)

Thus, in a finite $m$th stage, the precursor packings will not cover all space but have gaps that can be only covered by smaller spheres in the next stages. As the number of stages tends to be infinite, those gaps are eventually covered by spheres of size down to the infinitesimally small, i.e., $\eta_m \to 1$ as $m \to \infty$.

In this chapter, we consider two different types of volume scalings: a power-law scaling and an exponential scaling. For the power-law scaling, the cell volume in the $m$ stage is determined by

$$v^{(m)}/v^{(1)} = 1/m^p,$$  \hfill (7.36)

for a given scaling exponent $p > 1$. From this relation, it is straightforward to derive the relation between maximum cell volume $v^{(1)}$ and the prescribed insertion number $N$, and the prescribed covering fraction:

$$|V_F| = N v^{(1)} \zeta(p),$$  \hfill (7.37)

$$1 - \eta_m = \frac{1}{\zeta(p)} \sum_{j=m+1}^{\infty} 1/j^p,$$  \hfill (7.38)

where $\zeta(p)$ denotes the Riemann zeta function.

For the exponential scaling, cell volume in the $m$th stage is determined by

$$v^{(m)}/v^{(1)} = 1/q^{m-1},$$  \hfill (7.39)
for a given scaling base $q > 1$. The analogues of Eqs. (7.37) and (7.38) are

$$|V_F| = N v^{(1)} q/(q - 1),$$  \hspace{1cm} (7.40)\\
$$1 - \eta_m = 1/q^m.$$  \hspace{1cm} (7.41)

### 7.6.2 Theoretical analyses

While in the limit of $m \to \infty$ our coated-spheres model is predicted to be strongly hyperuniform (see Eq. (7.32)), our model in a finite $m$th stage will be nearly hyperuniform, rather than perfectly hyperuniform, due to the uncovered gaps ($\eta_m < 1$). To estimate the degree of hyperuniformity of our model in the $m$th stage, we consider the associated spectral density $\tilde{\chi}^{(m)}(k)$, given by

$$\tilde{\chi}^{(m)}(k) = \frac{1}{|V_F|} \left\langle \sum_{j=1}^{N_m} \tilde{m}(k; \phi^{1/d} R_j) e^{-ik \cdot x_j} - \phi \int_{V_F} dy e^{-ik \cdot y} \right\rangle^2,$$  \hspace{1cm} (7.42)

where $\phi$ denotes the local-cell packing fraction. Without any prior knowledge of cell-volume distribution, its rigorous bound can be obtained by using some simple inequalities (see Appendix 7.13.1):

$$\tilde{\chi}^{(m)}(k) \leq 2 F(k; \phi) + 2\phi^2 |V_F|(1 - \eta_m)^2,$$  \hspace{1cm} (7.43)

where

$$F(k; \phi) \equiv \frac{1}{|V_F|} \left[ \frac{\phi(1 - \phi^{2/d})}{2(2 + d)} \right]^2 \left\langle \sum_{j=1}^{N_m} v_1(R_j)(kR_j)^2 e^{-ik \cdot x_j} + \mathcal{O}(k^4) \right\rangle^2 \propto \phi^2(1 - \phi^{2/d})^2 |\mathbf{k}|^4, \quad |\mathbf{k}| \to 0.$$  \hspace{1cm} (7.44)

Here, the constant term $\phi^2 |V_F|(1 - \eta_m)^2$ is the largest volume-fraction fluctuations that can arise from uncovered gaps in the $m$th stage coated-spheres model. Importantly,
the constant term depends on $\phi(1 - \eta_m)$, i.e., the deviation between the local-cell packing fraction $\phi$ and the global packing fraction $\phi \eta_m$, which will vanish as the uncovered gaps become completely filled in the limit of $m \to \infty$. Thus, in this limit, the upper bound implies that our model becomes perfectly hyperuniform of class I, consistent with Eq. (7.32).

Although the upper bound (7.43) is rigorous, it is a gross overestimation compared to corresponding simulation results. Thus, we obtain a better estimate of Eq. (7.42) by assuming that the uncovered gaps are spatially uncorrelated, which effectively removes the ensemble average of their cross terms, yielding

$$
\tilde{\chi}_V^{(m)}(k) \approx F(k; \phi) + \frac{\phi^2}{|V_F|} \left\langle \sum_{j=N_{m}+1}^{\infty} v_1(R_j)^2 \right\rangle,
$$

(7.45)

where the summation term becomes dominant as $|k| \to 0$ because $F(k; \phi)$ shows a power-law scaling; see Appendix 7.13.2 for details.

Further estimation of the second term in Eq. (7.45) requires the cell-size distribution. Consider two distinct cell-size scalings discussed in Sec. 7.6.1: a power-law form (7.36) and an exponential functional form (7.39). For the power-law scaling, using Eqs. (7.37) and (7.38), the summation in Eq. (7.45) can be written as

$$
\left\langle \sum_{j=N_{m}+1}^{\infty} v_1(R_j)^2 \right\rangle = N \left(v^{(1)}\right)^2 \sum_{j=m+1}^{\infty} \frac{1}{j^{2p}} = v^{(1)}|V_F| \zeta(p) (1 - \eta_m)^2 f(m),
$$

(7.46)

where $f(m) \equiv \left(\sum_{j=m+1}^{\infty} j^{-2p}\right) / \left(\sum_{j=m+1}^{\infty} j^{-p}\right)^2$. Substituting Eq. (7.46) into Eq. (7.45) yields an approximation of the residual spectral density in the small-wavenumber limit as follows:

$$
\tilde{\chi}_V^{(m)}(k \to 0) \approx v^{(1)} f(m) \zeta(p) (\phi(1 - \eta_m))^2.
$$

(7.47)

For the exponential scaling, Eqs. (7.40) and (7.41) are used to simplify Eq. (7.45)
as follows:

\[
\left\langle \sum_{j=N_{m}+1}^{\infty} v_1(R_j) \right\rangle = N \left[ v_1^{(1)} \frac{q}{q^m(q-1)} \right]^2 \frac{(q-1)^2}{q^2 - 1} = |V_F| v_1^{(1)} \frac{q}{q + 1} (1 - \eta_m)^2, \tag{7.48}
\]

which results in the following leading-order term of the spectral density in the \(m\)th stage:

\[
\hat{\chi}_V^{(m)}(k \to 0) \approx v_1^{(1)} \frac{q}{q + 1} \left[ \phi(1 - \eta_m) \right]^2. \tag{7.49}
\]

### 7.6.3 Simulation results

![Simulation results for multiscale-disk tessellations.](image)

Figure 7.12: Simulation results for multiscale-disk tessellations. For the power-law scaling (7.36), (a) the semi-log plot of fraction of uncovered space \((1 - \eta_m)\) versus the number of stages \(m\), and (b) the log-log plot of total cell number \(N_m\) versus \((1 - \eta_m)\). For the exponential scaling (7.39), (c) the semi-log plot of \((1 - \eta_m)\) in the \(m\)th stage, and (d) the log-log plot of total cell number \(N_m\) as functions of \((1 - \eta_m)\). Here, we note that dashed lines represent prescribed covering fractions, given by Eqs. (7.38) and (7.41), and \(\delta \equiv \nu_{\text{max}}/|V_F|\).
For the purpose of illustration and simplicity, we specialize to two dimensions to generate multiscale-disk tessellations for the aforementioned cell-size scalings: a power-law (7.36) and an exponential (7.39) functional forms. Using the power-law functional forms, the simulations proceed up to the 400th stage, yielding a scaling exponent $p$ ranging from 1.5 to 1.8, and the ratios of the maximal cell volume to the sample volume $\delta = 10^{-3}$ and $10^{-4}$. For the exponential functional forms, the constructions proceed to achieve around covering fraction $\eta_m \approx 0.99$ for values of a scaling base $q=1.05$, 1.10, and 1.20, and the ratio $\delta = 10^{-3}$.

For both types of scaling functions, as the scaling parameters increases, the smaller
is the number of stages needed to achieve a prescribed covering fraction. Instead, for even larger scaling parameters, the precursor packings are more likely to be saturated [268], which often results in high computational costs. This is because, as the packing approaches to a saturation state, the voxel-list algorithm that we employ subdivides current voxels with an increasingly finer resolution, which requires increasingly larger computer memory and computational times. For this reason, we choose the scaling parameters smaller than 2.

To obtain multiscale-disk tessellations that nearly fill space ($\eta_m \approx 0.95$), these procedures need to continue up to around a few hundred stages [Fig. 7.12(a) and (c)]. For $m \lesssim 10$, cell volumes in the exponential scalings do not change much, compared to those in the power-law scalings. Thus, the tessellations in the exponential scalings can cover a larger fraction of the simulation box with a smaller number of stages than those for the power-law scaling. Instead, for the exponential scaling, the precursor packings more easily achieve saturation from relatively low covering fractions ($\eta_m \approx 0.65$). Then, the simulated covering fraction never keeps up with the prescribed covering fraction (7.41); see Fig. 7.12(c). However, the number of total cells $N_m$ required to reach to a certain covering fraction will be similar for both cases; see Fig. 7.12(b) and 7.12(d).

From these precursor polydisperse packings, we simulate the coated-disks model with the inclusion volume fraction $\phi = 0.5$. To ascertain their hyperuniformity, we compute the associated spectral densities $\tilde{\chi}^{(m)}(k)$. As shown in Figs. 7.13(a) and (c), the packings in the finite stages (around $\eta_m \approx 0.95$) are effectively hyperuniform. Importantly, this comes from the fact that the precursor packings have uncovered gaps, and thus the local-cell packing fraction $\phi$ and their global packing fraction $\phi \eta_m$ have a discrepancy. The consequent volume-fraction fluctuations are estimated by Eqs. (7.47) and (7.49). Figures 7.13(b) and (d) show that while the numerical results in $\tilde{\chi}_v^{(m)}(k = k_{\text{min}})$ and these theoretical estimations conform to each other for low
covering fractions ($\eta_m \lesssim 0.8$), they significantly deviate for relatively high covering fractions. This is because the uncovered gaps are no longer spatially uncorrelated in high covering fractions.

However, for high covering fractions, the residual spectral densities $\tilde{\chi}_V^{(m)}(k = k_{\text{min}})$ vanish with an error on the order of $(1 - \eta_m)^2$. Thus, our upper bound (7.43), which although is a gross overestimation, correctly predicts the scaling behavior of $\tilde{\chi}_V^{(m)}(k = k_{\text{min}})$. Both the theoretical and numerical results show that the coated-spheres model should be strongly hyperuniform in the limit of $m \to \infty$.

It is noteworthy that as the number $m$ of stages increases, the spectral densities in Fig. 7.13(a) and 7.13(c) tend to resemble one another for intermediate wavenumbers. In addition, the terms $\tilde{\chi}_V^{(m)}(k = k_{\text{min}})$, as shown in Figs. 7.13(b) and (d), collapse to a single curve that seemingly only depends on the fraction of uncovered space. From these observations, we surmise that the spectral densities of the multiscale coated-spheres structures ($\eta_m \to \infty$) are identical to one another, regardless of the cell-volume distributions of the precursor sphere packings. This universal behavior of spectral densities is apparently related to the fact that the coated-spheres structures have identical effective properties.

### 7.7 Fabrication of Our Designs

Disordered hyperuniform structures, constructed by numerical simulations have been produced via modern fabrication technologies [196, 193]. Here, we explicitly discuss how our designed hyperuniform packings (dispersions) can be fabricated via state-of-the-art technologies, such as 2D photolithographic [360] and 3D printing techniques [334, 269, 319].

Photolithography is a microfabrication technique that uses light to transfer a designed 2D pattern in the photomask on the photo-sensitive chemicals coated on
a flat substrate. Then, after a series of chemical treatments, the desired pattern is engraved on the material, or material is deposited on the pattern. These methods are widely used in industry and research because of their high efficiency and exact control over the shapes and sizes of the patterns. Instead, diffraction tends to round all sharp corners in the designed pattern, and the corner radii are associated with the minimum feature size. State-of-the-art photolithography techniques are capable of creating patterns on a 30-cm-diameter wafer with the minimum feature size down to 25 nm [360]. With equipment of 1.5 \( \mu m \) minimum feature size, one can readily fabricate our 2D hyperuniform disk packings (probably derived from Voronoi tessellations) that include more than one million particles.

![Figure 7.14: Fabrication of designed hyperuniform dispersions and matrices in three dimensions. (a) A portion of the designed dispersion of spherical particles (left) and the corresponding matrix (right) at \( \phi = 0.23 \), which are constructed from a 3D saturated RSA packing. (b) A portion of the designed dispersion of cubical particles (left) and the corresponding matrix (right). The hyperuniform matrices with spherical or cubical pores can be fabricated using 3D printing techniques.](image)

Three-dimensional hyperuniform packings can be fabricated using 3D additive
manufacturing techniques [334, 269, 319]. 3D printing refers to various processes that solidify materials layer by layer to create a 3D object (e.g., fused filament fabrication, stereolithography, and selective laser melting). A printed structure must be topologically connected such that it can be mechanically self-supporting after the procedure. Thus, the void (matrix) phase of our 3D hyperuniform packings (dispersions), shown in right panels in Fig. 7.14, can be readily printed [351]. Due to recent developments in 3D printing methods, some commercial desktop 3D printers can print a sample with dimensions $125 \times 125 \times 125 \text{ cm}^3$ in 50 hours with around 100 $\mu m$ XY-resolution and 20 $\mu m$ in Z-resolution. Setting the minimum pore size is 300 $\mu m$, such devices can readily fabricate our 3D hyperuniform sphere packings that include up to 50 million pores. If pore sizes are on the order of the resolutions, spherical pores will be suitable to reduce the effect of thermal deformations during the printing process.

### 7.8 Conclusions and Discussion

In summary, we have introduced the tessellation-based methodology to construct disordered hyperuniform packings (dispersions) in $d$-dimensional Euclidean space $\mathbb{R}^d$. Unlike many previous methods to generate disordered hyperuniform materials, this procedure is simple to implement as it only involves constraining the local-cell packing fraction, which is independent of the rest of a system. Furthermore, all computations to determine particle volumes can be exactly performed and easily parallelized. Importantly, this procedure also guarantees that the constructed packings are perfectly hyperuniform of class I in the infinite-system-size limit, as we analytically showed in Eqs. (7.26), (7.27), and (7.29). In this chapter, we have numerically implemented our methodology to two distinct types of disordered tessellations: Voronoi tessellations of nonhyperuniform progenitor packings and sphere tessellations.

In the case of Voronoi tessellations, we demonstrated that our methodology pro-
vides a remarkable mapping that converts virtually all samples of any statistically homoge-
neous point pattern, whether hyperuniform or nonhyperuniform, into perfectly hyperuniform packings. Furthermore, the fact that it is easy to create a Voronoi tessellation of a very large point pattern with the combination of our efficient procedure enables us to construct very large samples (of the order of $10^8$ particles) of perfectly hyperuniform disordered packings. Such large system sizes have not been possible for previous numerical methods.

In the case of sphere tessellations, we established that the optimal Hashin-Shtrikman multiscale dispersions are hyperuniform of class I. In addition to the fact that the spherical inclusions in such dispersions are “well-separated” from one another [303], hyperuniformity is another important structural attribute to attain optimal effective transport and elastic properties. In this regard, it is noteworthy that some disordered hyperuniform packings [355, 46] and cellular networks derived from hyperuniform systems [303] have been reported to possess (nearly) optimal effective transport and elastic properties. Thus, it will be interesting to investigate the physical properties of hyperuniform packings constructed by our procedure.

Furthermore, it is important to observe that our procedure allows many distinct types of tessellations as long as they meet the bounded-cell condition (7.18). Besides Voronoi tessellations and sphere tessellations studied in this chapter, examples include disordered isoradial graphs [28], Delaunay triangulations, “Delaunay-centroidal” tessellations [80, 303], dissected tessellations [84], and various generalizations of Voronoi tessellations, such as Laguerre tessellations [94, 130] and tessellations in Manhattan distances [166]. Since constructing progenitor packings is the most time-consuming step in our numerical implementations, one would increase sample sizes by employing other tessellations that can be efficiently generated.

We note that similar to our procedure, a 1D model [87] and the “equal-volume tessellation” [88] enable the generation of hyperuniform point mass patterns and hy-
peruniform point patterns from certain initial tessellations, respectively. Specifically, these two models place a point mass [87] and point particles [88] in each cell such that mass densities and number densities in the cells become identical, respectively. Thus, the hyperuniform systems in both models can be regarded as a zero-\( \phi \) limit of the hyperuniform packings in our procedure. Importantly, however, both previous models [87, 88] do not consider volume-fraction fluctuations, which is our central concern and accounted for by our procedure. In addition, the mass density of point masses in this 1D model does not change the structural characteristics of the resulting systems, which again is different from our methodology. We also note that the equal-volume tessellation is less versatile in its initial tessellations than our procedure because all cell volumes in an initial tessellation should be integer multiples of a common unit volume.

We have shown that our procedure enables a mapping that converts a nonhyperuniform packing into a hyperuniform one without changing the initial tessellation, i.e., the tessellation is an invariant under the transformation. In other words, packings that have identical tessellations can either be nonhyperuniform or hyperuniform by simply tuning local characteristics. It immediately follows that any tessellation statistic, including the distributions of nearest-neighbor distances and Voronoi-cell volumes, are identical for both the progenitor nonhyperuniform point patterns and their corresponding constructed hyperuniform packings. These results reinforce previous observations that local structural characteristics do not play an important role in determining the hyperuniformity of systems. For example, systems with substantial local clustering of particles can be hyperuniform [347], whereas disordered systems with appreciable short-range order often turn out to be non-hyperuniform [358]; see Ref. [300]. Moreover, our results also show that transitions from a nonhyperuniform state to a hyperuniform state can be achieved without correlated movements of particles/mass across all length scales. Such transitions stand in contrast to those in some
previous hyperuniform systems in thermal equilibrium [135, 321, 17, 352, 184, 46] as well as nonequilibrium [122, 287, 329, 169, 245, 307, 189], in which the transitions always involve collective rearrangements of the particles.

It should not go unnoticed that our methodology also allows ones to tune particle shapes, positions, and numbers within each cell with preserving the hyperuniformity of the constructed packings. For instance, one can engineer these two-phase systems at large length scales (i.e., \( k \ll 1 \)) by choosing nonspherical particles with various aspect ratios and placing them away from the centroids of the associated cells. However, at intermediate length scales the two-phase media are structurally similar to their progenitor patterns (see Sec. 7.5.2). One can tune local structures of the hyperuniform packings to achieve the “well-separated” condition, which is an important requirement to attain the optimal transport and mechanical properties (see Sec. 7.6). Moreover, the small-\( k \) scaling of the spectral density can either be \( \tilde{\chi}_V(k) \sim k^2 \) or \( \tilde{\chi}_V(k) \sim k^4 \) by engineering particle displacements \( \Delta \mathbf{X}_j \) with respect to the associated cell centroids (see Sec. 7.4). Due to this tunability, our methodology allows ones to design an enormous class of hyperuniform packings, including (nearly) optimal structures. Combining our computational designs with the aforementioned 2D and 3D fabrication techniques [157] is expected to accelerate the discovery of novel disordered hyperuniform two-phase materials.

There are several other open questions for future exploration. Could other types of initial tessellations lead to scaling behaviors of the spectral density besides quadratic or quartic shown here? To what extent can our procedure be generalized by relaxing the identical local-cell packing faction constraint (e.g., giving certain spatial correlations in local-cell packing fractions)? Can such generalized versions of our procedure construct “disordered” stealthy (i.e., \( \tilde{\chi}_V(k) = 0 \) for \( k < K \)) [17, 352, 353, 317] or class II hyperuniform (i.e., \( \tilde{\chi}_V(k) \sim k \), such as MRJ packings [138, 10])? If possible, this would certainly allow one to obtain more general scaling behaviors.
7.9 Appendix A: Voronoi Tessellations

We compute the Voronoi tessellations of a given point pattern in a \(d\)-dimensional cubic periodic simulation box (for \(d = 2, 3\)) of side length \(L\) via an open-source software packing (VORO++) [254]. To enhance the performance, we divide point patterns into several domains and compute Voronoi tessellation of each domain in parallel. Specifically, we select domains in the following steps (see Fig. 7.15):

1. Divide the simulation box into disjoint cubic regions of a fixed side length \(l\), called subdomains.

2. For each subdomain, make a marginal region that surrounds the subdomain. A subdomain and the associated marginal region comprises a domain. Each domain stores the position coordinates of all points that lie within it. We set a thickness as \(W = 6\rho^{-1/d}\), where \(\rho\) is the number density of the point pattern.

Then, we can compute the Voronoi cells in each subdomain in parallel. For point patterns with large holes, the thickness \(W\) of the marginal region should become not smaller than the largest hole diameter.

Since VORO++ is designed for three-dimensional geometries, its 2D implementation is performed in a quasi 3D simulation box whose height (in \(z\) component) is unity. To avoid any possible “memory leakage” in this 2D implementation, the number of particles within a domain should be smaller than \(10^5\).

7.10 Appendix B: Hyperuniform Packings Derived from Voronoi Tessellations

Here, we provide additional simulation data that are not presented in Sec. 7.5 in the main text.
Figure 7.15: Schematic of implementation of VORO++ library [254]. A point pattern in a periodic simulation box is sampled by domains of equal size (squares bounded by dashed lines). Each domain consists of a subdomain (a shaded region in the center) and a marginal region surrounding the subdomain. Subdomains are mutually disjoint and fully cover a simulation box. The thickness of the marginal region is denoted by $W$.

7.10.1 Progenitor packings for Voronoi tessellations

Here, we summarize the simulation parameters employed to generate progenitor packings that are used to obtain Voronoi tessellations.

To exactly generate saturated RSA packing, we employed the voxel-list algorithm, developed by Zhang and Torquato [358]. Using this algorithm with ten 2.6-GHz CPUs, it takes around 112 minutes to generate a 2D saturated RSA packing of $N \approx 10^7$. Under the same conditions, it takes around 14 minutes to generate a 3D saturated RSA packing of $N \approx 10^6$; see Ref. [157] for details. The number of sampled configurations are listed in Table. 7.1.

Equilibrium hard-sphere liquids in $\mathbb{R}^2$ and $\mathbb{R}^3$ are simulated via the Monte Carlo method in the canonical ensemble. Beginning with the initial triangular (FCC) lattice arrangements in $d = 2$ (3), we randomly remove some particles to have $N$ particles. Subsequently, we adjust the maximal distance of trial moves to satisfy that its average acceptance ratio for 10 MC cycles becomes $0.3 \pm 0.005$, where 1 MC cycle implies $N$ trial moves (i.e., one trial move per particle). Then, we keep the system evolving until it is equilibrated in such a manner that the relative standard deviation in the contact
Table 7.1: The number of progenitor packings used in the tessellation-based procedure. Packings are defined by initial packing fraction $\phi_{\text{init}}$ and particle number $N$. For saturated RSA packings with $\phi_{\text{init}} = \phi_{\text{sat}}$, the number of particles can vary for each realization. For reference, the saturated packing fractions of RSA packings are $\phi_{\text{sat}} = 0.5470735(28)$ and $0.3841307(21)$ in $d = 2$ and $3$, respectively [358]. Here, HSL stands for equilibrium hard-sphere liquids below the freezing point.

<table>
<thead>
<tr>
<th>Models</th>
<th>$\phi_{\text{init}}$</th>
<th>$N$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\phi_{\text{sat}}$</td>
<td>$10^3$</td>
</tr>
<tr>
<td>2D RSA</td>
<td>0.41025</td>
<td>0.27350</td>
</tr>
<tr>
<td>3D RSA</td>
<td>0.288</td>
<td>0.192</td>
</tr>
<tr>
<td>2D HSL</td>
<td>0.65</td>
<td>0.40</td>
</tr>
<tr>
<td>3D HSL</td>
<td>0.45</td>
<td>0.30</td>
</tr>
</tbody>
</table>

Table 7.2: The number of lattice packings with spatially uncorrelated vacancies, used in the tessellation-based procedure. Packings are defined by the vacancy concentration $c$ and the initial particle number $N_s$. Thus, particle number is $N = (1 - c)N_s$.

<table>
<thead>
<tr>
<th>Models</th>
<th>$N_s$</th>
<th>$c$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>0.1</td>
</tr>
<tr>
<td>$\mathbb{Z}^2$ lattice with vacancies</td>
<td>$10^6$</td>
<td>100</td>
</tr>
<tr>
<td></td>
<td>$25 \times 10^6$</td>
<td>50</td>
</tr>
<tr>
<td></td>
<td>$100 \times 10^6$</td>
<td>20</td>
</tr>
<tr>
<td>$\mathbb{Z}^3$ lattice with vacancies</td>
<td>$10^6$</td>
<td>100</td>
</tr>
<tr>
<td></td>
<td>$27 \times 10^6$</td>
<td>50</td>
</tr>
</tbody>
</table>

value of the radial distribution function $g_r(r = D^+)$ for four successive MC cycles becomes less than $10^{-3}$. After equilibration, we sample a configuration every 10000 MC cycles. The number of sampled configurations is listed in Table. 7.1. Using the same analysis in Ref. [84] in the main text, we obtain theoretical prediction of $\tilde{\chi}_V(0)$.
for 3D equilibrium hard-sphere liquids with the packing fraction $\phi_{\text{init}}$ as follows:

$$\tilde{\chi}_V(0) = \frac{\phi^2(1 - \phi_{\text{init}})^4}{\rho(1 + 4\phi_{\text{init}} + 4\phi_{\text{init}}^2 - 4\phi_{\text{init}}^3 + \phi_{\text{init}}^4)}.$$  \hspace{1cm} (7.50)

Figure 7.16: Simulation results for the spectral densities $\tilde{\chi}_V(k)$ of 3D equilibrium hard-sphere liquids with various values of packing fraction $\phi_{\text{init}}$ and sample size $N$. “Prediction” represents $\tilde{\chi}_V(0)$, obtained by Eq. (7.50).

A lattice packing with uncorrelated vacancies is generated by randomly removing $cN_s$ particles from an initial perfect $Z^d$-lattice packing of $N_s$ particles. The employed parameters are listed in Table 7.2.

7.10.2 Maximal packing fractions

TABLEs 7.3, 7.4, and 7.5 summarize maximal packing fractions of constructed sphere packings derived from Voronoi tessellations.

While there is a tendency that the maximal packing fraction slowly decreases as system size grows, we can straightforwardly show that it is bounded below for saturated RSA packings. For saturated RSA packings in $\mathbb{R}^d$ ($d > 1$), their maximal packing fractions is estimated as follows:

$$\phi_{\text{max}}^{(2)} = \frac{\min \left\{ \frac{v_1(R)}{|C_j|} \right\}}{\max \{|C_j|\}} = \frac{v_1(R)}{\max \{|C_j|\}}$$  \hspace{1cm} (7.51)

$$\frac{v_1(R)}{2^d v_1(R)} = \frac{1}{2^d}.$$  \hspace{1cm} (7.52)
Table 7.3: Maximal packing fractions of the packings constructed from the progenitor RSA packings of packing fraction \( \phi_{\text{init}} \). The maximal packing fractions \( \phi^{(2)}_{\text{max}} \) and \( \phi^{(1)}_{\text{max}} \) are defined by Eq. (7.19) and Fig. 7.4. Here, values in the square brackets represent the minimal values that we observed.

<table>
<thead>
<tr>
<th>( d )</th>
<th>( \phi_{\text{init}} )</th>
<th>( N )</th>
<th>( \phi^{(1)}_{\text{max}} )</th>
<th>( \phi^{(2)}_{\text{max}} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( 10^{-6} )</td>
<td>0.360(4)</td>
<td>0.3473</td>
<td>0.305(4)</td>
<td>0.2999</td>
</tr>
<tr>
<td>( 10^{-5} )</td>
<td>0.366(4)</td>
<td>0.3573</td>
<td>0.314(5)</td>
<td>0.2993</td>
</tr>
<tr>
<td>( 10^{-5} )</td>
<td>0.380(7)</td>
<td>0.3578</td>
<td>0.325(7)</td>
<td>0.3070</td>
</tr>
<tr>
<td>( 10^{-4} )</td>
<td>0.395(10)</td>
<td>0.3581</td>
<td>0.341(10)</td>
<td>0.3050</td>
</tr>
<tr>
<td>( 10^{-3} )</td>
<td>0.418(14)</td>
<td>0.3626</td>
<td>0.362(13)</td>
<td>0.3086</td>
</tr>
<tr>
<td>( 0.41025 )</td>
<td>0.217(12)</td>
<td>0.1792</td>
<td>0.1769(5)</td>
<td>0.1650</td>
</tr>
<tr>
<td>( 10^{-4} )</td>
<td>0.234(8)</td>
<td>0.2156</td>
<td>0.201(8)</td>
<td>0.1792</td>
</tr>
<tr>
<td>( 10^{-3} )</td>
<td>0.252(10)</td>
<td>0.2189</td>
<td>0.217(10)</td>
<td>0.1796</td>
</tr>
<tr>
<td>( 0.2735 )</td>
<td>0.106(5)</td>
<td>0.0921</td>
<td>0.076(5)</td>
<td>0.0655</td>
</tr>
<tr>
<td>( 10^{-4} )</td>
<td>0.116(6)</td>
<td>0.0909</td>
<td>0.084(4)</td>
<td>0.0707</td>
</tr>
<tr>
<td>( 10^{-3} )</td>
<td>0.132(8)</td>
<td>0.1017</td>
<td>0.094(5)</td>
<td>0.0793</td>
</tr>
<tr>
<td>( 10^{-4} )</td>
<td>0.150(11)</td>
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<td>0.106(7)</td>
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<td>0.167(16)</td>
<td>0.1030</td>
<td>0.123(10)</td>
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<tr>
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<td>0.0395</td>
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<tr>
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<td>0.0395</td>
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<td>0.083(10)</td>
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<td>0.0998</td>
<td>0.1083(1)</td>
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where \( R \) is the particle radius in a progenitor packing. In Eq. (7.52), we use the fact that any Voronoi cell of a saturated RSA packing should be fully covered by a sphere of volume \( 2^d v_1(R) \), i.e., \( |C_j| < 2^d v_1(R) \) for \( j = 1, \cdots, N \). Here, we note that \( \phi^{(2)}_{\text{max}} \leq \phi^{(1)}_{\text{max}} \leq \phi_{\text{max}} \).
Table 7.4: Maximal packing fractions of the constructed sphere packings according to the progenitor packings from equilibrium hard-sphere liquids. The maximal packing fractions $\phi_{\text{max}}^{(2)}$ and $\phi_{\text{max}}^{(1)}$ are defined by by Eq. (7.19) and Fig. 7.4. Here, the values in square brackets represent the minimal values that we observed.

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<th>$\phi_{\text{max}}^{(2)}$</th>
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<td>0.470(1) [0.3741]</td>
<td>0.428(1) [0.3360]</td>
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<tr>
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<td>0.5053(2) [0.3842]</td>
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<td>0.1567(8) [0.1308]</td>
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<td>0.2585(2) [0.1721]</td>
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<td>0.0551(3) [0.0453]</td>
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<td>0.0762(1) [0.0446]</td>
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<td>0.287(7) [0.2682]</td>
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<td>0.3599(4) [0.3064]</td>
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<td>0.3603(1) [0.2998]</td>
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<td>0.098(4) [0.0855]</td>
<td>0.079(4) [0.0691]</td>
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<tr>
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<td>0.0945(2) [0.0733]</td>
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<tr>
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<td>$10^6$</td>
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<td>0.1354(96) [0.0920]</td>
<td>0.1070(72) [0.0647]</td>
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7.10.3 Values of extrapolated $\tilde{\chi}_V(0)$

The limit values of $\lim_{k \to 0} \tilde{\chi}_V(k)$ for the constructed sphere packings are estimated by computing the spectral density $\tilde{\chi}_V(k_{\text{min}})$ at the smallest wavenumber and by extrapolating simulation results with a non-linear curve $y = \tilde{\chi}_V(0) + a_1 x^4$. In the non-linear regression, we take errors in $\tilde{\chi}_V(k)$ into account. Then, for RSA packings, equilibrium hard-sphere liquids, and their constructed sphere packings, values of hyperuniform
Table 7.5: Maximal packing fractions of the constructed sphere packings from imperfect \( \mathbb{Z}^d \) packings with uncorrelated point vacancy concentration \( c \). The maximal packing fractions \( \phi_{\text{max}}^{(2)} \) and \( \phi_{\text{max}}^{(1)} \) are defined by Eq. (7.19) and Fig. 7.4. Here, values in the square brackets represent the minimal values that we observed.

<table>
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<tr>
<th>( d )</th>
<th>( c )</th>
<th>( N_s )</th>
<th>( \phi_{\text{max}}^{(1)} )</th>
<th>( \phi_{\text{max}}^{(2)} )</th>
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<td>0.10</td>
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<td>0.229(7)</td>
<td>0.2142</td>
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<td>25 ( \times 10^6 )</td>
<td>0.236(11)</td>
<td>0.2081</td>
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<tr>
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<td>0.267(11)</td>
<td>0.2327</td>
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<td>100 ( \times 10^6 )</td>
<td>0.173(8)</td>
<td>0.1611</td>
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<td>25 ( \times 10^6 )</td>
<td>0.183(8)</td>
<td>0.1608</td>
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<td>0.208(11)</td>
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Metric \( H \) are computed as follows:

\[
H = \begin{cases} 
\frac{\tilde{\chi}_V(k_{\text{min}})}{\tilde{\chi}_V(\text{peak})}, & \text{Progenitor packings} \\
\frac{\tilde{\chi}_V(0)}{\tilde{\chi}_V(\text{peak})}, & \text{Constructed packings,}
\end{cases}
\]  

(7.53)

where \( \tilde{\chi}_V(\text{peak}) \) represents the spectral density at the first dominant non-Bragg peak.

Tables 7.6, 7.7, and 7.8 summarize simulation data.
Table 7.6: Values of hyperuniform metric $H$ for RSA packings ($d = 2, 3$) and corresponding constructed packings. Spectral density $\tilde{\chi}_V(k_{\text{min}})$ at the minimum wavenumber and the extrapolated values of $\tilde{\chi}_V(0)$ are summarized.

<table>
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<tr>
<th>$d$</th>
<th>$\phi_{\text{init}}$</th>
<th>$N$</th>
<th>Progenitor packing</th>
<th>$X_V(k_{\text{min}})$</th>
<th>$H$</th>
<th>Constructed packing</th>
<th>$X_V(k_{\text{min}})$</th>
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<tr>
<td>10^4</td>
<td>$\phi_{\text{sat}}$</td>
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<td>$2.29(54) \times 10^{-18}$</td>
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3D

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<th>$X_V(k_{\text{min}})$</th>
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Table 7.7: Values of hyperuniform metric $H$ for equilibrium hard-sphere liquids ($d = 2$ and $3$) and corresponding constructed packings. Spectral density $\tilde{\chi}_V(k_{min})$ at the minimum wavenumber and the extrapolated values of $\tilde{\chi}_V(0)$ are summarized.

<table>
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Table 7.8: Values of spectral density $\tilde{\chi}_V(k_{min})$ at the minimum wavenumber and the extrapolated values of $\tilde{\chi}_V(0)$ for the imperfect $\mathbb{Z}^d$-lattice packings and the constructed sphere packings.

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<th>$\tilde{\chi}_V(0)$</th>
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<td></td>
<td></td>
<td>$\tilde{\chi}<em>V(k</em>{min})$</td>
<td>$H$</td>
</tr>
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<td>$\tilde{\chi}_V(0)$</td>
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<td>6.14(54) $\times 10^{-18}$</td>
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<td>$25 \times 6.05(43) \times 10^{-6}$</td>
<td>4.56(36) $\times 10^{-16}$</td>
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<td>$5.13(26) \times 10^{-6}$</td>
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<td>5.80(21) $\times 10^{-10}$</td>
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### Appendix C: Multiscale-Disk Tessellations

Table 7.9: The prescribed cell number $N$ in every stage for our coated-disks models. The upper bound on cell volumes is denoted by $v_{\text{max}}$, and $|V_d|$ is the volume of the simulation box. For each parameter, we obtain 50 and 20 configurations when $v_{\text{max}}/|V_d| = 10^{-3}$ and $10^{-4}$, respectively.

| Type          | Parameter | $v_{\text{max}}/|V_d|$ | $10^{-3}$ | $10^{-4}$ |
|---------------|-----------|------------------------|----------|----------|
| Power-law     | 1.5       | 383                    | -        |          |
|               | 1.6       | 438                    | 4375     |          |
|               | 1.7       | 487                    | 4868     |          |
|               | 1.8       | 532                    | -        |          |
| Exponential   | 1.05      | 48                     | -        |          |
|               | 1.10      | 91                     | -        |          |
|               | 1.20      | 167                    | -        |          |

Table 7.9 lists the simulation parameters employed to construct multiscale-disk tessellations. Figure 7.17 summarizes the number of inserted disks in the $m$th stage when one uses the power-law scaling [i.e., $v^{(m)}/v^{(1)} = 1/m^p$]. In this figure, $N_m/N < 1$ at the $m$th stage implies that the procedure fails to insert a desired number of disks, i.e., the packing is saturated. Some stages with $N_m/N > 1$ may immediately follow because the algorithm tries to insert more spheres than $N$ to keep up with the scheduled covering fractions.

When $p = 1.5$, saturation hardly occurs so that the covering fraction is virtually identical to the theoretical values. However, the covering fraction increases slowly compared with cases of $p > 1.5$. For $p = 1.6$, saturation often occurs, but the procedure immediately keeps up the scheduled covering fraction. For $p > 1.6$, however, saturation occurs at very early stages, and once saturation occurs $N_m/N$ never becomes unity, i.e., the packings never keep up with the scheduled covering fraction.

Figure 7.18 is a magnification of Fig. 7.11.
Figure 7.17: (a) The number $N_m$ of spheres inserted in the $m$th stage in our multiscale-disk tessellations. (b) The inserted number $N_m$ as a function of cell-volumes $v^{(m)}$. Error bars represent sample standard deviations.

Figure 7.18: An enlarged portion ($\times 8$ magnification) of Fig. 7.11. This packing is generated via a power-law scaling with $p = 1.5$ up to the 400th stage.

### 7.12 Appendix D: Derivation of Eq. (7.24)

First, we rewrite the Fourier transform of the particle indicator function $\tilde{J}(k)$, given in Eq. (7.21):

$$\tilde{J}(k) = \sum_{j=1}^{N} e^{-ik \cdot x_j} \left\{ \tilde{m}(k; P_j) e^{-ik \cdot \Delta X_j} - \phi \tilde{m}(k; C_j) \right\}$$

$$= \sum_{j=1}^{N} e^{-ik \cdot x_j} \left\{ \left( \tilde{m}(k; P_j) - \phi \tilde{m}(k; C_j) \right) + \tilde{m}(k; P_j) \left( e^{-ik \cdot \Delta X_j} - 1 \right) \right\}.$$
Here, we substitute the form factors $\tilde{m}(k; P_j)$ and $\tilde{m}(k; C_j)$ with their Taylor expansions (7.22), which yields

$$\tilde{J}(k) = \sum_{j=1}^{N} \phi |C_j| e^{-i k \cdot x_j} \left\{ -\frac{k_\alpha k_\beta}{2} (M_{\alpha\beta}(P_j) - M_{\alpha\beta}(C_j)) + i k_\alpha k_\beta k_\gamma \left( M_{\alpha\beta\gamma}(P_j) - M_{\alpha\beta\gamma}(C_j) \right) \right\} + O(k^4)$$

(7.54)

$$= \phi \sum_{j=1}^{N} |C_j| e^{-i k \cdot x_j} \left\{ (e^{-i k \cdot \Delta X_j} - 1) - \frac{k_\alpha k_\beta}{2} (M_{\alpha\beta}(P_j) - M_{\alpha\beta}(C_j)) \right\} + \frac{i k_\alpha k_\beta k_\gamma}{6} (M_{\alpha\beta\gamma}(P_j) - M_{\alpha\beta\gamma}(C_j)) + \frac{k_\alpha k_\beta}{2} M_{\alpha\beta}(P_j) (e^{-i k \cdot \Delta X_j} - 1) + O(k^4)$$

$$= \phi \sum_{j=1}^{N} (e^{-i k \cdot \Delta X_j} - 1) |C_j| e^{-i k \cdot x_j} + \frac{1}{2} \phi \sum_{j=1}^{N} (M_{\alpha\beta}(P_j) - M_{\alpha\beta}(C_j)) |C_j| e^{-i k \cdot x_j} + \frac{i k_\alpha k_\beta k_\gamma}{6} \sum_{j=1}^{N} (M_{\alpha\beta\gamma}(P_j) - M_{\alpha\beta\gamma}(C_j)) + 3(\Delta X_j)_\gamma M_{\alpha\beta}(P_j)) |C_j| e^{-i k \cdot x_j} + O(k^4).$$

(7.55)

In Eq. (7.54), we used the identical local-cell packing fraction condition, i.e., $|P_j| = \phi |C_j|$ for $j = 1, \cdots, N$. In Eq. (7.55), $(\Delta X_j)_\gamma$ is the $\gamma$th Cartesian component of a vector $\Delta X_j$, and we apply the Taylor expansion $(e^{-i k \cdot \Delta X_j} - 1) = -i k_\gamma (\Delta X_j)_\gamma + O(k^2)$, which is a good approximation due to the bounded-cell condition (7.18).

### 7.13 Appendix E: Derivations of the spectral density of the $m$th stage coated-spheres model

Here, we compute upper bounds on the spectral density $\tilde{\chi}^{(m)}(k)$ defined in Eq. (7.42) in three cases of cell-volume distributions: unknown, a power-law scaling, and an exponential scaling.
7.13.1 Upper bounds of the spectral density

From Eq. (7.42),

\[ \tilde{\chi}^{(m)}(k) = \frac{1}{|V_F|} \left\langle \left\{ \sum_{j=1}^{N_m} [\tilde{m}(k; \phi^{1/d}R_j) - \phi \tilde{m}(k; R_j)] e^{-ik\cdot x_j} - \phi \sum_{j=N_m+1}^{\infty} \tilde{m}(k; R_j) e^{-ik\cdot x_j} \right\}^2 \right\rangle, \]

\[ = \frac{1}{|V_F|} \left\langle \left\{ \frac{\phi(1 - \phi^2/d)}{2(2 + d)} \sum_{j=1}^{N_m} v_1(R_j) (kR_j)^2 e^{-ik\cdot x_j} + O(k^4) - \phi \sum_{j=N_m+1}^{\infty} \tilde{m}(k; R_j) e^{-ik\cdot x_j} \right\}^2 \right\rangle. \]  

(7.56)

Now, note the following inequality for two complex numbers \( A \) and \( B \):

\[ |A + B|^2 \leq (|A| + |B|)^2 = |A|^2 + |B|^2 + 2|A||B| \]

\[ \leq 2(|A|^2 + |B|^2), \]

(7.57)

which is obtained by successively applying the triangle inequality and then the inequality of arithmetic and geometric means, and thus equality occurs if and only if \( A = B \). Using this inequality, we obtain a rigorous upper bound on Eq. (7.56):

\[ \tilde{\chi}^{(m)}(k) \leq \frac{2}{|V_F|} \left( \frac{\phi(1 - \phi^2/d)}{2(2 + d)} \right)^2 \left\langle \left\{ \sum_{j=1}^{N_m} v_1(R_j) (kR_j)^2 e^{-ik\cdot x_j} + O(k^4) \right\}^2 \right\rangle + \frac{2\phi^2}{|V_F|} \left\langle \left\{ \sum_{j=N_m+1}^{\infty} \tilde{m}(k; R_j) e^{-ik\cdot x_j} \right\}^2 \right\rangle. \]

(7.59)

Subsequently, we derive an upper bound on the second term in Eq. (7.59) by applying the triangle inequality:

\[ \left\langle \left\{ \sum_{j=N_m+1}^{\infty} \tilde{m}(k; R_j) e^{-ik\cdot x_j} \right\}^2 \right\rangle \leq \left\langle \left\{ \sum_{j=N_m+1}^{\infty} |\tilde{m}(k; R_j)| \right\}^2 \right\rangle \leq \left\langle \left\{ \sum_{j=N_m+1}^{\infty} v_1(R_j) \right\}^2 \right\rangle \]

\[ = |V_F|^2 (1 - \eta_m)^2, \]

(7.60)
where the inequality in Eq. (7.60) comes from the fact that $|\tilde{m}(k; R)| \leq v_1(R)$; see Eq. (7.9). We note that the last term in Eq. (7.60) can be regarded as the largest volume-fraction fluctuations contributed from the uncovered gaps. Combining Eqs. (7.59) and (7.60), we obtain a rigorous bound as follows:

$$
\tilde{\chi}_v^{(m)}(k) \leq 2 F(k; \phi) + 2\phi^2|V_F|(1 - \eta_m)^2, \quad (7.61)
$$

where $F(k; \phi)$ is defined by Eq. (7.44). We remark that this bound is derived without any prior knowledge of the cell-volume distribution.

### 7.13.2 Derivation of Eq. (7.45)

We can approximate Eq. (7.42) by assuming that its contributions from composite spheres (the first term in Eq. (7.56)) and uncovered gaps are uncorrelated, which effectively removes an ensemble average of their cross terms:

$$
\tilde{\chi}_v^{(m)}(k) \approx F(k; \phi) + \frac{\phi^2}{|V_F|} \left< \left| \sum_{j=N_m+1}^{\infty} \tilde{m}(k; \mathcal{R}_j) e^{-ik \cdot x_j} \right|^2 \right>. \quad (7.62)
$$

Here, the second term can be further simplified by assuming that the uncovered gaps are spatially uncorrelated:

$$
\frac{\phi^2}{|V_F|} \left< \left| \sum_{j=N_m+1}^{\infty} \tilde{m}(k; \mathcal{R}_j) e^{-ik \cdot x_j} \right|^2 \right> \approx \frac{\phi^2}{|V_F|} \left< \left| \sum_{j=N_m+1}^{\infty} \tilde{m}(k; \mathcal{R}_j) \right|^2 \right> \approx \frac{\phi^2}{|V_F|} \left< \left| \sum_{j=N_m+1}^{\infty} v_1(\mathcal{R}_j) \right|^2 \right>,
$$

where the last approximation comes from $|\tilde{m}(k; R)| \approx v_1(R) + \mathcal{O}(k^2)$ when $kR \ll 1$; see Eq. (7.9).
Chapter 8

Nonlocal Effective Electromagnetic Wave Characteristics of Composite Media: Beyond the Quasistatic Regime

8.1 Introduction

The theoretical problem of estimating the effective properties of multiphase composite media is an outstanding one and dates back to work by some of the luminaries of science, including Maxwell [202], Lord Rayleigh [187] and Einstein [69]. The preponderance of previous theoretical studies have focused on the determination of static effective properties (e.g., dielectric constant, elastic moduli and fluid permeability) using a variety of methods, including approximation schemes [202, 34, 32, 35], bounding techniques [236, 114, 21, 164, 210, 211, 296, 213] and exact series-expansion procedures [33, 76, 264, 293]. The latter set of investigations teaches us that an exact determination of an effective property, given the phase properties of the compos-
ite, generally requires an infinite set of correlation functions that characterizes the composite microstructure.

Our focus in this chapter is the determination of the effective dynamic dielectric constant tensor $\varepsilon_e(k_l, \omega)$ of a two-phase dielectric composite, which depends on the frequency $\omega$ or wavenumber $k_l$ of the incident radiation [266, 270]. From this effective property, one can determine the corresponding effective wave speed $c_e$ and attenuation coefficient $\gamma_e$. Virtually all previous homogenization estimates of $\varepsilon_e(k_l, \omega)$ apply only in the *quasistatic* or long-wavelength regime [139], i.e., applicable when $|k_l|\ell \ll 1$, where $\ell$ is a characteristic heterogeneity length scale. This spectral range is the realm of non-resonant dielectric behavior. Examples of such popular closed-form approximation formulas devised for spherical scatterers in a matrix include the Maxwell-Garnett [93, 253] and quasicrystalline [174, 270, 6] estimates, among others [266].

In the present investigation, we derive nonlocal homogenized constitutive relations from Maxwell’s equations to obtain exact expressions for the effective dynamic dielectric constant tensor $\varepsilon_e(k_l, \omega)$ of a macroscopically anisotropic two-phase medium of arbitrary microstructure that are valid well beyond the quasistatic regime, i.e., from the infinite-wavelength limit down to intermediate wavelengths ($0 \leq |k_l|\ell \lesssim 1$). This is accomplished by extending the strong-contrast expansion formalism, which has been used in the past exclusively for the static limit [296] and quasistatic regime [242], and establishing that the resulting homogenized constitutive relations are non-local in space (Sec. 8.4), i.e., the average polarization field at position $x$ depends on the average electric field at other positions around $x$. (Such nonlocal relations are well-known in the context of crystal optics in order to account for “spatial dispersion,” i.e., dependence of dielectric properties on wavevector [1].) The terms of

\[\text{A notable exception is the generalized coherent potential approximation for particle suspensions presented in Ref. [139]. However, this is not a closed-form formula for the effective dielectric constant and requires, as input, certain numerical simulations of the electric fields.}\]
the strong-contrast expansion are explicitly given in terms of integrals over products of Green’s functions and the \(n\)-point correlation functions of the random two-phase medium (defined in Sec. 8.2.1) to infinite order. This implies that multiple scattering to all orders is exactly treated for the range of wavenumbers for which our extended homogenization theory applies, i.e., \(0 \leq |k_I|\ell \lesssim 1\).

Due to the fast-convergence properties of such expansions, elaborated in Sec. 8.4.2, their lower-order truncations yield accurate closed-form approximate formulas for the effective dielectric constant that apply for a wide class of microstructures over the aforementioned broad range of incident wavelengths, volume fractions and contrast ratios (Sec. 8.5). Thus, we are able to accurately account for multiple scattering in the resonant realm (e.g., Bragg diffraction for periodic media), in contrast to the Maxwell-Garnett and quasicrystalline approximations, which are known to break down in this spectral range. These nonlocal strong-contrast formulas can be regarded to be approximate resummations of the expansions that still accurately capture multiple scattering effects to all orders via the nonlocal attenuation function \(F(Q)\) (Sec. 8.6). The key quantity \(F(Q)\) is a functional of the spectral density \(\tilde{\chi}_V(Q)\) (Sec. 8.2.2), which is straightforward to determine for general microstructures either theoretically, computationally or via scattering experiments. We employ precise full-waveform simulation methods (Sec. 8.7) to show that these microstructure-dependent approximations are accurate for both two-dimensional (2D) and three-dimensional (3D) ordered and disordered models of particulate composite media (Sec. 8.8). This validation means that they can be used to predict accurately the effective wave characteristics well beyond the quasistatic regime for a wide class of composite microstructures (Sec. 8.9) without having to perform full-blown simulations. This broad microstructure class includes particulate media consisting of identical or polydisperse particles of general shape (ellipsoids, cubes, cylinders, polyhedra) that may or not overlap, cellular networks as well as media without well-defined inclusions (see Sec. 8.4.2 for
Thus, our nonlocal formulas can be employed to accelerate the discovery of novel electromagnetic composites by appropriate tailoring of the spectral densities [298, 46] and then generating the microstructures that achieve them [46].

Although our strong-contrast formulas for the effective dynamic dielectric constant apply to periodic two-phase media, the primary applications that we have in mind are correlated disordered microstructures because they can provide advantages over periodic ones with high crystallographic symmetries [186, 341] which include perfect isotropy and robustness against defects [79, 196]. We are interested in both “garden-variety” models of disordered two-phase media [296] as well as exotic hyperuniform forms [313, 346, 300]. Hyperuniform two-phase systems are characterized by an anomalous suppression of volume-fraction fluctuations in the infinite-wavelength limit [313, 346, 300], i.e., the spectral density \( \tilde{\chi}_V(Q) \) obeys the condition

\[
\lim_{|Q| \to 0} \tilde{\chi}_V(Q) = 0.
\]  

Hyperuniform systems have attracted great attention over the last decade because of their deep connections to a wide range of topics that arise in physics [317, 354, 121, 245, 225, 194, 186, 343, 324, 300, 177, 104, 160], materials science [193, 339, 303, 46, 157], mathematics [97, 30, 316], and biology [137, 300] as well as for their emerging technological importance in the case of the disordered varieties [80, 196, 193, 179, 82, 163, 359, 300, 104].

We apply our strong-contrast formulas to predict the real and imaginary parts of the effective dielectric constant for model microstructures with typical disorder (nonhyperuniform) as well as those with exotic hyperuniform disorder (Sec. 8.3). We are particularly interested in exploring the dielectric properties of a special class of hyperuniform composites called disordered stealthy hyperuniform media, which are defined to be those that possess zero-scattering intensity for a set of wavevectors.
around the origin [321, 17, 317, 352, 46]. Such materials have recently been shown to be endowed with novel optical, acoustic, mechanical, and transport properties [242, 179, 355, 57, 102, 339, 303, 158]. Among other findings, we show that disordered hyperuniform media are generally less lossy than their nonhyperuniform counterparts. We also demonstrate that disordered stealthy hyperuniform particulate composites exhibit singular wave characteristics, including the capacity to act as low-pass filters that transmit waves ‘isotropically’ up to a selected wavenumber. They also can be engineered to exhibit refractive indices that abruptly change over a narrow range of wavenumbers by tuning the spectral density. Our results demonstrate that one can design the effective wave characteristics of a disordered composite, hyperuniform or not, by engineering the microstructure to possess tailored spatial correlations at prescribed length scales.

8.2 Background

8.2.1 \( n \)-Point Correlation Functions

A two-phase random medium is a domain of space \( \mathcal{V} \subseteq \mathbb{R}^d \) that is partitioned into two disjoint regions that make up \( \mathcal{V} \): a phase 1 region \( \mathcal{V}_1 \) of volume fraction \( \phi_1 \) and a phase 2 region \( \mathcal{V}_2 \) of volume fraction \( \phi_2 \) [296]. The phase indicator function \( \mathcal{I}^{(i)}(x) \) of phase \( i \) for a given realization is defined as

\[
\mathcal{I}^{(i)}(x) = \begin{cases} 
1, & x \in \mathcal{V}_i, \\
0, & x \notin \mathcal{V}_i. 
\end{cases}
\]  

(8.2)

The \( n \)-point correlation function \( S_{n}^{(i)} \) for phase \( i \) is defined by [312, 296]:

\[
S_{n}^{(i)}(x_1, \ldots, x_n) = \left\langle \prod_{j=1}^{n} \mathcal{I}^{(i)}(x_j) \right\rangle, \quad \text{(8.3)}
\]
where angular brackets denote an ensemble average over realizations. The function $S^{(i)}_{n}(x_1, \ldots, x_n)$ has a probabilistic interpretation: it gives the probability of finding the ends of the vectors $x_1, \ldots, x_n$ all in phase $i$. For statistically homogeneous media, $S^{(i)}_{n}(x_1, \ldots, x_n)$ is translationally invariant and, in particular, the one-point function is position-independent, i.e., $S^{(i)}_{1}(x_1) = \phi_i$.

### 8.2.2 Two-Point Statistics

For statistically homogeneous media, the two-point correlation function for phase 2 is simply related to that for phase 1 via the expression $S^{(2)}_{2}(r) = S^{(1)}_{2}(r) - 2\phi_1 + 1$, and hence the **autocovariance** function is given by

$$\chi_V(r) \equiv S^{(1)}_{2}(r) - \phi_1^2 = S^{(2)}_{2}(r) - \phi_2^2,$$

which we see is the same for phase 1 and phase 2. Thus, $\chi_V(r = 0) = \phi_1\phi_2$ and, assuming the medium possesses no long-range order, $\lim_{|r| \to \infty} \chi_V(r) = 0$. For statistically homogeneous and isotropic media, $\chi_V(r)$ depends only on the magnitude of its argument $r = |r|$, and hence is a radial function. In such cases, its slope at the origin is directly related to the **specific surface** $s$ (interface area per unit volume), i.e., asymptotically, we have [296]

$$\chi_V(r) = \phi_1\phi_2 - \beta(d)s|r| + \mathcal{O}(|r|^2),$$

where

$$\beta(d) = \frac{\Gamma(d/2)}{2\sqrt{\pi}\Gamma((d+1)/2)},$$

and $\Gamma(x)$ is the gamma function.

The nonnegative spectral density $\tilde{\chi}_V(Q)$, which is proportional to scattering in-
tensity [53], is the Fourier transform of $\chi_V(r)$, i.e.,

$$\tilde{\chi}_V(Q) = \int_{\mathbb{R}^d} \chi_V(r) e^{-iQ \cdot r} dr \geq 0, \quad \text{for all } Q,$$

(8.7)

where $Q$ represents the momentum-transfer wavevector. For statistically homogeneous media, the spectral density must obey the following sum rule [302]

$$\frac{1}{(2\pi)^d} \int_{\mathbb{R}^d} \tilde{\chi}_V(Q) dQ = \chi_V(r = 0) = \phi_1 \phi_2.$$  

(8.8)

For isotropic media, the spectral density only depends on $Q = |Q|$ and, as a consequence of (8.5), its large-$k$ behavior is controlled by the following power-law form:

$$\tilde{\chi}_V(Q) \sim \frac{\gamma(d) s}{Q^{d+1}}, \quad Q \to \infty,$$

(8.9)

where $\gamma(d) = 2^d \pi^{(d-1)/2} \Gamma((d+1)/2) \beta(d)$ is a $d$-dimensional constant and $\beta(d)$ is given by (8.6).

