Precision measurements of spin interactions with high density atomic vapors

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Abstract

Polarized atomic vapors can be used to detect fields interacting with a spin. Recent advances have extended the sensitivity of atomic magnetometers to a level favorable for fundamental physics research, and in many cases the sensitivity approaches quantum metrology limits.

In this thesis, we present a high density atomic K-3He comagnetometer, which features suppressed sensitivity to magnetic fields, but retains sensitivity to anomalous fields that couple differently than a magnetic field to electron and nuclear spins. The comagnetometer was used to measure interactions with a separate optically pumped 3He nuclear spin source. The 3He spin precession frequency in the comagnetometer was measured with a resolution of 18 pHz over the course of approximately one month, enabling us to constrain the anomalous spin-spin interaction between neutrons to be less than $2.5 \times 10^{-8}$ of their magnetic or less than $2 \times 10^{-3}$ of their gravitational interaction at a length scale of 50 cm. We set new laboratory bounds on the coupling strength of light pseudoscalar, vector and pseudovector particles to neutrons, and we consider the implications of our measurement to recently proposed models for unparticles and Goldstone bosons from spontaneous breaking of Lorentz symmetry.

We also describe theoretically and experimentally a quantum non-demolition (QND) measurement of atomic spin in the context of radio frequency magnetometer with hot alkali-metal vapors. Using stroboscopic probe light we demonstrate suppression of the probe back-action on the measured observable, which depends on the probe duty cycle and on the detuning of the probe modulation frequency from twice the alkali Larmor frequency. We study the dependence of spin-projection noise on the polarization for atoms with spin greater than 1/2 and develop a theoretical model that agrees well with the data. Finally, it is shown theoretically that QND measurements can improve the long-term sensitivity of atomic magnetometers with non-linear relaxation.
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To my parents
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Chapter 1

Introduction

The detection of fields that couple to the spins of particles has been of great interest for both practical and fundamental physics purposes. Measurement of magnetic fields have found applications in many areas, including navigation, geology, astrophysics, materials science, biomedicine and contraband detection. Although magnetic forces are the only experimentally observed macroscopic interactions between spins, the detection of new spin-spin forces would have a strong impact on our understanding of the physical world.

A variety of sensors have been developed to measure magnetic fields such as the Hall probe, fluxgate, inductive coil, and superconducting quantum interface device (SQUID). It has been half a century since optical pumping and probing techniques have been applied to atomic vapors for the detection of magnetic fields \cite{1}. The basic principle of atomic optical magnetometry is illustrated in Fig. 1.1: light resonant to an optical transition creates orientation and/or higher order atomic polarization moments; the spin ensemble perform Larmor precession in the presence of a magnetic field and the spin dynamics are monitored with a weak probe beam, whose optical properties (polarization or transmission) are modified by the spins (see \cite{36, 39} for recent reviews). The highest sensitivity has been realized with a linearly polarized

\footnote{The compass was probably the first magnetic field sensor that was used.}
probe beam that experiences Faraday paramagnetic rotation from the interaction with oriented atomic vapor.

Figure 1.1: (adapted from [169]) Schematic illustrating the basic principle of atomic optical magnetometry: a pump beam polarizes the atomic vapor and a probe beam monitors the spin dynamics.

Recent technological advancements, mainly the availability of robust, high power, easily tunable diode lasers, and the development of techniques for low spin relaxation rates, have enabled atomic optical magnetometers to surpass SQUIDs as the most sensitive magnetic sensors. Sensitivities below 1 fT/√Hz have been experimentally demonstrated with atomic magnetometers [114, 52, 128].

Their high sensitivity make atomic vapors particularly attractive as probes of new non-magnetic spin-dependent interactions. There are strong theoretical motivations to search for long-range fields that couple with fermion spins in the low energy limit. In addition to the original motivation for spin-dependent forces mediated by axions [112], a number of new theoretical ideas have been introduced recently, including para-photons [60], “unparticle” [129] and theories with spontaneous Lorentz violation [12].

Astrophysics and cosmology provide a valuable test bed for the applicability of these theories to the physical world [155] and many strong bounds on new spin-
dependent interactions have been placed based on astrophysical considerations. However, these bounds rely on model assumptions and recently there have been proposals to relax these constraints \([135, 61]\). More direct bounds come from laboratory experiments, where the conditions are well characterized and controlled, so that the results are more robust to interpretations (see \([108]\) for a review).

Overlapping spin-ensembles in a comagnetometer arrangement provide a well-suited platform to explore physics beyond the standard model. The two spin species of the comagnetometer system effectively allow for the control and cancelation of the magnetic field noise which could potentially mask the new spin coupling field to be measured. High density K-3He comagnetometers have been used as sensitive probes of Lorentz and CPT violating fields; these systems have demonstrated very high sensitivity on anomalous spin couplings, but suppressed response to magnetic fields \([116, 33]\).

As experimental limitations are overcome atomic magnetometers start to approach quantum limits, and in many cases of small-sized systems the quantum nature of measurement becomes apparent. In general, there are three fundamental sources of noise that could potentially limit the estimation of the magnetic field from measurements with an atomic optical magnetometer \([163]\): the spin projection noise which comes from the fact that if an atom is polarized in a particular direction, a measurement of the angular-momentum component in an orthogonal direction gives a random outcome; the photon shot noise of probe detection which originates from the random arrival of photons at the detector; and the a.c. Stark shift (light-shift) caused from the quantum polarization fluctuations of the probe beam, which effectively generates a noisy magnetic field in the direction of light propagation.

The light-shift noise is a manifestation of the fact that every physical measurement is necessarily invasive. The probe that is used to extract information from a quantum system introduces a Hamiltonian term that does not commute with all the
observables, thus disturbing the state of the system. Quantum non demolition (QND) measurements can be performed in such a way that the probe does not affect the evolution of the specific observable under investigation. A QND measurement eliminates the back-action of the probe on the detected observable, and drives the system to a squeezed state, where the uncertainty of the variable being monitored is reduced below the standard projection limit at the expense of an increase in the uncertainty of the conjugate variable. Spin squeezed states have attracted a great deal of interest in quantum metrology and quantum information processing [94].

Quantum non demolition measurements have been performed in atomic magnetometers with various measuring schemes (see [110, 39]). Although QND measurements have been shown to increase the bandwidth of atomic magnetometers without loss of sensitivity [170], it has been argued that spin squeezing in the presence of constant decoherence rate does not improve significantly on the long term sensitivity [13]; a substantial improvement in sensitivity seems to be possible only at timescales shorter than the spin-relaxation rate.

In this thesis we discuss precision measurements with atomic vapors. Chapter 2 is an introduction to optical pumping and probing, describing the interaction of light with atoms and physical processes that lead to spin relaxation. Chapter 3 presents a theoretical characterization of the K-$^3$He comagnetometer, based on a simple model where the dynamics are described with two coupled Bloch equations. In Chapter 4 the implementation of the K-$^3$He comagnetometer and the $^3$He spin source that creates the field to be measured by the comagnetometer are detailed. The results of the search for spin-spin interactions are presented in Chapter 5. Finally, Chapter 6 discusses a new quantum non demolition scheme in RF magnetometry that evades back-action; it presents systematic studies of spin noise, and explores theoretically the possibility of increasing the long-term sensitivity of atomic magnetometers using
QND measurements in a scenario of time varying spin relaxation due to spin-exchange collisions.
Chapter 2

Theoretical Background

In this chapter, we discuss the general features of an optical-atomic magnetometer. We begin with the energy structure of the alkali atoms and describe the atom-light interaction using a semiclassical picture. The pumping mechanism that creates polarization and the optical detection of spin motion through the Faraday paramagnetic effect are reviewed. We then consider the various mechanisms that relax the ground coherences and present the evolution of the atomic ground-electronic state that is relevant to the optical-atomic magnetometer. The simplified picture of Bloch equations is discussed and we briefly present quantum optical states and operators that are useful in describing the atom-light quantum interface.

2.1 Atomic Energy levels

Atomic magnetometers use alkali atoms in their ground electronic state for probing the magnetic field. To a high degree of accuracy alkali atoms can be described as having one unpaired electron. The ground electronic state has no orbital angular momentum, whereas the two first excited electronic states correspond to unit electronic orbital angular momentum (P shell). The P shell is split due to the fine interaction $H_{fn} \propto \mathbf{L} \cdot \mathbf{S}$ into two states with different total electronic angular momentum $\mathbf{J} = \mathbf{L} + \mathbf{S}$. The
state with $J = 1/2$ has lower energy than the state with $J = 3/2$. Since all naturally occurring alkali atoms have non-zero nuclear spins, they exhibit hyperfine structure. The hyperfine interaction is described by the Hamiltonian term $H = A_{hf} \mathbf{I} \cdot \mathbf{J}$ where $A_{hf}$ is the hyperfine interaction, $\mathbf{I}$ and $\mathbf{J}$ are the nuclear and electron spin respectively. The eigenstates of the Hamiltonian are $|F, m_F\rangle$, with $F = I + J, I + J - 1, \ldots |I - J|$ being the quantum number of the total atomic angular momentum $\mathbf{F} = \mathbf{I} + \mathbf{J}$ and $m_F$ is the projection of the total angular momentum in the quantization axis (taken typically to be the $z$ axis) $F_z = I_z + J_z$. Due to the hyperfine interaction the ground (and the first excited) electronic state is split into two hyperfine manifolds with $f = I + 1/2$ and $f = I - 1/2$ (the second excited state is split into 4 manifolds). Figure 2.1 shows the energy level diagram of an alkali atom with nuclear spin $I = 3/2$ (e.g. $^{39}$K).

![Energy level diagram of an alkali atom with nuclear spin $I = 3/2$.](image)

Figure 2.1: (taken from [169]) Energy structure of an alkali atom with nuclear spin $I = 3/2$. The effects of fine and hyperfine interaction are added in steps. The diagram is not drawn in scale.

### 2.2 The density matrix

A quantum mechanical system is described in its most general form with the density matrix $\rho$ [[63] [30]]. The density matrix is a positive ($\rho \geq 0$), normalized $\text{Tr}[\rho] = 1$, Hermitian operator ($\rho = \rho^\dagger$) and has all the information necessary to calculate the statistical properties of any observable $A$ through the relationship $\langle A \rangle = \text{Tr}[\rho A]$. 

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There are two ways of describing the time evolution of the statistical properties of the system. In one picture, called the Schrödinger picture, the observable operators do not have time dependence $A = A(0)$, and the dynamics of the system are described by the time evolution of the density matrix $\rho = \rho(t)$. Within this framework, the evolution of the density matrix is described by the equation:

$$\frac{d\rho}{dt} = \frac{1}{i\hbar}[H, \rho]$$

(2.1)

where $H$ is the Hamiltonian of the system. In the Heisenberg picture, it is the observables that change in time ($A = A(t)$) while the state remains fixed $\rho = \rho(0)$. In this case, the operator $A$ changes in time according to the equation:

$$\frac{dA}{dt} = -\frac{1}{i\hbar}[H, A]$$

(2.2)

Notice the minus sign in the commutator compared to (2.1). The Heisenberg and Schrödinger pictures are completely equivalent; we will find convenient in this chapter to work in the Schrödinger picture, but later when we will be discussing spin noise we will use the Heisenberg picture. Equation (2.1) describes the dynamics of a closed (i.e. isolated) system without considering the effect of the measurement. For this, we need to include an additional postulate (called the "projection postulate"). For concreteness, let us assume that we measure an observable $A$ that has a discrete spectrum with eigenvalues $a$ and let $P_a$ describe the operator that projects onto the subspace that is spanned by the eigenvectors of $A$ with eigenvalues $a$. Then the density matrix after the measurement of $A$ will be transformed according to:

$$\rho \mapsto \rho' = \frac{P_a \rho P_a}{\text{Tr}[\rho P_a]}$$

(2.3)
This conditional evolution of the density matrix plays a key role in quantum filtering and conditional squeezing and is going to be discussed in more detail in chapter 6.

- **Parts of density matrix with and without electron polarization.** For the description of various relaxation mechanisms, it will be convenient to separate the density matrix of alkali atoms in the ground state into two parts: one with electron polarization and one without. As explained in [9] the ground $\rho$ can be written as:

$$\rho = \phi + \Theta \cdot S$$  \hspace{1cm} (2.4)

where the part without electron polarization is:

$$\phi = \frac{1}{4} \rho + S \cdot \rho S$$ \hspace{1cm} (2.5)

and the part with electron polarization is:

$$\Theta \cdot S = \frac{3}{4} \rho - S \cdot \rho S$$ \hspace{1cm} (2.6)

where $\Theta = (\Theta_x, \Theta_y, \Theta_z)$ is an operator acting only on the nuclear degrees of freedom and $S$ is the electron spin operator. It is easy to show that $\text{Tr}[\Theta \cdot S] = \langle S \rangle$ and $\text{Tr}[\phi S] = 0$. For example:

$$\text{Tr} [\phi S_j] = \frac{1}{4} \text{Tr} [\rho S_j] + \text{Tr} [S_i \rho S_i S_j]$$

$$= \frac{1}{4} \langle S_j \rangle + \text{Tr} [S_i S_j S_i \rho] = \frac{1}{4} \langle S_j \rangle - \text{Tr} [\frac{1}{4} S_j \rho] = 0$$ \hspace{1cm} (2.7)

We use the convention that repeated indices imply a summation over these indices. In passing to the second line of (2.7) we used the trace property $\text{Tr}[AB] = \text{Tr}[BA]$. 

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and the spin identity \[ S_j S_k S_l = \frac{i}{8} \varepsilon_{jkl} I + \frac{1}{4}(S_j \delta_{kl} - S_k \delta_{jl} + S_l \delta_{jk}) \] \[ (2.8) \]

2.3 Evolution in the magnetic field

In the presence of a magnetic field \( \mathbf{B} \) along the \( z \) direction the ground state Hamiltonian is:

\[ H = A_{hf} I \cdot \mathbf{S} + g_e \mu_B S_z \cdot B_z - \frac{\mu_I}{I} I_z \cdot B_z \] \[ (2.9) \]

\( A_{hf} \) is the hyperfine splitting constant, \( g_e = 2.00232 \) is the electron gyromagnetic ratio, \( \mu_B = 9.2741 \times 10^{-21} \text{ erg G}^{-1} \) is the Bohr magneton and \( \mu_I \) is the nuclear magnetic moment. For magnetic fields less than a few hundred Gauss the magnetic interaction can be treated as a perturbation to the hyperfine interaction. Solving the perturbation equation to second order in \( B_z \) we find that for transitions with \( \Delta F = 0 \) (Zeeman transitions) and \( \Delta m_F = m_F - (m_F - 1) = 1 \) the resonance frequencies are \[ \omega_a = \frac{g_e \mu_B B_z}{\hbar [I]} - \frac{4(g_e \mu_B B_z)^2(m_F - 1/2)}{[I]^3 \hbar A_{hf}} \] \[ (2.10) \]

\[ \omega_b = -\frac{g_e \mu_B B_z}{\hbar [I]} + \frac{4(g_e \mu_B B_z)^2(m_F - 1/2)}{[I]^3 \hbar A_{hf}} \] \[ (2.11) \]

The first and second equation describe the resonant frequency for transitions in the hyperfine manifold with \( F = a = I + 1/2 \) and \( F = b = I - 1/2 \) respectively. Here and later we use the square bracket notation \([I] = 2I + 1\) for angular momentum quantum numbers. Note the difference in signs for the two hyperfine manifolds in (2.10) and (2.11). This means that alkali atoms at different hyperfine manifolds rotate in opposite directions in a static magnetic field. This turns out to be important in
the understanding of the relaxation caused by spin-exchange collisions between alkali atoms.

2.4 Interaction of alkali atoms with light

There are many excellent articles and books that describe the interaction of light with atoms [97, 96, 49, 180, 123, 98]. Here, we are only giving a brief description of the interaction for conditions relevant to the experiments of the thesis. For the optical wavelengths we can perform the dipole approximation [167] and write the Hamiltonian interaction term:

$$H = -\mathbf{D} \cdot \mathbf{E}$$ (2.12)

Here $\mathbf{D}$ is the dipole operator acting only on the electronic degrees of freedom:

$$\mathbf{D} = d + d^\dagger$$ (2.13)

$$d = \sum_{mS} |S_mS\rangle \langle S_mS| J_e |m_m_e\rangle \langle J_e m_m_e|$$ (2.14)

As can be seen from (2.14) $d$ can be thought of as an operator annihilating an atomic excitation (similarly $d^\dagger$ is an operator creating an atomic excitation). The electric field is given by (we dropped the spatial dependence) [123]:

$$\mathbf{E} = \sum_{k\sigma} (\mathcal{E}_{0k}^* \mathbf{e}_{k\sigma} \hat{a}_{k\sigma}^\dagger e^{i\omega_k t} + \mathcal{E}_{0k} \mathbf{e}_{k\sigma} \hat{a}_{k\sigma} e^{-i\omega_k t})$$ (2.15)

$$= \mathbf{E}'(-) + \mathbf{E}'(+)$$ (2.16)

where $\mathcal{E}_{0k}$ is the amplitude of mode $k$, $\sigma$ represents the polarization of the optical mode ($\pm$ in the circular basis), $\mathbf{e}_{k\sigma}$ is the polarization vector, $\omega$ is the angular frequency of

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\textsuperscript{1}Here, we wrote the interaction between light and the group of atoms that have velocity $\mathbf{v} = 0$ in the laboratory frame. For the general case of arbitrary $\mathbf{v}$, $\omega_k$ should be replaced by $\omega_k - \mathbf{k}\mathbf{v}$. 

the light, and $\hat{a}_{k\sigma}$, $\hat{a}_{k\sigma}^\dagger$ are respectively the annihilation and creation operator of the photon in mode $k$ and polarization $\sigma$. In the same manner, $\mathbf{E}^{(-)}$ and $\mathbf{E}^{(+)}$ denote the creation and annihilation operator of the electric field. From the definition it can be seen that $\mathbf{E}^{(-)} = (\mathbf{E}^{(+)} )^\dagger$. In (2.15) we used the quantized formulation of the electric field. Similarly, we can write a classical electric field (but without the creation and annihilation operators).

### 2.4.1 Polarizability Hamiltonian

The general equation for the evolution of the density matrix equation is complicated (even within the dipole approximation) and involves the time evolution of the atomic state in a very large Hilbert space. For transitions from the ground state of alkali atoms to a state of electronic spin $J_e$ the dimension is $[I]( [S] + [J_e] )$. Therefore for the D1 transition of $^{39}$K we have to solve the density matrix in a Hilbert space of dimension $2^{16}$. However, in this work we are interested in the evolution of the ground state (subspace of dimension $[I][S]$) and more specifically in the “quasi-steady state” \cite{97}, where we are concerned with timescales larger than the radiative lifetime of the atoms (typically on the order of a few nsec). Under the simplifying assumption of small excited state population, we can perform the adiabatic elimination \cite{75, 76, 180, 94} of the excited state and we can write an effective Hamiltonian term that determines the evolution of the ground state density matrix due to the interaction with monochromatic light \cite{98}:

\begin{equation}
\delta H = -\mathbf{E}^{(-)} \cdot \mathbf{\alpha} \cdot \mathbf{E}^{(+)} \tag{2.18}
\end{equation}

\begin{equation}
\frac{\text{i} \hbar}{\text{d}t} \rho = \delta H \rho - \rho \delta H^\dagger \tag{2.17}
\end{equation}

\footnote{This means that we need to work with $16 \times 16$ matrices}
\( \alpha \) is the atomic polarizability and is defined as:

\[
\alpha = \sum_{F_g F_e F_g'} \hat{P}_g \frac{d \hat{P}_e}{d^3 \hat{P}_e} \left( \sqrt{\frac{M_a}{2k_B T}} Z[x(F_e m_{F_e}; F_g' m_{F_g'}) + iy] \right) \tag{2.19}
\]

Here the operator \( P_f \) represents the projection operator in the manifold with atomic quantum number \( F \):

\[
\hat{P}_F = \sum_{m_F} |F, m_F \rangle \langle F, m_F |	ag{2.20}
\]

\( Z \) is the plasma dispersion function \( [73]\) defined by:

\[
Z(\zeta) = \frac{1}{\sqrt{\pi}} \int_{-\infty}^{\infty} du \frac{e^{-u^2}}{u - \zeta} \tag{2.21}
\]

and appears in (2.19) as a result of the average over atomic velocities following a Maxwellian distribution. The plasma-dispersion function (called in this sense as the profile function) is the most general way to describe the effects of Doppler, collisional and natural broadening.\(^3\) In Eq. (2.19) \( M_a \) is the mass of the atom, \( k_B \) is the Boltzmann constant, \( T \) is the absolute temperature, \( k \) is the wavevector of the mode of light interacting with the atoms and the arguments in the plasma-dispersion function are:

\[
x(F_e m_{F_e}; F_g' m_{F_g'}) = \frac{1}{k} \left( \frac{M_a}{2k_B T} \right)^{1/2} (\omega - \omega(F_e m_{F_e}; F_g' m_{F_g'})) \tag{2.22}
\]

\[
y = \frac{1}{k} \left( \frac{M_a}{2k_B T} \right)^{1/2} \Gamma \tag{2.23}
\]

Here \( \omega(F_e m_{F_e}; F_g' m_{F_g'}) \) is the difference in energies between the excited and ground state coupled through the light and \( \Gamma \) is the characteristic relaxation rate for optical coherences.\(^4\) If \( \Gamma_c \) is the collision rate and \( \Gamma_{sp} \) is the spontaneous emission rate

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\(^3\)This is known in literature as the Voigt profile.

\(^4\)This is not to be confused with the transverse relaxation rate \( 1/T_2 \) of the spin in the ground electronic state defined later.
then $\Gamma = \gamma_{sp}/2 + \gamma_c$. We should note that in writing (2.23) we assumed $\Gamma$ to be real. During collisions the electronic wavefunction of the atoms is deformed and that creates a collisional shift in the atomic energies. Here, we have accounted for these shifts by modifying appropriately the atomic frequencies $\omega_{fg}, \omega_{fe}$ and we keep $\Gamma \in \mathbb{R}$.

Before we continue to discuss the optical pumping, we summarize the approximations that led to the polarizability Hamiltonian (equation (2.17)-(2.19)). First, we note that equations (2.17) and (2.18) only contain the coherent scattering of photons and do not include the effect of spontaneous emission or buffer gas quenching that populate the ground state. These effects will be treated separately later. We also assumed negligible excited state population and we ignored the stimulated emission.

The condition for small excitation that led to the adiabatic approximation can be put more rigorously by the condition for the saturation parameter \( s_{\text{sat}} \):

\[
s_{\text{sat}} = \frac{\Omega^2}{2(\Delta^2 + \Gamma^2/4)} \ll 1 \tag{2.24}
\]

where $\Omega$ is the Rabi frequency of the transition defined as:

\[
\Omega = \frac{2\mathcal{E}_0 \langle S = 1/2 \mid d \mid J_e \rangle}{\hbar} \tag{2.25}
\]

To derive Eq. (2.18) we made the Rotating Wave Approximation, which consists of dropping out terms that oscillate with frequencies on the order of $\omega + \omega_{fe}/f_g$. This is the mathematical reason for keeping only the parts $E(\cdot) d$ and the part $d^\dagger E(\cdot)$ in the effective Hamiltonian.

Finally, for not too large magnetic fields (up to a few hundred Gauss) the Zeeman splitting of the hyperfine states is much smaller than the Doppler width of optical absorption and therefore we can ignore the dependence of the profile function $Z$ on the magnetic quantum number $m_f$ so from now on we are going to write $Z(F_e F_g)$. 


2.4.2 Light Shift and Absorption Operator

It can easily be seen that the effective Hamiltonian $\delta H$ in Equations (2.18) is not Hermitian. This is because it includes absorption of photons which results in disappearance of atoms from the ground state. We can decompose $\delta H$ into a Hermitian light-shift part:

$$\delta E = \frac{1}{2}(\delta H + \delta H^\dagger)$$ \hspace{1cm} (2.26)

and a Hermitian light-absorption part:

$$\delta \Gamma = \frac{i}{\hbar}(\delta H - \delta H^\dagger)$$ \hspace{1cm} (2.27)

so that:

$$\delta H = \delta E - i\hbar \delta \Gamma / 2$$ \hspace{1cm} (2.28)

The rate of disappearance from the ground state and equivalently the absorption rate of photons is given by:

$$-\text{Tr}\left[\frac{d\rho}{dt}\right] = \text{Tr}[\rho \delta \Gamma] = \langle \delta \Gamma \rangle$$ \hspace{1cm} (2.29)

2.4.3 Polarizability tensor

The polarizability (see Eq. (2.19)) is a dyad involving vector operators. As such, the polarizability in the irreducible representation can be decomposed into a scalar (isotropic), a vector (orientation$^5$) and a rank two (alignment) spherical tensor $\{98\}$:

$$\alpha = \sum_{L=0}^{2} \alpha^L$$ \hspace{1cm} (2.30)

$$= \sum_{F_gF'_gF_eLM} A^L (F_gF'_g)(-1)^M Q^L_M T^L_M (F_gF'_g)$$ \hspace{1cm} (2.31)

$^5$Here, we are adopting the convention of $[37]$ to name a rank 1 tensor as an orientation tensor and a rank 2 tensor as an alignment tensor.
The definition of the various operators in the above equation can be found in Appendix A. For an analytical discussion of the polarizability tensor the interested reader is referred to [98]. Here, we are only going to focus on certain limiting cases that are relevant to the experiments of the thesis. One important simplification comes when the profile function $Z(F_eF_g)$ has a very small dependence on the excited state quantum number $F_e$, so that for all the excited states the same profile factor applies:

$$Z(F_eF_g) \approx Z(F_g).$$

This is the case when the hyperfine separations of the excited state are small compared to the Doppler or collisional broadening or to the detuning. Then the various components of the polarizability take the form [98, 180]:

$$\alpha^0 = G \sum_{F_g} \hat{P}_{F_g} Z(F_g)$$

$$\alpha^1 = \frac{G}{2\sqrt{2}} [11 - 4J_e(J_e + 1)] \sum_M (-1)^M Q^1_M$$

$$\times \left[ \sum_{F_g} \hat{P}_{F_g} S_M \hat{P}_{F_g} Z(F_g) + \sum_{F_g \neq F'_g} \hat{P}_{F'_g} S_M \hat{P}_{F'_g} Z(F'_g) \right]$$

$$\alpha^2 \approx 0$$

The definition of factor $G$ can be found in (A.4) and $S_M$ represents the $M$ spherical component of the electron spin vector in the ground state. Appendix A discusses in more detail the tensor polarizability and the level of approximation involved when assuming $\alpha^2 \approx 0$. Using Equations (2.26) and (2.27) together with (2.32)-(2.34) and

$\text{In terms of the cartesian components the spherical components are: } S_{\pm 1} = \mp \frac{1}{\sqrt{2}} (S_x \pm iS_y), \quad S_0 = S_z.$
the properties of the inner and outer product (Equations (A.25)- (A.27)) we write:

\[ \delta E^0 = -|E_0|^2 G \sum_{F_g} \hat{P}_{F_g} Re[Z(F_g)] \] (2.35)

\[ \delta \Gamma^0 = \frac{2|E_0|^2 G}{\hbar} \sum_{F_g} \hat{P}_{F_g} \text{Im}[Z(F_g)] \] (2.36)

\[ \delta E^1 = -\frac{|E_0|^2 G}{4} [11 - 4J_e(J_e + 1)] \times \left\{ \sum_{F_g} \hat{P}_{F_g} S \hat{P}_{F_g} \text{Re}[Z(F_g)] \right. \\
+ \frac{1}{2} \sum_{F_g \neq F_g'} \left[ \hat{P}_{F_g} S \hat{P}_{F_g} Z(F_g') + Z^\dagger(F_g') \hat{P}_{F_g} S \hat{P}_{F_g} \right] \left. \right\} \cdot \mathbf{s} \] (2.37)

\[ \delta \Gamma^1 = \frac{|E_0|^2 G}{2} [11 - 4J_e(J_e + 1)] \times \left\{ \sum_{F_g} \hat{P}_{F_g} S \hat{P}_{F_g} \text{Im}[Z(F_g)] \right. \\
+ \frac{1}{2} i \sum_{F_g \neq F_g'} \left[ \hat{P}_{F_g} S \hat{P}_{F_g} Z(F_g') - Z^\dagger(F_g') \hat{P}_{F_g} S \hat{P}_{F_g} \right] \left. \right\} \cdot \mathbf{s} \] (2.38)

In the last equations we used the identity for the photon spin (unit) vector:

\[ \mathbf{s} = \frac{\mathbf{e}^\ast \times \mathbf{e}}{i} \] (2.39)

As mentioned before, we assumed a (classical) monochromatic light with electric field given by: \( \mathbf{E} = (\mathbf{E}_0^* e^{i\omega t} + \mathbf{E}_0 e^{-i\omega t}) \). In general the atomic polarizability tensor couples ground hyperfine levels with different quantum number for the total angular momentum, i.e. in the last summation terms of Equations (2.37) and (2.38) \( F_g \) can be different than \( F_g' \). These terms describe hyperfine coherences and have an effect on the evolution of the hyperfine coherence. For instance, due to the cross-hyperfine coupling in Eq. (2.37) intensity noise (e.g from quantum mechanical fluctuations) of the light can create noise at the hyperfine frequency. However, for the atomic
magnetometer described in this thesis we are only interested in Zeeman coherences
and also there is no coherent hyperfine excitation\textsuperscript{7}. It will be appropriate then to

drop out the cross-hyperfine terms in equations (2.37) and (2.38) and write:

\begin{align*}
\delta E^1 &= -\frac{|E_0|^2 G}{4} \left[ 11 - 4J_e(J_e + 1) \right] \left[ \sum_{F_g} \hat{P}_{F_g} \hat{S} \hat{P}_{F_g} \text{Re}[Z(F_g)] \right] \cdot \mathbf{s} \\
\delta \Gamma^1 &= \frac{|E_0|^2 G}{2} \left[ 11 - 4J_e(J_e + 1) \right] \left[ \sum_{F_g} \hat{P}_{F_g} \hat{S} \hat{P}_{F_g} \text{Im}[Z(F_g)] \right] \cdot \mathbf{s}
\end{align*}

It is convenient to write the light-shift (Equations (2.35) and (2.40) ) in a more
intuitive form:

\begin{align*}
\delta E &= \delta E_c + \delta A \mathbf{I} \cdot \mathbf{S} - \mathbf{L}_{ls} \cdot \mathbf{S} \\
\delta E_c &= -\frac{|E_0|^2 G}{2} \left[ \text{Re}[Z_\alpha] \frac{I + 1}{2I + 1} + \text{Re}[Z_\beta] \frac{I}{2I + 1} \right] \\
\delta A &= \frac{4|E_0|^2 G}{2I + 1} \text{Re}[Z_\alpha - Z_\beta] \\
\mathbf{L}_{ls} &= \frac{|E_0|^2 G}{4} \left[ 11 - 4J_e(J_e + 1) \right] \left[ \sum_{F_g} \hat{P}_{F_g} \text{Re}[Z(F_g)] \right] \cdot \mathbf{s}
\end{align*}

where $Z_\alpha$, $Z_\beta$ are the profile factors for the ground state hyperfine manifold $(S + 1/2)$
and $(S - 1/2)$ respectively.