### 8.2.3 Packings

We call a *packing* in $\mathbb{R}^d$ a collection of nonoverlapping particles [301]. In the case of a packing of identical spheres of radius $a$ at number density $\rho$, the spectral density $\tilde{\chi}_V(Q)$ is directly related to the structure factor $S(Q)$ of the sphere centers [296, 298]:

$$\tilde{\chi}_V(Q) = \phi_2 \tilde{\alpha}_2(Q; a) S(Q),$$

(8.10)

where

$$\tilde{\alpha}_2(Q; a) = \frac{1}{v_1(a)} \left( \frac{2\pi a}{Q} \right)^d J_{d/2}(Qa),$$

(8.11)
$J_\nu(x)$ is the Bessel function of the first kind of order $\nu$, $\phi_2 = \rho v_1(a)$ is the packing fraction (fraction of space covered by the spheres), and

$$v_1(a) = \frac{\pi^{d/2} a^d}{\Gamma(1 + d/2)}$$

is the $d$-dimensional volume of a sphere of radius $a$.

### 8.2.4 Hyperuniformity and Volume-Fraction Fluctuations

For typical disordered two-phase media, the variance $\sigma_v^2(R)$ for large $R$ goes to zero like $R^{-d}$ [188, 238, 296]. However, for hyperuniform disordered media, $\sigma_v^2(R)$ goes to zero asymptotically more rapidly than the inverse of the window volume, i.e., faster than $R^{-d}$, which is equivalent to the condition (8.1) on the spectral density. *Stealthy hyperuniform* two-phase media are a subclass of hyperuniform systems in which $\tilde{\chi}_v(Q)$ is zero for a range of wavevectors around the origin, i.e.,

$$\tilde{\chi}_v(Q) = 0 \quad \text{for} \quad 0 \leq |Q| \leq Q_U,$$

where $Q_U$ is some positive number.

As in the case of hyperuniform point configurations [313, 346, 347, 300], there are three different scaling regimes (classes) that describe the associated large-$R$ behaviors of the volume-fraction variance when the spectral density goes to zero as a power law $\tilde{\chi}_v(Q) \sim |Q|^\alpha$ as $|Q| \to 0$:

$$\sigma_v^2(R) \sim \begin{cases} R^{-(d+1)}, & \alpha > 1 \quad \text{(Class I)} \\ R^{-(d+1) \ln R}, & \alpha = 1 \quad \text{(Class II)}, \\ R^{-(d+\alpha)}, & 0 < \alpha < 1 \quad \text{(Class III)} \end{cases}$$

where the exponent $\alpha$ is a positive constant. Thus, the characteristics length of the
representative elementary volume for a hyperuniform medium will depend on the hyperuniformity class (scaling). Class I is the strongest form of hyperuniformity, which includes all perfect periodic packings as well as some disordered packings, such as disordered stealthy packings described in Sec. 8.3.4.

8.2.5 Popular Effective-Medium Approximations

Here we explicitly state the specific functional forms of an extended Maxwell-Garnett approximation and quasicrystalline approximation for the effective dynamic dielectric constant $\varepsilon_e(k_1)$ of isotropic media composed of identical spheres of dielectric constant $\varepsilon_2$ embedded in a matrix phase of dielectric constant $\varepsilon_1$. In Sec. 8.8, we compare the predictions of these formulas to those of our nonlocal approximations. The small-wavenumber expansions of these popular approximations are provided in Appendix 8.18. There we also provide the corresponding asymptotic behaviors of our strong-contrast approximations.

Maxwell-Garnett approximation

Maxwell-Garnett approximations (MGAs) [93, 253] are derived by substituting the dielectric polarizability of a single dielectric sphere into the Clausius-Mossotti equation [253], which consequently ignores the spatial correlations between the particles. In three dimensions, we utilize the following extended MGA that makes use of the exact electric dipole polarizability $\alpha_e(k_1)$ of a single dielectric sphere of radius $a$ [63]:

$$\frac{\varepsilon_e(k_1) - \varepsilon_1}{\varepsilon_e(k_1) + 2\varepsilon_1} = \phi_2 \frac{\alpha_e(k_1)}{a^3},$$

(8.15)

where

$$\alpha_e(k_1) = \frac{3i}{2k_1^3} \frac{m}{m \psi_1(mk_1a)} \frac{\psi_1(k_1a)}{\psi_1(mk_1a)} - \frac{\psi_1(k_1a)}{\psi_1(mk_1a)}.$$
\[
m \equiv \sqrt{\varepsilon_2 / \varepsilon_1}, \quad \psi_1(x) \equiv x j_1(x), \quad \xi_1(x) \equiv x h_1^{(1)}(x),
\]
the prime symbol (') denotes the derivative of a function, \( h_1^{(1)}(x) \) is the spherical Hankel function of the first kind of order 1, and \( j_1(x) \) is the spherical Bessel function of the first kind of order 1.

The 2D analog of Eq. (8.15) can be obtained by using the dynamic dielectric polarizability \( \alpha_e \) of a dielectric cylinder of radius \( a \) given in Ref. [271]:

\[
\frac{\varepsilon_e(k_1) - \varepsilon_1}{\varepsilon_e(k_1) + \varepsilon_1} = \frac{\phi_2}{2\pi \alpha_e(k_1) / a^2},
\]
where \( \alpha_e(k_1) = \frac{4(\varepsilon_2 - \varepsilon_1)}{ik_1^2 m \varepsilon_1} J_1(mk_1a) \left[ J_1'(mk_1a) H_1^{(1)}(k_1a) - m J_1(mk_1a) H_1^{(1)'}(k_1a) \right]^{-1},
\)
\( H_\nu^{(1)}(x) \) is the Hankel function of the first kind of order \( \nu \). Here, only TE polarization is considered, i.e., the electric field is perpendicular to the axis of the cylinder.

As with all MGA theories, formulas (8.15) and (8.16) neglect spatial correlations between the spheres and hence are only valid for low inclusion packing fractions. In the static limit, Eqs. (8.15) and (8.16) reduce to the Hashin-Shtrikman estimates \( \varepsilon_{HS} \); see relation (8.75).

**Quasicrystalline approximations**

The quasicrystalline approximation (QCA) for the quasistatic effective dynamic dielectric constant \( \varepsilon_e(k_1) \) employs the “effective” Green’s function of spherical scatterers up to the level of the pair correlation function \( g_2(r) \) [174, 270, 6]. However, this only accounts for the structure factor in the infinite-wavelength limit [6] [i.e., \( S(0) = 1 + \rho \int (g_2(r) - 1) \, dr \)] and consequently, spatial correlations at finite wavelengths are ignored. The QCA for \( d = 3 \) can be explicitly written as follows [270]:

\[
\phi_2^2 \beta_{21} \left[ \frac{\varepsilon_e(k_1) - \varepsilon_1}{\varepsilon_e(k_1) + 2\varepsilon_1} \right]^{-1} = \phi_2 - i \left\{ \frac{2}{3} \phi_2 S(0) (k_1a)^3 \right\} \beta_{21}, \quad \text{(8.17)}
\]

295
where $\beta_{21}$ is defined in Eq. (8.38). Interestingly, the QCA predicts that the hyperuniform composites $[S(0) = 0]$ will be transparent for all wavenumbers, which cannot be true for stealthy hyperuniform media [cf. (8.13)], since the transparency interval must be finite; see Sec. 8.8. Note that Eq. (8.17) is the complex conjugate of the one given in Ref. [270] so that it is consistent with the sign convention for the imaginary part $\text{Im}[\varepsilon_e]$ used here.

8.3 Model Microstructures

We consider four models of 2D and 3D disordered media to understand the effect of microstructure on the effective dynamic dielectric constant, two of which are nonhyperuniform (overlapping spheres and equilibrium hard-sphere packings) and two of which are hyperuniform (hyperuniform polydisperse packings and stealthy hyperuniform packings of identical spheres). The particles of dielectric constant $\varepsilon_2$ are distributed throughout a matrix of dielectric constant $\varepsilon_1$. We also compute the spectral density for each model, which is the required microstructural information to evaluate the nonlocal strong-contrast approximations discussed in Sec. 8.3.5.

Representative images of configurations of the four aforementioned models of 2D disordered particulate media are depicted in Fig. 8.1. Note that the degree of volume-fraction fluctuations decreases from the leftmost image to the rightmost one.

8.3.1 Overlapping Spheres

Overlapping spheres (also called fully-penetrable-sphere model) refer to an uncorrelated (Poisson) distribution of spheres of radius $a$ throughout a matrix [296]. For such nonhyperuniform models at number density $\rho$ in $d$-dimensional Euclidean space
Figure 8.1: Representative images of configurations of the four models of 2D disordered particulate media described in this section. These include (a) overlapping spheres, (b) equilibrium packings, (c) class I hyperuniform polydisperse packings, and (d) stealthy hyperuniform packings. For all models, the volume fraction of the dispersed phase (shown in black) is $\phi_2 = 0.25$. Note that (a) and (b) are not hyperuniform.

In $\mathbb{R}^d$, the autocovariance function is known analytically [296]:

$$\chi_V(r) = \exp(-\rho v_2(r;a)) - \phi_1^2,$$

(8.18)

where $\phi_1 = \exp(-\rho v_1(a))$ is the volume fraction of the matrix phase (phase 1), $v_1(a)$ is given by (8.12), and $v_2(r;a)$ represents the union volume of two spheres whose centers are separated by a distance $r$. In two and three dimensions, the latter is explicitly given respectively by

$$\frac{v_2(r;a)}{v_1(a)} = \begin{cases} 2\Theta(x-1) + \frac{2}{\pi} \left[ \pi + x(1-x^2)^{1/2} - \cos^{-1}(x) \right] \Theta(1-x), & d = 2 \\ 2\Theta(x-1) + \left(1 + \frac{3x}{2} - \frac{x^3}{2}\right) \Theta(1-x), & d = 3 \end{cases}$$

where $x \equiv r/2a$, and $\Theta(x)$ (equal to 1 for $x > 0$ and zero otherwise) is Heaviside step function. For $d = 2$ and $d = 3$, the particle phase (phase 2) percolates when $\phi_2 \approx 0.68$ (Ref. [240]) and $\phi_2 \approx 0.29$ (Ref. [246]), respectively. The corresponding spectral densities are easily found numerically by performing the Fourier transforms indicated in (8.7). In this work, we apply this model for $\phi_2$’s well below the percolation thresholds.
8.3.2 Equilibrium Packings

Another disordered nonhyperuniform model we treat are distributions of equilibrium (Gibbs) of identical hard spheres of radius \( a \) along the stable fluid branch [108, 296]. The structure factors of such packings are well approximated by the Percus-Yevick solution [108, 296], which is analytically solvable for odd values of \( d \). For \( d = 3 \), the Percus-Yevick solution gives the following expression for the structure factor \( S(Q) \) [296]:

\[
S(Q) = \left(1 - \rho \frac{16\pi a^3}{q^6} \left\{ [24a_1\phi_2 - 12(a_1 + 2a_2)\phi_2q^2 + (12a_2\phi_2 + 2a_1 + a_2\phi_2)q^4] \cos(q)
+ [24a_1\phi_2q - 2(a_1 + 2a_1\phi_2 + 12a_2\phi_2)q^3] \sin(q) - 24\phi_2(a_1 - a_2q^2) \right\} \right)^{-1},
\] (8.19)

where \( q = 2Qa \), \( a_1 = (1+2\phi_2)^2/(1-\phi_2)^4 \), and \( a_2 = -(1+0.5\phi_2)^2/(1-\phi_2)^4 \). Using this solution in conjunction with (8.10) yields the corresponding spectral density \( \tilde{\chi}_V(Q) \).

For \( d = 2 \), we obtain the spectral density from Monte Carlo generated disk packings [296].

8.3.3 Hyperuniform Polydisperse Packings

Class I hyperuniform packings of spheres with a polydispersity in size can be constructed from nonhyperuniform progenitor point patterns via a tessellation-based procedure [157, 156]. Specifically, we employ the centers of 2D and 3D configurations of identical hard spheres in equilibrium at a packing fraction 0.45 and particle number \( N = 1000 \) as the progenitor point patterns. One begins with the Voronoi tessellation [296] of these progenitor point patterns. We then rescale the particle in the \( j \)th Voronoi cell \( C_j \) without changing its center such that the packing fraction inside this cell is identical to a prescribed value \( \phi_2 < 1 \). The same process is repeated over all cells. The final packing fraction is \( \phi_2 = \sum_{j=1}^{N} v_1(a_j) / V_3 = \rho v_1(a) \), where \( \rho \) is the number density of particle centers and \( a \) represents the mean sphere radius.
In the small-$|Q|$ regime, the spectral densities of the resulting particulate composites exhibit a power-law scaling $\tilde{\chi}_V(Q) \sim |Q|^4$, which are of class I.

### 8.3.4 Stealthy Hyperuniform Packings

Stealthy hyperuniform particle systems, which are also class I, are defined by the spectral density vanishing around the origin, i.e., $\tilde{\chi}_V(Q) = 0$ for $0 < |Q| \leq Q_U$; see Eq.(8.13). We obtain the spectral density from realizations of disordered stealthy hyperuniform packings for $d = 2, 3$ that are numerically generated via the following two-step procedure. Specifically, we first generate such point configurations consisting of $N$ particles in a fundamental cell $\mathcal{F}$ under periodic boundary conditions via the collective-coordinate optimization technique [321, 17, 352], which amounts to finding numerically the ground-state configurations for the following potential energy:

$$\Phi(r^N) = \frac{1}{V_\delta} \sum_Q \tilde{v}(Q) S(Q) + \sum_{i<j} u(r_{ij}),$$  \hspace{1cm} (8.20)

where

$$\tilde{v}(Q) = \begin{cases} 
1, & Q_L < |Q| \leq Q_U \\
0, & \text{otherwise}
\end{cases}$$  \hspace{1cm} (8.21)

and a soft-core repulsive term [356]

$$u(r) = \begin{cases} 
(1 - r/\sigma)^2, & r < \sigma \\
0, & \text{otherwise}
\end{cases}$$  \hspace{1cm} (8.22)

In contrast to the usual collective-coordinate procedure [321, 17, 352], the interaction (8.20) used here also includes a soft-core repulsive energy (8.22), as done in Ref. [356]. Thus, the associated ground-state configurations are still disordered, stealthy and hyperuniform, and their nearest-neighbor distances are larger than the length
scale $\sigma$ due to the soft-core repulsion $u(r)$. Finally, to create packings, we follow Ref. [355] by circumscribing the points by identical spheres of radius $a < \sigma/2$ under the constraint that they cannot overlap (see Appendix 8.17 for certain results concerning stealthy “nonhyperuniform” packings in which $Q_L > 0$). The parameters used to generate these disordered stealthy packings are summarized in Appendix 8.19.1.

### 8.3.5 Spectral Densities for the Four Models

Here, we plot the spectral density $\tilde{\chi}_v(Q)$ for the four models at the selected particle-phase volume fraction of $\phi_2 = 0.25$. From the long- to intermediate-wavelength regimes ($Qa \lesssim 4$), their spectral densities are considerably different from one another. Overlapping spheres departs the most from hyperuniformity, followed by equilibrium packings. Stealthy packings suppress volume-fraction fluctuations to a greater degree than hyperuniform polydisperse packings over a wider range of wavelengths. In the small-wavelength regime ($Qa \gg 4$), all four curves tend to collapse onto a single curve, reflecting that fact that all four models are composed of spheres of similar sizes.

### 8.4 Exact Strong-Contrast Expansions and Non-locality

The original strong-contrast expansions for the effective dynamic dielectric constant obtained by Rechtsman and Torquato [242] were derived from homogenized constitutive relations that are local in space and hence are strictly valid only in the quasistatic regime. In this chapter, we follow the general strong-contrast formalism of Torquato [296] that was devised for the purely static problem [296] and show that it naturally leads to exact homogenized constitutive relations for the averaged fields that are nonlocal in space. The crucial consequences of this development are exact expressions for the effective dynamic dielectric constant tensor $\epsilon_e(k_R)$ for a macroscopically
Figure 8.2: Plots of the spectral density $\tilde{\chi}_V(Q)$ for the four models of 3D disordered media: overlapping spheres, equilibrium packings, class I hyperuniform polydisperse packings, and stealthy hyperuniform packings. In all cases, the volume fraction of the dispersed phase is $\phi_2 = 0.25$. For hyperuniform polydisperse packings, $a$ is the mean sphere radius. The other three models consist of identical spheres of radius $a$. Corresponding graphs of the spectral densities for the 2D models are provided in Appendix 8.20.

anisotropic medium of arbitrary microstructure into which a plane wave of wavevector $k_I$ is incident. These expressions for $\varepsilon_e(k_R)$ are valid from the infinite-wavelength limit down to wavelengths ($\lambda = 2\pi/|k_I|$) on the order of the heterogeneity length scale $\ell$. We explicitly show they necessarily require complete microstructural information, as embodied in the infinite set of $n$-point correlation functions (Sec. 8.2.1) of the composite. We also describe the fast-convergence properties of strong-contrast expansions and their consequences for extracting accurate approximations for $\varepsilon_e(k_R)$.

### 8.4.1 Strong-Contrast Expansions

Consider a macroscopically large ellipsoidal two-phase statistically homogeneous but anisotropic composite specimen in $\mathbb{R}^d$ embedded inside an infinitely large reference phase $I$ with a dielectric constant tensor $\varepsilon_I$. The microstructure is perfectly general, and it is assumed that the inhomogeneity length scale $\ell$ is much smaller than the
Figure 8.3: (a) Schematic of a large $d$-dimensional ellipsoidal, macroscopically anisotropic two-phase composite medium embedded in an infinite reference phase of dielectric constant tensor $\varepsilon_I$ (gray regions) under an applied electric field $\mathbf{E}_0(\mathbf{x}) = \tilde{\mathbf{E}}_0 \exp(i(\mathbf{k}_I \cdot \mathbf{x} - \omega t))$ of a frequency $\omega$ and a wavevector $\mathbf{k}_I$ at infinity. The wavelength $\lambda$ associated with the applied field can span from the quasistatic regime $(2\pi \ell / \lambda \ll 1)$ down to the intermediate-wavelength regime $(2\pi \ell / \lambda \lesssim 1)$, where $\ell$ is the inhomogeneity length scale. (b) After homogenization, the same ellipsoid can be regarded to be a homogeneous specimen with an effective dielectric constant $\varepsilon_e(\mathbf{k}_I, \omega)$, which depends on $\omega$ and $\mathbf{k}_I$. As noted in the main text, we omit the $\omega$ dependence of $\varepsilon_e$ because (without loss of generality) we assume a linear dispersion relation between $|\mathbf{k}_I|$ and $\omega$.

specimen size, i.e., $\ell \ll L$. The shape of this specimen is purposely chosen to be nonspherical since any rigorously correct expression for the effective property must ultimately be independent of the shape of the composite specimen in the infinite-volume limit. It is assumed that the applied or incident electric field $\mathbf{E}_0(\mathbf{x})$ is a plane wave of a angular frequency $\omega$ and wavevector $\mathbf{k}_I$ in the reference phase, i.e.,

$$\mathbf{E}_0(\mathbf{x}) = \tilde{\mathbf{E}}_0 \exp(i(\mathbf{k}_I \cdot \mathbf{x} - \omega t)).$$

(8.23)

Our interest is the exact expression for the effective dynamic dielectric constant tensor $\varepsilon_e(\mathbf{k}_I, \omega)$. Without loss of generality, we assume a linear dispersion relation in the reference phase, i.e., $k_I \equiv |\mathbf{k}_I| = \sqrt{\varepsilon_I \omega / c}$, where $c$ is the speed of light in vacuum, and thus we henceforth do not explicitly indicate the dependence of functions on $\omega$. The composite is assumed to be nonmagnetic, implying that the phase magnetic
permeabilities are identical, i.e., $\mu_1 = \mu_2 = \mu_0$. For simplicity, we assume real-valued, frequency-independent isotropic phase dielectric constants $\varepsilon_1$ and $\varepsilon_2$. Nonetheless, the composite can be generally lossy (i.e., $\varepsilon_e$ is complex-valued) due to scattering from the inhomogeneities in the local dielectric constant. It is noteworthy that our results can be straightforwardly extended to phase dielectric constants that are complex-valued (dissipative media), but this will not be done in this chapter.

Here we present a compact derivation of strong-contrast expansions. It follows the general formalism of Torquato [296] closely but departs from it at certain key steps when establishing the nonlocality of the homogenized constitutive relation. (A detailed derivation is given in Appendix 8.15). For simplicity, we take the reference phase $I$ to be phase $q$ (equal to 1 or 2). Under the aforementioned assumptions, the local electric field $\mathbf{E}(\mathbf{x})$ solves the time-harmonic Maxwell equation [242]:

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{x}) - k_q^2 \mathbf{E}(\mathbf{x}) = \left( \frac{\omega}{c} \right)^2 \mathbf{P}(\mathbf{x}),$$

where $\mathbf{P}(\mathbf{x})$ is the polarization field given by

$$\mathbf{P}(\mathbf{x}) \equiv [\varepsilon(\mathbf{x}) - \varepsilon_q] \mathbf{E}(\mathbf{x})$$

and

$$\varepsilon(\mathbf{x}) = (\varepsilon_p - \varepsilon_q) \mathcal{I}^{(p)}(\mathbf{x}) + \varepsilon_q$$

is the local dielectric constant, and $\mathcal{I}^{(p)}(\mathbf{x})$ is the indicator function for phase $p$ [cf. (8.2)]. The vector $\mathbf{P}(\mathbf{x})$ is the induced flux field relative to reference phase $q$ due to the presence of phase $p$, and hence is zero in the reference phase $q$ and nonzero in the “polarized” phase $p$ ($p \neq q$).

Using the Green’s function formalism, the local electric field can be expressed in
terms of the following integral equation [242, 296]:

\[ E(x) = E_0(x) + \int G^{(q)}(x - x') \cdot P(x') \, dx', \tag{8.27} \]

where the second-rank tensor Green’s function \( G^{(q)}(r) \) associated with the reference phase \( q \) is given by \(^2\)

\[ G^{(q)}(r) = -D^{(q)} \delta(r) + H^{(q)}(r), \tag{8.28} \]

\( D^{(q)} \) is a constant second-rank tensor that arises when one excludes an infinitesimal region around the position of the singularity \( x' = x \) in the Green’s function and \( H^{(q)}(r) \) represents the contribution outside of the “exclusion” region:

\[
H^{(q)}_{ij}(r) = i \frac{\pi}{2 \varepsilon_q} \left( \frac{k_q}{2 \pi r} \right)^{d/2} \left\{ \left[ k_q r \mathcal{H}^{(1)}_{d/2-1}(k_q r) - \mathcal{H}^{(1)}_{d/2}(k_q r) \right] \delta_{ij} + k_q r \mathcal{H}^{(1)}_{d/2+1}(k_q r) \hat{r}_i \hat{r}_j \right\},
\]

\[ d = 2 \]

\[
= \left\{ \begin{array}{l}
\frac{i}{4} \left[ k^2 q^2 \mathcal{H}_0^{(1)}(k_q r) - i k_q r \mathcal{H}_1^{(1)}(k_q r) \right] \delta_{ij} + \frac{k_q^2}{4} \mathcal{H}_2^{(1)}(k_q r) \hat{r}_i \hat{r}_j, \\
\frac{\exp(i k_q r)}{\varepsilon_q 4 \pi r} \left\{ [-1 + i k_q r + (k_q r)^2] \delta_{ij} + [3 - 3 i k_q r - (k_q r)^2] \hat{r}_i \hat{r}_j \right\}, \\
\end{array} \right.
\]

\[ d = 3 \tag{8.29} \]

where \( \hat{r} \equiv r/|r| \) is a unit vector directed to \( r \), and \( \mathcal{H}^{(1)}_{\nu}(x) \) is the Hankel function of the first kind of order \( \nu \). The Fourier transform of Eq. (8.28) is particularly simple and concise:

\[ \tilde{G}^{(q)}_{ij}(k) = \frac{1}{\varepsilon_q} \frac{k_q^2 \delta_{ij} - k_i k_j}{k^2 - k_q^2}. \tag{8.31} \]

Note that Eq. (8.31) is independent of the shape of the exclusion region, which stands in contrast to the shape-dependent Fourier transform of \( H^{(q)}_{ij}(r) \); see SM for details.

Use of (8.25) and (8.27) leads to an integral equation for the generalized cavity
Intensity field $\mathbf{F}(\mathbf{x})$:

$$
\mathbf{F}(\mathbf{x}) = \mathbf{E}_0(\mathbf{x}) + \int_\epsilon \mathbf{H}^{(q)}(\mathbf{x} - \mathbf{x}') \cdot \mathbf{P}(\mathbf{x}') \, d\mathbf{x}',
$$

(8.32)

where the integral subscript $\epsilon$ indicates that the integral is to be carried out by omitting the exclusion region and then allowing it to uniformly shrink to zero. Here $\mathbf{F}(\mathbf{x})$ is related directly to $\mathbf{E}(\mathbf{x})$ via

$$
\mathbf{F}(\mathbf{x}) = \left\{ \mathbf{I} + D^{(q)}[\varepsilon(\mathbf{x}) - \varepsilon_q] \right\} \cdot \mathbf{E}(\mathbf{x}).
$$

(8.33)

Using the definitions (8.25) and (8.33), we obtain a linear constitutive relation between $\mathbf{P}(\mathbf{x})$ and $\mathbf{F}(\mathbf{x})$:

$$
\mathbf{P}(\mathbf{x}) = L^{(q)}(\mathbf{x}) \cdot \mathbf{F}(\mathbf{x}),
$$

(8.34)

where

$$
L^{(q)}(\mathbf{x}) = [\varepsilon(\mathbf{x}) - \varepsilon_q] \cdot \left\{ \mathbf{I} + D^{(q)}[\varepsilon(\mathbf{x}) - \varepsilon_q] \right\}^{-1}.
$$

(8.35)

It is noteworthy that one is free to choose any convenient exclusion-region shape, provided that its boundary is sufficiently smooth. The choice of the exclusion-region shape is crucially important because it determines the type of modified electric field that results as well as the corresponding expansion parameter in the series expansion for the effective dynamic dielectric constant tensor. For example, when a spheroidal-shaped exclusion region at a position $\mathbf{x}$ in $\mathbb{R}^d$ is chosen to be aligned with the polarization vector $\mathbf{P}(\mathbf{x})$ at that position, we have

$$
D^{(q)} = \frac{A^*}{\varepsilon_q} \mathbf{I},
$$

(8.36)

where $\mathbf{I}$ is the second-rank identity tensor and $A^* \in [0, 1]$ is the depolarization factor for a spheroid [296] with the aforementioned alignment. In the special cases of a
sphere, disk-like limit, and needle-like limit, $A^* = 1/d, 1, 0$, respectively. Thus, for these three cases, Eq. (8.35) yields the following shape-dependent tensor:

$$L^{(q)}(x; A^*) = T^{(p)}(x) I \times \begin{cases} d\varepsilon_q\beta_{pq}, & A^* = 1/d \text{ (spherical)} \\ \varepsilon_q(1 - \varepsilon_q/\varepsilon_p), & A^* = 1 \text{ (disk-like)} \\ \varepsilon_q(\varepsilon_p/\varepsilon_q - 1), & A^* = 0 \text{ (needle-like)} \end{cases}, \quad (8.37)$$

where $\beta_{pq}$ is the dielectric polarizability defined by

$$\beta_{pq} \equiv \frac{\varepsilon_p - \varepsilon_q}{\varepsilon_p + (d - 1)\varepsilon_q}. \quad (8.38)$$

Moreover, in these three cases, the generalized cavity intensity field $F(x; A^*)$ reduces to

$$F(x; A^*) \rightarrow \begin{cases} E(x) + \frac{P(x)}{d\varepsilon_q}, & A^* = 1/d \text{ (spherical)} \\ \frac{D(x)}{\varepsilon_q}, & A^* = 1 \text{ (disk-like)} \\ E(x), & A^* = 0 \text{ (needle-like)} \end{cases}, \quad (8.39)$$

respectively, where $D(x)$ is the displacement field. Importantly, among these three cases, the original and modified strong-contrast expansions arise only when the exclusion region is taken to be a sphere, as will be elaborated below.

We will now show that our formalism yields an exact relation between the polarization field $P(x)$ and the applied field $E_0(x)$ that is nonlocal in space. It is more convenient at this stage to utilize a compact linear operator notation, which enables us to express the integral equation (8.32) as

$$F = E_0 + HP, \quad (8.40)$$

where have temporarily dropped the superscript $q$. Combination of this equation with
(8.34) yields the following integral equation for the polarization field

\[ \mathbf{P} = L\mathbf{E}_0 + L\mathbf{H}\mathbf{P}. \quad (8.41) \]

The desired nonlocal relation is obtained from (8.41) by successive substitutions:

\[ \mathbf{P} = S\mathbf{E}_0. \quad (8.42) \]

where

\[ S = [I - LH]^{-1}L. \quad (8.43) \]

More explicitly, the nonlocal relation (8.42) can be expressed as

\[ \mathbf{P}(1) = \int d^2 \mathbf{r} \mathbf{S}(1, 2) \cdot \mathbf{E}_0(2), \quad (8.44) \]

where boldface numbers 1, 2 are short-hand notations for position vectors \( r_1, r_2 \). Ensemble averaging (8.44) and invoking statistical homogeneity yields the convolution relation

\[ \langle \mathbf{P} \rangle (1) = \int d^2 \mathbf{r} \langle \mathbf{S} \rangle (1 - 2) \cdot \mathbf{E}_0(2), \quad (8.45) \]

where the operator \( \mathbf{S} \) depends on relative positions, i.e., \( \langle \mathbf{S} \rangle (1, 2) = \langle \mathbf{S} \rangle (1 - 2) \), and angular brackets denote an ensemble average. Formally, the nonlocal relation (8.45) is the same as the one given in Torquato [296] for the static problem, but nonlocality was not explicitly invoked there. Taking the Fourier transform of (8.45) yields a compact Fourier representation of this nonlocal relation, namely,

\[ \langle \tilde{\mathbf{P}} \rangle (\mathbf{k}) = \langle \tilde{\mathbf{S}} \rangle (\mathbf{k}) \cdot \tilde{\mathbf{E}}_0(\mathbf{k}), \quad (8.46) \]

where \( \langle f \rangle (\mathbf{k}) \equiv \int \langle f \rangle (\mathbf{x}) \exp(-i\mathbf{k} \cdot \mathbf{x}) d\mathbf{x} \). From Eq. (8.23), \( \tilde{\mathbf{E}}_0(\mathbf{k}) = \tilde{\mathbf{E}}_0 \delta(\mathbf{k} - \mathbf{k}_q) \), implying that the wavevector \( \mathbf{k} \) in Eq. (8.46) must be identical to \( \mathbf{k}_q \).
As in the static case [264, 296] and quasistatic regime [242], the ensemble-averaged operator $\langle S \rangle (r)$, which is given explicitly in Appendix 8.15 in terms of the $n$-point correlation functions and products of the tensor $H(r)$, depends on the shape of the macroscopic ellipsoidal composite specimen (see Fig. 8.3). This is due to the fact that $H$ decays like $r^{-d}$ for large $r$ and hence $\langle S \rangle (r)$ involves conditionally convergent integrals [296]. To avoid such conditional convergence issues, we follow previous strong-contrast formulations by seeking to eliminate the applied field $E_0$ in (8.45) in favor of the average cavity field $\langle F \rangle (r)$ in order to get a corresponding nonlocal homogenized constitutive relation between $\langle P \rangle (r)$ and $\langle F \rangle (r)$ or vice versa. Thus, solving for $E_0$ in (8.45) and substituting into the ensemble average of (8.40) yields

$$\langle F \rangle = [S^{-1} + H] \langle P \rangle.$$  \hfill (8.47)

Inverting this expression leads to the following nonlocal constitutive relation:

$$\langle P \rangle (1) = \int d^2 L_e^{(q)}(1 - 2) \cdot \langle F \rangle (2),$$  \hfill (8.48)

where $L_e^{(q)}(r)$ is a kernel that is derived immediately below and explicitly given by

$$L_e^{(q)}(r) \equiv \int dr' [\varepsilon_e(r') - \varepsilon_q I \delta(r')] \cdot \left[ I \delta(r - r') + D^{(q)} \cdot (\varepsilon_e(r - r') - \varepsilon_q I \delta(r - r')) \right]^{-1}.$$  \hfill (8.49)

We are not aware of any previous work that derives such an exact nonlocal homogenized constitutive relation (8.48) from first principles.

Note that the support $\ell_s$ of the kernel $L_e^{(q)}(r)$ relative to the incident wavelength $\lambda$ determines the degree of spatial dispersion. When $\lambda$ is finite, the relation between $\langle P \rangle (x)$ and $\langle F \rangle (x)$ in (8.48) is nonlocal in space. In the regime $\ell_s \ll \lambda$, the nonlocal relation (8.48) can be well approximated by the local relation...
\[ \langle P \rangle (x) \approx \left[ \int L_e^{(q)}(x') \, dx' \right] \cdot \langle F \rangle (x) . \] Indeed, in the static limit, \( L_e^{(q)}(r) \) tends to a Dirac delta function \( \delta(r) \), expression (8.48) becomes the \textit{position-independent} local relation

\[ \langle P \rangle = L_e^{(q)} \cdot \langle F \rangle \] (8.50)

derived earlier [296].

The nonlocal constitutive relation in direct space, Eq. (8.48), can be reduced to a linear product form in Fourier space by taking the Fourier transform of (8.48):

\[ \tilde{\langle P \rangle} (k_q) = L_e^{(q)}(k_q) \cdot \langle \tilde{F} \rangle (k_q) . \] (8.51)

The wavevector-dependent effective tensor \( L_e^{(q)}(k_q) \) is postulated (see discussion in Appendix 8.15.2) to be given by

\[ L_e^{(q)}(k_q) \equiv [\varepsilon_e(k_q) - \varepsilon_q I] \cdot \left\{ I + D^{(q)} : [\varepsilon_e(k_q) - \varepsilon_q I] \right\}^{-1} . \] (8.52)

This linear fractional form for \( L_e^{(q)}(k) \) is consistent with the one derived for the static limit [296] and for the quasistatic regime [242]. Taking the inverse Fourier transform of (8.52) yields its corresponding direct-space representation (8.49). Taking the Fourier transform of (8.47) yields

\[ \langle \tilde{F} \rangle (k_q) = \left[ \langle \tilde{S} \rangle (k_q)^{-1} + \tilde{H}(k_q) \right] \cdot \langle \tilde{P} \rangle (k_q) . \] (8.53)

Comparing (8.51) to (8.53), and specifically choosing a spherical exclusion-region, as discussed in Eq. (8.37), yields the desired exact strong-contrast expansions for general macroscopically anisotropic two-phase media:

\[ \phi_p^2 \beta_{pq}^2 [\varepsilon_e(k_q) + (d - 1)\varepsilon_q I] \cdot [\varepsilon_e(k_q) - \varepsilon_q I]^{-1} = \phi_p \beta_{pq} I - \sum_{n=2}^{\infty} A_n^{(p)}(k_q) \beta_{pq}^n , \] (8.54)

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where $A_n^{(p)}(k_q)$ is a wavevector-dependent second-rank tensor field that is a functional involving the set of correlation functions $S_1^{(p)}, S_2^{(p)}, \cdots, S_n^{(p)}$ (defined in Sec. 8.2.1) and products of the second-rank tensor field $H^{(q)}(r)$; see also Eq. (8.29). Specifically, for $n = 2$ and $n \geq 3$, these $n$-point tensors associated with the polarized phase $p$ are respectively given by

$$A_2^{(p)}(k_q) = d\varepsilon_q \int d\mathbf{r} \ H^{(q)}(r) \ e^{-i k_q \cdot r} \chi_\nu(r), \quad (8.55)$$

$$A_n^{(p)}(k_q) = d\varepsilon_q \left( \frac{-d\varepsilon_q}{\phi_p} \right)^{n-2} \int d\mathbf{r}_1 \cdots d\mathbf{r}_{n-1} \ H^{(q)}(r_1 - r_2) \ e^{-i k_q \cdot (r_1 - r_2)} \ H^{(q)}(r_2 - r_3) \ e^{-i k_q \cdot (r_2 - r_3)} \cdots H^{(q)}(r_{n-1} - r_n) \ e^{-i k_q \cdot (r_{n-1} - r_n)} \Delta_n^{(p)}(r_1, \cdots, r_n), \quad (8.56)$$

where $\int d\mathbf{r} \equiv \lim_{\varepsilon \to 0^+} \int_{|r| > \varepsilon} d\mathbf{r}$ and $\Delta_n^{(p)}$ is a position-dependent determinant involving correlation functions of the polarized phase $p$ up to the $n$-point level:

$$\Delta_n^{(p)}(r_1, \cdots, r_n) = \begin{vmatrix} S_2^{(p)}(r_1, r_2) & S_1^{(p)}(r_1) & \cdots & 0 \\ S_3^{(p)}(r_1, r_2, r_3) & S_2^{(p)}(r_2, r_3) & \cdots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ S_n^{(p)}(r_1, \cdots, r_n) & S_n^{(p)}(r_2, \cdots, r_n) & \cdots & S_2^{(p)}(r_{n-1}, r_n) \end{vmatrix}. \quad (8.57)$$

For macroscopically isotropic media, the effective dielectric tensor is isotropic, i.e., $\varepsilon_e(k_q) = \varepsilon_e(k_q) I$. The corresponding strong-contrast expansion for $\varepsilon_e(k_q)$ is obtained by taking trace of both sides of Eq. (8.54):

$$\phi_p^2 \beta_p^2 \varepsilon_e(k_q) + (d - 1) \varepsilon_q [\varepsilon_e(k_q) - \varepsilon_q]^{-1} = \phi_p \beta_p - \sum_{n=2}^\infty A_n^{(p)}(k_q) \beta_p^n, \quad (8.58)$$

where $\varepsilon_e(k_q) = \text{Tr}[\varepsilon_e(k_q)]/d; A_n^{(p)}(k_q) = \text{Tr}[A_n^{(p)}(k_q)]/d$ for $n \geq 2$ and $\text{Tr}[\ ]$ denotes the trace operation. Furthermore, for statistically isotropic media, the effective dielectric constant becomes independent of the direction of the wavevector, i.e., $\varepsilon_e(k_q) = \varepsilon_e(k_q)$. 

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Remarks:

1. Importantly, the strong-contrast expansion (8.54) is a series representation of a linear fractional transformation of the variable $\varepsilon_c(k_q)$ (left-hand side of the equation), rather than the effective dielectric constant tensor itself. The series expansion in powers of the polarizability $\beta_{pq}$ of this particular rational function of $\varepsilon_c(k_q)$ has important consequences for the predictive power of approximations derived from the expansion, as detailed in Sec. 8.4.2.

2. The fact that the exact expansion (8.54), extracted from our nonlocal relation (8.51), is explicitly given in terms of integrals over products of the relevant Green’s functions and the $n$-point correlation functions to infinite order, implies that multiple scattering to all orders is exactly treated for the range of wavenumbers for which our extended homogenization theory applies, i.e., $0 \leq |k_q| \ell \lesssim 1$.

3. Note that Eq. (8.54) represents two different series expansions: one for $q = 1$ and $p = 2$ and the other for $q = 2$ and $p = 1$.

4. The exact expansions represented by (8.54) are independent of the reference phase $q$ and hence independent of the wavevector $k_q$.

5. For $d = 2$, the strong-contrast expansion applies for TE polarization only. This implies that the electric field and wavevector are parallel to the plane or transverse to an axis of symmetry in a 3D system whose cross-sections are identical.

6. Formally, the original strong-contrast expansions that apply in the quasistatic regime [242] can be obtained from the nonlocal strong-contrast expansions (8.54) by simply replacing the exponential functions that appear in the expressions for the second-rank tensors $A^{(p)}_n(k_q)$, defined by (8.56), by unity.

7. For statistically isotropic media, the effective phase speed $c_e(k_q)$ and attenuation
coefficient $\gamma_e(k_q)$ are determined by the scalar effective dielectric constant $\varepsilon_e(k_q)$:

$$
\frac{c_e(k_q)}{c} = n_e(k_q)^{-1} = \text{Re} \left[ \frac{1}{\sqrt{\varepsilon_e(k_q)}} \right],
$$

(8.59)

$$
\frac{\gamma_e(k_q)}{c} = \kappa_e(k_q)^{-1} = \text{Im} \left[ \frac{1}{\sqrt{\varepsilon_e(k_q)}} \right],
$$

(8.60)

where $n_e(k_q)$ and $\kappa_e(k_q)$ are the effective refractive and extinction indices, respectively. The quantity $\exp(-2\pi\gamma_e/c_e)$ is the factor by which the incident-wave amplitude is attenuated for a period $2\pi/\omega$.

### 8.4.2 Convergence Properties and Accuracy of Truncated Series

The form of the strong-contrast expansion parameter $\beta_{pq}$ in (8.54) is a direct consequence of the choice of a spherical region excluded from the volume integrals in (8.54) due to singularities in the Green’s functions [242]. It is bounded by

$$
-\frac{1}{d-1} \leq \beta_{pq} \equiv \frac{\varepsilon_p - \varepsilon_q}{\varepsilon_p + (d-1)\varepsilon_q} \leq 1,
$$

which implies that the strong-contrast expansion (8.54) can converge rapidly, even for infinite contrast ratio $\varepsilon_p/\varepsilon_q \to \infty$. Other choices for the shape of the exclusion region will lead to different expansion parameters that will generally be bounded but can lead to expansions with significantly different convergence properties [296]. In Appendix 8.11, we present the corresponding expansions for disk-like and needle-like exclusion regions, which are exceptional cases that lead to slowly converging weak-contrast expansions with expansion parameter $(\varepsilon_p - \varepsilon_q)/\varepsilon_q$, and thus is unbounded when $\varepsilon_p/\varepsilon_q \to \infty$.

Importantly, in the purely static case, the expansion (8.54) becomes identical to one derived by Sen and Torquato [264] and its truncation after second-order terms
Figure 8.4: Schematic of the optimal multiscale “coated-spheres” model that realizes the isotropic Hashin-Shtrikman bounds on $\varepsilon_e$ [115]. Each composite sphere is composed of a spherical inclusion of one phase (dispersed phase) that is surrounded by a concentric spherical shell of the other phase such that the fraction of space occupied by the dispersed phase is equal to its overall phase volume fraction. The composite spheres fill all space, implying that their sizes range down to the infinitesimally small. When phase 2 is the disconnected inclusion (dispersed) phase, this two-phase medium minimizes and maximizes the effective static dielectric constant $\varepsilon_e$ for prescribed volume fraction and contrast ratio, when $\varepsilon_2/\varepsilon_1 > 1$ and $\varepsilon_2/\varepsilon_1 < 1$, respectively. It has recently been proved that these highly degenerate optimal Hashin-Shtrikman multiscale distributions of spheres are hyperuniform [157, 156].

[i.e., setting $A_n^{(p)} = 0$ for all $n \geq 3$] yields the generalized Hashin-Shtrikman bounds [115] derived by Willis [333] that are optimal since they are realized by certain statistically anisotropic composites in which there is a disconnected, dispersed phase in a connected matrix phase [207]. In the case of an isotropic effective dielectric constant $\varepsilon_e$, the optimal Hashin-Shtrikman upper and lower bounds for any phase-contrast ratio $\varepsilon_2/\varepsilon_1$ are exactly realized by the multiscale “coated-spheres” model, which is depicted in Fig. 8.4 in two dimensions. Affine transformations of the coated spheres in the $d$ orthogonal directions lead to oriented coated ellipsoids that are op-
timal for the macroscopically anisotropic case. The lower bound corresponds to the case when the high-dielectric-constant phase is the dispersed, disconnected phase and the upper bound corresponds to the instance in which the high-dielectric constant phase is the connected matrix. Thus, Torquato [289, 296] observed that the strong-contrast expansions (8.54) in the static limit can be regarded to be ones that perturb around such optimal composites, implying that the first few terms of the expansion can yield accurate approximations of the effective property for a class of particulate composites as well as more general microstructures, depending on whether the high-dielectric phase percolates or not. For example, even when $\varepsilon_2/\varepsilon_1 \gg 1$, the dispersed phase 2 can consist of identical or polydisperse particles of general shape (ellipsoids, cubes, cylinders, polyhedra) with prescribed orientations that may or not overlap, provided that the particles are prevented from forming large clusters compared to the specimen size. Moreover, when $\varepsilon_2/\varepsilon_1 \ll 1$, the matrix phase can be a cellular network [303]. Finally, for moderate values of the contrast ratio $\varepsilon_2/\varepsilon_1$, even more general microstructures (e.g., those without well-defined inclusions) can be accurately be treated. Importantly, we show that for the dynamic problem under consideration, the first few terms of the expansion (8.54) yield accurate approximations of $\epsilon_e(k_q)$ for a similar wide class of two-phase media (see Sec. 8.8). Analogous approximations were derived and applied for the quasistatic regime [242, 46].

We now show how lower-order truncations of the series (8.54) can well approximate higher-order functionals (i.e., higher-order diagrams) of the exact series to all orders in terms of lower-order diagrams. Such truncations are tantamount to approximate but resummations of the strong expansions, which enables multiple scattering and spatial dispersion effects to be accurately captured to all orders. Solving the left-hand side of (8.54) for $\epsilon_e$ yields the rational function in $\beta_{pq}$:

$$
\frac{\epsilon_e(k_q)}{\epsilon_q} = I + d\phi_p^2 \beta_{pq} \left[ \phi_p (1 - \phi_p \beta_{pq}) I - \sum_{n=2}^{\infty} A_{pq}^{(p)} (k_q) \beta_{pq}^{n-1} \right]^{-1}.
$$

(8.61)
Expanding (8.61) in powers of the scalar polarizability $\beta_{pq}$ yields the series

$$
\frac{\varepsilon_e(k_q)}{\varepsilon_q} = \sum_{n=0}^{\infty} B_n^{(p)}(k_q) \beta_{pq}^n,
$$

(8.62)

where the first several functionals $B_n^{(p)}(k_q)$ are explicitly given in terms of $A_0^{(p)}$, $A_1^{(p)}$, ..., $A_n^{(p)}$ as

$$
B_0^{(p)}(k_q) = I \\
B_1^{(p)}(k_q) = d\phi_p I \\
B_2^{(p)}(k_q) = d\left[ A_2^{(p)}(k_q) + \phi_p^2 I \right] \\
B_3^{(p)}(k_q) = \frac{d}{\phi_p} \left[ A_2^{(p)}(k_q)^2 + \phi_p A_3^{(p)}(k_q) + 2\phi_p^2 A_2^{(p)}(k_q) + \phi_p^4 I \right] \\
B_4^{(p)}(k_q) = \frac{d}{\phi_p^2} \left[ A_2^{(p)}(k_q)^3 + 2\phi_p A_2^{(p)}(k_q) \cdot A_3^{(p)}(k_q) + 3\phi_p^2 A_2^{(p)}(k_q)^2 + \phi_p^4 A_4^{(p)}(k_q) \\
+ 2\phi_p^3 A_3^{(p)}(k_q) + 3\phi_p^4 A_2^{(p)}(k_q) + \phi_p^6 I \right],
$$

where $T^n$ stands for $n$ successive inner products of a second-rank tensor $T$.

Let us now compare the exact expansion (8.62) to the one that results when expanding the truncation of the exact expression (8.54) for $[\varepsilon_e + (d-1)\varepsilon_q][\varepsilon_e - \varepsilon_q]^{-1}$ at the 2-point level:

$$
\frac{\varepsilon_e(k_q)}{\varepsilon_q} \approx I + d\phi_p \beta_{pq} \left[ (1 - \phi_p \beta_{pq}) I - A_2^{(p)}(k_q) \beta_{pq} / \phi_p \right]^{-1} \\
= \sum_{n=0}^{\infty} C_n^{(p)}(k_q) \beta_{pq}^n,
$$

(8.63)

(8.64)

where the $n$th-order functional $C_n^{(p)}(k_q)$ for any $n$ is given in terms of the volume fraction $\phi_p$ and $A_2^{(p)}(k_q)$, which has the following diagrammatic representation:

$$
A_2^{(p)}(k_q) = \begin{array}{c}
\text{Diagram}
\end{array}
$$

(8.65)
Here the solid and wavy lines joining two nodes represent the spatial correlation via \( \chi_V(r) \) and a wavevector-dependent Green’s function \( H^{(q)}(r) e^{-ik_q \cdot r} \) between the nodes, respectively. The black node indicates a volume integral and carries a factor of \( d\varepsilon_q \). The first several functionals \( C_n^{(p)}(k_q) \) are explicitly given as

\[
C_0^{(p)}(k_q) = I,
C_1^{(p)}(k_q) = d\phi_p I,
C_2^{(p)}(k_q) = d\left[A_2^{(p)}(k_q) + \phi_p^2 I\right],
C_3^{(p)}(k_q) = \frac{d}{\phi_p} \left[A_2^{(p)}(k_q) + \phi_p^2 I\right]^2,
C_4^{(p)}(k_q) = \frac{d}{\phi_p^2} \left[A_2^{(p)}(k_q) + \phi_p^2 I\right]^3.
\]

Thus, comparing (8.62) to (8.64), we see that truncation of the expansion (8.54) for \( [\varepsilon_e + (d-1)\varepsilon_q] \cdot (\varepsilon_e - \varepsilon_q)^{-1} \) at the two-point level actually translates into approximations of the higher-order functionals to all orders in terms of the first-order diagram \( \phi_p \) and the second-order diagram (8.65). This can be thought of as an approximate but accurate resummed representation of the exact expansion (8.62). Note that the approximate expansion (8.64) is exact through second order in \( \beta_{pq} \). Clearly, truncation of (8.54) at the three-point level (see Appendix 8.13) will yield even better approximations of the higher-order functionals.

### 8.5 Strong-Contrast Approximation Formulas

Here we describe lower-order truncations of the strong-contrast expansions that are expected to yield accurate closed-form formulas for \( \varepsilon_e(k_q) \) that apply over a broad range of wavelengths \( (k_q\ell \lesssim 1) \), volume fractions and contrast ratios for a wide class of microstructures.
8.5.1 Macroscopically Anisotropic Media

For the ensuing treatment, it is convenient to rewrite the expansions (8.54), valid for macroscopically anisotropic media in $\mathbb{R}^d$, in the following manner:

$$\phi_p^2 \beta_{pq}^2 \frac{\varepsilon_e(k_q)}{\varepsilon_e(k_q) - \varepsilon_q I} = \phi_p \beta_{pq} I - \sum_{n=2}^{M} A_n^{(p)}(k_q) \beta_{pq}^n + \mathcal{R}_M(k_q),$$  \hspace{1cm} (8.66)

where the $M$th-order remainder term is defined as

$$\mathcal{R}_M(k_q) \equiv \sum_{n=M+1}^{\infty} A_n^{(p)}(k_q) \beta_{pq}^n. \hspace{1cm} (8.67)$$

Truncating the exact nonlocal expansion (8.66) at the two- and three-point levels, i.e., setting $\mathcal{R}_2(k_q) = 0$ and $\mathcal{R}_3(k_q) = 0$, respectively, yields

$$\phi_p^2 \beta_{pq}^2 \frac{\varepsilon_e(k_q)}{\varepsilon_e(k_q) - \varepsilon_q I} = \phi_p \beta_{pq} I - A_2^{(p)}(k_q) \beta_{pq}^2,$$  \hspace{1cm} (8.68)

$$\phi_p^2 \beta_{pq}^2 \frac{\varepsilon_e(k_q)}{\varepsilon_e(k_q) - \varepsilon_q I} = \phi_p \beta_{pq} I - \left[A_2^{(p)}(k_q) \beta_{pq}^2 + A_3^{(p)}(k_q) \beta_{pq}^3\right],$$  \hspace{1cm} (8.69)

Compared to the quasistatic approximation [242], these nonlocal approximations substantially extend the range of applicable wavenumber, namely, $0 \leq |k_q| \ell \lesssim 1$.

8.5.2 Macroscopically Isotropic Media

All of the applications considered in this chapter, will focus on the case of macroscopically isotropic media, i.e., they are described by the scalar effective dielectric constant $\varepsilon_e(k_q) = \text{Tr}[\varepsilon_e(k_q)]/d$ but depends on the direction of the wavevector $k_q$. 
Strong-Contrast Approximation at the Two-Point Level

Solving Eq. (8.58) for the effective dielectric constant $\varepsilon_e(k_q)$ yields the strong-contrast approximation for macroscopically isotropic media:

$$\frac{\varepsilon_e(k_q)}{\varepsilon_q} = 1 + \frac{d\beta_{pq}\phi_p^2}{\phi_p(1 - \beta_{pq}\phi_p) - \beta_{pq} A_2^{(p)}(k_q)}$$

$$= 1 + \frac{d\beta_{pq}\phi_p^2}{\phi_p(1 - \beta_{pq}\phi_p) + \frac{(d-1)d\beta_{pq}}{2\pi^2d(d/2)} F(k_q)},$$

(8.70)

where $\beta_{pq}$ is defined in Eq. (8.38), $A_2^{(p)}(k_q) \equiv \text{Tr} \left[ A_2^{(p)}(k_q) \right]/d$, and $F(Q)$ is what we call the nonlocal attenuation function of a composite for reasons described below. The direct- and Fourier-space representations of $F(Q)$ are given as

$$F(Q) \equiv -\frac{2^{d/2} \Gamma(d/2)}{\pi} Q^2 \int_{\epsilon} \frac{i}{4} \left( \frac{Q}{2\pi r} \right)^{d/2-1} \mathcal{H}_{d/2-1}(Qr) e^{-iQ \cdot r} \chi_V(r) \, dr$$

(8.71)

$$= -\frac{\Gamma(d/2)}{2^{d/2}\pi^{d+1}} Q^2 \int \frac{\tilde{\chi}_V(q)}{|q + Q|^2 - Q^2} \, dq.$$  

(8.72)

The exponential $\exp(-iQ \cdot r)$ in Eq. (8.71) arises from the phase difference associated with the incident waves at positions separated by $r$. In the quasistatic regime, this phase factor is negligible, and Eq. (8.71) reduces to the local attenuation function $F(Q)$ (derived in Ref. [242] and summarized in Appendix 8.14.2) because it is barely different from unity over the correlation length associated with the autocovariance function $\chi_V(r)$. The strong-contrast approximation (8.70) was postulated in Ref. [158] on physical grounds. By contrast, this chapter derives it as a consequence of our exact nonlocal formalism (Sec. 8.4).

For statistically isotropic media, the effective dielectric constant as well as the attenuation function are independent of the direction of the incident wavevector $k_q$, and thus they can be considered as functions of wavenumber, i.e., $\varepsilon_e(k_q) = \varepsilon_e(k_q)$ and $F(k_q) = F(k_q)$. Then, the real and imaginary parts of Eq. (8.71) can be simplified.
as

\[
\text{Im}[F(Q)] = \begin{cases} 
-\frac{Q^2}{\pi} \int_0^{\pi/2} \tilde{\chi}_V(2Q \cos \phi) \, d\phi, & d = 2 \\
-\frac{Q}{2(2\pi)^{3/2}} \int_0^{2Q} q \tilde{\chi}_V(q) \, dq, & d = 3
\end{cases} 
\tag{8.73}
\]

\[
\text{Re}[F(Q)] = -\frac{2Q^2}{\pi} \text{p.v.} \int_0^\infty dq \frac{1}{q(Q^2 - q^2)} \text{Im}[F(q)], 
\tag{8.74}
\]

where Eq. (8.74) is valid for \( d = 2, 3 \). Following conventional usage, we say that a composite attenuates waves at a given wavenumber if the imaginary part of the effective dielectric constant is positive. Recall that attenuation in the present study occurs only because of multiple-scattering effects (not absorption). While it is the imaginary part of \( F(Q) \) that determines directly the degree of attenuation or, equivalently, \( \text{Im}[\varepsilon_e] \), we see from (8.74) that the real part of \( F(Q) \) is directly related to its imaginary part. It is for this reason that we refer to the complex function \( F(Q) \) as the (nonlocal) attenuation function.

**Modified Strong-Contrast Approximation at the Two-Point Level**

Here we extend the validity of the strong-contrast approximation (8.70) so that it is accurate at larger wavenumbers and hence better captures spatial dispersion. This is done by an appropriate rescaling of the wavenumber in the reference phase, \( k_q \), which we show is tantamount to approximately accounting for higher-order contributions in the remainder term \( R_2(k_q) \). Given that the strong-contrast expansion for isotropic media perturbs around the Hashin-Shtrikman structures (see Fig. 8.4) in the static limit, it is natural to use the scaling \( \sqrt{\varepsilon_{HS}/\varepsilon_q} k_q \), where \( \varepsilon_{HS} \) is the Hashin-Shtrikman estimate, i.e.,

\[
\varepsilon_{HS} \equiv \varepsilon_q \left[ 1 + \frac{d\phi_p \beta_{pq}}{1 - \phi_p \beta_{pq}} \right], 
\tag{8.75}
\]

which gives the Hashin-Shtrikman lower bound and upper bound if \( \varepsilon_p > \varepsilon_q \) and \( \varepsilon_p < \varepsilon_q \), respectively. This yields the following scaled strong-contrast approximation
for statistically isotropic media:

$$\frac{\varepsilon_e(k_q)}{\varepsilon_q} = 1 + \frac{d\beta_{pq}\phi_p^2}{\phi_p(1 - \beta_{pq}\phi_p)} + \frac{(d-1)\pi\beta_{pq}}{2^{d/2}\Gamma(d/2)} F\left(\sqrt{\frac{\varepsilon_{HS}k_q}{\varepsilon_q}}\right). \quad (8.76)$$

We now show that the scaled approximation (8.76) indeed provides good estimates of leading-order corrections of \( R_2(k_q) \) in powers of \( k_q \). To do so, we employ the concept of the averaged (effective) Green function \( \langle G(q)\rangle \) of an inhomogeneous medium which in principle accounts for the all multiple scattering events

$$\langle G^{(q)}(q)\rangle = \left(\frac{\omega}{c}\right)^2 \left\{ \left[ q^2 - k_e(\omega)^2 \right] I - qq \right\}^{-1}, \quad (8.77)$$

where \( k_e(\omega) \equiv \sqrt{\varepsilon_e(\omega)\omega/c} = \sqrt{\varepsilon_e(\omega)/\varepsilon_q}k_q \) is the effective wavenumber at a frequency \( \omega \) and \( \varepsilon_e(\omega) \) is the exact effective dynamic dielectric constant, assuming a well-defined homogenization description [266, 40]. Since exact complete microstructural information is, in principle, accounted for with the effective Green’s function (8.77), the exact strong-contrast expansion can be approximately equated to the one truncated at the two-point level with an attenuation function given in terms of the effective Green function, i.e.,

$$\phi_p^2 \beta_{pq}^2 \frac{\varepsilon_e(k_q)}{\varepsilon_q} + (d - 1)\varepsilon_q = \phi_p\beta_{pq} - A_2^{(p)}(k_q) \beta_{pq}^2 + R_2(k_q), \quad (8.78)$$

$$\approx \phi_p\beta_{pq} - A_2^{(p)}(k_e(\omega)) \beta_{pq}^2. \quad (8.79)$$

When the functional form of \( A_2^{(p)}(Q) \) or, equivalently, \( F(Q) \) is available, it is possible to solve Eq. (8.79) for \( \varepsilon_e(k_q) \) in a self-consistent manner. Instead, we show that by assuming \( k_e(\omega) \approx \sqrt{\varepsilon_{HS}/\varepsilon_q}k_q \), which results in Eq. (8.76), we obtain good estimates of the leading-order corrections of \( R_2(k_q) \) in powers of \( k_q \). Thus, the scaled approximation (8.76) provides better estimates of the higher-order three-point approximation (given in Appendix 8.13) than the unmodified strong-contrast approximation (8.63).
This can be easily confirmed in the quasistatic regime from the small-$k_q$ expansions of $A_2^{(p)}(k_q)$ and $A_3^{(p)}(k_q)$ given in Ref. [242]. We also confirm the improved predictive capacity of the scaled strong-contrast approximation to better capture dispersive characteristics for ordered and disordered models via comparison to FDTD simulations; see Figs. 8.7 and 8.8, and Appendix 8.20.

8.6 Results for the Nonlocal Attenuation Function

We report some general behaviors of the nonlocal attenuation function $F(Q)$ [cf. (8.71) or (8.72)] for nonhyperuniform and hyperuniform media for long and intermediate wavelengths. We also provide plots of both the real and imaginary parts of $F(Q)$ for the four models of disordered two-phase media considered in this chapter, which depends on wavenumber $Q$.

The function $F(Q)$ depends on the microstructure via the spectral density $\tilde{\chi}_V(Q)$. Thus, assuming that the latter has the power-law scaling $\tilde{\chi}_V(Q) \sim Q^\alpha$ as $Q \to 0$, the asymptotic behavior of $F(Q)$ in the long-wavelength limit ($Q \to 0$) follows as

$$\text{Im}[F(Q)] \sim \begin{cases} Q^d, & \text{nonhyperuniform} \\ Q^{d+\alpha}, & \text{hyperuniform} \end{cases}, \quad \text{as } Q \to 0, \quad (8.80)$$

$$\text{Re}[F(Q)] \sim Q^2, \quad \text{as } Q \to 0, \quad (8.81)$$

where we have used Eqs. (8.71) and (8.73). Recall that the exponent $\alpha$ lies in the open interval $(0, \infty)$ for hyperuniform systems (see Sec. 8.2.4). For nonhyperuniform systems studied here, we take $\alpha = 0$. The reader is referred to Appendix 8.16 for derivations of Eqs. (8.80) and (8.81). Importantly, in the quasistatic regime, the imaginary parts of the effective dielectric constant for both strong-contrast approximations, (8.70) and (8.76), are determined by the asymptotic behaviors $F(Q)$.
indicated in Eq. (8.80), i.e.,

\[ \text{Im}[\varepsilon_e(k_q)] \sim \text{Im}[F(k_q)] \sim \begin{cases} 
  k_q^d, & \text{nonhyperuniform} \\
  k_q^{d+\alpha}, & \text{hyperuniform}
\end{cases}, \quad \text{as } k_q \to 0. \quad (8.82) \]

Thus, hyperuniform media are less lossy than their nonhyperuniform counterparts in the quasistatic regime.

In the case of stealthy hyperuniform media [i.e., \( \bar{\chi}_V(Q) = 0 \) for \( 0 \leq Q < Q_U \)], the imaginary part of \( F(Q) \), defined by (8.73), is identically zero (transparent or lossless) for any space dimension \( d \) for a range of wavenumbers; specifically,

\[ \text{Im}[F(Q)] = 0, \quad \text{for } 0 \leq Q < Q_U/2. \quad (8.83) \]

(Appendix 8.17 describes how the local attenuation function \( F(Q) \) derived in Ref. [242] generally differs from its nonlocal counterpart.) The transparency interval in which \( \text{Im}[\varepsilon_e(k_q)] = 0 \) predicted by the two strong-contrast approximations [Eqs. (8.70) and (8.76)] are thus given by

\[ \text{Im}[\varepsilon_e(k_q)] = 0, \quad \text{for } \begin{cases} 
  0 \leq k_q < \frac{Q_U}{2}, & \text{[from Eq. (8.70)]}, \\
  0 \leq k_q < \frac{Q_U}{2(\varepsilon_{HS}/\varepsilon_q)^{1/2}}, & \text{[from Eq. (8.76)]},
\end{cases} \quad (8.84) \]

where \( \varepsilon_{HS} \) is given in Eq. (8.75). When \( \varepsilon_p > \varepsilon_q \), since \( \varepsilon_{HS} > \varepsilon_q \), the scaled approximation accurately predicts a narrower transparency interval than the unscaled variant, as verified in Sec. 8.8. Interestingly, the transparency interval obtained from the less accurate formula (8.70) agrees with the one obtained from previous simulation results for stealthy hyperuniform “point” scatterers [179], not finite-sized particles considered here.

Figure 8.5 shows \( F(Q) \) for the four distinct models of disordered particulate me-
dia in \( \mathbb{R}^3 \); overlapping spheres, equilibrium packings, stealthy hyperuniform packings, and hyperuniform polydisperse packings. (Its 2D counterpart is provided in Appendix 8.20.) We clearly see that these attenuation functions exhibit common large-\( Q \) behaviors, regardless of the microstructures. From the quasistatic to the intermediate-wavelength regimes (\( Qa < 1 \)), however, the attenuation characteristics (imaginary parts \( \text{Im}[F(Q)] \)) are considerably different from one model to another. For example, stealthy hyperuniform media are transparent up to a finite wavelength, and hyperuniform polydisperse packings exhibits much less attenuation than nonhyperuniform systems.

![Figure 8.5: The negatives of (a) the real and (b) imaginary parts of the nonlocal attenuation function \( F(Q) \), defined in Eq. (8.71), for the four models of 3D disordered composite media considered in this chapter. The inset in (b) is the log-log plot of the larger panel. The volume fraction of the dispersed phase for each model is \( \phi_2 = 0.25 \). The first three models consist of identical spheres of radius \( a \). For class I hyperuniform polydisperse particulate media, \( a \) is the mean sphere radius.](image)

### 8.7 Simulation Procedure to Compute Effective Dynamic Dielectric Constant

In Ref. [158], we established preliminary comparisons of strong-contrast approximation (8.70) and numerical simulations via the extended version of the Fast-Fourier-
Transform-based technique [217, 73]. Due to convergence issues, however, in this chapter, we employ a more reliable numerical technique, i.e., the finite-difference time-domain (FDTD) method [282], using an open-source software package [224]. We focus here on particulate media and take the matrix to be the reference medium (phase 1) and the particles to be the polarized phase (phase 2).

Figure 8.6: Schematic of the general simulation setup for either (a) periodic or (b) non-periodic consisting of $N$ spheres of radius $a$ in a matrix. In both cases, Gaussian pulses of electric fields propagate from the planar sources (shown in red lines) to the packings (shown in black circles). The wavenumber (spectrum) of the pulses spans between $\min[k_1]$ and $\max[k_1]$. Periodic boundary conditions are applied along all directions, except for the propagation direction $\hat{x}$. The perfectly matched layers (PML, shown in blue) of thickness $L_{\text{pml}}$ are placed at the both ends of the simulation box to absorb any reflected and transmitted waves.

The general simulation setup is schematically illustrated in Fig. 8.6. It is carried out in two steps:

1. Obtain the steady-state spatial distributions of electric field $E_y(r, \omega)$ and electric displacement field $D_y(r, \omega)$ at a given frequency $\omega$ (or the corresponding wavenumber $k_1$). Specifically, the planar source generates Gaussian pulses of electric fields whose wavenumber $k_1$ spans between $\min[k_1]$ and $\max[k_1]$. Using the aforementioned MEEP package [224], we compute time evolution of elec-
tric field \( E_y(r, t) \) and electric displacement field \( D_y(r, t) \) for a period of time \( 6\pi \sqrt{\varepsilon_1}/\{c \min[k_1]\} \). We then compute the temporal Fourier transforms of these fields inside packings. The values of the simulation parameters (indicated in Fig. 8.6) for the 2D and 3D ordered and disordered models studied in this article are summarized in Appendix 8.19.2.

2. Post-process \( E_y(r, \omega) \) and \( D_y(r, \omega) \) to estimate the effective dielectric constant \( \varepsilon_e(k_1, \omega) \) so that it satisfies the following equations:

\[
\varepsilon_e(k_1) = \overline{\varepsilon_e(k_1)}, \quad \varepsilon_e(k_1) = \frac{\tilde{D}_y(k_e)}{\tilde{E}_y(k_e, \omega)}, \quad k_e = \sqrt{\overline{\varepsilon_e(k_1)}} \frac{\omega}{c},
\]

(8.85)

where \( k_e \) represents the effective wavenumber at macroscopic length scales, and

\[
\tilde{D}_y(q, \omega) \equiv \frac{1}{|V|} \int_V D_y(r, \omega) e^{-iq \hat{x} \cdot r} \, dr,
\]

\[
\tilde{E}_y(q, \omega) \equiv \frac{1}{|V|} \int_V E_y(r, \omega) e^{-iq \hat{x} \cdot r} \, dr,
\]

where a chosen region \( V \) inside the composite. This task is carried by numerically finding a minimizer of \( |\varepsilon_e - \overline{\varepsilon_e}|^2 \) with an initial guess \( \overline{\varepsilon_e} = \varepsilon_{\text{HS}} \) via the Broyden-Fletcher-Goldfarb-Shanno (BFGS) nonlinear optimization algorithm [183], where \( \varepsilon_{\text{HS}} \) is the Hashin-Shtrikman estimate given by Eq. (8.75).

8.8 Comparison of Simulations of \( \varepsilon_e(k_1) \) to Various Approximations Formulas

In this section, we compare our simulations of the effective dynamic dielectric constant \( \varepsilon_e(k_1) \) for various 2D and 3D ordered and disordered model microstructures to the predictions of the strong-contrast formulas as well as to conventional approximations, such as MGA (8.16) and QCA (8.17). Most of these models provide stringent tests of
the predictive power of the approximations at finite wavenumbers because they are characterized by nontrivial spatial correlations at intermediate length scales.

### 8.8.1 2D and 3D Periodic Media

We first carry out our FDTD simulations for the effective dynamic dielectric constant $\varepsilon_e(k_1)$ of 2D and 3D periodic packings (square and simple-cubic lattice packings), which necessarily depends on the direction of the incident wave $k_1$. While these periodic packings are macroscopically isotropic, due to their cubic symmetry, they are statistically anisotropic (see SM for details). For simplicity, we only consider the case where $k_1$ is aligned with one of the minimal lattice vectors, i.e., $\Gamma$-$X$ direction in the first Brillouin zone. Such periodic models enable us to validate our simulations because $\varepsilon_e(k_1)$ also can be accurately extracted from the lowest two photonic bands that are calculated via MPB, an open-source software package [142]. The results from the band-structure calculations and our FDTD simulations show excellent agreement. In particular, our simulations accurately predict two salient dielectric characteristics that must be exhibited by periodic packings: transparency up to a finite wavenumber associated with the edge of the first Brillouin zone (i.e., Im[$\varepsilon_e$] = 0 for $0 \leq |k_1| \lesssim \pi$), and resonance-like attenuation due to Bragg diffraction within the photonic bandgap $^3$ (i.e., a peak in Im[$\varepsilon_e$] or, equivalently, a sharp transition in Re[$\varepsilon_e$] $^4$). Thus, our numerical homogenization scheme is valid down to intermediate wavelengths (see Appendix 8.19.4 for comparison of the band-structure and FDTD computations).

Importantly, while our strong-contrast approximations Eqs. (8.70) and (8.76) account for directionality of the incident waves, the MGA and QCA are independent of the direction of $k_1$. In Fig. 8.7, the FDTD simulation results are compared with

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$^3$Here, the large value of the imaginary part Im[$\varepsilon_e$] is due to a small penetration depth of evanescent waves.

$^4$The Kramers-Kronig relations (8.88) and (8.89) dictate that a resonance phenomenon in the dielectric response, that is, a sharp peak in Im[$\varepsilon_e$], must correspond to a sharp transition in Re[$\varepsilon_e$], and vice versa.
Figure 8.7: Comparison of the predictions of the strong-contrast formulas, Eqs. (8.70) and (8.76), to the Maxwell-Garnett [Eqs. (8.16) and (8.15)] and QCA (8.17) approximations for the effective dynamic dielectric constant $\epsilon_e(k_1)$ of periodic packings to our corresponding computer simulation results. We consider (a) 3D simple cubic lattice and (b) 2D square lattice of packing fraction $\phi_2 = 0.25$ and contrast ratio $\epsilon_2/\epsilon_1 = 4$. Here $k_1$ is the wavenumber in the reference (matrix) phase along the $\Gamma$-$X$ direction, and $L$ is the side length of a unit cell.

While all approximations agree with the FDTD simulations in the quasistatic regime, the MGA and QCA fail to capture properly two key features: no loss of energy up to a finite wavenumber and resonance-like attenuation in the band gaps. Each strong-contrast approximation captures both of these salient characteristics. However, it is noteworthy that the scaled strong-contrast approximation [Eq. (8.76)] agrees very well with the FDTD simulations. For contrast ratios $\epsilon_2/\epsilon_1 < 1$, FDTD simulations are also in very good agreement with the predictions of strong-contrast approximations for a wide range of wavenumbers, as detailed in Appendix 8.20.
8.8.2 Disordered Nonhyperuniform and Hyperuniform Media

To test the predictive capacity of approximation formulas for $\varepsilon_e(k_1)$ for disordered media as measured against simulations, we choose to study two distinctly different models: disordered nonhyperuniform packings (equilibrium packings) and disordered stealthy hyperuniform disordered packings [i.e., $\tilde{\chi}_V(Q) = 0$ for $0 \leq Qa < 1.5$] for both 2D and 3D. Again, we compare our simulation results to the MGAs [Eqs. (8.16) and (8.15)], QCA (8.17), strong-contrast approximation (8.70), and the scaled counterpart (8.76). The conventional approximations fail to capture spatial dispersion effects. Specifically, the MGA neglects any microstructural information, except for the particle shape, and thus cannot account for long-range correlations, such as the lossless property of stealthy hyperuniform media. By contrast, while the QCA formula yields better estimates of $\text{Im}[\varepsilon_e]$ for nonhyperuniform systems, it cannot generally capture the correct transparency characteristics of hyperuniform systems, e.g., it incorrectly predicts $\text{Im}[\varepsilon_e(k_1)] = 0$ for all wavenumbers, regardless of whether the medium is stealthy hyperuniform or nonstealthy hyperuniform; see Fig. 8.8 (b).

On the other hand, the scaled strong-contrast approximation provides excellent estimates of $\varepsilon_e(k_1)$ for both disordered models, even for large wavenumbers ($0 \leq k_1 a \leq 1$); see Fig. 8.8. Moreover, the predictions of both strong-contrast approximations accurately capture the salient microstructural differences between the nonhyperuniform and hyperuniform models because they incorporate spatial correlations at finite wavelengths via the spectral density $\tilde{\chi}_V(Q)$. For example, they properly predict that stealthy hyperuniform media are lossless up to a finite wavenumber, even if at different cut-off values; see Eq. (8.84). Corresponding 2D results are presented in Appendix 8.20 because they are qualitatively the same as the 3D results.
Figure 8.8: Comparison of the predictions of the strong-contrast formulas, Eqs. (8.70) and (8.76), to the MGA (8.15) and QCA (8.17) approximations for the effective dynamic dielectric constant $\varepsilon_e(k_1)$ of 3D disordered sphere packings to our corresponding computer simulation results. We consider (a) equilibrium packings and (b) stealthy hyperuniform packings $[\chi_V(Q) = 0$ for $0 \leq Qa < 1.5]$ of sphere radius $a$, packing fraction $\phi_2 = 0.25$, and phase contrast ratio $\varepsilon_2/\varepsilon_1 = 4$. Here $k_1$ is the wavenumber in the reference (matrix) phase, and the error bars in the FDTD simulations represent the standard errors over independent configurations.

### 8.9 Predictions of Strong-Contrast Approximations for Disordered Particulate Media

Having established the accuracy of the scaled strong-contrast approximation (8.76) for ordered and disordered media in the previous section, we now apply it to the four different disordered models discussed in Sec. 8.3 in order to study how $\varepsilon_e(k_1)$ varies with the microstructure. We first study how $\varepsilon_e(k_1)$ varies with $k_1$ at a fixed contrast ratio $\varepsilon_2/\varepsilon_1 = 10$ for the four models; see Fig. 8.9. According to Eq. (8.82), nonhyperuniform and hyperuniform media in the quasistatic regime have the different scalings, i.e., $\text{Im}[\varepsilon_e(k_1)] \sim k_1^d$ and $\text{Im}[\varepsilon_e(k_1)] \sim k_1^{d+\alpha}$, respectively, where $\alpha > 0$ for hyperuniform systems. This implies that hyperuniform media are less lossy than their nonhyperuniform counterparts as $k_1$ tends to zero, as seen in the insets of Fig. 8.9. Moreover, beyond the quasistatic regime, each model exhibits “effective” trans-
parency for a range of wavenumbers that depends on the microstructure. For 2D and 3D models, hyperuniform polydisperse packings tend to be effectively transparent for a wide range of wavenumbers compared to the nonhyperuniform ones, while the stealthy hyperuniform systems are perfectly transparent for the widest range of wavenumbers, as established in Eq. (8.84) and Sec. 8.8. For each model, the “effective” transparency spectral range must be accompanied by normal dispersion (i.e., an increase in $\text{Re}[\varepsilon_e(k_1)]$ with $k_1$) [1] because our strong-contrast approximation is consistent with the Kramers-Kronig relations (see Appendix 8.12). Moreover, we see that anomalous dispersion (i.e., a decrease in $\text{Re}[\varepsilon_e(k_1)]$ with $k_1$) occurs at wavenumbers larger but near the respective transition between the effective transparency and appreciable attenuation, which again is dictated by the Kramers-Kronig relations. The specific anomalous dispersion behavior is microstructure-dependent.

Figure 8.9: Predictions of the scaled strong-contrast approximation (8.76) for the effective dynamic dielectric constant $\varepsilon_e(k_1)$ of the four models of disordered media at volume fraction $\phi_2 = 0.25$ and contrast ratio $\varepsilon_2/\varepsilon_1 = 10$: (a) three dimensions and (b) two dimensions. The inset in the lower panel is the log-log plot of the larger panel.

We now examine how the imaginary part $\text{Im}[\varepsilon_e]$ varies with the contrast ratio $\varepsilon_2/\varepsilon_1$ for the disordered models for a given large wavenumber $k_1$ inside the transparency interval for 2D and 3D stealthy hyperuniform systems. These results are summarized in Fig. 8.10. The disparity in the attenuation characteristics across microstructures
Overlapping spheres
Equilibrium packings
Stealthy hyperuniform packings; \( Q_U a = 1.5 \)
Hyperuniform polydisperse packings
\( d=3, \phi_2 = 0.25, (\varepsilon_{HS}/\varepsilon_1)^{1/2} k_1 a = 0.7 \)

Figure 8.10: Predictions of the strong-contrast approximation (8.76) for the effective dynamic dielectric constant \( \varepsilon_e (k_1) \) of the four disordered models, as per Fig. 8.9, as function of dielectric-contrast ratio \( \varepsilon_2/\varepsilon_1 \) at packing fraction \( \phi_2 = 0.25 \) and wavenumber (a) \( \sqrt{\varepsilon_{HS}/\varepsilon_1} k_1 a = 0.7 \) in three dimensions and (b) \( \sqrt{\varepsilon_{HS}/\varepsilon_1} k_1 a = 0.6 \) in two dimensions.

widens significantly as the contrast ratio increases. Clearly, overlapping spheres are the lossiest systems. Hyperuniform polydisperse packings can be nearly as lossless as stealthy hyperuniform ones.

Stealthy hyperuniform packings, \( \varepsilon_2/\varepsilon_1 = 4 \)

\( -\gamma_e (k_1) / \varepsilon_1 \)

\( c_e (k_1) / \varepsilon_1 \)

\( \phi_2 = 0.25, Q_U a = 1.33 \)

\( \phi_2 = 0.4, Q_U a = 1.5 \)

Figure 8.11: Predictions of the scaled strong-contrast approximation (8.76) for the effective wave speed \( c_e \) and the negative of the attenuation coefficient \( \gamma_e \) as a function of \( k_1 \) for 3D stealthy hyperuniform sphere packings of contrast ratio \( \varepsilon_2/\varepsilon_1 = 4 \) at two different packing fractions: \( \phi_2 = 0.4 \) with \( Q_U a = 1.5 \) and \( \phi_2 = 0.25 \) with \( Q_U a \approx 1.33 \). The inset is a magnification of the lower panel.
We also study the effect of packing fraction $\phi_2$ on the effective phase speed $c_e(k_1)$ and effective attenuation coefficient $\gamma_e(k_1)$, as defined by Eqs. (8.59) and (8.60), respectively. For concreteness, we focus on 3D stealthy hyperuniform packings. We first generate such packings at a packing fraction $\phi_2 = 0.4$ and $Q_U a = 1.5$, as described in Sec. 8.3.4. Without changing particle positions, we then shrink particle radii to attain a packing fraction $\phi_2 = 0.25$, whose stealthy regime is now $Q_U a \approx 1.33$. The coefficients $c_e(k_1)$ and $\gamma_e(k_1)$ for these packings with $\epsilon_2/\epsilon_1 = 4$ are estimated from the scaled approximation (8.76); see Fig. 8.11. It is seen that the waves propagate significantly more slowly through the denser medium due to an increase in multiple scattering events. Moreover, the transparency intervals (wavenumber ranges where the effective attenuation coefficients vanish) is larger for the packing with the higher stealthy cut-off value $Q_U a = 1.5a (\phi_2 = 0.4)$, as predicted by Eq. (8.84).

8.10 Conclusions and Discussion

All previous closed-form homogenization estimates of the effective dynamic dielectric constant apply only at long wavelengths (quasistatic regime) and for very special macroscopically isotropic disordered composite microstructures, namely, nonoverlapping spheres or spheroids in a matrix. In this chapter, we have laid the theoretical foundation that has enabled us to substantially extend previous work in both its generality and applicability. First, we derived exact homogenized constitutive relations for the effective dynamic dielectric constant tensor $\varepsilon_e(k_q)$ that are nonlocal in space from first principles. Second, our strong-contrast representation of $\varepsilon_e(k_q)$ exactly accounts for complete microstructural information (infinite set of $n$-point correlation functions) for arbitrary microstructures and hence multiple scattering to all orders for the range of wavenumbers for which our extended homogenization theory applies, i.e., $0 \leq |k_q| \ell \lesssim 1$ (where $\ell$ is a characteristic heterogeneity length scale). Third,
we extracted from the exact expansions accurate nonlocal closed-form approximate formulas for $\varepsilon_e(k_q)$, relations (8.70) and (8.76), which are resummed representations of the exact expansions that incorporate microstructural information through the spectral density $\tilde{\chi}_V(Q)$, which is easily ascertained for general microstructures either theoretically, computationally or via scattering experiments. Depending on whether the high-dielectric phase percolates or not, the wide class of microstructures that we can treat includes particulate media consisting of identical or polydisperse particles of general shape (ellipsoids, cubes, cylinders, polyhedra) with prescribed orientations that may or not overlap, cellular networks as well as media without well-defined inclusions (Sec. 8.4.2). Our approximations account for multiple scattering across a range of wavenumbers Fourth, we carried out precise full-waveform simulations for various 2D and 3D models of ordered and disordered media to validate the accuracy of our nonlocal microstructure-dependent approximations for wavenumbers well beyond the quasistatic regime.

Having established the accuracy of the scaled strong-contrast approximation (8.76), we then applied it to four models of 2D and 3D disordered media (both nonhyperuniform and hyperuniform) to investigate the effect of microstructure on the effective wave characteristics. Among other findings, we showed that disordered hyperuniform media are generally less lossy than their nonhyperuniform counterparts. We also found that our scaled formula (8.76) accurately predicts that disordered stealthy hyperuniform media possess a transparency wavenumber interval $[0, 0.5 Q_U(\varepsilon_{HS}/\varepsilon_q)^{-1/2}) [\text{cf. (8.84)}]$, where most nonhyperuniform disordered media are opaque. Note that, using multiple-scattering simulations, Leseur, Pierrat, and Carminati [179] were the first to show that stealthy hyperuniform systems should exhibit a transparency interval, but for “point” scatterers, not finite-sized scatterers considered here. Interestingly, their transparency-interval prediction coincides with the one predicted by our less accurate strong-contrast formula [cf. (8.84)].
The accuracy of our nonlocal closed-form formulas has important practical implications, since one can now use them to accurately and efficiently predict the effective wave characteristics well beyond the quasistatic regime of a wide class of composite microstructures without having to perform computationally expensive full-blown simulations. Thus, our nonlocal formulas can be used to accelerate the discovery of novel electromagnetic composites by appropriate tailoring of the spectral densities and then constructing the corresponding microstructures by using the Fourier-space inverse methods [46]. For example, from our findings in the present study, it is clear that stealthy disordered particulate media can be employed as low-pass filters that transmit waves ‘isotropically’ up to a selected wavenumber. Moreover, using the spectral densities of the type found by Chen and Torquato [46] for stealthy hyperuniform packings (characterized by a peak value at $Q = Q_U$ with intensities that rapidly decay to zero for larger wavenumbers) and formula (8.76), one can design materials with refractive indices that abruptly change over a narrow range of wavenumbers. Of course, one could also explore the design space of effective wave properties of nonhyperuniform disordered composite media for potential applications.

### 8.11 Appendix A: Different Expansions as a Result of Different Exclusion-Region Shapes

To get a sense of how the resulting expansions change due to the choice of the exclusion-region shape, we consider the aforementioned oriented spheroidal exclusion region in the two limiting disk-like and needle-like cases. Comparing the expansion parameters in the limit cases given in Eq. (8.37) to the strong-contrast expansion with a spherical exclusion-region given in Eq. (8.58), one can obtain the counterparts of Eq. (8.58) with disk-like and needle-like exclusion-regions.

Specifically, we replace parameters in Eq. (8.58) according to the following map-
pings:

\[ \beta_{pq} \rightarrow (\varepsilon_p - \varepsilon_q)/(d\varepsilon_p), \quad \frac{\varepsilon_e(k_q) + (d-1)\varepsilon_q}{\varepsilon_e(k_q) - \varepsilon_q} \rightarrow \frac{d\varepsilon_e(k_q)}{\varepsilon_e(k_q) - \varepsilon_q}, \quad \text{disk-like,} \]

\[ \beta_{pq} \rightarrow (\varepsilon_p - \varepsilon_q)/(d\varepsilon_q), \quad \frac{\varepsilon_e(k_q) + (d-1)\varepsilon_q}{\varepsilon_e(k_q) - \varepsilon_q} \rightarrow \frac{d\varepsilon_q}{\varepsilon_e(k_q) - \varepsilon_q}, \quad \text{needle-like,} \]

resulting in the following expansions, respectively,

\[ \phi_p^2 \left( \frac{\varepsilon_p - \varepsilon_q}{d\varepsilon_p} \right)^2 \frac{d\varepsilon_e(k_q)}{\varepsilon_e(k_q) - \varepsilon_q} = \phi_p \left( \frac{\varepsilon_p - \varepsilon_q}{d\varepsilon_p} \right) - \sum_{n=2}^{\infty} A_n^{(p)}(k_q; A^* = 1) \left( \frac{\varepsilon_p - \varepsilon_q}{d\varepsilon_p} \right)^n, \quad (8.86) \]

\[ \phi_p^2 \left( \frac{\varepsilon_p - \varepsilon_q}{d\varepsilon_q} \right)^2 \frac{d\varepsilon_q}{\varepsilon_e(k_q) - \varepsilon_q} = \phi_p \left( \frac{\varepsilon_p - \varepsilon_q}{d\varepsilon_q} \right) - \sum_{n=2}^{\infty} A_n^{(p)}(k_q; A^* = 0) \left( \frac{\varepsilon_p - \varepsilon_q}{d\varepsilon_q} \right)^n. \quad (8.87) \]

Here the functionals \( A_n^{(p)}(k_q; A^*) \) are identical to Eqs. (8.55) and (8.56), except for the exclusion-region shape.

### 8.12 Appendix B: Kramers-Kronig Relations

Kramers-Kronig relations connect the real and imaginary parts of any complex function that is analytic in the upper half-plane and meets mild conditions [134, 214]. Since causality in a dielectric response function of a homogeneous material implies such analyticity properties, the Kramers-Kronig relations enable one to directly link the real part of a response function to its imaginary part or vice versa, even if the real or imaginary parts are only available in a finite frequency range [134, 214]. This implies that when a heterogeneous material can be treated as a homogeneous material with a dynamic effective dielectric constant \( \varepsilon_e(k_q) \), Kramers-Kronig relations
immediately apply to the exact strong-contrast expansion (8.66), i.e.,

\[ \text{Re}[\varepsilon_e(kq)] = \varepsilon_e(\infty) + \frac{2}{\pi} \text{p.v.} \int_0^\infty dq \frac{q \text{Im}[\varepsilon_e(q)]}{q^2 - k_q^2}, \tag{8.88} \]

\[ \text{Im}[\varepsilon_e(kq)] = -\frac{2k_q}{\pi} \text{p.v.} \int_0^\infty dq \frac{\text{Re}[\varepsilon_e(q)] - \varepsilon_e(\infty)}{q^2 - k_q^2}, \tag{8.89} \]

where we assume a linear dispersion relation in the reference phase (i.e., \( k_q = \sqrt{\varepsilon_q} \omega/c \)) and \( \lim_{\omega \to \infty} \varepsilon_e(\omega) = \varepsilon_q \) is real-valued. The Kramers-Kronig relations may or may not be obeyed when the strong-contrast expansion is truncated at the two-point level, yielding (8.70).

Here we analytically show that the effective dielectric constant \( \varepsilon_e(kq) \) for isotropic media obtained from either the unscaled or scaled strong-contrast formulas [see Eqs. (8.70) and (8.76)] also satisfies the Kramers-Kronig relations. We focus on the “unmodified” strong-contrast approximation because it has the same behavior as the scaled variant (8.76). We define a complex function \( f(kq) \equiv \varepsilon_e(kq) - \varepsilon_q \), which is equal to \( [a + b F(kq)]^{-1} \) for the strong-contrast approximation, where \( a \) and \( b \) are constant real numbers. The following three properties of \( F(kq) \) \( F(kq) \) is an analytic function in the upper half-plane of complex variable \( k_q \); \( F(kq) \) diverges like \( |k_q| \) as \( |k_q| \) goes to infinity; \( \text{Re}[F(kq)] \) and \( \text{Im}[F(kq)] \) are even and odd functions of \( k_q \), respectively], shown in Secs. 8.16 and 8.16.4, lead \( f(kq) \) to possess the three properties:

(i) \( \varepsilon_e(kq) \) is an analytic function in the upper half-plane of \( k_q \);

(ii) \( f(kq) = \varepsilon(kq) - \varepsilon_q \) vanishes like \( 1/|k_q| \) as \( |k_q| \) goes to infinity; and

(iii) \( \text{Re}[f(kq)] \) and \( \text{Im}[f(kq)] \) are even and odd functions of \( k_q \), respectively.

Note that any complex function \( f(q) \) exhibiting properties (i) and (ii) satisfies the following relations, which correspond to the Kramer-Kronig relations in general
By combining these relations with the property (iii) of $f(k_q)$, one can immediately show that the strong-contrast approximations (both unmodified and scaled ones) meet the Kramer-Kronig relations given in Eqs. (8.88) and (8.89). This result makes sense because the strong-contrast approximations or, equivalently, the nonlocal attenuation function $F(Q)$ come from $G^{(q)}(x, x')$ given in Eq. (8.28) that is the temporal Fourier transform of the retarded Green function $G^{(q)}(x, t, x', t')$ [134] accounting for causality.

![Equilibrium packing, $\phi_2=0.25$, $\varepsilon_2/\varepsilon_1=4$](image)

Figure 8.12: Numerical verification of the Kramers-Kronig relations for the (unscaled) strong-contrast approximation. We consider 3D equilibrium packing of packing fraction $\phi_2 = 0.25$ and contrast ratio $\varepsilon_2/\varepsilon_1 = 4$. In the upper panel, we compare the real part $\text{Re}[\varepsilon_e(k_1)]$ of the approximation to that evaluated from the Kramers-Kronig relation (8.88) and the imaginary part of the strong-contrast approximation. In the lower panel, we compare the imaginary part $\text{Im}[\varepsilon_e(k_1)]$ of the approximation to that evaluated from (8.89) and the real part of the approximation. The predictions from the approximation and its transform via Kramers-Kronig relations show excellent agreement.

For illustrative purpose, we compare the effective dielectric constant $\varepsilon_e(k_q)$ from the strong-contrast approximation with its transformation via the Kramers-Kronig relations. We consider 3D equilibrium packings of packing fraction $\phi_2 = 0.25$ and
contrast ratio $\varepsilon_2/\varepsilon_1 = 4$; see Fig. 8.12. Specifically, in the upper panel of Fig. 8.12, we present the predictions of $\text{Re}[\varepsilon_e(k_1)]$ from the approximation and those evaluated by using the Kramers-Kronig relation (8.88) and $\text{Im}[\varepsilon_e(k_1)]$ from the approximation. Similarly, in the lower panel, we compare $\text{Im}[\varepsilon_e(k_1)]$ from the approximation with the application of Eq. (8.89) to $\text{Re}[\varepsilon_e(k_1)]$ from the approximation. Computing the Kramers-Kronig relations, we carry out numerical integrals with an upper limit of $q = 400$. In both panels, the strong-contrast approximation and its transformations via the Kramers-Kronig relations show excellent agreement, which numerically confirms our proof. Note that in the upper panel, there are small deviations between the approximation and its transform via the Kramers-Kronig relations. Such differences result from the fact that a finite upper limit is used to compute Eq. (8.88), and the numerator $q \text{Im}[\varepsilon_e(q)]$ in the integrand of Eq. (8.88) goes to zero slowly as $q$ increases.

8.13 Appendix C: Strong-Contrast Approximation at the Three-Point Level

Here we present the strong-contrast approximation at the three-point level for a spherical exclusion region. It is obtained from Eq. (8.58) by setting $A_n^{(p)} = 0$ for $n \geq 4$ and by solving it in $\varepsilon_e(k_q)$:

\[
\frac{\varepsilon_e(k_q)}{\varepsilon_q} = 1 + \frac{d \beta_{pq} \phi_p^2}{\phi_p(1 - \beta_{pq} \phi_p) + (d - 1)\pi/[2^{d/2} \Gamma(d/2)] \beta_{pq} F(k_q) - \beta_{pq}^2 A_3^{(p)}(k_q)},
\]

(8.90)

where the local three-point parameter is given as
\[ A_3^{(p)}(k_q) = -\frac{(d \varepsilon_q)^2}{\phi_p} \int_{\mathbb{R}^d} dx_1 dx_2 \frac{1}{d} \text{Tr} \left[ \mathbf{H}^{(q)}(x_1 - x_2) e^{-i k_q (x_1 - x_2)} \cdot \mathbf{H}^{(q)}(x_2 - x_3) e^{-i k_q (x_2 - x_3)} \right] \times \Delta_3^{(p)}(x_1, x_2, x_3) \] (8.91)

\[ = -\frac{1}{\phi_p (2\pi)^{2d}} \int dq_1 dq_2 \frac{1}{q_1^2 - k_q^2} \frac{1}{q_2^2 - k_q^2} \left\{ (d - 1) k_q^2 q_1^2 + q_1^2 q_2^2 [d(q_1 \cdot q_2)^2 - 1] \right\} \times \tilde{\Delta}_3^{(p)}(q_1 + k_q, q_2 + k_q), \] (8.92)

where, due to the statistical homogeneity,

\[ \tilde{\Delta}_3^{(p)}(q_1, q_2) \equiv \int dr_1 dr_2 e^{-i q_1 \cdot r_1} e^{-i q_2 \cdot r_2} \left[ S_2^{(p)}(r_1) S_2^{(p)}(r_2) - \phi_p S_3^{(p)}(r_1, r_2) \right]. \]

Note that Eq. (8.92) is obtained from Eq. (8.91) via Parseval’s theorem. The static limit of Eq. (8.91) for statistically isotropic media is given by [296]

\[ A_3^{(p)}(0) = (d - 1) \phi_p (1 - \phi_p) \zeta_p = \left( \frac{d}{\Omega_d} \right)^2 \int_{\mathbb{R}^d} dr ds d(\hat{r} \cdot \hat{s})^2 \left[ S_3^{(p)}(r, s, t) - S_2^{(p)}(r) S_2^{(p)}(s) / \phi_p \right], \] (8.93)

where \( \Omega_d \) is the surface area of a unit sphere in \( \mathbb{R}^d \), \( t \equiv |r - s| \) and the parameter \( \zeta_p \) lies in the closed interval \([0, 1]\).

**8.14 Appendix D: Original Local Strong-Contrast Expansions**

Here we state the original local strong-contrast expansions obtained by Rechtsman and Torquato [242] and then discuss approximations extracted from them. The expansions are valid only in the quasistatic regime because they are derived from a homogenized constitutive relation that is local in space. The problem setup is the same
as the one discussed in Sec. IV in the main text. The original Rechtsman-Torquato quasistatic strong-contrast expansion for the effective dielectric constant tensor $\varepsilon_e$ is given by

$$(\phi_p \beta_{pq})^2 \left[ \frac{\varepsilon_e(k_q) - \varepsilon_q I}{\varepsilon_e(k_q) + (d - 1)\varepsilon_q I} \right]^{-1} = \phi_p \beta_{pq} I - \sum_{n=2}^{\infty} A_n^{(p)}(k_q) \beta_{pq}^n, \quad (8.94)$$

where the dielectric polarizability $\beta_{pq}$ is given in Eq. (8.38), and the $n$-point local parameters $A_n^{(p)}(k_q)$ are defined as

$$A_2^{(p)}(k_q) = d\varepsilon_q \int \mathcal{H}^{(q)}(r_1) \chi_v(r_1) \, dr_1, \quad (8.95)$$

$$A_n^{(p)}(k_q) = d\varepsilon_q \left( -\frac{d\varepsilon_q}{\phi_p} \right)^{n-2} \int \mathcal{H}^{(q)}(x_1 - x_2) \cdots \mathcal{H}^{(q)}(x_{n-1} - x_n)$$

$$\times \Delta_n^{(p)}(x_1, \cdots, x_n) \, dx_2 \cdots dx_n, \quad n \geq 3, \quad (8.96)$$

where $\int_\epsilon \, dr = \lim_{\epsilon \to 0^+} \int_{|r| > \epsilon} \, dr$ for a spherical exclusion region, the explicit formula of the second-rank tensor field $\mathcal{H}^{(q)}(r)$ is given in Eq. (8.29), and $\Delta_n^{(p)}(x_1, \cdots, x_n)$ is a position-dependent determinant involving up to the $n$-point correlation function associated with the polarized phase $p$ of composites, given in Eq. (59) in the main text.

**Remarks:**

1. The effective dielectric constant tensor $\varepsilon_e$ depends on the wavenumber $k_q$ of the applied electric field in the reference phase. Importantly, while the resulting tensor $\varepsilon_e$ can be anisotropic, it is expected to be independent of the propagation direction of the applied field.

2. The $n$-point parameters $A_n^{(p)}(k_q)$ given in Eqs. (8.95) and (8.96) are different from those for the nonlocal strong-contrast expansions beyond the quasistatic regime; see Sec. 8.15.
3. The homogenized relation for the original strong-contrast expansion is local in space:

\[ \langle P \rangle (x) = L_e^{(q)}(k = 0) \cdot \langle F \rangle (x), \]

where ensemble averages \( \langle P \rangle (x) \) and \( \langle F \rangle (x) \) are constants of position. The reader is referred to Eq. (8.132) for the nonlocal counterpart that is derived in this chapter.

4. The second-rank tensor field \( H^{(q)}(r) \) given in Eq. (8.29) is related to \( t^{(p)}(r) \) given in Eq. (19) in Ref. [242] as follows:

\[ t^{(p)}(r) = \Omega_d \varepsilon_q H^{(q)}(r), \]

where \( \Omega_d \) is the surface area of a \( d \)-dimensional unit sphere.

### 8.14.1 Truncated Approximations

For the reasons that we discussed in Sec. IV in the main text, truncation of expansions (8.94) at the \( n \)-point level gives an accurate approximation of \( \varepsilon_e \) for two-phase composites. We present the two- and three-point approximations of the original strong-contrast expansions, derived in Ref. [242], for the macroscopically isotropic media. In this case, the effective dielectric constant tensor \( \varepsilon_e(k_q) \) can be reduced to a scalar quantity \( \varepsilon_e(k_q) \) by taking the trace, i.e., \( \varepsilon_e(k_q) = \text{Tr}[\varepsilon_e(k_q)]/d \).

The two-point approximation is obtained by setting \( A_n^{(p)} = 0 \) for \( n \geq 3 \) in Eq. (8.94), taking trace of its both sides, and solving them in \( \varepsilon_e(k_q) \):

\[
\frac{\varepsilon_e(k_q)}{\varepsilon_q} = 1 + \frac{d\beta_{pq}\phi_p^2}{\phi_p(1 - \beta_{pq}\phi_p) - \beta_{pq} A_2^{(p)}(k_q)}
= 1 + \frac{d\beta_{pq}\phi_p^2}{\phi_p(1 - \beta_{pq}\phi_p) + (d - 1)\pi/[2^{d/2} \Gamma(d/2)]\beta_{pq} F(k_q)},
\]

(8.98)
where $\Gamma(x)$ is the Gamma function, $\beta_{pq}$ is defined in Eq. (8.38), and $A_n^{(p)} \equiv \text{Tr} [ A_n^{(p)} ] / d$ for $n = 2, \cdots$. Here the local attenuation function $\mathcal{F}(Q)$ is defined as

$$
\mathcal{F}(Q) \equiv -\frac{2^{d/2} \Gamma(d/2)}{\pi} Q^2 \int \frac{Q}{2\pi} \mathcal{H}_{d/2-1}(Qr) \chi_v(r) \, dr \quad (8.99)
$$

$$
= -\frac{\Gamma(d/2)}{2^{d/2} \pi^{d+1}} Q^2 \int \frac{\hat{\chi}_v(q)}{|q|^2 - Q^2} \, dq, \quad (8.100)
$$

where $\mathcal{H}_{\nu}^{(1)}(x)$ is the Hankel function of the first kind of order $\nu$, and Eq. (8.100) is obtained by applying the Parseval theorem to Eq. (8.99). Importantly, we note that the integrand in Eq. (8.100) depends on the shape of the exclusion-region; see Sec. 8.15.1 for details.

Analogously, the three-point approximation is obtained from Eq. (8.94) as follows:

$$
\frac{\varepsilon_e(k_q)}{\varepsilon_q} = 1 + \frac{d\beta_{pq} \phi_p^2}{\phi_p (1 - \beta_{pq} \phi_p) - \beta_{pq} A_2^{(p)}(k_q) - \beta_{pq}^2 A_3^{(p)}(k_q)}, \quad (8.101)
$$

where the three-point local parameter is given as

$$
A_3^{(p)}(k_q) \equiv -\frac{(d\varepsilon_q)^2}{\phi_p} \int \frac{1}{d} \text{Tr} \left[ H^{(q)}(x_1 - x_2) \cdot H^{(q)}(x_2 - x_3) \right] \Delta_3^{(p)}(x_1, x_2, x_3) \quad (8.102)
$$

$$
= -\frac{1}{\phi_p (2\pi)^2} \int \text{d}q_1 \text{d}q_2 \frac{1}{q_1^2 - k_q^2} \frac{1}{q_2^2 - k_q^2} \left\{ (d-1)^2 k_q^4 + q_1^2 q_2^2 \left[ d(q_1 \cdot \mathbf{q}_2)^2 - 1 \right] \right\} \Delta_3^{(p)}(q_1, q_2), \quad (8.103)
$$

where, due to the statistical homogeneity,

$$
\Delta_3^{(p)}(q_1, q_2) = \int \text{d}r_1 \text{d}r_2 e^{-i\mathbf{q}_1 \cdot \mathbf{r}_1} e^{-i\mathbf{q}_2 \cdot \mathbf{r}_2} \left[ S_2^{(p)}(r_1) S_2^{(p)}(r_2) - \phi_p S_3^{(p)}(r_1, r_2) \right].
$$
8.14.2 Properties of the Local Attenuation Function $\mathcal{F}(Q)$

Assuming both $\varepsilon_1$ and $\varepsilon_2$ are real-valued, the imaginary part of the local attenuation function $\mathcal{F}(Q)$ determines the attenuation characteristics of a composite predicted by the two-point approximation (8.98). In this subsection, we investigate some generic and microstructure-dependent properties of $\mathcal{F}(Q)$. We compare $\mathcal{F}(Q)$ to its nonlocal counterpart defined in Eq. (8.71) in Sec. 8.17.

For statistically isotropic media, the real and imaginary parts of local attenuation function, defined in Eq. (8.99), can be simplified as

\begin{align*}
\text{Im}[\mathcal{F}(Q)] &= -\lim_{\epsilon \to 0^+} \int_{\epsilon}^{\infty} dr \, Q \, \chi_V(r) \, (Qr)^{d/2} \, J_{d/2-1}(Qr) \quad (8.104) \\
&= -\frac{Q^d}{(2\pi)^{d/2}} \tilde{\chi}_V(Q), \quad (8.105) \\
\text{Re}[\mathcal{F}(Q)] &= \lim_{\epsilon \to 0^+} \int_{\epsilon}^{\infty} dr \, Q \, \chi_V(r) \, (Qr)^{d/2} \, Y_{d/2-1}(Qr) \quad (8.106) \\
&= -\frac{2Q^2}{\pi} \, \text{p.v.} \, \int_{0}^{\infty} dq \, \frac{1}{q(Q^2 - q^2)} \, \text{Im}[\mathcal{F}(q)], \quad (8.107)
\end{align*}

where $J_{\nu}(x)$ [$Y_{\nu}(x)$] is the Bessel function of the first kind [the second kind] of order $\nu$, and p.v. stands for the Cauchy principal value of an integral. For a finite $Q$, the limits of integrals in Eqs. (8.104) and (8.106) can be simplified to $\int_{0}^{\infty} dr$ because the integrands do not possess a singularity at the origin. Equation (8.107) is obtained from Eqs. (8.104) and (8.106) by utilizing the following identity $Y_{\nu-1}(Qr) = \text{p.v.} \int_{0}^{\infty} dq \, \frac{q^{\nu}}{(Q^2 - q^2)} \, J_{\nu-1}(qr)$, for positive real numbers $Q$, $r$, and $\nu$.

Using the formulas above, we investigate the asymptotic behaviors of $\mathcal{F}(Q)$ in both quasistatic (small-$Q$) and large-$Q$ regimes. For the imaginary part $\text{Im}[\mathcal{F}(Q)]$, those expressions can be easily obtained from Eq. (8.105) and the asymptotic behaviors of the spectral density $\tilde{\chi}_V(Q)$. For either nonhyperuniform or hyperuniform isotropic

\[343\]
media, the spectral density has the following power-law scalings:

\[
\tilde{\chi}_V(Q) \sim \begin{cases} 
Q^\alpha, & \text{in the quasistatic regime}, \\
Q^{-(d+1)}, & \text{in the large-}Q \text{ regime},
\end{cases}
\] (8.108)

where the exponent \(\alpha\) lies in the interval \((0, \infty)\) for hyperuniform systems, and \(\alpha = 0\) for nonhyperuniform systems. Combining Eqs. (8.105) and (8.108) immediately gives

\[
\text{Im}[\mathcal{F}(Q)] \sim \begin{cases} 
Q^{d+\alpha}, & \text{in the quasistatic regime}, \\
Q^{-1}, & \text{in the large-}Q \text{ regime}.
\end{cases}
\] (8.109)

By contrast, for both hyperuniform and nonhyperuniform systems, the real part exhibits common asymptotic behaviors

\[
\text{Re}[\mathcal{F}(Q)] \sim Q^2, \quad \text{in the quasistatic regime},
\] (8.110)
\[
\text{Re}[\mathcal{F}(Q)] \to \frac{2^{d/2} \Gamma(d/2)}{\pi} \phi_p (1 - \phi_p), \quad \text{in the large-}Q \text{ regime}.
\] (8.111)

Expression (8.110) is obtained from Eq. (8.106) by using the Taylor expansion of \(Y_\nu(x)\) around the origin:

\[
\text{Re}[\mathcal{F}(Q)] \sim Q^2 \int_0^\infty dr \, r \chi_V(r) \sim Q^2.
\] (8.112)

In order to obtain Eq. (8.111), we use Eqs. (8.105) and (8.107) to write

\[
\text{Re}[\mathcal{F}(Q)] = \frac{2Q^2}{\pi} \frac{1}{(2\pi)^{d/2}} \text{p.v.} \int_0^\infty dq \, \frac{q^{d-1}}{Q^2 - q^2} \tilde{\chi}_V(q) = \frac{2}{\pi} \frac{1}{(2\pi)^{d/2}} \text{p.v.} \int_0^\infty dq \, \frac{q^{d-1}}{1 - (q/Q)^2} \tilde{\chi}_V(q).
\]

Since \(q^{d-1} \tilde{\chi}_V(q)\) vanishes like \(1/q^2\) for large \(q\), the singularity contribution in this
integral can be neglected in the large-$Q$ limit

$$\lim_{Q \to \infty} \text{Re}[\mathcal{F}(Q)] \approx \frac{2}{\pi} \frac{1}{(2\pi)^{d/2}} \int_0^\infty dq \, q^{d-1} \tilde{\chi}_V(q) = \frac{2^{d/2} \Gamma(d/2)}{\pi} \phi_p (1 - \phi_p),$$

where we note that the integral above is related to the inverse Fourier transform of $\tilde{\chi}_V(q)$ at $r = 0$, i.e.,

$$\frac{1}{(2\pi)^d} \int_0^\infty dq \, \frac{2\pi^{d/2}}{\Gamma(d/2)} q^{d-1} \tilde{\chi}_V(q) = \chi_V(0) = \phi_p (1 - \phi_p).$$

We present the local attenuation function $\mathcal{F}(Q)$ for the five models three-dimensional disordered composite media, discussed in the main text; see Fig. 8.13. In the inset of the lower panel of this figure, one can see how the power-law scalings vary with models in the quasistatic regime.

![Figure 8.13](image)

Figure 8.13: Evaluation of negatives of (a) the real and (b) negative imaginary parts of the local attenuation function $\mathcal{F}(Q)$, defined in Eq. (8.99), for the four models of three-dimensional disordered particulate media. Each model has the same volume fraction for the dispersed phase $\phi_2 = 0.25$. The inset in (b) is the log-log plot of the larger panel. The first three models consist of spheres of radius $a$. For class I hyperuniform packings via tessellation-based procedure, $a$ is the mean sphere radius, i.e., $\phi_2 = \rho \, v_1(a)$, where $\rho$ is number density of particle centers, and $v_1(a)$ is the volume of a $d$ dimensional sphere of radius $a$. 
8.15 Appendix E: Derivations of Nonlocal Strong-Contrast Expansion

Here we present a detailed derivation of the nonlocal strong-contrast expansion presented in the main text. Aforementioned, we consider a macroscopically large ellipsoidal two-phase composite specimen in $\mathbb{R}^d$ embedded inside an infinitely large reference phase of dielectric constant tensor $\varepsilon_I$. We assume that the microstructure is perfectly general, and the inhomogeneity length scales $\ell$ are much smaller than the size of specimen $L$, i.e., $\ell \ll L$. The shape of this specimen is purposely chosen to be non-spherical since any rigorously correct expression for the effective stiffness tensor must ultimately be independent of the shape of the composite specimen in the infinite-volume limit. The local dielectric constant tensor $\varepsilon(x)$ of a two-phase composite is written as

$$\varepsilon(x) \equiv \varepsilon_1 I^{(1)}(x) + \varepsilon_2 I^{(2)}(x),$$  \hspace{1cm} (8.113)

where $\varepsilon_i$ is the dielectric constant tensor of phase $i = 1, 2$, and $I^{(i)}(x)$ is the the indicator of phase $i$ (see Sec. II in the main text). It is assumed that the incident electric waves $E_0(x)$ is a plane wave of an angular frequency $\omega$ and wavevector $k_I(\omega)$ in the reference phase; see Eq. (8.23). The associated wavelength $\lambda = 2\pi/|k_I|$ must lie between the inhomogeneity length scales $\ell$ and the specimen size $L$. Granting that the effective-medium description is valid for the composite, we obtain the corresponding dielectric constant tensor $\varepsilon_e(\omega, k_I)$.

In the ensuing derivation, we make the following three assumptions on the phase properties:

(a) the reference phase and phases 1, 2 are perfect insulators and dissipationless so that $\varepsilon_i$'s are purely real-valued and frequency-independent [i.e., $\varepsilon_i(\omega) = \varepsilon_i$ and...
\[ \text{Im}[\varepsilon_i] = 0 \text{ for } i = 1, 2, I; \]

(b) the dielectric constant of the reference phase is isotropic, i.e., \( \varepsilon_I = \varepsilon_I \mathbf{I} \); and

(c) the magnetic permeability tensors of all phases are identical (i.e., \( \mu_1 = \mu_2 = \mu_I = \mu_0 \mathbf{I} \)), where \( \mu_0 \) is the magnetic permeability of vacuum.

The assumptions (a) and (b) implies the linear dispersion relation in the reference phase [i.e., \( k_I(\omega) \equiv |\mathbf{k}_I(\omega)| = \sqrt{\varepsilon_I \omega / c} \)], where \( c \) is the speed of light in vacuum, and thus we henceforth do not explicitly indicate the dependence of functions on \( \omega \).

We derive some important integral equations for local fields in Sec. 8.15.1. We derive a nonlocal homogenized relation associated with the strong-contrast expansions, equivalent to the standard constitutive relation in Sec. 8.15.2. We then derive the strong-contrast expansions for an arbitrarily-shaped exclusion region and any reference phase in Sec. 8.15.3. In Sec. 8.15.4, we then simplify the expansions derived in Sec. 8.15.3 by assuming a spherical exclusion region and reference phase \( q \) (\( = 1, 2 \)).

### 8.15.1 Integral Equation for Cavity Electric field

We derive the equation of motion for the electric field inside a composite at a given angular frequency \( \omega \). For this purpose, we begin with the governing frequency-dependent wave equation that is obtained from the Maxwell equations and assumptions (a) and (c):

\[
\nabla \times \nabla \times \mathbf{E}(\mathbf{x}) - \omega^2 \mu_0 \varepsilon_0 \varepsilon(\mathbf{x}) \cdot \mathbf{E}(\mathbf{x}) = 0,
\]

where we have used separation of variables \( \mathbf{E}(\mathbf{x}, t) \rightarrow \mathbf{E}(\mathbf{x}) e^{-i\omega t} \), \( \mu_0 \) is the magnetic permeability of vacuum, and \( \varepsilon_0 \) is the dielectric permittivity of vacuum. We rewrite this homogeneous differential equation with respect to the reference phase as follows:

\[
\nabla \times \nabla \times \mathbf{E}(\mathbf{x}) - \omega^2 \mu_0 \varepsilon_0 \varepsilon_I \cdot \mathbf{E}(\mathbf{x}) = \omega^2 \mu_0 \varepsilon_0 \mathbf{P}(\mathbf{x}),
\]

(8.114)
where \( P(x) \) is the *induced flux polarization field* given by

\[
P(x) \equiv [\varepsilon(x) - \varepsilon_I] \cdot E(x).
\]

(8.115)

Assuming the isotropy of the reference phase, one can simplify Eq. (8.114)

\[
\nabla \times \nabla \times E(x) - k_I^2 E(x) = \left( \frac{\omega}{c} \right)^2 P(x),
\]

(8.116)

where \( k_I \equiv \omega \sqrt{\varepsilon_I \mu_0} = \sqrt{\varepsilon_I \omega / c} \) is the wavenumber of electromagnetic waves inside the reference phase.