The light-shift term \(\delta E_c\) simply shifts the energy levels and does not provide any
state dependent information. This term, although not important to magnetometers
or clocks, it is crucial in realizing spatially dependent potentials for atoms (e.g., in
optical lattices). The second term in the right hand side of Eq. (2.42) changes the
hyperfine transition frequency causing a shift in the hyperfine structure, but does not
affect the Zeeman coherences (and therefore the magnetometer is not affected either).

Finally, the last term in Eq. (2.42) is formally the same as interaction with a magnetic

\textsuperscript{7}This can be created for instance with the application of a weak RF field at the hyperfine frequency
or with modulating coherently the intensity of the light.
field, so that the light-shift can be treated as an effective magnetic field defined in Eq. (2.45). Obviously, the Zeeman structure and consequently the magnetometer operation are affected by this part of the light-shift.

2.4.4 Polarizability at high-pressures/large detunings

So far, we have made no assumptions about how the Doppler broadening compares with the collisional broadening or the detuning. In the case of high buffer-gas pressures (in the range of a few hundred Torrs) or detunings much larger than the Doppler broadening we can take advantage of the Plasma-Dispersion function $Z(\zeta)$ \[73\] property that for $|\zeta| \gg 1$:

$$Z(\zeta) = -\frac{1}{\zeta} \quad (2.46)$$

The profile factor then takes the simple form:

$$Z(F_g) = -k \sqrt{\frac{2k_B T}{M_a}} \frac{1}{(\omega - \omega_{F_g}) + i\Gamma} \quad (2.47)$$

By simple substitution of (2.47) to the polarizability equation (2.32) - (2.33) we find:

$$\alpha^0 = -\frac{r_e c^2 f_{osc}}{2\omega} \sum_{F_g} \frac{\hat{P}_{F_g}}{(\omega - \omega_{F_g}) + i\Gamma} \quad (2.48)$$

$$\alpha^1 = -\frac{r_e c^2 f_{osc}}{4\sqrt{2\omega}} [11 - 4J_e(J_e + 1)] \sum_M (-1)^M Q_{1,M}$$

$$\times \left[ \sum_{F_g} \frac{\hat{P}_{F_g} S_M \hat{P}_{F_g}}{(\omega - \omega_{F_g}) + i\Gamma} \right] \quad (2.49)$$

where we used Eq. (A.4) and the classical electron radius $r_e = \frac{e^2}{m_e c^2}$.

In addition, when the collisional broadening or detuning is much larger than the ground state hyperfine separation we can drop the dependence of $Z(F_g)$ on $F_g$ (all

19
The approximation of neglecting the hyperfine contribution to $\alpha$ starts to fail at low pressures for the heavier alkali-metal atoms, especially Rb and Cs.
\( \alpha = \alpha_R + i\alpha_I \) relate to each other through the Kramers-Kronig transform \[106\]:

\[
\begin{align*}
\alpha_R(\omega) &= \frac{P}{\pi} \int_{-\infty}^{\infty} \frac{\alpha_I(\omega')d\omega'}{\omega' - \omega} \\
\alpha_I(\omega) &= -\frac{P}{\pi} \int_{-\infty}^{\infty} \frac{\alpha_R(\omega')d\omega'}{\omega' - \omega}
\end{align*}
\] (2.57) (2.58)

The symbol \( P \) denotes the principal part of the integral. The above equations follow from the casual connection between the polarization and the electric field. One consequence of the Kramers-Kronig relationship is that by measuring the absorption profile of the medium (imaginary component of the polarizability) we can find out the refractive index (related to the real part of polarizability).

2.4.5 Summary

Here, we give a brief summary of some of the important results obtained previously. Within the dipole, rotating wave approximation and for negligible excited state population the interaction Hamiltonian between atoms and light takes the simple form:

\[ \delta H = \delta E_v - \frac{i}{2} \delta \Gamma. \]

We introduced the index \( v \) in the light-shift operator to emphasize the fact that it refers to \textit{virtual} transitions as opposed to real transitions (discussed in the Section [2.5]). When the pressure broadening is much larger than the Doppler broadening and when we can ignore the ground and excited hyperfine structure, the
light-shift and absorption can be written as:

$$\delta E_v = \hbar \delta \Omega \left[ 1 + \frac{11 - 4J_e(J_e+1)}{4} S \cdot s \right]$$  \hspace{1cm} (2.59)

$$\delta \Gamma = R \left[ 1 + \frac{11 - 4J_e(J_e+1)}{4} S \cdot s \right]$$  \hspace{1cm} (2.60)

$$\delta \Omega = \frac{c|E_0|^2}{2\pi \hbar \nu} r_e f_{osc} c D(\nu)$$  \hspace{1cm} (2.61)

$$R = \frac{c|E_0|^2}{2\pi \hbar \nu} r_e f_{osc} c \frac{\Gamma/2\pi}{(\nu - \nu_0)^2 + (\Gamma/2\pi)^2}$$  \hspace{1cm} (2.62)

$$D(\nu) = \frac{\nu - \nu_0}{(\nu - \nu_0)^2 + (\Gamma/2\pi)^2}$$  \hspace{1cm} (2.63)

Here, we expressed the quantities in frequency space instead of the angular frequency ($\omega = 2\pi \nu$), and $\nu_0$ is the resonant transition frequency. Equation (2.62) gives the pumping rate per unpolarized atom. The above equations can be expressed in terms of the photon flux $\Phi$ (photons/time) and the area of the beam $A_b$, taking into account that:

$$\frac{\Phi}{A_b} = \frac{c|E_0|^2}{2\pi \hbar \nu}$$  \hspace{1cm} (2.64)

For $J_e = 1/2$ (D1 transition) and $J_e = 3/2$ (D2 transition) the term multiplying $S \cdot s$ is 2 and -1 respectively.

We note that so far we have assumed monochromatic light. This is a very good approximation for a laser light field. The extension to non-monochromatic light is straightforward. For example, if $\tilde{\Phi}(\nu)$ is the photon flux density then the pumping rate is:

$$R = \int_{-\infty}^{\infty} \frac{\tilde{\Phi}(\nu)}{A_b} r_e f_{osc} c \frac{\Gamma/2\pi}{(\nu - \nu_0)^2 + (\Gamma/2\pi)^2} d\nu$$  \hspace{1cm} (2.65)

### 2.4.6 Effective Magnetic-like field from light

Equation (2.61) describes how light field (with non-zero photon spin) couples with the atomic spin; the effect is formally equivalent with the coupling of a magnetic
field \( \mathbf{L} \) to the electron spin. By equating the vector light-shift energy shift (the term proportional to \( \mathbf{S} \) in (2.59)) with the Hamiltonian term for a magnetic field \( (g_e\mu_B \mathbf{L} \cdot \mathbf{S}) \), the light-shift magnetic-like field is found:

\[
\mathbf{L} = \Phi \frac{r_e f_{osc}(\nu)}{A_b} \frac{[11 - 4J_e(J_e + 1)]}{8\gamma_e} \mathbf{s}
\]  

(2.66)

where \( \gamma_e = g_e\mu_B/\hbar \) is the electron gyromagnetic ratio.

### 2.5 Repopulation Pumping

In the previous section we studied the evolution of the ground state due to coherent interaction with a light field. We did not take into account how the ground state is influenced from the decay of the excited state. This pumping effect due to the transfer of population from the excited to the ground state is called repopulation pumping. A simple case that demonstrates the principle is illustrated in Fig. 2.2 (taken from [96]). A completely polarized excited \( ^2P_{1/2} \) state decays spontaneously to the ground \( ^2S_{1/2} \) state. The probability for decaying to the \( m_g = -1/2 \) is twice as high as the probability of decaying to the \( m_g = 1/2 \). This way, the ground state will be partially polarized and the repopulation pumping would be proportional to the spontaneous decay rate.

![Figure 2.2](taken from [96]) Spontaneous emission from a spin-polarized excited state leads to partial polarization of the ground state.

\[ ^2P_{1/2} \quad ^2S_{1/2} \quad ^2P_{1/2} \quad ^2S_{1/2} \]

\[ -1/2 \quad 2/3 \quad 1/3 \]

\[ +1/2 \quad 1/2 \quad -1/2 \quad +1/2 \]

\[ ^2P_{1/2} \quad ^2S_{1/2} \quad ^2P_{1/2} \quad ^2S_{1/2} \]

\[ -1/2 \quad 2/3 \quad 1/3 \]

\[ +1/2 \quad 1/2 \quad -1/2 \quad +1/2 \]

\[ ^2P_{1/2} \quad ^2S_{1/2} \quad ^2P_{1/2} \quad ^2S_{1/2} \]

\[ -1/2 \quad 2/3 \quad 1/3 \]

\[ +1/2 \quad 1/2 \quad -1/2 \quad +1/2 \]

\[ ^2P_{1/2} \quad ^2S_{1/2} \quad ^2P_{1/2} \quad ^2S_{1/2} \]

\[ -1/2 \quad 2/3 \quad 1/3 \]

\[ +1/2 \quad 1/2 \quad -1/2 \quad +1/2 \]

\[ ^2P_{1/2} \quad ^2S_{1/2} \quad ^2P_{1/2} \quad ^2S_{1/2} \]

\[ -1/2 \quad 2/3 \quad 1/3 \]

\[ +1/2 \quad 1/2 \quad -1/2 \quad +1/2 \]

\[ ^2P_{1/2} \quad ^2S_{1/2} \quad ^2P_{1/2} \quad ^2S_{1/2} \]

\[ -1/2 \quad 2/3 \quad 1/3 \]

\[ +1/2 \quad 1/2 \quad -1/2 \quad +1/2 \]

\[ ^2P_{1/2} \quad ^2S_{1/2} \quad ^2P_{1/2} \quad ^2S_{1/2} \]

\[ -1/2 \quad 2/3 \quad 1/3 \]

\[ +1/2 \quad 1/2 \quad -1/2 \quad +1/2 \]

\[ ^2P_{1/2} \quad ^2S_{1/2} \quad ^2P_{1/2} \quad ^2S_{1/2} \]

\[ -1/2 \quad 2/3 \quad 1/3 \]

\[ +1/2 \quad 1/2 \quad -1/2 \quad +1/2 \]

\[ ^2P_{1/2} \quad ^2S_{1/2} \quad ^2P_{1/2} \quad ^2S_{1/2} \]

\[ -1/2 \quad 2/3 \quad 1/3 \]

\[ +1/2 \quad 1/2 \quad -1/2 \quad +1/2 \]

\[ ^2P_{1/2} \quad ^2S_{1/2} \quad ^2P_{1/2} \quad ^2S_{1/2} \]

\[ -1/2 \quad 2/3 \quad 1/3 \]

\[ +1/2 \quad 1/2 \quad -1/2 \quad +1/2 \]

\[ ^2P_{1/2} \quad ^2S_{1/2} \quad ^2P_{1/2} \quad ^2S_{1/2} \]

\[ -1/2 \quad 2/3 \quad 1/3 \]

\[ +1/2 \quad 1/2 \quad -1/2 \quad +1/2 \]

\[ ^2P_{1/2} \quad ^2S_{1/2} \quad ^2P_{1/2} \quad ^2S_{1/2} \]

\[ -1/2 \quad 2/3 \quad 1/3 \]

\[ +1/2 \quad 1/2 \quad -1/2 \quad +1/2 \]

\[ ^2P_{1/2} \quad ^2S_{1/2} \quad ^2P_{1/2} \quad ^2S_{1/2} \]

\[ -1/2 \quad 2/3 \quad 1/3 \]

\[ +1/2 \quad 1/2 \quad -1/2 \quad +1/2 \]

\[ ^2P_{1/2} \quad ^2S_{1/2} \quad ^2P_{1/2} \quad ^2S_{1/2} \]

\[ -1/2 \quad 2/3 \quad 1/3 \]

\[ +1/2 \quad 1/2 \quad -1/2 \quad +1/2 \]

\[ ^2P_{1/2} \quad ^2S_{1/2} \quad ^2P_{1/2} \quad ^2S_{1/2} \]

\[ -1/2 \quad 2/3 \quad 1/3 \]

\[ +1/2 \quad 1/2 \quad -1/2 \quad +1/2 \]

\[ ^2P_{1/2} \quad ^2S_{1/2} \quad ^2P_{1/2} \quad ^2S_{1/2} \]

\[ -1/2 \quad 2/3 \quad 1/3 \]

\[ +1/2 \quad 1/2 \quad -1/2 \quad +1/2 \]

\[ ^2P_{1/2} \quad ^2S_{1/2} \quad ^2P_{1/2} \quad ^2S_{1/2} \]

\[ -1/2 \quad 2/3 \quad 1/3 \]

\[ +1/2 \quad 1/2 \quad -1/2 \quad +1/2 \]

\[ ^2P_{1/2} \quad ^2S_{1/2} \quad ^2P_{1/2} \quad ^2S_{1/2} \]

\[ -1/2 \quad 2/3 \quad 1/3 \]

\[ +1/2 \quad 1/2 \quad -1/2 \quad +1/2 \]

\[ ^2P_{1/2} \quad ^2S_{1/2} \quad ^2P_{1/2} \quad ^2S_{1/2} \]

\[ -1/2 \quad 2/3 \quad 1/3 \]

\[ +1/2 \quad 1/2 \quad -1/2 \quad +1/2 \]

\[ ^2P_{1/2} \quad ^2S_{1/2} \quad ^2P_{1/2} \quad ^2S_{1/2} \]

\[ -1/2 \quad 2/3 \quad 1/3 \]

\[ +1/2 \quad 1/2 \quad -1/2 \quad +1/2 \]
In the experiments of this thesis, the repopulation mechanism differs from the simple example of Fig. 2.2. Here only a brief explanation of the repopulation pumping will be mentioned. The interested reader is referred to [9] for a detailed derivation. As discussed in Section 2.7, in order to avoid radiation trapping we use N\textsubscript{2} buffer gas in the experiments described in this thesis. The main decay mechanism of the excited alkali-atoms is the quenching collisions which transfer polarization from the excited to the ground state. The duration of a quenching collision (on the order of \(10^{-12}\) sec) is long enough for the Coulomb and spin-orbit interactions to destroy (most of) the electronic polarization of the alkali atom, but it is short enough for the hyperfine interaction not to change the nuclear spin. This way, a quenching collision transfers the nuclear polarization of the excited state to the ground state with very small change, but does not transfer any electron polarization (i.e. the ground state has zero electron polarization due to repopulation pumping). In order then to describe the repopulation pumping of the ground state, it is sufficient to see how the nuclear polarization of the excited states evolves. There are three mechanisms that affect the excited state: J-damping collisions that destroy the total electronic angular momentum, quenching collisions and the hyperfine interaction that couples the electronic with the nuclear degrees of freedom. Collisions of alkali-atoms with both inert and molecular gas contribute to the J-damping collisions, and they typically happen at a rate of \((1 - 10) \times 10^{10}\) sec\(^{-1}\) (depending on the buffer gas pressure). This is much faster than the characteristic excited state hyperfine timescale (on the order of \(10^{-9}\) sec), so that the hyperfine interaction does not have sufficient time to depolarize the nuclear polarization [9]. In other words, the electronic angular momentum changes directions randomly so frequently that it becomes effectively zero for the slow hyperfine interaction. As shown in [9] the evolution of the ground state due to the...
repopulation pumping at high buffer gas pressures is:

\[
\frac{d\rho}{dt} = R(\phi - \frac{s \cdot \Theta}{2}) + \frac{1}{i\hbar}[\delta E_r, \rho]
\]  

(2.67)

Here \(\delta E_r\) is called the light-shift due to real transition [182] and is on the order of \(\omega_f \bar{m} T_Q R\), where \(\omega_f \bar{m}\) is the Zeeman frequency and \(1/T_Q\) is the quenching rate. This light-shift results from the fact that a (nuclear) coherence that has passed through the excited state and returned to the ground state has acquired a different phase compared to the coherence of atoms that have not been excited. In general, this plays a small role in optical pumping experiments.

### 2.6 Total Optical Pumping

Combing the depopulation and repopulation Hamiltonian we find the net evolution of the density matrix due to optical pumping at high buffer gas pressures:

\[
\frac{d\rho}{dt} = R[\phi(1 + 2s \cdot S) - \rho] + \frac{1}{i\hbar}[\delta E_{op}, \rho]
\]  

(2.68)

where:

\[
\delta E_{op} = \delta E_r + \delta E_v
\]  

(2.69)

We note that for the conditions of the thesis experiment the light-shift due to real transitions, proportional to \(\omega_f \bar{m} T_Q R\) can be ignored and Eq. (2.66) describes adequately the effective magnetic field from light-shifts.

Comparing Eq. (2.68) with Eq. (2.132) it can be seen that the optical pumping effect on the the density matrix evolution is the same as spin exchange at a rate \(R\) with fictitious alkali-metal atoms of electronic spin \(s/2\).
2.7 Radiation Trapping and Quenching

In optical pumping experiments, it is important to suppress the spontaneous emission from the atoms. The reason is that a fluorescent photon has a random polarization with respect to the laboratory frame, which can be resonantly absorbed by the atoms and depolarize them. This is called radiation trapping and can be a significant source of depolarization, especially in a high density vapor (see [140] for an extensive overview of the subject. However, buffer gases may quench the excited atoms without the emission of a fluorescent photon. Studies have shown that direct conversion of the excitation energy to kinetic energy of the colliding atoms is highly unlikely, but the rotational and vibrational degrees of freedom of a molecular buffer gas can absorb the excitation energy of the alkali atom. This non-radiative decay becomes more efficient when the excitation energy resonantly matches the rovibrational levels of the molecular buffer gas. A particularly attractive buffer gas molecule for this purpose is $\text{N}_2$, which exhibits high efficiency quenching and also does not react with the alkali atoms. As discussed in [169] the probability of an atom for radiative decay is given by:

$$Q = \frac{1}{1 + \frac{p_Q}{\tilde{p}_Q}}$$

where $p_Q$ is quenching gas pressure and $\tilde{p}_Q$ is the characteristic pressure. For $\text{N}_2$ $\tilde{p}_Q \approx 6 \text{ Torr}$ [136], so for the experimental value of $p_Q \approx 50 \text{ Torr}$ the probability of radiative decay is $\approx 10\%$.

---

10 This is not to say that the fluorescent light from a polarized light is random; instead the fluorescence retains information about the polarization state of the emitting atom. This results in partial rather than complete depolarization of the absorbing atom. The depolarizing effect is further reduced by the fact that only the electronic part of angular momentum is affected by this process, whereas the nuclear part remains unaffected.
2.8 Light propagation in an atomic ensemble

In this section we examine the evolution of light due to the polarizability Hamiltonian. For now, we are going to stay within the classical description of light.

We consider a plane wave propagating along the $\hat{z}$ direction with electric field given by:

$$E = E_0(z, t)e^{ikz - \omega t}$$ (2.71)

where $\omega$ and $k = \frac{\omega}{c}$ are respectively the temporal and spatial frequencies and $E_0(z, t)$ is the electric field amplitude that can vary slowly with the propagation distance and time. In this formulation $E_0$ is a vector and has the information about the polarization of light. From Maxwell’s equations we get [90, 106]:

$$\left( \frac{\partial^2}{\partial z^2} - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right) E = \frac{4\pi}{c^2} \frac{\partial^2}{\partial t^2} P$$ (2.72)

Here, $P$ is the polarization per unit volume vector, and is related to the polarizability tensor and the electric field:

$$P = [A] \text{Tr}[\rho\alpha]E = [A] \text{Tr}(\alpha)E$$ (2.73)

where $[A]$ is the alkali metal density. Substituting for the polarization in Eq. (2.72) and noting that the dominant spatial and temporal dependence of the electric field is expressed by the exponentials $e^{ikz}$ and $e^{-i\omega t}$ so that:

$$|kE_0| \gg |\frac{\partial E_0}{\partial z}|, |\omega P| \gg |\frac{\partial E_0}{\partial z}|$$ (2.74)

we can write [97]:

$$\left( \frac{\partial}{\partial z} + \frac{1}{c} \frac{\partial}{\partial t} \right) E_0 = i2\pi k[A]\langle\alpha\rangle E_0$$ (2.75)
Ignoring retardation effects and substituting for the susceptibility tensor the above equation is written:

\[
\frac{\partial}{\partial z} E_0 = i2\pi k [A] \langle \alpha \rangle E_0
\]  

(2.76)

In the most general case of anisotropically polarized medium \(E_0\) can have both longitudinal (along the direction of propagation) and transverse (perpendicular to the direction of propagation) components. Mathematically, this can be seen from the fact that even for initially transverse polarization the cross product in (2.56) may generate a longitudinal field. However, the longitudinal components are many orders of magnitude smaller than the transverse, and they can be safely ignored. Therefore, we will concentrate on the “transverse” polarizability defined as [97]:

\[
\langle \alpha_{\perp} \rangle = T \langle \alpha_{\perp} \rangle T
\]

(2.77)

\[
T = 1 - nn
\]

(2.78)

where \(1 = i_0^0 + i_1 (i_1)^* + (i_{-1} i_{-1})^*\) is the unit dyadic and \(n\) is the propagation direction of light.

We seek to find the eigenvectors of the polarizability: that is, the electric field in Eq. (2.70) that remains parallel to itself after the application of the (transverse) polarizability tensor. The polarizability can be written in the form:

\[
\alpha = \alpha^0 \left( T + i \frac{1 - 4 J_e (J_e + 1)}{2\sqrt{2}} \sum (-1)^M S_M T Q_{-M}^1 T \right)
\]

(2.79)

Taking into account that \(T i_\mu i_\nu T = 0\) for \(\mu = 0\) or \(\nu = 0\) (the z axis was taken to be the longitudinal direction), it is straightforward to prove that:

\[
\langle \alpha_{\perp} \rangle = \alpha^0 \left[ i_1 (i_1)^* + i_{-1} (i_{-1})^* \right]
\]

\[
+ \alpha^0 \frac{1 - 4 J_e (J_e + 1)}{4} \langle S_0 \rangle \left[ i_1 (i_1)^* - i_{-1} (i_{-1})^* \right]
\]

(2.80)
Form the above equation it is obvious that the eigenvectors of the transverse polarizability are the spherical basis vectors $i_{\pm 1}$. These correspond to left and right circularly polarized light.

The solution to Eq. (2.76) is then

$$E_0(z) = e^{i2\pi[Ak]n\alpha_0} \left[i_1(i_1)^* + i_{-1}(i_{-1})^* e^{-i2\pi k[A]n\alpha_0} \right] E_0(0)$$

where we introduced the gyrotropic part of the polarizability:

$$\alpha_{gt} = \alpha_0 \frac{11 - 4J_e(J_e + 1)}{4}$$

Equation (2.81) is general and describes the effect of atoms in light (optical rotation and absorption).

### 2.8.1 Faraday Rotation

If we can neglect the imaginary component of $\alpha_0$ (i.e. $|\omega - \omega_{S\rightarrow J_e}| \gg \Gamma$), Eq. (2.81) corresponds to a rotation of the polarization axis of the light by an angle

$$\theta_{FR}(z) = [A] r_e c_fosc \frac{11 - 4J_e(J_e + 1)}{8} \frac{2\pi(\omega - \omega_{S\rightarrow J_e})}{[(\omega - \omega_{S\rightarrow J_e})^2 + \Gamma^2]} \langle n \cdot S \rangle_z$$

This effect is called the electron paramagnetic Faraday rotation and is used in order to monitor the polarization of the medium in an optical atomic magnetometer.

The sign and absolute value of the Faraday rotation depend on the excited state electron angular momentum ($J_e$). We see that for the D1 and D2 transitions the sign of $\theta$ is opposite. To the extent that there is small absorption (i.e. light is far

---

11Equation (2.81) is valid for uniform alkali polarization. In the general case of varying atomic spin the product $\langle n \cdot S \rangle_z$ has to be replaced by: $\int_0^z dz \langle n \cdot S \rangle_z$

12If we also include the imaginary part of $\alpha_{gt}$ describing the absorption, the effect of atoms on the state of light is more complicated than a simple optical rotation; in fact, linearly polarized incident light will acquire in general an elliptical polarization.
detuned from resonance), the contribution of the different electronic states to the Faraday effect is additive. For a light detuned from the D1 and D2 line the angle of polarization rotations is:

\[ \theta_{\text{FR}}(z) = [A] r_e c \left[ f_{\text{osc}}^{D1} D(\nu) \big|_{D1} - \frac{1}{2} f_{\text{osc}}^{D2} D(\nu) \big|_{D2} \right] \langle S_z \rangle z \]  

(2.84)

\[ = \frac{1}{2} [A] r_e c \left[ f_{\text{osc}}^{D1} D(\nu) \big|_{D1} - \frac{1}{2} f_{\text{osc}}^{D2} D(\nu) \big|_{D2} \right] \langle P_z \rangle z \]  

(2.85)

where \( P_z = 2S_z \) is the electron polarization in the light propagation direction.

### 2.8.2 Optical Absorption

For frequencies close to the optical resonance (i.e. \( |\omega - \omega_{S\rightarrow J_e}| \sim \Gamma \)) the light field experiences attenuation \( \left( \text{light absorption is expressed mathematically in the imaginary part of } \alpha \right) \) which cannot be neglected. It is straightforward to show that for circularly polarized light the intensity \( \propto E_0 E_0^* \) propagation is described by:

\[ \frac{dI^\pm(z)}{dz} = -[A] \sigma I^\pm(z) \]  

(2.86)

\[ \sigma = \frac{r_e c f_{\text{osc}} \Gamma / 2\pi}{(\nu - \nu_0)^2 + (\Gamma/2\pi)^2} \left[ 1 - \frac{11 - 4J_e (J_e + 1)}{4} \langle S \cdot s \rangle \right] \]  

(2.87)

where \( s \) is the photon spin (2.39) and \( \sigma \) is the absorption cross section (for circularly polarized light). In the case where atomic polarization results from the pumping action of the light field the \( s \) and \( \langle S \rangle \) point in the same direction and the absorption cross section can be written:

\[ \sigma = \frac{r_e c f_{\text{osc}} \Gamma / 2\pi}{(\nu - \nu_0)^2 + (\Gamma/2\pi)^2} \left[ 1 - \frac{11 - 4J_e (J_e + 1)}{8} P_z \right] \]  

(2.88)

\(^{13}\text{We consider alkali media in their ground electronic state.}\)
where $P_z$ is the alkali polarization (assumed to vary from 0 to 1) along the direction of light propagation.

### 2.9 Usefulness of Spherical Tensor Representation of $\rho$

As discussed in A.3, the density matrix can be decomposed into irreducible spherical (or equivalently polarization) operators. The importance of this representation in optical pumping experiments and magnetometry lies on how light and magnetic fields couple with the different multipoles of $\rho$. Static or oscillating magnetic fields simply rotate the atomic ensemble, but do not change the multipolarity of the distribution. In addition, light couples through only a second rank tensor to the atomic ensemble, and therefore only the dipole and quadrupole polarization (but not higher order polarizations) determine the propagation of light. In the particular case that the second-rank part of polarizability can be ignored, it will be enough to find the evolution of the dipole polarization in order to describe the experiment. Furthermore, under isotropic conditions which are often encountered in optical pumping experiments with not very high magnetic fields, each multipole polarization is decoupled from the rest and relaxes with a characteristic decay time. This can be a significant simplification, as will be shown later.

### 2.10 Relaxation of Polarized Atoms

There are various mechanisms that can destroy or change the polarization of optically pumped alkali-atoms. The most important of these are collisional dynamics of alkali-atoms with other alkali-atoms or buffer gases, spatial diffusion to depolarizing walls and through magnetic field gradients. We will focus on the relaxation of the
ground state, since the experiments of this thesis are based on the evolution of the ground state density matrix. In general, the relaxation mechanism can affect the mean value of angular momentum along the quantization axis and/or the coherences between states (non-diagonal elements). We call the first longitudinal relaxation and the second transverse. A more technical definition of the longitudinal and transverse relaxation can be given based on the expansion of the density matrix in the basis of irreducible spherical tensors (or eigenpolarization) $T_{LM}$; longitudinal refers to relaxation of the $M = 0$ tensors, while transverse refers to relaxation of $|M| \geq 1$ tensors.

2.10.1 Collisional Relaxation

Because of the collisions an additional term $V$ in the Hamiltonian should be added. This interaction depends on the internuclear distance between the colliding atoms and the spin state of the alkali-atom. It may also depend on the internal state of the perturbing atom and on the collisional velocity. We should point out that the collisional spin interaction $V$ that appears in the evolution of the density matrix should be an average over all the different time histories for each atom in the ensemble.

An important aspect of the collisional dynamics is their very fast timescales, typically on the order of $10^{-12}$ sec (with the important exception of collisions leading to Van Der Waals molecules, which we are not going to consider). As mentioned before for the quenching collisions this leads to approximating the collision as sudden with respect to quantities that evolve typically at much longer timescales (e.g. the nuclear angular momentum for many of the collisions).

Each mechanism is associated with a characteristic rate, that effectively measures the strength of the relaxation interaction. The rate can be written in the form:

$$R_v = n \langle \sigma v \rangle$$

(2.89)
where $n$ is the density of the perturbing gas, $\sigma$ is the collisional cross section and has units of area, and $v$ is the velocity in the center of mass frame. The $\langle \rangle$ denotes average over the distribution of the collisional parameters (relative velocity, effective impact parameter). It is customary in literature to write the above equation as:

$$R_r = n\sigma \bar{v}$$

(2.90)

where $\sigma$ is the effective cross section, and $\bar{v}$ is the mean velocity of the colliding pair in the center of mass frame:

$$\bar{v} = \sqrt{\frac{8k_B T}{\pi M}}$$

(2.91)

where the reduced mass $M$ depends on the mass $M_a$ of the alkali-atom and on the mass $M'$ of the colliding partner:

$$\frac{1}{M} = \frac{1}{M'} + \frac{1}{M_a}$$

(2.92)

We note that in (2.90) the effective cross section has a temperature dependence.

There has been extensive theoretical work on how to calculate from first principles the interaction and rate for various collisions. The interested reader is referred to [97, 98, 193] and references therein for a thorough discussion of the topic. However, there are a lot of uncertainties associated with the theoretical calculation and one may find significant differences between the theoretical prediction and the experiment. Here we simply mention the most important (for our experimental conditions\footnote{For example we ignore the relaxation due to the formation of short-lived Van Der Waals molecules, which are significant for Xe buffer gas but not for He.}) collisional contribution to the Hamiltonian.


**Hyperfine-Shift Interaction**

The hyperfine coupling constant of the ground state is proportional to the probability density of the valence electron of the alkali-atom to be at the site of the nucleus, that is: $A_{hf} \propto |\psi(0)|^2 \text{[97]}$. During the collision, the electronic wavefunction is perturbed and the hyperfine constant is modified. Therefore, we can write:

$$V = \delta A_{hf} \mathbf{I} \cdot \mathbf{S} \quad (2.93)$$

This interaction leads to pressure shifts in the microwave resonances (which is of special importance in the atomic clock physics), but has a negligible effect on the Zeeman populations (at least for not very large magnetic fields).

**Spin-Rotation Interaction**

The spin-rotation interaction has the form \text{[194]}:

$$V_{NS} = \gamma_r \mathbf{N} \cdot \mathbf{S} \quad (2.94)$$

and couples the electron spin $\mathbf{S}$ with the relative angular momentum $\mathbf{N}$ of the colliding pair of atoms. This interaction randomizes only the electronic part of the angular momentum, while the nuclear part remains unaffected (S-damping collisions), and the evolution of the density matrix is written:

$$\frac{d\rho}{dt} = -\frac{1}{T_{NS}}(\rho - \phi) \quad (2.95)$$

Under conditions typical for optical pumping experiments, the spin-rotation interaction is the main relaxation mechanism due to buffer gases without nuclear spin (e.g. $\text{N}_2$ and $^4\text{He}$).
Spin-Spin Rotation interaction

Spin destruction cross sections due to alkali-alkali collisions have been measured to be much larger than the cross sections in alkali-buffer gas collisions. It has been hypothesized [24] that the interaction that couples the electron spin with the relative angular momentum of the colliding pair has the form:

\[ V_{SS} = \frac{2}{3} \lambda_{SS} \cdot (3 \hat{R} \hat{R} - 1) \cdot S \]  

(2.96)

where \( S = S_1 + S_2 \) is the total electronic spin of the colliding alkali atoms, and \( \hat{R} \) is the unit vector along the internuclear axis. As for the spin-rotation interaction, only the electronic angular momentum is affected in the collision (S-damping collision) and the evolution of the density matrix is similar to (2.95) with the appropriate replacement for the rate:

\[ \frac{d\rho}{dt} = -\frac{1}{T_{SS}}(\rho - \phi) \]  

(2.97)

Although the current theory is not entirely consistent with experiments [9], the last equation is still an excellent approximation for the ground state evolution due to spin-destruction collisions between alkali-atoms.

Dipolar interaction

In a binary collision between two spins \( K_1 \) and \( K_2 \) (not necessarily the same species) the magnetic dipole interaction couples the spins to the relative orbital angular momentum of the colliding atoms. The interaction is the one between magnetic dipoles:

\[ V_{dd} = \frac{\mu_1 \mu_2}{r^3 K_1 K_2} [K_1 \cdot K_2 - 3(K_1 \cdot \hat{r})(K_2 \cdot \hat{r})] \]

(2.98)

Here, \( \mu_1 \) and \( \mu_2 \) are the magnetic moments of the spins \( K_1 \) and \( K_2 \) respectively, \( r \) is the distance and \( \hat{r} \) the unit vector connecting the spins. The scattering of two
spins from the same species according to the above interaction has been treated in [145]. For example, the magnetic dipolar scattering for a gas of spin-1/2 particles with no interaction at all (except for the magnetic dipole-dipole interaction) leads to a relaxation rate:

\[
\frac{1}{T_{dd}} = \frac{32}{\hbar^4} n \mu \sqrt{8 \pi mk_B T}
\]

where \(n\) is the density of the spins, \(T\) is the temperature and \(k_B\) the Boltzmann constant.

**Spin Exchange collisions**

Spin-exchange processes can have a significant effect on the polarization of optically pumped alkali-metal atoms (and noble gases). We defer to Sections 2.12 and 2.13 for a discussion of the topic.

**2.10.2 Diffusion**

In general, the density matrix depends on time and position within the cell. We should therefore add a diffusional term in the evolution of the density matrix given by:

\[
\frac{\partial \rho}{\partial t} = D \nabla^2 \rho
\]

where \(D\) is the diffusion constant. The value of \(D\) depends on the type and pressure of buffer-gas (for a single species buffer-gas \(D\) is inversely proportional to pressure), and there is also a weak temperature dependence. For the above diffusion equation to be a good approximation the mean free path of the atoms \(\lambda\) should be much smaller than the characteristic length of the cell or any characteristic length-scale (\(l\)) of the normal Hamiltonian (which in the most general case may depend on the spatial location of the atoms), and the diffusional time scale \(\lambda/\bar{v}\) should be much shorter than any other characteristic rate (other relaxation rate or Larmor precession). If this condition does
not hold then the atoms move ballistically rather than diffusively. In our experiments, the condition for diffusion $\lambda \ll l$ holds and we can use the diffusion equation.

In order to solve the diffusion equation we need to specify (besides the initial $t=0$ condition) boundary conditions at the walls of the cell. The boundary conditions depend on the type of the wall surface, but in general they can be described with the accommodation factor $\tilde{A}$ which captures the details of the interaction of the polarized atoms with the walls and is defined by the equation [97]:

$$J_{+} - J_{-} = \tilde{A}J_{+}$$

(2.101)

The current density to the surface $J_{+}$ and returning back from it $J_{-}$ are [134]:

$$J_{+} = [A]\bar{\nu}\left(\frac{1}{4} - \frac{\lambda}{6} n \cdot \nabla\right) \rho$$

(2.102)

$$J_{-} = [A]\bar{\nu}\left(\frac{1}{4} + \frac{\lambda}{6} n \cdot \nabla\right) \rho$$

(2.103)

where $n$ is the unit vector normal to the surface and $[A]$ is the atomic density. We can then write:

$$\tilde{A}\rho = -\frac{2\lambda}{3}(2 - \tilde{A})n \cdot \nabla \rho$$

(2.104)

When an alkali atom diffuses to the wall, it gets adsorbed and adheres to the wall, and eventually after some mean time $\tau_{ad}$ (typically in the range of $10^{-5} - 10^{-7}$ sec [54]) it escapes back to the vapor. During the adsorption time the atom experiences fluctuating (due to the motion of the atoms) electric and magnetic fields from the atoms, ions and molecules of the glass surface, which can cause complete or partial depolarization. Surface coatings that cause only partial depolarization have been found and used in many experiments. However, with the recent notable exception of octadecyltrichlorosilane (OTS), their application has been limited to low

\[\text{Paraffin, octadecyltrichlorosilane and several silane coatings are among those that have been used}\]
temperatures (up to 80°C). In the experiments of this thesis we did not use any surface coating, so that for our purposes the walls are considered as completely depolarizing. Then, the density matrix at the walls corresponds to completely unpolarized atoms, so that the boundary condition (2.104) is written:

\[ \rho = \frac{1}{\text{Tr}[\hat{I}]} = \rho_{UP} \]  

where \( \hat{I} \) is the unit matrix and \( \rho_{UP} \) is the density matrix for completely unpolarized atoms.

**Wall Relaxation**

We seek to find a simple rate equation to account for the effect of diffusion to the walls of the completely depolarizing walls of the cell. The starting point is the diffusion Eq. (2.100) with the boundary condition (2.105). The solution of (2.100) and (2.105) can be written in the form:

\[ \rho(t, r) = \sum_i (\tilde{\rho}_i - \rho_{UP}) e^{-i k_D r - \Gamma_D t} \]  

(2.106)

The dummy index \( i \) denotes the various eigenmodes of the diffusion equation. The spatial frequency \( k_D \) is related to the relaxation rate \( \Gamma_D \) through:

\[ |k_D|^2 = \frac{\Gamma_D}{D} \]  

(2.107)

In Eq. (2.106) the “amplitude” \( \tilde{\rho}_i \) depends on the initial condition:

\[ \tilde{\rho}_i = \rho_{UP} + \int d^3 r \rho(0, r) e^{ik_D r} \]  

(2.108)
Because of the boundary condition (2.105) the spatial frequencies are discrete, and so are the relaxation rates \( \Gamma_{D_i} \). In most of the experiments, it is adequate to keep only the slowest diffusion mode, which has the smallest relaxation rate and the largest amplitude for (close to) homogeneous initial conditions. Then (2.100) reduces to:

\[
\frac{d\rho}{dt} = -\Gamma_d(\rho - \rho_{UP}) \tag{2.109}
\]

where the spatial dependence of \( \rho \) has been dropped, and \( \Gamma_d = k_{D0}^2 D \) is the diffusional damping rate. For a spherical cell of radius \( l \):

\[
k_{D0} = \pi / l \tag{2.110}
\]
\[
\Gamma_d = \frac{D\pi^2}{l^2} \tag{2.111}
\]

while for a rectangular parallelepiped cell of dimensions \((l_x, l_y, l_z)\)

\[
k_{D0} = \pi \sqrt{\frac{1}{l_x^2} + \frac{1}{l_y^2} + \frac{1}{l_z^2}} \tag{2.112}
\]
\[
\Gamma_d = D\pi^2 \left( \frac{1}{l_x^2} + \frac{1}{l_y^2} + \frac{1}{l_z^2} \right) \tag{2.113}
\]

**Relaxation due to Magnetic Field Gradients**

In its most general form, the effect of magnetic field gradients can be complicated and depends on the motion (diffusional or ballistic) of the atoms, the Larmor frequency and the boundary conditions on the walls of the cell. As explained in [42] we can distinguish two regimes depending on the value of the parameter:

\[
\kappa = \frac{\Omega L l^2}{D} = \frac{\tau_D}{\tau_L} \tag{2.114}
\]
where $\Omega_L$ is the average Larmor frequency, $l$ is the characteristic length scale of the cell, $\tau_D = l^2/D$ is the timescale of diffusion through the cell and $\tau_L = 1/\Omega_L$ is the characteristic precession time. We are going to focus on the regime where the parameter $\kappa \gg 1$, that is, the precession time is short compared to the diffusion time $\tau_D$.

We consider weak magnetic field gradients, i.e. $|\nabla B|/|B| \ll 1$, where $B$ is the local field. The total field can be written as the sum of a strong homogeneous field $B_0 = \frac{1}{V} \int_V B \, dV$ and a weak field $B_1(r)$ that depends on position. For concreteness, we take the direction of the homogeneous field to be the $z$ direction and $x$ and $y$ are the transverse directions (in the lab frame).

It will be instructive to sketch a derivation of the longitudinal relaxation rate. The Brownian motion causes atoms to experience a randomly fluctuating magnetic field, so that to the normal homogeneous Hamiltonian we can add the perturbation:

$$V_{MG} = -\hbar \gamma \mathbf{K} \cdot \mathbf{B}_{eff}(t)$$ (2.115)

where $\mathbf{K}$ is the spin, $\gamma$ the gyromagnetic ratio and $\mathbf{B}_{eff}$ is the effective fluctuating field \[96\]:

$$\mathbf{B}_{eff} = \frac{v \cdot [(\nabla B) \times B]}{\gamma |B|^2}$$ (2.116)

Here $v$ is the atomic velocity and is a random variable with an exponentially decreasing correlation function and correlation time $\tau_D$. We can use equations (2.121)-(2.124) to calculate the effect of the perturbation. It will be convenient to expand the density matrix in terms of eigenpolarizations in the spherical basis $T_{LM}$. As explained in \[96, 95\] for isotropic conditions (zero or very weak magnetic field) the different eigenpolarizations ($2^L$ poles) relax independently and the relaxation rate for each of the $2^L$ poles depends on the multipolarity ($l$) of the weak interaction $V$ in Eq. (2.121). For

---

16 Although we did perform experiments at very small magnetic fields, these experiments were conducted at high buffer gas pressures and the diffusion time was long (on the order of seconds).
a magnetic perturbation the multipolarity of the interaction is \( l = 1 \) and the independence of eigenpolarization relaxation is preserved even at finite magnetic fields. As discussed above in our experiments we are interested in the dipole relaxation \((L = 1)\). After a bit of algebra we find that dipole longitudinal \((L = 1, M = 0 \text{ in the spherical tensor } T_{LM})\) relaxation rate due to magnetic field gradient is:

\[
\frac{1}{T_{1,D}} \approx D \frac{(|\nabla B_{1x}|^2 + |\nabla B_{1y}|^2)}{|B_0|^2} \tag{2.117}
\]

Similarly, the origin of the transverse relaxation is the loss of phase coherence as the diffusing atoms experience different magnetic field. To calculate the transverse relaxation a more complete theory described in \([42, 100, 137]\) is required. The theory, valid only when the main relaxation mechanism of the atoms is diffusion through magnetic field gradients\(^{17}\), solves the diffusion equation with a source (i.e. inhomogeneous diffusion equation), where the source term (which has the information of the magnetic field gradients) is added in a perturbative way. The dimension of the cell and the boundary conditions at the walls affect the solution and therefore the transverse relaxation rate. For a spherical cell of radius \(l\), with non-depolarizing walls the dipole \((L=1)\) transverse relaxation is:

\[
\frac{1}{T_{2,D}} \approx \frac{8l^4|\gamma \nabla B_0|}{175D} \tag{2.118}
\]

For alkali atoms the boundary condition for non-depolarizing walls is not satisfied under the conditions of our experiments. However, even for completely depolarizing walls \((2.118)\) gives a good estimate of the transverse relaxation. We note that in the high pressure regime \((\kappa \gg 1)\), the longitudinal relaxation scales linearly with the

\(^{17}\)Equivalently the diffusion time through the cell should be smaller than the transverse relaxation caused from all mechanisms.
diffusion constant (inversely proportional to pressure), while the transverse relaxation is inversely proportional to the diffusion constant (proportional to pressure).

Besides the decoherence, the atoms (statistically) also pick an extra phase as they diffuse through the gradients. This is expressed as a frequency shift which at high pressures can be written $[42]$:

$$
\delta \Omega_L \approx \frac{l^2}{10\gamma|B_0|} (|\nabla B_{1y}|^2 + |\nabla B_{1y}|^2)
$$

Typically, this is only a small correction to the Larmor frequency and can always be included in an effective magnetic field.

## 2.11 Relaxation of Polarized Noble Gas Atoms

Similar relaxation mechanisms exist also for the nuclear spin of polarized noble gas atoms. Typically, the nuclear spin does not couple to external degrees of freedom as strongly as the electron spin and the relaxation rates for polarized noble gas are usually much smaller than those for alkali atoms. This makes the effect of certain relaxation mechanisms (that are comparatively too small to influence the polarization of alkali atoms) to the evolution of noble gas polarization pronounced. For example, nuclear spins with $I \geq 1$ couple with electric field gradients, leading to quadrupolar relaxation (see for example $[81]$). However, in this thesis we used noble gas with nuclear spin $I = 1/2$, and therefore we don’t need to consider the quadrupolar relaxation.

Furthermore, it has been demonstrated that even without surface coatings certain types of glasses depolarize only weakly the nuclear spin of noble gas $[165]$. Then (2.111) and (2.113) are only lower limits of the relaxation.
2.12 Spin-Exchange Collisions between Alkali-Metal Atoms and Noble Gas Atoms

The presence of noble gas atoms with non-zero nuclear spin leads to a type of spin exchange interaction between the electron spin of an alkali-atom and the nuclear spin of the noble gas\(^{18}\):

\[ V_{KS} = \alpha \mathbf{K} \cdot \mathbf{S} \]  \hspace{1cm} (2.120)

The interaction coefficient \( \alpha \) is small enough to consider the interaction \( V_{KS} \) as a weak perturbation. Then, as explained in [1], the density matrix equation to second order is governed by the equation:

\[ \frac{d\tilde{\rho}}{dt} = -\langle \int_0^\infty [\tilde{V}(t), [\tilde{V}(t-\tau), \tilde{\rho}]]d\tau \rangle \]  \hspace{1cm} (2.121)

Here \( \langle \rangle \) denotes average over the atomic ensemble, and \( \tilde{\rho} \) and \( \tilde{V} \) are the density matrix and collisional interaction respectively in the interaction representation:

\[ \tilde{\rho} = e^{iH_{0}t}\rho e^{-iH_{0}t} \]  \hspace{1cm} (2.122)
\[ \tilde{V} = e^{iH_{0}t}V e^{-iH_{0}t} \]  \hspace{1cm} (2.123)

where \( H_{0} \) is the non-perturbing normal Hamiltonian of the atom. For sudden collisions we can write Eq. (2.121) in the form [97]:

\[ \frac{d\rho}{dt} = \frac{1}{i\hbar}[H, \rho] + \frac{1}{2\hbar^{2}}[V_{F}(0), [V_{F}(0), \rho]] + \frac{1}{i\hbar}[V_{F}(0), \rho] \]  \hspace{1cm} (2.124)

and \( V_{F}(0) \) is the zero component of the Fourier Transform of \( V(t) \).

\(^{18}\)In Eq. (2.120) only the dominant isotropic magnetic-dipole hyperfine interaction was included. For a more precise treatment one has to account for the anisotropic magnetic-dipole hyperfine and electric quadrupole interaction (see [194]). It has been pointed out that anisotropic spin-exchange interactions put a limit on the noble gas polarization attainable through spin-exchange optical pumping [192].
After a bit of algebra it can be shown that when the nuclear spin of the noble gas is \( K = 1/2 \) the spin-exchange interaction leads to a density matrix evolution (for the ground state of alkali-atoms) that is given to first order by:

\[
\frac{d\rho}{dt} = \frac{1}{T_{KS}} [\phi(1 + 4\langle K \rangle \cdot S) - \rho] + \frac{1}{i\hbar}[\delta E_{KS}, \rho]
\]  

(2.125)

The frequency shift operator has an effect similar to a magnetic field and is:

\[
\delta E_{KS} = \frac{8\pi g_e \mu_B \mu_K}{3K} \tilde{\kappa}_{KS}[X] \langle K \rangle \cdot S
\]  

(2.126)

where \([X]\) is the noble-gas density and the enhancement factor \( \tilde{\kappa}_{KS} \) is a dimensionless parameter that depends on the colliding pair and weakly on temperature \([158, 9]\). In addition to the spin exchange collisions, the polarized noble gas generates a magnetic field (as predicted from the classical electromagnetic theory) which contributes to the energy shift. In literature \([91, 164, 158]\) it is customary to define an enhancement factor \( \kappa \) in such a way that, besides the collisional interaction, it also includes the (typically smaller) effect of the long-range magnetic dipolar interaction; in particular, the enhancement factor \( \kappa \) is defined as the ratio of the mean effective magnetic field experienced by the alkali atoms (due to the overlapping noble gas ensemble) to the macroscopic field \((8\pi/3)(\mu_K[K])\langle K \rangle/K\) which would be produced by the noble gas in a spherical cell at the same density and polarization \([164, 91]\). That is, in a spherical geometry the overall frequency shift of alkali metals due to the presence of polarized noble gas is:

\[
\delta E_{KS} = \frac{8\pi g_e \mu_B \mu_K}{3K} \kappa[X] \langle K \rangle \cdot S
\]  

(2.127)
Similarly, the noble gas (with spin quantum number $K = 1/2$) experiences a frequency shift due to the polarized alkali-metal ensemble:

$$
\delta E_{KS} = \frac{8\pi g_e \mu_B \mu_K}{3K} \kappa[A] \langle S \rangle \cdot K
$$

We note that although $\tilde{\kappa}_{KS}$ (which describes a contact interaction) does not depend on the geometry of the ensemble, the enhancement factor $\kappa$ (through the magnetization classical magnetic field term) is affected by the shape of the ensemble.

### 2.13 Spin-Exchange Collisions between Pairs of Alkali Metal Atoms

During collisions between alkali-atoms a strong exchange interaction is present:

$$
V_{SE} = \eta S_1 \cdot S_2
$$

where $S_1$ and $S_2$ are the electron spins of the colliding atoms. In general, the colliding atoms can be different species or different isotopes. The above interaction is of electrostatic origin and therefore its strength is much larger than the other relaxation interactions. The singlet and triplet states of the dimer that is formed during the collision have different energies (typically $\approx 0.5$ eV near the classical turning points) and they can be described by an interaction potential of the form:

$$
V(r) = V_0(r) + \eta(r) S_1 \cdot S_2.
$$

In general the colliding atomic pair will be in a superposition of the singlet and triplet state of the dimer; because of the different energies of the two states the singlet and triplet amplitudes will evolve at different rates, so that the spin states of the two atoms may change after the collision. However, the exchange interaction preserves the total spin $[S_1 + S_2, V_{SE}] = 0$. Therefore, for same species collisions the spin-exchange mechanism contributes to the transverse relaxation but
does not cause longitudinal relaxation. Figure 2.3 shows an example where the total spin angular momentum is conserved, but the angular momentum is redistributed in the ground state sublevels.

![Diagram](image)

Figure 2.3: (Taken from [TK]) Spin-exchange collisions between alkali atoms conserve the total angular momentum projection \( m_{F1} + m_{F2} \) but the level populations are redistributed.

The alkali spin-exchange interaction is strong enough and cannot be treated as a weak perturbation for the equations (2.121) and (2.124) to be applicable. A more elaborate theory using partial-wave analysis of the scattering or by a classical path analysis is required (see [99, 97] and references therein). The result of this analysis leads to a density matrix equation:

\[
\frac{d\rho}{dt} = \frac{1}{T_{SE,12}} \left[ \phi (1 + 4 \langle S_2 \rangle \cdot S_1) - \rho \right] + \frac{1}{i\hbar} \left[ \delta E_{SE,12}, \rho \right] \quad (2.130)
\]

\[
\delta E_{SE,12} = \frac{2\hbar \kappa_{SE}}{T_{SE,12}} \langle S_2 \rangle \cdot S_1 \quad (2.131)
\]

For a single species, single isotope the evolution of the density matrix is:

\[
\frac{d\rho}{dt} = \frac{1}{T_{SE}} \left[ \phi (1 + 4 \langle S \rangle \cdot S) - \rho \right] + \frac{1}{i\hbar} \left[ \delta E_{SE}, \rho \right] \quad (2.132)
\]

\[
\delta E_{SE} = \frac{2\hbar \kappa_{SE}}{T_{SE}} \langle S \rangle \cdot S \quad (2.133)
\]
Although there are many optical pumping experiments that use two or more alkali species [46 47], in this thesis we performed experiments with a single species and (to a good approximation) single isotope alkali atomic vapor ($^{39}$K). We will therefore concentrate on Equations (2.132)-(2.133).

It is worth noting that the equation for the density matrix evolution is non-linear, which can add significant complication to the problem. However, as will be discussed later there are certain limiting cases (that are frequently encountered in optical pumping experiments), where the alkali spin-exchange relaxation can take a simple form.

### 2.14 Spin Temperature Distribution

In optical pumping experiments, we frequently encounter situations where the spin-exchange rate is much more rapid than any other relaxation or pumping rate. For example, because of the large spin-exchange cross-section, this is typically the case for high alkali density. Although the discussion of this section can be generalized to the case of multiple alkali (and noble gas) spin species, here we will limit ourselves to the experimentally relevant situation of very rapid spin-exchange rate between alkali-alkali atoms of single species. Under such conditions and when there are no coherences (so that the density matrix is determined by the occupational probability of the Zeeman sublevels) the ground density can be very well described by the spin-temperature distribution:

$$\rho_{ST}(\beta) = \frac{e^{\beta I_z} e^{\beta S_z}}{Z S T} = \frac{e^{\beta F_z}}{Z I Z S}$$  \hspace{1cm} (2.134)

where $z$ is the longitudinal axis of polarization $\langle S \rangle = \langle S_z \rangle \hat{z}$. In the above equation $\beta$ is called the spin-temperature and $Z = Z_S Z_I$ is the partition function of the distribution.

We emphasize that here we refer to the collisional spin-exchange rate ($R = n\sigma_{SE}\bar{v}$) and we discriminate it from the relaxation rate due to spin-exchange collisions.
For a spin of integer or half-integer quantum number $K$ the partition function $Z_K$ is written:

$$Z_K = \sum_{m=\pm K}^{K} e^{m\beta} = \frac{\sinh (\beta[K]/2)}{\sinh(\beta/2)}$$

(2.135)

The temperature parameter is related to the longitudinal spin polarization, which for a spin $K$ is defined to be:

$$P_K = \langle K_z \rangle / K$$

(2.136)

It can be readily shown that:

$$P_S = 2\langle S_z \rangle = \tanh \frac{\beta}{2}$$

(2.137)

or equivalently:

$$\beta = \ln \frac{1 + P_S}{1 - P_S}$$

(2.138)

Taking into account that for spin 1/2 [188] and cartesian spin component $S_i$:

$$e^{\beta S_i} = \hat{I} + \cosh \frac{\beta}{2} + 2S_i \sinh \frac{\beta}{2}$$

(2.139)

the spin temperature distribution (2.134) can be written in the form:

$$\rho_{ST} = \phi(\hat{I} + 4\langle S_z \rangle S_z)$$

(2.140)

where $\phi = e^{\beta I_z}/(2Z_I)$ is the part of $\rho_{ST}$ with no electron polarization (2.5). From the above equation it is obvious that the spin-temperature distribution is the steady state solution of (2.132), i.e. $\rho_{ST}$ is invariant to spin-exchange collisions between alkali-atoms. Similarly, the spin-temperature distribution is stationary with respect to the evolution due to optical pumping (2.68). Also, it can be shown that:

$$[g\mu B_F z + A_{hf} \mathbf{I} \cdot \mathbf{S}, \rho_{ST}] = 0$$

(2.141)
where $g\mu B_z F_z$ is the Hamiltonian term due to interaction with a longitudinal static magnetic field. Equation (2.141) indicates that $\rho_{ST}$ is invariant to the hyperfine and longitudinal magnetic interaction.

We note that the spin-temperature distribution is the distribution that maximizes entropy under the constraint of a specific longitudinal polarization.

As mentioned before, the spin temperature distribution does not account for any coherences. However this does not limit its usefulness, because for small transverse excitation the density matrix can be considered as a perturbation to spin-temperature distribution and we can expand $\rho$ in a perturbative way around $\rho_{ST}$, which can lead to significant simplifications. Furthermore, although $\rho_{ST}$ is not (time) invariant under certain relaxation mechanisms (e.g. diffusion to the walls), when the spin-exchange rate is much more rapid than these relaxation rates (as is usually the case) we can approximate the longitudinal density matrix (that does not include coherences) with the spin-temperature distribution $\rho = e^{\beta F_z}$, where the spin-temperature parameter $\beta$ depends on time.

For atoms described (approximately) by the spin-temperature distribution we will find it convenient to define the paramagnetic "slowing down" coefficient [9]

$$q = \frac{\langle F_z \rangle}{\langle S_z \rangle} = 2\langle \mathbf{F} \cdot \mathbf{F} - F_z^2 \rangle = 1 + 2\langle \mathbf{I} \cdot \mathbf{I} - I_z^2 \rangle$$  \hspace{1cm} (2.142)

The coefficient $q$ depends on the spin-temperature and can be written as a function of the nuclear spin ($I$) and the electron polarization ($P$). For $I = 3/2$ (e.g. $^{39}$K) $q = 1 + (5 + P^2)/(1 + P^2)$. From the definition it can be seen that $q$ measures how the total atomic angular momentum is distributed between the electron and the nucleus.

\footnote{In [9] a slightly different definition of the paramagnetic coefficient $\epsilon$ is used. The two coefficients are related through the equation $q = 1 + \epsilon$}
2.15 Density Matrix Evolution

Before proceeding, for clarity we write down the evolution of the ground density matrix of alkali-atoms including the most significant processes (for the experiments of the thesis) that affect the atomic state:

$$\frac{\partial \rho}{\partial t} = \frac{1}{i\hbar}[A_{hf} \mathbf{S} \cdot \mathbf{I} + g_e \mu_B \mathbf{S} \cdot \mathbf{B}, \rho] + \frac{1}{i\hbar} [\delta E_{op} + \delta E_{SE} + \delta E_{KS}, \rho]$$

$$+ \frac{1}{T_{SE}} [\phi(1 + 4\langle \mathbf{S} \rangle \cdot \mathbf{S}) - \rho] + \frac{1}{T_{KS}} [\phi(1 + 4\langle \mathbf{K} \rangle \cdot \mathbf{S}) - \rho]$$

$$+ R[\phi(1 + 2\mathbf{s} \cdot \mathbf{S}) - \rho] + \frac{1}{T_{SD}} (\phi - \rho) - \Gamma_d (\rho - \rho_{UP}) + D \nabla^2 \rho \quad (2.143)$$

Here, we used the rate $1/T_{SD}$ to describe all the electron spin relaxation mechanisms.

2.16 Bloch equations

The full dynamical state of the atomic ensemble is given by the evolution of the density matrix. A significant simplification over the density matrix equation was proposed by Felix Bloch [25] who suggested that the spin evolution of the atomic ensemble in many cases can be described with phenomenological rate equations. The generic form of the Bloch equations for the magnetization $\mathbf{M}$ can be written:

$$\frac{d\mathbf{M}}{dt} = \gamma \mathbf{M} \times \mathbf{B} - \frac{\mathbf{M}_t}{T_2} - \frac{M_z - M_0}{T_1} \hat{z} \quad (2.144)$$

Alternatively, we can include explicitly the mechanism that creates polarization and write:

$$\frac{d\mathbf{M}}{dt} = \gamma \mathbf{M} \times \mathbf{B} - \frac{\mathbf{M}_t}{T_2} - \frac{M_z}{T_1} \hat{z} + R_p \hat{s}_p \quad (2.145)$$

Here, $\gamma$ is the effective gyromagnetic ratio of the atomic species, $\hat{z}$ is the longitudinal direction in the laboratory frame specified by a large static magnetic field or the
pumping direction, \( M_0 = R_p T_1 \) is the steady state longitudinal magnetization \( \mathbf{M}_t \). \( R_p \) is the rate of magnetization creation along the direction defined by the unit vector \( \hat{s}_p \). We point out that the magnetization \( \mathbf{M} \) is a classical vector. In (2.144) and (2.145) the phenomenological rates \( 1/T_1 \) and \( 1/T_2 \) (different in general) were introduced to describe the approach of longitudinal and transverse magnetization respectively to equilibrium. As shown in [66], Bloch equations have a solid theoretical justification for a two level system, but even for spins higher than 1/2 they have been proven exceedingly fruitful in the study of magnetic resonance phenomena.

Since in an optical alkali magnetometer we monitor the spin motion through the electron paramagnetic Faraday interaction, it will be convenient for the description of the experiments to express the Bloch equations explicitly in terms of the electron spin rather than the more abstract magnetization (that is we replace \( \mathbf{M} \) in (2.144) and (2.145) with \( \langle \mathbf{S} \rangle \)). For the noble gas species, we use the nuclear spin for the magnetization. We note that for the Bloch equations to describe electron spin or diamagnetic atoms the gyromagnetic ratio in (2.144) and (2.145) should be negative.

2.16.1 Steady state solution of the Bloch equation

Before proceeding, it is interesting to examine the steady state solution predicted by the Bloch equations in the presence of a dc magnetic field \( B_z = -\omega_0/\gamma \) and an oscillating transverse field of amplitude \( B_1 \) and (angular) frequency \( \omega \). The interested reader is referred to [1,169] for a thorough discussion on the topic.