When external electric field \( E_0(x) \) is incident to the composite, the consequent local electric field \( E(x) \) can be computed by using the dyadic Green’s function \( G^{(I)}(x, x') \) satisfying the following equation:

\[
\nabla \times \nabla \times G^{(I)}(x, x') - k_I^2 G^{(I)}(x, x') = \left( \frac{\omega}{c} \right)^2 I \delta(x - x'),
\]

(8.117)

\[
G^{(I)}(x, x') \to 0, \quad |x' - x| \to \infty.
\]

Using the Green’s method, the local electric field is written as

\[
E(x) = E_0(x) + \int G^{(I)}(x, x') \cdot P(x') \, dx',
\]

(8.118)

\[
= E_0(x) + \underbrace{-D^{(I)} \cdot P(x)}_{\text{inside the exclusion-region}} + \underbrace{\int H^{(I)}(x, x') \cdot P(x') \, dx'}_{\text{outside the exclusion-region}},
\]

(8.119)

where we note that due to the singular nature of Green’s function around the origin (i.e., \( x - x' = 0 \)), the integral (8.118) should be separated into two parts; one is the integral inside an infinitesimal exclusion region around the origin, and another is the integral outside the exclusion region (denoted by \( \int_{x} dx' \)). Thus, the Green’s function
can be written concisely as

\[ G^{(I)}(x, x') = -D^{(I)} \delta(x - x') + H^{(I)}(x - x'), \tag{8.120} \]

where the second-rank constant tensor \( D^{(I)} \) depends on the shape of exclusion region.

For a spherical exclusion region in \( \mathbb{R}^d \),

\[ D^{(I)} = \frac{1}{d\varepsilon I}, \tag{8.121} \]

where \( I \) is the second-rank identity tensor.

It is useful to provide explicit formulas for Eq. (8.120) in both direct- and Fourier-space representations. In the direct-space representation (outside the “exclusion region”), the second-rank tensor field \( H^{(I)}(r) \), where \( r \equiv x - x' \), is given by Eq. (8.29) whose the superscript \( q = I \). The Fourier representation of Eq. (8.120), however, is concise and simple [see Eq. (8.31)] which can be obtained from the Fourier transform of Eq. (8.117) by using the orthogonality of two tensors \( \Pi \equiv \mathbf{q}\mathbf{q}/|\mathbf{q}|^2 \) and \( I - \Pi \). It is important to note that Eq. (8.31) is independent of the “exclusion region.”

We now express the integral equation (8.119) more compactly in linear operator form

\[ \mathbf{E} = \mathbf{E}_0 + G^{(I)} \mathbf{P}. \tag{8.122} \]

Excluding the contribution from a exclusion region in Eq. (8.119), we obtain the integral equation for the cavity intensity field \( \mathbf{F}(x) \):

\[
\mathbf{F} = \mathbf{E} + D^{(I)} \cdot \mathbf{P} = \mathbf{E}_0 + H^{(I)} \mathbf{P} = \left[ I + D^{(I)} \cdot (\varepsilon(x) - \varepsilon_I) \right] \cdot \mathbf{E}. \tag{8.125}
\]
Using the definition (8.115) and expression (8.125), one obtains a linear constitutive relation between $P(x)$ and $F(x)$:

$$P(x) = L^{(I)}(x) \cdot F(x), \quad (8.126)$$

where

$$L^{(I)}(x) = (\varepsilon(x) - \varepsilon_I) \cdot [I + D^{(I)} \cdot (\varepsilon(x) - \varepsilon_I)]^{-1} = L_1^{(I)} \Theta^{(1)}(x) + L_2^{(I)} \Theta^{(2)}(x), \quad (8.127)$$

and, for phase $p$ $(=1,2)$,

$$L_p^{(I)} = (\varepsilon_p - \varepsilon_I) \cdot [I + D^{(I)} \cdot (\varepsilon_p - \varepsilon_I)]^{-1}. \quad (8.128)$$

**Remarks:**

1. The definition of $G^{(I)}(x, x')$, given in Eq. (8.117), is different from that given in Ref. [242]. Specifically, the Green’s function in this chapter (8.120) is a multiplication of that in Ref. [242] with $(\omega/c)^2$. By doing so, Eq. (8.120) converges to its static counterpart in the static limit (i.e., $\omega \to 0$).

2. The reference phase $I$ employed in the Green’s function $G^{(I)}(x, x')$ can be different from phases 1 and 2. In the main text, however, we take phase $q$ $(= 1, 2)$ as the reference phase for simplicity.

3. Equation (8.120) can be written more explicitly as

$$G^{(I)}(x, x') = \begin{cases} -D^{(I)} \delta(x - x'), & \text{outside the exclusion-region} \\ H^{(I)}(x - x'), & \text{inside the exclusion-region}. \end{cases}$$
Due to this definition, while $H^{(I)}(r)$ in the direct-space is independent of a choice of exclusion-region, the Fourier transform $\tilde{H}^{(I)}(q)$ depends on the shape of the exclusion region

$$
\tilde{H}^{(I)}(q) = \int dr \, e^{-iq \cdot r} \, H^{(I)}(r) = \int_\epsilon dr \, e^{-iq \cdot r} \, H^{(I)}(r)
$$

$$
= \tilde{G}^{(I)}(q) + D^{(I)}, \quad (8.129)
$$

where we have used Eq. (8.120).

### 8.15.2 Equivalence of Two Nonlocal Relations

In the nonlocal strong-contrast formalism, the following nonlocal homogenized constitutive relation is employed:

$$
\langle P \rangle^{(I)}(k) = L_e^{(I)}(k) \cdot \langle F \rangle^{(I)}(k), \quad (8.130)
$$

where the constant tensor $L_e^{(I)}(k)$ is written explicitly as

$$
L_e^{(I)}(k) \equiv [\varepsilon_e(k) - \varepsilon_I] \cdot \left[ I + D^{(I)} \cdot (\varepsilon_e(k) - \varepsilon_I) \right]^{-1}. \quad (8.131)
$$

Here, $k$ is a wavevector, $\langle f \rangle^{(I)}(k) \equiv \int \langle f \rangle(x) \exp(-i k \cdot x) \, dx$, $\langle f \rangle(x)$ is a ensemble average of field $f(x)$ that varies with position. We note that a relation like Eq. (8.130) is nonlocal in space because it is written in terms of a convolution operation

$$
\langle P \rangle(x) = \int dx' \, L_e^{(I)}(x - x') \cdot \langle F \rangle(x'), \quad (8.132)
$$

implying that $\langle P \rangle(x)$ at position $x$ is determined by $\langle F \rangle(x')$ in vicinity of $x$. In this subsection, we show that the relation (8.130) is equivalent to the following popular
constitutive relation:
\[
\langle \mathbf{D} \rangle (\mathbf{k}) = \varepsilon_0 \varepsilon_e (\mathbf{k}) \cdot \langle \mathbf{E} \rangle (\mathbf{k}),
\]  
(8.133)

where \( \langle \mathbf{D} \rangle (\mathbf{k}) \) is the spatial Fourier transform of an ensemble average of the electric displacement field

\[
\mathbf{D}(\mathbf{x}) \equiv \varepsilon_0 \mathbf{E}(\mathbf{x}) + \mathbf{P}(\mathbf{x}) = \varepsilon_0 \varepsilon(\mathbf{x}) \cdot \mathbf{E}(\mathbf{x}),
\]  
(8.134)

and \( \mathbf{P}(\mathbf{x}) \) is the electric polarization density.

To do so, we rewrite the induced flux polarization field \( \langle \mathbf{P} \rangle (\mathbf{k}) \) and the cavity intensity field \( \langle \mathbf{F} \rangle (\mathbf{k}) \) in terms of \( \langle \mathbf{E} \rangle (\mathbf{k}) \), and then eliminate \( \langle \mathbf{E} \rangle (\mathbf{k}) \) between these two expressions. We begin with expression the induced flux polarization field and cavity intensity field in terms of the electric displacement field \( \mathbf{D}(\mathbf{x}) \) and electric field \( \mathbf{E}(\mathbf{x}) \). Using Eqs. (8.115), (8.123), and (8.134), the followings are obtained

\[
\mathbf{P}(\mathbf{x}) = \frac{1}{\varepsilon_0} \mathbf{D}(\mathbf{x}) - \varepsilon_I \mathbf{E}(\mathbf{x}),
\]  
(8.135)

\[
\mathbf{F}(\mathbf{x}) = \mathbf{E}(\mathbf{x}) + \mathbf{D}^{(f)} \cdot \mathbf{P}(\mathbf{x}).
\]  
(8.136)

After averaging Eq. (8.135) over an ensemble, taking its spatial Fourier transform, and substituting Eq. (8.133) into it to eliminate \( \langle \mathbf{D} \rangle (\mathbf{k}) \), we obtain the following expression

\[
\langle \mathbf{P} \rangle (\mathbf{k}) = \langle \mathbf{D} \rangle (\mathbf{k}) / \varepsilon_0 - \varepsilon_I \langle \mathbf{E} \rangle (\mathbf{k})
= [\varepsilon_e (\mathbf{k}) - \varepsilon_I \mathbf{I}] \cdot \langle \mathbf{E} \rangle (\mathbf{k}).
\]  
(8.137)
Analogously, we re-express Eq. (8.136) in terms of $\langle E(k) \rangle$ by using Eq. (8.137):

$$
\langle F(k) \rangle = \langle E(k) \rangle + D^{(I)} \cdot \langle P(k) \rangle
$$

$$
= \left\{ I + D^{(I)} \cdot \left[ \varepsilon_e(k) - \varepsilon I I \right] \right\} \cdot \langle E(k) \rangle. \tag{8.138}
$$

Now, the linear relation (8.130) is obtained by inverting the right-side of Eq. (8.138) and substituting it to Eq. (8.137). Later, we will show that the wavevector $k$ in Eq. (8.130) is indeed identical to wavevector of the incident waves $k_I$.

### 8.15.3 Strong-Contrast Expansion in General Cases

In this subsection, we derive an exact expression for the effective dielectric constant tensor $\varepsilon_e$ for an arbitrary reference phase $I (\neq 1, 2)$ and an arbitrarily-shaped exclusion region. To do so, we derive an explicit expression for the constant tensor $L_e^{(I)}(k_I)$ in the nonlocal homogenized relation (8.132) with an incident electric field (8.23).

For this purpose, we first find explicit expressions for $\langle P(k) \rangle$ and $\langle F(k) \rangle$ in terms of the applied field $\tilde{E}_0$ from the integral equation (8.124). We then find an explicit expression for the effective constant tensor $L_e^{(I)}$ by eliminating $\tilde{E}_0$ between these two expressions. Keeping in mind that the tensors $L^{(I)}$, $L_e^{(I)}$, and $H^{(I)}$ are associated with the reference phase $I$, we shall temporarily drop the superscript $I$ when referring these tensors in the following derivation. Applying Eq. (8.124) to Eq. (8.126) yields

$$
P = LE_0 + LHP. \tag{8.139}
$$

Iterative substitution of this expression with the polarization field in the right-hand side gives

$$
P = (I + LH + LHLH + \cdots)LE_0 = [I - LH]^{-1}LE_0 = SE_0.
$$
More explicitly, we write out the equation above as

\[
P(1) = \int d1' \left[ L(1) \delta(1 - 1') + L(1) H(1, 1') L(1') \\
+ \int d2 L(1) H(1, 2) L(2) H(2, 1') L(1') + \cdots \right] \cdot E_0(1') .
\]

\[
= \int d1' S(1, 1') \cdot E_0(1') ,
\]

(8.140)

where \( \delta(x) \) is the Dirac delta function in \( \mathbb{R}^d \), and boldface numbers \( 1', 1, 2, \cdots \) are short-hand notations for position vectors \( r_{1'}, r_1, r_2, \cdots \). The two-point tensor operator \( S(1, 1') \) explicitly reads

\[
S(1, 1') = L(1) \delta(1 - 1') + L(1) H(1, 1') L(1') \\
+ \int d2 L(1) H(1, 2) L(2) H(2, 1') L(1') + \cdots .
\]

(8.141)

For a statistically homogeneous medium, an ensemble average of the two-point operator \( S(1, 1') \) becomes dependent on relative positions, i.e., \( \langle S \rangle (1, 1') = \langle S \rangle (1 - 1') \), and an ensemble average of Eq. (8.140) can be written as a convolution:

\[
\langle P \rangle (1) = \int d1' \langle S \rangle (1 - 1') \cdot E_0(1') .
\]

(8.142)

The nonlocal relation (8.142) can be simplified in the Fourier space as follows:

\[
\langle \tilde{P} \rangle (k) = \langle \tilde{S} \rangle (k) \cdot \tilde{E}_0(k) .
\]

(8.143)

Here it is crucial to note that from Eq. (8.23),

\[
\tilde{E}_0(k) = \tilde{E}_0 \delta(k - k_f) ,
\]

(8.144)

implying that the wavevector \( k \) in Eq. (8.143) must be identical to \( k_f \). From Eq.
where \( \langle \mathbf{S}(\mathbf{k}_f) \rangle \) is explicitly written as

\[
\langle \mathbf{S}(\mathbf{k}_f) \rangle = \int \, d(1 - 1') e^{-i\mathbf{k}_f \cdot (1 - 1')} \left[ L(1) \delta(1 - 1') + L(1) H(1, 1') L(1') + \cdots \right] = \int \, d(1 - 1') e^{-i\mathbf{k}_f \cdot (1 - 1')} \left[ \sum_{p_1=1}^2 L_{p_1} \left\langle \mathcal{I}^{(p_1)}(1) \right\rangle \delta(1 - 1') + \sum_{p_1,p_2=1}^2 L_{p_1} H(1 - 1') L_{p_2} \left\langle \mathcal{I}^{(p_1)}(1) \mathcal{I}^{(p_2)}(1') \right\rangle + \sum_{p_1,p_2,p_3=1}^2 L_{p_1} H(1 - 2) L_{p_2} H(2 - 1') L_{p_3} \left\langle \mathcal{I}^{(p_1)}(1) \mathcal{I}^{(p_2)}(2) \mathcal{I}^{(p_3)}(1') \right\rangle + \cdots \right] = \langle \mathbf{A}_1 \rangle + \langle \mathbf{A}_2 \rangle + \langle \mathbf{A}_3 \rangle + \cdots , \tag{8.146}
\]

where

\[
\langle \mathbf{A}_1 \rangle = \langle \mathbf{L}(1) \rangle = \phi_1 \mathbf{L}_1 + \phi_2 \mathbf{L}_2 , \tag{8.147}
\]

\[
\langle \mathbf{A}_2 \rangle = \sum_{p_1,p_2=1}^2 \int \, d(1 - 2) L_{p_1} H(1, 2) e^{-i\mathbf{k}_f \cdot (1 - 2)} L_{p_2} \left\langle \mathcal{I}^{(p_1)}(1) \mathcal{I}^{(p_2)}(2) \right\rangle , \tag{8.148}
\]

\[
\langle \mathbf{A}_3 \rangle = \sum_{p_1,p_2,p_3=1}^2 \int \, d(1 - 3) L_{p_1} H(1, 2) e^{-i\mathbf{k}_f \cdot (1 - 2)} L_{p_2} H(2, 3) e^{-i\mathbf{k}_f \cdot (2 - 3)} L_{p_3} \times \left\langle \mathcal{I}^{(p_1)}(1) \mathcal{I}^{(p_2)}(2) \mathcal{I}^{(p_3)}(3) \right\rangle . \tag{8.149}
\]

We then obtain an expression for \( \langle F \rangle (1) \) from an ensemble average of Eq. (8.124):

\[
\langle F \rangle (1) = \mathbf{E}_0(1) + \int \, d(1 - 2) H(1 - 2) \cdot \langle \mathbf{P} \rangle (2) . \tag{8.150}
\]

Its spatial Fourier transform gives

\[
\langle \mathbf{F} \rangle (\mathbf{k}_f) = \mathbf{E}_0(\mathbf{k}_f) + \tilde{\mathbf{H}}(\mathbf{k}_f) \cdot \langle \mathbf{P} \rangle (\mathbf{k}_f) = \left[ \langle \mathbf{S} \rangle (\mathbf{k}_f) \right]^{-1} + \tilde{\mathbf{H}}(\mathbf{k}_f) \cdot \langle \mathbf{P} \rangle (\mathbf{k}_f) , \tag{8.151}
\]

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where we have eliminated the term $\tilde{E}_0(k_I)$ by substituting Eq. (8.143). Eventually, comparing the above expression to Eq. (8.130) gives an expression of the constant tensor $L_e$ at given frequency $\omega$ and wavevector $k_I$:

$$[L_e(k_I)]^{-1} = (S)(k_I)^{-1} + \tilde{H}(k_I)$$

$$= [I + D \cdot (\varepsilon_e(k_I) - \epsilon_I I)] \cdot [\varepsilon_e(k_I) - \epsilon_I I]^{-1},$$

(8.152)

where we have employed Eq. (8.131). More explicitly, Eq. (8.152) reads

$$[L_e(k_I)]^{-1} = \left(\begin{array}{ccc} A_1 & A_2 & A_3 \\ \end{array}\right) + \ldots$$

$$= \left(\begin{array}{ccc} I + A_2 A_1^{-1} + A_3 A_1^{-1} + \ldots \end{array}\right) + \int d(-2) H(1 - 2) e^{-ik_I(1-2)}$$

(8.153)

where $A_1$, $A_2$, and $A_3$ are given in Eqs. (8.147), (8.148), and (8.149), respectively.

Expression (8.153) can be rewritten as

$$\langle L(1) \rangle \cdot [L_e(k_I)]^{-1} \cdot \langle L(1) \rangle$$

$$= \langle L(1) \rangle \cdot \left\{ [\varepsilon_e(k_I) - \epsilon_I I] \cdot [I + D \cdot (\varepsilon_e(k_I) - \epsilon_I I)]^{-1} \right\}^{-1} \cdot \langle L(1) \rangle$$

$$= \langle L(1) \rangle - \left[ A_2 - A_1 \int d(-2) H(1 - 2) e^{-ik_I(1-2)} A_1 \right]$$

$$= A_1^{(p)}(k_I)(L_2 - L_1)^2/(d\epsilon_I)$$

$$= A_2^{(p)}(k_I)(L_2 - L_1)^3/(d\epsilon_I)^2$$

$$+ \ldots$$

$$= \langle L(1) \rangle - \sum_{n=2}^{\infty} A_n^{(p)}(k_I)(L_2 - L_1)^n/(d\epsilon_I)^n,$$

(8.154)

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where $T^n$ stands for $n$ successive inner products of a second-rank tensor $T$. Using the following identity

$$\langle T^{(p_1)}(1) T^{(p_2)}(2) \rangle - \langle T^{(p_1)}(1) \rangle \langle T^{(p_2)}(2) \rangle = \begin{cases} \chi_v(1 - 2), & p_1 = p_2, \\ - \chi_v(1 - 2), & p_1 \neq p_2 \end{cases}$$

we simplify the second-order term as follows:

$$A_2^{(p)}(k) (L_2 - L_1)^2/(d \epsilon_I) \equiv \begin{array}{c} \begin{array}{c} A_2 \\ A_1 \end{array} \end{array} - \begin{array}{c} \begin{array}{c} A_1 \\ \varepsilon \end{array} \end{array} \int d(1 - 2) H(1 - 2) e^{-ik_I(1-2)} \begin{array}{c} \begin{array}{c} A_1 \\ A_2 \end{array} \end{array}$$

$$= \sum_{p_1, p_2=1}^2 \int d(1 - 2) L_{p_1} \cdot H(1 - 2) e^{-ik_I(1-2)} \cdot L_{p_2} \left[ \langle T^{(p_1)}(1) T^{(p_2)}(2) \rangle - \langle T^{(p_1)}(1) \rangle \langle T^{(p_2)}(2) \rangle \right]$$

$$= \int d(1 - 2) (L_2 - L_1) \cdot H(1 - 2) e^{-ik_I(1-2)} \cdot (L_2 - L_1) \chi_v(1 - 2), \quad (8.155)$$

$$A_3^{(p)}(k) (L_2 - L_1)^3/(d \epsilon_I)^2 \equiv \begin{array}{c} \begin{array}{c} A_3 \\ \varepsilon \end{array} \end{array} - \begin{array}{c} \begin{array}{c} A_2 \\ \varepsilon \end{array} \end{array} \begin{array}{c} \begin{array}{c} A_1 \\ \varepsilon \end{array} \end{array}^{-1} \begin{array}{c} \begin{array}{c} A_2 \\ \varepsilon \end{array} \end{array}$$

$$= \sum_{p_1, p_2, p_3=1}^2 \int d1 d2 L_{p_1} \cdot H(1 - 2) e^{-ik_I(1-2)} \cdot L_{p_2} \cdot H(2 - 3) e^{-ik_I(2-3)} \cdot L_{p_3} \left[ \langle T^{(p_1)}(1) T^{(p_2)}(2) T^{(p_3)}(3) \rangle \right]$$

$$- \sum_{p_1, p_2, p_3, p_4=1}^2 \int d1 \left[ L_{p_1} \cdot H(1 - 2) e^{-ik_I(1-2)} \cdot L_{p_2} \left[ \langle T^{(p_1)}(1) T^{(p_2)}(2) \rangle \right] \right] \cdot \langle L(2) \rangle^{-1}$$

$$\cdot \int d2 \left[ L_{p_3} \cdot H(2 - 3) e^{-ik_I(2-3)} \cdot L_{p_4} \left[ \langle T^{(p_3)}(2) T^{(p_4)}(3) \rangle \right] \right]. \quad (8.156)$$

Note that it is highly nontrivial to simplify the $n$-th order term because the inner products of $\langle L(1) \rangle^{-1}$ and other tensors are generally non-commutable.

**Remarks:**

1. In principle, the effective dielectric constant tensor $\epsilon_e$ in the full exact expansion (8.154) is independent of the choice of reference phase.
2. It is important to note that in the original local strong-contrast expansions [242] that applies to the quasistatic regime, the authors employed a local relation \( \langle P(1) \rangle = \langle S \rangle(k = 0) \cdot E_0(1) \), implying that electric polarization at one position is determined solely by the electric field at that position. Therefore, one would not expect the original strong-contrast approximations to provide good estimates for relatively large wavelengths. When the wavelength is comparable or smaller than the inhomogeneity length scales \( \ell \), however, the electric polarization \( P \) at one position \( 1 \) is determined by the electric field at a different position (nonlocality). Since the constitutive relation (8.142) takes such spatial dispersion effects into account [134, 1], the nonlocal strong-contrast approximation derived here gives more accurate estimates from long- to intermediate-wavelength regime.

### 8.15.4 Expansions for the spherical exclusion region

In this subsection, we derive the strong-contrast expansions that are presented in the main text. Specifically, we make two additional assumptions to simplify expansions (8.154) in Sec. 8.15.3: the reference phase is taken to be one phase \( q \) of the composite [i.e., \( I = q (= 1, 2) \)]; and the exclusion region is spherical. Then, we have simplified expressions for some tensor quantities:

\[
D^{(I)} \rightarrow D^{(q)} = \frac{1}{d\varepsilon_q} I, \tag{8.157}
\]

\[
\tilde{H}_{ij}^{(I)}(q) \rightarrow \frac{q^2 \delta_{ij} - q_i q_j + (d - 1) k_q^2 \delta_{ij}}{d\varepsilon_q(q^2 - k_q^2)}, \tag{8.158}
\]

\[
L_p^{(I)} \rightarrow \begin{cases} 
L^{(q)} \equiv d\varepsilon_q \beta_{pq} I, & p_1 \neq q, \\
0, & p_1 = q
\end{cases}, \tag{8.159}
\]

\[
\langle L^{(I)}(1) \rangle \rightarrow \phi_p L^{(q)} \ (q \neq p). \tag{8.160}
\]
Similarly, the nonlocal relation (8.151) is written as

$$\langle F \rangle(k_q) = [L_e(k_q)]^{-1} \cdot \langle P \rangle(k_q), \quad (8.161)$$

where

$$[L_e(k_q)]^{-1} = \langle S \rangle(k_q)^{-1} + \hat{H}(k_q)$$

$$= \left\{ [\varepsilon_e(k_q) - \varepsilon_q I] \cdot [\varepsilon_e(k_q) + (d-1)\varepsilon_q I]^{-1} \right\}^{-1}. \quad (8.162)$$

To solve Eq. (8.162), we first simply the expression for $\langle S \rangle(k_q)^{-1}$. Substituting Eqs. (8.158)-(8.160) to the Fourier transform of $\langle S \rangle(1 - 1')$ given in (8.145) leads to

$$\langle S \rangle(k_q) = \int d(1 - 1') e^{-i\mathbf{k}_q \cdot (1 - 1')} \left\{ [L(1) \delta(1 - 1') + L(1) H(1, 1') L(1')] + \int d^2 L(1) H(1, 2) L(2) H(2, 1') L(1') + \cdots \right\}$$

$$= \int d(1 - 1') e^{-i\mathbf{k}_q \cdot (1 - 1')} \left[ \phi_p (d\varepsilon_q \beta_{pq} I) \delta(1 - 1') + (d\varepsilon_q \beta_{pq})^2 H(1 - 1') S_2^{(p)}(1 - 1') \right.$$  

$$\left. + \int d^2 (d\varepsilon_q \beta_{pq})^3 H(1 - 2) \cdot H(2 - 1') \cdot S_3^{(p)}(1 - 1', 2 - 1') + \cdots \right]$$

$$= \phi_p (d\varepsilon_q \beta_{pq} I) + (d\varepsilon_q \beta_{pq})^2 \int dx_1 H(x_1 - x_2) e^{-i\mathbf{k}_q \cdot (x_1 - x_2)} S_2^{(p)}(x_1 - x_2)$$

$$+ (d\varepsilon_q \beta_{pq})^3 \int dx_1 dx_2 H(x_1 - x_2) e^{-i\mathbf{k}_q \cdot (x_1 - x_2)} \cdot H(x_2 - x_3) e^{-i\mathbf{k}_q \cdot (x_2 - x_3)}$$

$$\times S_3^{(p)}(x_1, x_2, x_3) + \sum_{n=4}^{\infty} (\mathbb{A}_n), \quad (8.163)$$

where we note that $S_n^{(p)}(x_1, \cdots, x_{n-1}, x_n) = S_n^{(p)}(x_1 - x_n, \cdots, x_{n-1} - x_n)$ due to statistical homogeneity, and

$$\mathbb{A}_n \equiv (d\varepsilon_q \beta_{pq})^n \int dx_1 \cdots dx_{n-1} H(x_1 - x_2) e^{-i\mathbf{k}_q \cdot (x_1 - x_2)} \cdots H(x_{n-1} - x_n) e^{-i\mathbf{k}_q \cdot (x_{n-1} - x_n)}$$

$$\times S_n^{(p)}(x_1, x_2, \cdots, x_n), \quad \text{for } n \geq 2. \quad (8.164)$$
An explicit expression for the nonlocal strong-contrast expansions is obtained by substituting Eq. (8.163) into Eq. (8.162) and by following the calculations analogous to Eqs. (8.153) and (8.154):

\[
\begin{align*}
(\phi_p \beta_{pq})^2 \left[ \frac{\varepsilon_e(k_q) - \varepsilon_q I}{\varepsilon_e(k_q) + (d-1)\varepsilon_q I} \right]^{-1} \\
- \frac{(\phi_p d\varepsilon_q \beta_{pq})^2}{d\varepsilon_q} (\phi_p d\varepsilon_q \beta_{pq})^{-1} \left\{ I - \left[ \mathbb{A}_2 + \mathbb{A}_3 + \cdots \right] (\phi_p d\varepsilon_q \beta_{pq})^{-1} \\
+ \left[ \mathbb{A}_2 + \mathbb{A}_3 + \cdots \right] (\phi_p d\varepsilon_q \beta_{pq})^{-2} - \left[ \mathbb{A}_2 + \mathbb{A}_3 + \cdots \right]^3 (\phi_p d\varepsilon_q \beta_{pq})^{-3} + \cdots \right\} \\
+ \frac{(\phi_p d\varepsilon_q \beta_{pq})^2}{d\varepsilon_q} \int dx_1 H(x_1) e^{-ik_q \cdot x_1}
\right) = \phi_p \beta_{pq} I - \frac{1}{d\varepsilon_q} \left[ \mathbb{A}_2 - (d\varepsilon_q \beta_{pq})^2 \phi_p^2 \int d\varepsilon_q \int dx_1 H(r_1) e^{-ik_q \cdot x_1} \right] - \frac{1}{d\varepsilon_q} \left[ \mathbb{A}_3 - \mathbb{A}_2 \mathbb{A}_2 (\phi_p d\varepsilon_q \beta_{pq})^{-1} \right] \\
- \frac{1}{d\varepsilon_q} \left[ \mathbb{A}_4 - (\mathbb{A}_2 \mathbb{A}_3 + \mathbb{A}_3 \mathbb{A}_2) (\phi_p d\varepsilon_q \beta_{pq})^{-1} + \mathbb{A}_2 \mathbb{A}_2 \mathbb{A}_2 (\phi_p d\varepsilon_q \beta_{pq})^{-2} \right] + \cdots.
\end{align*}
\]

(8.165)

Substituting the definitions of \( \mathbb{A}_n \) given in Eq. (8.164) into the above expression leads to

\[
(\phi_p \beta_{pq})^2 \left[ \frac{\varepsilon_e(k_q) - \varepsilon_q I}{\varepsilon_e(k_q) + (d-1)\varepsilon_q I} \right]^{-1} = \phi_p \beta_{pq} I - \sum_{n=2}^{\infty} A_n^{(p)}(k_q) \beta_{pq}^n,
\]

which is identical to the series given in Eq. (8.54).

8.16 Appendix F: Properties of Nonlocal Attenuation Function \( F(Q) \)

For macroscopically isotropic media in \( \mathbb{R}^d \), nonlocal attenuation function \( F(Q) \) is the key microstructure-dependent parameter in the strong-contrast approximation, which
is related to the second-rank tensor \( A_{2}^{(p)}(k_q) \) given in Eq. (8.55):

\[
A_{2}^{(p)}(k_q) \equiv \frac{1}{d} \text{Tr} \left[ A_{2}^{(p)} (k_q) \right] = -\frac{(d-1)\pi}{2^{d/2} \Gamma(d/2)} F(k_q). \tag{8.166}
\]

For a statistically anisotropic two-phase medium, the nonlocal attenuation function is explicitly given in Eqs. (8.71) and (8.72). Note that (8.72) is the Fourier representation of Eq. (8.71), which can be immediately obtained from the Parseval theorem by utilizing the fact that 1/\[q^2 - Q^2\] is the radial Fourier transform of \((i/4)[Q/(2\pi r)]^{d/2-1} H_{d/2-1}^{(1)}(Qr)\).

For statistically isotropic media, the nonlocal attenuation function depends on wavenumber \(Q\), instead of wavevector \(Q\). Its imaginary and real parts can be simplified as Eqs. (8.73) and (8.74), respectively.

In this section, we derive and present some important properties of the nonlocal attenuation function. We first derive \(F(Q)\) for hypercubic lattice packings that are macroscopically isotropic but statistically anisotropic in Sec. 8.16.1. We then investigate general properties of \(F(Q)\) for statistically isotropic media. Specifically, in Sec. 8.16.2-8.16.4, we derive some of its analytic properties, its alternative formulas [Eqs. (8.73) and (8.74)], and its asymptotic expressions, respectively.

### 8.16.1 Formulas for Hypercubic Lattice Packings

For a hypercubic lattice \((Z^d)\) packing in \(\mathbb{R}^d\), its effective dielectric constant tensor possesses the cubic symmetry but has \(k_q\)-dependence because \(Z^d\) is statistically anisotropic, i.e., \(\epsilon_e(k_q) = \epsilon_e(k_q) I\). In this section, we derive the analytic expression of \(\epsilon_e(k_q)\) for \(Z^d\) packings \((d = 2, 3)\) using the general definition (8.72) of the attenuation function. For concreteness, we take the matrix and reference phases as phase 1, and the particle phase as phase 2.

We first obtain the attenuation function of \(Z^d\) sphere packings. For a \(Z^d\) sphere
packing of packing fraction $\phi_2$ and lattice constant $L$, its spectral density is written as

$$\tilde{\chi}_V(Q) = \phi_2 \tilde{\alpha}_2(Q; a) S(Q) = (2\pi)^d \rho \phi_2 \tilde{\alpha}_2(Q; a) \sum_{G \in \mathbb{Z}^d^* \setminus 0} \delta(Q - G/L), \quad (8.167)$$

where $\rho = L^{-d}$ is number density, $a$ is particle radius, $\mathbb{Z}^d^*$ is the reciprocal lattice of the unit $\mathbb{Z}^d$ lattice, $\tilde{\alpha}_2(Q; a)$ is given in Eq. (8.11), and $S(Q)$ is the structure factor of a hypercubic lattice. Applying Eq. (8.167) to Eq. (8.72) yields

$$F(Q) = -\frac{2^{d/2} \Gamma(d/2)}{\pi} \rho^2 (QL)^2 \sum_{G \in \mathbb{Z}^d^* \setminus 0} \tilde{m}(|G|/L; a)^2 \frac{G \cdot (G + 2QL)}{G \cdot (G + 2QL)}, \quad (8.168)$$

if the Laue condition of $L\mathbb{Z}^d$ lattice is unmet [i.e., $G \cdot (G + 2QL) \neq 0$]. Here, $\mathbb{Z}^d^*$ stands for the reciprocal lattice of $\mathbb{Z}^d$. Note that Eq. (8.168) is real-valued.

Using Eq. (8.168), we obtain two-point approximation of $\varepsilon_e(k_1)$ for a hypercubic lattice packing:

$$\frac{\varepsilon_e(k_1)}{\varepsilon_1} = 1 + d\phi_2^2 \beta_{21} \left[ \phi_2(1 - \phi_2\beta_{21}) + (d - 1)(\rho k_1 L)^2 \beta_{21} \sum_{G \in \mathbb{Z}^d^* \setminus 0} \tilde{m}(|G|/L; a)^2 \frac{G \cdot (G + 2k_1 L)}{G \cdot (G + 2k_1 L)} \right]^{-1}, \quad (8.169)$$

where $\beta_{21}$ is defined in Eq. (8.38). We use Eq. (8.169) with $k_1 = k_1 \hat{x}$ to estimate $\varepsilon_e$ of periodic packings along the $\Gamma$-$X$ direction (see Sec. VIII of the main text).

### 8.16.2 Analytic Properties

We now show that $F(Q)$ has the following analytic properties for statistically isotropic media in $\mathbb{R}^d$.

1. $F(Q)$ is an analytic function of a complex variable $Q$ in the *upper half-plane*, i.e., $\text{Im}[Q] \geq 0$;
2. Re\([F(Q)]\) and Im\([F(Q)]\) are even and odd functions of real variable \(Q\), respectively.

To do so, we start by rewriting Eq. (8.71) as follows:

\[
F(Q) = -i2^{(d-2)/2} \Gamma(d/2) Q^2 \int_0^\infty r \mathcal{H}_{d/2-1}^{(1)}(Qr) J_{d/2-1}(Qr) \chi_v(r) \, dr
= -i2^{(d-2)/2} \Gamma(d/2) \int_0^{\text{sgn}(Q)\infty} x \mathcal{H}_{d/2-1}^{(1)}(x) J_{d/2-1}(x) \chi_v(x/Q) \, dx ,
\]

(8.170)

where \(\text{sgn}(Q)\) is the sign of a real number \(Q\), and we have used the formula for the radial Fourier transform in \(\mathbb{R}^d\)

\[
\tilde{f}(Q) = \int f(r) e^{-iQ \cdot r} \, dr = (2\pi)^{d/2} \int_0^\infty r^{d-1} f(r) \frac{J_{(d-2)/2}(Qr)}{(Qr)^{(d-2)/2}} \, dr .
\]

Property 1 follows from the observation that, if Eq. (8.170) is regarded as a function in the complex \(Q\) plane, this integral is well-defined for \(\text{Im}[Q] \geq 0\) because as \(|x|\) increases at a constant angle \(\arg x\) (or, equivalently, as \(r\) goes to infinity with \(Q\) fixed),

\[
x \mathcal{H}_{d/2-1}^{(1)}(x) J_{d/2-1}(x) \sim \begin{cases} 
\frac{1}{\pi} e^{2i[x-(d-1)\pi/4]}, & -\pi < \arg x < 0 \\
\frac{1}{\pi}, & 0 < \arg x < \pi \end{cases}
\]

as \(|x| \to \infty\).

Property 2 immediately follows from the fact that the real and imaginary parts of \(ix \mathcal{H}_{d/2-1}^{(1)}(x) J_{d/2-1}(x)\) are even and odd function of \(x\), respectively, for \(d = 2, 3\).

### 8.16.3 Alternative Formulas

In this subsection, we derive Eqs. (8.73) and (8.74) from a general Fourier-space representation of \(F(Q)\) given in Eq. (8.72). In three dimensions, without loss of
generality, we can take $Q = Q \hat{z}$. We write Eq. (8.72) in spherical coordinates:

$$\int dq \frac{\tilde{\chi}_V(q)}{|q + Q \hat{z}^2|} = \int_0^\infty dq q^2 \int_0^\pi d\theta \sin \theta \left[ \int_0^{2\pi} d\phi \right] \frac{\tilde{\chi}_V(q)}{q(q + 2Q \cos \theta)}$$

$$= 2\pi \int_{-1}^1 dx \int_0^\infty dq \frac{q \tilde{\chi}_V(q)}{q + 2Qx}$$

$$= \pi \left[ \int_{-1}^1 dx \int_0^\infty dq \frac{q \tilde{\chi}_V(q)}{q + 2Qx} + \int_{-\infty}^{-1} (-1) dx' \int_{0}^{-\infty} dq' (-1) \frac{-q' \tilde{\chi}_V(-q')}{q' - 2Qx'} \right]$$

$$= \pi \int_{-1}^1 dx \int_{-\infty}^\infty dq \frac{q \tilde{\chi}_V(q)}{q + 2Qx},$$

where we use $\tilde{\chi}_V(q) = \tilde{\chi}_V(-q)$ to obtain Eq. (8.172). Since $F(Q)$ is analytic for $\text{Im}[Q] \geq 0$ (see Sec. 8.16.2), applying the following identity

$$\int_{-\infty}^\infty dx \frac{f(x)}{x - x'} = \lim_{\epsilon \to 0^+} \int_{-\infty}^\infty dx \frac{f(x)}{x - (x' \pm i\epsilon)} = \text{p.v.} \int_{-\infty}^\infty dx \frac{f(x)}{x - x'} \pm i\pi f(x')$$

(8.173)

to Eq. (8.172) yields

$$F(Q) = -\frac{Q^2}{(2\pi)^{5/2}} \int_{-1}^1 dx \int_{-\infty}^\infty dq \frac{q \tilde{\chi}_V(q)}{q + 2x(Q + i\epsilon)}$$

$$= -\frac{Q^2}{(2\pi)^{5/2}} \int_{-1}^1 dx \left[ \text{p.v.} \int_{-\infty}^\infty dq \frac{q \tilde{\chi}_V(q)}{q + 2xQ} + i\pi \text{sgn}(x) (2Qx) \tilde{\chi}_V(2Qx) \right]$$

$$= -\frac{Q^2}{(2\pi)^{5/2}} \int_{-1}^1 dx \left[ \text{p.v.} \int_{-\infty}^\infty dq \frac{q \tilde{\chi}_V(q)}{q + 2xQ} + i\pi 2Q|x| \tilde{\chi}_V(2Qx) \right]$$

$$= -\frac{4Q^2}{(2\pi)^{5/2}} \int_0^1 dx \left[ \text{p.v.} \int_0^\infty dq \frac{q^2 \tilde{\chi}_V(q)}{q^2 - (2xQ)^2} \right] - \frac{iQ}{2(2\pi)^{3/2}} \int_0^{2Q} dq' q' \tilde{\chi}_V(q'),$$

(8.174)

where $\text{p.v.}$ stands for the Cauchy principal value of an improper integral. Since each integral in Eq. (8.174) is real-valued, the real and imaginary parts of $F(Q)$ can be
written as

\[
\text{Re}[F(Q)] = -\frac{4Q^2}{(2\pi)^{5/2}} \int_0^1 dx \left[ \text{p.v.} \int_0^\infty dq \frac{q^2 \tilde{\chi}_\nu(q)}{q^2 - (2qx)^2} \right], \quad (8.175)
\]

\[
\text{Im}[F(Q)] = -\frac{Q}{2(2\pi)^{3/2}} \int_0^{2Q} dq \ q \tilde{\chi}_\nu(q), \quad (8.176)
\]

respectively.

When computing \( F(Q) \) from the isotropic spectral density obtained from experiments or numerical simulations, it is easy to compute the imaginary part given in Eq. (8.176) but difficult to compute the real part (8.175). For this reason, we derive an alternative formula for the real part [Eq. (8.175)]. We first interchange the order of integrals in Eq. (8.175), change the integration variables such that \( q \rightarrow 2q'x \) and \( x \rightarrow q''/2q' \), and replace the integral over \( q'' \) with Eq. (8.176) to obtain

\[
\text{Re}[F(Q)] = -\frac{4Q^2}{(2\pi)^{5/2}} \text{p.v.} \int_0^\infty dq \int_0^1 dx \frac{q^2 \tilde{\chi}_\nu(q)}{q^2 - (2Qx)^2} \]
\[
= -\frac{2Q^2}{(2\pi)^{5/2}} \text{p.v.} \int_0^\infty dq' \frac{1}{q'^2 - Q^2} \int_0^{2Q'} dq'' q'' \tilde{\chi}_\nu(q'') \]
\[
= -\frac{2Q^2}{\pi} \text{p.v.} \int_0^\infty dq \frac{\text{Im}[F(q)]}{q(Q^2 - q^2)}, \quad (8.177)
\]

Thus, we have derived Eqs. (8.73) and (8.74) for three dimensions.

In two dimensions, we rewrite the integral in Eq. (8.72) in cylindrical coordinates:

\[
\int dq \frac{\tilde{\chi}_\nu(q)}{|q + Q| - Q^2} = \int_0^{2\pi} d\phi \int_0^\infty dq \frac{\tilde{\chi}_\nu(q)}{q + 2Q \cos \phi} \]
\[
= 2 \int_0^{\pi/2} d\phi \int_0^\infty dq \frac{\tilde{\chi}_\nu(q)}{q + 2Q \cos \phi} = 2 \int_0^{\pi/2} d\phi \int_0^\infty dq \left[ \frac{1}{q + 2Q \cos \phi} + \frac{1}{q - 2Q \cos \phi} \right] \tilde{\chi}_\nu(q) \]
\[
= 2 \int_0^{\pi/2} d\phi \int_{-\infty}^{\infty} dq \left[ \frac{1}{q + 2(Q + i\epsilon) \cos \phi} + \frac{1}{q - 2(Q + i\epsilon) \cos \phi} \right] \\Theta(q) \tilde{\chi}_\nu(q), \quad (8.178)
\]
where $\epsilon$ is an infinitesimal positive number, and $\Theta(x) \equiv \left\{ \begin{array}{ll} 1, & x > 0 \\ 0, & \text{otherwise} \end{array} \right.$ is Heaviside step function. Applying the identity (8.173) to the integral over $q$ in Eq. (8.178) gives

$$
\int_{-\infty}^{\infty} dq \left[ \frac{1}{q + 2(Q + i\epsilon) \cos \phi} + \frac{1}{q - 2(Q + i\epsilon) \cos \phi} \right] \Theta(q) \tilde{\chi}_V(q) = \text{p.v.} \int_{0}^{\infty} dq \frac{2q \tilde{\chi}_V(q)}{q^2 - (2Q \cos \phi)^2} + i\pi \tilde{\chi}_V(2Q \cos \phi). \tag{8.179}
$$

Combining Eq. (8.179) to Eqs. (8.178) and (8.72), we obtain

$$
\text{Re}[F(Q)] = -\frac{Q^2}{\pi^3} \int_{0}^{\pi/2} d\phi \left[ \text{p.v.} \int_{0}^{\infty} dq \frac{2q \tilde{\chi}_V(q)}{q^2 - (2Q \cos \phi)^2} \right], \tag{8.180}
$$

$$
\text{Im}[F(Q)] = -\frac{Q^2}{\pi^2} \int_{0}^{\pi/2} d\phi \tilde{\chi}_V(2Q \cos \phi). \tag{8.181}
$$

As we done in three dimensions, we derive an alternative expression for Eq. (8.180). We first interchange the order of integrals, change variables of integration $q \rightarrow 2q' \cos \phi$, and then substitute the integral over $\phi$ with Eq. (8.181):

$$
\text{Re}[F(Q)] = -\frac{Q^2}{\pi^3} \text{p.v.} \int_{0}^{\infty} dq \int_{0}^{\pi/2} d\phi \frac{2q \tilde{\chi}_V(q)}{q^2 - (2Q \cos \phi)^2} \tag{8.182}
$$

$$
= -\frac{2Q^2}{\pi^3} \text{p.v.} \int_{0}^{\infty} dq' \frac{q'}{q'^2 - Q^2} \int_{0}^{\pi/2} d\phi \tilde{\chi}_V(2q' \cos \phi) \tag{8.183}
$$

$$
= -\frac{2Q^2}{\pi} \text{p.v.} \int_{0}^{\infty} dq \frac{\text{Im}[F(q)]}{q(Q^2 - q^2)}. \tag{8.184}
$$

Thus, we have derived Eqs. (8.73) and (8.74) for two dimensions.

Remarks:

1. Similar to the Kramers-Kronig relations, the expression (8.74) is in fact a natural consequence of the analytic properties of $F(Q)$.

2. In practice, it is nontrivial to numerically compute the Cauchy principal value in Eq. (8.74). To avoid such problems, we change this integral into a more
numerically tractable form by utilizing the fact that \( \text{Im}[F(Q)] \sim Q \) as \( Q \to \infty \) (see Sec. 8.16.4):

\[
\text{Re}[F(Q)] \approx -\frac{2Q^2}{\pi} \left[ \text{p.v.} \int_0^M \frac{1}{q(Q^2 - q^2)} \text{Im}[F(q)] \, dq + \frac{\text{Im}[F(M)]}{M} \int_M^\infty \frac{1}{Q^2 - q^2} \, dq \right]
\]

\[
= -\frac{2Q}{\pi} \left( \int_0^M \frac{\text{Im}[F(q)]}{(Q + q)q} \, dq + \int_0^M \frac{\text{Im}[F(q)] - \text{Im}[F(Q)]}{Q^2 - q^2} \, dq \right)
\]

\[
+ \frac{1}{2Q} \left\{ \text{Im}[F(Q)] - \frac{Q}{M} \text{Im}[F(M)] \right\} \ln \left| \frac{M + Q}{M - Q} \right|,
\]

(8.185)

where \( M \) is an upper limit of the integral.

### 8.16.4 Asymptotic Behaviors

In this subsection, we derive general asymptotic expressions for \( F(Q) \) in the quasistatic (small-\( Q \)) and large-\( Q \) regimes. The reader is referred to Eqs. (83) and (84) in the main text for the small-\( Q \) behaviors of \( \text{Im}[F(Q)] \) and \( \text{Re}[F(Q)] \), respectively. The large-\( Q \) asymptotic behaviors are

\[
\text{Im}[F(Q)] \sim Q + \mathcal{O}(Q^{-1}),
\]

(8.186)

\[
\text{Re}[F(Q)] \sim \text{const.}(< 0),
\]

(8.187)

regardless of the functional form of the spectral density.

The small-\( Q \) expressions are easy to derive. For Eq. (84) in the main text, it can be derived from Eq. (8.170):

\[
\text{Re}[F(Q)] = 2^{(d-2)/2} \Gamma(d/2) Q^2 \int_0^\infty r Y_{d/2-1}(Qr) J_{d/2-1}(Qr) \chi_r(r) \, dr
\]

\[
= 2^{(d-2)/2} \Gamma(d/2) Q^2 \int_0^\infty \left[ -\frac{r \Gamma(d/2 - 1)}{\pi \Gamma(d/2)} + \mathcal{O}(Q^{d-2}r^{d-1}) \right] \chi_r(r) \, dr
\]

\[
\sim -Q^2 \int_0^\infty r \chi_r(r) \, dr \sim -Q^2, \text{ as } Q \to 0^+.
\]
The small-$Q$ behavior of $\text{Im}[\varepsilon_e(Q)]$, given in Eq. (83) in the main text, can be easily derived from Eq. (8.73).

The large-$Q$ behaviors are slightly more complicated to derive. To show Eq. (8.186), we note that $\text{Im}[F(Q)]/Q$ given in Eq. (8.73) is identical to the integral of $\tilde{\chi}_V(q)$ on a $d$-dimensional spherical surface of radius $Q$ centered at $q = Q\hat{x}$ up to a proportional constant. Furthermore, for any $Q > 0$, this integral is bounded above by the volume integral of a non-negative function $\tilde{\chi}_V(q)$ that converges to a finite value $(2\pi)^d\chi_V(0)$, i.e.,

$$
\left| \frac{\text{Im}[F(Q)]}{Q} \right| \propto \int_{|q-Q\hat{x}|=Q} dq \tilde{\chi}_V(q) < \int_{\mathbb{R}^d} dq \tilde{\chi}_V(q) = (2\pi)^d\chi_V(0).
$$

These two observations lead us to conclude that $\text{Im}[F(Q)]/Q$ converges to a negative constant as $Q$ goes to infinity, which is identical to Eq. (8.186). A simple asymptotic analysis of Eqs. (8.74) and (8.186) yields Eq. (8.187).

### 8.17 Appendix G: Comparisons of Local and Nonlocal Attenuation Functions

Here we compare the behaviors of the “local” attenuation function $F(Q)$ derived in Ref. [242] for the quasistatic strong-contrast to its nonlocal counterpart $F(Q)$ [cf. (8.71) or (8.72)] derived in this chapter. Both the local and nonlocal attenuation functions depend on the spectral density but their functional behaviors are generally different across wavenumbers. We also provide plots of both the real and imaginary parts of the nonlocal attenuation function $F(Q)$ for the five models of disordered two-phase media considered in this chapter.

The local attenuation $F(Q)$ is defined in Eqs. (8.99) and (8.100). In contrast to the nonlocal attenuation function $F(Q)$, the local attenuation function only depends
on the wavenumber $Q$, instead of the wavevector $Q$. For statistically isotropic media, its real and imaginary parts are given in Eqs. (8.107) and (8.105), respectively.

As expected, the local and nonlocal attenuation functions, $\mathcal{F}(Q)$ and $F(Q)$, are identical in the quasistatic (small-$Q$) regime. Specifically, the small-$Q$ behaviors of $\text{Im}[\mathcal{F}(Q)]$ and $\text{Im}[F(Q)]$ are given in Eq. (8.109) and Eq. (83) in the main text. The small-$Q$ behaviors of $\text{Re}[\mathcal{F}(Q)]$ and $\text{Re}[F(Q)]$ are given in Eq. (8.110) and Eq. (84) in the main text. Beyond the quasistatic regime, however, both types of attenuation functions become increasingly different as the wavenumber $k_q$ increases, with concomittant distinctly different attenuation characteristics. In the large-$Q$ regime, the local and nonlocal attenuation functions exhibit considerably different behaviors:

\[
\text{Im}[\mathcal{F}(Q)] \sim Q^{-1}, \quad \text{Re}[\mathcal{F}(Q)] \to \frac{2^{d/2} \Gamma(d/2)}{\pi} \phi_p (1 - \phi_p) (> 0), \quad \text{as } Q \to \infty \quad (8.188)
\]

\[
\text{Im}[F(Q)] \sim Q, \quad \text{Re}[F(Q)] \to \text{const.} \quad (< 0), \quad \text{as } Q \to \infty. \quad (8.189)
\]

The reader is referred to Secs. 8.14.2 and 8.16 for derivations.

![Figure 8.14: Comparison of negatives of (a) the real and (b) imaginary parts of the nonlocal attenuation function $F(Q)$ [see Eq. (8.170)] and its local counterpart $\mathcal{F}(Q)$ [see Eq. (8.99)] for 3D disordered models. We consider (left panel) stealthy hyperuniform packings and (right panel) stealthy nonhyperuniform packings of packing fraction $\phi_2 = 0.25$ and identical spheres of radius $a$.](image)

We compare both attenuation functions $\mathcal{F}(Q)$ and $F(Q)$ for stealthy hyperuniform packings; see Fig. 8.14 for 3D cases. While these two functions are identical
in the quasistatic regime, they become increasingly different from one another as the wavenumber increases. For example, Eqs. (8.73) and (8.105) immediately show that for stealthy hyperuniform systems [i.e., \( \tilde{\chi}_V(Q) = 0 \) for \( Q < Q_U \)], the imaginary parts of these two attenuation functions are identically zero (transparent or lossless) up to finite but quite different range of wavenumbers; specifically,

\[
\begin{align*}
\text{Im}[F(Q)] &= 0, \quad \text{where } Q < Q_U/2, \\
\text{Im}[F(Q)] &= 0, \quad \text{where } Q < Q_U.
\end{align*}
\] (8.190)

We also present a plot comparing both local and nonlocal attenuation functions [Eqs. (8.99) and (8.170), respectively] for stealthy nonhyperuniform dispersions [i.e., \( \tilde{\chi}_V(Q) = 0 \) for \( 0 < Q_L < Q < Q_U \)]. Specifically, we consider three-dimensional stealthy nonhyperuniform sphere packings of radius \( a \) and packing fraction \( \phi_2 = 0.25 \), and they are stealthy in the region of \( 1.0 < Qa < 1.5 \). As shown in Fig. 8.14, both attenuation functions are identical only in the quasistatic regime, as observed in the stealthy hyperuniform packings. Beyond that regime, importantly, the local attenuation incorrectly predicts that these media are transparent for \( 1 < k_qa < 1.5 \), the nonlocal attenuation function successfully captures the fact that they are not transparent.

8.18 Appendix H: Comparisons of the Strong-Contrast Approximation with the Popular Effective-Medium Approximations

We compare the asymptotic behaviors of the “unmodified” strong-contrast approximation to those of the popular effective-medium approximations [i.e., the Maxwell-Garnett approximation (MGA) and the quasicrystalline approximation (QCA)] that
are employed in Sec. III in the main text. For simplicity, we consider statistically isotropic sphere packings of sphere radius $a$ in three dimensions, where the matrix (reference) phase is taken as phase 1. We analytically demonstrate that in the quasistatic (small-$k_1$) and dilute regimes, MGA and QCA become consistent with the strong-contrast approximation.

We first consider the expressions in the quasistatic regime for these three approximations. For the strong-contrast approximation, by taking the Taylor expansions of Eqs. (8.73) and (8.74) in terms of $Q$, we obtain

$$F(Q) = -\frac{1}{\sqrt{2\pi}} \left[ 2 \int_0^\infty r \chi_v(r) \, dr \, Q^2 + \frac{i}{2\pi} \tilde{\chi}_v(0) Q^3 \right] + O(Q^4). \quad (8.191)$$

Substituting Eq. (8.191) and the identity $\tilde{\chi}_v(0) = (4\pi a^3/3)\phi_2 S(0)$ into the strong-contrast approximation gives the following small-$k_1$ asymptotic expression:

$$\epsilon_e(k_1)/\epsilon_1 = 1 + \frac{3\phi_2 \beta_{21}}{1 - \phi_2 \beta_{21}} + 3\phi_2 \left( \frac{\beta_{21}}{1 - \phi_2 \beta_{21}} \right)^2 \times \left[ \frac{2}{\phi_2} \int_0^\infty r \chi_v(r) \, dr \, k_1^2 + i \frac{2}{3} S(0) (k_1 a)^3 \right] + O(k_1^4), \quad (8.192)$$

where $\beta_{21}$ is given in Eq. (8.38). The analogous expressions for the MGA and QCA are

$$\epsilon_e(k_1)/\epsilon_1 = 1 + \frac{3\phi_2 \beta_{21}}{1 - \phi_2 \beta_{21}} + 3\phi_2 \beta_{21}^2 \left[ -\frac{1 - 4\beta_{21}}{5} (k_1 a)^2 + \frac{2}{3} i (k_1 a)^3 \right] + O(k_1^4), \quad (8.193)$$

and

$$\epsilon_e(k_1)/\epsilon_1 = 1 + \frac{3\phi_2 \beta_{21}}{1 - \phi_2 \beta_{21}} + 3\phi_2 \left( \frac{\beta_{21}}{1 - \phi_2 \beta_{21}} \right)^2 S(0) \times i \frac{2}{3} (k_1 a)^3, \quad (8.194)$$

respectively. We note that the static limits of these three approximations are identical.
to the Hashin-Shtrikman estimate $\varepsilon_{\text{HS}}$ given in Eq. (8.75). Furthermore, the leading-order terms of $\text{Im}[\varepsilon_e(k_1)]$ given in the strong-contrast approximation (8.192) and the QCA (8.194) are identical.