We assume a circularly polarized transverse field \( \mathbf{B} = B_1 [\hat{x} \cos \omega t + \hat{y} \sin \omega t] \). The solution is most easily derived in the rotating frame, the (non-inertial) frame that rotates around the \( B_z \) at the frequency \( \omega \). With appropriate choice of phase, Eq. (2.144)

\[ \text{In writing the Bloch equations we implicitly assumed that the longitudinal field } (B_z) \text{ is significantly larger than the transverse field. For a more general case of large (oscillating) transverse field see [1].} \]
becomes in the rotating frame:

\[ \frac{dM}{dt} = \gamma M \times B_{\text{eff}} - \frac{\tilde{M}_x \hat{i} + \tilde{M}_y \hat{j}}{T_2} - \frac{M_z - M_0}{T_1} \hat{k} \]  
\[(2.146)\]

\[ B_{\text{eff}} = \left( \omega - \omega_0 \right) \frac{\hat{k}}{\gamma} + B_1 \hat{i} = \frac{\Delta \omega}{\gamma} \frac{\hat{k}}{\gamma} + B_1 \hat{i} \]  
\[(2.147)\]

where \( \hat{i} = \hat{x} \cos \omega t + \hat{j} \sin \omega t, \hat{j} = -\hat{x} \sin \omega t + \hat{j} \cos \omega t \) and \( \hat{k} = \hat{z} \) are the unit vectors in the rotating frame, and \( \tilde{M}_x \) and \( \tilde{M}_y \) are the transverse components of \( M \) in that frame. It is straightforward to show [1] that:

\[ \tilde{M}_x = \frac{\Delta \omega \gamma B_1 T_2}{1 + (\Delta \omega T_2)^2 + \gamma^2 B_1^2 T_1 T_2} M_0 \]  
\[(2.148)\]

\[ \tilde{M}_y = \frac{\gamma B_1 T_2}{1 + (\Delta \omega T_2)^2 + \gamma^2 B_1^2 T_1 T_2} M_0 \]  
\[(2.149)\]

\[ M_z = \frac{1 + (\Delta \omega T_2)^2}{1 + (\Delta \omega T_2)^2 + \gamma^2 B_1^2 T_1 T_2} M_0 \]  
\[(2.150)\]

In the laboratory frame the transverse components are:

\[ M_x = \tilde{M}_x \cos \omega t - \tilde{M}_y \sin \omega t \]  
\[(2.152)\]

\[ M_y = \tilde{M}_x \sin \omega t + \tilde{M}_y \cos \omega t \]  
\[(2.153)\]

In actual experiments, linearly polarized transverse fields are often applied (e.g. a solenoidal coil creates a linearly polarized magnetic field). In the case of large Larmor frequency \( \omega_0 \gg (\gamma B_1, 1/T_1, 1/T_2) \) the counter-rotating component \( \rightarrow B_1/2 \) can be neglected and the steady state magnetization is given by equations \((2.148)-(2.153)\) with the conversion \( B_1 \rightarrow B_1/2 \) (see [1]).

\[ ^{22}\]The linearly polarized field can be decomposed into circularly polarized components of opposite helicity (rotating and counter-rotating).
For $\omega_0 \sim 1/T_1$ the counter-rotating component has a non-negligible effect on $M$ and the analytical solution to (2.144) and (2.145) cannot be given in terms of elementary functions. Assuming the oscillating field leads to small perturbation of the longitudinal polarization so that the longitudinal polarization can be considered constant, the Bloch equations become linear and one can show that in the steady state:

\[
M_x = (1 - i\omega T_2) M_t, \quad B_t = \hat{x} B_1 e^{i\omega t} \quad (2.154)
\]
\[
M_y = \omega_0 T_2 M_t, \quad B_t = \hat{y} B_1 e^{i\omega t} \quad (2.155)
\]
\[
M_x = \omega_0 T_2 M_t, \quad B_t = \hat{y} B_1 e^{i\omega t} \quad (2.156)
\]
\[
M_y = - (1 - i\omega T_2) M_t, \quad B_t = \hat{y} B_1 e^{i\omega t} \quad (2.157)
\]
\[
M_t = \frac{\gamma B_1 M_0 T_2}{(i + \omega T_2)^2 - (\omega T_2)^2} \quad (2.158)
\]

A particularly interesting case arises in the limit of vanishing frequency $\omega \to 0$. In practice this corresponds to fields that change very slowly at timescales $\sim T_2$ (i.e. $\frac{dB_t}{dt} \ll 1/T_2$). The system of Bloch equations can be solved analytically (by setting to zero the left hand side of Equations (2.144)); it is straightforward to show that in the steady state:

\[
M_x = M_0 \frac{-\tilde{B}_y + \tilde{B}_x \tilde{B}_z}{1 + \tilde{B}_x^2 + \tilde{B}_y^2 + \tilde{B}_z^2} \xrightarrow{\tilde{B} \ll 1} -M_0 \tilde{B}_y \quad (2.159)
\]
\[
M_y = M_0 \frac{\tilde{B}_x + \tilde{B}_y \tilde{B}_z}{1 + \tilde{B}_x^2 + \tilde{B}_y^2 + \tilde{B}_z^2} \xrightarrow{\tilde{B} \ll 1} M_0 \tilde{B}_x \quad (2.160)
\]
\[
M_z = M_0 \frac{1 + \tilde{B}_z^2}{1 + \tilde{B}_x^2 + \tilde{B}_y^2 + \tilde{B}_z^2} \xrightarrow{\tilde{B} \ll 1} M_0 \quad (2.161)
\]
\[
\tilde{B} = \gamma T_1 B \quad (2.162)
\]

\[23\]Within the linear approximation the response to an arbitrary excitation can be calculated as a superposition of the solutions Eq. (2.154)-(2.158).
In the above equations we set \( T_1 = T_2 \) (which is the case for low magnetic field \( \gamma |B| \sim 1/T_1, 1/T_2 \)).

### 2.16.2 Estimation of \( T_1 \) and \( T_2 \)

By using the full density matrix equation \( T_1 \) and \( T_2 \) can be calculated (in principle) from the various relaxation rates mentioned in the previous section. Similarly, the effective gyromagnetic ratio \( \gamma \) can be derived from the electron (or nuclear in the case of noble gas) gyromagnetic ratio. In general, the phenomenological parameters \( T_1, T_2 \) and \( \gamma \) depend on the strength of the magnetic field, the spin-exchange rate and the polarization of the ensemble. Staying within the conditions relevant to the experiments of the thesis, we are interested in the regime where the density matrix is described to leading order by the spin-temperature distribution (2.134) and the weak transverse fields (if they exist at all) create small deviations from the spin-temperature distribution. As far as the strength of the static magnetic field is concerned, we are going to consider two limiting cases: magnetic field leading to Larmor frequency \( \alpha \) much smaller and \( \beta \) significantly larger than the spin-exchange rate.

#### \( T_1 \) estimation

The parameter \( T_1 \) is estimated from the rate of longitudinal relaxation. Multiplying (2.143) with \( F_z \) and taking the trace for the spin temperature distribution (2.134) we find:

\[
\frac{d}{dt} q \langle S_z \rangle = -\frac{1}{T_{SD}} \langle S_z \rangle + R \left( \frac{s_z}{2} - \langle S_z \rangle \right) - \frac{\langle K_z \rangle}{T_{KS}} - \Gamma_d q \langle S_z \rangle
\]

The paramagnetic coefficient \( q \) was defined in (2.142). In the above equation we used the fact that the pumping light is along the longitudinal \( z \) direction and that for the spin temperature distribution (Equation (2.134)), which does not include transverse excitation, the expectation values of the atomic and nuclear spins are all longitudinal.
and also $\rho_{ST}$ is invariant to spin-exchange collisions between alkali atoms. This leads to a significant simplification, since the spin-exchange nonlinear terms in the density matrix evolution do not enter.

The slowing down factor is not a sensitive function of polarization (e.g. it ranges from 4 to 6 for atoms with $I = 3/2$); in applications where the polarization does not change appreciably (as is the case in the steady state magnetometry) we can (to first order) drop out the time dependence of the coefficient $q$ and write Eq. (2.164) in the form:

$$\frac{d}{dt} \langle S_z \rangle = -\frac{1}{qT_{SD}} \langle S_z \rangle + \frac{R}{q} \left( \frac{s_z}{2} - \langle S_z \rangle \right) - \frac{\langle K_z \rangle}{qT_{KS}} - \Gamma_d \langle S_z \rangle$$

(2.165)

Before proceeding, we need to justify further the use of the longitudinal spin temperature distribution (2.134) in deriving (2.164). In the experiments performed in this thesis, the transverse excitation (if existed at all) was very small; it is appropriate then to write the full density matrix as $\rho = \rho_{ST} + \epsilon \tilde{\rho}$, with $\epsilon \ll 1$, so that the contribution of $\rho_{ST}$ to $\langle S_z \rangle$ is much larger than the one from $\tilde{\rho}$, which can therefore be neglected. For a derivation of the longitudinal relaxation in the regime where this approximation fails the interested reader is referred to [9] (the analysis is limited for low polarization). It is shown there that in general (when the spin-exchange rate is comparable to other relaxation rates) the longitudinal relaxation depends also on the spin-exchange rate between alkali atoms.

**$T_2$ Estimation:**

The estimation of the phenomenological parameter $T_2$ can be quite complex, and no general derivation is known to the writer. Since the spin temperature distribution $\rho_{ST}$ does not include any coherences, we need to know the full density matrix to describe the evolution of $\langle S_{x,y} \rangle$. A more elaborate calculation is therefore required. In [9] a

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\[24\] In fact, we only need to know the dipole polarization component of the density matrix as mentioned above.
solution of the density matrix equation is presented in detail. The problem is formulated in terms of the transverse relaxation operator Λ that acts on the Hilbert space spanned from the Liouville vectors \(|fm_F+1⟩⟨fm_F|\) (we neglect hyperfine coherences and are only interested to one photon Zeeman transitions); the eigenvalues of Λ are found, with the real and imaginary part of the eigenvalues representing the damping and precession frequency of coherences respectively. A similar calculation restricted to the low polarization regime is performed in [99]. There, the density matrix equation is solved treating the spin-exchange and Zeeman (magnetic) contributions as perturbations to the hyperfine interaction (the extension to include spin-destruction mechanisms is straightforward). The solution of the density matrix is expressed in terms of the polarization operators (rather than the \(|fm⟩⟨f′m′|\) of [9]), which allows for a closed form analytic solution (but only applicable to low polarization atomic ensembles). In [23] a low polarization estimation of \(T_1\) and \(T_2\) is presented; rate equations for the mean value of the total spin at each hyperfine manifold of the ground state are derived and the projection theorem \(⟨S⟩ = (⟨F_α⟩ - ⟨F_β⟩)/2[I]\) is then used to retrieve the Bloch equations.

The interested reader is referred to the above mentioned works for a detailed calculation of the phenomenological parameters. Here, we are simply going to mention the result in cases pertinent to the experiments of this thesis.

**Low magnetic field:** A significant simplification applies when the precession rate around a static magnetic field is much smaller than the spin-exchange rate between alkali atoms. The spin-exchange and hyperfine interactions are isotropic; when the mechanisms that break the spherical symmetry (e.g. magnetic field and pumping light) are only small perturbations\(^{25}\) to the isotropic Hamiltonian (e.g. hyperfine and spin-exchange interaction) \(T_2\) and \(T_1\) become equivalent and Eq. (2.164) can also be used.

\(^{25}\) The pumping rate for typical magnetometer applications is on the order of the total spin-destruction rate and therefore much smaller than the spin-exchange rate.
to describe the transverse relaxation. We emphasize that in this low magnetic field case spin-exchange collisions between alkali-atoms do not contribute (to first order) to the relaxation (transverse or longitudinal) of the atoms. A more detailed analysis shows that the contribution of the spin-exchange rate is second order to magnetic field in the low polarization regime [99, 6]:

$$\frac{1}{T_{SE}^2} = \frac{\omega_0^2 T_{SE} [I] (1 - C_q^2)}{2C_q}$$  \hspace{1cm} (2.166)

$$C_q = \frac{[I]^2 + 2}{3[I]}$$  \hspace{1cm} (2.167)

$$\omega_0 = \frac{g_e \mu_B B_0}{[I]} \hbar$$  \hspace{1cm} (2.168)

The independence (to first order) of spin relaxation from alkali spin-exchange collisions at low magnetic field results in atomic magnetometers with very narrow linewidths and subsequently high sensitivities [6, 114, 52, 189, 115] to spin coupling fields. We call this regime where alkali spin-exchange rate is much larger than the Larmor frequency as the Spin Exchange Relaxation Free or SERF regime.

**Large Magnetic field**  When the static magnetic field is large enough so that the resonant frequencies $\omega_{f,\bar{m}} = \hbar^{-1}|E_{f,m} - E_{f,m-1}|$ are well resolved from each other we can write a simple equation for the transverse relaxation rate as explained in [9] and [110]:

$$\gamma_{f\bar{m}} = \left( \frac{1}{T_{SE}} + \frac{1}{T_{SD}} + R + \frac{2\langle K_z \rangle}{T_{KS}} \right) \left( \frac{3[I]^2 + 1 - 4\bar{m}^2}{4[I]^2} \right) - \left( \frac{P}{T_{SE}} + R_{S_z} \right) \bar{m} \frac{(-1)^{\alpha_f - f}}{[I]} \frac{[J]^2 - 4\bar{m}^2 Q_{\bar{m}}}{4[I]^2} \frac{Q_{\bar{m}}}{T_{SE}}$$  \hspace{1cm} (2.169)

Here we used the quantum number $\bar{m} = m - 1/2$ to describe a $\Delta m = 1$ ground state Zeeman transition and $Q_{\bar{m}}$ is the probability that the nuclear spin has the azimuthal
quantum number $\bar{m}$ for the spin-temperature distribution:

$$Q_{\bar{m}} = \frac{2P(1 + P)^{I+\bar{m}}(1 - P)^{I-\bar{m}}}{(1 + P)[I] - (1 - P)[I]}$$  \hspace{1cm} (2.170)$$

We note that the approach to (transverse) equilibrium is not described by a single exponential, but rather each Zeeman transition decays with a characteristic timescale.

For intermediate static magnetic fields with $1/T_{SE} \ll \omega_0 \ll \omega_{hf}$ (here, $\omega_{hf}$ is the hyperfine frequency) there is no simple general analytic solution. However, in the two limiting (but very interesting too) cases of low and high polarization there is a simple expression for the smallest damping rate. As shown in [99] for small polarization $P \to 0$:26

$$\frac{1}{T_2} = \frac{1 + I + 2I^2}{I^2} R_{SD} + \frac{R_{SE}}{8}$$  \hspace{1cm} (2.171)$$

and in the limit of high polarization $P \to 1$, for an atomic species with nuclear spin $I = 3/2$ we have [9, 163]:

$$\frac{1}{T_2} = \frac{R_P}{4} + \frac{R_{SD}}{2} + (1 - P)R_{SE}G(\omega_0, R_{SE})$$  \hspace{1cm} (2.172)$$

$$G(\omega_0, R_{SE}) = \text{Re} \left[ \frac{R_{SE} + 4i\omega_0^2/\pi \nu_{hf}}{5R_{SE} + 8i\omega_0^2/\pi \nu_{hf}} \right]$$  \hspace{1cm} (2.173)$$

Here, $R_{SD}$ is the sum of all the electron spin-destruction rates and $\nu_{hf}$ is the hyperfine frequency. In our experiment we had $\omega^2/R_{SE}\nu_{hf} \ll 1$, in which case $G(\omega_0, R_{SE}) \approx 1/5$.

We emphasize that in the calculation of $T_2$ we used the slowest relaxation mode. Other modes decay much faster and generally have negligible contribution to the magnetometer response.

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26We neglect the pumping rate, since in the low polarization regime $R_{SD} >> R$. 

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2.16.3 Effective Gyromagnetic Ratio:

For low magnetic field $\omega_0 T_{SE} \ll 1$ the precession rate $\omega$ depends on polarization and can be written as a function of the slowing down coefficient defined in \[2.142\] [6 162]:

$$\gamma = \frac{g_e \mu_B}{q \hbar} \quad (2.174)$$

The above equation has a simple intuitive explanation. We consider the precession rate in the presence of a static magnetic field and we note that the net precession rate can be thought as the weighted average of the opposite frequencies in the two hyperfine manifolds, so that:

$$\omega = \omega_0 \frac{\langle F_\alpha \rangle - \langle F_\beta \rangle}{\langle F_\alpha \rangle + \langle F_\beta \rangle} \quad (2.175)$$

From the projection theorem $S_{\alpha,\beta} = (-1)^{\alpha-f} F_{\alpha,\beta}$ and the definition of slowing down coefficient \[2.142\] we arrive at Eq. \(2.174\).

The above argument for the effective precession rate holds only in the limit of rapid spin-exchange collisions between alkali atoms ($\omega_0 T_{SE} \ll 1$). For static magnetic fields large enough so that $\omega_0 T_{SE} \gg 1$ it can be shown [9] that the gyromagnetic ratio is:

$$\gamma = \frac{g_e \mu_B}{[I] \hbar} \quad (2.176)$$

Here, we neglected the much weaker coupling of the nuclear spin to the magnetic field.
2.16.4 Two species spin-exchange collisions

When two different\(^{27}\) atomic spin systems are present, the effect of spin-exchange collisions between different species (i.e. between alkali-metal and noble gas atoms with nonzero nuclear spin) has to be included. Multiplying both sides of Eq. \((2.130)\) with \(S\) we find after a bit of algebra\(^{28}\):

\[
\frac{d\langle S \rangle}{dt} = g_S \mu_B \lambda M_K \frac{\langle K \rangle}{K} \times \langle S \rangle + R_{se}^{en} (\langle K \rangle - \langle S \rangle) \quad (2.177)
\]

Here, \(R_{se}^{en}\) is the spin exchange rate per alkali-metal atom, \(M_K = [X] \mu_K\) is the noble gas magnetization (the product of nuclear magnetic moment \(\mu_K\) with the noble gas density \([X]\)), \(g_e \approx 2\) is the electron g-factor, \(\mu_B\) the Bohr magneton, and \(\lambda = 8\pi \kappa/3\) (in CGS units) is characteristic of the interaction of the alkali-atom with the noble gas\(^{29}\).

An analogous rate equation holds for \(\langle K \rangle\):

\[
\frac{d\langle K \rangle}{dt} = \frac{\mu_K}{K} \lambda M_S \frac{\langle S \rangle}{S} \times \langle K \rangle + R_{se}^{ne} (\langle S \rangle - \langle K \rangle) \quad (2.178)
\]

Here, \(R_{se}^{ne}\) is the spin exchange rate per noble gas atom and \(M_S = [A] g_S \mu_B S\) is the alkali atom magnetization (the product of the electron magnetic magnetic \(g_S \mu_B S \approx \mu_B\) with the alkali density). At sufficiently high pressures (practically above an atmosphere for \(^3\)He) when the contribution of van der Waals molecules is negligible the enhancement factor \(\kappa\) (and subsequently the coefficient \(\lambda\)) have the same value in equations \((2.177)\) and \((2.178)\) \([164]\).

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\(^{27}\)In the previous paragraphs we described how spin exchange collisions between pairs of same species alkali-atoms influence \(T_2\).

\(^{28}\)In deriving \((2.177)\) the following identities are useful \([97]\): \(S_i S_j S_l = |S(S+1) - 1|\) and \(\epsilon_{ijk} S_i S_j S_l = i [ \{ S_i, S_k \} - \delta_{ik} S_l(S+1)]\) (as usual the summation over repeated indices is understood).

\(^{29}\)Besides the spin-exchange collisional effect the classical magnetic field from the noble gas magnetization is included in the definition of \(\lambda\), as discussed in Section \(2.12\).
The rates $R_{se}^{en}$ and $R_{se}^{ne}$ are closely related; from the principle of detailed balance:

$$[A]R_{se}^{en} = [K]R_{se}^{ne}. $$

We stress that the above equations are correct for $K = S = 1/2$. In the more general case of arbitrary spins a modification of the above equations is required [97].

2.17 Quantum Optical States and Operators

We complete the theoretical background with a brief discussion of the quantum optical states and operators that are relevant to the work of this thesis. A detailed discussion on this topic can be found in [123, 130, 173, 76, 94], where a more formal derivation is provided which takes into account the transverse profile of the beam.

2.17.1 Electric field operator

We will consider monochromatic fields described by:

$$\mathbf{E} = \mathbf{E}^+ + \mathbf{E}^- $$

$$\mathbf{E}^+(l, t) = \sqrt{\frac{2\pi \hbar \omega}{A_b c}} \sum_j e^j \hat{a}^j(l, t) e^{-i\omega t + ikl} $$

$$\mathbf{E}^-(l, t) = \sqrt{\frac{2\pi \hbar \omega}{A_b c}} \sum_j \mathbf{e}^j \hat{a}^{j\dagger}(l, t) e^{i\omega t - ikl} $$

where the summation occurs over the polarization modes ($j = \pm 1$), $c$ is the speed of light, $e^j$ the polarization unit vector, $A_b$ the area of the beam, $\omega$ the light angular frequency, $k = \omega/c$, $l$ measures along the direction of propagation, $\hat{a}(l, t)$ and $\hat{a}^{\dagger}(l, t)$ are the lowering and raising operators which may vary (slowly compared to the exponential $e^{i\omega t - klt}$) with time and distance. With the normalization used in (2.180) and
the operators $\hat{a}^\dagger$, $\hat{a}$ satisfy the following equations (2.181):

$$\left[\hat{a}_m(t, l), \hat{a}_j^\dagger(t', l')\right] = \delta(t - t' \pm \frac{l - l'}{c}) \delta_{mj}$$  \hspace{1cm} (2.182)

$$\langle \hat{a}_j^\dagger(t, l) \hat{a}_j(t, l) \rangle = \Phi_j(t, l)$$  \hspace{1cm} (2.183)

$$\langle \hat{a}_j^\dagger(t, l) \hat{a}_j(t, l) \rangle = \Phi_j(t, l)$$  \hspace{1cm} (2.184)

with $\Phi_j$ being the photon flux (photons/sec) in the $j$ polarization mode, and the choice of sign in the delta function depends on the direction of light propagation.

Breaking up the beam into slices (modes) of (spatial) width $c \times \Delta t$ and defining Fock states for this mode $|n\rangle$, with $n = \Phi \times \Delta t$ (2.173), we get:

$$\hat{a} \sqrt{\Delta t} |n\rangle = \sqrt{n} |n - 1\rangle$$  \hspace{1cm} (2.185)

$$\hat{a}^\dagger \sqrt{\Delta t} |n\rangle = \sqrt{n + 1} |n + 1\rangle$$  \hspace{1cm} (2.186)

Under the approximation discussed in Section 2.8, the equation of motion for the lowering operator is:

$$\left[\frac{\partial}{\partial t} - \frac{1}{c} \frac{\partial}{\partial t} \right] \hat{a}_j(t, l) = \frac{i}{\hbar c} \left[ \hat{H}_{int}(t, l), \hat{a}_j(t, l) \right]$$  \hspace{1cm} (2.187)

$$\hat{H}_{int}(t, l) = - \sum_{j, m} \hat{E}_j(t, l) \hat{a}_{jm}(\omega, t, l) \hat{E}_{m}^+(t, l)$$  \hspace{1cm} (2.188)

where $\hat{a}_{jm}(t, l)$ is the polarizability (second rank) tensor described in 2.4.1. A similar equation holds for $\hat{a}^\dagger$.

2.17.2 Polarization Basis

In anticipation of the work presented in chapter 6 where the light is propagating in the $+x$ Cartesian direction, we will find convenient to define the spherical or circular
basis (irreducible representation of vectors) as:

\[ e_+ = -\frac{1}{\sqrt{2}} (e_y + ie_z) \]  \hspace{1cm} (2.189)

\[ e_- = \frac{1}{\sqrt{2}} (e_y - ie_z) \]  \hspace{1cm} (2.190)

The + and – indices refer to positive and negative spin respectively (or left and right
circular polarization). For the polarization analysis, it is useful to define basis vectors
e_y' and e_z' that form (with e_x) a Cartesian system rotated by 45° around the x axis
with respect to e_y and e_z:

\[ e_y' = \frac{1}{\sqrt{2}} (e_y + e_z) \]  \hspace{1cm} (2.191)

\[ e_z' = \frac{1}{\sqrt{2}} (-e_y + e_z) \]  \hspace{1cm} (2.192)

### 2.17.3 Stokes Representation

With the above bases conventions, the Stokes operators (which are the quantum
mechanical analogue of the classical Stokes parameters [27]) are defined:

\[ \hat{\Xi}_1 = \hat{a}_{+}^{\dagger} \hat{a}_{z'} - \hat{a}_{y'}^{\dagger} \hat{a}_{y'} \]  \hspace{1cm} (2.193)

\[ \hat{\Xi}_2 = \hat{a}_{+}^{\dagger} \hat{a}_{+} - \hat{a}_{-}^{\dagger} \hat{a}_{-} \]  \hspace{1cm} (2.194)

\[ \hat{\Xi}_3 = \hat{a}_{z}^{\dagger} \hat{a}_{z} - \hat{a}_{y}^{\dagger} \hat{a}_{y} \]  \hspace{1cm} (2.195)

\[ \hat{\Xi}_0 = \hat{a}_{z}^{\dagger} \hat{a}_{z} + \hat{a}_{y}^{\dagger} \hat{a}_{y} \]  \hspace{1cm} (2.196)

For notational clarity we dropped the explicit dependence on time and distance; it is
understood that the Stokes components are functions of t and l.

Clearly, \( \hat{\Xi}_0 \) measures the total photon flux (of both polarization modes); \( \hat{\Xi}_2 \) measures
the photon flux difference of the two polarization modes in the circular basis
and \( \hat{\Xi}_1 \) and \( \hat{\Xi}_3 \) measure the flux difference in linearly polarized bases.
Using equations (2.182) and (2.189)-(2.192) one can show after a bit of algebra:

\[
\left[ \hat{\xi}_j(t, l), \hat{\xi}_k(t', l) \right] = 2i \epsilon_{jkm} \hat{\xi}_m(t, l) \delta(t - t') \tag{2.197}
\]

\[
\left[ \hat{\xi}_j, \hat{\xi}_0 \right] = 0 \tag{2.198}
\]

where \( \epsilon_{jlm} \) is the Levi-Civita symbol and Einstein summation is assumed.

### 2.17.4 Coherent states

A coherent state of light \( |\alpha\rangle \) can be defined in terms of the Fock states of a particular polarization mode as:

\[
|\alpha\rangle = e^{-|\alpha|^2/2} \sum_{n=0}^{\infty} \frac{\alpha^n}{\sqrt{n!}} |n\rangle \tag{2.199}
\]

where \( \alpha \) is a complex number. It is straightforward to show that:

\[
\hat{a} \sqrt{\Delta t} |\alpha\rangle = \alpha |\alpha\rangle \tag{2.200}
\]

\[
\langle \alpha | \hat{a}^{\dagger} \sqrt{\Delta t} = \langle \alpha | \alpha^* \tag{2.201}
\]

\[
\langle \alpha | \hat{a}^{\dagger} \hat{a} |\alpha\rangle = \frac{|\alpha|^2}{\Delta t} \tag{2.202}
\]

Of particular interest in estimating quantum noise properties is the vacuum coherent field mode, for which \([76]\):

\[
\langle \hat{a}(t) \hat{a}^{\dagger}(t') \rangle = \delta(t - t') \tag{2.203}
\]

\[
\langle \hat{a}^{\dagger}(t) \hat{a}(t') \rangle = 0 \tag{2.204}
\]
Chapter 3

Comagnetometer Theory

In this chapter we give a theoretical treatment of K-\(^3\)He comagnetometer. The coupled spin dynamics are described within the Bloch formalism, and the steady state and dynamical behavior are studied. It is shown that the comagnetometer is insensitive to magnetic interactions but retains sensitivity to anomalous spin-coupling fields and gyroscopic motion.

3.1 Description of the Comagnetometer

In general, the comagnetometer consists of overlapping spin polarized ensembles. There have been developed various types of comagnetometers with different polarized atomic species that have been applied in gyroscopic applications and in the search of physics beyond the standard model (see \([\text{[160, 202, 19, 84, 115, 189, 17]}]\)). We will here describe the K-\(^3\)He comagnetometer which was used in the experiment of this thesis. We happily acknowledge that \([\text{[116]}]\) was the primary reference of the present analysis and an invaluable help for our understanding.

\(^1\)From now on we will use [TK] to refer to [116].
3.1.1 Dynamics of Overlapping Spin Ensembles

Although the more rigorous density matrix approach can be performed, we will use the simplified Bloch picture (Section 2.16) to describe the dynamics. For the $^3\text{He}$ (atomic species with spin 1/2) spin ensemble the Bloch and density matrix representations are equivalent. For K (of atomic spin $F = 2, 1$), the Bloch picture is only an approximation to the full density matrix equation; however, as was discussed previously (see Section 2.16), the Bloch equations (with the introduction of the appropriate phenomenological parameters) describe the evolution of the spin ensemble in spin-temperature equilibrium to a good approximation.

The dynamics of the spin ensembles are determined by magnetic and potentially anomalous, spin-coupling fields, and by the various relaxation and pumping mechanisms. The K spin experiences an environmental magnetic field $B$ and light-shift from the light (pump and probe beam). The pump and probe beam have mean photon spin $s_{pu}$ and $s_m$ (see Eq. (2.39)), and generate spin orientation with rates $R_{pu}$ and $R_m$ respectively. Diffusion to the walls and spin-destruction collisions happen at rate $R_{sd}$. Besides the spin destruction collisions, the effect of the $^3\text{He}$ spin ensemble on K can be separated into a relaxation/pumping spin-exchange mechanism with rate $R_{en}^se$ and a total effective magnetic field $\lambda M_n P_n$, where $P_n = \langle K \rangle / K$ is the nuclear polarization, $M_n = [\text{He}]\mu_{\text{He}}, [\text{He}]$ is the $^3\text{He}$ density and $\mu_B = -1.07455 \times 10^{-26}$ JT$^{-1}$ is the $^3\text{He}$ magnetic moment (see Section 2.12). The effective magnetic field $\lambda M_n P_n$ includes contribution from both the classical magnetic field due to (approximately uniform) spherical $^3\text{He}$ magnetization and the spin-exchange frequency shift described by Eq. (2.126); in [21] $\lambda$ for the K-$^3\text{He}$ spherically symmetric (as is approximately

---

$^2$The magnetic field generated by the macroscopic nuclear magnetization is not included in $B$.

$^3$As was discussed in Section 2.4.4, when the alkali atoms are at high pressure (the case of our experiment) the tensor interaction can be neglected in the light-atom interaction.
the case in the experiment) system was measured to be:

\[
\lambda = \frac{8\pi}{3}\kappa_0 = \frac{8\pi}{3}(1.85 + 0.00878T(\circ K))
\] (3.1)

where \( T \) is the temperature in Kelvin. Similarly, the nuclear spin is subjected to the environmental magnetic field \( \mathbf{B} \) and is destroyed with total rate \( R_{sd}^{n} \); from the noble gas perspective the presence of the alkali atoms results in a total effective magnetic field \( \lambda M_S P_e \), where \( P_e = \langle S \rangle / S \) is the electron polarization, and a pumping/relaxation spin-exchange mechanism with rate \( R_{se}^{ne} \). The pump and light fields do not interact with the nuclear spin to a very good approximation.

The coupled dynamics can then be approximated by the following set of Bloch equations:

\[
\frac{dP_e}{dt} = \frac{\gamma_e}{q} (\mathbf{B} + \lambda M_n \mathbf{P}_n + \mathbf{L} + \mathbf{b}_e) \times \mathbf{P}_e + \mathbf{\Omega} \times \mathbf{P}_e
\]

\[
+ \frac{1}{q} (R_{pu} \mathbf{s}_{pu} + R_{en}^{en} \mathbf{P}_n + R_{ms} \mathbf{s}_{pr} - R_{tot}^{e} \mathbf{P}_e)
\] (3.2)

\[
\frac{dP_n}{dt} = \gamma_n (\mathbf{B} + \lambda M_e \mathbf{P}_e + \mathbf{b}_n) \times \mathbf{P}_n + \mathbf{\Omega} \times \mathbf{P}_n
\]

\[
+ R_{se}^{ne} (\mathbf{P}_e - \mathbf{P}_n) - R_{sd}^{n} \mathbf{P}_n
\] (3.3)

Here, \( \gamma_e \) and \( \gamma_n \) are electron and \(^3\)He nuclear gyromagnetic ratio (which are defined to be positive numbers), \( q \) is the paramagnetic coefficient defined in (2.142) and is a function of the electron polarization. The above Bloch equations were written in the laboratory frame, where the non-inertial frame of reference introduces “inertial forces”; for the case of our experiment and for time scales relevant to the measurement process the non-inertial motion can be very well approximated by the uniform rotation of the Earth around its axis. The Hamiltonian that describes the dynamics of an arbitrary spin \( \mathbf{M} \) in a rotating frame with angular velocity \( \mathbf{\Omega} \) acquires an extra inertial term \( \mathbf{M} \cdot \mathbf{\Omega} \) \([124]\), which results in the terms \( \mathbf{\Omega} \times \mathbf{P}_e \) and \( \mathbf{\Omega} \times \mathbf{P}_n \) in (3.2) and (3.3).
In the equations above, $b_e$ and $b_n$ describe anomalous fields that couple to the electron spin and nuclear spin respectively according to the Hamiltonian:

$$H = g_S \mu_B S \cdot b_e - \frac{\mu_K}{K} S \cdot b_n$$

(3.4)

In general, there are theoretical predictions for various anomalous fields that couple to the spins of particles. Although the co-magnetometer of the experiment can in principle be used for various spin couplings [34], in this thesis we will present studies of the phenomenological Hamiltonian (3.4).

Despite their notational simplicity, the coupled dynamics (3.2) and (3.3) exhibit rich behavior. Following the literature of K-3He co-magnetometer, the z-axis is defined to be the nominal direction of the pump beam and the x-axis as the nominal direction of the probe beam. The probe beam (which to a very good approximation interacts only with the alkali atoms) is then nominally measuring the x-component of the electron polarization $P_e^x$. The longitudinal direction is then along the z-axis and the transverse axes are the x and y axes.

### 3.2 Steady State Solution

The steady state solution can be found by setting $\frac{dP^e}{dt} = \frac{dP^n}{dt} = 0$ and solving for $P^e$ and $P^n$. The resulting $6 \times 6$ system of equations is non-linear and an analytic solution is not known to the writer (and certainly even if it exists it will not be illuminating). A series of approximations make the solution more tractable, retaining the main features that determine up to high accuracy the behavior of the coupled dynamics.

First, it is a very good approximation to assume that the longitudinal components $P^e_z$ and $P^n_z$ are not affected by the presence of transverse components in the
(generalized) spin-coupling fields $(B, L, \Omega, b^e, b^n)$. Equivalently the angle $\theta$ of the polarization vectors $P^e$ and $P^n$ with respect to the z-axis is small enough so that $\cos \theta \approx 1$. Solving for $P^e_z$ and $P^n_z$ we find:

\[ P^e_z = \frac{R_{pu} s_{pu} (R_{sd} + R_{se}^e)}{R_{sd} (R_{pu} + R_{sd} + R_{se}^e) + R_{se}^e (R_{pu} + R_{sd})} \approx \frac{R_{pu} s_{pu}}{R_{sd} + R_{se}^e + R_{pu}} \]  

(3.5)

\[ P^n_z = \frac{R_{pu} s_{pu} R_{ne}^e}{R_{sd} (R_{pu} + R_{sd} + R_{se}^e) + R_{se}^e (R_{pu} + R_{sd} + R_{pu})} \approx P^n_z \approx P^e_z \]  

(3.6)

The approximate forms result from the condition: $R_{se}^n / R_{sd}^n \ll 1$, see Table 3.1. Since $P^e_x, P^e_y \ll P^e_z$ we can make the approximation $q(P^e) \approx q(P^e_z)$.

The resulting $4 \times 4$ system of equations for the transverse components of electron and nuclear polarization becomes linear and the standard methods of linear algebra can be performed to find the solution. It is convenient to write the Bloch equations as a system of equations for $\tilde{P}^e = P^e_x + iP^e_y$ and $\tilde{P}^n = P^n_x + iP^n_y$ in the matrix form:

\[ 0 = A \begin{pmatrix} \tilde{P}^e \\ \tilde{P}^n \end{pmatrix} + C \]  

(3.7)

The matrices $A$ and $C$ are:

\[ A = \begin{pmatrix} -\tilde{R}_{tot}^e + i\omega_e & \tilde{R}_{tot}^e - i\omega_en \\ R_{se}^e - i\omega_{ne} & -\tilde{R}_{tot}^n + i\omega_n \end{pmatrix} \]  

(3.8)

\[ C = \begin{pmatrix} \tilde{b}_y^e - i\tilde{b}_x^e \\ \tilde{b}_y^n - i\tilde{b}_x^n \end{pmatrix} \]  

(3.9)

\[ ^4 \text{The angle for } P^e \text{ and } P^n \text{ does not need be the same (in fact it is unlikely to be the same).} \]
where:

\[
\omega_e = \frac{\gamma_e (B_z + \lambda M_n P^n_z + L_z + b^e_z)}{q} + \Omega_z
\]  
(3.10)

\[
\omega_n = \gamma_n (B_z + \lambda M_e P^e_z + b^n_z) + \Omega_z
\]  
(3.11)

\[
\tilde{R}_{tot}^e = \frac{R_{tot}^e}{q} = \frac{R_{se}^e + R_{pa} + R_{sd} + R_{pr}}{q}
\]  
(3.12)

\[
R_{tot}^n = R_{se}^n + R_{sd}^n
\]  
(3.13)

\[
\tilde{b}_y^e = P_z^e \left( \gamma_e \frac{b^e_y + L_y + B_y}{q} + \Omega_y \right)
\]  
(3.14)

\[
\tilde{b}_x^e = P_z^e \left( \gamma_e \frac{b^e_x + L_x + B_x}{q} + \Omega_x \right)
\]  
(3.15)

\[
\tilde{b}_y^n = P_z^n \left[ \gamma_n (b^n_y + B_y) + \Omega_y \right]
\]  
(3.16)

\[
\tilde{b}_x^n = P_z^n \left[ \gamma_n (b^n_x + B_x) + \Omega_x \right]
\]  
(3.17)

\[
\tilde{R}_{se}^n = R_{se}^n / q
\]  
(3.18)

\[
\omega_{en} = \gamma_e \lambda M_n P^e_z / q
\]  
(3.19)

\[
\omega_{ne} = \gamma_n \lambda M_e P^n_z
\]  
(3.20)

The experimentally measured signal \((P^e_x)\) is the real value of \(\tilde{P}^e\). Although an analytic solution can be written, the form is too complicated. However, the different importance of the various terms appearing in the equations allows us to proceed to further approximations for a physically transparent solution.

As will be described in the following sections, the longitudinal field at the point of co-magnetometer operation can be written as:

\[
B_z = B_c + \delta B_z
\]  
(3.21)

\[
B_c = -\lambda M^n P^n_z - \lambda M^e P^e_z - \Omega_z / \gamma_n
\]  
(3.22)
with $|\delta B_z| \ll |B_c|$. For the experimentally realized comagnetometers $M^n P^n_z \gg M^e P^e_z \gg \Omega_z/\gamma_n$ and Eq. (3.22) can be approximated by:

$$B_c \approx -\lambda M^n P^n_z$$  \hspace{1cm} (3.23)

In addition: $B_c \gg (L_z, b^e_z, b^n_z)$.

We emphasize that $B_c$ depends on $\Omega_z$, contrary to what is claimed in [TK]. We define $B_c$ (the compensation point) as the total magnetic field along the pump direction ($B_z$) that minimizes (in fact zeroes) the sensitivity of the measured signal on the magnetic field perpendicular to pump and probe ($B_y$). Mathematically, this is expressed with the relation: $B_z = B_c \Rightarrow \partial P_m/\partial B_y = 0$, where $P_m$ is the measured electron polarization from the probe beam.

A major simplification in the solution arises from neglecting the contribution of the pumping/relaxation part of the spin-exchange interaction between the alkali and the noble gas atoms in the transverse spin components. From Table 3.1 $\tilde{R}^{en}_{se} \ll \omega_{en}$ and $\tilde{R}^{ne}_{se} \ll \omega_{ne}$ so that we can set $\tilde{R}^{en}_{se} \approx R^{ne}_{se} \approx 0$ in (3.8) (but without setting $R^{en}_{se} \approx 0$ in the definition of $R^{e}_{tot}$). Physically, the approximation implies that as far as the spin-exchange collisions are concerned the dynamics are mainly determined by the effective magnetic field experienced by the spins. Furthermore, $R^{n}_{tot} \ll \omega_{en}$ and the dependence of $A$ on $R^{n}_{tot}$ can be dropped.

The simplified solution for $P^e_x$ (real part of $\tilde{P}^e$) can be written compactly:

$$P^e_x = R^{e}_{tot} \left( \tilde{v}^e_x + \frac{\omega_{en} \tilde{v}^e_y}{\omega_n} \right) + \left( \tilde{v}^e_y + \frac{\omega_{en} \tilde{v}^e_x}{\omega_n} \right) \left( \omega_e - \frac{\omega_{en} \omega_{ne}}{\omega_n} \right)$$  \hspace{1cm} (3.24)

\footnote{As was explained in Sections 2.12 and 2.16.4 the terms $\lambda M^n P^n_z$ and $\lambda M_e P^e_z$ come to a large extent from the spin-exchange collisions. In the described approximation this contribution of the spin-exchange mechanism is not ignored.}
If the probe beam forms an angle $\alpha$ with the x-axis ($\alpha \ll 1$) then the measured polarization is:

$$P_m = \cos \alpha P^e_x + \sin \alpha P^e_z \approx P^e_x + \alpha P^e_z$$  \hspace{1cm} (3.25)

Combining Equations (3.10)-(3.22) and (3.24)-(3.25) one can show that to lowest order in small quantities:

$$P_m \approx \frac{P^e_z \gamma e R^e_{\text{tot}}}{R^e_{\text{tot}}^2 + \gamma_e^2 \left( \delta B_z + L_z - \bar{\Omega}_z \right)^2}$$

$$\times \left[ b_y^e - \left( 1 - \frac{\delta B_z}{B_c} \right) b_y^n - \left( \frac{1}{\gamma_{\text{eff}}} - \frac{\delta B_z}{\gamma_n B_c} \right) \Omega_y + L_y \right]$$

$$+ \frac{\gamma_e R^e_{\text{tot}}}{R^e_{\text{tot}}} \left( \delta B_z + L_z - \bar{\Omega}_z \right) \left( b_x^e - b_x^n + L_x - \frac{\Omega_x}{\gamma_{\text{eff}}} \right)$$

$$+ \frac{\delta B_z}{B_c} B_y + \frac{\gamma_e \delta B_z}{B_c R^e_{\text{tot}}} \left( \delta B_z + L_z - \bar{\Omega}_z \right) B_x$$

$$+ \frac{R_m}{\gamma_e P^e_z s_m} + \alpha P^e_z$$  \hspace{1cm} (3.26)

In the above equation, we used the definitions:

$$\gamma_{\text{eff}} = \frac{\gamma e \gamma n}{\gamma e - q \gamma n}$$  \hspace{1cm} (3.27)

$$\bar{\Omega}_z = \frac{\Omega_z}{\gamma_{\text{eff}}}$$  \hspace{1cm} (3.28)

and made the approximations:

$$(b_x^e, b_y^n) + \bar{\Omega}_z \approx \bar{\Omega}_z$$  \hspace{1cm} (3.29)

$$B_c + \delta B_z + \lambda M_e P^e_x + \Omega_z / \gamma_n \approx B_c$$  \hspace{1cm} (3.30)
3.2.1 Refinements in steady state signal

Here, we briefly summarize the main corrections to Eq. (3.26). Including the effect of nonzero $R_{se}^n$, $R_{se}^{ne}$, $R_{tot}^n$, after a bit of algebra, we have:

$$P_m \approx \frac{P_z^e \gamma_e R_{tot}^e}{R_{tot}^e + \gamma_c^2 \left[ \delta B_z (1 + \mathcal{K}) + L_z - \bar{\Omega}_z \right]^2} \times \left\{ b_y^e - \left( 1 - \frac{\delta B_z}{B_c} \right) b_y^n - \left( \frac{1}{\gamma_{eff}} - \frac{\delta B_z}{\gamma_n B_c} \right) \Omega_y + L_y + \frac{\delta B_z}{B_c} B_y + \frac{R_m}{\gamma_e P_{ez}^e} \right\}$$

$$\times \left[ 1 - C_{se}^{ne} - D_{se}^{en} - F_{sd}^n + O(10^{-6}) \right]$$

$$+ \frac{\gamma_e}{R_{tot}^e} \left[ \delta B_z (1 + \mathcal{K}) + L_z - \bar{\Omega}_z \right] (1 - 2D_{se}^{en} - 2F_{sd}^n) (b_x^e + L_x)$$

$$- \frac{\gamma_e}{R_{tot}^e} \left[ (1 - \frac{\delta B_z}{B_c} - 2C_{se}^{ne}) \left( \delta B_z + L_z - \bar{\Omega}_z \right) + \delta B_z \right]$$

$$+ \left( 1 - \frac{\delta B_z}{B_c} \right) R_{tot}^e P_{tot}^n + \left( 1 - \frac{\delta B_z}{B_c} \right) R_{tot}^e R_{tot}^{en} \right] b_x^n$$

$$+ \left[ \frac{\gamma_e \delta B_z}{B_c R_{tot}^e} \left( \delta B_z + L_z - \bar{\Omega}_z \right) + \left( 1 - \frac{\delta B_z}{B_c} \right) \frac{R_{se}^e}{\gamma_e \lambda M^n P_z^e} \right]$$

$$- \frac{R_{tot}^n}{\gamma_n \lambda M^n P_z^n} \left( 1 - \frac{\delta B_z}{B_c} \right) B_x$$

$$+ \left[ \frac{\gamma_e}{R_{tot}^e} \left( \delta B_z + L_z - \bar{\Omega}_z \right) \left( \frac{1}{\gamma_{eff}} + \frac{\delta B_z}{\gamma_n B_c} \right) - \frac{\gamma_e \delta B_z}{\gamma_{eff} R_{tot}^e} \mathcal{K} \right]$$

$$+ 2 \left( \frac{\gamma_{e} C_{se}^{ne}}{\gamma_{eff} R_{tot}^e} - \frac{q D_{se}^{en}}{R_{tot}^e} - \frac{q F_{sd}^n}{R_{tot}^e} \right) \left( \delta B_z + L_z - \bar{\Omega}_z \right)$$

$$- \frac{R_{se}^{en}}{\gamma_e \gamma_n \lambda M^n P_z^e} \left( 1 - \frac{\delta B_z}{B_c} \right) - \frac{R_{tot}^n}{\gamma_n \lambda M^n P_z^n} \left( 1 + \frac{2 \delta B_z}{B_c} \right) \right\} \Omega_x$$

$$+ \alpha P_z^e$$

(3.31)
where we used:

\[
C_{se}^{en} = \frac{P_n^{en} R_{se}^{en}}{P_z^{en} R_{tot}^{en}} \sim 10^{-3} \quad (3.32)
\]

\[
C_{se}^{me} = \frac{\gamma_e P_e^{en} R_{se}^{en}}{\gamma_n P_n^{en} R_{tot}^{en}} \sim 10^{-3} \quad (3.33)
\]

\[
D_{se}^{en} = \frac{M_e^{en} R_{se}^{en}}{M_n^{en} R_{tot}^{en}} \sim 10^{-6} \quad (3.34)
\]

\[
F_{sd}^{en} = \frac{\gamma_e P_e^{en} M_e^{en} R_{tot}^{en}}{\gamma_n P_n^{en} M_n^{en} R_{tot}^{en}} \sim 10^{-6} \quad (3.35)
\]

\[
K = \frac{M_e^{en} P_e^{en}}{M_n^{en} P_n^{en}} \sim 10^{-3} \quad (3.36)
\]

Equation (3.22) is modified to:

\[
B_c = -\lambda M_n P_n^{en} - \lambda M_e P_e^{en} - \Omega_z/\gamma_n + \left( \frac{P_n^{en} R_{se}^{en}}{P_z^{en} \Delta \omega_e} + \frac{\gamma_e \Delta \omega_e}{\gamma_n \Delta \omega_e} \right) \left( L_z - \tilde{\Omega}_z \right) \quad (3.37)
\]

\[
\approx -\lambda M_n P_n^{en} \left( 1 + \frac{\lambda M_e P_e^{en} + \Omega_z/\gamma_n}{\lambda M_n P_n^{en}} + O(10^{-9}) \right) \quad (3.38)
\]

In the limit of zero \( \delta B_z \), \( L \), \( \alpha \) and \( s_m \), and also dropping the terms proportional to \( D_{se}^{en} \) and \( F_{sd}^{en} \) the steady state signal is:

\[
P_m \approx \frac{P_e^{en} R_{tot}^{en}}{R_{tot}^{en}^2 + \gamma_e^2 \Omega_z^2}
\times \left\{ \left( \frac{b_e^y - b_n^y}{\gamma_{eff}} \right) \left( 1 - C_{se}^{me} \right) - \frac{\gamma_e \tilde{\Omega}_z}{R_{tot}^{en}} \left( 1 - 2C_{se}^{me} \right) \left( b_e^x - b_n^x \right) \right. \\
+ \left( \frac{R_n^{en}}{\gamma_n B_c} + \frac{\gamma_e P_{se}^{en}}{\gamma_e B_c} \right) b_n^y - \left( \frac{R_{se}^{en}}{\gamma_e \lambda M_n P_n^{en}} + \frac{R_n^{en}}{\gamma_n \lambda M_n P_n^{en}} \right) B_x \\
+ \left[ -\frac{\gamma_e}{\gamma_{eff} \Omega_z} \left( 1 + 2C_{se}^{me} \right) \tilde{\Omega}_z - \frac{1}{\gamma_n \lambda M_n} \left( \frac{R_{se}^{en}}{\gamma_e P_e^{en}} + \frac{R_{tot}^{en}}{\gamma_n P_n^{en}} \right) \right] \Omega_x \left\} \right.
\]

(3.39)
3.2.2 Sensitivity of the comagnetometer

It is instructive to summarize the dependence of the measured co-magnetometer signal on various parameters: magnetic and anomalous spin-coupling fields, light-shifts, uniform rotation, probe circular polarization and angle $\alpha$. In the following equations we keep only the main contributions.

**Anomalous field**:

\[ S(b_{y}^{e}) \approx P_{e}^{e} \frac{\gamma_{e} b_{y}^{e}}{R_{tot}^{e}} \]  \hspace{1cm} (3.40)

\[ S(b_{y}^{n}) \approx -P_{e}^{e} \frac{\gamma_{e} b_{y}^{n}}{R_{tot}^{e}} \]  \hspace{1cm} (3.41)

\[ S(b_{x}^{e}) \approx P_{e}^{e} \frac{\gamma_{e} (\delta B_{z} + L_{z} - \tilde{\Omega}_{z})}{R_{tot}^{e}} \gamma_{e} b_{x}^{e} \]  \hspace{1cm} (3.42)

\[ S(b_{x}^{n}) \approx -P_{e}^{e} \left[ \frac{\gamma_{e} (\delta B_{z} + L_{z} - \tilde{\Omega}_{z})}{R_{tot}^{e}} + \frac{R_{n}^{m} + R_{se}^{m} P_{n}^{m}}{\gamma_{n} B_{c} P_{n}^{e}} \right] \gamma_{e} b_{x}^{n} \]  \hspace{1cm} (3.43)

\[ S(b_{z}^{e}) \approx P_{e}^{e} \left( \frac{\gamma_{e} L_{x}}{R_{tot}^{e}} + \frac{\gamma_{e} \Omega_{x}}{\gamma_{n} R_{tot}^{e}} \right) \gamma_{e} b_{z}^{e} \]  \hspace{1cm} (3.44)

\[ S(b_{z}^{n}) \approx -P_{e}^{e} \left( \frac{\gamma_{e} L_{x}}{R_{tot}^{e}} + \frac{\gamma_{e} \Omega_{x}}{\gamma_{n} R_{tot}^{e}} \right) \gamma_{e} b_{z}^{n} \]  \hspace{1cm} (3.45)

**Magnetic field**:

\[ S(B_{y}) \approx P_{e}^{e} \frac{\delta B_{z} \gamma_{e} B_{y}}{B_{c} R_{tot}^{e}} \]  \hspace{1cm} (3.46)

\[ S(B_{x}) \approx P_{e}^{e} \left[ \frac{\gamma_{e} (\delta B_{z} + L_{z} - \tilde{\Omega}_{z})}{R_{tot}^{e}} \frac{\delta B_{z}}{B_{c}} - \frac{R_{n}^{m}}{\gamma_{n} B_{c}} - \frac{R_{se}^{m} P_{n}^{m}}{\gamma_{e} B_{c} P_{n}^{e}} \right] \gamma_{e} B_{x} \]  \hspace{1cm} (3.47)

\[ S(\delta B_{z}) \approx P_{e}^{e} \left( \frac{B_{y}}{B_{c}} + \frac{\gamma_{e} L_{x}}{\gamma_{n} B_{c}} + \frac{\Omega_{y}}{\gamma_{n} B_{c}} + \frac{\gamma_{e} \Omega_{x}}{\gamma_{eff} R_{tot}^{e}} \right) \gamma_{e} \delta B_{z} \]  \hspace{1cm} (3.48)

\(^6\)For the signal dependence on $b_{y}^{e}$ and $b_{y}^{n}$ we relaxed the approximation $(b_{y}^{e}, b_{y}^{n}) \ll \tilde{\Omega}_{z}$. The dependence on $b_{x}^{e}$ and $b_{x}^{n}$ can be found from (3.31) by the substitutions: $\Omega_{z} \rightarrow \Omega_{z} + \gamma_{n} b_{z}^{n}$ and $L_{z} \rightarrow L_{z} + b_{x}^{e} - (\frac{q \gamma_{n}}{\gamma_{e}}) \tilde{\Omega}_{z}$.
Lightshift:

\[ S(L_y) \approx P_z^e \frac{\gamma_e L_y}{R_{tot}^e} \quad (3.49) \]
\[ S(L_x) \approx P_z^e \frac{\gamma_e \left( \delta B_z + L_z - \tilde{\Omega}_z \right)}{R_{tot}^e} \frac{\gamma_e L_z}{R_{tot}^e} \quad (3.50) \]
\[ S(L_z) \approx -P_z^e \left( \frac{\gamma_e L_z}{R_{tot}^e} + \frac{\gamma_e}{\gamma_{eff}} \frac{\Omega_z}{R_{tot}^e} \right) \frac{\gamma_e L_z}{R_{tot}^e} \quad (3.51) \]
\[ S(L_z) \approx -P_z^e \left( \frac{\gamma_e L_z}{R_{tot}^e} + \frac{\gamma_e}{\gamma_{eff}} \frac{\Omega_z}{R_{tot}^e} \right) \frac{\gamma_e L_z}{R_{tot}^e} \quad (3.52) \]

Uniform Rotation:

\[ S(\Omega_y) \approx P_z^e \frac{\gamma_e \Omega_y}{\gamma_{eff} R_{tot}^e} \sim \quad (3.53) \]
\[ S(\Omega_x) \approx P_z^e \left[ \frac{\gamma_e \left( \delta B_z + L_z - \tilde{\Omega}_z \right)}{R_{tot}^e} \right] - \frac{P_n^e \gamma_{en}}{P_z^e \gamma_e B_c - \gamma_{en} B_c} \frac{\gamma_e \Omega_x}{\gamma_{eff} R_{tot}^e} \quad (3.54) \]
\[ S(\Omega_z) \approx -P_z^e \left( \frac{\gamma_e L_x}{R_{tot}^e} + \frac{\gamma_e}{\gamma_{eff}} \frac{\Omega_x}{R_{tot}^e} \right) \frac{\gamma_e \Omega_z}{\gamma_{eff} R_{tot}^e} \quad (3.55) \]

Probe Circular Polarization:

\[ S(s_m) \approx \frac{R_m}{R_{tot}} s_m \quad (3.56) \]

Probe-probe angle:

\[ S(\alpha) \approx P_z^e \alpha \quad (3.57) \]

From Equations (3.40)-(3.55) it can be seen that the sensitivity of the comagnetometer signal to magnetic fields (in all directions) is suppressed compared to the anomalous field in the y-direction (direction perpendicular to the plane formed by the pump and probe beams). Similarly, the sensitivity to the x and z components of the anomalous spin coupling field, light-shift and uniform rotation is reduced compared to the respective y components.
<table>
<thead>
<tr>
<th>Variable</th>
<th>Notation</th>
<th>Typical Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^3$He gyromagnetic ratio</td>
<td>$\gamma_n$</td>
<td>3.24 kHz/G</td>
</tr>
<tr>
<td>electron gyromagnetic ratio</td>
<td>$\gamma_e$</td>
<td>2.8 MHz/G</td>
</tr>
<tr>
<td>K slowing down factor in SERF</td>
<td>$q$</td>
<td>5.2</td>
</tr>
<tr>
<td>Temperature</td>
<td>$T$</td>
<td>166°C</td>
</tr>
<tr>
<td>K number density</td>
<td>$n_e$</td>
<td>$3.1 \times 10^{13}$ cm$^{-3}$</td>
</tr>
<tr>
<td>$^3$He number density</td>
<td>$n_{He}$</td>
<td>$3.0 \times 10^{20}$ cm$^{-3}$ (11.2 amg)</td>
</tr>
<tr>
<td>K polarization field</td>
<td>$\lambda M^e P^e_z$</td>
<td>7 $\mu$G</td>
</tr>
<tr>
<td>$^3$He polarization field</td>
<td>$\lambda M^e P^m_z$</td>
<td>5.3 mG</td>
</tr>
<tr>
<td>K probe beam pumping rate</td>
<td>$R_m$</td>
<td>82 sec$^{-1}$</td>
</tr>
<tr>
<td>K-$^3$He spin-exchange rate</td>
<td>$R^m_{se}$</td>
<td>17 sec$^{-1}$</td>
</tr>
<tr>
<td>K-K spin destruction rate</td>
<td>$R^e_{KK}$</td>
<td>22 sec$^{-1}$</td>
</tr>
<tr>
<td>K-He spin destruction rate</td>
<td>$R^e_{KHe}$</td>
<td>44 sec$^{-1}$</td>
</tr>
<tr>
<td>K total spin destruction rate</td>
<td>$R^e_{sd}$</td>
<td>165 sec$^{-1}$</td>
</tr>
<tr>
<td>K pumping rate</td>
<td>$R^e_{pu}$</td>
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<tr>
<td>K total relaxation rate</td>
<td>$R^e_{tot}$</td>
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<tr>
<td>$^3$He-K spin-exchange rate</td>
<td>$R^m_{se}$</td>
<td>$1.8 \times 10^{-6}$ sec$^{-1}$</td>
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<tr>
<td>$^3$He spin destruction rate</td>
<td>$R^m_{sd}$</td>
<td>$1/11.7$ h$^{-1}$</td>
</tr>
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<td>Pump photon spin</td>
<td>$(s^pu_x, s^pu_y, s^pu_z)$</td>
<td>$(10^{-4}, 10^{-4}, 0.99)$</td>
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<tr>
<td>Probe photon spin</td>
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<td>K polarization</td>
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</tr>
<tr>
<td>$^3$He polarization</td>
<td>$P^m_n$</td>
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<tr>
<td>Applied compensation field</td>
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<tr>
<td>Magnetic field offsets</td>
<td>$B_x, B_y, \delta B_z$</td>
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<tr>
<td>Light-shifts</td>
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</tr>
<tr>
<td>Pump-probe angle</td>
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<td>$&lt;10$ $\mu$rad</td>
</tr>
<tr>
<td>Gyroscopic Signal</td>
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<tr>
<td>Anomalous fields</td>
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<td>Probe Area</td>
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</tr>
<tr>
<td>Probe Power</td>
<td>$P_m$</td>
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<tr>
<td>Probe Detuning</td>
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</tr>
<tr>
<td>Cell Diameter</td>
<td>$d_c$</td>
<td>2.5 cm</td>
</tr>
</tbody>
</table>

Table 3.1: Typical values relevant to the thesis experiment of the parameters appearing in the Bloch Equations (see Sections 4.3 and B.1).
3.2.3 An intuitive model

The simplified steady state solution (3.26) can be understood as the response to spin coupling fields of K in the SERF regime (see Section 2.16.2); in fact this is how the steady state solution to the Bloch equations was derived in [TK] and in [34]. Without going into a detailed mathematical derivation based on this picture, an intuitive understanding of the magnetic field insensitivity can be developed from Fig. 3.1. In the comagnetometer operation point, the noble gas magnetization (including the contribution from the spin-exchange frequency shift) cancels (approximately) the applied magnetic field in the z direction (see Fig. 3.1(a)), so that alkali metal atoms are in the SERF regime where the sensitivity to spin-coupling fields is (in principle) large. For magnetic transverse excitations that happen adiabatically (slowly with respect to the precession time) $^3$He follows the direction of the total magnetic field and the transverse magnetic field that K experiences is reduced from the noble gas magnetization, see Fig. 3.1(b): this reduction is more effective as the ratio of transverse magnetic perturbation to effective $^3$He magnetic field (or equivalently the holding field in the compensation point) becomes smaller. Note that although the nuclear magnetization does not compensate for longitudinal perturbations, K (being in the SERF regime) only exhibits second order sensitivity to fluctuations in the $B_z$. The noble gas does not react to $b^n$ and the alkali metal responds to first order in the y-component of the field (see Section 2.16.2); for nucleon coupling field $b^n$ $^3$He transverse orientation is left uncompensated to K (which does not experience $b^n$) and the magnetic field of the noble gas affects the signal. Similar arguments hold for the gyroscopic and light-shift signal, and in general to any field that couples to electron and nuclear spin in a manner different than magnetic field.
Figure 3.1: (Adapted from [TK]) The comagnetometer is insensitive to magnetic fields. (a) From the K perspective, the $^3$He magnetization (pointing opposite to the nuclear spin) cancels the externally applied magnetic field. (b) The noble gas magnetization responds to a transverse magnetic field perturbation and adiabatically follows the total field, canceling to first order in $|B_t|/|B_z|$ the magnetic field experienced by K.