We now further assume the dilute limits (i.e., $\phi_2 \to 0$) of Eqs. (8.192), (8.193), and (8.194). For typical disordered packings in the dilute regime, $S(Q) \approx 1$, and thus $\tilde{\chi}_v(Q) \approx \phi_2 \tilde{\alpha}_2(Q,a)$; see Sec. IIC in the main text. Combining this approximation and Eq. (8.72) gives

$$F(Q) = -\frac{3\phi_2}{128\sqrt{2\pi}(Qa)^3} \left\{ \frac{1}{3}[64(Qa)^3 + 12Qa\cos(4Qa) - 3\sin(4Qa)] + i[32(Qa)^4 - 8(Qa)^2 - 1 + \cos(4Qa) + 4Qa\sin(4Qa)] \right\}$$

$$= -\frac{\phi_2}{\sqrt{2\pi}} \left[ \frac{4}{5}(Qa)^2 + i\frac{2}{3}(Qa)^3 \right] + O(Q^4). \quad (8.195)$$

Thus, the leading-order expressions in $\phi_2$ of the strong-contrast approximation (8.192) is

$$\frac{\varepsilon_e(k_1)}{\varepsilon_1} = 1 + 3\phi_2\beta_{21} + 3\phi_2^2\tilde{\alpha}_{21} \left[ \frac{4}{5}(k_1a)^2 + \frac{2}{3}i(k_1a)^3 \right] + O(\phi_2^2). \quad (8.196)$$

In the same regime, the analogous expressions for the MGA [Eq. (8.193)] and the QCA [Eq. (8.194)] are given as

$$\varepsilon_e(k_1)/\varepsilon_1 = 1 + 3\phi_2\beta_{21} + 3\phi_2^2\beta_{21}^2 \left[ -\frac{1}{5}(k_1a)^2 + \frac{2}{3}i(k_1a)^3 \right] + O(\phi_2^2), \quad (8.197)$$

$$\varepsilon_e(k_1)/\varepsilon_1 = 1 + 3\phi_2\beta_{21} + 3\phi_2^2\beta_{21}^2 \times i\frac{2}{3}(k_1a)^3 + O(\phi_2^2), \quad (8.198)$$

respectively. Unlike Eqs. (8.192)-(8.194) in the quasistatic regime, the leading-order terms of $\text{Im}[\varepsilon_e(k_1)]$ given in Eqs. (8.196), (8.197), and (8.198) are identical. This difference arises from the fact that the MGA neglects the spatial correlations of particles
and is consequently operative in the dilute regime.

8.19 Appendix I: Simulation Details

Here we provide additional details about simulation procedures and values of the simulation parameters that we used. We list parameters employed to generate sphere packings for computing the spectral density and the attenuation functions in Sec. 8.19.1. We list the parameters employed in the finite-difference time-domain (FDTD) simulations in Sec. 8.19.2. In Sec. 8.19.3, we describe the numerical homogenization estimates obtained by FDTD simulations. In Sec. 8.19.4, we confirm the validity of the aforementioned scheme.

8.19.1 Parameters for Numerically Generated Packings

Here, we list the parameters employed in generating sphere packings numerically. These packings have been used to compute the spectral density (Table 8.1) and carry out FDTD simulations (Table 8.2).

We numerically generate packings for disordered stealthy hyperuniform/nonhyperuniform packings for \( d = 2, 3 \), equilibrium packings for \( d = 2 \), and hyperuniform polydisperse packings for \( d = 2, 3 \) to compute the spectral density \( \tilde{\chi}_V(Q) \); see Sec. III in the main text. For each model, we generate \( N_c \) different packings of particle radius \( a \), \( N \) particles, and number density \( \rho \) in a periodic fundamental cell. Here, we list these parameters as well as some other relevant parameters.

Stealthy hyperuniform/nonhyperuniform packings are generated via the collective-coordinate optimization technique. For these models, parameters \( Q_L \) and \( Q_U \) define the stealthy regions, and \( \sigma \) represents the diameter of the repulsion region of each particle. For stealthy packings (or point patterns), it is useful to define the \( \chi \) parameter, which the ratio of constrained degrees of freedom to total number of degrees of freedom.
freedom [317, 352], i.e.,
\[
\chi \equiv \frac{\mathcal{M}}{d(N - 1)}.
\] (8.199)

For \(0 < \chi < 1/2\), they are highly degenerate and disordered, whereas for \(1/2 < \chi < 1\) they crystallize [352].

Equilibrium packings for \(d = 2\) are generated via Monte Carlo simulations. We obtain hyperuniform polydisperse packings by applying the tessellation-based procedure [157, 156] to equilibrium packings of \(\phi_2 = 0.45\). Simulation parameters employed to generate these systems are listed in Table 8.1.

Table 8.1: Parameters of disk/sphere packings used to compute \(\tilde{\chi}_V(Q)\). We generate realizations of disordered stealthy hyperuniform packings (SHP) and disordered nonhyperuniform packings (SNHP), equilibrium packings (EP), and hyperuniform polydisperse packings (HPP). For each model, \(N_c\) is the number of distinct packings, \(N\) particle number, \(a\) is particle radius, \(\rho\) is the number density, and \(\phi_2\) is the packing fraction. For hyperuniform polydisperse packings, \(a\) stands for the mean particle radius, i.e., \(a \equiv [\phi_2/v_1(1)]^{1/d}\). Quantities \(Q_L, Q_U,\) and \(\sigma\) are parameters used in the collective-coordinate optimization method; see Sec. III in the main text. The \(\chi\) parameter is defined in Eq. (8.199).

<table>
<thead>
<tr>
<th>Systems</th>
<th>Parameters</th>
<th>(N)</th>
<th>(\rho)</th>
<th>(N_c)</th>
<th>((Q_L a, Q_U a))</th>
<th>(\sigma)</th>
<th>(a)</th>
<th>(\chi)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2D SHP ((\phi_2 = 0.25))</td>
<td>1000</td>
<td>1</td>
<td>300</td>
<td>(0, 1.3)</td>
<td>0.57</td>
<td>0.2801</td>
<td>0.4214</td>
<td></td>
</tr>
<tr>
<td>3D SHP ((\phi_2 = 0.25))</td>
<td>1000</td>
<td>1</td>
<td>300</td>
<td>(0, 1.5)</td>
<td>0.8</td>
<td>0.3903</td>
<td>0.1582</td>
<td></td>
</tr>
<tr>
<td>3D SHP ((\phi_2 = 0.4))</td>
<td>1000</td>
<td>1</td>
<td>300</td>
<td>(0, 1.5)</td>
<td>0.92</td>
<td>0.4571</td>
<td>0.1031</td>
<td></td>
</tr>
<tr>
<td>2D SNHP ((\phi_2 = 0.25))</td>
<td>1000</td>
<td>1</td>
<td>300</td>
<td>(0.9, 1.3)</td>
<td>0.57</td>
<td>0.2801</td>
<td>0.2202</td>
<td></td>
</tr>
<tr>
<td>3D SNHP ((\phi_2 = 0.25))</td>
<td>1000</td>
<td>1</td>
<td>300</td>
<td>(1.0, 1.3)</td>
<td>0.8</td>
<td>0.3903</td>
<td>0.1154</td>
<td></td>
</tr>
<tr>
<td>2D EP ((\phi_2 = 0.25))</td>
<td>1000</td>
<td>1</td>
<td>300</td>
<td>-</td>
<td>-</td>
<td>0.2801</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>2D HPP ((\phi_2 = 0.25))</td>
<td>1000</td>
<td>1</td>
<td>300</td>
<td>-</td>
<td>-</td>
<td>0.2801</td>
<td>-</td>
<td></td>
</tr>
<tr>
<td>3D HPP ((\phi_2 = 0.25))</td>
<td>1000</td>
<td>1</td>
<td>300</td>
<td>-</td>
<td>-</td>
<td>0.3903</td>
<td>-</td>
<td></td>
</tr>
</tbody>
</table>

Table 8.2 lists the parameters to generate packings employed in FDTD simulations. We consider periodic packings (square and simple-cubic lattice packings) and some disordered packings (equilibrium packing and stealthy hyperuniform packings) of packing fraction \(\phi_2 = 0.25\) in two and three dimensions. For each model, we generate \(N_c\) distinct packings of unit number density (\(\rho = 1\)) and \(N\) particles.
Table 8.2: Parameters of disk/sphere packings that are numerically generated for FDTD simulations. We consider simple square/cubic lattice packings, disordered stealthy hyperuniform packings, and equilibrium packings in two and three dimensions. For each model, $N_c$ distinct packings are considered, each of which contains $N$ particles of radius $a$ and has number density $\rho$. All packings have the identical packing fraction $\phi_2 = 0.25$. Quantities $Q_U$ and $\sigma$ are the parameters for the collective-coordinate optimization method; see Sec. III in the main text.

<table>
<thead>
<tr>
<th>Systems \ Parameters</th>
<th>$N$</th>
<th>$\rho$</th>
<th>$N_c$</th>
<th>$Q_U a$</th>
<th>$\sigma$</th>
<th>$a$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Square lattice packings</td>
<td>25</td>
<td>1</td>
<td>1</td>
<td>-</td>
<td>-</td>
<td>0.2801</td>
</tr>
<tr>
<td>Cubic lattice packings</td>
<td>25</td>
<td>1</td>
<td>1</td>
<td>-</td>
<td>-</td>
<td>0.3908</td>
</tr>
<tr>
<td>2D Stealthy hyperuniform packings</td>
<td>100</td>
<td>1</td>
<td>20</td>
<td>1.3</td>
<td>0.57</td>
<td>0.2801</td>
</tr>
<tr>
<td>3D Stealthy hyperuniform packings</td>
<td>1000</td>
<td>1</td>
<td>10</td>
<td>1.5</td>
<td>0.8</td>
<td>0.3908</td>
</tr>
<tr>
<td>2D Equilibrium packings</td>
<td>100</td>
<td>1</td>
<td>20</td>
<td>-</td>
<td>-</td>
<td>0.2801</td>
</tr>
<tr>
<td>3D Equilibrium packings</td>
<td>1000</td>
<td>1</td>
<td>10</td>
<td>-</td>
<td>-</td>
<td>0.3908</td>
</tr>
</tbody>
</table>

8.19.2 Parameters for FDTD simulations

Table 8.3: Parameters employed in FDTD simulations via MEEP (see Sec. VII in the main text). Here $\Delta x$ is the grid resolution, $\rho$ is the number density of each configuration, and $\min[k_1]$ and $\max[k_1]$ refer to the minimal and the maximal wavenumbers of the Gaussian pulses in the matrix phase, respectively. We use different parameters for periodic and disordered models.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>2D Periodic</th>
<th>3D Periodic</th>
<th>2D Disordered</th>
<th>3D Disordered</th>
</tr>
</thead>
<tbody>
<tr>
<td>$L$</td>
<td>1</td>
<td>1</td>
<td>10</td>
<td>10</td>
</tr>
<tr>
<td>$\Delta x \rho^{1/d}$</td>
<td>1/60</td>
<td>1/60</td>
<td>1/60</td>
<td>1/40</td>
</tr>
<tr>
<td>$L_{pml}$</td>
<td>5</td>
<td>5</td>
<td>2.5</td>
<td>10</td>
</tr>
<tr>
<td>$L_{padd}$</td>
<td>1</td>
<td>1</td>
<td>5</td>
<td>5</td>
</tr>
<tr>
<td>$\min[k_1]a$</td>
<td>0.1a/L</td>
<td>0.1a/L</td>
<td>0.1</td>
<td>0.1</td>
</tr>
<tr>
<td>$\max[k_1]a$</td>
<td>4.0a/L</td>
<td>4.0a/L</td>
<td>1.1</td>
<td>1.1</td>
</tr>
</tbody>
</table>

We list the parameters employed to carry out FDTD simulations. Using the numerically generated packings (see Table 8.2), we perform FDTD simulations via MEEP, an open-source software package [224]. The simulation setup is depicted in Sec. VII in the main text. For simplicity, we take $\varepsilon_1 = 1$ throughout the simulation. Table 8.3 lists the parameters employed the simulation via MEEP.
8.19.3 Computation of $\varepsilon_e$ from FDTD simulations

Here, we provide a detailed description of numerical homogenization estimates obtained from FDTD simulations. To understand this step, we employ the concept of macroscopic fields, which has been used to explain the atomic origin of dielectric constants of materials [134].

At macroscopic length scales, local electric field $E^{(\text{local})}(x; \omega)$ at a frequency $\omega$ in a composite, a solution of Eq. (8.114), can be coarse-grained to macroscopic electric field $E^{(\text{macro})}(x; \omega)$ defined as

$$E^{(\text{macro})}(x; \omega) \equiv f(x) \otimes E^{(\text{local})}(x; \omega) = \int f(x - x') E^{(\text{local})}(x'; \omega) \, dx',$$

where $\otimes$ denotes the convolution operator, $f(x)$ is a certain weighting function satisfying $\int f(x) \, dx = 1$. Note that the support of $f(x)$ is larger than the inhomogeneity length scales $\ell$ of a composite. Analogously, we can also define the macroscopic electric displacement field $D^{(\text{macro})}(x; \omega)$ in terms of its local counterpart $D^{(\text{local})}(x; \omega)$.

When the effective-medium description is valid and the medium is macroscopically isotropic (i.e., $\varepsilon_e = \varepsilon_e I$), $E^{(\text{macro})}(x; \omega)$ should meet the effective wave equation

$$\nabla \times \nabla \times E^{(\text{macro})}(x; \omega) - k_e^2 E^{(\text{macro})}(x; \omega) = 0,$$

where $k_e$ is the effective wavenumber in a composite at a frequency $\omega$. The corresponding effective dielectric constant $\varepsilon_e$ is

$$\varepsilon_e = (k_e c / \omega)^2,$$

where $c$ is the speed of light in vacuum.

From Eq. (8.200), we can write the $y$ components of macroscopic electric and
electric displacement fields propagating through a composite in the $+x$ direction as

$$
E_y^{(\text{macro})}(x, \omega) \sim E_y^{(\text{macro})}(\omega) e^{ik_e x},
$$

$$
D_y^{(\text{macro})}(x, \omega) \sim D_y^{(\text{macro})}(\omega) e^{ik_e x},
$$

respectively. The convolution theorem yields that the spatial Fourier transforms of these macroscopic fields are given as

$$
\tilde{E}_y^{(\text{macro})}(k_e, \omega) \sim E_y^{(\text{macro})}(\omega) = \tilde{f}(k_e) \tilde{E}_y^{(\text{local})}(k_e, \omega),
$$

$$
\tilde{D}_y^{(\text{macro})}(k_e, \omega) \sim D_y^{(\text{macro})}(\omega) = \tilde{f}(k_e) \tilde{D}_y^{(\text{local})}(k_e, \omega),
$$

where the Fourier transform is defined in the region $V$ of a composite as

$$
\tilde{f}(q) \equiv |V|^{-1} \int_V f(x') e^{-iq\cdot x'} \, dx',
$$

and $|V|$ is the volume of $V$. Thus, the effective dielectric constant can be written as

$$
\varepsilon_e = \frac{D_y^{(\text{macro})}(\omega)}{E_y^{(\text{macro})}(\omega)} = \frac{\tilde{D}_y^{(\text{macro})}(k_e, \omega)}{\tilde{E}_y^{(\text{macro})}(k_e, \omega)} = \frac{\tilde{D}_y^{(\text{local})}(k_e, \omega)}{\tilde{E}_y^{(\text{local})}(k_e, \omega)},
$$

(8.202)

regardless of a choice of the weighting function $f(x)$.

Note that the effective dielectric constants given in Eqs. (8.201) and (8.202) are identical provided the exact value of $k_e$. Since it is not easy to compute $k_e$ in practice, however, we numerically solve the following equation of a complex wavenumber $q$:

$$
\left| \frac{\tilde{D}_y^{(\text{local})}(q, \omega)}{\tilde{E}_y^{(\text{local})}(q, \omega)} - \left( \frac{qc}{\omega} \right)^2 \right|^2 = 0,
$$

(8.203)

where the first and second terms come from Eqs. (8.202) and (8.201), respectively. The estimated effective dielectric constant $\varepsilon_e$ is obtained from $(q'c/\omega)^2$, where $q'$ is a solution of Eq. (8.203). Importantly, one can recover the conventional volume-
averaging homogenization estimates from relation (8.202) by taking $k_e = 0$, i.e.,

$$\varepsilon_e(k_1) = \frac{\tilde{D}_y(0, \omega)}{\tilde{E}_y(0, \omega)}.$$ 

Importantly, we do not use this simple volume-averaging estimate since it is no longer valid for intermediate wavelengths ($k_1 \rho^{-1/d} \gtrsim 0.4$), where $\rho$ is number density of the particle centers [105].

### 8.19.4 Validity of Extraction Method

Here we confirm the validity of the numerical homogenization estimates from FDTD simulations (see Sec. VII in the main text and Sec. 8.19.3) in the intermediate-wavelength regime. For this purpose, we carry out the simulations for 2D and 3D periodic packings (square and simple-cubic lattice packings) of packing fraction $\phi_2 = 0.25$. As noted in Sec. 8.16.1, their effective dielectric constant depend on the direction of the incident wavevector $k_1$. For simplicity, we only consider the case where $k_1$ is aligned with one of the minimal lattice vectors, i.e., $\Gamma$-$X$ direction in the first Brillouin zone. Such models enable us to validate the accuracy of our FDTD simulations because $\varepsilon_e(k_1)$ also can be accurately extracted from the lowest two photonic bands, which are calculated via MPB, an open-source software package [142]. Specifically, the band structures can be converted to $\varepsilon_e(\omega)$ at a given frequency $\omega$:

$$\varepsilon_e(\omega) = \left(\frac{cK}{\omega}\right)^2,$$

where $K$ is the Bloch wavenumber along the $\Gamma$-$X$ direction. Furthermore, such periodic packings must show two salient dielectric characteristics: see Sec. VIII in the main text.

We carry out FDTD simulations for 2D square lattice packings of $\varepsilon_2/\varepsilon_1 = 1/4, 4$, and 3D simple-cubic lattice packing of $\varepsilon_2/\varepsilon_1 = 4$; see Sec. 8.19.2 for simulation pa-
rameters. As shown in Fig. 8.15, the corresponding FDTD simulation results show excellent agreement with the band-structure calculations and accurately capture both of the aforementioned features up to $k_1L = 4$. This implies that our numerical homogenization estimates from FDTD simulations are valid from the infinite-wavelength limit down to intermediate wavelengths.

Figure 8.15: Comparison of $\varepsilon_e(k_1)$ for periodic packings of $\phi_2 = 0.25$ estimated from the band-structure (via MPB) and FDTD simulations along the $\Gamma$-$X$ direction. Three systems are considered: (a) square lattice packing of $\varepsilon_2/\varepsilon_1 = 1/4$, (b) square lattice packing of $\varepsilon_2/\varepsilon_1 = 4$, and (c) simple cubic lattice packing of $\varepsilon_2/\varepsilon_1 = 4$. Note that the band-structure calculations are omitted in the photonic bandgaps. Both types of simulations show excellent agreement, implying that our homogenization estimates from FDTD simulations are valid down to intermediate wavelengths ($k_1L < 4$ and $k_1a \lesssim 1.1$).

8.20 Appendix J: Results for Two-Dimensional Systems

Here, we present some plots for two-dimensional models, which are not presented in the main text for brevity. Figure 8.16 shows the spectral densities and associated nonlocal attenuation functions for the four disordered models in two dimensions. All models have the same volume fraction of dispersed phase $\phi_2 = 0.25$.

Figure 8.17 compares the (both unmodified and scaled) strong-contrast approximations and the Maxwell-Garnett approximation (MGA) for the effective dielectric constant $\varepsilon_e(k_1)$ of a 2D square lattice packing to the corresponding FDTD simu-
Figure 8.16: Plots of (a) spectral density, and (b) the real and (c) imaginary parts of the nonlocal attenuation function for the four models of two-dimensional disordered particulate composite media. All models have the same volume fraction of dispersed phase \( \phi_2 = 0.25 \). Here, the first three models consist of identical spheres of radius \( a \). For class I hyperuniform packings via tessellation-based procedure, \( a \) is the mean sphere radius, i.e., \( \phi_2 = \rho v_1(a) \).

This packing has the packing fraction \( \phi_2 = 0.25 \) and contrast ratio \( \varepsilon_2/\varepsilon_1 = 1/4 \). Similar to the results presented in the main text, the MGA shows good estimates only in the quasistatic regime. For the strong-contrast approximations, unlike the cases of \( \varepsilon_2/\varepsilon_1 > 1 \), the unmodified approximation provides better estimates for \( \text{Re}[\varepsilon_e(k_1)] \) than the scaled counterpart up to the edge of first photonic band. However, the scaled approximation correctly predicts that the frequency at which the Bragg diffraction occurs lies within the first photonic bandgap.

Figure 8.18 compares FDTD simulation results to the MGA and the (both unmodified and scaled) strong-contrast approximations for 2D disordered models: equilibrium packings and stealthy hyperuniform packings. Each model has packing fraction \( \phi_2 = 0.25 \) and contrast ratio \( \varepsilon_2/\varepsilon_1 = 4 \). These results are qualitatively the same as the three-dimensional results discussed in the main text. Among the three approximations, the scaled strong-contrast approximation shows the best predictive power up to \( k_1a = 0.6 \).
Figure 8.17: Comparison of the predictions of the both scaled and unscaled strong-contrast formulas and the MGA for $\varepsilon_e(k_1)$ of 2D square lattice packing to the corresponding FDTD simulation results. Packing fraction is $\phi_2 = 0.25$, and contrast ratio is $\varepsilon_2/\varepsilon_1 = 1/4$. Here $k_1$ is the wavenumber in the matrix phase along the $\Gamma$-$X$ direction, and $L$ is the side length of a unit cell.

Figure 8.18: Comparison of FDTD simulation results to the predictions from the strong-contrast formulas and the MGA for the effective dynamic dielectric constant $\varepsilon_e(k_1)$ of two-dimensional disordered disk packings. We consider (a) equilibrium packing and (b) stealthy hyperuniform packings [$\tilde{\chi}_V(Q) = 0$ for $Qa < 1.3$] of particle radius $a$, packing fraction $\phi_2 = 0.25$, and phase contrast ratio $\varepsilon_2/\varepsilon_1 = 4$. Here $k_1$ is the wavenumber in the matrix phase, and the error bars in FDTD simulations represent the standard errors over independent configurations.
Chapter 9

Effective Elastic Wave Characteristics of Composite Media

9.1 Introduction

The theoretical determination of the effective elastic wave characteristics of multiphase composite media is of great importance in geophysics [26, 170, 201], exploration seismology [267, 256], diagnostic sonography [258], crack diagnosis [281, 107], architectural acoustics [326] and acoustic metamaterials [344], among many examples. Such effective elastic properties generally depend on the phase properties, phase volume fractions \( \phi_i \), frequency \( \omega \) or wavenumber \( k_l \) of the incident elastic waves, and an infinite set of correlation functions that characterizes the composite microstructure [147, 149, 144]. There have been numerous theoretical/computational attempts to estimate the effective elastic wave characteristics [147, 149, 144, 148, 139, 153, 266, 248]. However, the preponderance of previous closed-form approximation formulas for the effective elastodynamic properties apply only in the quasistatic regime [144, 153], i.e.,
applicable when $k_I \ell \ll 1$, where $\ell$ is a characteristic heterogeneity length scale\(^1\), and
under restrictive conditions. One such closed-form approximation is the Gaunaurd-Überall approximation \cite{153}, which we employ to compare to simulation data and our nonlocal formulas described below.

Our focus in this chapter is the theoretical determination of the effective dynamic stiffness tensor $C_e(k_I, \omega)$ of a two-phase elastic composite, which depends on the frequency $\omega$ or wavevector $k_I$ of the incident elastic waves beyond the quasistatic regime; see Fig 9.1. From this effective property, one can determine the corresponding effective wave speeds $c_{eL,T}$ and attenuation coefficients $\gamma_{eL,T}$. To achieve this goal, we first generalize the strong-contrast expansion formalism that has been employed to treat the static elastic problem \cite{293, 292, 296} to the elastodynamic problem in the quasistatic regime by establishing homogenized constitutive relations that are local in space. Because of the interplay between longitudinal and transverse waves and the complexity of the fourth-rank tensors that are involved, this task is considerably more challenging than the derivation of its dielectric counterparts \cite{242, 308}. The terms of the resulting quasistatic strong-contrast expansions are explicitly given in terms of integrals over products of Green’s functions and the $n$-point correlation functions $S^{(i)}_n(x_1, \cdots, x_n)$ of the random two-phase medium to infinite order. Here, the quantity $S^{(i)}_n(x_1, \cdots, x_n)$ gives the probability of finding $n$ points at positions $x_1, \cdots, x_n$ simultaneously in phase $i$. This implies that multiple scattering to all orders is exactly treated in the long-wavelength or quasistatic regime. Due to the fast-convergence properties of strong-contrast expansions, their lower-order truncations yield accurate closed-form approximate formulas for the effective dynamic moduli that apply for a wide class of microstructures. Postulated nonlocal variants of these formulas are resummed representations of the strong-contrast expansions that still accurately cap-

\(^1\)Some multiple-scattering approximations for effective elastic waves are accurate beyond the quasistatic regime; see Ref. \cite{248} and references therein. However, these formulas require complicated scattering coefficients of individual scatterers.
ture multiple scattering to all orders via the microstructural information embodied in the spectral density $\tilde{\chi}_v(Q)$. The quantity $\tilde{\chi}_v(Q)$ is the Fourier transform of the autocovariance function $\chi_v(r) \equiv S_2^{(i)}(r) - \phi_i^2$, where $r \equiv x_2 - x_1$, which can be easy to ascertain for general microstructures theoretically, computationally, or via scattering experiments [54].

We also verify the accuracy of the postulated approximations via full-waveform simulations for certain benchmark models. This validation allows us to use them to predict the effective elastic wave characteristics accurately well beyond the quasistatic regime for a wide class of composite microstructures without computationally expensive full-blown simulations. As discussed in Sec. 9.2, such a broad microstructure class includes particulate composites consisting of identical or polydisperse particles of arbitrary shapes (ellipsoids, cylinders, polyhedra) that may or not overlap, cellular networks as well as systems without well-defined inclusions. Such broad applicability is a notable advantage of our formulas over other multiple-scattering approximations, such as Keller’s approximation [148, 59]. Thus, our postulated formulas can be employed to accelerate the discovery of novel elastodynamic composites by appropriate tailoring of the spectral densities [298, 46] and then generating the microstructures satisfying them [46], as elaborated in Sec. 9.7. Thus, our postulated formulas can be employed to accelerate the discovery of novel elastodynamic composites by appropriate tailoring of the spectral densities [298, 46] and then generating the microstructures satisfying them [46].

While our strong-contrast formulas for the effective dynamic elastic moduli can be applied to periodic two-phase media, the primary applications are spatially correlated disordered media because they can provide advantages over periodic ones with high crystallographic symmetries [186, 341], including perfect isotropy and robustness.

$^2$Keller’s approximation is derived for the simplified case in which only longitudinal waves propagate in a very special system: colloidal suspensions of spherical particles in which the fluid has zero shear modulus. Such systems can be treated with the scalar Helmholtz equation, which is to be contrasted with our treatment of the full tensor elastodynamic treatment.
against defects [79, 196]. We are interested in both “garden-variety” models [296] as well as exotic hyperuniform forms [313, 346, 300] of disordered two-phase media. Hyperuniform two-phase systems are characterized by an anomalous suppression of volume-fraction fluctuations in the infinite-wavelength limit [313, 346, 300], i.e., the spectral density $\tilde{\chi}_V(Q)$ obeys the condition

$$\lim_{|Q| \to 0} \tilde{\chi}_V(Q) = 0.$$  \hspace{1cm} (9.1)$$

Disordered hyperuniform systems have attracted considerable attention over the last decade because of their relevance on a broad spectrum of topics in various fields [317, 354, 121, 245, 225, 194, 186, 343, 324, 300, 177, 104, 160, 137, 300, 362, 97, 30, 316, 193, 339, 303, 46, 157] as well as for their emerging technological importance [80, 196, 193, 179, 82, 163, 359, 300, 104]; see discussion in Sec. 8.1.

We apply our nonlocal strong-contrast formulas to predict the real and imaginary parts of the effective elastic moduli for model microstructures that possess some typical disorder (nonhyperuniform) as well as those with exotic hyperuniform disorder (Sec. 9.5). We are particularly interested in exploring the elastic properties of a special class of hyperuniform composites called disordered stealthy hyperuniform media, which are defined to be those that possess zero-scattering intensity for a set of wavevectors around the origin [321, 17, 317, 352, 46], i.e.,

$$\tilde{\chi}_V(Q) = 0, \text{ for } 0 \leq Q \leq Q_U,$$  \hspace{1cm} (9.2)

where $Q \equiv |Q|$. Disordered stealthy hyperuniform materials have been shown to exhibit novel optical, acoustic, mechanical, and transport properties [242, 179, 355, 57, 102, 339, 303, 158]. Among other results, we show here that disordered hyperuniform media are generally less lossy than their nonhyperuniform counterparts. We also demonstrate that disordered stealthy hyperuniform particulate composites ex-
hibit novel wave characteristics, including the capability to act as low-pass filters that transmit elastic waves isotropically without loss up to a selected wavenumber. Our results demonstrate that one can design the effective wave characteristics of a disordered composite, hyperuniform or not, by engineering spatial correlations of microstructure at prescribed length scales.

In Sec. 9.2, we present the strong-contrast formalism to derive corresponding expansions of the effective elastic wave characteristics of macroscopically anisotropic or isotropic two-phase media in the quasistatic regime. In Sec. 9.3, we extract strong-contrast approximations from the exact expansions. In Sec. 9.4, we extend the validity of the strong-contrast approximations for the effective dynamic moduli so that they apply well beyond the quasistatic regime. The accuracy of these nonlocal approximations is verified by comparison to full-waveform simulations for certain benchmark models. In Sec. 9.5, we describe four models of disordered composites that we treat in the chapter, two of which are nonhyperuniform and two of which are hyperuniform. In Sec. 9.6, we investigate the microstructure-dependence of the effective elastic wave characteristics for these models. Finally, we provide concluding remarks in Sec. 9.7.

9.2 Exact Strong-Contrast Expansions

Here we extend the general strong-contrast formalism that was devised for the purely static elastic problem [293, 292, 296] to the elastodynamic problem in the long-wavelength (quasistatic) regime. We first present a compact derivation of the expansions for the effective stiffness tensor $C_e(k,\omega)$ for a macroscopically anisotropic medium (Sec. 9.2.1) and then specialize them to a macroscopically isotropic medium (Sec. 9.2.2). Detailed derivations are given in Appendix 9.10.
Figure 9.1: (a) Schematic of a large ellipsoidal, macroscopically anisotropic two-phase composite medium embedded in an infinite reference phase of mass density $\rho_I$ and stiffness tensor $C_I$ (gray regions) under an applied elastic waves $\mathbf{e}_0(\mathbf{x}) = \mathbf{e}_0 \exp(i(\mathbf{k}_I \cdot \mathbf{x} - \omega t))$ of frequency $\omega$. The wavelength $\lambda$ associated with the applied wave can span from the quasistatic regime ($2\pi\ell/\lambda \ll 1$) down to the intermediate-wavelength regime ($2\pi\ell/\lambda \lesssim 1$), where $\ell$ is the inhomogeneity length scale. (b) After homogenization, the same ellipsoid can be regarded to be a specimen of homogeneous medium with an effective stiffness tensor $C_e(\mathbf{k}_I, \omega)$, which depends on $\omega$ and $\mathbf{k}_I$. As noted in the main text, we omit the $\omega$ dependence of $C_e(\mathbf{k}_I, \omega)$ because (without loss of generally) we assume a linear dispersion relation between $|\mathbf{k}_I|$ and $\omega$.

9.2.1 Macroscopically Anisotropic Media

Here we follow closely the strong-contrast formalism of Torquato [292, 296] but apply it to derive the analogous series expansions for the effective dynamic moduli. We consider a macroscopically large ellipsoidal specimen of two-phase statistically homogeneous but anisotropic composite in $\mathbb{R}^d$ embedded inside an infinitely large reference phase $I$ with mass density $\rho_I$ and stiffness tensor $C_I$; see Fig. 9.1 The microstructure is perfectly general, and its inhomogeneity length scale $\ell$ is much smaller than the specimen size, i.e., $\ell \ll L$. The shape of this specimen is purposely chosen as nonspherical since any rigorously correct expression for the effective property must ultimately be independent of the shape of the composite specimen in the infinite-volume limit.
For a two-phase medium, we define the indicator function for phase \( i \) as

\[
I^{(i)}(x) \equiv \begin{cases} 
1, & \text{x lies in phase } i \\
0, & \text{otherwise}
\end{cases}, \quad \text{for } i = 1, 2. 
\] (9.3)

For statistically homogeneous media, its ensemble average is simply the phase volume fraction, i.e., \( \phi_i \equiv \langle I^{(i)}(x) \rangle \) so that \( \phi_1 + \phi_2 = 1 \). The local stiffness tensor \( C(x) \) of such a medium can be written as

\[
C(x) \equiv C_1 I^{(1)}(x) + C_2 I^{(2)}(x), 
\] (9.4)

where \( C_i \) denotes the stiffness tensor of phase \( i \) \((= 1, 2)\). For simplicity, we take the reference phase to be phase \( q \) \((\text{equal to } 1 \text{ or } 2)\).

In the ensuing discussion, we make the following three assumptions on the phase properties.

(a) Phase 1 and phase 2 are elastically isotropic, i.e.,

\[
C_i = dK_i \Lambda_h + 2G_i \Lambda_s, \quad (i = 1, 2) \tag{9.5}
\]

where \( K_i \) and \( G_i \) are bulk and shear moduli of phase \( i \) \((= 1, 2)\), respectively. Here, the \textit{hydrostatic projection tensor} \( \Lambda_h \) and \textit{shear projection tensor} \( \Lambda_s \) are constant fourth-rank tensors given by

\[
(\Lambda_h)_{ijkl} \equiv \frac{1}{d} \delta_{ij} \delta_{kl}, \tag{9.6}
\]

\[
(\Lambda_s)_{ijkl} \equiv \frac{1}{2} (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}) - \frac{1}{d} \delta_{ij} \delta_{kl}, \tag{9.7}
\]

where \( \delta_{ij} \) is the Kronecker delta symbol. The tensor \( \Lambda_h \) projects onto fields that are isotropic everywhere, whereas the tensor \( \Lambda_s \) projects onto fields that
are trace-free (see the SM for useful identities).

(b) Each phase is dissipationless, namely, the elastic moduli $K_i$ and $G_i$ for $i = 1, 2$ are real-valued and frequency-independent.

(c) The mass densities of both phases are identical, i.e.,

$$\rho_1 = \rho_2 = \rho_e. \quad (9.8)$$

Assumption (a) enables us to decompose the elastic waves in phase $i$ ($= 1, 2$) into longitudinal and transverse waves with their respective wave speeds $c_{Li}$ and $c_{Ti}$, which are defined by

$$c_{Li}^2 \equiv \left[ K_i + 2(1 - 1/d)G_i \right]/\rho_i, \quad c_{Ti}^2 \equiv G_i/\rho_i. \quad (9.9)$$

Under this assumption, the Poisson ratio $\nu_i$ of phase $i$ is expressed as [296]

$$\nu_i = \frac{dK_i - 2G_i}{d(d - 1)K_i + 2G_i}, \quad (9.10)$$

and bounded in the range of $-1 \leq \nu_i \leq 1/(d - 1)$ [296, 366]. Assumption (b) means that these speeds are independent of frequency $\omega$, implying the following linear dispersion relations:

$$\omega = c_{Li}/k_{Li} = c_{Ti}/k_{Ti} \quad (i = 1, 2), \quad (9.11)$$

where $k_{Li}$ and $k_{Ti}$ are longitudinal and transverse wavenumbers, respectively. Assumption (c) is achievable for many pairs of solid materials; see discussion in Ref. [158].

We suppose that the applied or incident elastic strain field $\epsilon_0(x)$ is a plane wave of

---

3In this chapter, ‘wave speed’ always refers to the speed associated with the phase of the wave. This term is used instead of ‘phase speed’ because ‘phase’ in this chapter refers to a constituent material of a composite.
an angular frequency $\omega$, well-defined propagation direction $\hat{\mathbf{k}}$ in the reference phase, and the associated wavelength $\lambda$. Our interest is in deriving an exact expression for the effective stiffness tensor $C_e(\omega)$ or, equivalently, $C_e(k_{Lq})$ in the quasistatic regime ($\ell \ll \lambda$), where $k_{Lq}$ is the longitudinal wavenumber (9.11) in the reference phase. While each phase is dissipationless, as stated in (b), the composite is generally lossy (i.e., $C_e$ is complex-valued) due to scattering from the inhomogeneities. Nonetheless, our results can be straightforwardly extended to viscoelastic media (with complex-valued moduli), but this will not be done in this chapter.

Under the assumptions (a)-(c), the local displacement field $\mathbf{u}(\mathbf{x})$ solves the time-harmonic wave equation:

$$\omega^2 u_i + \left(c_{Lq}^2 - c_{Tq}^2\right) \frac{\partial^2 u_k}{\partial x_i \partial x_k} + c_{Tq}^2 \frac{\partial^2 u_i}{\partial x_j \partial x_i} = -\frac{\partial P_{ij}(\mathbf{x})}{\partial x_j}, \quad (9.12)$$

where the Einstein summation is implied, $c_{Lq}$ and $c_{Tq}$ are given in Eq. (9.9), and $P_{ij}(\mathbf{x})$ is the induced stress polarization field given by

$$P(\mathbf{x}) \equiv \rho_q^{-1} [\mathbf{C}(\mathbf{x}) - \mathbf{C}_q] : \mathbf{\epsilon}(\mathbf{x}), \quad (9.13)$$

and $\mathbf{\epsilon}(\mathbf{x})$ is the local strain tensor [366]. The symmetric second-rank tensor $P(\mathbf{x})$ is the induced field relative to the reference phase $q$, and hence is nonzero only in the “polarized” phase $p$ ($p \neq q$).

Following Torquato [292, 296], we use a Green’s function formalism to solve the wave equation (9.12) for $\mathbf{u}$ for an arbitrary macroscopically anisotropic two-phase medium:

$$\mathbf{u}(\mathbf{x}) = \mathbf{u}_0(\mathbf{x}) + \int g^{(q)}(\mathbf{x} - \mathbf{x'}) \cdot [\nabla \cdot P(\mathbf{x'})] \, d\mathbf{x'}, \quad (9.14)$$

where $\mathbf{u}_0(\mathbf{x})$ is related to the applied strain $\mathbf{\epsilon}_0$, and $g^{(q)}(\mathbf{r})$ is the the second-rank
Green’s function associated with (9.12). Taking the symmetric part of the gradient of \( u(x) \) gives an integral equation for the local strain tensor \( \epsilon(x) \):

\[
\epsilon(x) = \epsilon_0(x) + \int G^{(q)}(x - x') : P(x') \, dx',
\]

where the fourth-rank Green function \( G^{(q)}(r) \) associated with the reference phase \( q \) is given by \[296\]

\[
G^{(q)}(r) = -D^{(q)}(r) + H^{(q)}(r),
\]

\( r \equiv x - x' \), \( D^{(q)}(r) \) is a constant fourth-rank tensor that arises when one excludes an infinitesimal volume around the singularity of the Green function at \( x' = x \), and \( H^{(q)}(r) \) is the contribution outside of this “exclusion” region.

The fourth-rank tensor \( H^{(q)}(r) \) is symmetric under the following index changes, i.e.,

\[
H^{(q)}_{ijkl} = H^{(q)}_{jikl} = H^{(q)}_{ijlk} = H^{(q)}_{klij},
\]

and its explicit expression is given by

\[
H^{(q)}_{ijkl}(r) = \frac{-i\pi}{2(2\pi)^{d/2} \omega^{d-1} r^{d+1}} \left\{ \left[ r^{d/2+1}_L \mathcal{H}^{(1)}_{d/2+1}(r_L) - r^{d/2+1}_T \mathcal{H}^{(1)}_{d/2+1}(r_T) \right] (dA_h + 2I) \\
+ r^{d/2+2}_T \mathcal{H}^{(1)}_{d/2}(r_I) I - \left[ r^{d/2+2}_L \mathcal{H}^{(1)}_{d/2+2}(r_L) - r^{d/2+2}_T \mathcal{H}^{(1)}_{d/2+2}(r_T) \right] \left[ 2 T_1(r) + 4 T_2(r) \right] \\
- r^{d/2+3}_T \mathcal{H}^{(1)}_{d/2+1}(r_L) T_2(r) + \left[ r^{d/2+3}_L \mathcal{H}^{(1)}_{d/2+3}(r_L) - r^{d/2+3}_T \mathcal{H}^{(1)}_{d/2+3}(r_T) \right] T_3(r) \right\}.
\]

(9.18)

where \( r_L \equiv k_{Lq} r \), \( r_T \equiv k_{Tq} r \), \( I \) is the fourth-rank identity tensor, and \( \mathcal{H}^{(1)}_{\nu}(x) \) is the Hankel function of the first kind of order \( \nu \). The three fourth-rank tensors \( T_i(r) \) for
\( i = 1, 2, 3 \) are defined, in component form, as

\[
(T_1)_{ijkl}(\mathbf{r}) \equiv \frac{1}{2} (\delta_{ij} \hat{r}_k \hat{r}_l + \hat{r}_i \hat{r}_j \delta_{kl}),
\]
\( (9.19) \)

\[
(T_2)_{ijkl}(\mathbf{r}) \equiv \frac{1}{4} (\hat{r}_i \delta_{jk} \hat{r}_l + \hat{r}_j \delta_{ik} \hat{r}_l + \hat{r}_i \delta_{jl} \hat{r}_k + \hat{r}_j \delta_{il} \hat{r}_k),
\]
\( (9.20) \)

\[
(T_3)_{ijkl}(\mathbf{r}) \equiv \hat{r}_i \hat{r}_j \hat{r}_k \hat{r}_l,
\]
\( (9.21) \)

\( \hat{r} \equiv r/|r| \), and \( \hat{r}_i \) is the \( i \)th component of \( \hat{r} \). Formulas for the traces of \( H^{(q)}(\mathbf{r}) \) are provided in the SM. Note that (9.18) reduces to its static counterpart given in Refs. [293, 292, 296] up to a multiplicative factor \( \rho_q \). The constant tensor \( D^{(q)} \) depends on the “exclusion-region” shape. Due to the fast-convergence properties of the resulting expansion discussed below and in Refs. [293, 296, 308], we choose a spherical-exclusion region, for which

\[
D^{(q)} = \frac{\rho_c \Lambda_h}{dK_q + 2(d-1)G_q} + \frac{\rho_c d(K_q + 2G_q) \Lambda_s}{G_q (d + 2)[dK_q + 2(d-1)G_q]}
\]
\[
= \frac{1}{dc_{\nu_q}^2} \Lambda_h + \frac{1}{d + 2} \left( \frac{2}{dc_{\nu_q}^2} + \frac{1}{c_{\tau_q}^2} \right) \Lambda_s.
\]
\( (9.22) \)

The integral equation (9.15) is written in a compact linear operator form as

\[
\epsilon = \epsilon_0 + GP.
\]
\( (9.23) \)

Excluding the contribution from the exclusion region in (9.23), we define *generalized cavity strain* tensor:

\[
f = \epsilon_0 + HP.
\]
\( (9.24) \)

Use of (9.23), (9.24), and (9.13) demonstrates that \( P \) and \( f \) are directly related as follows:

\[
P(x) = \left[ L^{(q)} I^{(p)}(x) \right] : f(x), \quad (p \neq q)
\]
\( (9.25) \)
where

\[ L^{(q)} \equiv \frac{(C_p - C_q)}{\rho_q} : \left[ I + D^{(q)} : \frac{(C_p - C_q)}{\rho_q} \right]^{-1} \]

\[ = dc_{L_q}^2 \left[ \kappa_{pq} \Lambda_h + \frac{(d + 2)c_{T_q}^2}{dc_{L_q}^2 + 2c_{T_q}^2 \mu_{pq} \Lambda_s} \right], \tag{9.26} \]

\( \kappa_{pq} \) and \( \mu_{pq} \) are the scalar polarizabilities for bulk and shear moduli, respectively, defined by

\[ \kappa_{pq} = \frac{K_p - K_q}{K_p + 2(d - 1)G_q/d}, \tag{9.27} \]

\[ \mu_{pq} = \frac{G_p - G_q}{G_p + [dK_q/2 + (d + 1)(d - 2)G_q/d] G_q/(K_q + 2G_q)}. \tag{9.28} \]

Note that Eqs. (9.22) and (9.26) are identical to their static counterparts [292, 294, 296] up to a multiplicative factor \( \rho_q \).

We now find a series expansion for the following homogenized constitutive relation

\[ \langle P \rangle (x) = L^{(q)}_{e}(k_{L_q}) : \langle f \rangle (x), \tag{9.29} \]

where \( \langle \cdot \rangle \) denotes an ensemble average, and the effective constant tensor \( L^{(q)}_{e}(k_{L_q}) \) is explicitly given as

\[ L^{(q)}_{e}(k_{L_q}) = \left[ C_e(k_{L_q}) - C_q \right]/\rho_e : \left[ I + D^{(q)} : \left[ C_e(k_{L_q}) - C_q \right]/\rho_e \right]^{-1}. \tag{9.30} \]

To do so, we solve \( P(x) \) in terms of \( \epsilon_0 \) by iteratively substituting (9.24) and (9.25). We then obtain the relation (9.29) from the aforementioned expansion and an ensemble average of (9.24) by eliminating \( \epsilon_0 \) in order to avoid conditional convergence problems.

Following the strong-contrast formalism of Torquato [292, 296], we obtain an ex-
pression of the effective tensor $L^{(q)}_e$ in the form of series expansion:

$$\phi_p^2 L^{(q)}_e : [L^{(q)}_e (k_{Lq})]^{-1} = \phi_p^2 L^{(q)}_e : \left\{ I + D^{(q)} : [C_e(k_{Lq}) - C_q] / \rho_e \right\} : \left\{ [C_e(k_{Lq}) - C_q] / \rho_e \right\}^{-1}$$

$$= \phi_p I - \sum_{n=2}^{\infty} B_n^{(p)}(k_{Lq}) ,$$

(9.31)

where

$$B_2^{(p)}(k_{Lq}) = \int dx_2 \, U^{(q)}(x_1 - x_2) \chi_v(x_1, x_2) ,$$

(9.32)

$$B_n^{(p)}(k_{Lq}) = (-1)^n (\phi_p)^{-(n-2)} \int dx_2 \cdots dx_n \, U^{(q)}(x_1 - x_2) : U^{(q)}(x_2 - x_3) : \cdots : U^{(q)}(x_{n-1}, x_n) \times \Delta_n^{(p)}(x_1, x_2, \cdots, x_n) , \quad n \geq 3,$$

(9.33)

$$U^{(q)}(r) \equiv L^{(q)} : H^{(q)}(r) ,$$

(9.34)

and $\Delta_n^{(p)}$ is a position-dependent determinant involving up to the $n$-point correlation function associated with the dispersed phase $p$, i.e.,

$$\Delta_n^{(p)}(x_1, \cdots, x_n) = \begin{vmatrix}
S_2^{(p)}(x_1, x_2) & S_1^{(p)}(x_1) & \cdots & 0 \\
S_3^{(p)}(x_1, x_2, x_3) & S_2^{(p)}(x_2, x_3) & \cdots & 0 \\
\vdots & \vdots & \ddots & \vdots \\
S_n^{(p)}(x_1, \cdots, x_n) & S_{n-1}^{(p)}(x_2, \cdots, x_n) & \cdots & S_2^{(p)}(x_{n-1}, x_n)
\end{vmatrix} .$$

(9.35)

Here, $S_n^{(p)}(x_1, \cdots, x_n)$ is the $n$-point correlation function defined as

$$S_n^{(p)}(x_1, \cdots, x_n) \equiv \langle I^{(p)}(x_1) \cdots I^{(p)}(x_n) \rangle ,$$

(9.36)

which gives the probability for simultaneously finding $n$ points at $x_1, x_2, \cdots, x_n$ in phase $p$ [255, 296]. Here, it is important to note that the integrals (9.32) and (9.33) are absolutely convergent because while $H^{(q)}(r)$ decays as $r^{-d}$ for large $r$, $\Delta_n^{(p)}$ identically vanishes at the boundary of the specimen [296]. The detailed derivation of
the strong-contrast expansion (9.31) is given in the SM. Importantly, the exact se-
ries expansion (9.31) accounts for complete microstructural information (infinite set
of \( n \)-point correlation functions) and hence multiple scattering to all orders in the
quasistatic regime.

**Remarks:**

1. Importantly, the strong-contrast expansion (9.31) is a series representation of
a linear fractional transformation of the effective stiffness tensor \( \mathbf{C}_e(k_{\mathbf{q}}) \) (left-
hand side). The series expansion in powers of the polarizabilities \( \kappa_{pq} \) and \( \mu_{pq} \)
of this particular rational function of \( \mathbf{C}_e(k_{\mathbf{q}}) \) has important consequences for
the predictive power of approximations derived from the expansion. While this
desirable feature is briefly discussed below, the reader is referred to Ref. [308]
for detailed explanations for the corresponding electromagnetic problem.

2. The homogenized constitutive relation (9.29) is *local in space* [i.e., \( \langle \mathbf{P} \rangle(\mathbf{x}) \) at
point \( \mathbf{x} \) depends on \( \langle \mathbf{f} \rangle(\mathbf{x}) \) at the same position \( \mathbf{x} \)] and strictly valid in the
long-wavelength regime. In such a regime, the effective elastic moduli are in-
dependent of the direction of incident waves, as shown in the expansion (9.31).
For shorter wavelengths, however, the associated relation must be nonlocal in
space [i.e., \( \langle \mathbf{P} \rangle(\mathbf{x}) \) at point \( \mathbf{x} \) depends on \( \langle \mathbf{f} \rangle(\mathbf{x}') \) at positions around \( \mathbf{x} \)], which
can result in “wavevector”-dependent effective elastic moduli, as was rigorously
shown for the analogous electromagnetic wave problem [308].

3. Note that the expansion (9.31) represents two different series: one for \( q = 1 \)
and \( p = 2 \) and the other for \( q = 2 \) and \( p = 1 \).

4. In the static limit (\( \omega \to 0 \)), the series (9.31) reduce to one derived for the static
strong-contrast expansions [293, 292, 296].

5. In contrast to its static counterpart, the \( n \)-point microstructure-dependent ten-
rors \(B^{(p)}_{n}\left(k_{Lq}\right)\) given in Eqs. (9.32) and (9.33) are functions of a frequency \(\omega\) of the elastic waves or, equivalently, the longitudinal and transverse wavenumbers \(k_{Lq}\) and \(k_{Tq}\). Throughout this chapter, we use \(k_{Lq}\) as an independent variable, instead of \(\omega\) or \(k_{Tq}\), for the following three reasons. First, the tensor \(H^{(q)}(r)\) as well as the effective stiffness tensors are conveniently written in terms of \(k_{Lq}\). Second, \(k_{Lq}\) is directly proportional to \(\omega\) and \(k_{Tq}\) [cf. (9.9)]. Furthermore, \(k_{Lq}\) is directly related to a length scale, which is suitable for describing microstructural information rather than the temporal quantity \(\omega\).

6. The exact expansions (9.31) are independent of the reference phase \(q\) and hence of the associated wavenumber \(k_{Lq}\).

7. Note that the strong-contrast formalism for the elastodynamic problem shares similar mathematical structure to the electromagnetic counterpart [242, 308]. In both cases, the wave equations can be simplified to the Helmholtz equation [i.e., \((\nabla^2 + k^2)u(x) = 0\)], which results in integral operator descriptions of their expansions being formally identical. However, there are important fundamental distinctions between the two problems. Among other things, while electromagnetic waves have only transverse propagation modes, elastic waves always have both transverse and longitudinal modes with different wave speeds. The interplay between these two propagation modes makes the theoretical determination of the effective elastodynamic properties generally more complex than its electromagnetic counterpart.

### 9.2.2 Macroscopically Isotropic Media

Here we assume that the composite is macroscopically isotropic. In this case, the effective stiffness tensor \(C_e\) can be expressed in the effective bulk and shear moduli (denoted by \(K_e\) and \(G_e\), respectively). Then, the series expansion (9.31) can be
reduced to
\[
\phi_p^2 \left[ \frac{\kappa_{pq}}{\kappa_{eq}(k_{Lq})} \Lambda_h + \frac{\mu_{pq}}{\mu_{eq}(k_{Lq})} \Lambda_s \right] = \phi_p I - \sum_{n=2}^{\infty} B_n^{(p)}(k_{Lq}) . \tag{9.37}
\]

Utilizing the properties of two tensors \( \Lambda_h \) and \( \Lambda_s \) (see the SM), one can separate (9.37) into two expansions by taking the quadruple inner products of \( \Lambda_h \) and \( \Lambda_s \) with (9.37). One is associated with the effective bulk modulus, and the other is related to the effective shear modulus:

\[
\kappa_{eq}(k_{Lq}) \equiv \frac{K_e(k_{Lq}) - K_q}{K_e(k_{Lq}) + 2(d-1)G_q/d} = \frac{\phi_p^2 \kappa_{pq}}{\phi - \sum_{n=2}^{\infty} C_n^{(p)}(k_{Lq})} , \tag{9.38}
\]

\[
\mu_{eq}(k_{Lq}) \equiv [G_e(k_{Lq}) - G_q] \left\{ G_e(k_{Lq}) + [dK_q/2 + (d+1)(d-2)G_q/d]G_q/(K_q + 2G_q) \right\}^{-1}
= \frac{\phi_p^2 \mu_{pq}}{\phi - \sum_{n=2}^{\infty} D_n^{(p)}(k_{Lq})} , \tag{9.39}
\]

respectively, where \( C_n^{(p)}(k_{Lq}) \equiv B_n^{(p)}(k_{Lq}) : \Lambda_h \) and \( D_n^{(p)}(k_{Lq}) \equiv 2[(d+2)(d-1)]^{-1} B_n^{(p)}(k_{Lq}) : \Lambda_s \).

Note that \( C_n^{(p)}(k_{Lq}) \) and \( D_n^{(p)}(k_{Lq}) \) involve the powers \( \kappa_{pq}^m \mu_{pq}^{n-m} \), where an integer \( m \) lies between 0 and \( n \).

Assuming that the composite is passive (i.e., it does not generate mechanical energy), and the time-harmonic factor of waves is \( e^{-i\omega t} \), the imaginary parts of the effective elastic moduli must be non-positive, implying that

\[
\text{Im}[K_e(k_{Lq})] \leq 0, \quad \text{Im}[G_e(k_{Lq})] \leq 0,
\]

for any non-negative \( k_{Lq} \). In light of these properties, we have

\[
\text{Im} \left[ \kappa_{pq} \sum_{n=2}^{\infty} C_n^{(p)}(k_{Lq}) \right] \leq 0, \quad \text{Im} \left[ \mu_{pq} \sum_{n=2}^{\infty} D_n^{(p)}(k_{Lq}) \right] \leq 0. \tag{9.40}
\]
The effective elastic wave characteristics, including wave speeds $c_e^{L,T}$ and attenuation coefficients $\gamma_e^{L,T}$, are directly related to the effective moduli as follows:

\begin{align*}
c_e^L + i\gamma_e^L &\equiv \sqrt{[K_e(k_{Lq}) + 2(1 - 1/d) G_e(k_{Lq})]}/\rho_e, \quad (9.41) \\
c_e^T + i\gamma_e^T &\equiv \sqrt{G_e(k_{Lq})}/\rho_e, \quad (9.42)
\end{align*}

where $\rho_e = \rho_p = \rho_q$, and the superscripts $L$ and $T$ denote longitudinal and transverse waves, respectively. Note that $\exp(-2\pi\gamma_e^L/c_e^L)$ and $\exp(-2\pi\gamma_e^T/c_e^T)$ represent the factors by which the amplitudes of the incident waves are attenuated inside the composite for a period of time $2\pi/\omega$. Thus, if $\gamma_e^L = \gamma_e^T = 0$ at certain wavenumbers (or frequencies), the composite is perfectly transparent, i.e., elastic waves propagate without any loss.

**Remarks:**

1. Any statistically isotropic medium is macroscopically isotropic, but the converse is not true. For example, while cubic lattice packings are statistically anisotropic, they are macroscopically isotropic due to the cubic symmetry (see Sec. 9.4.2).

2. The dynamic strong-contrast expansions represented by (9.38) and (9.39) possess fast-convergence properties for a wide class of microstructures, even at extreme phase contrast ratios (see Ref. [308] for detailed explanations). Such convergence properties are attributed to the following two aspects. First, even for extreme contrast ratios $K_p/K_q$ or $G_p/G_q$, the two expansion parameters $\kappa_{pq}$ and $\mu_{pq}$ are bounded by

\begin{align*}
-\infty < -\left[\frac{d^2(d - 1)}{1 + \nu_q} - d(d - 1)^2\right]^{-1} &\leq \kappa_{pq} < 1, \\
-\frac{2d}{(d - 2)(d + 1)} \leq \left[\frac{d}{2} - \frac{3(d + 2)(2\nu_q - 1)}{2d(5\nu_q - 4)}\right]^{-1} &\leq \mu_{pq} < 1,
\end{align*}
where \( \nu_q \) is the Poisson ratio of the reference phase \( q \). Secondly, as Torquato [293, 292] observed, the strong-contrast expansions in the static limit can be regarded to be ones that perturb around the wide class of optimal structures [293, 292, 305]. The reader is referred to Refs. [293, 292] for details. It suffices to note here that such optimal two-phase media are characterized by a disconnected dispersed phase that is distributed throughout a connected matrix. These observations imply that the first few terms of the expansions (9.38) and (9.39) can yield accurate approximations of the effective properties for a class of particulate composites as well as more general microstructures, even for extreme contrast ratios, provided that the dispersed phase is prevented from forming large clusters compared to the specimen size. Depending on whether the high-stiffness phase percolates or not, this broad microstructure class includes particulate media consisting of identical or polydisperse particles of general shape (ellipsoids, cubes, cylinders, polyhedra) that may or not overlap, cellular networks [305] as well as media without well-defined inclusions. The reader is referred to Ref. [308] for a more complete discussion of this issue.