3.3 Dynamic Response

In the previous sections we derived the steady state solution of the K-$^3$He comagnetometer, which does not contain information about the dynamic response of the system. The dynamics are most conveniently studied when writing the equations in the matrix form:

$$\frac{d}{dt} \mathbf{P} = \mathbf{A} \cdot \mathbf{P} + \mathbf{C}$$  \hspace{1cm} (3.58)

where $\mathbf{A}$ and $\mathbf{C}$ were defined in (3.8) and (3.9) respectively, and the vector $\mathbf{P}$ is:

$$\mathbf{P} = \begin{pmatrix} \tilde{P}^e \\ \tilde{P}^n \end{pmatrix} = \begin{pmatrix} P_x^e + i P_y^e \\ P_x^n + i P_y^n \end{pmatrix}$$  \hspace{1cm} (3.59)

The formal solution to Eq. (3.59) is:

$$\mathbf{P} = \mathbf{A}^{-1} \mathbf{C} + \sum_{i=1}^{2} \exp(\lambda_i t) \mathbf{A}_i$$  \hspace{1cm} (3.60)
where $A_i$ is the Frobenius covariant of $A$ corresponding to the eigenvalue $\lambda_i$. The eigenvalues $\lambda_i$ are complex numbers that determine the dynamical behavior the system. The two eigenvalues are:

$$\lambda_{1,2} = -\frac{R^e_{\text{tot}} + R^n_{\text{tot}}}{2} + i\frac{\omega_e + \omega_n}{2} \pm \frac{1}{2} \sqrt{x_r + iy_i}$$ (3.61)

with:

$$x_r = \left(\tilde{R}^e_{\text{tot}} - R^n_{\text{tot}}\right)^2 - 4\omega_en\omega_{ne} - (\omega_e - \omega_n)^2 + 4\tilde{R}_{se}^n R_{ne}^n$$ (3.62)

$$y_i = -2\left(\tilde{R}^e_{\text{tot}} - R^n_{\text{tot}}\right)(\omega_e - \omega_n) - 4\left(R_{se}^n\omega_{en} + \tilde{R}_{se}^n\omega_{ne}\right)$$ (3.63)

The general dynamical solution contains two separate oscillations with different frequencies (imaginary part of the eigenvalue) and decay rates (real part of the eigenvalue) respectively that depend on the $B_z$ field (through $\omega_e$ and $\omega_n$). The square root in (3.61) can be written:

$$\kappa = \sqrt{x_r + iy_i} = \frac{1}{\sqrt{2}} \left(\sqrt{\sqrt{x_r^2 + y_i^2} + x_r + \text{Sign}[y_i]i\sqrt{x_r^2 + y_i^2 - x_r}}\right)$$ (3.64)

For large $B_z$ such that $B_z \gg (|\lambda M^n P^n_z|, |\lambda M^e P^n_e|, \tilde{R}^e_{\text{tot}}/\gamma_n, R^n_{\text{tot}}/\gamma_n)$ the eigenvalues can be approximated (keeping the most significant terms) to be:

$$\lambda_{1,B\infty} = -\tilde{R}^e_{\text{tot}} + i\gamma_e B_z/q$$ (3.65)

$$\lambda_{2,B\infty} = -R^n_{\text{tot}} + i\gamma_n B_z$$ (3.66)

The two eigenvalues correspond to the decoupled oscillatory motions of alkali atoms and noble gas independent of each other. It is interesting to find the $B_z$ field that results in the fastest approach to equilibrium\footnote{We examine the longer lived eigenmodes as a function of the longitudinal magnetic field.} that is, the value of $B_z$ that maximizes...
\[
\min ||\text{Real}[\lambda_1, \lambda_2]|| \text{ or equivalently minimizes the real part of the square root in (3.61).}
\]

Depending on the sign of:

\[
x_{r0} = \left( \tilde{R}_{tot}^e - R_{tot}^n \right)^2 - 4\omega_{en}\omega_{ne} - \frac{4 \left( R_{se}^e\omega_{en} + \tilde{R}_{se}^e\omega_{ne} \right)^2}{\left( \tilde{R}_{tot}^e - R_{tot}^n \right)^2} + 4\tilde{R}_{se}^e R_{se}^n
\]

\[
\approx \left( \tilde{R}_{tot}^e - R_{tot}^n \right)^2 - 4\omega_{en}\omega_{ne}
\]

(3.67)

(3.68)

the condition for the fastest decay is:

\[
\omega_e - \omega_n \approx \begin{cases} 
\frac{-2(R_{se}^e\omega_{en} + \tilde{R}_{se}^e\omega_{ne})}{\tilde{R}_{tot}^e + R_{tot}^n}, & x_{r0} < 0 \\
\frac{-2(R_{se}^e\omega_{en} + \tilde{R}_{se}^e\omega_{ne}) \left( \tilde{R}_{tot}^e + R_{tot}^n \right)^2}{4\omega_{en}\omega_{ne}}, & x_{r0} \geq 0
\end{cases}
\]

(3.69)

For an arbitrary longitudinal magnetic field \(B_z\) the effect of \(\kappa\) on the decay rate to equilibrium can be found when comparing \(\text{Real}[\kappa]\) with \(\tilde{R}_{tot}^e + R_{tot}^n \approx \tilde{R}_{tot}^e\). For small values of \(K_r = \text{Real}[\kappa]/\tilde{R}_{tot}^e\) the longer-lived oscillation (eigenvalue in (3.61) with the smallest absolute value of the real part) decays at a rate on the order of \(\tilde{R}_{tot}^e/2\), while for values of \(K_r\) approaching unity, the longer-lived oscillation continues for timescales significantly larger than \(1/R_{tot}^e\). Dropping the (small) terms proportional to \(R_{se}^e, R_{se}^n\) and \(R_{tot}^n\) we can write:

\[
K_r = \frac{\text{Real}[\kappa]}{\tilde{R}_{tot}^e} \approx \frac{1}{\sqrt{2}} \sqrt{1 - s^2 - \eta + \sqrt{4s^2 + (1 - s^2 - \eta)^2}}
\]

(3.70)

where:

\[
s = \frac{\omega_e - \omega_n}{\tilde{R}_{tot}^e}
\]

(3.71)

\[
\eta = \frac{4\omega_{en}\omega_{ne}}{\tilde{R}_{tot}^e} = \frac{4q\lambda^2 M^e M^n P_z^e P_z^n}{\tilde{R}_{tot}^e}
\]

(3.72)
The parameter $\eta$ depends on the particular implementation of the comagnetometer (electron and nuclear polarization and density, gyromagnetic ratio of the noble gas) and is always a positive number, while $s$ is a function of $B_z$ and can be tuned by changing the applied longitudinal magnetic field. In Fig. 3.2 we plot the slowest decay rate as a function of $s$ for different parameters $\eta$. For a given value of $s$, the decay to equilibrium is faster the larger the value of $\eta$ is. In practice, large $\eta$ can be realized with high density of alkali atoms and noble gas.

After a bit of algebra, one can show that in the limit $\omega_e/R_{\text{tot}}^e \ll 1$ (condition which is realized in the thesis experiment) the slow relaxation rate ($r_d$) and the corresponding eigenfrequency $\omega_r$ can be approximated to be:

$$r_d \approx \frac{R_{\text{tot}}^e \omega_e \omega_n}{R_{\text{tot}}^e + \omega_n^2}$$  \hspace{1cm} (3.73)

$$\omega_r \approx \omega_n + \frac{\omega_e \omega_n}{R_{\text{tot}}^e + \omega_n^2}$$  \hspace{1cm} (3.74)

The alkali and noble gas density affect also the parameter $s$. One has to do a more careful analysis, but in general, to within certain limits, at the compensation point larger densities result in faster decay.

---

8The alkali and noble gas density affect also the parameter $s$. One has to do a more careful analysis, but in general, to within certain limits, at the compensation point larger densities result in faster decay.
It is an essential aspect of the comagnetometer that at the operation point, where the field along the pump direction is close to the compensation point ($B_z = B_c + \delta B_z$), the decay of the coupled ensemble is mainly determined by the alkali relaxation, resulting in decay timescales typically on the order of a few hundreds msec. This allows the application of various methods that depend on the quasi-static response to perturbations (see Section 4.2).

### 3.4 AC Response to Transverse Magnetic Fields

Equations (3.26) and (3.31) describe the response to dc (zero frequency) fields. Here, we explore the steady state response to an ac transverse magnetic field. Applying a field $\mathbf{B} = (B_0 x \hat{x} + B_0 y \hat{y}) e^{-i\omega t}$ of angular frequency $\omega$, the steady state (after the transients have decayed) of the coupled ensemble is:

$$-i\omega \mathbf{P} = -\mathbf{A} \cdot \mathbf{P} + \mathbf{C}_\omega$$

or

$$\mathbf{P} = (\mathbf{A} - i\omega \mathbb{I})^{-1} \mathbf{C}_\omega$$

where $\mathbf{P} = (P^e_x, P^e_y, P^n_x, P^n_y)^\top$ and

$$\mathbf{C}_\omega = \begin{pmatrix}
\gamma_e P_z B_{0y} / q \\
-\gamma_e P_z B_{0x} / q \\
\gamma_n P^n_z B_{0y} \\
-\gamma_n P^n_z B_{0x}
\end{pmatrix}$$

83
\[ A = \begin{pmatrix}
\tilde{R}_e^{\text{tot}} & \omega_e & -\tilde{R}_e^{\text{en}} & -\omega_{en} \\
-\omega_e & \tilde{R}_e^{\text{total}} & \omega_{en} & -\tilde{R}_e^{\text{en}} \\
-R_{ne}^{\text{en}} & -\omega_{ne} & R_{n}^{\text{en}} & \omega_n \\
\omega_{ne} & -R_{ne}^{\text{en}} & -\omega_n & R_{n}^{\text{en}}
\end{pmatrix} \]  \hspace{1cm} (3.78)

Note that here the polarization vector has dimension 4 with real elements (instead of dimension 2 with complex elements); similarly for the matrix \( A \) and the vector \( \mathbf{C}_\omega \). In Eq. (3.77) the fields \( b^n, b^e \) and \( \Omega \) were ignored. It is straightforward to show that at the compensation point the measured signal can be approximated (setting \( R_{se}^{\text{en}} \) and \( R_{se}^{\text{ne}} \) to zero) to be:

\[
P_e^x \approx \frac{\gamma_e P_z^e \omega \left[ B_{0y} \omega \left( \tilde{R}_e^{\text{tot}} - i\omega \right) - B_{0x} \omega (\omega_0 + \omega_{e0}) + i\tilde{R}_e^{\text{tot}} \omega_0 \right]}{q \left[ \tilde{R}_e^{\text{tot}} (\omega - \omega_0) + i\omega (\omega_{e0} + \omega_0 - \omega) \right] \left[ \tilde{R}_e^{\text{tot}} (\omega + \omega_0) - i\omega (\omega_{e0} + \omega_0 + \omega) \right]} e^{-i\omega t} \]

(3.79)

where \( \omega_0 \approx \gamma_n \lambda M^n P_z^n \) and \( \omega_{e0} \approx \gamma_e \lambda M^e P_z^e / q \). For low frequencies (\( \omega \ll (\omega_0, \omega_{e0}) \)):

\[
P_e^x \approx \frac{i\gamma_e P_z^e \omega}{R_{e}^{\text{tot}} \omega_0} \left[ \frac{B_{0y} \omega}{R_{e}^{\text{tot}} \omega_0} + O \left( \frac{\omega}{\omega_0} \right)^2 \right] B_{0x} e^{-i\omega t} + \left[ \frac{-\gamma_e P_z^e \omega^2}{R_{e}^{\text{tot}} \omega_0^2} + O \left( \frac{\omega}{\omega_0} \right)^3 \right] B_{0y} e^{-i\omega t} \]

(3.80)

and for high frequencies (\( \omega \gg (\omega_0, \omega_{e0}, R_{e}^{\text{tot}}) \)):

\[
P_e^x \approx \frac{\gamma_e P_z^e}{R_{e}^{\text{tot}}} \left[ \frac{(\omega_{e0} + \omega_0) R_{e}^{\text{tot}}}{q \omega^2} B_{0x} \right] + \left[ \frac{i R_{e}^{\text{tot}} \omega_0}{q \omega} B_{0y} \right] e^{-i\omega t} \]

(3.81)

### 3.5 Fundamental Noise

It is interesting to explore the fundamental limit of the comagnetometer. Here, we present a phenomenological derivation, without too much emphasis on rigorosity.
3.5.1 Atomic Spin Noise

We use a semi-classical approach, and modify the Bloch equations ((3.2) and (3.3)) to include noise terms (Langevin forces), which originate from relaxation processes and the quantum fluctuation of the probe polarization (light-shift noise) as explained in Chapter 6. We consider the performance of the co-magnetometer at the compensation point when the angle $\alpha$, the classical probe light-shift and probing photon spin vector $s_{pr}$ and the external transverse magnetic fields $B_t$ have been zeroed. In addition, the uniform rotation and the anomalous fields are set to zero ($\Omega = 0, b^e = b^n = 0$). Furthermore, the external magnetic field is treated classically (no quantum mechanical fluctuations); however, we include the effective magnetic-like quantum noise associated with the fluctuations of the nuclear and electron spins.

We then write the Bloch equations (with the Langevin noise terms) in the Ito form:

$$dP = -APdt + d\mathcal{F}$$

with $P$ and $A$ defined in the previous section and:

$$d\mathcal{F} = \begin{pmatrix}
\sqrt{\xi \frac{R_pr}{N_e}} dW_1(t) \\
\sqrt{\xi \frac{R^n_{tot}}{N_e}} dW_2(t) + \delta L_{pr} dW_3(t) \\
\sqrt{\frac{2R^n_{tot}}{N_n}} dW_4(t) \\
\sqrt{\frac{2R^n_{tot}}{N_n}} dW_5(t)
\end{pmatrix}$$

where $W_{1-5}(t)$ are classical Wiener processes, for which the following equations hold:

$$dW_i(t)dW_j(t) = \delta_{ij}dt$$

$$dW_i(t)^{N+2} = 0, \quad N > 0$$

$$dW_i(t)dt = 0$$
The term proportional to $\delta L_{pr}$ refers to the probe light-shift noise\textsuperscript{9}, which for unsqueezed light and under the conditions of large detuning and high pressure can be written [163]:

$$\delta L_x = r_c f_{osc} D(\nu) \frac{\sqrt{\Phi} P_e^c}{A} \frac{q}{Z}$$

(3.87)

where $\Phi$ is the number of photons per unit time, and $A$ is the area of the beam. The number of alkali atoms and noble gas atoms in the measurement volume are $N_e$ and $N_n$ respectively. The coefficient $\xi$ is a dimensionless numerical factor which depends on the longitudinal polarization as discussed in Chapter 6\textsuperscript{10} for $P_{z}^e = 1 \rightarrow \xi = 1/2$, while for $P_{z}^e = 0.5$ (which is realized in the thesis experiment) $\xi = 0.65$. We emphasize the semiclassical character of the above derivation, in which the variables are treated as commuting (classical) quantities (instead of non-commuting operators) and the quantum nature appears only in the phenomenology of the equations\textsuperscript{10}. Since $W_2(t)$ and $W_3(t)$ are classical Wiener processes we can write:

$$\sqrt{\xi P_{tot}^c/N_e} dW_2(t) + \delta L_{pr} dW_3(t) = \sqrt{\xi P_{tot}^c/N_e + \delta L_{pr}^2} dW_6(t)$$

(3.88)

with $W_6(t)$ being a classical Wiener process with the properties of equations (3.84)-(3.86). We can then write the stochastic differential Eq. (3.82) in the convenient, compact form:

$$dP = -A P dt + B dW$$

(3.89)

where:

$$B = \begin{pmatrix}
\sqrt{\xi P_{tot}^n/N_e} & 0 & 0 & 0 \\
0 & \sqrt{\xi P_{tot}^c/N_e + \delta L_{pr}^2} & 0 & 0 \\
0 & 0 & \sqrt{2 P_{tot}^n/N_n} & 0 \\
0 & 0 & 0 & \sqrt{2 P_{tot}^n/N_n}
\end{pmatrix}$$

(3.90)

\textsuperscript{9}The pump beam is tuned on resonance and assuming no wavelength fluctuations the pump beam does not contribute to the light-shift noise.

\textsuperscript{10}See Chapter 6 for a justification of the semiclassical model.
and \( d\mathcal{W} = (dW_1(t), dW_6(t), dW_4(t), dW_5(t))^\top \). Equations (3.82) and (3.89) describe a multivariate Ornstein-Uhlenbeck process [75, 83]. The spectral density matrix (Fourier transform of the correlation function) in the stationary state is given by [75]:

\[
S(\omega) = \frac{1}{2\pi} \left( \mathbf{A} + i\omega \right)^{-1} \mathbf{B} \mathbf{B}^\top \left( \mathbf{A}^\top - i\omega \right)^{-1}
\]

(3.91)

The spectrum matrix is a \( 4 \times 4 \) matrix; the non-diagonal elements describe the cross-correlations while the diagonal elements are the spectra of \( P_e^x, P_e^y, P_n^x \) and \( P_n^y \). Equation (3.91) describes the two sided spectrum (\( \omega \) can be positive or negative); a more experimentally relevant quantity is the one sided spectral density in the frequency domain \( G(f) = 4\pi S(\omega) \) for \( f \geq 0 \) and \( G(f) = 0 \) for \( f < 0 \), where \( \omega = 2\pi f \). We are interested in the spectrum of \( P_e^x \), which is the measured quantity in the experiment. The analytic form for the measured spin noise spectrum \( G_m(\omega) \) is rather complicated, but a few approximations lead to a more tractable form. In a typical alkali-noble gas comagnetometer the number of alkali atoms is many orders of magnitude smaller than the noble gas atoms; we can then ignore the Langevin forces in \( \mathbf{B} \) that come from noble gas relaxation. Also, in matrix \( \mathbf{A} \), we set \( \tilde{R}_\text{se}, R_{\text{se}}^\text{n} \) and \( R_{\text{tot}}^n \) to zero, as was explained in detail in Section 3.2. After a bit of algebra, we get for the measured spin noise spectrum:

\[ G_m(\omega) \]

11The eigenvalues of \( \mathbf{A} \) should have positive real part for a steady state to exist. As was discussed previously this is the case when the comagnetometer is at the operation point (close to the compensation point).
spin noise spectrum $G_m(f)$:

$$G_m(f) \approx \frac{2C}{N_e V}$$

(3.92)

$$C = N_e \left( f^2 q \omega_{e0} \delta L_{pr} \right)^2 + q R_{tot}^e \xi \left[ (f^2 - f_0^2)^2 \left( 4\pi^2 f^2 q^2 + R_{tot}^e \right)^2 ight]$$

(3.93)

$$V = \left( f^2 - f_0^2 \right)^2 \left( 4\pi^2 q^2 f^2 + R_{tot}^e \right)^2 + 8\pi q f_0 f^4 \omega_{e0}^3 + f^4 \omega_{e0}^4$$

(3.94)

where $f_0 = \omega_0/2\pi \approx \gamma_n \lambda M^n P^n_z / 2\pi$. In Fig. 3.3 we plot the spin noise spectrum as a function of frequency, using the values of table 3.1, the spectrum has been converted to effective anomalous magnetic field noise (from polarization noise) using the calibration factor $(\gamma_e P_e^z / R_{tot}^e)^2$. For the resolution of the figure there is no difference between the approximate spectrum (Equations (3.92)-(3.94)) and the spectrum that includes the dependence on $R_{se}^e, R_{se}^{ne}$ and $R_{tot}^e$.

It is interesting to estimate the noise spectrum in the low frequency limit. It can be shown that to first order in $f, R_{se}^e, R_{se}^{ne}$ and $R_{tot}^e$ we get:

$$W_{f \to 0}(f) \approx \frac{2q\xi}{N_e R_{tot}^e} (1 - 2C_{se}^{ne} - 2D_{se}^{ne} - 2F_{sd}^{ne}) \approx \frac{2q\xi}{N_e R_{tot}^e}$$

(3.95)

where $C_{se}^{ne}, D_{se}^{ne}$ and $F_{sd}^{ne}$ were defined in Eq. (3.32)-(3.35). The noise in electron polarization along the direction of the probe beam (Eq. (3.95)) is translated to uncertainty (per unit bandwidth) in the estimation of uniform rotation along the y-axis (axis perpendicular to the pump and probe direction) or the anomalous, magnetic-like field in the y-axis. From Equations (3.95) and (3.26) we find the fundamental limit\textsuperscript{12} (associated with the spin projection noise) of the comagnetometer as a gyroscope or gyroscope.

\textsuperscript{12}The limit is reported in per square root of unit bandwidth. The reader is referred to the appendix for a brief explanation.
Figure 3.3: Theoretical estimate (solid blue curve) of the effective magnetic field noise spectrum for the thesis experiment by numerical solution of Eq. (3.91). The dashed red curve is the noise assuming uncertainty is electron polarization $\delta P^e_x = \sqrt{\frac{2q\xi}{N_eR^e_{tot}}}$ as discussed in the main document. The values of Table 3.1 were used in this calculation. The peak at $\approx 21 \text{ Hz}$ corresponds to the resonance frequency of the coupled ensemble (see Eq. (3.74)).

anomalous, spin-coupling field sensor:

$$\delta \Omega_y \approx \frac{\gamma_n}{\gamma_e P^e_z} \sqrt{\frac{2q\xi R^e_{tot}}{N_e}} \quad (3.96)$$

$$\delta b^e_y, \delta b^n_y \approx \frac{1}{\gamma_e P^e_z} \sqrt{\frac{2q\xi R^n_{tot}}{N_e}} \quad (3.97)$$

The same result for the comagnetometer noise (associated with the spin noise) in the low frequency regime is reached if we simply equate the uncertainty $\delta P^e_x$ with the spin projection noise of an ensemble of alkali atoms with spin nuclear spin $I = 3/2$; in this case $\delta P^e_x = \sqrt{\frac{2q\xi}{N_eR^e_{tot}}}$, which is the same as the approximate Eq. (3.95). In fact, this is not surprising, since in the derivation of (the second part of) Eq. (3.95) the noise from the noble gas is neglected and the low frequency limit effectively simplifies the dynamics.
3.5.2 Photon Shot Noise

The above equations for the comagnetometer noise include only the contribution from the spin projection noise. In actual experiment we need to add (in quadrature) the photon shot noise, which depends on the type of probe light (squeezed or coherent). In this section coherent light is assumed, but the use of quantum optics formalism makes the extension to squeezed light straightforward. Two types of polarization measurement are studied: balanced polarimetry and lock-in detection with the use of Photoelastic modulator. Equations for the angular noise are derived, which can be converted to equivalent polarization noise using equation (see Section 2.8.1):

\[
\phi = \frac{1}{2} |A| t_r c f_{osc} D(\nu) P_x^c
\]  

Balanced Polarimetry Detection

The basic concept is illustrated in Fig. 3.4 (a). The strong local oscillator field\(^{13}\) \(\hat{b}(t)\) with electric field in the horizontal direction \(H\) is overlapped with the quantum field \(\hat{a}(t)\) with electric field in the vertical direction \(V\). The polarizing cube splits the light into the diagonal \(\hat{c}(t)\) (45°) and antidiagonal \(\hat{d}(t)\) (−45°) modes and the differential power \(S\) of the two modes is measured for time \(T\) [76, 94]:

\[
\hat{c}(t) = \left(\hat{a}(t) + \hat{b}(t)\right) / \sqrt{2} \\
\hat{d}(t) = \left(-\hat{a}(t) + \hat{b}(t)\right) / \sqrt{2} \\
S(T) = \frac{1}{T} \int_0^T \left(\hat{c}(t)^\dagger \hat{c}(t) - \hat{d}(t)^\dagger \hat{d}(t)\right) \, dt
\]

For simplicity we assume ideal photodetectors (100% efficiency). It is convenient to write the local oscillator in the form: \(\hat{b} = \hat{b} + \hat{v}\), where \(\hat{b}(t) \approx \beta(t)\) is very large and can be approximated by a complex number (\(\beta\)), and \(\hat{\nu}(t)\) includes the fluctuations

\(^{13}\)The operators refer to the amplitude of the electric field as was explained in Section 2.17.1
(quantum and possibly classical) of the local oscillator. Then, Eq. (3.101) is written:

\[
S(T) = \frac{1}{T} \int_0^T \left[ \beta^*(t) \dot{\hat{a}}(t) + \beta(t) \hat{a}^\dagger(t) + \dot{\upsilon}(t) \hat{a}(t) + \upsilon(t) \hat{a}^\dagger(t) \right] dt
\]  

(3.102)

The last two terms can be neglected compared to the first two (\(\beta(t)\) is very large). Assuming a single frequency local oscillator (the laser field is a very good approximation to it), i.e. \(\beta(t) = |\beta|e^{-i\chi}\), \(\chi\) being the phase of the oscillator, after a bit of algebra it can be shown that the expected measured signal is:

\[
\langle S(T) \rangle \approx 2|\beta|\text{Real} [e^{i\chi} \langle \tilde{a} \rangle] 
\]  

(3.103)

where \(\tilde{a} = \frac{1}{T} \int_0^T \hat{a}(t) \, dt\). The variance of the detected signal is:

\[
\delta S^2(T) = \langle S^2(T) \rangle - \langle S(T) \rangle^2 \approx \frac{|\beta|^2}{T} 
\]  

(3.104)

In deriving Eq. (3.104) we assumed vacuum coherent field for \(\hat{a}(t)\) and the properties for a coherent vacuum field mentioned in Section 2.17.4. To associate the measured noise \(\delta S(T)\) with angular noise we note that the local oscillator mode rotated by the small angle \(\delta \phi\) results in signal:

\[
\langle S(T) \rangle \approx 2|\beta|^2 \delta \phi 
\]  

(3.105)

The noise in Eq. (3.104) corresponds to angular noise:

\[
\delta \phi \approx \frac{1}{2\sqrt{|\beta|^2 T}} = \frac{1}{2\sqrt{\Phi_m T}} 
\]  

(3.106)

where \(\Phi_m = \langle \hat{b}^\dagger \hat{b} \rangle\) is the flux of photons (photons per unit time) of the incident light field (local oscillator). The (one sided) angular noise spectrum (in frequency domain)

\footnote{From Eq. (3.102) and setting \(\langle \hat{a}(t) \rangle \approx \beta \delta \phi\)}
is equal to:

\[
\delta \phi_{BW}(f) = \frac{1}{\sqrt{2\Phi_m}} \tag{3.107}
\]

where we used \( BW = 1/(2T) \). It can be shown that in the case of non-ideal photodetectors (e.g. of equal sensitivity \( \eta \)) the noise equations have to be modified according to the rule: \( \Phi_m \mapsto \eta \Phi_m \).

Figure 3.4: Homodyne (a) and heterodyne (b) detection schemes. PB: Polarizing Beamsplitter, PD: Photodetector, PEM: Photoelastic Modulator.

The classical treatment of photon counting as a Poisson process \([75]\) leads to the same result for the photon shot noise of the coherent field (Equation (3.106)). The detected photons \( N_1 \) and \( N_2 \) in time \( T \) by the two detectors are random variables following Poisson distribution with mean and variance are:

\[
\langle N_1 \rangle = \langle N_1^2 \rangle - \langle N_1 \rangle^2 = N_1 = \eta \Phi_m T/2 \tag{3.108}
\]

\[
\langle N_2 \rangle = \langle N_2^2 \rangle - \langle N_2 \rangle^2 = N_2 = \eta \Phi_m T/2 \tag{3.109}
\]

The measurement of polarization rotation \( \phi \) is:

\[
\phi = \frac{N_1 - N_2}{2(N_1 + N_2)} \tag{3.110}
\]
and the uncertainty is:

\[ \delta \phi = \sqrt{\left( \frac{\partial \phi}{\partial N_1} \delta N_1 \right)^2 + \left( \frac{\partial \phi}{\partial N_2} \delta N_2 \right)^2} \]  

(3.111)

\[ = \frac{\sqrt{N_2^2 \delta N_1^2 + N_1^2 \delta N_2^2}}{(N_1 + N_2)^2} \]  

(3.108)

\[ eq:N:mean2:sec: \]  

\[ eq:PSN:ch:Comag:Theory \]

\[ 1 \]  

(3.112)

**Lock-in detection**

As shown in Fig. 3.4 (b), angular measurement can be performed with the use of a quarter-wave plate with the two axes of retardation (fast and slow) in the horizontal and vertical direction, a photoelastic modulator in the diagonal direction applying a small oscillating retardation \( \psi(t) = \alpha_m \cos \omega_m t \), and a linear polarizer with the axis of transmission oriented in the vertical direction. The strong local oscillator (probe light) \( \hat{b}(t) \) has horizontal polarization, while the quantum field \( \hat{a}(t) \) is vertically polarized. The transmitted field is \[ \hat{c}(t) = \frac{1}{2} \left[ \hat{a}(t) \left( -1 + e^{i\psi(t)} \right) + i \hat{b}(t) \left( 1 + e^{i\psi(t)} \right) \right] \]  

(3.113)

The signal to be measured (with the lock-in technique) is:

\[ S(T) = \frac{\sqrt{2}}{T} \int_0^T \hat{c}(t) \hat{c}(t) \cos(\omega t) \, dt \]  

(3.114)

In the above equation it is assumed that \( T \gg 2\pi/\omega \). For symbolic calculation it is convenient to assume that \( T = 2\pi k/\omega, k \in \mathbb{N} \) and \( k \gg 1 \). The \( \sqrt{2} \) factor normalizes the lock-in output to the rms value of the harmonic component. Following arguments similar to those for the balanced polarimetry detection, it can be shown that the

\[ ^{15} \]  

For concreteness we assumed that the slow axis of the quarter-wave plate is oriented in the horizontal direction.
signal is:

\[
\langle S(T) \rangle \approx \frac{1}{\sqrt{2T}} \int_0^T \langle \beta^*(t)\hat{a}(t) + \beta(t)\hat{a}^\dagger(t) \rangle \sin[\alpha_m \cos(\omega_m t)] \cos(\omega t) \, dt
\]

\[
+ \frac{1}{\sqrt{2T}} \int_0^T |\beta|^2 [1 - \cos [\alpha_m \cos(\omega_m t)]] \cos(\omega t) \, dt
\]

(3.115)

\[
\langle S(T) \rangle_{\omega_m} \approx \sqrt{2}\beta^2 \delta J_1(\alpha_m) \approx \frac{|\beta|^2 \alpha_m \delta \phi}{\sqrt{2}} [1 + O(\alpha_m^2)] , \quad \omega = \omega_m
\]

(3.116)

\[
\langle S(T) \rangle_{2\omega_m} \approx \frac{|\beta|^2 J_2(\alpha_m)}{\sqrt{2}} \approx \frac{|\beta|^2 \alpha_m^2}{8\sqrt{2}} [1 + O(\alpha_m^2)] , \quad \omega = 2\omega_m
\]

(3.117)

Similarly, the noise is:

\[
\delta S^2(T) = \frac{1}{2T} \int_0^T |\beta|^2(t) \sin(\alpha_m \cos(\omega_m t)) \cos(\omega t)^2 \, dt
\]

\[
+ \frac{1}{T^2} \int_0^T dt' dt |\beta|^2(t)|\beta|^2(t') \sin \left( \frac{\alpha_m \cos(\omega_m t)}{2} \right) \sin \left( \frac{\alpha_m \cos(\omega_m t')}{2} \right)^2 \cos \omega t \cos \omega t'
\]

(3.118)

and for \( \omega = \omega_m \) becomes:

\[
\delta S^2(T) \approx \frac{|\beta|^2}{8T\alpha_m} [\alpha_m - J_1(2\alpha_m) + 2\alpha_m J_2(2\alpha_m)]
\]

(3.119)

\[
\approx \frac{3}{16T} \alpha_m^2 |\beta|^2 [1 + O(\alpha_m^2)]
\]

(3.120)

where \( J_n(x) \) is the Bessel function of the first kind \([2]\). The angular noise is:

\[
\delta \phi \approx \sqrt{\frac{3}{2} \frac{1}{2 \sqrt{\Phi_p T}}}
\]

(3.121)

\[
\delta \phi_{BW}(f) \approx \sqrt{\frac{3}{2} \frac{1}{\sqrt{2\Phi_p}}}
\]

(3.122)

Note that angular noise in the lock-in measurement is a factor of \( \sqrt{3/2} \approx 1.225 \) larger than in the balanced polarimetry case.
Using the values of Table 3.1, the expected photon shot noise is $\sim 0.3 \text{ fT/}\sqrt{\text{Hz}}$, which is approximately a factor of three larger than the atom shot noise (see Section 4.6.6 for the noise in the actual experiment).