### 9.3 Approximations at the Two- and Three-Point Levels

Due to the fast-convergence properties of strong-contrast expansions, their truncations at low orders should yield accurate approximations for the effective bulk and shear moduli for the aforementioned wide class of microstructures over a broad range of volume fractions and contrast ratios; see also Ref. [308] for additional details. In what follows, we present such approximations by truncating the strong-contrast expansions after the second- and third-order terms, respectively. Detailed derivations are provided in Sec. I in the SM.
9.3.1 Strong-Contrast Approximations at the Two-Point Level

Truncating (9.38) and (9.39) at the two-point level and solving the left-hand sides of these truncated series for $K_e$ and $G_e$, respectively, yields

\[
\frac{K_e(k_{Lq})}{K_q} = 1 + \left[ \frac{c_{Lq}^2}{c_{Lq}^2 - 2(1 - 1/d) c_{Tq}^2} \right] \frac{\phi_p^2 \kappa_{pq}}{\phi_p(1 - \phi_p \kappa_{pq}) - C_2^{(p)}(k_{Lq})},
\]

(9.43)

\[
\frac{G_e(k_{Lq})}{G_q} = 1 + \left[ \frac{d(d + 2) c_{Lq}^2/2}{d c_{Lq}^2 + 2 c_{Tq}^2} \right] \frac{\phi_p^2 \mu_{pq}}{\phi_p(1 - \phi_p \mu_{pq}) - D_2^{(p)}(k_{Lq})},
\]

(9.44)

where $C_2^{(p)}(k_{Lq})$ and $D_2^{(p)}(k_{Lq})$ are defined respectively as

\[
C_2^{(p)}(k_{Lq}) = \frac{\pi}{2^{d/2} \Gamma(d/2)} F(k_{Lq}) \kappa_{pq},
\]

(9.45)

\[
D_2^{(p)}(k_{Lq}) = \frac{\pi}{2^{d/2} \Gamma(d/2)} \frac{d c_{Lq}^2 F(k_{Tq}) + 2 c_{Tq}^2 F(k_{Lq})}{d c_{Lq}^2 + 2 c_{Tq}^2} \mu_{pq},
\]

(9.46)

and the local attenuation function $F(Q)$ is defined as

\[
F(Q) \equiv -\frac{2^{d/2} \Gamma(d/2)}{\pi} Q^2 \int \frac{i}{4} \left( \frac{Q}{2 \pi r} \right)^{d-2} \mathcal{H}_{d/2-1}(Qr) \chi_V(r) \, dr
\]

(9.47)

\[
= -\frac{\Gamma(d/2)}{2^{d/2} \pi^{d+1}} Q^2 \int \frac{\tilde{\chi}_V(\mathbf{q})}{|\mathbf{q}|^2 - Q^2} \, d\mathbf{q},
\]

(9.48)

where $\Gamma(x)$ is the gamma function, $\chi_V(r) \equiv S_2^{(p)}(r) - \phi_p^2$ is the radial autocovariance function, and the spectral density $\tilde{\chi}_V(Q)$ is its Fourier transform. Some important properties of $F(Q)$ are given in Appendix 9.9. We discuss how to evaluate $F(Q)$ in Appendix 9.13. Use of these properties of $F(Q)$ immediately show that in the static limit ($\omega = 0$), the parameters $C_2^{(p)}(0)$ and $D_2^{(p)}(0)$ are identically zero, which is consistent with previous studies [294, 296].

Remarkably, $F(Q)$ also appears in the quasistatic strong-contrast approximations for the electromagnetic characteristics [242, 308]. This commonality between the two
wave problems at the two-point level allowed us to establish cross-property relations for the effective elastic and electromagnetic wave characteristics in Ref. [158]; see Chapter 10.

### 9.3.2 Strong-Contrast Approximations at the Three-Point Level

Truncating (9.38) and (9.39) at the three-point level and solving the left-hand sides of these truncated series for $K_e$ and $G_e$, respectively, yields

\[
\frac{K_e(k_{Lq})}{K_q} = 1 + \left[ \frac{c_{Lq}^2}{c_{Lq}^2 - 2(1 - 1/d)c_{Tq}^2} \right] \frac{\phi_p \frac{\phi_p^2 \kappa_{pq}}{\mu_{pq}}}{\phi_p (1 - \phi_p \mu_{pq}) - [C_2^{(p)}(k_{Lq}) + C_3^{(p)}(k_{Lq})]},
\]

(9.49)

\[
\frac{G_e(k_{Lq})}{G_q} = 1 + \left[ \frac{d(d + 2) c_{Lq}^2 / 2}{d c_{Lq}^2 + 2 c_{Tq}^2} \right] \frac{\phi_p \frac{\phi_p^2 \mu_{pq}}{\mu_{pq}}}{\phi_p (1 - \phi_p \mu_{pq}) - [D_2^{(p)}(k_{Lq}) + D_3^{(p)}(k_{Lq})]},
\]

(9.50)

where the explicit formulas for the three-point parameters $C_3^{(p)}(k_{Lq})$ and $D_3^{(p)}(k_{Lq})$ are given respectively as

\[
C_3^{(p)}(k_{Lq}) = -\frac{k_{Lq}^{d+2}}{2^{d+2 - 1} \pi^{d-2}} \int d \mathbf{r} \int d \mathbf{s} \left\{ \kappa_{pq} \mu_{pq} \frac{(d + 2) c_{Tq}^2}{d c_{Lq}^2 + 2 c_{Tq}^2} \mathcal{H}_{d/2+1}^{(1)}(k_{Lq} \mathbf{r}) \mathcal{H}_{d/2+1}^{(1)}(k_{Lq} \mathbf{s}) \right. \\
\left. \times \mathcal{P}_2(\mathbf{r} \cdot \mathbf{s}) + \kappa_{pq}^2 \mathcal{H}_{d/2-1}^{(1)}(k_{Lq} \mathbf{r}) \mathcal{H}_{d/2-1}^{(1)}(k_{Lq} \mathbf{s}) \right\} \left[ S_3^{(p)}(\mathbf{r}, \mathbf{s}, \mathbf{t}) - \frac{S_2^{(p)}(\mathbf{r}) S_2^{(p)}(\mathbf{s})}{\phi_p} \right],
\]

(9.51)
\[ D_3^{(p)}(k_{Lq}) = \frac{2}{d-1} \frac{k_{Lq}^{d+2}}{22^d \pi^{d-2} d c_{Lq}^2 + 2 c_{Tq}^2} \int \int \frac{dr}{r^{d/2-1}} \frac{ds}{s^{d/2-1}} \]

\[ \times \left( - \kappa_{pq} \hat{P}_2(\hat{r} \cdot s) \mathcal{H}_{d/2+1}^{(1)}(k_{Lq} r) \frac{\mathcal{H}_{d/2+1}^{(1)}(k_{Lq} s)}{(c_{Tq} / c_{Lq})^{d/2+3}} \right) \]

\[ \times \left( - \kappa_{pq} \hat{P}_2(\hat{r} \cdot s) \mathcal{H}_{d/2+1}^{(1)}(k_{Lq} r) \frac{\mathcal{H}_{d/2+1}^{(1)}(k_{Lq} s)}{(c_{Tq} / c_{Lq})^{d/2+3}} \right) \]

\[ \times \left[ \frac{\mathcal{H}_{d/2+1}^{(1)}(k_{Lq} s)}{(c_{Tq} / c_{Lq})^{d/2+3}} \right] \]

\[ + \frac{d-2}{4(d+4)} \hat{P}_2(\hat{r} \cdot s) \left[ 4 \mathcal{H}_{d/2+1}^{(1)}(k_{Lq} r) + d \frac{\mathcal{H}_{d/2+1}^{(1)}(k_{Tq} r)}{(c_{Tq} / c_{Lq})^{d/2+3}} \right] \]

\[ \times \left[ 4 \mathcal{H}_{d/2+1}^{(1)}(k_{Lq} s) + d \frac{\mathcal{H}_{d/2+1}^{(1)}(k_{Tq} s)}{(c_{Tq} / c_{Lq})^{d/2+3}} \right] \]

\[ + \frac{d-1}{2(d+2)} \left[ 2 \mathcal{H}_{d/2-1}^{(1)}(k_{Lq} r) + d \frac{\mathcal{H}_{d/2-1}^{(1)}(k_{Tq} r)}{(c_{Tq} / c_{Lq})^{d/2+3}} \right] \]

\[ \times \left[ 2 \mathcal{H}_{d/2-1}^{(1)}(k_{Lq} s) + d \frac{\mathcal{H}_{d/2-1}^{(1)}(k_{Tq} s)}{(c_{Tq} / c_{Lq})^{d/2+3}} \right] \}

\[ \times \left[ S_3^{(p)}(\mathbf{r}, \mathbf{s}, \mathbf{t}) - \frac{S_2^{(p)}(\mathbf{r}) S_2^{(p)}(\mathbf{s})}{\phi_p} \right] \]

Here \( \mathbf{t} \equiv \mathbf{r} - \mathbf{s} \), and

\[ \hat{P}_4(t) \equiv t^4 - \frac{6}{d+4} t^2 + \frac{3}{(d+2)(d+4)}, \]  

\[ \hat{P}_2(t) \equiv dt^2 - 1. \]
In the static limit ($\omega = 0$), the three-point parameters (9.51) and (9.52) reduce to

\[
C_3^{(p)}(0) = \frac{(d + 2) c_{T_q}^2}{d c_{L_q}^2 + 2 c_{T_q}^2} \frac{\kappa_{pq} \mu_{pq}}{\pi^d} \int \int \frac{dr\, ds}{r^d s^d} \frac{\Gamma(d/2 + 1)^2}{\pi^d} \left[ S_3^{(p)}(r, s) - \frac{S_2^{(p)}(r) S_2^{(p)}(s)}{\phi_p} \right]
\]

\[
= \frac{(d - 1)(d + 2) c_{T_q}^2}{d c_{L_q}^2 + 2 c_{T_q}^2} \kappa_{pq} \phi_q \phi_p \zeta_p,
\]

(9.55)

\[
D_3^{(p)}(0) = \frac{2}{d - 1} \frac{c_{T_q}^2}{d c_{L_q}^2 + 2 c_{T_q}^2} \frac{\Gamma(d/2 + 1)^2}{\pi^d} \int \int \frac{dr\, ds}{r^d s^d} \left[ \kappa_{pq} \hat{P}_2(\hat{r} \cdot \hat{s}) + \frac{(d + 2) c_{T_q}^2}{d c_{L_q}^2 + 2 c_{T_q}^2} \mu_{pq} \right]
\]

\[
\times \left\{ \frac{d - 2}{d + 4} \frac{d c_{L_q}^2 + 4 c_{T_q}^2}{4 c_{T_q}^4} \hat{P}_2(\hat{r} \cdot \hat{s}) + \frac{d^2(d + 2)^2}{4} \frac{(c_{L_q}^2 - c_{T_q}^2)^2}{c_{T_q}^4} \hat{P}_4(\hat{r} \cdot \hat{s}) \right\}
\]

\[
\times \left[ S_3^{(p)}(r, s, t) - \frac{S_2^{(p)}(r) S_2^{(p)}(s)}{\phi_p} \right]
\]

\[
= \frac{2 c_{T_q}^2}{d c_{L_q}^2 + 2 c_{T_q}^2} \phi_p \phi_q \mu_{pq} \left\{ \kappa_{pq} \zeta_p + \frac{(d + 2) c_{T_q}^2 \mu_{pq}}{4 d c_{L_q}^2 + 2 c_{T_q}^2} \left[ \frac{d(d - 2)}{4} \frac{2 c_{L_q}^2 - (1 - 4/d) c_{T_q}^2}{c_{T_q}^2} \zeta_p \right.ight.
\]

\[
\left. \left. + \frac{d^8}{4(d + 2)} \frac{(c_{L_q}^2 - c_{T_q}^2)^2}{c_{T_q}^4} \eta_p \right] \right\},
\]

(9.56)

where the parameters $\eta_p$ and $\zeta_p$ lie in a closed interval $[0, 1]$; see Ref. [296] and reference therein.

### 9.4 Improved Approximations at the Two-point Level

In order to extend the series expansions and approximations discussed in Sec. 9.2.1-9.3 beyond the quasistatic regime, one needs to generalize the strong-contrast expansion formalism to theories of nonlocal elasticity (see a recent review [140]) from first principles, as we did for the electrodynamic problem in Ref. [308]. Unlike the dielectric problems, however, such generalizations are nontrivial in the case of the elastodynamic problem because an elastically isotropic medium generally possesses multiple elastic wavenumbers at a given frequency $\omega$. 

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9.4.1 Nonlocal Strong-Contrast Approximation

Based on the following two observations, we postulate nonlocal strong-contrast approximations for the effective elastodynamic properties at the two-point level that are expected to be accurate beyond the quasistatic regime. First, the local strong-contrast expansions for the elastodynamic and electromagnetic problems are similar in that the local attenuation function \( F(Q) \), given by (9.47), appears in the local strong-contrast approximations of the effective dielectric constant that was rigorously derived in Ref. [242]; see also Ref. [308]. Second, guided by our exact formulation of the nonlocal effective electromagnetic characteristics [308], such generalizations at the two-point level are tantamount to replacing the wavenumber-dependent local attenuation function \( F(Q) \), defined in (9.47), with the wavevector-dependent nonlocal attenuation function \( F(Q) \) defined by [158, 308]

\[
F(Q) \equiv -\frac{2^{d/2} \Gamma(d/2)}{\pi} Q^2 \int \frac{i}{4} \left( \frac{Q}{2\pi r} \right)^{d/2-1} \mathcal{H}^{(1)}_{d/2-1}(Qr) e^{-iQ \cdot r} \chi_v(r) \, dr \tag{9.57}
\]

\[
= -\frac{\Gamma(d/2)}{2^{d/2} \pi^{d+1}} Q^2 \int \frac{\bar{\chi_v}(q)}{|q + Q|^2 - Q^2} \, dq. \tag{9.58}
\]

Unlike \( F(Q) \), \( F(Q) \) accounts for the contribution from spatial variation of the sinusoidal incident waves \( \exp(-iQ \cdot r) \) and thus more accurately estimates the scattering effects of waves associated with wavevector \( Q \) from the long- to intermediate-wavelength regimes. (Important properties of \( F(Q) \) for a statistically isotropic medium are provided in Appendix 9.9.) From these two observations, it is reasonable to assume that one can extend the range of applicable wavelengths by replacing \( F(Q) \) in the local strong-contrast approximations at the two-point level [Eqs. (9.43) and (9.44)] with \( F(Q) \), which are numerically verified in Sec. 9.4.2. The resulting ap-
proximations are given respectively by

\[ K_e(k_{Lq}) = 1 + \left[ \frac{c_{Lq}^2}{c_{Lq}^2 - 2(1 - 1/d) c_{Tq}^2} \right] \frac{\phi_p^2 \kappa_{pq}}{\phi_p (1 - \phi_p \kappa_{pq}) - \frac{2\pi}{2^{d/2} \Gamma(d/2)} F(k_{Lq}) \kappa_{pq}}, \tag{9.59} \]

\[ G_e(k_{Lq}) = 1 + \left[ \frac{d(d + 2) c_{Lq}^2 / 2}{d c_{Lq}^2 + 2 c_{Tq}^2} \right] \frac{\phi_p^2 \mu_{pq}}{\phi_p (1 - \phi_p \mu_{pq}) - \frac{\pi}{2^{d/2} \Gamma(d/2)} \frac{d c_{Lq}^2 F(k_{Tq}) + 2 c_{Lq}^2 F(k_{Lq})}{d c_{Lq}^2 + 2 c_{Tq}^2} \mu_{pq}}, \tag{9.60} \]

where \( k_{Lq} \) and \( k_{Tq} \) are the longitudinal and transverse wavevectors of the incident waves, respectively, and \( F(Q) \) is given in (9.57). We emphasize that the nonlocal strong-contrast approximations for both elastic and electromagnetic properties share a common microstructure-dependent parameter \( F(Q) \), which enabled us to establish cross-property relations linking those properties in Ref. [158]. Note that, as we shown in a recent paper [308], the analytic properties of \( F(|Q|) \) lead the nonlocal approximations (9.59) and (9.60) satisfies Kramers-Kronig relations for elastic waves [27, 226]. (These nonlocal approximations were first postulated in Ref. [158] on physical grounds for establishing the cross-property relations.)

For a statistically isotropic composite, as shown in Chapter 8, \( F(Q) \) becomes a radial function of wavenumber \( Q \) and is written as [158, 308]

\[ \text{Im}[F(Q)] = \begin{cases} \frac{-Q^2}{\pi^2} \int_0^{\pi/2} \tilde{X}_v(2Q \cos \phi) \, d\phi, & d = 2 \\ \frac{-Q}{2(2\pi)^{3/2}} \int_0^{2Q} q \, \tilde{X}_v(q) \, dq, & d = 3 \end{cases} \tag{9.61} \]

\[ \text{Re}[F(Q)] = -\frac{2Q^2}{\pi} \text{p.v.} \int_0^\infty dq \frac{1}{q(Q^2 - q^2)} \text{Im}[F(q)], \tag{9.62} \]

where Eq. (9.62) is valid for \( d = 2, 3 \). As shown in (9.59) and (9.60), the imaginary part of \( F(|Q|) \) directly determines the degree of attenuation, i.e., \( \text{Im}[K_e] \) and \( \text{Im}[G_e] \) or, equivalently, \( \gamma_e^L \) and \( \gamma_e^T \) defined in (9.41) and (9.42). In the quasistatic regime, as-
Figure 9.2: Comparison of the predictions of the local strong-contrast approximations [Eqs. (9.43) and (9.44)], the nonlocal variants [Eqs. (9.59) and (9.60)] and GUA [Eqs. (9.66) and (9.67)] for the effective dynamic bulk $K_e(k_{Lq})$ and shear $G_e(k_{Lq})$ moduli of periodic packings to the corresponding simulation results. We consider 3D cubic lattice packing of packing fraction $\phi_2 = 0.05$, contrast ratios $K_2/K_1 = G_2/G_1 = 2$, and Poisson ratio $\nu_1 = 1/3$. Here, $k_{Lq}$ is the longitudinal wavenumber in the reference phase along the $\Gamma$-$X$ direction, and $L$ is the nearest-neighbor distance.

Assuming that the spectral density has the power-law scaling $\tilde{\chi}_V(Q) \sim Q^\alpha$, the effective attenuation coefficients $\gamma_{e,T}^L(k_{Lq})$ exhibit

$$\gamma_{e,T}^L(k_{Lq}) \sim \text{Im}[F(k_{Lq})]$$

$$\sim \begin{cases} 
  k_{Lq}^3, & \text{nonhyperuniform } (\alpha = 0) \\
  k_{Lq}^{3+\alpha}, & \text{hyperuniform } (\alpha > 0)
\end{cases}, \text{ as } k_{Lq} \to 0^+,$$

where nonhyperuniform systems take $\alpha = 0$, whereas hyperuniform ones take $\alpha > 0$ (see Appendix 9.9). Thus, hyperuniform media are less lossy than their nonhyperuniform counterparts as the wavenumber tends to zero. Remarkably, the stealthy hyperuniform media are perfectly transparent up to a finite wavenumber:

$$\gamma_{e,T}^L(k_{Lq}) = 0, \text{ if } 0 \leq k_{Lq} \leq \frac{c_{Tq}}{c_{Lq}} \frac{Q_U}{2},$$

where $c_{Tq}/c_{Lq} = \sqrt{(1-2\nu_q)/[2(1-\nu_q)]}$, and $\nu_q$ is the Poisson ratio of the reference phase $q$. 
9.4.2 Comparison of Simulations to Various Approximations

Here we compare various approximations formulas for the effective dynamic elastic moduli to computer simulations, which are highly nontrivial calculations. In particular, we utilize our fast-Fourier-transform (FFT) numerical scheme presented elsewhere [158]. This procedure extends the one first devised for the effective static elastic moduli [217] in order to treat elastodynamics. The reader is referred to the SM and Ref. [158] for details.

In order to ensure convergence of the simulation procedure, we choose to study simple cubic lattice packings in which identical spheres of radius $a$ of phase 2 are embedded in the matrix phase (phase 1). While the periodic packings are macroscopically isotropic, due to cubic symmetry, they are statistically anisotropic, implying that effective properties can depend on the direction of the incident wave $k_{L1}$. For simplicity, we only consider the case where $k_{L1}$ is aligned with one of the minimal lattice vectors, i.e., $\Gamma$-$X$ direction in the first Brillouin zone. Simple cubic lattice packings also provide stringent tests of the predictive power of the approximations at finite wavenumbers because they exhibit two salient and nontrivial elastic properties due to spatial correlations at intermediate length scales: transparency up to finite wavenumbers associated with the edges of the first Brillouin zone (i.e., $\text{Im}[K_e] = 0$ for $0 \leq k_{L1} \lesssim \pi$ and $\text{Im}[G_e] = 0$ for $0 \leq k_{T1} \lesssim \pi$), and resonance-like attenuation due to Bragg diffraction within the phononic bandgap (i.e., a peak in the imaginary parts or, equivalently, a sharp transition in the real parts).

We perform simulations for the case of simple cubic lattice of spheres in a matrix in which the packing fraction is $\phi_2 = 0.05$, contrast ratios are $K_2/K_1 = G_2/G_1 = 2$, and the Poisson ratio of the reference phase is $\nu_1 = 1/3$. In Fig. 9.2, we compare the simulation results to the predictions from the strong-contrast approximations [Eqs. (9.43) and (9.44) for local approximations, and Eqs. (9.59) and (9.60) for the nonlocal counterparts] as well as the Gaunaurd-Überall approximation (GUA) [(9.66)]
and (9.67)]. While all approximations agree with the simulations in the quasistatic regime, the GUA and local strong-contrast approximations fail to capture properly two key features: no loss of energy up to finite wavenumbers and resonance-like attenuation in the band gaps. However, the nonlocal strong-contrast approximations capture these two features and agree well with the simulation results, even beyond the quasistatic regime.

9.5 Model Microstructures

Here, we describe the four models of 3D disordered two-phase media that are statistically isotropic to study the microstructure-dependence of effective elastic properties:

- Overlapping spheres;
- Equilibrium packings;
- Class I hyperuniform polydisperse packings; and
- Disordered stealthy hyperuniform packings.

The models include two nonhyperuniform systems (overlapping spheres and equilibrium packings) and two hyperuniform systems (class I hyperuniform polydisperse packings and stealthy hyperuniform packings). In each mode, spherical particles of phase 2 are distributed throughout a matrix phase (phase 1). Detailed description of each model is provided in Sec. 8.3.
Figure 9.3: Predictions of scaled effective (a) longitudinal and (b) transverse wave characteristics, $c_{eL,T}^e$ and $\gamma_{eL,T}^e$, from approximations (9.59) and (9.60) for the four 3D models of disordered composites of spheres of radius $a$ and $\phi_2 = 0.25$. The Poisson ratios of the matrix and dispersed phases are $\nu_1 = 0.4$ and $\nu_2 = 0.25$, respectively, and the phase contrast ratios are $K_2/K_1 = 10$, $G_2/G_1 = 28$, which correspond to glass beads in an epoxy matrix [263]. Here, $k_{L_1}$ is the longitudinal wavenumber in the reference phase, and $c_{eL}$ and $c_{eT}$ are longitudinal and transverse wave speeds, respectively. The insets in the lower panels are log-log plots of the respective larger panels.

9.6 Predictions from Strong-Contrast Approximations

Having established the accuracy of the nonlocal strong-contrast approximations, (9.59) and (9.60), for simple cubic lattice packings in Sec. 9.4.2, we now apply them to predict the effective elastodynamic characteristics of the four different disordered models discussed in Sec. 9.5. Specifically, we study how the effective elastic moduli $[K_e(k_{L_1}), G_e(k_{L_1})]$, wave speeds $c_{eL,T}^e(k_{Lq})$, and attenuation coefficients $\gamma_{eL,T}^e(k_{Lq})$ vary with the microstructure. For simplicity, we take the matrix phase to be the reference phase (phase 1) and the dispersed phase to be the polarized phase (phase 2).

Figure 9.3 shows the scaled effective wave characteristics [i.e., $c_{eL}^e/c_{eL}$ and $\gamma_{eL}^e/c_{eL}$ for longitudinal waves and $c_{eT}^T/c_{eT}$ and $\gamma_{eT}^T/c_{eT}$ for transverse waves] vary with $k_{Lq}$ at fixed phase properties $K_2/K_1 = 10$, $G_2/G_1 = 28$, and $\nu_1 = 0.4$ for the four models. While all models are effectively lossless (i.e., small values of $\gamma_e$) for a range of
wavenumber around the origin, they become increasingly lossy as the wavenumber increases; see the lower panels of Fig. 9.3. In the quasistatic regime, as shown in the insets of Fig. 9.3, hyperuniform and nonhyperuniform exhibit qualitatively different attenuation characteristics [cf. (9.63)]: hyperuniform composites generally tend to be less lossy than their nonhyperuniform counterparts. Remarkably, stealthy hyperuniform media can be perfectly lossless, even well beyond the quasistatic regime; see Eq. (9.64). Such microstructure-dependence of the effective attenuation behaviors vividly demonstrates that \( \gamma_{L,T} \) can be engineered by the spatial correlations of composites.

Figure 9.4: Predictions of the nonlocal strong-contrast approximation (9.60) for the negative of the imaginary part of the effective shear modulus \( \text{Im}[G_e(k_{Lq})] \) of the four disordered models, as per Fig. 9.3, as function of contrast ratio \( G_2/G_1 \) at volume fraction \( \phi_2 = 0.25 \) and wavenumber \( k_{L1} a = 0.3 \). The Poisson ratios of the matrix and dispersed phases are fixed at \( \nu_1 = 0.4 \) and \( \nu_2 = 0.25 \).

We now examine how the imaginary part \( \text{Im}[G_e] \) varies with the contrast ratio \( G_2/G_1 \) for the disordered models for a given large wavenumber \( k_{Lq} \) inside the transparency interval (wavenumber ranges where the imaginary parts of the effective bulk and shear moduli vanish) given in (9.64) for the stealthy hyperuniform packing. Here, we fix the phase Poisson ratios to be \( \nu_1 = 0.4 \) and \( \nu_2 = 0.25 \), as we did for the case shown in Fig. 9.3. These results are summarized in Fig. 9.4. The disparity in the attenuation characteristics across microstructures widens significantly as the contrast
ratio increases. Clearly, overlapping spheres are the lossiest systems. Hyperuniform polydisperse packings can be nearly as lossless as stealthy hyperuniform ones. Unlike the imaginary part, the real part \( \text{Re}[G_e] \) is virtually independent of model microstructure and thus is not shown in this chapter. We also do not include the corresponding plot for \( K_e \) because its behavior is qualitatively similar to that of \( G_e \).

![Graphs showing predictions of the nonlocal strong-contrast approximations for effective bulk and shear moduli, and effective Poisson ratio as a function of \( k_{L_1} \).](image)

Figure 9.5: Predictions of the nonlocal strong-contrast approximations (9.59) and (9.60) for the effective (a) bulk \( K_e \) and (b) shear \( G_e \) moduli, and (c) effective Poisson ratio \( \nu_e \) as a function of \( k_{L_1} \) for 3D stealthy hyperuniform packings of contrast ratio \( G_2/G_1 = 10 \) at two different packing fractions: \( \phi_2 = 0.4 \) and \( Q_{UA} = 1.5 \) and \( \phi_2 = 0.25 \) and \( Q_{UA} \approx 1.33 \). The Poisson ratios of the matrix and dispersed phases are \( \nu_1 = 1/3 \) and \( \nu_2 = -1 \) (i.e., \( K_2/K_1 = 0 \)), respectively. In the lower panels of each figure, the negatives of the corresponding loss tangents [cf. (9.65)] are plotted. The insets in (a) and (b) are magnifications of the respective lower panels.

Since stealthy hyperuniform packings exhibit novel physical properties, such as perfect transparency, we further study the effect of packing fraction \( \phi_2 \) on their effective elastic moduli \( K_e(k_{L_1}) \) and \( G_e(k_{L_1}) \) and the effective Poisson ratio \( \nu_e(k_{L_1}) \). Specifically, we are interested in examining stealthy hyperuniform packings consist-
ing of auxetic particles of $\nu_2 = -1$ and a matrix phase with $\nu_1 = 1/3$ and $G_2/G_1 = 10$. Auxetic (negative Poisson ratio) materials laterally dilate (shrink) in response to axial elongation (contraction) [172], and are known to have superior energy-absorbing properties [260]. We first generate such packings at a packing fraction $\phi_2 = 0.4$ and $Q_U a = 1.5$, as described in Sec. 8.3.4. Without changing particle positions, we then shrink the sphere radii to attain a packing fraction $\phi_2 = 0.25$, whose stealthy regime is now $Q_U a \approx 1.33$.

In Fig. 9.5, we plot the effective bulk and shear moduli as well as the effective Poisson ratio using approximations (9.59) and (9.60), and Eq. (9.10). To quantify the damping characteristics of such composites, we also include in this figure the corresponding loss tangents defined by

$$\tan \delta_{X_e} \equiv \frac{\text{Im}[X_e]}{\text{Re}[X_e]},$$

for some general effective property $X_e$, which are frequently measured in experiments. For the bulk and shear moduli, the loss tangents represent the ratios of mechanically attenuated energy to the stored elastic energy [5]. We see that these stealthy dispersions are effectively auxetic, i.e., $\text{Re}[\nu_e] < 0$ [see Fig. 9.5(c)]. Figure 9.5(a) reveals that such stealthy auxetic composites have exceptionally large loss tangent values in the intermediate-wavelength regime, compared to typical values ($\lesssim 10^{-1}$ as in the cases in Fig. 9.3), which implies that they are excellent energy absorbers, as expected. The transparency interval (i.e., $\tan \delta_{K_e} = \tan \delta_{G_e} = 0$) is slightly larger for the higher density packing with the higher stealthy cut-off value $Q_U a = 1.5a$, as predicted by Eq. (9.64). The complex Poisson ratio implies that the lateral and axial vibrations are out of phase. The absolute value of $\tan \delta_{\nu_e}$ is approximately proportional to the difference between the shear and bulk loss factors (i.e., degrees of energy loss due to shear and compression); see Ref. [237].
9.7 Conclusions and Discussion

Closed-form approximations of the effective dynamic elastic moduli derived previously only apply at long wavelengths (quasistatic regime) and for very special macroscopically isotropic disordered composite microstructures [153], namely, nonoverlapping spheres or spheroids in a matrix. In this chapter, we have provided the theoretical underpinnings to substantially extend previous work in both its generality and applicability. First, we derived exact homogenized constitutive relations for the effective dynamic elastic stiffness tensor \( C_e(k_{Lq}) \) from first principles that are local in space. Second, our strong-contrast representation of \( C_e(k_{Lq}) \) exactly accounts for complete microstructural information (\( n \)-point correlation functions for \( n \geq 1 \)) for general microstructures and hence multiple scattering to all orders in the quasistatic regime. Third, we extracted from the exact expansions accurate local closed-form approximate formulas for \( K_e(k_{Lq}) \) and \( G_e(k_{Lq}) \), relations (9.59) and (9.60), which are resummed representations of the exact expansions that incorporate microstructural information through the spectral density \( \tilde{\chi}_V(Q) \), which is easily ascertained for general microstructures either theoretically, computationally or via scattering experiments. Depending on whether the high-stiffness phase percolates or not, the wide class of microstructures that we can treat includes particulate media consisting of identical or polydisperse particles of general shape (ellipsoids, cubes, cylinders, polyhedra) with prescribed orientations that may or not overlap, cellular networks as well as media without well-defined inclusions (Sec. 9.2). Fourth, we extended these local approximations beyond the quasistatic regime by postulating nonlocal formulas based on the similarities between electrodynamic and elastodynamic problems and our rigorous formulation of the nonlocal effective dynamic dielectric properties [308]. We carried out precise full-waveform elastodynamic simulations for certain 3D benchmark models to validate the accuracy of our nonlocal formulas for wavenumbers well beyond the quasistatic regime, i.e., \( 0 \leq k_{Lq} \ell \lesssim 1 \) (where \( \ell \) is a characteristic heterogeneity.
Having verified the accuracy of the postulated strong-contrast approximations (9.59) and (9.60) for dispersions, we then applied them to the four disordered model microstructures in three dimensions (both nonhyperuniform and hyperuniform) to investigate the microstructure-dependence of the effective elastic wave characteristics. We demonstrated that disordered hyperuniform media are generally less lossy than their nonhyperuniform counterparts. We also found that our approximations predict that disordered hyperuniform media possess a transparency wavenumber interval (9.64) around which most nonhyperuniform media exhibit strong attenuation. We note that using finite-element method calculations and supercell techniques, Gkantzounis, Amoah, and Florescu [102] showed that 2D stealthy hyperuniform packings should exhibit a transparency interval for elastic waves, which are qualitatively consistent with our predictions.

The accuracy of our nonlocal closed-form formulas has important practical implications since one can now use them to accurately and efficiently predict the effective wave characteristics well beyond the quasistatic regime of a wide class of composite microstructures without carrying out computationally expensive full-blown simulations. Thus, our nonlocal formulas can be used to accelerate the discovery of novel elastodynamic composites by appropriate tailoring of the spectral densities and then constructing the corresponding microstructures by using Fourier-space inverse methods [46]. For example, from our findings, it is clear that stealthy disordered particulate media can be utilized as low-pass filters that transmit elastic waves “isotropically” up to a selected wavenumber. Of course, one could also explore the design space of effective elastic wave properties of nonhyperuniform disordered composite media for potential applications.

There are interesting open problems for future exploration. Could the exact local strong-contrast expansions, such as (9.31) and (9.37), be generalized to the cases in
which the mass densities of both phases are different, i.e., $\rho_1 \neq \rho_2$? This is a highly nontrivial extension. One possible approach to answer this question is to introduce the concept of the dynamic matrix, which is used to derive dispersion relations for elastic waves in simple harmonic lattices [7] in order to separate the local mass density $\rho(\mathbf{x})$ and displacement field $\mathbf{u}(\mathbf{x})$. Another challenging problem is the derivation of strong-contrast expansions of the effective dynamic elastic moduli from the first principles in the manner obtained for the electromagnetic problem [308]. This problem is also quite challenging partly because, unlike the electromagnetic waves, one needs to account for the interplay between longitudinal and transverse propagation modes of elastic waves at a given frequency. Finally, it desirable to formulate full-waveform elastodynamic simulations for two-phase media that are more efficient than the dynamic FFT scheme used here.

9.8 Appendix A: Gaunaurd-Überall Approximation

Here we state explicit formulas for the Gaunaurd-Überall approximation (GUA) for the effective dynamic elastic moduli of an isotropic medium composed of identical spheres of radius $a$ in the quasistatic regime. The particles are in phase 2 of mass density $\rho_2$ and elastic moduli $K_2$ and $G_2$, and they are embedded in a matrix phase of $\rho_1$, $K_1$ and $G_1$.

Since the GUA accounts solely for scatterings from a single particle in the mean-field treatments, it can be regarded as the elastodynamic counterpart of the Maxwell-Garnett approximation [253]. The explicit formulas for the effective bulk and shear
moduli are given respectively as [153]

\[
\frac{K_e(k_{L_1}) - K_1}{K_e(k_{L_1}) + 4G_1/3 - [\Gamma_e R^2(k_{L_1}, a)^2 - iR^3(k_{L_1}, a)^3(K_e(k_{L_1}) - K_1)]/3}
= \frac{\phi_2\kappa_{21}}{1 - [\Gamma_2(k_{L_1}, a)^2/(3K_2 + 4G_1) - i(k_{L_1}, a)^2\kappa_{21}/3]}, \tag{9.66}
\]

\[
\frac{G_e(k_{L_1}) - G_1}{G_e(k_{L_1}) + \frac{[3K_1/2+4G_1/3]G_1}{K_1+2G_1}} = \phi_2\mu_{21}, \tag{9.67}
\]

where \(\kappa_{21}\) and \(\mu_{21}\) are given in (9.27) and (9.28), respectively, \(\rho_e = \rho_1 + \phi_2(\rho_2 - \rho_1)\), \(R\) represents the radius of a specimen, which is often set to be zero [153], and \(\Gamma_i\) (for \(i = 2, e\)) are given as

\[
\Gamma_i = K_1 - \frac{3}{2}K_i - \frac{2}{3}G_1 + \frac{\rho_i}{2\rho_1} \frac{3K_1 + 4G_1}{3K_i + 4G_i} \left[ K_i + \frac{4}{5}(G_1 + \frac{2}{3}G_i) \right]. \tag{9.68}
\]

### 9.9 Appendix B: Properties of the Attenuation Functions

Here, we present asymptotic behaviors of both attenuation functions \(F(Q)\) and \(F(Q)\), defined by (9.47) and (9.57), respectively, for a statistically isotropic composite. We then briefly discuss the transparency condition (9.64) for stealthy hyperuniform media. Both are functionals of the spectral density \(\tilde{\chi}_v(Q)\) and identical in the quasistatic regime. Specifically, assuming that the spectral density has the power-law
scaling $\chi_v(Q) \sim Q^\alpha$ as $Q \to 0$, the attenuation functions become

$$\text{Re}[F(Q)] = \text{Im}[F(Q)] \sim \begin{cases} Q^d, & \text{nonhyperuniform (}\alpha = 0\text{)} \\ Q^{d+\alpha}, & \text{hyperuniform (}\alpha > 0\text{)} \end{cases}, \quad \text{as } Q \to 0, \quad (9.69)$$

$$\text{Re}[F(Q)] = \text{Re}[F(Q)] \sim Q^2, \quad \text{as } Q \to 0, \quad (9.70)$$

where $\alpha = 0$ for nonhyperuniform systems, and $\alpha > 0$ for hyperuniform systems. In the large-$Q$ regime, both types of attenuation functions exhibit considerably different scalings:

$$\text{Im}[F(Q)] \sim Q^{-1}, \quad \text{Re}[F(Q)] \to \frac{2^{d/2} \Gamma(d/2)}{\pi} \phi_p (1 - \phi_p) \quad (> 0), \quad \text{as } Q \to \infty \quad (9.71)$$

$$\text{Im}[F(Q)] \sim Q, \quad \text{Re}[F(Q)] \to \text{const.} \quad (< 0), \quad \text{as } Q \to \infty, \quad (9.72)$$

regardless of whether the composites are hyperuniform or not. The reader is referred to Ref. [308] (or Chapter 8) for derivations.

A two-phase composite is effectively lossless for elastic waves at a given frequency $\omega$ if and only if $\text{Im}[K_e(k_{Lq})] = 0$ and $\text{Im}[G_e(k_{Lq})] = 0$, which are equivalent to $\text{Im}[F(k_{Lq})] = 0$ when using the nonlocal approximations (9.59) and (9.60). One can show that these conditions are satisfied in the transparency interval (9.64) for stealthy hyperuniform media [cf. (9.2)].

### 9.10 Appendix C: Derivation of Local Strong-Contrast Expansions

Here we present a detailed derivation of the local strong-contrast expansion presented in the main text. Aforementioned, we consider a macroscopically large ellipsoidal two-phase composite specimen in $\mathbb{R}^d$ embedded inside an infinitely large reference phase
of mass density $\rho_I$ and stiffness tensor $C_I$. We assume that the microstructure is perfectly general, and the inhomogeneity length scales $\ell$ are much smaller than the size of specimen $L$, i.e., $\ell \ll L$. The shape of this specimen is purposely chosen to be non-spherical since any rigorously correct expression for the effective stiffness tensor must ultimately be independent of the shape of the composite specimen in the infinite-volume limit. The local stiffness tensor $C(x)$ and mass density $\rho(x)$ of a two-phase composite are written respectively as

$$C(x) \equiv C_1 \mathcal{I}^{(1)}(x) + C_2 \mathcal{I}^{(2)}(x), \quad (9.73)$$

$$\rho(x) \equiv \rho_1 \mathcal{I}^{(1)}(x) + \rho_2 \mathcal{I}^{(2)}(x), \quad (9.74)$$

where for phase $i (= 1, 2)$, $C_i$ is the stiffness tensor, $\rho_i$ is the mass density, and $\mathcal{I}^{(i)}(x)$ is the phase indicator (see Sec. II in the main text). Now suppose the incident plane strain waves $\epsilon_0(x)$ of an angular frequency $\omega$ and wavevector $k_I(\omega)$ in the reference phase, i.e.,

$$\epsilon_0(x) = \tilde{\epsilon}_0 \exp(i(k_I(\omega) \cdot x - \omega t)). \quad (9.75)$$

The associated wavelengths $\lambda = 2\pi/|k_I|$ must lie between the inhomogeneity length scales $\ell$ and the specimen size $L$.\footnote{Rigorously speaking, this qualitative description is not accurate because elastic wave speeds generally vary with their polarizations (e.g., longitudinal or transverse).} Granting that the effective-medium description is valid for the composite, we obtain the corresponding stiffness tensor $C_e(k_I, \omega)$.

In the ensuing derivation, we take the reference phase to be phase $q (= 1, 2)$ and make the following five assumptions, in which three are associated with phase properties, one is related to wavelengths, and the last assumes Hooke’s law:

(a) Phase 1 and phase 2 are elastically isotropic; see Eq. (9.5).

(b) Each phase is dissipationless, namely, the elastic moduli $K_i$ and $G_i$ for $i = 1, 2$ are real-valued and frequency-independent.
(c) The mass densities of both phases are identical, i.e., \( \rho_1 = \rho_2 = \rho_e \).

(d) The long-wavelength (quasistatic) regime is assumed, i.e., \( \lambda \gg \ell \).

(e) We assume infinitesimal deformations, implying the Hooke’s law.

Implications of assumptions (a)-(c) are discussed in Sec. II in the main text. Among them, we only state here that assumptions (a) and (b) imply the linear dispersion relation in the reference phase [i.e., \( k_{Lq} (\omega) = \omega / c_{Lq} \) and \( k_{Tq} (\omega) = \omega / c_{Tq} \) for longitudinal and transverse waves, respectively]. Henceforth we replace the argument \( \omega \) in functions with \( k_{Lq} \) and do not explicitly indicate the \( \omega \) dependence.

We derive some important integral equations for local fields in Sec. 9.10.1. We then derive the local strong-contrast expansions for a macroscopically anisotropic medium in Sec. 9.10.2. In Sec. 9.10.3, we then simplify the expansions derived in Sec. 9.10.2 by assuming the composite is macroscopically isotropic.

### 9.10.1 Integral Equations for the Cavity Strain Field

Here we first derive general expressions for an anisotropic reference phase \( q \) and then simplify the results by assuming (a). Due to assumption (e), we can use the Hooke’s law to relate the local stress \( \boldsymbol{\tau}(x) \) to the local strain \( \boldsymbol{\epsilon}(x) \)

\[
\boldsymbol{\tau}(x) = \mathbf{C}(x) : \boldsymbol{\epsilon}(x),
\]  

(9.76)

where \( \mathbf{C}(x) \) is given in (9.73), and the strain tensor is the symmetric part of the gradient of displacement field \( \mathbf{u}(x) \), i.e.,

\[
\boldsymbol{\epsilon}(x) \equiv \frac{1}{2} \left[ \nabla \mathbf{u} + (\nabla \mathbf{u})^T \right].
\]  

(9.77)
Using separation of variables \( u(x, t) \to u(x) e^{-i\omega t} \), one can write the time-harmonic equation of motion of the volume element at \( x \) as

\[
-\omega^2 \rho(x) u(x) = \nabla \cdot \tau(x). \tag{9.78}
\]

We now introduce the *induced stress polarization field* defined by

\[
P(x) \equiv \left( \frac{C(x)}{\rho(x)} - \frac{C_q}{\rho_q} \right) : \epsilon(x), \tag{9.79}
\]

which is a symmetric, second-rank tensor that is non-zero only in the “polarized” phase \( p(\neq q) \). Using Eqs. (9.78) and (9.79), we obtain an inhomogeneous elastic wave equation in the reference phase whose source term arises from the inhomogeneity of the stiffness and mass density. This wave equation is written in component form as

\[
\omega^2 \ddot{u}_i(x) + \frac{(C_q)_{ijkl}}{\rho_q} \frac{\partial^2 \ddot{u}_l(x)}{\partial x_j \partial x_k} = -\frac{\partial P_{ij}}{\partial x_j} + \frac{1}{\rho(x)} \left[ \frac{1}{\rho(x)} \right] C_{ijkl}(x) \epsilon_{kl}, \tag{9.80}
\]

where \( \ddot{u}(x) \equiv u(x) - u_0(x) \) is the displacement field in excess of the displacement field applied at infinity \( u_0(x) \). Importantly, comparing with the static counterpart [292, 296], Eq. (9.80) has an extra term \( \nabla \left[ \rho(x)^{-1} \right] \cdot \tau(x) \) on the right-hand side which represents the force per area on the interface between phases \( p \) and \( q \) due to the internal stress. In this chapter, we have removed this term by assuming (c) because this simplification makes the dynamic problem formally identical to the static strong-contrast expansions studied in Refs. [292, 296]. Thus, we follow the static strong-contrast expansion formalism closely in the ensuing derivation.

Using the Green function formalism, the excessive displacement field \( \ddot{u}_i(x) \) can be
expressed as

$$\ddot{u}_i(x) = \int dx' g^{(q)}_{ij}(x, x') \left[ \frac{\partial P_{jk}(x')}{\partial x'_k} \right],$$  \hspace{1cm} (9.81)

where $g^{(q)}_{ij}(x, x')$ is the infinite-space Green’s function of elastic wave equation with respect to the reference phase $q$ that satisfies

$$\omega^2 g^{(q)}_{km}(x, x') + \frac{(C_q)_{ijkl}}{\rho_q} \frac{\partial^2 g^{(q)}_{im}(x, x')}{\partial x_j \partial x_k} = -\delta_{kn} \delta(x - x'),$$  \hspace{1cm} (9.82)

$$g^{(q)}_{ij}(x, x') \to 0, \ |x - x'| \to \infty,$$

where we note that $g^{(q)}_{ij}(x, x') = g^{(q)}_{ij}(x - x').$

To obtain an expression for strain tensor, we take the symmetric part of the gradient of Eq (9.81) given as (see Sec. 9.11 for details)

$$\epsilon_{ij}(x) - (\epsilon_0)_{ij}(x) = \int G^{(q)}_{ijkl}(x - x') P_{kl}(x') \, dx',$$

$$= \underbrace{\int_\epsilon -D^{(q)}_{ijkl} P_{kl}(x) \, dx'}_{\text{inside the exclusion-region}} + \underbrace{\int_\epsilon H^{(q)}_{ijkl}(x - x') P_{kl}(x') \, dx'}_{\text{outside the exclusion-region}},$$

(9.84)

where we note that, due to the singular nature of Green’s function around the origin (i.e., $x - x' = 0$), the integral (9.83) should be separated into two parts; one is the integral inside an infinitesimal exclusion region around the origin, and another is the integral outside the exclusion region (denoted by $\int_\epsilon \, dx'$). Thus, the fourth-rank Green’s function can be written concisely as

$$G^{(q)}(x, x') = -D^{(q)} \delta(x - x') + H^{(q)}(x - x'),$$

(9.85)

where the fourth-rank constant tensor $D^{(q)}$ depends on the shape of exclusion region. The fourth-rank tensor field $H^{(q)}_{ijkl}(r)$ is associated with the double gradient of the
dyadic Green function $g_{ij}^{(q)}(r)$ [cf. (9.82)] given as

$$H_{ijkl}^{(q)}(r) = \frac{1}{4} \left[ \frac{\partial^2 g_{ik}^{(q)}}{\partial x_j \partial x_l} + \frac{\partial^2 g_{jk}^{(q)}}{\partial x_i \partial x_l} + \frac{\partial^2 g_{il}^{(q)}}{\partial x_j \partial x_k} + \frac{\partial^2 g_{jl}^{(q)}}{\partial x_i \partial x_k} \right], \quad (9.86)$$

which is symmetric under the following index exchanges, i.e.,

$$H_{ijkl}^{(q)}(r) = H_{jikl}^{(q)}(r) = H_{ijkl}^{(q)}(r) = H_{klij}^{(q)}(r). \quad (9.87)$$

Excluding the contribution from a exclusion-region in Eq. (9.84), we obtain an integral equation for the cavity strain tensor $f(x)$ given as

$$f(x) \equiv \epsilon(x) + D^{(q)} : P(x) = \epsilon_0(x) + \int \epsilon' \, H^{(q)}(x - x') : P(x') \quad (9.88)$$

$$= \{ I + D^{(q)} : [C(x) - C_q] / \rho_q \} : \epsilon(x), \quad (9.89)$$

where the relation (9.90) is obtained from (9.88) and (9.79). The cavity strain field is the elasticity analog of the Lorentz electric field used in the static strong-contrast expansion for the effective dielectric tensor [33, 289].

Eliminating $\epsilon_0$ in Eqs. (9.90) and (9.79) yields the following relation:

$$P(x) = L^{(q)}(x) : f(x), \quad (9.91)$$

where the fourth-order tensor $L^{(q)}(x)$ is a linear fractional transformation of $C(x)$ given by

$$L^{(q)}(x) \equiv [C(x) - C_q] / \rho_q : \left\{ I + D^{(q)} : [C(x) - C_q] / \rho_q \right\}^{-1} \quad (9.92)$$

$$= L^{(q)} \mathcal{I}^{(p)}(x), \quad (9.93)$$
where $L^{(q)} \equiv (C_p - C_q)/\rho_q : \left[ I + D^{(q)} : (C_p - C_q)/\rho_q \right]^{-1}$. Note that $L^{(q)}$ is identical to its static counterpart.

We now assume phase 1 and 2 are isotropic [cf. (a)] to provide explicit formulas for $g^{(q)}(r), H^{(q)}(r), D^{(q)},$ and $L^{(q)}$ which are used in the main text. The reader is referred to Sec. 9.11 for detail of derivations. The dyadic Green function defined in Eq. (9.82) can be simplified as

$$g_{ij}^{(q)}(r) = A_d(r) \hat{r}_i \hat{r}_j + B_d(r) \delta_{ij},$$

where $r \equiv |r|, \hat{r} \equiv r/|r|, \hat{r}_i$ is the $i$th component of $\hat{r}$,

$$A_d(r) = \frac{-i\pi}{2\omega^2(2\pi)^{d/2} r^d} \left[ (k_{Lq} r)^{d/2+1} H_{d/2+1}^{(1)}(k_{Lq} r) - (k_{Tq} r)^{d/2+1} H_{d/2+1}^{(1)}(k_{Tq} r) \right],$$

$$B_d(r) = \frac{i\pi}{2\omega^2(2\pi)^{d/2} r^d} \left[ (k_{Lq} r)^{d/2} H_{d/2}^{(1)}(k_{Lq} r) - (k_{Tq} r)^{d/2} H_{d/2}^{(1)}(k_{Tq} r) + (k_{Tq} r)^{d/2+1} H_{d/2-1}^{(1)}(k_{Tq} r) \right],$$

and $H_{\nu}^{(1)}(x)$ is the Hankel function of the first kind of order $\nu$. Here, $k_{Lq}$ and $k_{Tq}$ are wave speeds of longitudinal and transverse elastic waves in the reference phase $q$, respectively; see definitions in the main text. For $d = 3$, (9.94) is simply expressed as

$$g_{ij}^{(q)}(r) = \frac{1}{4\pi \omega^2 r^3} \left\{ e^{ik_{Lq} r}(1 - i k_{Lq} r) - e^{ik_{Tq} r}[1 - i k_{Tq} r - (k_{Tq} r)^2] \right\} \delta_{ij} + \left\{ e^{ik_{Lq} r}[-3 + i3 k_{Lq} r + (k_{Lq} r)^2] + e^{ik_{Tq} r}[3 - i3 k_{Tq} r - (k_{Tq} r)^2] \right\} \hat{r}_i \hat{r}_j.$$
up to a multiplicative factor $\rho_q$:

\[
\lim_{\omega \to 0^+} H^{(q)}(r) = \frac{\Gamma(d/2)}{4\pi^{d/2}} \frac{1}{d c_{L_q}^2} \left[ \alpha_q d \Lambda_h - 2dI - 2d\alpha_q T_1(r) + 2d(d - \alpha_q) T_2(r) \\
+ d(d + 2)\alpha_q T_3(r) \right],
\]

(9.98)

where $\alpha_q \equiv dK_q/G_q + (d - 2) = d(c_{L_q}^2 - c_{T_q}^2)/c_{T_q}^2$. The Fourier transforms of the Green function $G^{(q)}_{ijkl}(r)$ and $H^{(q)}_{ijkl}(r)$ are simple:

\[
\tilde{G}^{(q)}_{ijkl}(q) = -\frac{q^4}{\omega^2} \left( \frac{1}{q^2 - k_{L_q}^2} - \frac{1}{q^2 - k_{T_q}^2} \right) \hat{q}_i \hat{q}_j \hat{q}_k \hat{q}_l \\
- \frac{1}{c_{T_q}^2} \frac{1}{q^2 - k_{T_q}^2} \frac{1}{4} \{ \hat{q}_j \delta_{jk} \hat{q}_l + \hat{q}_j \delta_{jl} \hat{q}_k + \hat{q}_j \delta_{ik} \hat{q}_l + \hat{q}_i \delta_{il} \hat{q}_k \},
\]

(9.99)

and $\tilde{H}^{(q)}_{ijkl}(q)$ is given in Eq. (9.102).

The constant tensors $D^{(q)}$ and $L^{(q)}$ depend on the exclusion-region shape. For a spherical exclusion-region in $\mathbb{R}^d$, $D^{(q)}$ defined in Eq. (9.84) writes explicitly as

\[
D^{(q)} = \frac{\rho_e \Lambda_h}{dK_q + 2(d - 1)G_q} + \frac{\rho_e d(K_q + 2G_q) \Lambda_s}{G_q(d + 2)[dK_q + 2(d - 1)G_q]} \\
= \frac{1}{dc_{L_q}^2} \Lambda_h + \frac{1}{d + 2} \left( \frac{2}{dc_{L_q}^2} + \frac{1}{c_{T_q}^2} \right) \Lambda_s,
\]

(9.100)

where $c_{L_q}$ and $c_{T_q}$ are the longitudinal and transverse wave speeds; see the definitions in the main text. Thus, the corresponding constant tensor $L^{(q)}$ [(9.93)] is explicitly written as

\[
L^{(q)} = dc_{L_q}^2 \left[ \kappa_{pq} \Lambda_h + \frac{(d + 2)c_{T_q}^2}{dc_{L_q}^2 + 2c_{T_q}^2(\mu_{pq} \Lambda_s)} \right],
\]

(9.101)

where $\kappa_{pq}$ is the polarizability for bulk modulus and $\mu_{pq}$ is the polarizability for shear modulus, which are defined in Eqs. (9.27) and (9.28), respectively.

**Remarks:**
1. The reference phase $q$ employed in the Green function $G^{(q)}(r)$ can be different from phase 1 or 2. In this chapter, however, we focus on the cases where $q$ equals 1 or 2 for simplicity.

2. In rigorous sense, equation (9.85) should be written as

$$G^{(q)}(x, x') = \begin{cases} 
-D^{(q)}(x) \delta(x - x'), & \text{outside the exclusion region}, \\
 H^{(q)}(x) - H^{(q)}(x'), & \text{inside the exclusion region}.
\end{cases}$$

Due to this definition, while $H^{(q)}(r)$ in the direct-space is independent of the exclusion-region shape, the Fourier transform $\tilde{H}^{(q)}(q)$ does depend on the exclusion-region shape in the following manner

$$\tilde{H}^{(q)}(q) = \int d^3 r e^{-iqr} H^{(q)}(r) = \int d^3 r e^{-iqr} H^{(q)}(r)$$

$$= \tilde{G}^{(q)}(q) + D^{(q)}. \quad (9.102)$$

9.10.2 Local Strong-Contrast Expansions in General Cases

Here we derive the local strong-contrast expansions for the effective stiffness tensor of a macroscopically anisotropic medium that are strictly valid in the long-wavelength (quasistatic) regime. To do so, we derive an exact series expansion for the effective tensor $L_e^{(q)}$ in a local homogenized relation

$$\langle f \rangle (x) = L_e^{(q)} : \langle P \rangle (x) \quad (9.103)$$

with an incident strain field (9.75). When the effective-medium description is valid, the local homogenized relation (9.103) can be obtained from (9.91) by using the following mapping

$$C_p \rightarrow C_e(L_q), \quad T^{(p)}(x) \rightarrow 1,$$
and thus

\[ L_e^{(q)}(k_{Lq}) = \left[ C_e(k_{Lq}) - C_q \right] / \rho_e : \left[ I + D^{(q)} : (C_e(k_{Lq}) - C_q) / \rho_e \right]^{-1}. \]  

(9.104)

For this purpose, we first find explicit expressions for \( \langle P \rangle (x) \) and \( \langle f \rangle (x) \) in terms of the applied field \( \epsilon_0 \) from the integral equation (9.89). We then find an explicit expression for the effective constant tensor \( L_e^{(q)} \) by eliminating \( \epsilon_0 \) between these two expressions. Keeping in mind that the tensors \( L^{(q)}, L_e^{(q)} \), and \( H^{(q)} \) are associated with the reference phase \( q \), we shall temporarily drop the superscript \( q \) when referring these tensors in the ensuing derivation.

We rewrite (9.89) in a compact linear operator form:

\[ f = \epsilon_0 + HP. \]  

(9.105)

Combination of this equation with (9.91) yields

\[ P = \mathcal{L} \epsilon_0 + \mathcal{L} HP. \]  

(9.106)

The desired relation between \( P \) and \( \epsilon_0 \) can be obtained by iteratively substituting (9.106)

\[ P = (I + \mathcal{L} H + \mathcal{L} H \mathcal{L} H + \cdots) \mathcal{L} \epsilon_0 = [I - \mathcal{L} H]^{-1} \mathcal{L} \epsilon_0 = T \epsilon_0, \]  

(9.107)

which writes out more explicitly as

\[ P(1) = \mathcal{L}(1) : \epsilon_0(1) + \int_{\epsilon} d1' \mathcal{L}(1) : H(1, 1') : \mathcal{L}(1') : \epsilon_0(1') \]
\[ + \int_{\epsilon} d2 d1' \mathcal{L}(1) : H(1, 2) : \mathcal{L}(2) : H(2, 1') : \mathcal{L}(1') : \epsilon_0(1') + \cdots \]
\[ = \int_{\epsilon} d1' T(1, 1') : \epsilon_0(1'), \]  

(9.108)
where the boldface numbers $1, 2, \cdots$ are short-hand notations for position vectors $\mathbf{r}_1, \mathbf{r}_2, \cdots$. For a statistically homogeneous medium, an ensemble average of the two-point operator $\mathbf{T}(\mathbf{1}, \mathbf{1}')$ becomes dependent on relative positions, i.e., $\langle \mathbf{T} \rangle (\mathbf{1}, \mathbf{1}') = \langle \mathbf{T} \rangle (\mathbf{1} - \mathbf{1}')$, and an ensemble average of Eq. (9.108) can be written as a convolution:

$$
\langle \mathbf{P} \rangle (\mathbf{1}) = \int_\epsilon d\mathbf{1}' \langle \mathbf{T} \rangle (\mathbf{1} - \mathbf{1}') : \epsilon_0(\mathbf{1}') .
$$

(9.109)

The two-point operator $\langle \mathbf{T} \rangle (\mathbf{1} - \mathbf{1}')$ is explicitly written as

$$
\langle \mathbf{T} \rangle (\mathbf{1} - \mathbf{1}') = \left[ I \left\langle \mathcal{T}^{(p)}(\mathbf{1}) \right\rangle + \mathbf{L} : \mathbf{H}(\mathbf{1} - \mathbf{1}') \left\langle \mathcal{T}^{(p)}(\mathbf{1}) \mathcal{T}^{(p)}(\mathbf{1}') \right\rangle 
+ \int_\epsilon d\mathbf{2} \mathbf{L} : \mathbf{H}(\mathbf{2} - \mathbf{1}') \mathbf{L} : \mathbf{H}(\mathbf{1} - \mathbf{1}') \left\langle \mathcal{T}^{(p)}(\mathbf{1}) \mathcal{T}^{(p)}(\mathbf{2}) \mathcal{T}^{(p)}(\mathbf{1}') \right\rangle + \cdots \right] : \mathbf{L}
= \left[ I S_1^{(p)}(\mathbf{1}) + \mathbf{U}(\mathbf{1} - \mathbf{1}') S_2^{(p)}(\mathbf{1}, \mathbf{1}') + \int_\epsilon d\mathbf{2} \mathbf{U}(\mathbf{1} - \mathbf{2}) : \mathbf{U}(\mathbf{2} - \mathbf{1}') S_3^{(p)}(\mathbf{1}, \mathbf{2}, \mathbf{1}') 
+ \cdots \right] : \mathbf{L},
$$

(9.110)

where $\mathbf{U}(\mathbf{r}) \equiv \mathbf{L} : \mathbf{H}(\mathbf{r})$, and $S_n^{(p)}(\mathbf{1}, \cdots, \mathbf{1}')$ is the $n$-point correlation function of phase $p \neq q$ (see Sec. II in the main text). Importantly, the relation (9.109) is nonlocal in space [i.e., $\langle \mathbf{P} \rangle (\mathbf{x})$ at $\mathbf{x}$ depends on $\epsilon_0(\mathbf{x}')$ at different positions around $\mathbf{x}$] in general. In the quasistatic regime, however, the applied strain tensor will barely change over the correlation length scales over which $\langle \mathbf{T} \rangle (\mathbf{1} - \mathbf{1}')$ vanishes, allowing us to approximate (9.109) as follows:

$$
\langle \mathbf{P} \rangle (\mathbf{1}) \approx \left[ \int_\epsilon d\mathbf{1}' \langle \mathbf{T} \rangle (\mathbf{1} - \mathbf{1}') \right] : \epsilon_0(\mathbf{1}) .
$$

(9.111)

We obtain a local homogenized constitutive relation between $\langle \mathbf{f} \rangle (\mathbf{x})$ and $\langle \mathbf{P} \rangle (\mathbf{x})$ by eliminating the term $\epsilon_0(\mathbf{x})$ in (9.105) by substituting (9.111):

$$
\langle \mathbf{f} \rangle = \epsilon_0 + \mathbf{H} \langle \mathbf{P} \rangle = \left( (\langle \mathbf{T} \rangle)^{-1} + \mathbf{H} \right) \langle \mathbf{P} \rangle .
$$

(9.112)
Comparing (9.112) with (9.103) gives

\[
\left[ L^{(q)}(k_{L_q}) \right]^{-1} = \langle T \rangle^{-1} + H, \tag{9.113}
\]

which can be written explicitly as

\[
L : \left[ L^{(q)}(k_{L_q}) \right]^{-1} = L : \left\{ \int_\epsilon d1' \langle T \rangle (1 - 1') \right\}^{-1} + \int_\epsilon d1' H(1 - 1')
\]

\[
= \frac{I}{S_1^{(p)}(1)} - \int_\epsilon d1' U(1, 1') \left[ \frac{S_2^{(p)}(1, 1') - S_1^{(p)}(1) S_1^{(p)}(1')}{S_1^{(p)}(1) S_1^{(p)}(1') - S_2^{(p)}(1, 2) S_2^{(p)}(2, 1') / S_1^{(p)}(2)} \right]
\]

\[
= \int_\epsilon d2 d1' U(1, 2) : U(2, 1') \left[ \frac{S_3^{(p)}(1, 2, 1') - S_2^{(p)}(1, 2) S_2^{(p)}(2, 1') / S_1^{(p)}(2)}{S_1^{(p)}(1) S_1^{(p)}(1') - S_2^{(p)}(1, 2) S_2^{(p)}(2, 1') / S_1^{(p)}(2)} \right]
\]

\[
\times \left[ S_1^{(p)}(2) S_3^{(p)}(3) S_2^{(p)}(1, 2, 3, 1') - S_1^{(p)}(2) S_2^{(p)}(1, 2) S_3^{(p)}(2, 3, 1') - S_1^{(p)}(3) S_3^{(p)}(1, 2, 3) S_2^{(p)}(3, 1') + S_2^{(p)}(1, 2) S_2^{(p)}(2, 3) S_2^{(p)}(3, 1') \right] + \cdots. \tag{9.114}
\]

The resulting series expansion is

\[
\phi_p^2 L^{(q)} : \left[ L^{(q)}(k_{L_q}) \right]^{-1} = \phi_p I - \sum_{n=2}^{\infty} B_n^{(p)}(k_{L_q}),
\]

which is the series given in Eq. (9.31).

### 9.10.3 Local Strong-Contrast Expansions for Macroscopically Isotropic Media

Here we assume that the composite is macroscopically isotropic, i.e.,

\[
C_e(k_{L_q}) \equiv d K_e(k_{L_q}) \Lambda_h + 2 G_e(k_{L_q}) \Lambda_s,
\]

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and thus the series expansion (9.31) can be rewritten as

$$\phi_p^2 \left[ \frac{\kappa_{pq}}{\kappa_{eq}(k_{Lq})} \Lambda_h + \frac{\mu_{pq}}{\mu_{eq}(k_{Lq})} \Lambda_s \right] = \phi_p I - \sum_{n=2}^{\infty} B_n^{(p)}(k_{Lq}).$$ \hspace{1cm} (9.115)

Using properties of the projection tensors $\Lambda_h$ and $\Lambda_s$ (Sec. 9.12.2) yields the strong-contrast expansions for the effective bulk and shear moduli given respectively as

$$\kappa_{eq}(k_{Lq}) = \frac{K_e(k_{Lq}) - K_q}{K_e(k_{Lq}) + 2(d-1)G_q/d} = \frac{\phi_p^2 \kappa_{pq}}{\phi_p - \sum_{n=2}^{\infty} C_n^{(p)}(k_{Lq})},$$ \hspace{1cm} (9.116)

$$\mu_{eq}(k_{Lq}) = \frac{G_e(k_{Lq}) - G_q}{G_e(k_{Lq}) + [dK_q/2 + (d+1)(d-2)G_q/d]G_q/(K_q + 2G_q)} = \frac{\phi_p^2 \mu_{pq}}{\phi_p - \sum_{n=2}^{\infty} D_n^{(p)}(k_{Lq})},$$ \hspace{1cm} (9.117)

where $C_n^{(p)}(k_{Lq}) \equiv B_n^{(p)}(k_{Lq}) : \Lambda_h$ and $D_n^{(p)}(k_{Lq}) \equiv 2[(d+2)(d-1)]^{-1} B_n^{(p)}(k_{Lq}) : \Lambda_s$.

In what follows, we derive explicit formulas of $C_n^{(p)}(k_{Lq})$ and $D_n^{(p)}(k_{Lq})$ for $n = 2, 3$.

**Approximation at the two-point level**

Here we derive the explicit formulas for $C_2^{(p)}(k_{Lq})$ and $D_2^{(p)}(k_{Lq})$ that are important for the local strong-contrast approximation at the two-point level. To do so, we remind the formulas for $C_2^{(p)}(k_{Lq})$, $D_2^{(p)}(k_{Lq})$, and $U^{(q)}(r)$. From (9.32), the two-point parameters are written respectively as

$$C_2^{(p)}(k_{Lq}) \equiv B_2^{(p)}(k_{Lq}) : \Lambda_h = \frac{1}{d} \int dr \, U^{(q)}_{ijkh}(r) \chi_v(r),$$ \hspace{1cm} (9.118)

$$D_2^{(p)}(k_{Lq}) \equiv \frac{2}{(d+2)(d-1)} B_2^{(p)}(k_{Lq}) : \Lambda_s = \frac{2}{(d+2)(d-1)} \int dr \left[ U^{(q)}_{ijkh}(r) - \frac{1}{d} U^{(q)}_{ikkh}(r) \right] \chi_v(r),$$ \hspace{1cm} (9.119)
where

\[
U_{ijkl}^{(q)}(r) \equiv L_{ijmn}^{(q)} H_{mnkl}^{(q)}(r) \\
= d c_{Lq}^2 \left[ \left( \kappa_{pq} - \frac{(d + 2) c_{Tq}^2}{dc_{Lq}^2 + 2 c_{Tq}^2} \mu_{pq} \right) \delta_{ij} H_{mnkl}^{(q)}(r) + \frac{(d + 2) c_{Tq}^2}{dc_{Lq}^2 + 2 c_{Tq}^2} \mu_{pq} H_{mnkl}^{(q)}(r) \right],
\]

(9.120)

and \( H^{(q)}(r) \) is given in Eq. (9.18).

To compute (9.118) and (9.119), we first obtain \( U_{iikk}^{(q)}(r) \) and \( U_{ikik}^{(q)}(r) \) from (9.120)

\[
U_{iikk}^{(q)}(r) = d c_{Lq}^2 \left[ \left( \kappa_{pq} - \frac{(d + 2) c_{Tq}^2}{dc_{Lq}^2 + 2 c_{Tq}^2} \mu_{pq} \right) H_{mnkk}^{(q)}(r) + \frac{(d + 2) c_{Tq}^2}{dc_{Lq}^2 + 2 c_{Tq}^2} \mu_{pq} H_{iikk}^{(q)}(r) \right] \\
= d c_{Lq}^2 \kappa_{pq} H_{iikk}^{(q)}(r) = - \frac{i d \pi}{2(2\pi)^{d/2} \omega^{d+2}} \kappa_{pq} r_{L}^{d/2+3} H_{d/2-1}^{(1)}(r_{L}) ,
\]

(9.121)

\[
U_{ikik}^{(q)}(r) = d c_{Lq}^2 \left[ \left( \kappa_{pq} - \frac{(d + 2) c_{Tq}^2}{dc_{Lq}^2 + 2 c_{Tq}^2} \mu_{pq} \right) \frac{1}{d} H_{mnkk}^{(q)}(r) + \frac{(d + 2) c_{Tq}^2}{dc_{Lq}^2 + 2 c_{Tq}^2} \mu_{pq} H_{iikk}^{(q)}(r) \right] \\
= \frac{1}{d} U_{iikk}^{(q)}(r) + d c_{Lq}^2 \frac{(d + 2) c_{Tq}^2}{dc_{Lq}^2 + 2 c_{Tq}^2} \mu_{pq} \left[ H_{ikik}^{(q)}(r) - \frac{1}{d} H_{iikk}^{(q)}(r) \right] \\
= \frac{1}{d} U_{iikk}^{(q)}(r) - \frac{i d \pi}{2(2\pi)^{d/2} \omega^{d+2}} \frac{(d - 1)(d + 2)}{2} c_{Lq}^2 c_{Tq}^2 \mu_{pq} \\
\times \left[ \frac{1}{d} r_{L}^{d/2+3} H_{d/2-1}^{(1)}(r_{L}) + \frac{1}{2} r_{T}^{d/2+3} H_{d/2-1}^{(1)}(r_{T}) \right] ,
\]

(9.122)

where we have used the properties of \( H^{(q)}(r) \) (cf. Sec. 9.12.3), \( r_{L} \equiv k_{Lq} r \), and
Using (9.121) and (9.122), we simplify Eqs. (9.118) and (9.119) as follow:

\[
C_2^{(p)}(k_Lq) = \frac{\pi}{2^{d/2} \Gamma(d/2)} k_{pq} \kappa_{pq} \left[ -i \int_0^\infty \, dr \, (k_{Lq} r)^{d/2} k_{Lq} \mathcal{H}^{(1)}_{d/2-1}(k_{Lq} r) \chi_V(r) \right] = \frac{\pi}{2^{d/2} \Gamma(d/2)} k_{pq} \mathcal{F}(k_{Lq}) \tag{9.123}
\]

\[
D_2^{(p)}(k_Lq) = \frac{\pi}{2^{d/2} \Gamma(d/2)} \frac{c_{Lq}^2}{d^2 c_{Lq}^2 + 2 c_{Tq}^2} \mu_{pq} \left[ - \frac{c_{Tq}}{d^2} i \int_0^\infty \, dr \, (k_{Tq} r)^{d/2} k_{Tq} \mathcal{H}^{(1)}_{d/2-1}(k_{Tq} r) \right]
\]

\[
= \frac{\pi}{2^{d/2} \Gamma(d/2)} \left[ \frac{2 c_{Tq}^2 \mathcal{F}(k_{Lq})}{d^2 c_{Lq}^2 + 2 c_{Tq}^2} \right] \mu_{pq}, \tag{9.124}
\]

where \( \Gamma(x) \) is the Gamma function, and the local attenuation function \( \mathcal{F}(Q) \) is defined in Eqs. (9.47). The real and imaginary parts of \( \mathcal{F}(Q) \) are respectively given as

\[
\text{Im}[\mathcal{F}(Q)] = - \lim_{\epsilon \to 0^+} \int_\epsilon^\infty \, dr \, Q \chi_V(r) (Q r)^{d/2} J_{d/2-1}(Qr) = - \frac{Q^d}{(2\pi)^{d/2}} \tilde{\chi}_V(Q), \tag{9.125}
\]

\[
\text{Re}[\mathcal{F}(Q)] = \lim_{\epsilon \to 0^+} \int_\epsilon^\infty \, dr \, Q \chi_V(r) (Q r)^{d/2} Y_{d/2-1}(Qr) = - \frac{2Q^2}{\pi} \text{p.v.} \int_0^\infty dq \frac{1}{q(Q^2 - q^2)} \text{Im}[\mathcal{F}(q)], \tag{9.126}
\]

where \( J_\nu(x) \) \([Y_\nu(x)]\) is the Bessel function of the first kind \[the second kind\] of order \( \nu \), and p.v. stands for the Cauchy principal value of an integral.

**Approximation at the three-point level**

Here we derive the explicit formulas for \( C_3^{(p)}(k_{Lq}) \) and \( D_3^{(p)}(k_{Lq}) \) that are important for the local strong-contrast approximation at the three-point level. These three-point
parameters are defined respectively as

\[ C_3^{(p)}(k_{L_q}) \equiv B_3^{(p)}(k_{L_q}) \cdot \Lambda_h \]

\[ = \frac{-1}{\phi_p} \int d^2 d3 \left[ U^{(q)}(1 - 2) : U^{(q)}(2 - 3) \right] \cdot \Lambda_h \Delta_3^{(p)}(1, 2, 3), \quad (9.129) \]

\[ D_3^{(p)}(k_{L_q}) \equiv \frac{2}{(d + 2)(d - 1)} B_3^{(p)}(k_{L_q}) \cdot \Lambda_s \]

\[ = \frac{-1}{(d + 2)(d - 1)} \frac{1}{\phi_p} \int d^2 d3 \left[ U^{(q)}(1 - 2) : U^{(q)}(2 - 3) \right] \cdot \Lambda_s \Delta_3^{(p)}(1, 2, 3), \quad (9.130) \]

respectively. To do so, we first obtain expressions for \( U^{(q)}(r) : U^{(q)}(s) \) from (9.120):

\[ U_{ijmn}^{(q)}(r) U_{mnkl}^{(q)}(s) = \left( \frac{dc_{L_q}}{2} \right)^2 \left\{ \left( \kappa_{pq} - \frac{(d + 2)c_{T_q}^2}{2c_{L_q}^2 + 2c_{T_q}^2 + 2\mu_pq} \right)^2 \frac{1}{d^2} \delta_{ij} H_{m'm'nn'(s)}^{(q)} \right\} \]

\[ + \frac{(d + 2)c_{T_q}^2}{d^2} \frac{2}{2c_{L_q}^2 + 2c_{T_q}^2 + 2\mu_pq} \left( \kappa_{pq} - \frac{(d + 2)c_{T_q}^2}{d^2} \frac{2}{2c_{L_q}^2 + 2c_{T_q}^2 + 2\mu_pq} \right) \left[ d H_{m'm'nn'(s)}^{(q)} H_{m'm'nn'(s)}^{(q)} + H_{m'm'nn'(s)}^{(q)} H_{m'm'nn'(s)}^{(q)} \right] \]

\[ \left. + \left( \frac{(d + 2)c_{T_q}^2}{2c_{L_q}^2 + 2c_{T_q}^2 + 2\mu_pq} \right)^2 H_{ijmn}^{(q)}(r) H_{mnkl}^{(q)}(s) \right\}. \quad (9.131) \]

Using (9.131), we obtain

\[ \left[ U^{(q)}(r) : U^{(q)}(s) \right] \cdot \Lambda_h = (\Lambda_h)_{ijkl} U_{ijmn}^{(q)}(r) U_{mnkl}^{(q)}(s) \]

\[ = \frac{(dc_{L_q}^2)^2}{d} \left\{ \left( \kappa_{pq} - \frac{(d + 2)c_{T_q}^2}{2c_{L_q}^2 + 2c_{T_q}^2 + 2\mu_pq} \right)^2 \frac{1}{d} H_{m'm'nn'(s)}^{(q)} \right\} \]

\[ + \frac{(d + 2)c_{T_q}^2}{d^2} \frac{2}{2c_{L_q}^2 + 2c_{T_q}^2 + 2\mu_pq} \left[ \kappa_{pq} - \frac{(d + 2)c_{T_q}^2}{d^2} \frac{2}{2c_{L_q}^2 + 2c_{T_q}^2 + 2\mu_pq} \right] \left[ d H_{m'm'nn'(s)}^{(q)} H_{m'm'nn'(s)}^{(q)} \right] \]

\[ + \left. H_{m'm'nn'(s)}^{(q)} H_{m'm'nn'(s)}^{(q)} \right] + \left( \frac{(d + 2)c_{T_q}^2}{2c_{L_q}^2 + 2c_{T_q}^2 + 2\mu_pq} \right)^2 H_{ijmn}^{(q)}(r) H_{mnkl}^{(q)}(s) \}

\[ = \frac{(dc_{L_q}^2)^2}{d} \left\{ \left( \kappa_{pq} - \frac{(d + 2)c_{T_q}^2}{2c_{L_q}^2 + 2c_{T_q}^2 + 2\mu_pq} \right) \kappa_{pq} \frac{1}{d} H_{ijmn}^{(q)}(r) H_{mnkl}^{(q)}(s) \right\} \]

\[ + \frac{(d + 2)c_{T_q}^2}{d^2} \frac{2}{2c_{L_q}^2 + 2c_{T_q}^2 + 2\mu_pq} H_{ijmn}^{(q)}(r) H_{mnkl}^{(q)}(s) \]. \quad (9.132)\]
Substituting the expressions for $\mathbf{H}^{(q)}(\mathbf{r})$ in Sec. 9.12.3 into (9.132) gives

$$
\left[ U^{(q)}(\mathbf{r}) : U^{(q)}(\mathbf{s}) \right] \Delta_n = U^{(q)}_{i_{\mu\nu\kappa\lambda}}(\mathbf{r}) U^{(q)}_{m_{\mu'n'n'\kappa\lambda}}(\mathbf{s}) - \frac{1}{\lambda} U^{(q)}_{i_{\mu\nu\kappa\lambda}}(\mathbf{r}) U^{(q)}_{m_{\mu'n'n'\kappa\lambda}}(\mathbf{s})
$$

and

$$
\left[ U^{(q)}(\mathbf{r}) : U^{(q)}(\mathbf{s}) \right] \Delta_n = \left[ \kappa_{pq} - \frac{(d + 2)c^2_{T_q}}{d c_{L_q}^2 + 2 c^2_{T_q}} \mu_{pq} \right] \frac{1}{d} (k_{L_q} r) H_{d/2-1}^{(1)}(k_{L_q} r) (k_{L_q} s) H_{d/2-1}^{(1)}(k_{L_q} s)
$$

where we have used the recurrence relation of $\mathcal{H}_d^{(1)}(x)$; see (9.159). Use of (9.129) and (9.134) yields the expression of $C_3^{(p)}(k_{L_q})$ given in Eq. (9.51).

Using (9.131), we now compute

$$
\left[ U^{(q)}(\mathbf{r}) : U^{(q)}(\mathbf{s}) \right] \Delta_n = U^{(q)}_{i_{\mu\nu\kappa\lambda}}(\mathbf{r}) U^{(q)}_{m_{\mu'n'n'\kappa\lambda}}(\mathbf{s}) - \frac{1}{\lambda} U^{(q)}_{i_{\mu\nu\kappa\lambda}}(\mathbf{r}) U^{(q)}_{m_{\mu'n'n'\kappa\lambda}}(\mathbf{s})
$$

and

$$
\left[ U^{(q)}(\mathbf{r}) : U^{(q)}(\mathbf{s}) \right] \Delta_n = \left[ \kappa_{pq} - \frac{(d + 2)c^2_{T_q}}{d c_{L_q}^2 + 2 c^2_{T_q}} \mu_{pq} \right] \frac{1}{d} (k_{L_q} r) H_{d/2-1}^{(1)}(k_{L_q} r) (k_{L_q} s) H_{d/2-1}^{(1)}(k_{L_q} s)
$$

where we have used the recurrence relation of $\mathcal{H}_d^{(1)}(x)$; see (9.159). Use of (9.129) and (9.134) yields the expression of $C_3^{(p)}(k_{L_q})$ given in Eq. (9.51).

Using (9.131), we now compute

$$
\left[ U^{(q)}(\mathbf{r}) : U^{(q)}(\mathbf{s}) \right] \Delta_n = U^{(q)}_{i_{\mu\nu\kappa\lambda}}(\mathbf{r}) U^{(q)}_{m_{\mu'n'n'\kappa\lambda}}(\mathbf{s}) - \frac{1}{\lambda} U^{(q)}_{i_{\mu\nu\kappa\lambda}}(\mathbf{r}) U^{(q)}_{m_{\mu'n'n'\kappa\lambda}}(\mathbf{s})
$$

and

$$
\left[ U^{(q)}(\mathbf{r}) : U^{(q)}(\mathbf{s}) \right] \Delta_n = \left[ \kappa_{pq} - \frac{(d + 2)c^2_{T_q}}{d c_{L_q}^2 + 2 c^2_{T_q}} \mu_{pq} \right] \frac{1}{d} (k_{L_q} r) H_{d/2-1}^{(1)}(k_{L_q} r) (k_{L_q} s) H_{d/2-1}^{(1)}(k_{L_q} s)
$$

where we have used the recurrence relation of $\mathcal{H}_d^{(1)}(x)$; see (9.159). Use of (9.129) and (9.134) yields the expression of $C_3^{(p)}(k_{L_q})$ given in Eq. (9.51).
Here, the two terms $H_{m'mnm'}^{(q)}(r) H_{m'm'n'n'}^{(q)}(s)$ and $H_{m'mmn}^{(q)}(r) H_{nmn}^{(q)}(s)$ are easy to compute by using formulas in Sec. 9.12.3. After some tedious calculations, the last term $H_{ikmn}^{(q)}(r) H_{mnik}^{(q)}(s)$ can be rewritten by use of the formulas in Table 9.2 as follows:

$$
H_{ikmn}^{(q)}(r) H_{mnik}^{(q)}(s) = \frac{-\pi^2}{4(2\pi)^d r^{d+2}s^{d+2} \omega^d} \left[ M_0(r, s) + (\hat{r} \cdot \hat{s})^2 M_2(r, s) + (\hat{r} \cdot \hat{s})^4 M_4(r, s) \right],
$$

(9.136)

where

$$
M_0(r, s) \equiv \left[ r_L^{d/2+3} \mathcal{H}_{d/2+3}^{(1)}(r_L) - r_T^{d/2+3} \mathcal{H}_{d/2+3}^{(1)}(r_T) \right]
\times \left[ 3s_L^{d/2+1} \mathcal{H}_{d/2+1}^{(1)}(s_L) - 3s_T^{d/2+1} \mathcal{H}_{d/2+1}^{(1)}(s_T) + s_T^{d/2+2} \mathcal{H}_{d/2}^{(1)}(s_T) \right]
+ \left[ r_L^{d/2+2} \mathcal{H}_{d/2+2}^{(1)}(r_L) - r_T^{d/2+2} \mathcal{H}_{d/2+2}^{(1)}(r_T) \right]
\times \left[ -6s_L^{d/2+2} \mathcal{H}_{d/2}^{(1)}(s_L) - 2(d - 1)s_T^{d/2+2} \mathcal{H}_{d/2}^{(1)}(s_T) + s_T^{d/2+3} \mathcal{H}_{d/2+1}^{(1)}(s_T) \right]
+ r_T^{d/2+3} \mathcal{H}_{d/2+1}^{(1)}(r_T) \left[ -s_L^{d/2+2} \mathcal{H}_{d/2}^{(1)}(s_L) - \frac{d - 1}{2} s_T^{d/2+2} \mathcal{H}_{d/2}^{(1)}(s_T) + \frac{1}{4} s_T^{d/2+3} \mathcal{H}_{d/2+1}^{(1)}(s_T) \right]
+ \left[ r_L^{d/2+1} \mathcal{H}_{d/2+1}^{(1)}(r_L) - r_T^{d/2+1} \mathcal{H}_{d/2+1}^{(1)}(r_T) \right] s_L^{d/2+3} \mathcal{H}_{d/2-1}^{(1)}(s_L)
+ \left[ 2r_L^{d/2+1} \mathcal{H}_{d/2+1}^{(1)}(r_L) - 2 r_T^{d/2+1} \mathcal{H}_{d/2+1}^{(1)}(r_T) + r_T^{d/2+2} \mathcal{H}_{d/2}^{(1)}(r_T) \right]
\times \left[ s_L^{d/2+3} \mathcal{H}_{d/2-1}^{(1)}(s_L) + \frac{d - 1}{2} s_T^{d/2+3} \mathcal{H}_{d/2-1}^{(1)}(s_T) \right],
$$

(9.137)

$$
M_2(r, s) \equiv - \left[ r_L^{d/2+3} \mathcal{H}_{d/2+3}^{(1)}(r_L) - r_T^{d/2+3} \mathcal{H}_{d/2+3}^{(1)}(r_T) \right] \left[ s_L^{d/2+2} \mathcal{H}_{d/2+2}^{(1)}(s_L) - s_T^{d/2+2} \mathcal{H}_{d/2+2}^{(1)}(s_T) \right]
+ 6 \left[ r_L^{d/2+2} \mathcal{H}_{d/2+2}^{(1)}(r_L) - r_T^{d/2+2} \mathcal{H}_{d/2+2}^{(1)}(r_T) \right] \left[ s_L^{d/2+3} \mathcal{H}_{d/2+1}^{(1)}(s_L) - s_T^{d/2+3} \mathcal{H}_{d/2+1}^{(1)}(s_T) \right]
+ r_T^{d/2+3} \mathcal{H}_{d/2+1}^{(1)}(r_T) s_L^{d/2+3} \mathcal{H}_{d/2+1}^{(1)}(s_L)
+ \left[ r_L^{d/2+3} \mathcal{H}_{d/2+1}^{(1)}(r_L) - \frac{d - 6}{4} r_T^{d/2+3} \mathcal{H}_{d/2+1}^{(1)}(r_T) \right] s_T^{d/2+3} \mathcal{H}_{d/2+1}^{(1)}(s_T),
$$

(9.138)
\[ M_4(r, s) \equiv \left[ r_L^{d/2+3} \mathcal{H}_{d/2+3}^{(1)}(r_L) - r_T^{d/2+3} \mathcal{H}_{d/2+3}^{(1)}(r_T) \right] \left[ s_L^{d/2+3} \mathcal{H}_{d/2+3}^{(1)}(s_L) - s_T^{d/2+3} \mathcal{H}_{d/2+3}^{(1)}(s_T) \right], \]

(9.139)

where \( r_L \equiv k_{Lq} r \), \( r_T \equiv k_{Tq} r \), \( s_T \equiv k_{Lq} s \), and \( s_T \equiv k_{Tq} s \). We substitute the newly obtained expression for Eq. (9.135) into (9.130) and then rearrange it in terms of \( \hat{P}_4(\hat{r} \cdot \hat{s}) \) and \( \hat{P}_2(\hat{r} \cdot \hat{s}) \), which are defined in Eqs. (9.53) and (9.54), respectively. After painstaking algebraic calculations, we obtain the expression of \( D_3^{(p)}(k_{Lq}) \) given in Eq. (9.52).

9.11 Appendix D: Derivation of Green Functions for an Isotropic Medium

In this section, we derive the explicit formulas for the Green functions employed in this chapter.

9.11.1 Dyadic Green Function \( g^{(q)}(r) \)

Here we derive the explicit expression for the dyadic Green function \( g^{(q)}_{ij}(x, x') = g^{(q)}_{ij}(x - x') \) associated with the elastic wave equation considered in Sec. II in the main text. The Green function satisfies the following equation:

\[ \omega^2 g^{(q)}_{ij}(r) + \left( c_{Lq}^2 - c_{Tq}^2 \right) \frac{\partial^2 g^{(q)}_{kj}(\mathbf{r})}{\partial x_i \partial x_k} + c_{Tq}^2 \frac{\partial^2 g^{(q)}_{ij}(\mathbf{r})}{\partial x_i \partial x_l} = -\delta_{ij} \delta(\mathbf{r}), \]

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where \( \mathbf{r} \equiv \mathbf{x} - \mathbf{x}' \). We reduce this partial differential equation into a system of linear equations by taking the Fourier transform on both sides;

\[
-\delta_{ij} = \omega^2 \hat{g}^{(q)}_{ij}(q) - \left( c_{Lq}^2 - c_{Tq}^2 \right) q_i q_k \hat{g}^{(q)}_{kj}(q) - c_{Tq}^2 q_i q_l \hat{g}^{(q)}_{lj}(q)
\]

\[
\left[ - \left( c_{Lq}^2 - c_{Tq}^2 \right) q_i q_k + \left( \omega^2 - c_{Tq}^2 q_i q_l \right) \delta_{ik} \right] \hat{g}^{(q)}_{kj}(q)
\]

\[
\left[ \left( -c_{Lq}^2 q^2 + \omega^2 \right) \Pi_{ik} + \left( \omega^2 - c_{Tq}^2 q^2 \right) \left( \delta_{ik} - \Pi_{ik} \right) \right] \hat{g}^{(q)}_{kj}(q),
\]  
(9.140)

where \( \Pi_{ij} \equiv \hat{q}_i \hat{q}_j \) is the projection operator on a given wavevector \( \mathbf{q} \). Equation (9.140) implies that \( g^{(q)}(q) \) is the inverse of \(- (c_{Lq}^2 k^2 + \omega^2) \Pi - (\omega^2 - c_{Tq}^2 k^2) (I - \Pi)\), and thus using the following properties of projection operators

\[
\Pi \cdot \Pi = \Pi, \quad (I - \Pi) \cdot (I - \Pi) = I - \Pi, \quad (I - \Pi) \cdot \Pi = 0,
\]

one can easily find the explicit formula for the Fourier transform \( g^{(q)}(k) \):

\[
\hat{g}^{(q)}_{ij}(q) = \left[ \frac{1}{c_{Lq}^2 q^2 - \omega^2} \Pi_{ij} + \frac{1}{c_{Tq}^2 q^2 - \omega^2} \left( \delta_{ij} - \Pi_{ij} \right) \right]
\]

\[
= \left( \frac{1}{c_{Lq}^2 q^2 - \omega^2} - \frac{1}{c_{Tq}^2 q^2 - \omega^2} \right) \Pi_{ij} + \frac{1}{c_{Tq}^2 q^2 - \omega^2} \delta_{ij}
\]

\[
= \frac{1}{\omega^2} \left( \frac{1}{q^2 - k_{Lq}^2} - \frac{1}{q^2 - k_{Tq}^2} \right) q_i q_j + \frac{1}{c_{Tq}^2 q^2 - k_{Tq}^2} \delta_{ij},
\]  
(9.141)

where \( k_{Tq} \equiv \omega / c_{Tq} \) and \( k_{Lq} \equiv \omega / c_{Lq} \). Using the relation between the Fourier transform of derivatives, we can write

\[
\hat{g}^{(q)}_{ij}(q) = -\frac{1}{\omega^2} \text{F.T.} \left\{ \frac{\partial^2}{\partial x_i \partial x_j} \text{F.T.}^{-1} \left[ \frac{1}{q^2 - k_{Lq}^2} - \frac{1}{q^2 - k_{Tq}^2} \right] \right\} + \frac{1}{c_{Tq}^2 q^2 - k_{Tq}^2} \delta_{ij},
\]

(9.142)

where F.T. and F.T.\(^{-1}\) represent the Fourier and inverse Fourier transforms, respectively.
We note that the inverse Fourier transform of \((k^2 - k_{Lq}^2)^{-1}\) corresponds to the Green function \(g_H(r; k_{Lq})\) of the Helmholtz equation with a wavenumber \(k_{Lq}\) in infinite space \(\mathbb{R}^d\) given in

\[
\frac{\partial^2 g_H(r; k_{Lq})}{\partial x_i^2} + k_{Lq}^2 g_H(r; k_{Lq}) = -\delta(r),
\]

(9.143)

where \(r \equiv x - x'\). In direct space, an explicit formula for this Green function is well-known:

\[
g_H(r; k) = \frac{i}{4} \left( \frac{k}{2\pi|r|} \right)^{d/2-1} \mathcal{H}_{d/2-1}^{(1)}(k|r|),
\]

(9.144)

which reduces for \(d = 1, 3\) as

\[
g_H(r; k) = \begin{cases} 
  i \frac{e^{ik|r|}}{2|k|^2}, & d = 1 \\
  e^{ik|r|} \frac{4\pi}{4\pi|r|}, & d = 3
\end{cases}
\]

(9.145)

Using (9.144) and the following identities

\[
\frac{\partial r^{-\alpha}}{\partial x_i} = -\alpha \hat{r}_i r^{-\alpha-1},
\]

(9.146)

\[
\frac{\partial \hat{r}_j}{\partial x_j} = \frac{1}{r}(\delta_{ij} - \hat{r}_i \hat{r}_j),
\]

(9.147)

one can explicitly write out the dyadic Green function as follows:

\[
g_{ij}^{(q)}(x, x') = -\frac{1}{\omega^2} \frac{\partial^2}{\partial x_i \partial x_j} \left[ g_H(r; k_{Lq}) - g_H(r; k_{Tq}) \right] + \frac{k_{Tq}^2}{\omega^2} g_H(r; k_{Tq}) \delta_{ij}
\]

(9.148)

\[
= \frac{1}{\omega^2} \left[ g_H''(r; k_{Tq}) - g_H''(r; k_{Lq}) - \frac{1}{r} \left( g_H'(r; k_{Tq}) - g_H'(r; k_{Lq}) \right) \right] \hat{r}_i \hat{r}_j
\]

\[
+ \frac{1}{\omega^2} \left[ \frac{1}{r} \left( g_H'(r; k_{Tq}) - g_H'(r; k_{Lq}) \right) + k_{Tq}^2 g_H(r; k_{Tq}) \right] \delta_{ij}
\]

(9.149)

\[
= A_d(r) \hat{r}_i \hat{r}_j + B_d(r) \delta_{ij},
\]

(9.150)

where the prime symbol \('\) denotes the derivative with respect to \(r\), and \(A_d(r)\) and \(B_d(r)\) are constants.
\( B_d(r) \) are defined in Eqs. (9.95) and (9.96), respectively.

### 9.11.2 Fourth-Rank Green Function \( G^{(q)}(r) \)

We derive the fourth-rank Green function employed in (9.83). To do so, we first need to derive the integral equation (9.83) from (9.81).

\[
\epsilon_{ij}(x) - (\epsilon_0)_{ij}(x) = \frac{1}{2} \left[ \frac{\partial \tilde{u}_i(x)}{\partial x_j} + \frac{\partial \tilde{u}_j(x)}{\partial x_i} \right] = \frac{1}{2} \int_\epsilon dx' \left( \frac{\partial g^{(q)}_{ik}}{\partial x_j} + \frac{\partial g^{(q)}_{jk}}{\partial x_i} \right) \frac{\partial P_{kl}(x')}{\partial x'_l}
\]

\[
\frac{1}{2} \int_\epsilon dx' \frac{\partial}{\partial x'_l} \left[ \left( \frac{\partial g^{(q)}_{ik}}{\partial x_j} + \frac{\partial g^{(q)}_{jk}}{\partial x_i} \right) P_{kl}(x') \right] - \frac{1}{2} \int_\epsilon dx' \frac{\partial}{\partial x'_l} \left( \frac{\partial g^{(q)}_{ik}}{\partial x_j} + \frac{\partial g^{(q)}_{jk}}{\partial x_i} \right) P_{kl}(x'),
\]

(9.151)

where we have used integration by parts to remove the divergence applied to the polarization tensor. Applying the divergence theorem to the first integral in (9.151) yields

\[
\epsilon_{ij}(x) - (\epsilon_0)_{ij}(x) = \frac{1}{2} \oint d\alpha' \hat{r}_l \left[ \left( \frac{\partial g^{(q)}_{ik}}{\partial x_j} + \frac{\partial g^{(q)}_{jk}}{\partial x_i} \right) P_{kl}(x') \right] - \frac{1}{2} \int_\epsilon dx' \frac{\partial}{\partial x'_l} \left( \frac{\partial g^{(q)}_{ik}}{\partial x_j} + \frac{\partial g^{(q)}_{jk}}{\partial x_i} \right) P_{kl}(x'),
\]

(9.152)

where \( \oint d\alpha' \hat{r}_l \) denotes the surface integral over the exclusion-region boundary.

Use of (9.150), (9.146), and (9.147) gives an expression for the integrand in the first integral (9.152):

\[
\left( \frac{\partial g^{(q)}_{ik}}{\partial x_j} + \frac{\partial g^{(q)}_{jk}}{\partial x_i} \right) = \left[ (\hat{r}_i \delta_{jk} + \hat{r}_j \delta_{ik} + 2\hat{r}_k \delta_{ij}) \frac{A_d(r)}{r} + (\hat{r}_j \delta_{ik} + \hat{r}_i \delta_{jk}) B'_d(r) \right]
\]

\[+ 2\hat{r}_i \hat{r}_j \hat{r}_k \left[ A'_d(r) - \frac{2}{r} A_d(r) \right].
\]

(9.153)
Thus, for a spherical exclusion-region, the first integral in (9.152) is written as

\[
\frac{1}{2} \lim_{r \to 0^+} r^{d-1} \left\{ \frac{A_d(r)}{r} \int_r d\Omega_d \, \hat{r}_i \delta_{jk} + \hat{r}_j \delta_{ik} + 2\hat{r}_k \delta_{ij} \right\} P_{kl}(x') + B'_d(r) \int_r d\Omega_d \, \hat{r}_i \delta_{jk} + \hat{r}_j \delta_{ik} + 2\hat{r}_k \delta_{ij} \right\} P_{kl}(x') \\
+ 2 \left[ A'_d(r) - \frac{2}{r} A_d(r) \right] \int_r d\Omega_d \, \hat{r}_i \hat{r}_j \hat{r}_k \hat{r}_l P_{kl}(x') \right\} \\
= \frac{1}{2} \left\{ \lim_{r \to 0^+} \left[ r^{d-2} A_d(r) \right] \lim_{r \to 0^+} \int_r d\Omega_d \, \hat{r}_i \delta_{jk} + \hat{r}_j \delta_{ik} + 2\hat{r}_k \delta_{ij} \right\} P_{kl}(x') \\
+ \lim_{r \to 0^+} \left[ r^{d-1} B'_d(r) \right] \lim_{r \to 0^+} \int_r d\Omega_d \, \hat{r}_i \delta_{jk} + \hat{r}_j \delta_{ik} + 2\hat{r}_k \delta_{ij} \right\} P_{kl}(x') \\
+ 2 \lim_{r \to 0^+} r^{d-1} \left[ A'_d(r) - \frac{2}{r} A_d(r) \right] \lim_{r \to 0^+} \int_r d\Omega_d \, \hat{r}_i \hat{r}_j \hat{r}_k \hat{r}_l P_{kl}(x') \right\} \\
= - \frac{1}{(2 + d) d} \left( \frac{2}{c_L^2} - \frac{d}{c_T^2} \right) P_{ij}(x) - \frac{1}{(2 + d) d} \left( \frac{1}{c_L^2} - \frac{1}{c_T^2} \right) P_{kk}(x) \delta_{ij} \\
= - \frac{1}{d c_L^2} \left( \Lambda_h \right)_{ijkl} + \frac{1}{d + 2} \left( \frac{2}{d c_L^2} + \frac{1}{d c_T^2} \right) \left( \Lambda_s \right)_{ijkl} \right\} P_{kl}(x) \\
\equiv -D^{(q)}_{ijkl} P_{kl}(x), \quad (9.157)
\]

where \( A_d(r) \) and \( B_d(r) \) are given in (9.95) and (9.96), respectively, and we have used expressions in Sec. 9.12.5.

The second term in (9.152) is obtained by using \(-\frac{\partial}{\partial x_i} \to \frac{\partial}{\partial x'_{i}}\) and then using the fact that \( P_{kl} \) is a symmetric tensor:

\[
- \frac{1}{2} \int_\epsilon \frac{dx'}{\frac{\partial g^{(q)}_{i k}}{\partial x'_l} + \frac{\partial g^{(q)}_{j k}}{\partial x'_l}} P_{kl}(x') = \frac{1}{2} \int_\epsilon \frac{dx'}{\frac{\partial g^{(q)}_{i k}}{\partial x'_l} + \frac{\partial g^{(q)}_{j k}}{\partial x'_l}} P_{kl}(x') \\
= \frac{1}{4} \left[ \frac{\partial^2 g^{(q)}_{i k}}{\partial x_j \partial x_l} + \frac{\partial^2 g^{(q)}_{j k}}{\partial x_i \partial x_l} + \frac{\partial^2 g^{(q)}_{i k}}{\partial x_j \partial x_k} + \frac{\partial^2 g^{(q)}_{j k}}{\partial x_i \partial x_k} \right] P_{kl}(x') \frac{dx'}{H^{(q)}_{ijkl}(x - x')} P_{kl}(x') dx'. \\
\]  

(9.158)

Thus, the fourth-rank tensor \( H^{(q)}(r) \) meets (9.87). Combination of (9.157) and (9.158) results in (9.84).

**Remarks:**
1. The constant $D^{(q)}$ also can be computed by using (9.102) in the limit of $|q| \to 0^+$. 

# 9.12 Appendix E: Useful Identities

In this section, we list some useful identities that are frequently used in this chapter.

## 9.12.1 Recurrence Relation

Here, we present a useful recurrence relation associated with Bessel functions as well as Hankel functions that is heavily used to simplify formulas in Sec. 9.10.3. Specifically, for all $\nu > 0$,

$$2\nu Z_\nu(x) = xZ_{\nu-1}(x) + xZ_{\nu+1}(x),$$

(9.159)

where $Z_\nu(x)$ corresponds to Bessel function of the first kind $J_\nu(x)$, Bessel function of the second kind $Y_\nu(x)$, and Hankel function of the first kind $H^{(1)}_\nu(x)$. From this recurrence relation, one can also obtain the following identity

$$\frac{3}{d(d+2)}x^2Z_{d/2+3}(x) + \frac{6}{d}xZ_{d/2+2}(x) + 3Z_{d/2+1}(x) = \frac{3}{d(d+2)}x^2Z_{d/2-1}(x).$$

## 9.12.2 Properties of Projection Tensors

Here, we briefly discuss important properties of the hydrostatic projection tensor $\Lambda_h$ and the shear projection tensor $\Lambda_s$; see the main text for definitions. While the former $\Lambda_h$ projects onto fields that are isotropic everywhere, whereas the latter $\Lambda_s$ projects onto fields that are trace-free. These two tensors are useful due to the following
identities:

\[ \Lambda_h + \Lambda_s = I, \]  
\[ \Lambda_h : \Lambda_s = \Lambda_s : \Lambda_h = 0, \]  
\[ \Lambda_h : \Lambda_h = \Lambda_h, \]  
\[ \Lambda_s : \Lambda_s = \Lambda_s, \]  
\[ \Lambda_h : \Lambda_h = \Lambda_h: I = 1, \]  
\[ \Lambda_s : \Lambda_s = \Lambda_s: I = \frac{(d-1)(d+2)}{2}, \]  

where the symmetric fourth-order identity tensor \( I \) is defined in a component form as

\[ I_{ijkl} \equiv \frac{1}{2} (\delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}). \]

9.12.3 Properties of \( H^{(q)}(r) \)

We list traces of the fourth-rank tensor \( H^{(q)}(r) \) given in (9.18):

\[ H_{ijkl}^{(q)}(r) = \frac{i \pi}{2(2\pi)^{d/2}} \frac{1}{\sqrt{2}^d r_{d/2+2}} \left[ r_L^{d/2+2} \left[ -H_{d/2+1}^{(1)}(r_L) \delta_{kl} + r_L H_{d/2+1}^{(1)}(r_L) \hat{r}_k \hat{r}_l \right] \right], \]

\[ H_{ijij}^{(q)}(r) = \frac{-i \pi}{2(2\pi)^{d/2}} \frac{1}{\sqrt{2}^d r_{d/2+2}} \left[ r_L^{d/2+3} H_{d/2-1}^{(1)}(r_L) + \frac{d-1}{2} r_T^{d/2+3} H_{d/2-1}^{(1)}(r_T) \right], \]

where \( r_L \equiv k_{Lq} r \) and \( r_T \equiv k_{Tq} r \). In order to obtain these formulas, we apply the recurrence relations of Hankel functions given in (9.159) to (9.18).

9.12.4 Tensor Contractions

Here, we present some formulas for the inner products (contractions) of tensors employed to express \( H^{(q)}(r) \) given in (9.18). These formulas are heavily used in Sec.
9.10.3. For convenience, we defined the following two fourth-rank tensors:

\[(T_a)_{ijkl}(r) \equiv \delta_{ij} \hat{r}_k \hat{r}_l,\]  
\[(T_b)_{ijkl}(r) \equiv \hat{r}_i \hat{r}_j \delta_{kl}.\]  

(9.170)  
(9.171)

Double and quadruple contractions of two fourth-rank tensors are listed in Table 9.1 and 9.2, respectively.

Table 9.1: Double contractions of two four-rank tensors (i.e., \(A : B\) or, equivalently, \(A_{ijmn}B_{mnlk}\)) in \(\mathbb{R}^d\). Here, \(T_a\) and \(T_b\) are defined in (9.170) and (9.171), respectively.

<table>
<thead>
<tr>
<th>A</th>
<th>B</th>
<th>(\Lambda_h)</th>
<th>I</th>
<th>(T_1(r))</th>
<th>(T_2(r))</th>
<th>(T_3(r))</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\Lambda_h)</td>
<td>(\Lambda_h)</td>
<td>(\Lambda_h)</td>
<td>(\Lambda_h)</td>
<td>(\frac{1}{2}(\Lambda_h + T_a))</td>
<td>(\frac{1}{2}T_a)</td>
<td>(\frac{1}{2}T_a)</td>
</tr>
<tr>
<td>I</td>
<td>(\Lambda_h)</td>
<td>I</td>
<td>(T_1)</td>
<td>(\frac{1}{2}T_1)</td>
<td>(\frac{1}{2}(\Lambda_h + T_3))</td>
<td>(\frac{1}{2}(T_a + T_3))</td>
</tr>
<tr>
<td>(T_1(r))</td>
<td>(\frac{1}{2}(\Lambda_h + T_b))</td>
<td>(T_1)</td>
<td>(\frac{1}{2}T_1)</td>
<td>(\frac{1}{2}(\Lambda_h + T_3))</td>
<td>(\frac{1}{2}(T_a + T_3))</td>
<td>(\frac{1}{2}(T_a + T_3))</td>
</tr>
<tr>
<td>(T_2(r))</td>
<td>(\frac{1}{2}T_b)</td>
<td>(T_2)</td>
<td>(\frac{1}{2}(T_3 + T_b))</td>
<td>(\frac{1}{2}(T_2 + T_3))</td>
<td>(T_3)</td>
<td>(T_3)</td>
</tr>
<tr>
<td>(T_3(r))</td>
<td>(\frac{1}{2}T_b)</td>
<td>(T_3)</td>
<td>(\frac{1}{2}(T_3 + T_b))</td>
<td>(T_3)</td>
<td>(T_3)</td>
<td>(T_3)</td>
</tr>
</tbody>
</table>

Table 9.2: Quadruple contractions of two fourth-rank tensors (i.e., \(A:B\) or, equivalently, \(A_{ijkl}B_{ijkl}\)) in \(\mathbb{R}^d\).

<table>
<thead>
<tr>
<th>A</th>
<th>B</th>
<th>(\Lambda_h)</th>
<th>I</th>
<th>(T_1(r))</th>
<th>(T_2(r))</th>
<th>(T_3(r))</th>
<th>(T_1(s))</th>
<th>(T_2(s))</th>
<th>(T_3(s))</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\Lambda_h)</td>
<td>1</td>
<td>1</td>
<td>(\frac{1}{d})</td>
<td>(\frac{1}{d})</td>
<td>1</td>
<td>(\frac{1}{d})</td>
<td>(\frac{1}{d})</td>
<td>(\frac{1}{d})</td>
<td>(\frac{1}{d})</td>
</tr>
<tr>
<td>I</td>
<td>(d(d+1)/2)</td>
<td>1</td>
<td>(\frac{d+1}{2})</td>
<td>(\frac{d+1}{2})</td>
<td>1</td>
<td>(\frac{d+1}{2})</td>
<td>(\frac{d+1}{2})</td>
<td>1</td>
<td>(\hat{r} \cdot \hat{s})^2</td>
</tr>
<tr>
<td>(T_1(r))</td>
<td>1</td>
<td>1</td>
<td>(\frac{d+1}{2})</td>
<td>1</td>
<td>(\frac{d+1}{2})</td>
<td>(\frac{d+1}{2})</td>
<td>(\hat{r} \cdot \hat{s})^2</td>
<td>(\hat{r} \cdot \hat{s})^2</td>
<td></td>
</tr>
<tr>
<td>(T_2(r))</td>
<td>(\frac{d+1}{2})</td>
<td>(\frac{d+1}{2})</td>
<td>1</td>
<td>(\frac{d+1}{2})</td>
<td>(\hat{r} \cdot \hat{s})^2</td>
<td>(\hat{r} \cdot \hat{s})^2</td>
<td>(\hat{r} \cdot \hat{s})^2</td>
<td></td>
<td></td>
</tr>
<tr>
<td>(T_3(r))</td>
<td>(\frac{d+1}{2})</td>
<td>(\frac{d+1}{2})</td>
<td>1</td>
<td>1</td>
<td>(\hat{r} \cdot \hat{s})^2</td>
<td>(\hat{r} \cdot \hat{s})^2</td>
<td>(\hat{r} \cdot \hat{s})^2</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

9.12.5 Integral over Orientations

It is useful to compute integrals of tensors over orientations in \(\mathbb{R}^d\) for \(d \geq 3\) when one computes \(C_3^{(p)}(k_{L_q})\) and \(D_3^{(p)}(k_{L_q})\) in Sec. 9.10.3. In \(d\)-dimensional spherical
coordinates, the integral of a given tensor $U(\hat{r})$ over orientations can be written as

$$\oint d\Omega U(\hat{r}) = \int_0^\pi d\theta \sin^{d-2}(\theta) \left[ \int_0^{2\pi} d\phi \prod_{i=1}^{d-3} \int_0^\pi d\theta_i \sin(\theta_i) \ U(\hat{r}) \right]$$

where $\hat{e}_d$ is a unit vector along the rotation axis for the azimuthal angle $\phi$ (e.g., $\hat{z}$ for $d = 3$), and $\oint_{\hat{r} \cdot \hat{e}_d = \cos \theta} d\Omega(\hat{r})$ [shortly, $\oint_\theta d\Omega(\hat{r})$] stands for an integral over a unit sphere satisfying the condition $\hat{r} \cdot \hat{e}_d = \cos \theta$.

Using an identity

$$\int_0^\pi d\theta \sin^n \theta = \sqrt{\pi} \frac{\Gamma((n+1)/2)}{\Gamma(n/2 + 1)},$$

one derives the following integrals:

$$\oint_\theta d\Omega = \frac{2\sqrt{\pi}^{d-1}}{\Gamma((d-1)/2)} = \Omega_{d-1},$$

$$\oint_\theta d\Omega \hat{r}_i = \Omega_{d-1} \cos \theta(\hat{e}_d)_i,$$

$$\oint_\theta d\Omega \hat{r}_i \hat{r}_j = \frac{\Omega_{d-1}}{d-1} \left[ \sin^2 \theta \delta_{ij} + (d \cos^2 \theta - 1)(\hat{e}_d)_i(\hat{e}_d)_j \right],$$

$$\oint_\theta d\Omega \hat{r}_i \hat{r}_j \hat{r}_k \hat{r}_l = \frac{\Omega_{d-1}}{d-1} \left\{ \frac{1}{d+1} \sin^4 \theta (\delta_{ij} \delta_{kl} + \delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk}) + \sin^2 \theta \left( 1 - \frac{d+2}{d+1} \sin^2 \theta \right) [2 T_1(\hat{e}_d) + 4 T_2(\hat{e}_d)] + \left[ (d - 1) \cos^4 \theta - 6 \sin^2 \theta + \frac{6d + 9}{d+1} \sin^4 \theta \right] (\hat{e}_d)_i (\hat{e}_d)_j (\hat{e}_d)_k (\hat{e}_d)_l \right\},$$

where $\Omega_d$ is the surface area of a unit sphere in $\mathbb{R}^d$. Use of (9.174)-(9.177) gives the

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orientation integrals of the following fourth-rank tensors $\Lambda_h, I, T_1(r), T_2(r), T_3(r)$ as

\[
\oint d\Omega \Lambda_h = \Omega_{d-1} \Lambda_h, \quad \oint d\Omega I = \Omega_{d-1} I
\]

(9.178)

\[
\oint d\Omega T_1(r) = \frac{\Omega_{d-1}}{d-1} [d(1 - \cos^2 \theta) \Lambda_h + (d \cos^2 \theta - 1) T_1(\hat{e}_d)]
\]

(9.179)

\[
\oint d\Omega T_2(r) = \frac{\Omega_{d-1}}{d-1} [(1 - \cos^2 \theta) I + (d \cos^2 \theta - 1) T_2(\hat{e}_d)]
\]

(9.180)

\[
\oint d\Omega T_3(r) = \frac{\Omega_{d-1}}{d-1} \left\{ \frac{1}{d+1} \sin^4 \theta (d \Lambda_h + 2 I) + \sin^2 \theta \left( 1 - \frac{d+2}{d+1} \sin^2 \theta \right) \left[ 2 T_1(\hat{e}_d) + 4 T_2(\hat{e}_d) \right] + \left[ (d-1) \cos^4 \theta - 6 \sin^2 \theta + \frac{6d+9}{d+1} \sin^4 \theta \right] T_3(\hat{e}_d) \right\}. \quad (9.181)
\]

Substituting (9.174)-(9.177) into the orientation integral (9.172) gives

\[
\oint d\Omega = \Omega_d, \quad (9.182)
\]

\[
\oint d\Omega \hat{r}_i = 0, \quad (9.183)
\]

\[
\oint d\Omega \hat{r}_i \hat{r}_j = \frac{\Omega_d}{d} \delta_{ij}, \quad (9.184)
\]

\[
\oint d\Omega \hat{r}_i \hat{r}_j \hat{r}_k \hat{r}_l = \frac{\Omega_d}{d(d+2)} \left( \delta_{ij} \delta_{kl} + \delta_{ik} \delta_{jl} + \delta_{il} \delta_{jk} \right) = \frac{\Omega_d}{d+2} \left( [\Lambda_h]_{ijkl} + \frac{2}{d} [I]_{ijkl} \right)
\]

(9.185)

\[
= \frac{\Omega_d}{d} \left( [\Lambda_h]_{ijkl} + \frac{2}{d+2} [\Lambda_s]_{ijkl} \right). \quad (9.186)
\]

Use of (9.184) and (9.186) gives

\[
\oint d\Omega T_1(r) = \Omega_d \Lambda_h, \quad (9.187)
\]

\[
\oint d\Omega T_2(r) = \frac{\Omega_d}{d} I = \frac{\Omega_d}{d} (\Lambda_h + \Lambda_s), \quad (9.188)
\]

\[
\oint d\Omega T_3(r) = \frac{\Omega_d}{d+2} \left( \Lambda_h + \frac{2}{d} I \right) = \Omega_d \left( \frac{1}{d} \Lambda_h + \frac{2}{d(d+2)} \Lambda_s \right), \quad (9.189)
\]

\[
\oint d\Omega T_a(r) = \oint d\Omega T_b(r) = \Omega_d \Lambda_h, \quad (9.190)
\]

where $T_a(r)$ and $T_b(r)$ are defined in Eqs. (9.170) and (9.171), respectively.
9.13 Appendix F: Evaluation of the Attenuation Functions

For general disordered two-phase media that are statistically isotropic, it is often difficult to numerically compute \( \text{Re}[\mathcal{F}(Q)] \) or \( \text{Re}[F(Q)] \) by using Eqs. (9.128) and (9.62), respectively. We note that Appendix 8.16 discuss alternative expressions that are easy to integrate.