### 3.6 Response To Transients

An important feature of the comagnetometer sensor is the response to magnetic field transients and time varying rotations. The general case of arbitrary perturbation is studied with the use of Green’s function formalism \[197\] for the linearized Bloch equations:

$$\frac{d\mathbf{P}}{dt} = \mathbf{A} \mathbf{P} + B_p$$  \hspace{1cm} (3.123)

and the boundary condition $\mathbf{P}(t = 0) = 0$ (before the application of any perturbation the transverse electron and nuclear polarization is zero). $\mathbf{A}$ and $\mathbf{P}$ were defined in Section 3.4 and the perturbation vector (that makes the equation “inhomogeneous”) is:

$$B_p = \begin{pmatrix}
P^e_z (\gamma e B_y/q + \Omega_y) \\
-P^e_z (\gamma e B_x/q + \Omega_x) \\
P^n_z (\gamma n B_y + \Omega_y) \\
-P^n_z (\gamma n B_x + \Omega_x)
\end{pmatrix}$$  \hspace{1cm} (3.124)

The general solution of the homogeneous equation ((3.123) with $B_p = 0$) is given by:

$$\mathbf{P}_h = c_1 \mathbf{P}_1 e^{\lambda_1 t} + c_2 \mathbf{P}_2 e^{\lambda_2 t} + c_3 \mathbf{P}_3 e^{\lambda_3 t} + c_4 \mathbf{P}_4 e^{\lambda_4 t}$$  \hspace{1cm} (3.125)

where $c_{1-4}$ are constants to be determined from the boundary and continuity conditions, $\lambda_{1-4}$ are the eigenvalues of $\mathbf{A}$ and $\mathbf{P}_{1-4}$ the corresponding eigenvectors. Neglect-
The Green’s function is equal to:

\[ G(t, \xi) \] 

at time \( \xi \) found in Section 3.3

ing the contribution of \( R_{\text{se}}^n \), \( R_{\text{se}}^m \) and \( R_{\text{sd}}^n \), the eigenvalues\(^{16}\) are:

\[
\lambda_1 = \frac{1}{2} \left[ -\tilde{R}_{\text{tot}}^e + i (\omega_c + \omega_0) - \sqrt{x_r + iy_r} \right] \\
\lambda_2 = \frac{1}{2} \left[ -\tilde{R}_{\text{tot}}^e + i (\omega_c + \omega_0) + \sqrt{x_r + iy_r} \right] \\
\lambda_3 = \frac{1}{2} \left[ -\tilde{R}_{\text{tot}}^e - i (\omega_c + \omega_0) - \sqrt{x_r - iy_r} \right] \\
\lambda_3 = \frac{1}{2} \left[ -\tilde{R}_{\text{tot}}^e - i (\omega_c + \omega_0) + \sqrt{x_r - iy_r} \right]
\]

and the eigenvectors are:

\[
P_1 = \begin{pmatrix} 1 \\ i \\ \frac{-i\tilde{R}_{\text{tot}}^e - \omega_c + \omega_0 + i\sqrt{x_r + iy_r}}{2\omega_c} \\ \frac{\tilde{R}_{\text{tot}}^e - i(\omega_c - \omega_0) - \sqrt{x_r + iy_r}}{2\omega_c} \end{pmatrix}, \\
P_2 = \begin{pmatrix} 1 \\ i \\ \frac{-i\tilde{R}_{\text{tot}}^e - \omega_c + \omega_0 - i\sqrt{x_r + iy_r}}{2\omega_c} \\ \frac{\tilde{R}_{\text{tot}}^e - i(\omega_c - \omega_0) + \sqrt{x_r + iy_r}}{2\omega_c} \end{pmatrix}
\]

\[
P_3 = \begin{pmatrix} 1 \\ -i \\ \frac{i\tilde{R}_{\text{tot}}^e - \omega_c + \omega_0 - i\sqrt{x_r - iy_r}}{2\omega_c} \\ \frac{\tilde{R}_{\text{tot}}^e + i(\omega_c - \omega_0) - \sqrt{x_r - iy_r}}{2\omega_c} \end{pmatrix}, \\
P_4 = \begin{pmatrix} 1 \\ -i \\ \frac{i\tilde{R}_{\text{tot}}^e - \omega_c + \omega_0 + i\sqrt{x_r - iy_r}}{2\omega_c} \\ \frac{\tilde{R}_{\text{tot}}^e + i(\omega_c - \omega_0) + \sqrt{x_r - iy_r}}{2\omega_c} \end{pmatrix}
\]

Since the system is first-order, the Green’s function \( G^v(t, \xi) \) should be continuous at time \( \xi \) when the delta perturbation is applied, i.e. \( (B_x, B_y, \Omega_x, \Omega_y) = \delta(t - \xi) \).

The superscript \( v \) is used to denote the vectorial character of the Green’s function.

The Green’s function is equal to:

\[
G^v(t, \xi) = \begin{cases} 
0, & 0 < t < \xi \\
\sum_{i=1}^{4} c_i \xi_p e^{\lambda_i t}, & t > \xi
\end{cases}
\]

\(^{16}\)Of course these are the same as the eigenvalues of the 2 × 2 matrix \( A \) with complex elements found in Section 3.
The coefficients $c_1^{\xi}$ are the solutions of the continuity equation: $G^v(t,\xi^+) - G^v(t,\xi^-) = B_p$, with one of the $B_x$, $B_y$, $\Omega_x$ and $\Omega_y$ set to unity and the rest to zero depending on which perturbation is investigated (due to the linear character of Eq. (3.123) each perturbation can be studied separately). The measured signal for an arbitrary time varying perturbation $p(t)$ is given:

$$P^e_x(t) = \int_0^\infty G^e_x(t,\xi)p(\xi)\,d\xi$$  \hspace{1cm} (3.132)$$

where $G^e_x(t,\xi)$ is the first element of $G^v(t,\xi)$ associated with the electron polarization along the direction of the probe beam. It can be shown that the Green’s function has the property:

$$G^e_x(t,\xi) = G^e_x(t-\xi).$$

In particular, we are interested in the integral of the comagnetometer response for a time interval during which a transverse magnetic field transient or a time varying rotation were applied. The end limit of integration is taken to be significantly larger than the time when the transient or the rotation ended, so that the integration interval can be extended to infinity. It is then straightforward to show:

$$\int_0^\infty P^e_x(t)|_{B_x} \, dt = 0, \quad \int_0^\infty P^e_x(t)|_{B_y} \, dt = 0, \quad \int_0^\infty P^e_x(t)|_{\Omega_x} \, dt = 0 \quad (3.133)$$

$$\int_0^\infty P^e_x(t)|_{\Omega_y} \, dt = -\frac{\gamma_e P^e_z}{\gamma_{eff} R_{tot}} \int_0^\infty p(\xi)\,d\xi \quad (3.134)$$

The integral of the signal is not affected by transverse magnetic field transients or rotation around the probe axis, and is proportional to the total angle of rotation around the $y$-axis independent of the time dependence of $\Omega_y$ \cite{115}; even for a very fast rotation the comagnetometer tracks the rotation angle \cite{116}. This makes the comagnetometer particularly attractive for gyroscopic applications.

\footnote{The subscript in $P^e_x$ denotes which perturbation is applied.}

\footnote{There is however a delay which is determined by the characteristic relaxation time.}
Figure 3.5: The comagnetometer signal and angular estimation (integral of the signal over time) is insensitive to transverse magnetic field transients. Here $B_y$ transient is shown and has been divided by a factor of 10 for plotting purposes.

In Fig. 3.5 the comagnetometer response to a $B_y$ transient is simulated using the values of 3.1. The comagnetometer signal quickly relaxes to zero after the decay of the transient, and the angular reading (integral of the signal over time) is not affected by the magnetic field perturbation.
Chapter 4

Comagnetometer Implementation

The experimental implementation of the K-\(^3\)He comagnetometer is presented in this chapter. The optical, electrical and mechanical elements that constitute the setup are described, and the procedure of preparing the system for measurement is analyzed. The performance of the comagnetometer in the measurement of an anomalous field is characterized. The comagnetometer implementation of this thesis was based on the one described in [TK], but with a few important modifications. We then describe and evaluate the performance of the \(^3\)He spin source which provides the anomalous field to be measured.

4.1 Comagnetometer Experimental Setup

A schematic of the experimental setup is shown in Fig. 4.1.

4.1.1 Cell

The atomic ensembles (K and \(^3\)He) are contained in an approximately spherical, aluminosilicate glass (Corning 1720) cell of 2.4 cm outer diameter with thin walls (esti-
mated to be approximately 0.2 mm). Aluminosilicate glass\(^1\), being dense, is relatively impermeable and can keep large pressures of \(^3\)He over many months with low losses \([166]\). In addition, this kind of glass shows negligible discoloration (darkening) from the exposure to alkali metals at the temperatures of the experiment \([127]\), and the transmission of light through the cell remains high, limited only by the reflection losses on the glass-air interface (\(\sim 84\%\) for a glass with refractive index 1.528). The small wall thickness keeps the birefringence and the light absorption of the cell low, without compromising the mechanical endurability of the cell to withstand the high atomic pressure. The cell size is small enough so that the available heating and pumping power can create high density and polarization throughout the cell and the effect from magnetic field inhomogeneities is suppressed, but (the cell size is) large enough so that the relaxation rate from diffusion to the wall is insignificant and the lensing effect of the cell weak. A spherical cell reduces (or even eliminates to the extent that the polarization is uniform) noble gas relaxation related to magnetic inhomogeneities from the \(^3\)He magnetization, which in practice is the limiting factor for high polarizations. A sphere has the additional advantage that exhibits better mechanical properties to stress than any other shape \([185]\). The largest deviation of the surface from the sphere occurs where the main body is attached to the stem. It is therefore important to construct a thin neck\(^2\), which in the case of the thesis experiment was a few mm. The asphericity of the contact point is significantly alleviated by placing the liquid alkali metal at the entrance of the stem, thus forming an effective plug for the cell neck. We found that in certain cases, when the cell was heated to the required temperature, the plug was moved from the start of the neck to deeper into the stem, probably due to pressure gradient in the two sides of the meniscus. Although

\(^1\)In aluminosilicate glass some silica (SiO\(_2\)) is replaced by alumina (Al\(_2\)O\(_3\)). The composition of Corning 1720 aluminosilicate (the type of glass used in the experiment) can be found in \([159]\).

\(^2\)This requirement applies to the inner part neck that gives shape to the atomic vapor; the outer neck should in fact be relatively thick to give extra strength to the mechanically weak point of contact with the main body.
we haven’t developed a standardized procedure to appropriately position the plug so that it does not move during the experiment, it is likely that for this it is helpful to retain approximately uniform temperature throughout the cell when placing the plug. In addition, constructing a conical stem cell (with the narrow part at the point of contact with the main cell body) harnesses the surface tension for preventing motion of the plug [53]. Creating a temperature gradient between the main cell and the stem with additional heating of the stem creates differential pressure that also acts against unwanted displacement of the liquid plug.

The cell was filled with K in natural abundance (93.26% $^{39}$K and 6.73% $^{41}$K), $^3$He and molecular N$_2$. The two isotopes of K have the same nuclear spin ($I = 3/2$) and for the high pressure conditions (where the hyperfine structure is not resolved) of the experiment their atomic properties related to the measurement are practically the same, so that they can be treated as one species. The cell is filled with enough K so that when heated a droplet with radius of a few mm is formed, adequate to constitute the stem plug. However, adding more potassium than what is required to reach the saturated vapor (at the temperature of operation) makes more likely the formation of K droplets at the coldest regions of the cell (see Fig. 4.3 (a)). The pressure of N$_2$ is estimated (from the filling process) to be 46 Torr at room temperature. Based on the reported in literature for $\tilde{p}_Q$ (Section 2.7) [136] the probability for radiative decay is $\approx 11\%$, and the effect of radiation trapping in polarization is significantly suppressed.

There is a theoretical advantage to have high density of noble gas: typically, large values of $\omega_{en}\omega_{ne} \propto [\text{He}]$ where $[\text{He}]$ is the noble gas density lead to faster damping at the compensation point (see Section 3.3), and large $B_n \approx \lambda n M_n P_z^n$ results in stronger suppression of noise terms associated with magnetic field and light-shift fluctuations (see Section 3.2). In addition, inhomogeneities in the alkali pumping rate,

$^3$Potassium is solid at room temperature and has to be heated above 63°C to become liquid and be moved to the neck of the cell.
Figure 4.2: Schematic for filling the comagnetometer cell with $^3$He. Most the $^3$He in the string is concentrated inside the cell when it is brought into contact with liquid $^4$He. The amount of $^3$He is estimated from the measured pressure (before using the liquid $^4$He) and the volume of the string.

electron polarization and comagnetometer sensitivity, associated with propagation effects or variations in intensity profile of the pump beam, are reduced as the alkali optical linewidth is broadened from collisions with the noble gas atoms (see Section 4.6.3). Taking the above into consideration, the cell was filled with approximately 12 atmospheres at room temperature ($\approx 11.2$ amagat$^4$) of $^3$He. The above number is an estimate based on the filling process (see Fig. 4.2): the cell connected (through the stem) to the main part of the filling chamber is brought in contact with liquid $^4$He so that most of the $^3$He (and all $N_2$) vapor is concentrated inside the cell (coldest region), and the cell is carefully detached from the chamber with a glass torch. From the conservation of the number of atoms, the pressure of noble gas in the cell at (absolute) temperature $T$ is estimated to be: $P_n \approx \frac{T}{T_0} \left[ P_1 + (P_1 - P_2) \frac{V_{ch}}{V_{cl}} \right]$, where $V_{ch}$ and $V_{cl}$ are the volumes of the chamber and the cell respectively, $P_1$ and $P_2$ are respectively the pressure gauge readings before and after cooling the cell with the liquid $^4$He, and $T_0$ is the temperature of the chamber.

\begin{footnote}
$^4$One amagat, named after the French physicist Emile Hilaire Amagat, is defined as the density of an ideal gas at standard conditions of pressure (1 atm) and temperature ($T=0$ °C), equal to $2.69 \times 10^{19}$ cm$^{-3}$ [194].
\end{footnote}
Figure 4.3: (taken from [TK]) (a) Photo of the cell used in [TK]. The thesis experiment was performed with a similar cell. The comagnetometer cell is firmly held in place by its stem. Droplets of K can be seen on the lower surface. During the experiment K in liquid phase forms the plug at the neck of cell. The 10 kΩ RTD temperature sensor can be seen close to the cell. (b) The double-walled oven of the experiment made of Nema Grade G-7. The heater wires (not shown here) are glued with a thermally conductive cement on the outer surface of the inner compartment (vector tip). (c) The cooling jacket, made of thermally conductive epoxy that surrounds the oven, is cooled with chilled water.

4.1.2 Oven

The cell is heated in a double-walled oven (Fig. 4.3 (b)) made of NEMA Grade G-7, a fiber glass and silicone resin laminate appropriate for high temperature applications (maximum temperature is 220° C). The thermal power is provided by six resistive wire heaters driven by AC current ($I_{rms} \sim 0.4$ A) and glued to the six outer oven sides with high thermal conductivity cement, which distributes homogeneously the heating power in each side. Special care is devoted to minimize the loop area of the heaters by using small diameter twisted-pair wire; this way the magnetic field from the heating current is suppressed. In addition, the frequency of the AC current (around 200 kHz) is well above the comagnetometer bandwidth and the response to the residual magnetic field is suppressed. By abruptly turning off the heater the total effective magnetic field on the comagnetometer (when the oven is at the operating temperature) was estimated to be below 600 fT. Fluctuations in the heating current
on the order of one part per thousand can be achieved\(^5\) over the time scales relevant for the experiment (approximately 10 sec), so that the effect of the heater to the measured signal is estimated to be below 0.6 fT\(^6\). In principle the six heaters allow for an independent control of the thermal power on each side of the oven; however for the experiment of this thesis a single temperature controller was used since this way an acceptable temperature homogeneity was realized. The total heater resistance is approximately 115 Ω and the power dissipated to maintain the temperature of operation is on the order of 50 Watts. There is also a separate heater around the neck of the stem, which allows additional, localized to stem, heating to control the position of the neck plug. Although this feature was not used in the experiment of this thesis (the oven temperature distribution and the careful positioning of the plug made the use of the heater unnecessary), it has been proven useful in other similar experiments \([33, 34]\).

The circuit schematic for the heater operation can be seen in Fig. 4.4. The temperature sensor is a 10 kΩ\(^7\) platinum resistive temperature device (RTD), which is placed in the inner oven chamber. The large resistance RTD and the corresponding enhanced temperature sensitivity allow for a small current excitation with insignificant effect on the comagnetometer; a Cryo-Con temperature controller (Model 32B) measures the RTD resistance in a four point contact measurement scheme by providing a 10 μA constant DC current, and applies a low frequency (timescale < 0.1 sec) PID controller to the reading. The PID output controls the amplitude of an oscillator (\(\sim 200\) kHz frequency), which in turn drives a high power (and current) transistor and the energy is coupled through a transformer to the heaters. The transformer decouples the heaters from the low frequency noisy currents generated by the transistor

\(^{5}\)This depends on the thermal stability of the oven and on the stability of the current source.
\(^{6}\)The heater also contributes to magnetic field gradients.
\(^{7}\)That is the nominal resistance (resistance at 0°C).
and oscillator circuit, and also steps up the voltage to the appropriate range for the heaters.

![Schematic diagram of the feedback loop that controls the temperature of the oven. Only high frequency AC current is flowing in the heaters.](image)

Figure 4.4: Schematic of the feedback loop that controls the temperature of the oven. Only high frequency AC current is flowing in the heaters.

We note that the design of the oven is not optimized for the adopted heating scheme\(^8\). The use of a low thickness, high thermal conductivity (non metallic, non magnetic) material instead of the low conductivity G-7 and air (\(\approx 0.29\) W/Km and \(\approx 0.025\) W/Km respectively) would result in better control of the system\(^9\).

The oven is thermally insulated with one-inch thickness polyimide foam, a high temperature foam of low conductivity (\(\sim 0.02\) W/Km)\(^{10}\). Due to the high thermal resistivity of the foam the heater power is effectively transferred to the cell, which remains isolated from environmental temperature fluctuations. The insulated oven is placed inside a box made of high thermal conductivity epoxy and water-cooled to avoid increase of the temperature inside the magnetic shields (Fig. 4.3 (c)).

Special care is devoted to the structural rigidity of the oven in order to avoid mechanical drifts. In addition, the mechanical instability from turbulent convective air currents is suppressed by preventing air leakage through interfaces of significantly

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\(^8\)For this experiment, the oven used in [TK] was modified. There, the oven was heated with warm air and the double walls isolated the cell from the turbulent air.

\(^9\)In newer experiments in the Romalis group at Princeton, the oven is constructed from ceramic Boron Nitride (heat conductivity \(\approx 120\) W/K m)\(^{[81, 34, 52]}\).

\(^{10}\)The foam heat conductivity is a combination of the gaseous conduction within the pores (dominant mechanism at atmospheric pressures, \(\sim 80\%\)), conduction through the solid structure (of density significantly smaller than that of the bulk material) and radiative transfer \([41]\).
different temperatures; the seams of the oven are pasted with Fomblin, a high temperature vacuum grease, and O-rings seal the openings for the pump and probe light.

4.1.3 Magnetic Fields and Shields

The oven-cooled epoxy box assembly is firmly rested inside a NEMA grade G-10 tube, around which magnetic field and gradient coils are wrapped. Cosine winding [107] is used to produce homogeneous magnetic fields in the transverse direction. Five layers of high $\mu$-metal magnetic shields surround the assembly, providing a shielding factor of $\approx 10^6$ to quasi-static magnetic fields [TK]. The space between the shields is filled with melamine foam, a soundproofing foam to suppress acoustic noise[12] (see Fig. 4.5). The structure is rigidly held together with a Ti frame and radial Ti rods which penetrate the shields and support the G-10 tube. In order to avoid Ti-Fe/Ti-Ni thermocouple junctions and galvanic action noise at the contacts (which could result in noisy currents in the shields) the magnetic shields are electrically isolated from the support structure.

The magnetic field in the comagnetometer is generated from the sum of a computer controlled current and a separate ultra-stable current source with a mercury battery reference (Fig. 4.6). Both sources have three independent channels that allow control of the magnetic field in the x, y and z axis. The current source is manually tuned to a rough longitudinal compensation and transverse field zeroing, while the computer output applies the field modulations and finely adjusts the magnetic fields to the point of optimum performance through the use of automated routines (see Section 4.2). A 16-bit National Instruments analog output card provides a voltage (scale $\pm 10$ V) which after filtering and buffering is converted to current on a resistor: $100 \text{k}\Omega$.

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11 A nickel-iron alloy (typically $\approx 80\% \text{ Ni}, 15 \% \text{ Fe}$ and other metals).
12 For instance, acoustic vibrations can get excited from turbulent air or at the opening for the pump and probe beam.
Figure 4.5: (taken from [TK]) Five layers of $\mu$-metal shields provide a shielding factor of $10^6$. The oven is firmly rested on the green G-10 tube, which is the frame for the magnetic field coils. The assembly is rigidly supported by a Ti frame. The white layer in between the magnetic shields is soundproofing foam.

for the x and y channel, and 20 k$\Omega$ for $B_{\parallel}$. The filter is of the RC type, with two options for the resistor corresponding to $\approx 0.008$ Hz and $\approx 11$ Hz corner frequency. An electronic switch (computer activated) switches between the short time constant ($\sim 14$ msec) during change of fields and the long time constant ($\sim 20$ sec) during signal measurement.

### 4.1.4 Vacuum tubes

The large temperature gradient between the oven and the room create turbulent convective air currents through the beam ports in the magnetic shields and oven; in addition, acoustic waves might be generated in the magnetic shields and oven openings. The resultant air density fluctuations affect the position (and angle) of the light beams (pump and probe), thus contributing to signal noise. For this, the pump and probe light travel through the layers of magnetic shielding and the oven inside

\footnote{The digitization error is $\sim 3$ nA and $\sim 15$ nA or $\sim 0.3$ pT and 1.4 pT for the transverse and longitudinal fields respectively; this noise level is significantly reduced by the filters.}
evacuated glass tubes (pressure < 1 Torr), which eliminate (for the corresponding section of the optical path) beam motion noise.

### 4.1.5 Comagnetometer Optical Setup

#### Pump Beam

The pump laser is a single mode distributed feedback (DFB) laser diode (GaAs semiconductor laser diode from Eagleyard). In order to emit light in the D1 line of K, the laser diode is cooled to approximately -18°C with a two stage cooling scheme: a thermoelectric (Peltier) cooler attached onto the laser diode wafer chip, and running water (≈ 20°C) at the hot side of the Peltier cooler. Unlike to the lasers used in [TK], the DFB laser has narrow linewidth (HWHM 40 MHz according to specifications), allows for smooth, wide range wavelength tuning, and exhibits low wavelength noise and drift: driven with a stable current source (δi < 0.1 mA) and temperature controlled within 1 mK, the expected wavelength fluctuation is smaller than 0.1 pm.

The laser light is directed through an optical isolator and appropriate lenses to a tapered amplifier (Optica TA 100) for power amplification. Using appropriate
combination of cylindrical lenses and a $\lambda/2$ wave plate the output of the amplifier
is mode matched (beam waist and correction for astigmatism) with the input of a
single mode polarization maintaining fiber\textsuperscript{16}. The light propagation through the fiber
is robust to temperature fluctuations and mechanical and other disturbances; the
output of the fiber (single mode $\text{TEM}_{00}$) is left to expand to a beam waist of 2 cm
(diameter) and roughly collimated by a lens mounted on a translation stage for beam
steering. The beam after the lens has a small divergence (estimated to be $< 1^\circ$) in
order to illuminate the whole cell (of diameter $\approx 2.4$ cm). The spherical aberration
of the collimating lens is used to smooth the intensity gradients of the beam (due to
the Gaussian profile), resulting in more homogeneous pumping rate across the cell.
The polarization homogeneity is also enhanced from the reduced optical depth at
the off center edges of the cell (due to its spherical shape\textsuperscript{17}). A beam sampler with
one surface AR-coated to minimize ghosting directs a fraction of the pump beam
(after the collimating lens) to a photodiode, and the measured intensity is used to
control the pump intensity through an Integral controller (bandwidth of a few tenths
of Hz), adjusting the current of the amplifier; this way the intensity is effectively
decoupled from the wavelength of the pump laser. A $\lambda/4$ wave-plate converts the
light polarization to circular (the helicity determines the orientation of the holding
field $B_z$). The fiber output, collimating lens and beam sampler are rested on a single
axis translation stage to facilitate beam alignment.

By creating a robust reference point (output of the fiber\textsuperscript{18}), the optical fiber
reduces significantly the path length of the pump beam compared to [TK]. Air density

\textsuperscript{16}A polarization maintaining fiber has strong built-in birefringence; due to the high birefringence,
(small) disturbances along the fiber (e.g. mechanical stress, temperature drift) do not couple ef-
effectively the fiber eigenmodes (linearly polarized light along the axes of birefringence) and the
polarization of light is preserved, provided that the input light is matched with one of the fiber
eigenmodes \cite{118}.

\textsuperscript{17}Due to the lensing effect from the cell walls the pump beam off the center is not aligned with the
holding magnetic field (nominally in the z-direction) and the absorption increases \cite{115}. However,
this effect is very small (see Fig. \textbf{4.9 (b)}).

\textsuperscript{18}In addition, the collimating lens, approximately one focal length from the fiber output, sup-
presses the effect of mechanical motion of the fiber on the beam.
fluctuations (for instance generated from convective currents, or acoustic waves) affect the pump beam only in the short region (20 cm) between the fiber output and the vacuum tube; this is to be contrasted with [TK] where the optical path length (before the vacuum tube) is 100 cm. In addition, the use of optical fiber reduces the effect of the flap flexing (e.g. from thermal stress) on the beam position\(^\text{19}\) (see Fig. 4.12).

As shown in Fig. 4.7, misalignment of the pump beam and cell center results in light-shift along the y-direction (direction perpendicular to the pump and probe beam) due to the retroflection of the light off the walls of the cell and the metallic (shinny) liquid K at the stem. Since the comagnetometer signal has first-order sensitivity on \(L_y\) fluctuations (Eq. (3.49)), it was argued in [TK] that wavelength and beam motional noise can have an effect on the measurement. From Section (2.4.6) it can be seen that the light-shift sensitivity on wavelength for a light field on resonance (D1, \(J_e = 1/2\)) is:

\[
\left| \frac{dL_y}{d\lambda} \right| = \frac{I_0 r_e c f_{osc}}{h \gamma_e \Gamma_2^2} \tag{4.1}
\]

where \(I_0\) is the pump beam intensity and \(\Gamma\) is the optical linewidth (HWHM) of K D1 transition. Assuming that one thousand of the pump light incident on the back side of the cell contributes to the net light-shift\(^\text{20}\), for typical conditions of the experiment (3.1): \(|dL_y/d\lambda| < 0.6 \text{ fT/pm}\) so that over the time scales of the experiment (\(\sim 10\) sec) this noise source does not seem to be significant\(^\text{21}\). Similarly, the light-shift noise resulting from beam motion is estimated to be: \(< 0.1 \text{ fT/} 10 \mu \text{m}\)\(^\text{22}\) and its contribution to the overall noise is expected to be minor.

\(^{19}\)The flap flexing moves the pump beam mainly in the vertical direction, causing extra noise. The characteristic time scale for the fluctuation is expected to be significantly longer than this for the measured signal (\(\sim 10\) sec).

\(^{20}\)Although typically the reflection losses are \(\sim 10\%\) (taking into account the curvature of the cell and the shiny liquid K that blocks the stem), the fraction of the reflected light that differentially breaks the symmetry in the transverse direction (in particular along the y axis) is not expected to be larger than 1%.

\(^{21}\)The wavelength stability is better than 0.5 pm and the measured overall comagnetometer noise is \(\approx 0.75\) fT.

\(^{22}\)For this estimate, a uniform pump intensity covering a large (70%) fraction of the cell and 5 pm deviation from the resonance are assumed.
For a pump beam well centered with respect to the cell the net light-shift from retroflection is zero. (b) A misaligned in the y-axis pump beam causes a net vertical light-shift from back reflection off the cell walls.

We note that typically the pumping power is on the order of a few mW; this level of light power has an insignificant effect on the cell heating\textsuperscript{23} [195].

**Probe beam**

The probe laser is identical to the one used for the pump beam, with similar temperature (and wavelength) control. The probe laser is tuned to 769.640 nm (that is $\approx 237$ GHz blue shifted with respect to resonance); this wavelength is a compromise between the requirement of large optical rotation due to electron spin (which results to reduced sensitivity on beam motion, photon shot noise and electronic noise), and the requirements for low photon absorption and probe-induced relaxation and reduced probe light-shift sensitivity to wavelength and photon spin fluctuations.

The probe beam goes through a Faraday isolator to protect the laser from optical feedback. Pairs of cylindrical lenses correct for the intrinsic astigmatism, creating a roughly (as can be perceived by eye\textsuperscript{24}) elliptical beam of area $\sim 0.3 \text{ cm}^2$ (ellipsis axis $\sim 0.5 \text{ cm}$ and $\sim 0.2 \text{ cm}$). A high extinction linear polarizer ($< 10^{-6}$), Casix Glan-Laser Prism\textsuperscript{25}, located after the last mirror, ensures high degree of linear polarization.

\textsuperscript{23}The light energy is converted to (alkali) atomic energy and eventually through the collisions with the N$_2$ buffer gas to heat.

\textsuperscript{24}At the time of the experiment, there was no beam profiler available. This is only an approximate estimate.

\textsuperscript{25}This type of polarizer (effectively a Glan-Taylor polarizer [143] with escape windows for the rejected beam) is based on the total internal reflection at the boundary of a birefringent material with air, where the transmitted ray passes through nearly at Brewster angle.
A glass (quartz) plate, one inch in thickness, where mechanical stress can be applied both manually (through a pair of screws) and electronically (through a piezoelectric actuator), is used to cancel to first order residual circular polarization of the beam. Compared to other types of glass, quartz has low expansion coefficient and therefore exhibits reduced birefringent sensitivity to temperature, and also has relatively large photoelastic response (Pockels’ elasto-optic constants $p_{11}$ and $p_{12}$ are $+0.121$ and $+0.270$ respectively \[59\]).

\[ E_i = E_0 \cos \phi \hat{p} + E_0 \sin \phi \hat{s} \\
E_t = t_p E_0 \cos \phi \hat{p} + t_s E_0 \sin \phi \hat{s} \]

Figure 4.8: (a) Light at the boundary of different optical materials experiences different transmission for the s and p polarization. (b) (Taken from [TK]) Probe light incident off center to the cell experiences optical rotation. In the “sweet spot” light travels through the cell with no change of polarization; for a spherical cell this corresponds to normal incidence. (c) Schematic for light incident off center to a sphere of radius $R$. The angle $\chi$ is defined with respect to the polarization axis of the incident beam, and $\rho$ is the distance from the sphere-light intersection to the point of sphere-center projection onto the plane perpendicular to the incident light ray.