9.14 Appendix G: Simulation Details

Here we provide additional details about simulation procedures and values of the simulation parameters that we used. We list parameters employed to numerically generate sphere packings for computing the spectral density and the attenuation functions in Sec. 9.14.1. In Sec. 9.14.2, we describe the full-waveform simulations that we employed in the main text.

9.14.1 Parameters for Numerically Generated Packings

We numerically generate packings in three dimensions for disordered stealthy hyperuniform packings and hyperuniform polydisperse packings to compute the spectral density \( \tilde{\chi}_V(Q) \). For each model, we generate \( N_c \) different packings of particle radius \( a \), \( N \) particles, and number density \( \rho \) in a periodic fundamental cell. Here, we list these parameters as well as some other relevant parameters; see Table 9.3.

Stealthy hyperuniform packings are generated via the collective-coordinate optimization technique. For this model, a parameter \( Q_U \) defines the stealthy regions, and \( \sigma \) represents the diameter of the repulsion region of each particle. For stealthy hyperuniform packings (or point patterns), it is useful to define the \( \chi \) parameter, which the ratio of constrained degrees of freedom to total number of degrees of freedom.
\[ \chi \equiv \frac{\mathcal{M}}{d(N - 1)}. \]  

(9.191)

For \( 0 < \chi < 1/2 \), they are highly degenerate and disordered, whereas for \( 1/2 < \chi < 1 \) they crystallize [352].

Table 9.3: Parameters of sphere packings used to compute \( \tilde{\chi}_V(Q) \). We generate realizations of disordered stealthy hyperuniform packings (SHP) and hyperuniform polydisperse packings (HPP) in three dimensions. For each model, \( N_c \) is the number of distinct packings, \( N \) particle number, \( a \) is particle radius, \( \rho \) is the number density, and \( \phi_2 \) is the packing fraction. For hyperuniform polydisperse packings, \( a \) stands for the mean particle radius, i.e., \( a \equiv [\phi_2 / v_1(1)]^{1/d} \). Quantities \( Q_U \) and \( \sigma \) are parameters used in the collective-coordinate optimization method; see Sec. III in the main text. The \( \chi \) parameter is defined in Eq. (9.191).

<table>
<thead>
<tr>
<th>Systems \ Parameters</th>
<th>( N )</th>
<th>( \rho )</th>
<th>( N_c )</th>
<th>( Q_U a )</th>
<th>( \sigma )</th>
<th>( a )</th>
<th>( \chi )</th>
</tr>
</thead>
<tbody>
<tr>
<td>3D SHP (( \phi_2 = 0.25 ))</td>
<td>1000</td>
<td>1</td>
<td>300</td>
<td>1.5</td>
<td>0.8</td>
<td>0.3908</td>
<td>0.1582</td>
</tr>
<tr>
<td>3D SHP (( \phi_2 = 0.4 ))</td>
<td>1000</td>
<td>1</td>
<td>300</td>
<td>1.5</td>
<td>0.92</td>
<td>0.4571</td>
<td>0.1031</td>
</tr>
<tr>
<td>3D HPP (( \phi_2 = 0.25 ))</td>
<td>1000</td>
<td>1</td>
<td>300</td>
<td>-</td>
<td>-</td>
<td>0.3908</td>
<td>-</td>
</tr>
</tbody>
</table>

9.14.2 Full-Waveform Simulations

We employ the dynamic extension of the fast-Fourier transform-based homogenization scheme devised for the purely static problem [217, 73]. We note that such dynamic extensions were first employed in Ref. [158]. The simulation results presented in the main text are obtained with setting the number of voxels in each side as \( N_1 = N_2 = N_3 = 101 \).
Algorithm 1: Fast-Fourier transform-based homogenization scheme for the effective dynamic bulk and shear moduli. The applied strain $\epsilon_0$ and the associated wavevector $k$ depend on the elastic moduli to compute:

$(\epsilon_0)_{ij} = \delta_{ij}$ with $k = k_{\epsilon_1} \hat{x}$, for $K_e$, $(\epsilon_0)_{ij} = 1 - \delta_{ij}$ with $k = k_{\tau_1} \hat{x}$, for $G_e$.

Result: $K_e$ or $G_e$ at a given frequency $\omega$

Set tolerance $\Delta = 10^{-9}$ and mass density $\rho_1 = \rho_2 = 1$.

Assign bulk modulus $K_i$ and shear modulus for phase $i$ ($i = 1, 2$).

Voxelize a two-phase medium in a periodic fundamental cell into $N_1 \times N_2 \times N_3$ voxels.

1. Set $m = 1$, $\Delta_{\text{div}} = 1$, $\epsilon^{(0)}(x) e^{-ik \cdot x} = \epsilon_0$, and $P^{(0)}(x) e^{-ik \cdot x} = [C(x) - C_1]: [\epsilon^{(0)}(x) e^{-ik \cdot x}]$.

   while $\Delta_{\text{div}} > \Delta$ do

   i. $\tilde{P}^{(m-1)}(q + k) = \text{FFT}[P^{(m-1)}(x) e^{-ik \cdot x}]$.

   ii. Compute $\tilde{\sigma}(q + k) = C_1 : \tilde{\epsilon}^{(m-1)}(q + k) + \tilde{P}^{(m-1)}(q + k)$ and the consequent error in wave equation:

   \[
   \Delta_{\text{div}} = \left\| \omega^2 \rho_1 \epsilon^{(m)}_{ij}(q') - \frac{1}{2} \left[ \sigma_{ij}' \tilde{\sigma}_{ij}(q') + \epsilon_{ij}' \tilde{\epsilon}_{ij}(q') \right] \right\|
   \]

   where $q' \equiv q + k$, and for a second-rank tensor $\tilde{T}_{ij}(q)$,

   \[
   \| \tilde{T}_{ij}(q) \| \equiv \left[ \frac{1}{N_1 N_2 N_3} \sum_q \sum_{i,j=1}^3 |\tilde{T}_{ij}(q)|^2 \right]^{1/2}.
   \]

   Stop if $\Delta_{\text{div}} < \Delta$.

   iii. Using the Green function $\tilde{G}^{(q)}(q)$ given in Eq. (9.99), compute

   \[
   \tilde{\epsilon}^{(m)}(q + k) = \begin{cases} 
   N_1 N_2 N_3 \epsilon_0, & q = 0 \\
   \tilde{G}^{(q)}(q + k) : \tilde{P}^{(m-1)}(q + k), & \text{otherwise}.
   \end{cases}
   \]

   iv. $\epsilon^{(m)}(x) e^{-ik \cdot x} = \text{IFFT}[\tilde{\epsilon}^{(m)}(q + k)]$.

   v. Compute $P^{(m)}(x) e^{-ik \cdot x} = [C(x) - C_1] : [\epsilon^{(m)}(x) e^{-ik \cdot x}]$.

   vi. Set $m \rightarrow m + 1$.

   end

2. Compute the effective moduli:

$K_e = \text{Tr}[(\sigma)]/\text{Tr}[(\epsilon)]$, $G_e = \frac{1}{2} \sum_{i,j=1}^d \langle \sigma \rangle_{ij} / \left\{ \sum_{i,j=1}^d \langle \epsilon \rangle_{ij} - \text{Tr}[(\sigma)] \right\}$,

where $\text{Tr}[]$ denotes the trace, and $\langle \sigma \rangle = \tilde{\sigma}(k)$, $\langle \epsilon \rangle = \tilde{\epsilon}^{(m)}(k)$.
Chapter 10
Multifunctional Composites for Elastic and Electromagnetic Wave Propagation

10.1 Introduction

When the domain (inhomogeneity) length scale $\ell$ of a heterogeneous medium is much smaller than the system size, the medium can be regarded to be a homogeneous material with effective physical properties, such as the thermal (electric) conductivity $\sigma_e$, dielectric tensor $\epsilon_e$, or stiffness tensor $C_e$ [255, 213, 296]. Such effective properties depend on the phase properties, phase volume fractions $\phi_i$, and higher-order microstructural information [22, 55, 289, 264, 292, 242, 296, 255, 213]. Heterogeneous materials are ideally suited to achieve multifunctionality, since the best features of different materials can be combined to form a new material that has a broad spectrum of desired properties [290, 99, 306, 296, 304, 208, 191, 325]. Because the effective properties of a heterogeneous material reflect common morphological information, knowledge of one effective property can provide useful information about a different

All of the previous applications of cross-property relations for multifunctional design have focused on the static transport and elastic properties. Remarkably, however, nothing is known about analogous cross-property relations for various effective dynamic properties, each of which is of great interest in its own right. For example, in the case of propagation of electromagnetic waves in two-phase media, the key properties of interest is the frequency-dependent dielectric constant, which is essential for a wide range of applications, including remote sensing of terrain [318], investigation of the microstructures of biological tissues [270], probing artificial materials [363], studying wave propagation through turbulent atmospheres [284], investigation of electrostatic resonances [204], and design of materials with desired optical properties [270, 336]. An equally important dynamic situation occurs when elastic waves propagate through a heterogeneous medium, which is of great importance in geophysics [26, 170], exploration seismology [267], diagnostic sonography [258], crack diagnosis [281], architectural acoustics [326] and acoustic metamaterials [344].

Our study is motivated by the increasing demand for multifunctional composites with desirable wave characteristics for a specific bandwidth (i.e., a range of frequencies). Possible applications include sensors that detect changes in moisture content and water temperature [70], thin and flexible antennas [4], materials that efficiently convert acoustic waves into electrical energy [206], materials that can attenuate low-frequency sound waves and exhibit excellent mechanical strength [283], and materials with negative modulus in the presence of magnetic fields [340]; see Ref. [182] and references therein.

However, systematic designs of multifunctional materials with desirable elastody-
Figure 10.1: Schematics illustrating multifunctional applications of heterogeneous materials. Elastic and electromagnetic waves at two different wavenumbers (a) $k_I$ and (b) $k_{II}$ incident to, inside of and transmitted from a composite material (a large ellipse) consisting of a matrix phase (shown in yellow) and a dispersed phase (shown in cyan). Parallel lines and sinusoidal curves represent elastic and electromagnetic waves, respectively. (a) For an elastic wave with a wavenumber $k_I$, while the wavefronts inside this material experience microscopic disturbances, they effectively behave like a plane wave inside a homogeneous material with an effective wavenumber $(k_e)_I = \omega_I \left[ c_e^L + i \gamma_e^L \right]^{-1}$ and effective elastic moduli $K_e$ and $G_e$; see definition (10.3). Analogously, for an electromagnetic wave, this material behaves like a homogeneous material with an effective dielectric constant $\epsilon_e$. For instance, both elastic and electromagnetic waves are attenuated due to scattering if this composite has a non-zero scattering intensity at $k_I$ [i.e., $\gamma_e^L < 0$ and $\text{Im}[\epsilon_e] > 0$]. (b) For both types of waves (red) of a wavenumber $k_{II}$, this composite can be effectively transparent, if it has a zero-scattering intensity at $k_{II}$ [i.e., $\gamma_e^L = 0$ and $\text{Im}[\epsilon_e] = 0$].

Dynamic and electromagnetic properties have yet to be established. In this chapter, we utilize the accurate microstructure-dependent formulas for the effective dynamic dielectric constant $\epsilon_e$ and effective dynamic bulk $K_e$ and shear $G_e$ moduli derived in Chapters 8 and 9 to derive cross-property relations that link effective electromagnetic properties to effective elastodynamic properties. We show that these formulas can accurately capture the dynamic responses of composites for a wide range of wavelengths, i.e., beyond the long-wavelength limitation of conventional approximations; see Sec. 10.5.1 and Chapters 8 and 9. Such cross-property relations facilitate multifunctional designs. Two striking multifunctional design applications are schematically illustrated in Fig. 10.1.
We establish accurate cross-property relations linking the effective elastic and electromagnetic wave characteristics by utilizing the aforementioned microstructure-dependent formulas and by eliminating the common microstructural parameter $F(Q)$, called nonlocal attenuation function, defined in Eq. (10.5). Thus, these results enable one to determine the response of a composite to electromagnetic waves from the corresponding response to acoustic/elastic waves and vice versa. The resulting cross-property relations will have practical implications, as discussed in Secs. 10.5.3 and 10.6.

The primary applications we have in mind are disordered microstructures, both exotic and “garden” varieties because they can provide advantages over periodic ones with high crystallographic symmetries, which include perfect isotropy and robustness against defects. Such disordered media have recently been exploited for applications involving photonic bandgap materials [80, 196], gradient-index photonic metamaterials [336], compact spectrometers [243], random lasers [332, 57], bone replacement [241, 223], and impact-absorbers [92, 199].

We are particularly interested in studying the wave characteristics of exotic disordered two-phase media, such as disordered \textit{hyperuniform} and/or \textit{stealthy} ones, and their potential applications. Hyperuniform two-phase systems are characterized by anomalously suppressed volume-fraction fluctuations at long wavelengths [313, 346, 300] such that

$$\lim_{Q \to 0} \tilde{\chi}_V(Q) = 0,$$

where $Q \equiv |Q|$ refers to a wavenumber. Here, the spectral density $\tilde{\chi}_V(Q)$ is the Fourier transform of the autocovariance function $\chi_V(r) \equiv S_2^{(i)}(r) - \phi_i^2$, where $\phi_i$ is the volume fraction of phase $i$, and $S_2^{(i)}(r)$ gives the probability of finding two points separated by $r$ in phase $i$ at the same time [296, 255]. One special class of hyperuniform two-phase systems is disordered \textit{stealthy hyperuniform} media that are defined by zero-scattering intensity for a set of wavevectors around the origin.
Such materials are endowed with novel physical properties, including that they are transparent to electromagnetic waves down to a finite wavelength. We also explore the wave characteristics of disordered stealthy nonhyperuniform media that possess zero-scattering intensity for a set of wavevectors that do not include the origin.

In Sec. 10.6, we describe how our microstructure-dependent estimates enable one to design materials that have the targeted attenuation coefficients $\gamma^{L,T}$ for a range of wavenumbers (or, equivalently, frequencies) via inverse-problem approaches. Using the stealthy hyperuniform materials, we explicitly demonstrate that such engineered materials can serve as filters for elastic waves which selectively absorb or transmit waves “isotropically” for a prescribed range of wavenumbers. Furthermore, we show that we can engineer composites that exhibit anomalous dispersion, yielding resonance-like attenuation in $\gamma^{L,T}$.

In Sec. 10.2, we present background definitions and assumptions. In Sec. 10.3, we summarize the nonlocal strong-contrast approximations derived in Chapters 8 and 9. In Sec. 10.7, we briefly describe the four models of disordered two-phase dispersions. In Sec. 10.5, we discuss cross-property relations and the applications. Finally, we provide concluding remarks in Sec. 10.6.

10.2 Preliminaries

We consider two-phase heterogeneous materials in $d$-dimensional Euclidean space $\mathbb{R}^d$. For simplicity, the results reported here mainly focus on the case of $d = 3$. We also make three assumptions on the phase dielectric properties: (a) the dielectric tensors of both phases are isotropic, (b) their dielectric constants are real-valued and independent of frequency, and (c) their magnetic permeabilities are identical.

The three analogous assumptions for the elastodynamic problem are (a) both
phases are elastically isotropic, (b) their elastic moduli are real numbers independent of frequency, and (c) they have identical mass densities ($\rho_1 = \rho_2$). The last assumption is achievable for many pairs of solid materials, e.g., nickel, copper, and cobalt have mass densities about 8.9 g/cm$^3$, and tin and manganese have mass densities about 7.2 g/cm$^3$ [7], but they have considerably different elastic moduli.

When these assumptions are met, inside each domain of phase $p$ (= 1, 2), the elastic wave equation is given as [173]

$$\omega^2 u_i + \left( c_{Lp}^2 - c_{Tp}^2 \right) \frac{\partial^2 u_i}{\partial x_i \partial x_l} + c_{Tp}^2 \frac{\partial^2 u_i}{\partial x_l \partial x_l} = 0,$$

where a displacement field oscillates sinusoidally with a frequency $\omega$ [i.e., $u_i(x, t) = u_i(x) e^{-i\omega t}$], indices span integers between 1 and $d$, and the Einstein summation is implied. Here, $c_{Lp}$ and $c_{Tp}$ represent the longitudinal and transverse wave speeds$^1$ in phase $p$, respectively, and they are given as

$$c_{Lp}^2 \equiv \frac{[K_p + 2(1 - 1/d)G_p]}{\rho_p}, \quad c_{Tp}^2 \equiv \frac{G_p}{\rho_p},$$

where $K_p$ and $G_p$ are the bulk modulus and the shear modulus of phase $p$, respectively. For a frequency $\omega$, the corresponding longitudinal and transverse wavenumbers for elastic waves in phase $p$ (=1,2) are denoted by

$$k_{Lp} \equiv \frac{\omega}{c_{Lp}}, \quad \text{and} \quad k_{Tp} \equiv \frac{\omega}{c_{Tp}}, \quad (10.2)$$

respectively. Henceforth, we take “reference” and “polarized” phases to be phase 1 and 2, respectively; see Refs. [292, 296].

Formulas for the effective dielectric constant $\epsilon_e(k_1)$ and the effective elastic moduli $K_e(k_{Lq})$ and $G_e(k_{Lq})$ also lead to estimates of the effective wave characteristics,

---

$^1$Henceforth, ‘wave speeds’ always refer to the phase speeds, because the term ‘phase’ is reserved for a constituent material in this chapter.
including effective wave speeds $c_e$ and attenuation coefficient $\gamma_e$. For electromagnetic and elastic waves, the analogous quantities are given by

\begin{align}
\frac{c_e}{c_1} + i\frac{\gamma_e}{c_1} &= \sqrt{\epsilon_1/\epsilon_e},
\frac{c_e^L}{c_1} + i\frac{\gamma_e^L}{c_1} &= \sqrt{K_e + 2(1 - 1/d)G_e}/\rho_e,
\frac{c_e^T}{c_1} + i\frac{\gamma_e^T}{c_1} &= \sqrt{G_e/\rho_e},
\end{align}

where $c_1$ is the wave speed of electromagnetic waves in the reference phase, and $\rho_e$ is the effective mass density ($\rho_e = \rho_1 = \rho_2$). Note that, for the scaled attenuation coefficients $\gamma_e/c_e$, $\gamma_e^L/c_e^L$, and $\gamma_e^T/c_e^T$, a quantity $\exp[-2\pi\gamma_e/c_e]$ represents the factor by which the amplitude of the incident wave is attenuated for a period of time $2\pi/\omega$.

### 10.3 Microstructure-Dependent Approximation Formulas

#### 10.3.1 Effective Dielectric Constant

Here, we employ the nonlocal strong-contrast approximation of the effective dielectric constant derived in Chapter 8. This approximation is valid down to intermediate wavelengths ($k_1\ell \lesssim 1$).

For macroscopically isotropic media in $d$-dimensional Euclidean space $\mathbb{R}^d$, this formula depends on a functional $A_2(Q)$ involving $\tilde{\chi}_\nu(Q)$ [308]:

\begin{align}
\frac{\epsilon_e(k_1)}{\epsilon_1} &= 1 - \frac{d\beta\phi_2^2}{\phi_2(\beta\phi_2 - 1) - \frac{(d-1)\pi}{2^{(d-1)/2}\Gamma(d/2)} F(Q) \beta},
\end{align}

where $k_1$ is the wavenumber of the electromagnetic waves in the reference phase (phase 1), $d$ is the dimension, $\beta \equiv (\epsilon_2 - \epsilon_1)/[\epsilon_2 + (d - 1)\epsilon_1]$ is the dielectric “polarizability,” and $\epsilon_1$ and $\epsilon_2$ are the dielectric constants of phases 1 and 2, respectively. Here $F(Q)$
is what we call the *nonlocal attenuation function* defined by [158, 308]

\[
F(Q) = -i \frac{\Gamma(d/2)}{2\pi^{d/2}} Q^{d/2+1} \int \frac{\mathcal{H}^{(1)}_{d/2}(Qr)}{r^{d/2-1}} e^{-iQ \cdot r} \chi_V(r) \, dr
\]

(10.5)

\[
= -\frac{\Gamma(d/2)}{2^{d/2+1}\pi^{d/2}} Q^2 \int \frac{\tilde{\chi}_V(q)}{|q + Q|^2 - Q^2} \, dq,
\]

(10.6)

where \(Q\) is the wavevector of the incident waves, and \(\mathcal{H}^{(1)}_\nu(x)\) is the Hankel function of the first kind of order \(\nu\). Physically, the attenuation function \(F(Q)\) incorporates the contributions from all diffracted waves due to single, elastic scattering events when the wavenumber of the incident waves is \(Q\). The reader is referred to Chapters 8 and 9 for a derivation of \(F(Q)\). Numerical simulations of \(\epsilon_e(k_1)\) shown in Sec. 8.8 validate the high-predictive power of Eqs. (10.4) for a wide range of incident wavelengths, which popular approximation schemes [270, 198] cannot predict.

For statistically isotropic media in three dimensions, the nonlocal attenuation function can be rewritten as

\[
\text{Im}[F(Q)] = -\frac{Q}{2(2\pi)^{3/2}} \int_0^{2Q} q \, \tilde{\chi}_V(q) \, dq,
\]

(10.7)

\[
\text{Re}[F(Q)] = -\frac{2Q^2}{\pi} \text{p.v.} \int_0^{\infty} dq \frac{1}{q(Q^2 - q^2)} \, \text{Im}[F(q)],
\]

(10.8)

where p.v. stands for the Cauchy principal value. We elaborate on how to compute the attenuation function in Sec. 8.16.

### 10.3.2 Effective Elastic Moduli

Here, we employ the nonlocal strong-contrast approximations at the two-point level for \(K_e(k_L)\) and \(G_e(k_L)\) derived in Chapter 9. These approximations are valid in the
intermediate-wavelength regime \((k_{Lq} \ell \lesssim 1)\), and their explicit formulas are given as

\[
\frac{K_e(k_{Lq})}{K_1} = 1 - \frac{\kappa \phi_2^2 [1 + 2(1 - 1/d)G_1/K_1]}{[C_2(k_{Lq}) + \phi_2 (\kappa \phi_2 - 1)]}, \quad \tag{10.9}
\]

\[
\frac{G_e(k_{Lq})}{G_1} = 1 - \frac{(d + 2)\mu \phi_2^2 [1 + 2(1 - 1/d)G_1/K_1]}{2(1 + 2G_1/K_1) [D_2(k_{Lq}) + \phi_2 (\mu \phi_2 - 1)]}, \quad \tag{10.10}
\]

where \(C_2(k_{Lq})\) and \(D_2(k_{Lq})\) are \(k_{Lq}\)-dependent microstructural parameters given by

\[
C_2(k_{Lq}) = \frac{\pi}{2^{d/2} \Gamma(d/2)} F(k_{Lq}) \kappa, \quad \tag{10.11}
\]

\[
D_2(k_{Lq}) = \frac{\pi}{2^{d/2} \Gamma(d/2)} \frac{dc_{Lq}^2 F(k_{Tq}) + 2 c_{Tq}^2 F(k_{Lq})}{dc_{Lq}^2 + 2 c_{Tq}^2} \mu, \quad \tag{10.12}
\]

where \(F(Q)\) is the nonlocal attenuation function given in Eqs. (10.7) and (10.8).

The reader is referred to Chapter 9 for a derivation of these relations. Computer simulations in Sec. 9.14.2 verify that these modified formulas accurately predict microstructure-dependence of \(K_e(k_{Lq})\) and \(G_e(k_{Lq})\) down to intermediate wavelengths, where conventional approximation schemes \([153]\) are no longer valid.

Note that the approximations [Eqs. (10.9) and (10.10)] are conveniently written in terms of the wavenumber \(k_{Lq}\) associated with the reference phase. Furthermore, this wavenumber is directly proportional to the frequency \(\omega\) [Eq. (10.2)] and more suitable to describe microstructural information rather than the temporal quantity \(\omega\). For these reasons, we henceforth use the longitudinal wavenumber \(k_{Lq}\), instead of \(\omega\) or \(k_{Tq}\), as an independent variable for the effective elastic properties.

### 10.3.3 Static Limit

In the long-wavelength limit \((Q \to 0)\), the attenuation function vanishes as \(F(Q) \sim O(Q^2)\), which enables us to recover the previous corresponding static results \([289, 292]\) in which the effective properties are real-valued and identical to the Hashin-Shtrikman bounds on \(\epsilon_e, K_e,\) and \(G_e\) \([113, 296, 213]\), respectively. Note that these static limits
are also identical to the static Maxwell-Garnet approximations (or Hashin-Shtrikman estimates); see Chapters 8 and 9.

### 10.3.4 Long-Wavelength Regime

The effective dynamic properties \( \epsilon_e(k_1) \), \( K_e(k_{Lq}) \), and \( G_e(k_{Lq}) \) are generally complex-valued, implying that the associated waves propagating through this medium are attenuated. Such attenuation occurs due to scattering from the inhomogeneity even when both phases are dissipationless (i.e., real-valued, as we assume throughout this study).

In the long-wavelength regime \( (Q\ell \ll 1) \), we now demonstrate that attenuation is stronger in a nonhyperuniform medium than in a hyperuniform one by comparing their leading-order terms of the effective attenuation coefficients. The imaginary parts of effective properties and, importantly, the associated effective attenuation coefficients \( \gamma_e \) are approximately proportional to \( \text{Im}[F(Q)] \), which is easily evaluated from the spectral density by using Eq. (10.7). When the spectral density follows the power-law form \( \tilde{\chi}_V(Q) \approx a_1 + a_2 Q^\alpha \), where for nonhyperuniform media \( a_1 > 0 \) and hyperuniform media \( a_1 = 0 \), \( a_2 > 0 \), and \( \alpha > 0 \), \( \gamma_e \) in the limit \( Q \to 0 \) is given by

\[
\gamma_e \sim \begin{cases} 
Q^3, & \text{nonhyperuniform}, \\
Q^{3+\alpha}, & \text{hyperuniform}.
\end{cases}
\]

Thus, hyperuniform media are less dissipative than nonhyperuniform media due to the complete suppression (in the former) of single scattering events in the long-wavelength limit. Note for stealthy hyperuniform media [\( \tilde{\chi}_V(Q) = 0 \) for \( 0 < Q < Q_U \)], which is the strongest form of hyperuniformity, \( \gamma_e = 0 \) in the same limit; see Sec. 10.3.5 for details.
10.3.5 Transparency Conditions

Our formulas [Eqs. (10.9) and (10.10)] predict that heterogeneous media can be transparent for elastic waves \([\gamma_{e}^{L,T}(k_{Lq}) = 0]\) if

\[
\text{Im}[F(k_{Lq})] = \text{Im}[F(k_{Tq})] = 0. \tag{10.13}
\]

Physically, these conditions imply that single scattering events of elastic waves at the corresponding frequency are completely suppressed. For stealthy hyperuniform media that satisfy \(\tilde{\chi}_{V}(Q) = 0\) in \(0 < Q < Q_{U}\), the transparency conditions (10.13) are simply given as

\[
0 < k_{Lq} a < \left(\frac{c_{Tq}}{c_{Lq}}\right)Q_{U}a/2,
\]

where \(c_{Tq}/c_{Lq} = \sqrt{(1-2\nu_{1})/[2(1-\nu_{1})]}\), where \(\nu_{1}\) is the Poisson ratio of phase 1 [cf. Eq. (9.64) in Sec. 9.4.1]. For electromagnetic waves, the condition (10.13) is simplified as \(\text{Im}[F(k_{1})] = 0\). Thus, the aforementioned stealthy hyperuniform media are transparent to the electromagnetic waves \((\gamma_{e} = 0)\) for \(0 < k_{1}a < Q_{U}a/2\); see Es. (8.83) in Sec. 8.6. We will make use of these interesting properties in Sec. 10.3.2 and 10.6.

10.4 Models of Dispersions

We investigate four different 3D models of disordered dispersions of identical spheres of radius \(a\) with \(\phi_{2} = 0.25\).

- Overlapping spheres;
- Equilibrium hard spheres (Equilibrium packings);
- Stealthy hyperuniform dispersions; and
- Stealthy nonhyperuniform dispersions.
Figure 10.2: Evaluation of $\tilde{\chi}_V(Q)$ (a) and the attenuation function $F(Q)$ (b,c) for four different models of 3D dispersions: stealthy hyperuniform dispersions, stealthy nonhyperuniform dispersions, overlapping spheres, and equilibrium hard spheres. The inset in (b) is a magnification of the larger panel. Since all of these dispersions are composed of spheres of radius $a$, their microstructures resemble one another at the small length scales $r < a$, indeed all of the spectral densities effectively collapse onto a single curve for $Qa \gg 5$.

These models include two typical disordered ones (overlapping spheres and equilibrium hard spheres) and two exotic disordered ones (stealthy hyperuniform dispersions and stealthy nonhyperuniform dispersions). Detailed descriptions of the first three models are provided in Sec. 8.3.

Stealthy nonhyperuniform dispersions are defined by $\tilde{\chi}_V(Q) = 0$ for $Q_L < Q < Q_U$. We numerically find realizations of these systems [$N = 10^3$ and $\tilde{\chi}_V(Q) = 0$ for $1.0 < Qa < 1.5$] via the collective-coordinate optimization technique [321, 17, 352]; see Sec. 8.3.4. Its spectral density is ascertained from the numerically generated configurations. We tabulate parameters employed to generate the disordered stealthy hyperuniform and disordered stealthy nonhyperuniform dispersions in Table 10.1.

Values of the complex-valued attenuation function $F(Q)$ [see Eqs. (10.7) and
Table 10.1: Parameters for disordered stealthy hyperuniform and disordered stealthy nonhyperuniform dispersions in $\mathbb{R}^3$.

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Stealthy hyperuniform</th>
<th>Stealthy nonhyperuniform</th>
</tr>
</thead>
<tbody>
<tr>
<td>Particle number $N$</td>
<td>1000</td>
<td>1000</td>
</tr>
<tr>
<td>$\rho$</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>The number of configurations</td>
<td>300</td>
<td>300</td>
</tr>
<tr>
<td>$(Q_L a, Q_U a)$</td>
<td>(0, 1.5)</td>
<td>(1.0, 1.5)</td>
</tr>
<tr>
<td>$\sigma$</td>
<td>0.8</td>
<td>0.8</td>
</tr>
<tr>
<td>$a$</td>
<td>0.3908</td>
<td>0.3908</td>
</tr>
</tbody>
</table>

(10.8)] for the four aforementioned models of dispersions are presented in Fig. 10.2. Their imaginary parts are directly obtained from the spectral density based on Eq. (10.7). The associated real parts are then computed from an approximation of Eq. (10.8); see Appendix 8.16. For various types of dispersions, while the values of $\tilde{\chi}_V(Q)$ up to intermediate wavenumbers ($Q a \lesssim 3$) are considerably different from one another, all of the curves approximately collapse onto a single curve for $Q a \gg 5$; see Fig. 10.2(a).

### 10.5 Results

We first derive the microstructure-dependent formulas for the effective dynamic dielectric constant, bulk modulus, and shear modulus that apply from infinite wavelengths down to intermediate wavelengths. Then we use these estimates to establish cross-property relations between them by eliminating a common microstructural parameter among them. Using these formulas, we estimate the effective elastic wave characteristics and cross-property relations for four different 3D models of disordered two-phase dispersions, including two typical nonhyperuniform ones. Finally, we discuss how to employ the newly established cross-property relations in designing multifunctional materials.

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Figure 10.3: Estimates of scaled effective elastic wave characteristics for 3D dispersions of rigid spheres of radius $a$ in a compressible matrix phase with Poisson ratio $\nu_1 = 1/3$ (i.e., $K_2/K_1 = G_2/G_1 = \infty$). Here, $k_{Lq}$ is the wavenumber of longitudinal waves in reference phase (phase 1), and $c_{Lq}$ and $c_{Tq}$ are elastic wave speeds of longitudinal and transverse waves, respectively, in phase 1. (a,c) Effective wave speeds and (b,d) effective attenuation coefficients are plotted in terms of $k_{Lq}a$. The insets in (b) and (d) are magnifications of the larger panels, respectively. In these insets, we see that stealthy hyperuniform dispersions have a zero-$\gamma_{L,S}^e$ regime in $k_{Lq}a < 0.375$, but others do not, which vividly demonstrates that $\gamma_{L,S}^e$ can be engineered by the spatial correlations of composites.

10.5.1 Effective Elastic Wave Characteristics

We now investigate the aforementioned effective elastic wave characteristics of four different models of 3D dispersions, using the strong-contrast approximations [Eqs. (10.9) and (10.10)]. In striking contrast to the other models, stealthy hyperuniform dispersions are transparent for both longitudinal and transverse elastic waves down to a finite wavelength. This result clearly demonstrates that it is possible to design disordered composites that exhibit nontrivial attenuation behaviors by appropriately manipulating their spatial correlations.

We first determine the phase elastic moduli of the aforementioned four models of composites. Since this parameter space of phase moduli is infinite, we consider two extreme cases: a compressible matrix phase (phase 1) with a Poisson ratio $\nu_1 = 1/3$
Figure 10.4: Estimates of scaled effective elastic wave characteristics for 3D dispersions of spherical cavities of radius $a$ in a compressible matrix phase with Poisson ratio $\nu_1 = -0.1$ (i.e., $K_2/K_1 = G_2/G_1 = 0$) by the strong-contrast approximations (10.9) and (10.10). Here, $k_{Lq}$ is the wavenumber of longitudinal waves in phase 1, and $c_{Lq}$ and $c_{Tq}$ are wave speeds of longitudinal and transverse waves, respectively, in phase 1. (a,c) Effective wave speeds and (b,d) effective attenuation coefficients are plotted in terms of $k_{Lq}a$. For stealthy hyperuniform dispersions, $\gamma_{e,L,T} = 0$ when $k_{Lq}a \lesssim 0.55$.

that contains a rigid dispersed phase (phase 2), i.e., $K_2/K_1 = G_2/G_1 = \infty$ (Fig. 10.3) and a compressible matrix with $\nu_1 = -0.1$ that contains cavities, i.e., $K_2/K_1 = G_2/G_1 = 0$ (Fig. 10.4). Investigating these two extreme cases will still provide useful insight into the wave characteristics in intermediate regimes of phase moduli.

While the Poisson ratio of the compressible matrix phase can take any value in the allowable interval of $-1 \leq \nu_1 < 1/2$, we examine two different values of $\nu_1 = 1/3$ (typical of many materials), and $-0.1$. Negative Poisson ratio (“auxetic”) materials laterally dilate (contract) in response to axial elongation (contraction) [212]. While we present the estimated effective elastic moduli up to $k_{Lq}a = 3$, our approximations are, in principle, valid down to the intermediate-wavelength regime ($k_{Lq}a < 1.5$).

We estimate the scaled effective wave propagation properties of the models of 3D dispersions considered in Fig. 10.2. For each of the aforementioned cases of phase properties, four different models have similar effective wave speeds but significantly
different attenuation coefficients. Instead $c^L_e(k_{Lq})$ and $c^T_e(k_{Lq})$ depend largely on the phase properties. For rigid dispersed phase (Fig. 10.3), the effective wave speeds $c^L,T_e$ are generally faster than those in phase 1 but tend to decreases with $k_{Lq}$ at most frequencies. By contrast, when the dispersed phase consists of cavities, the wave speeds are slower than those in phase 1 and increases with $k_{Lq}$ as $k_{Lq} \approx 1$; see Fig. 10.4.

In both cases shown in Figs. 10.3 and 10.4, stealthy hyperuniform dispersions are transparent to both longitudinal and transverse waves in $0 < k_{Lq} a \lesssim 0.4$, as predicted in Eq. (10.13). Such composites can be employed to design of low-pass filters for elastic as well as electromagnetic waves. By contrast, the stealthy nonhyperuniform dispersions do not attain zero attenuation at any finite wavelength because these systems can suppress scatterings at only specific directions.

### 10.5.2 Cross-Property Relations

It is desired to design composites with prescribed elastic and electromagnetic wave characteristics, as schematically illustrated in Fig. 10.1. The rational design of such multifunctional characteristics can be greatly facilitated via the use of cross-property relations for these different effective properties, which we derive here.

We first obtain a cross-property relation between the effective dynamic bulk modulus and effective dynamic dielectric constant from Eqs. (10.4) and (10.9) by eliminating $F(Q)$ between them:

$$
\frac{K_e(k_{Lq})}{K_1} = 1 + \frac{6\beta \kappa (\nu_1 - 1) \phi_2}{\nu_1 + 1} \frac{\epsilon_e(k_{Lq})}{\epsilon_1 - 1} \frac{1}{2\beta + \kappa + (3\beta \kappa \phi_2 - 2\beta - \kappa) \epsilon_e(k_{Lq})/\epsilon_1},
$$

(10.14)

where the effective properties $K_e$ and $\epsilon_e$ must be at the same wavenumber (i.e., $k_{Lq} = k_1$) but possibly at different frequencies, as illustrated in Fig. 10.1. Remarkably, this cross-property relation depends only on the phase properties, regardless of
Figure 10.5: Cross-property relation between the effective dielectric constant $\epsilon_e$ and the effective bulk modulus $K_e$ for the four models of 3D dispersions with $\phi_2 = 0.25$, each of which consists of a compressible matrix with $\nu = 1/3$ and incompressible inclusions (see Fig. 10.3) and the ratio of phase dielectric constants is $\epsilon_2/\epsilon_1 = 10$. Left: surface plots, evaluated from Eq. (10.14), represent the surface on which $\epsilon_e$ and the real (the upper panels) and imaginary (the lower panels) parts of $K_e$ of any composites with the prescribe phase properties must lie. Contour lines (black dotted lines) are at level spacing 0.1. Right: contour plots are the top views of the surface plots on the left panels. We note that the surface plots for other values of the Poisson ratio $\nu_1$ and the contrast ratio $\epsilon_2/\epsilon_1$ are qualitatively similar except for the position of a simple pole; see Figs. 10.7-10.8 in Appendix 10.7. On these surfaces, the locus of points (solid lines with/without markers) represents effective properties of each dispersion model as the wavenumber $k_1$ increases from $k_1a = 0$ to $k_1a = 5$ (in the directions of arrows). Note that all the models attain the Hashin-Shtrikman bounds (blue stars) at $k_1a = 0$.

the microstructures of composites. Intuitively speaking, such cross-property relations can be established because the effective properties depend on the interference pattern of the associated waves, which are commonly determined by wavelengths and microstructures.

The real and imaginary parts of this cross-property relation (10.14) are separately
Table 10.2: Evaluation of the effective shear moduli $G_e/G_1$ for dispersions of spheres of radius $a$ from the strong-contrast approximation (10.10) and the cross-property relation (10.15). Phase moduli are identical to those considered in Fig. 10.3 (i.e., $K_2/K_1 = G_2/G_1 = \infty$ and $\nu_1 = 1/3$), and $\epsilon_2/\epsilon_1 = 10$. Four 3D models are considered: overlapping spheres (OS), equilibrium hard spheres (EHS), stealthy hyperuniform dispersions (SHD), and stealthy nonhyperuniform dispersions (SNHD). Here, $c_{L_q}$ and $c_{T_q}$ are the longitudinal and transverse elastic wave speeds in the reference phase (phase 1), respectively, and $k_{L_q}$ is the wavenumber of longitudinal elastic waves in phase 1.

<table>
<thead>
<tr>
<th>Models</th>
<th>$k_{L_q}$</th>
<th>$\epsilon_e(k_{L_q})/\epsilon_1$</th>
<th>$\epsilon_e(c_{L_q}/c_{T_q})/\epsilon_1$</th>
<th>$G_e(k_{L_q})/G_1$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>From (10.15)</td>
<td>From (10.10)</td>
<td></td>
</tr>
<tr>
<td>OS</td>
<td>0.3</td>
<td>1.724 $+8.239 \times 10^{-3}$</td>
<td>1.808 $+1.455 \times 10^{-2}$</td>
<td>1.650 $-i 2.941 \times 10^{-2}$</td>
</tr>
<tr>
<td></td>
<td>0.5</td>
<td>1.776 $+4.135 \times 10^{-2}$</td>
<td>1.843 $+3.420 \times 10^{-1}$</td>
<td>1.585 $-i 8.904 \times 10^{-2}$</td>
</tr>
<tr>
<td>EHS</td>
<td>0.3</td>
<td>1.708 $+1.723 \times 10^{-3}$</td>
<td>1.758 $+1.637 \times 10^{-2}$</td>
<td>1.676 $-i 8.139 \times 10^{-2}$</td>
</tr>
<tr>
<td></td>
<td>0.5</td>
<td>1.737 $+8.938 \times 10^{-3}$</td>
<td>1.888 $+1.240 \times 10^{-1}$</td>
<td>1.618 $-i 3.614 \times 10^{-2}$</td>
</tr>
<tr>
<td>SHD</td>
<td>0.3</td>
<td>1.704 $+1.510 \times 10^{-2}$</td>
<td>1.743 $+3.477 \times 10^{-1}$</td>
<td>1.683 $-i 1.894 \times 10^{-2}$</td>
</tr>
<tr>
<td></td>
<td>0.5</td>
<td>1.727 $+2.739 \times 10^{-2}$</td>
<td>1.915 $+9.130 \times 10^{-1}$</td>
<td>1.615 $-i 2.469 \times 10^{-2}$</td>
</tr>
<tr>
<td>SNHD</td>
<td>0.3</td>
<td>1.716 $+9.594 \times 10^{-3}$</td>
<td>1.755 $+4.643 \times 10^{-2}$</td>
<td>1.675 $-i 2.302 \times 10^{-2}$</td>
</tr>
<tr>
<td></td>
<td>0.5</td>
<td>1.742 $+3.726 \times 10^{-2}$</td>
<td>1.875 $+1.908 \times 10^{-1}$</td>
<td>1.611 $-i 5.638 \times 10^{-2}$</td>
</tr>
</tbody>
</table>

The surface plots on the left panels in Fig. 10.5 depict the hypersurface on which any possible pairs of $\epsilon_e(k_1)$ and $K_e(k_{L_q} = k_1)$ of a composite must lie when its phase properties and $\phi_2$ are prescribed. The black dotted lines in the upper and lower panels are contour lines of $\text{Re}[K_e/K_1]$ and $\text{Im}[K_e/K_1]$ at level spacing 0.1, respectively. The right panels in Fig. 10.5 represent the top views of the associated surface plots on the left panels. We note that the resulting surface plots have a simple pole at $\epsilon_e = \epsilon_{\text{pole}}$ whose position is determined by phase properties and packing fraction $\phi_2$; see also Figs. 10.7 and 10.8. In Fig. 10.5, the locus of points (shown in solid lines) depicts the effective dielectric constants and bulk moduli of the four different models of 3D dispersions as a dimensionless wavenumber $k_1a (= k_{L_q}a)$ varies from 0 to 5 along with the arrows. Since these points should lie on the surfaces as depicted in Fig. 10.5, one can indirectly determine the wavenumber-dependent $K_e$ by measuring $\epsilon_e$ at different wavenumbers (or frequencies), and vice versa.

Similarly, we can obtain cross-property relations that links $\epsilon_e$ to $G_e$ or $G_e$ to $K_e$. 465
The former case is explicitly given as

$$\frac{G_e(k_{Lq})}{G_1} = 1 - 15(\nu_1 - 1)\phi_2\mu\left((5\nu_1 - 4)\left[\phi_2(1 + 2\mu) - \frac{1 + 2\beta}{\beta}\right]\right) + \frac{3\phi_2}{3c_{Lq}^2 + 2c_{Tq}^2} \left[\frac{3c_{Lq}^2}{\epsilon_e(c_{Lq}k_{Lq}/c_{Tq})/\epsilon_1 - 1} + \frac{2c_{Tq}^2}{\epsilon_e(k_{Lq})/\epsilon_1 - 1}\right]^{-1}, \quad (10.15)$$

which depends on values of the effective dielectric constant $\epsilon_e$ at two different wavenumbers $k_1 = k_{Lq}$ and $c_{Lq}k_{Lq}/c_{Tq}$, making it difficult to graphically depict this cross-property relation. Instead, we list in Table 10.2 values of $G_e$ that are computed from both Eqs. (10.10) and (10.15). Furthermore, by combining Eqs. (10.14) and (10.15), one can also establish cross-property relations that link the effective dielectric constant to the effective elastic wave characteristics, i.e., $c_{e,L,T}^L$ and $\gamma_{L,T}^L$.

10.5.3 Sound-Absorbing and Light-Transparent Materials

To illustrate how our results can be applied for novel multifunctional material design, we engineer composites that are transparent to electromagnetic waves at infrared wavelengths (long wavelengths) but absorb sound at certain frequencies. Importantly, designing such materials is not possible by using standard approximations [153, 270, 198] and quasi-static cross-property relations [290, 99, 306, 296, 304, 208, 191, 325]. Such engineered materials could be used as heat-sinks for central processing units (CPUs) and other electrical devices subject to vibrations or sound-absorbing housings [182]. A similar procedure can be applied to design composites for exterior components of spacecraft [136] and building materials [106]. We will further discuss possible applications in Sec. 10.6.

We take advantage of the fact that stealthy hyperuniform dispersions are transparent down to a finite wavelength ($k_1 \leq Q_U/2$); see Fig. 10.6. We then find polarizabilities $\beta$ and $\kappa$ that result in high attenuation coefficient $\gamma_e^L$ at $k_{Lq} a = k' a \equiv 1.3$. 

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Figure 10.6: Multifunctional design of materials that are transparent at infrared wavelengths but absorb sound at certain acoustic frequencies. In order to attain such materials, we exploit exotic 3D stealthy hyperuniform dispersions. (a) Contour plot of the distance between the real part of the effective dielectric constant at a target frequency (the associated wavenumber is $k'a = 1.3$) and the simple pole $\epsilon_{\text{pole}}$ as a function of the dielectric $\beta$ and bulk modulus $\kappa$ polarizabilities. We choose $\beta = 0.17$ and $\kappa = -12$ (shown in a blue disk). (b) The cross-property relation between $\epsilon_e/\epsilon_1$ and $\text{Im}[K_e/K_1]$ for the chosen parameters $\beta = 0.17$ and $\kappa = -12$. The effective static dielectric constant is shown as a blue star. The effective dielectric constant at the target frequency and the pole are shown in the black and red circles, respectively. The associated composite consists of a nearly incompressible matrix phase with $\nu_1 = 23/49$ that containing cavities ($K_2 = G_2 = 0$), and its contrast ratio of the dielectric constants is $\epsilon_2/\epsilon_1 \approx 1.63$. (c) The imaginary part of the resulting effective dynamic dielectric constant as a function of the wavenumber $k_1a$. (d) The resulting scaled attenuation coefficient for the longitudinal elastic waves as a function of the longitudinal wavenumber $k_{Lq}$.

This is achieved when $\epsilon_e(k')$ is close to the simple pole $\epsilon_{\text{pole}}$ of Eq. (10.14); see Fig. 10.6(a). Figure 10.6(b) shows the cross-property relation (10.14) with the chosen polarizabilities, i.e., $\beta = 0.17$ and $\kappa = -12$. Phase properties corresponding to these polarizabilities are degenerate, and we choose $\epsilon_2/\epsilon_1 \approx 1.63$, $\nu_1 = 23/49$, and $K_2 = G_2 = 0$. We see from Fig. 10.6(c)-(d) that the resulting materials are indeed
transparent to electromagnetic waves at long wavelengths but exhibit resonance-like attenuation of sound at \( k_{Lq} a \approx 1.0 \).

### 10.6 Conclusions and Discussion

We have obtained accurate approximations for the effective dynamic dielectric constant \( \epsilon_e \) and the effective dynamic elastic moduli \( K_e \) and \( G_e \) of two-phase composites that depend on the microstructure via the spectral density \( \tilde{\chi}_V(Q) \), which is easily computed theoretically/computationally or accessible via scattering experiments; see Eqs. (10.4), (10.9), and (10.10). These formulas are superior in predicting these effective dynamic properties compared to commonly used approximations, such as Maxwell-Garnett and quasicrystalline approximations [153, 270, 198], as verified by computer simulations in Chapters 8 and 9. Unlike these conventional approximations, our formulas are accurate for a wide range of incident wavelengths for a broad class of dispersions.

Using the approximations (10.4), (10.9), and (10.10), we have shown that hyperuniform composites can have desirable attenuation properties both for electromagnetic and elastic waves. We analytically showed that hyperuniform media are less dissipative than nonhyperuniform ones in Sec. 10.3.3. Remarkably, stealthy hyperuniform media are dissipationless (i.e., \( \gamma_e = 0 \)) down to a finite wavelength, as shown in Fig. 10.3. Such composites can be employed to low-pass filters for elastic and electromagnetic waves.

Using Eqs. (10.4), (10.9), and (10.10), we also established cross-property relations (10.14) and (10.15) that link the effective dynamic dielectric constant \( \epsilon_e(k_1) \) to the effective dynamic bulk modulus \( K_e(k_{Lq}) \) and shear modulus \( G_e(k_{Lq}) \), respectively. Thus, when it is difficult to directly measure \( K_e \) or \( G_e \), they can be indirectly evaluated from these cross-property relations by measuring the wavenumber-dependent
dielectric constants [280, 83], as demonstrated in Fig. 10.5 and Table 10.2, and vice versa. For example, one can use them to indirectly determine physical/chemical properties for construction materials [280, 83] and oil-exploration [39].

Our cross-property relations also have important practical implications for the rational design of multifunctional composites [208, 296, 306, 191, 325, 304, 182] that have the desired dielectric properties for a particular range of electromagnetic wavelengths and elastic properties for a certain range of elastodynamic wavelengths. The validation of our formulas via computer simulations justifies their use for the design of novel multifunctional materials without having to perform full-blown simulations. In particular, we described how to engineer a sound-absorbing composite that is transparent to light via our cross-property relations, which again could not be done using previous approximation formulas [153, 270, 198, 290, 99, 306, 296, 304, 208, 191, 325]. This is done by exploiting the exotic structural properties of stealthy hyperuniform dispersions; see Fig. 10.6. Such engineered materials could be used as heat-sinks for CPUs and other electrical devices subject to vibrations because they enable radiative cooling while suppressing prescribed mechanical vibrations. Another application is a sound-absorbing housing for an engine or a motor, which can efficiently convert cyclic noise into electric energy [182] and allow radiative cooling. It is natural to extend to the aerospace industry where low-frequency engine noise is prevalent [182]. A similar procedure can be applied to design composites with high stiffness that absorb electromagnetic waves at certain wavenumbers for use as exterior components of spacecraft [136] and building materials [106].

With the aid of our microstructure-dependent formulas [Eqs. (10.4), (10.9), and (10.10)], one can employ inverse-design approaches [297] to design composites. We recall that inverse-design approaches enable one to prescribe the effective properties of composites and then find the microstructures that achieve them. For example, one would first prescribe the material phases and then compute the desired effective prop-
erties (say, attenuation coefficients $\gamma_e$ for a given bandwidth) via the microstructure-dependent formulas. Then, one backs out the corresponding spectral density from the attenuation function, which would correspond to a particular microstructure, if realizable. The latter can be constructed by using previously established Fourier-space construction techniques [321, 17, 352, 46]. Finally, one can generate simulated microstructures via modern fabrication methods, such as 3D printing [334] or 2D photolithographic technologies [360]. The same inverse techniques also can be employed to design multifunctional composites using cross-property relations. Remarkably, such inverse approaches were not possible with previously known approximations, such as Maxwell-Garnett and quasicrystalline formulas [153, 270, 198] because they are independent of microstructures.

It is instructive to briefly discuss how to measure the wavenumber-dependent effective properties in experiments. Here, for brevity, we focus on the dielectric constants because the same reasoning applies to the elastodynamic case (Eqs. (10.9) and (10.10)). Clearly, the property $\epsilon_e(k_1)$ is identical to the frequency-dependent one because a wavenumber $k_1$ in the reference phase can be converted to a frequency $\omega$ via the dispersion relation of the reference phase [i.e., $\omega = \omega(k_1)$]. The frequency-dependent effective dielectric constant $\epsilon_e(\omega)$ can be measured via conventional techniques, such as perturbation methods (measuring changes in a resonance frequency of a resonator due to a specimen) or transmission techniques (measuring the transmission/reflection by a specimen); see Ref. [285] and references therein. However, when using cross-property relations, it is crucial to covert the independent variable of the effective properties from frequency $\omega$ to the associated wavenumbers ($k_1$ and $k_{Lq}$) according to the dispersion relations of the reference phase.

While we primarily focused on three-dimensional two-phase media, our microstructure-dependent formulas (Eqs. (10.4), (10.9), and (10.10)) are valid for $d \geq 2$. Furthermore, the cross-property relations (Eqs. (10.14) and (10.15)) can be extended to any
dimension $d$ with minor modifications.

Based on a previous study on the static case [100], it is relatively straightforward to generalize our microstructure-dependent formulas to composites whose dispersed phase is a piezoelectric (i.e., mechanical stress can induce an electric voltage in the solid material). Such extensions can be profitably utilized in the optimal design of materials for elastic wave energy-harvesting to power small electrical devices [344].

10.7 Appendix A: Other Plots of the Cross-Property Relation Between $\epsilon_e$ and $K_e$

We present plots depicting cross-property relations for some other phase properties beyond the ones described in Fig. 10.3; see Figs. 10.7 and 10.8. We note that contours vary with phase properties $\epsilon_p, K_p, G_p$ for $p = 1, 2$ regardless of microstructures.
Figure 10.7: Cross-property relations between the effective dielectric constant $\epsilon_e$ and the effective bulk modulus $K_e$ in the case of a compressible matrix with $\nu_1 = 1/3$ containing an incompressible dispersed phase. The ratios of phase dielectric constants are (a) $\epsilon_2/\epsilon_1 = 10$ and (b) $\epsilon_2/\epsilon_1 = 0.25$. Contour plots, evaluated from Eq. (14) in the main text, represent the surface on which $\epsilon_e$ and the real (the upper panel) and the imaginary (the lower panel) parts of $K_e$ at a prescribed wavenumber $k_{Lq}$ should lie. We then overlap cross-property relations for four models of 3D disordered dispersions with $\phi_2 = 0.25$: stealthy hyperuniform (SHU), stealthy nonhyperuniform (SNHU), overlapping spheres (OVS), and equilibrium hard spheres (EHS). These curves start from the Hashin-Shtrikman bounds (blue stars) at $k_{Lq} a = 0$ to $k_{Lq} a = 5$. 
Figure 10.8: Cross-property relations between the effective dielectric constant $\epsilon_e$ and the effective bulk modulus $K_e$ in the case of a compressible matrix with $\nu_1 = -0.1$ containing cavities. The ratios of phase dielectric constants are (a) $\epsilon_2/\epsilon_1 = 10$ and (b) $\epsilon_2/\epsilon_1 = 0.25$. The phase properties considered in (a) are identical to those in Fig. 4 in the main text. Contour plots, evaluated from Eq. (14) in the main text, represent the surface on which $\epsilon_e$ and the real (the upper panel) and the imaginary (the lower panel) parts of $K_e$ at a prescribed wavenumber $k_L$ should lie. We then overlap cross-property relations for four models of 3D disordered dispersions with $\phi_2 = 0.25$: stealthy hyperuniform (SHU), stealthy nonhyperuniform (SNHU), overlapping spheres (OVS), and equilibrium hard spheres (EHS). These curves start from the Hashin-Shtrikman bounds (blue stars) at $k_L a = 0$ to $k_L a = 5$. 
Bibliography