Special care is devoted to align the probe beam so that it goes through the “sweet spot” of the cell [TK]; in this particular configuration the probe optical rotation due to the glass cell (not including the effect of atomic spins) remains insensitivity to first order to beam motion. Light incident at the boundary of two different optical materials (e.g. glass and air) experiences different (amplitude) transmittance $t_p$ and $t_s$ for the linear polarization in the plane of incidence\[26\] (p polarization) and the linear

\[26\] The plane of incidence is defined by the light propagation vector and the vector perpendicular to the boundary surface
polarization perpendicular to the plane of incidence (s polarization) respectively\(^{27}\).

The difference in \(t_s\) and \(t_p\) results in an effective optical rotation when the incident light polarization is a superposition of the s and p polarization\(^{28}\). This can easily be seen in the simple case depicted in Fig. 4.8(a); it is straightforward to show that the optical rotation \(\delta \phi_{opt}\) is given by:

\[
\delta \phi = \arctan \left( \tan \phi \frac{t_s}{t_p} \right) - \phi_0
\]

(4.2)

where \(\phi_0\) is the incident polarization angle and \(t_s, t_p\) are functions of the incident angle \(\theta\). For \(\theta = 0\) (normal incidence) \(t_s = t_p\) and there is no optical rotation, while for \(\theta \neq 0\) optical rotation in nonzero. For a perfectly spherical cell the condition of normal incidence (beam going undeflected through the cell center) results in zero optical rotation and minimum sensitivity (zero to first order) to beam motion (Fig. 4.8(b)). It can be shown that for the light ray of Fig. 4.8(c) the optical rotation due to the boundary can be approximated:

\[
\delta \phi \approx \frac{(\rho/R)^2(n_1 - n_2)^2 \sin(2\chi)}{4n_2^2}
\]

(4.3)

where \(\chi\) is the angle with respect to the axis of incident polarization. For an extended beam, it is straightforward to show that zero first order sensitivity is realized when the center of the (cylindroidally symmetric) beam goes undeviated through the center of the sphere. We note that in the actual experiment, due to the asphericity of the cell, the “sweet spot” is located slightly off-center, and is found by modulating the beam position in both vertical and horizontal dimension and minimizing the response.

\(^{27}\)The amplitude transmittance and reflectance as a function of the incident angle and the linear polarization are known in literature as Fresnel coefficients \(\left[27\right]\).

\(^{28}\)Note that \(t_s\) and \(t_p\) are real. As a consequence for linearly polarized incident light the transmitted light is also linear.
Figure 4.9: (a) (Modified from [TK]) Schematic of the probe beam steering optics that allow independent control of the beam angle and position at the cell center. (b) The cell acts a weakly negative lens. Note that the probe beam covers only a fraction of the cell (not shown here).

The optical design for steering the probe beam is similar to the one described in [TK] (see Fig. 4.9 (a)). A deviator and a pair of lenses allow for independent control of the probe beam propagation axis and the beam position at the cell. The optical setup can be characterized with the help of the “ABCDEF” matrix formalism, applied to paraxial light ray propagation in misaligned systems. The probe beam is steered by translating the first lens in the transverse plane, and therefore the analysis of the setup has to include the effect of transverse misalignment of optical elements. Here, we follow the procedure of [172], defining an overall reference optical axis, which in general is different from the element optic axis. The light ray is described by the the vector \( v = (x, \theta, 1)^\top \), where \( x \) and \( \theta \) are the offset and the angle respectively from the reference optic axis. The translation matrix is given by:

\[
M_t(d) = \begin{pmatrix}
1 & d & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1
\end{pmatrix}
\]

(4.4)

where \( d \) is the distance along the propagation axis traveled by the beam. The effect of lens of focal length \( f \) with its optical center offsetted by \( y \) from the propagation
axis is described (in the paraxial approximation) by:

\[ M_l(f, y) = \begin{pmatrix} 1 & 0 & 0 \\ -1/f & 1 & y/f \\ 0 & 0 & 1 \end{pmatrix} \]  \quad (4.5)

The beam deviator, a pair of matching convex and concave lenses and index-matching fluid to allow the two lenses to slip smoothly past each other, affects the angle \( \theta \) of light ray with respect to the optic axis. The angle and position of the beam at the cell is:

\[ v_c = M_t(d_3) M_l(f_2, y_2) M_t(d_2) M_l(f_1, y_1) M_t(d_1) v_0 \]  \quad (4.6)

The various parameters of the above equation are defined in Fig. 4.9. Under the constraints of \( d_2 = f_1 + f_2 \) (the two lenses are in the Keplerian magnifying telescope arrangement), second lens with no offset from the optical axis \( (y_2 = 0) \), and \( d_{tot} = d_1 + d_2 + d_3 \), the position at the cell \( x_c \) becomes independent of the initial angle with the axis \( \theta_0 \) \((dx_c/d\theta_0 = 0)\) when:

\[ d_1 = \frac{(f_1 + f_2)^2 - f_1 d_{tot}}{f_2^2 - f_1^2} \]  \quad (4.7)

In this case the position and angle at the cell are:

\[ x_c = \frac{f_2(f_1 + f_2)^2 - d_{tot} f_1 f_2}{f_1 (f_2^2 - f_1^2)} y_1 - \frac{f_2}{f_1} x_0 \]  \quad (4.8)

\[ \theta_c = -\frac{y_1 + \theta_0 f_1}{f_2} \]  \quad (4.9)

In the experiment of this thesis a combination of \( f_1 = 100 \text{ mm} \) and \( f_2 = 250 \text{ mm} \) lenses were used.
A beam sampler directs a fraction of the beam to a position detector (quadrant photodiode, with two axis resolution) located at a distance $d_3$ from the second lens (equal to the cell distance), and the detector output through an integral controller (bandwidth of a few Hz) adjusts a piezo-actuated mirror to retain the beam fixed at the position detector (Fig. 4.1). For negligible noise in the optical path from the beam sampler to the position detector and to the cell (distance of $d_3 = 568$ mm) the feedback loop maintains the beam at the “sweet spot” of the cell.

Contrary to the case of [TK], in the current experiment there was no need to find the comagnetometer background (that is, the measured probe optical rotation from sources other than the atomic spin), and, as a consequence, we performed the experiment without adjusting the pump-probe angle and the beam deviator was not used.

The comagnetometer cell is a weakly negative lens (see Fig. 4.9b); for an outer diameter of $D = 24.0$ mm, thickness of $t = 0.2$ mm, and glass refractive index $n = 1.528$ a spherical cell effectively acts as a diverging lens with focal length $f_{\text{cell}} \approx -982$ mm. The defocussing effect of the cell is compensated by a positive lens $f = +500$ mm in close proximity with the cell ($\approx 30$ mm).

The measurement sensitivity is enhanced by adopting a polarization modulation scheme and lock-in detection of the signal, as depicted in Fig. 4.1. Measuring angles with respect to the direction of the probe polarization axis when there is no optical rotation from the atomic spins (e.g. when the atoms are unpolarized), the quarter wave plate and the photoelastic modulator (PEM) are oriented at $0^\circ$ and $45^\circ$ respectively and the final polarizer acts as an analyzer at $90^\circ$, i.e. the transmission axis

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29 From (3.26) it can be seen that, except for the last term, the signal sensitivity does not depend on the pump-probe angle $\alpha$. The term $\alpha P_z$ contributes to the signal noise primarily through fluctuations of the angle $\alpha$, resulting in a noise source that approximately does not depend on the average value of $\alpha$.

30 This was calculated using the OSLO software (student version) for optimization of optical systems.

31 By this we mean that the principal axes (fast or slow) of the wave plate and PEM form $0^\circ$ and $45^\circ$ respectively with the polarization axis of the probe light.
is perpendicular to the nominal linear polarization of probe light. The phase retardation of the PEM is modulated at $f_m = 50$ kHz frequency with amplitude (depth of modulation) $\alpha_m \sim 0.09$ rad. For zero optical rotation from the atomic medium, the $\lambda/4$ wave plate has no effect on the light polarization and the linearly polarized light incident on the PEM acquires a polarization component in the transmission axis of analyzer which results in a photodetector signal ($\propto |E|^2$) oscillating at even harmonics of the modulation frequency (e.g. no first harmonic). When the spins induce optical rotation, the polarization of light through the quarter wave plate becomes elliptical and the PEM creates an interference effect between the two polarization components (at $0^\circ$ and $90^\circ$) which is recorded in the odd harmonics of the photodiode signal. A quantitative analysis is most easily done with the use of the Jones matrix calculus \[^{[104]}\]. It is straightforward to show that the detected signal is given by:

$$S_m = I_0 \sin \left[ \theta_s + \frac{\alpha_m \cos(\omega_m t)}{2} \right]$$

$$\approx I_0 \left[ \frac{\alpha_m^2}{8} + \frac{\alpha_m \theta_s \cos(\omega_m t)}{8} + \frac{\alpha_m^2}{8} \cos(2\omega_m t) \right] + \mathcal{O}(\theta_s^2) + \mathcal{O}(\alpha_m^3) \quad (4.12)$$

where $I_0$ is the incident light intensity scaled by the efficiency (e.g. there are reflection losses on the various surfaces) and $\theta_s$ is the optical rotation angle from the spins to be measured. Equation \((4.12)\) is an expansion for small $\theta_s$ and $\alpha_m$; it can be seen that the first harmonic (with respect to the modulation frequency) is proportional to the optical rotation angle $\theta_s$. The above equations assume that the optical elements have their nominal values; however, in the actual experiment there are deviations from the
ideal case, and for small deviations the detected signal to minimum order is:

\[
\frac{S_m}{I_0} \approx \frac{\alpha_m^2}{8} \left[ 1 + 4\theta_{\text{pol}}\theta_m - 4\theta_s\theta_{\text{wp}} + 4\theta_m\theta_{\text{wp}} - 2\alpha_{0m} (\theta_s + \theta_{\text{wp}}) \right] \\
+ \left[ 2\theta_{\text{wp}} (\theta_s - \theta_{\text{pol}}) + \alpha_{0m} (\theta_s + \theta_{\text{wp}}) \right] + \left[ \frac{\alpha_{0m}}{2} + \theta_s + \theta_{\text{wp}} \right] \alpha_m \cos (\omega_m t) \\
+ \left[ 1 + 4\theta_{\text{pol}}\theta_m - 4\theta_s\theta_{\text{wp}} + 4\theta_m\theta_{\text{wp}} - 2\alpha_{0m} (\theta_s + \theta_{\text{wp}}) \right] \frac{\alpha_m^2}{8} \cos (2\omega_m t)
\] (4.13)

where $\theta_{\text{pol}}, \theta_m$ and $\theta_{\text{wp}}$ are the misalignments in the orientation of the polarization, PEM and wave plate respectively, and $\alpha_{0m}$ is the residual quasi-static phase retardation in the PEM. Equation (4.13) shows that the first harmonic depends on the angle of the $\lambda/4$ wave plate $\theta_{\text{wp}}$ (in a manner similar to the optical rotation angle $\theta_s$); this property is used in the experiment to calibrate the measured photodetector signal with respect to light optical rotation, and to zero the first harmonic offset.

The actual photodiode output has a non-negligible DC offset (the part that cannot be described as harmonic component raised to some power). In Fig. 4.10 a typical photodiode signal is shown in the time domain; the ratio of the DC offset (which does not carry any useful information) to the second (with respect to the photoelastic modulation) harmonic amplitude (peak to peak) is approximately 1:3. The measured offset is very large to be explained by Eq. (4.13), and it is believed that it results from inhomogeneities in the optical components.

There is a first order dependence of the first harmonic amplitude on the residual PEM phase retardation, which varies with the position on the PEM optical element. To minimize the effect of beam position noise on the measured signal (first harmonic) a positive lens $f_p = 150$ mm focuses$^{32}$ the light on the PEM.

In the experiment, a zero-order $\lambda/4$ wave plate$^{33}$ was used, which exhibits a smaller sensitivity on temperature and light wavelength compared to a multi-order wave plate.

---

$^{32}$The linear polarizer used for the analyzer (Glan-Laser polarizer) has a limited acceptance angle (angular field is 6°), which places a constraint on the focal length of the lens.

$^{33}$A zero-order wave plate is constructed by combing two multi-order wave plates; the fast axis of one is aligned with the slow axis of the other and the total optical path length difference is $\lambda/4$.  

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Figure 4.10: Photodiode output in the time domain. There is a large DC offset which is believed to be due to the inhomogeneities in the optical components.

The electronic circuit for the photodiode amplifier is depicted in Fig. 4.11. A two stage amplifier scheme is used to meet the requirements for large gain (to suppress electronic noise) and relatively high bandwidth (BW $\sim 100$ kHz to record the second harmonic signal\[34\]); a voltage amplifier is added after the current-to-voltage converter, resulting in an effective total transresistance: $R_t = (1 + R_2/R_1) R_f$. We note that the photodiode is not reversely biased (with a battery) to decrease the photodiode capacitor (and increase the gain bandwidth - a 10 V reverse bias could result in a approximately 5-fold increase of the bandwidth-); this was done in order to avoid complications associated with voltage drop of the battery during the continuous operation of the comagnetometer\[35]. As discussed in [85], the two stage amplifier scheme compromises the electronic noise. An analysis of the circuit with typical noise terms (from the datasheet of the electronic elements) is described in Fig. 4.11 (c). The dominant (net output) noise term is the amplified (amplification $R_2/R_1$) total noise of the current to voltage amplifier ($e_{t,1}$)(which is dominated by the Johnson noise of the resistor $R_f$ and the input voltage noise of the amplifier), and the overall signal to noise ratio (SNR) only depends on the feedback resistor $R_f$ (that is the overall SNR is to a good approximation equal to the SNR of the first stage). In combination

\[34\] The second harmonic signal is a good measure of the light intensity.

\[35\] The use of a power supply to provide the biasing voltage would require significant filtering to avoid noise coupling [85].
with the total effective feedback capacitor (photodiode capacitor $C_d$ plus amplifier capacitor $C_{am,1}$ plus feedback capacitor $C_1$: $C_f = C_d + C_{am,1} + C_1 \approx 90 \, \text{pF}$), $R_f$ sets the bandwidth of the system ($\text{BW} = 1/(2\pi R_f C_f)$). In the experiment of the thesis $R_f = 549 \, \text{k\Omega}$, $R_1 = 2 \, \text{k\Omega}$ and $R_2 = 30.1 \, \text{k\Omega}$, corresponding to $\text{BW} \approx 300 \, \text{kHz}$ and transresistor $R_t \approx 8 \times 10^6 \, \text{k\Omega}$.

The amplifier output is read by a digital lock-in amplifier (Stanford SRS 830) with a time constant of 3 msec and the first harmonic (with respect to the PEM modulation frequency) is recorded in a computer. We note that the lock-in scheme reduces the effect of electronic noise\footnote{Electronic noise is significantly higher at low frequencies (of a few Hz) compared to the noise at the modulation frequency (50 kHz).} on the recorded signal (first harmonic); however the modulation technique does not alter the sensitivity to other sources of low frequency noise (e.g. light intensity, wavelength, and beam position noise).

In [TK] a Faraday rotator was used to modulate the optical polarization and provide the basis for the lock-in detection. Although fundamentally (that is, as far as the photon shot noise is concerned) the Faraday rotator and photoelastic modulator schemes are identical, the one adopted in this thesis is experimentally more robust and easier implemented. In particular, unlike to the case of Faraday rotator the PEM does not require large current (in fact no current at all) resulting in minimal heating and electromagnetic interference; in addition, there is no need for magnetic shielding and overall the setup is more compact.

**Enclosure**

The pump and probe optical setups are enclosed by 0.5 inches thickness Lexan (a polycarbonate resin thermoplastic) boxes, which significantly reduce air currents (i.e. convective currents, air drafts, and acoustic waves) and beam motion noise associated with air density fluctuations. Rubber seals are used in the seams and contacts to the optical table to provide tight air insulation. Besides isolating the optical elements and
\[ R_f = 549 \, \text{k}\Omega \]
\[ R_1 = 2 \, \text{k}\Omega \]
\[ R_2 = 30.1 \, \text{k}\Omega \]
\[ C_1 = 1 \, \text{pF} \]
\[ C_2 = 20 \, \text{pF} \]
\[ I_{nb,1} = 0.4 \, \text{pA/Hz} \]
\[ I_{nb,2} = 0.4 \, \text{pA/Hz} \]
\[ e_{am,1} = 8 \, \text{pF} \]
\[ e_{am,2} = 8 \, \text{pF} \]
\[ e_{R_1} = \sqrt{4k_BT R_1} \]
\[ e_{R_2} = \sqrt{4k_BT R_2} \]
\[ e_{I_d} = \frac{I_{nb,1}}{\sqrt{1 + (2\pi f R_1 C_1)^2}} \]
\[ e_{I_d} = \frac{I_{nb,2}}{\sqrt{1 + (2\pi f R_2 C_2)^2}} \]

\[ C_f = C_d + C_{am,1} + C_1 \]

\[ \epsilon_{v_1}^2 = \left[ \sqrt{4k_BT R_f} \right]^2 + \left[ \frac{I_{nb,1} R_f}{\sqrt{1 + (2\pi f R_1 C_1)^2}} \right]^2 + \left[ \frac{I_{nb,2} R_2}{\sqrt{1 + (2\pi f R_2 C_2)^2}} \right]^2 \]

\[ \epsilon_{v_2}^2 = \left[ \sqrt{4k_BT R_2} \right]^2 + \left[ \frac{I_{nb,2} R_2}{\sqrt{1 + (2\pi f R_2 C_2)^2}} \right]^2 + \left[ \frac{\sqrt{4k_BT R_1 R_2}}{R_1 \sqrt{1 + (2\pi f R_1 C_1)^2}} \right]^2 \]

Figure 4.11: PD: Photodiode (OSI Optoelectronics, PIN-5D), OP1: Operational Amplifier (OPA627), OP2: Operational Amplifier (OP27), \( I_d \): Photodiode dark current, \( C_d \): Photodiode Capacitance, \( e_{am} \): Amplifier input voltage noise, \( C_{am} \): Amplifier input capacitance, \( I_{nb} \): Input bias current noise, \( e_R = \sqrt{4k_BT R} \): Johnson noise of the resistor, \( k_B \): Boltzmann constant, \( T \): Absolute temperature. (a) Photodiode amplifier circuit. A two-stage scheme is used. (b) Values for the various electronic components. For the noise sources, typical values from the data sheet are reported. (c) Equivalent noise circuit of the current to voltage converter and the voltage amplifier. The total output noise is dominated by the Johnson noise of \( R_f \) and input noise of the first operational amplifier.
beam path from (air) room perturbations, the enclosure contributes to maintaining a robust thermal steady state and decreases the Rayleigh number (proportional to the thickness/height of the fluid layer) well below the critical value for convective flow \[126\]. The Lexan boxes that surround the probe (signal) and pump (as part of intensity feedback) photodetectors are painted black to avoid light leaking from the spin source setup (see Section 4.7). In more recent comagnetometer experiments, the whole optical setup is under vacuum \[34, 33\], eliminating issues arising from beam motion due to air density fluctuations. Alternatively to vacuum, the use of $^4$He with atomic polarizability (in the optical frequencies) more than an order of magnitude smaller than effective air polarizability, reduces the effect of density fluctuations on the beam position$^{37}$.

**Optical table and the rest**

The experiment is performed in a dedicated room, in the basement of the Princeton Physics department, providing a relatively immune environment to external temperature perturbations. Special care is taken to mount the optical components rigidly for mechanical stability. An optical table (TMC), with pneumatic vibration isolation providing damping of both vertical and horizontal motion, resonant frequency $\sim 2$ Hz, is used in the experiment. The relatively low center of mass (see Fig. 4.12) results in increased gravitational stability (resistance to tilt), and the honeycomb structure of the table top contributes to the mechanical and thermal rigidity$^{38}$. The table top is made of non-magnetic$^{39}$ stainless steel; the use of Super Invar (readily available) with low thermal expansion coefficient would be more preferable for the experiment, despite being slightly magnetic.

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$^{37}$Compared to vacuum, $^4$He has the advantage of relatively large heat conduction.

$^{38}$A clear explanation of the optical table characteristics can be found in the TMC website: [http://www.techmfg.com/techbkgd/intro.htm](http://www.techmfg.com/techbkgd/intro.htm)

$^{39}$The non-magnetic character is not essential, since the comagnetometer is magnetically shielded and the homogeneity of the magnetic fields in the spin-source are much more affected by the comagnetometer magnetic shields.
Electronic components (e.g. laser drivers, lock-in amplifier) that cause vibrations (originating from the fans) are placed on a self which is not rested on the optical table. To reduce vibration coupling, the connecting cables are long enough to provide a low tension (i.e. not stiff) connection.

### 4.2 Comagnetometer Field and Light-shift adjustment

In the search of anomalous fields, it is of fundamental importance to minimize the effect of noise sources in the comagnetometer signal. In addition to building a robust apparatus (see previous Section), enhanced noise performance is realized by appropriately tuning the magnetic field and light-shift experienced by the coupled spin ensembles.

---

*We remind that the term anomalous is used to identify spin-coupling fields of different nature than magnetic fields that have not been observed so far.*
From Eq. (3.26) it can be seen that fluctuations or drift in e.g. $B_x$ or $B_y$ have no effect (to first order) on the comagnetometer signal provided that $\delta B_z$ is zero. Similar arguments also hold for the pump and probe light-shifts. As discussed in [TK], a lock-in technique is implemented to reliably tune the comagnetometer at the point of optimum operation. In particular, a low frequency (much smaller than the comagnetometer resonance frequency $f_0 \approx |\gamma_n \lambda M^n|$ and the inverse of the characteristic relaxation time (see Section 3.3)), square wave modulation excites the comagnetometer and the quasi-steady-state response is measured. The modulation scheme separates the comagnetometer contribution to the measured signal from the contribution of the optical elements in the polarization rotation of probe light.

4.2.1   Zeroing of $\delta B_z$

From Eq. (3.26) it can be seen that to first order:

$$\frac{\partial P_m}{\partial B_y} \propto \delta B_z$$

which suggests that the quasi-steady-state response to a small $B_y$ modulation is (approximately) proportional to the residual field $\delta B_z$. Experimentally, the derivative of the comagnetometer signal with respect to $B_y$ can be approximated by subtracting the (steady state) comagnetometer signals corresponding to $B_y$ fields that differ by $2\Delta B_y$, and dividing this difference by $2\Delta B_y$.

The $\delta B_z$ zeroing process is illustrated in Fig. 4.13. The longitudinal field is increased by $\Delta B_z$ and a smoothed square wave $B_y$ modulation is applied: the $B_y$ field is smoothly increased by $\Delta B_y$ and after the transients have decayed the comagne-
tometer signal is measured by averaging over some time $t_{avg}$; similarly, the average value of the steady state comagnetometer signal is measured for a $-\Delta B_y$ change and the difference $\Delta P_{m,1}$ in the average response to $B_y$ modulation is calculated. The longitudinal magnetic field is decreased by $\Delta B_z$ and the modulation procedure is repeated ($\Delta P_{m,2}$). From linear interpolation (or extrapolation depending on the initial magnetic field) of the two measurements the magnetic field $B_z$ for zero response to $B_y$ modulation ($\Delta P_m = 0$) is estimated.

The applied smoothed square wave modulation has the form:

$$\Delta B_y(t) = \Delta B_y \tanh [s \sin (2\pi f_m t)]$$

(4.15)

where $\Delta B_y$ and $f_m$ are the amplitude and frequency of modulation respectively. The parameter $s$ characterizes the sharpness (duration) of transition; this has to be slow enough so that it does not excite significant transverse oscillation (that is the Fourier components of $\Delta B_y(t)$ have negligible power in the resonance frequency, which for the thesis experiment is 21 Hz).
Table 4.1: Parameter values used in the zeroing routines.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Zeroing Routine</th>
</tr>
</thead>
<tbody>
<tr>
<td>$s$</td>
<td>8</td>
<td></td>
</tr>
<tr>
<td>$f_m$</td>
<td>0.1 Hz</td>
<td>All</td>
</tr>
<tr>
<td>$t_m$</td>
<td>2 sec</td>
<td></td>
</tr>
<tr>
<td>$R_{tot}^e/\gamma_e$</td>
<td>20 $\mu$G</td>
<td></td>
</tr>
<tr>
<td>$\Delta B_z$</td>
<td>3 $\mu$G</td>
<td>$\delta B_z$</td>
</tr>
<tr>
<td>$\Delta B_y$</td>
<td>2 $\mu$G</td>
<td>$B_y$</td>
</tr>
<tr>
<td>$\Delta B_z$</td>
<td>1 $\mu$G</td>
<td>$B_y$</td>
</tr>
<tr>
<td>$\Delta B_x$</td>
<td>5 $\mu$G</td>
<td></td>
</tr>
<tr>
<td>$\Delta B_y$</td>
<td>1 $\mu$G</td>
<td></td>
</tr>
</tbody>
</table>

The linear approximation for finding the zero response, and the estimation of the derivative $\partial P_m/\partial B_y$ from finite differences hold in the region where the detuning from the compensation point and the transverse fields and pump light-shift are small compared to $R_{tot}^e/\gamma_e$ (in particular, the error of estimation to lowest order is quadratic e.g. $(\gamma_e \delta B_z/R_{tot}^e)^2$; this places a limit on the magnitude of modulations $\Delta B_y$ and $\Delta B_z$. Small modulations have little effect on the comagnetometer polarization, which could be masked from signal drifts during the zeroing operation. In practice, the limit of how small the modulations should be is set by the comagnetometer signal noise.

The values of modulation parameters are given in Table 4.1.

4.2.2 Zeroing of the transverse fields.

The zeroing of the transverse fields can be performed in a likewise manner as for $\delta B_z$.

We note that for small deviation from the optimum comagnetometer state, and to

$^{44}$Effectively, the zeroing routine depends on the validity of the approximation: $\partial^2 P_m/\partial B_y \partial B_z \approx (\Delta P_{m,1} - \Delta P_{m,2}) / (4\Delta B_y \Delta B_z)$. $^{45}$The detuning of the fields also affects the comagnetometer noise; however for detunings from the optimum point < 50 $\mu$G the noise is not significantly changed.
the lowest order is small quantities:

$$\frac{\partial P_m}{\partial B_z} \approx \frac{\gamma_e P_e}{B_c R_{\text{tot}}} q_y$$  \tag{4.16}

$$\frac{\partial^2 P_m}{\partial B_z^2} \approx \frac{2 P_e \gamma_e^2}{B_c R_{\text{tot}}^2} q_x$$  \tag{4.17}

\[ q_y = B_y + \frac{\gamma_e B_c}{R_{\text{tot}}} L_x + \Omega_y / \gamma_n \]  \tag{4.18}

\[ q_x = B_x + \frac{\Omega_x}{\gamma_n} - \frac{\gamma_e B_c}{R_{\text{tot}}} L_y + \frac{\gamma_e B_c}{R_{\text{tot}}^2} \frac{\Omega_y}{\gamma_{\text{eff}} R_{\text{tot}}} \]  \tag{4.19}

The quantity \( q_y \) is zeroed by adjusting the \( B_y \) field to minimize (zero) the response to \( B_z \) modulation (see Fig. 4.14). The \( L_x \) term, decoupled from the magnetic field, is independently zeroed with an appropriate routine, so that zeroing \( q_y \) corresponds to adjusting the environmental field in the \( y \) axis to \( B_y = -\Omega_y / \gamma_n < 3 \text{ nG} \).

Similarly, the zeroing of \( q_x \) results in \( B_x \approx -\Omega_x / \gamma_n - \gamma_e B_c \Omega_y / (\gamma_{\text{eff}} R_{\text{tot}}) \sim 0.9 \mu\text{G} \) (\( L_y \) is small since nominally no light field is in the \( y \) direction). The finite value of the \( B_x \) field does not compromise the noise performance of the comagnetometer. In fact, the sensitivity to noise depends on the derivatives of \( P_m \) with respect to the
fluctuating parameters, so that the zeroing of $q_x$ ($q_y$) instead of $B_x$ ($B_y$) provides optimum comagnetometer performance. The experimental implementation of $q_x$ zeroing is illustrated in Fig. 4.15: the $\delta B_z$ field is modulated between zero (found with the application of the appropriate routine) and a finite value $\Delta B_z$, and the $B_x$ field is tuned to zero the response. In general, the comagnetometer response to the asymmetric modulation depends also on $q_y$; therefore, the $q_x$ routine is applied only after $q_y$ has been zeroed, and there is no first order dependence of the comagnetometer signal on $\delta B_z$.

We note that the $q_x$ zeroing may become more robust to uncertainty in the $q_y$ term by approximating the second derivative in (4.17) with the experimentally measurable finite difference:

$$\frac{\partial^2 P_m}{\partial B_z^2} \approx P_m (+\Delta B_z) - 2P_m(0) + P_m (-\Delta B_z)$$  \hspace{1cm} (4.20)

which does not depend on $q_y$.

![Figure 4.15: Routine for zeroing $B_x$.](image)
4.2.3 Zeroing of the Light-shifts

In principle, the pump and probe light-shift can be zeroed by exploiting the comagnetometer signal dependence on the $L_x L_y$ term. To lowest order:

\[
\frac{\partial P_m}{\partial L_x} \approx \frac{\gamma_e^2 P_e}{R_{\text{tot}}^2} \left( \delta B_z + L_z - \frac{\Omega_z}{\gamma_{eff}} \right) \left( \delta B_{z=0} \right) \frac{\gamma_e^2 P_e}{R_{\text{tot}}^2} \left( L_z - \frac{\Omega_z}{\gamma_{eff}} \right) \tag{4.21}
\]

and routines similar to those described above can be applied to zero $L_x$ and $L_z$. The probe light-shift ($L_x$) is modulated using the stress plate (Section 4.1.5) that affects the probe photon spin $s_m$, while the pump light-shift $L_z$ is adjusted by changing the light wavelength.\textsuperscript{46} Experimentally, it was found that the zeroing routines based on Equations (4.21) and (4.22) are not robust, giving inconsistent results: the changes in the pump wavelength that are required from the routines are large enough to have a significant effect on the pumping rate (and overcome the short term comagnetometer noise), leading to spin polarization and effective field drifts; in addition, there is a considerable settling time (on the order of a few seconds) to step changes of the pump wavelength, making the zeroing routine susceptible to environmental drifts.

In the experiment of this thesis, no automated zeroing routine for the pump light-shift was applied. From the absorption spectrum (see Section 4.6.1) the absorption resonance frequency is estimated and the pump light is set at this wavelength using a wavemeter (Agilent / HP 86120B); no further adjustment is performed, except for periodical (approximately every week, when the experiment was not running) manual verification (with the wavemeter) that the wavelength has not drifted from the resonance condition. The accuracy of $L_z$ zeroing is limited by the uncertainty of the fit ($\sim 2$ pm), and corresponds to $\delta L_z \sim 0.1 \mu G$ (see Eq. (2.66)). The (approximate)

\textsuperscript{46}The wavelength modulation is preferred to intensity modulation, because of the smaller effect on pumping rate for the same light-shift change.
zeroing of $L_z$ is also demonstrated in Fig. 4.18 where the response to (smoothed square wave) $B_y$ modulation is plotted as a function of the $\delta B_z$ field: whether the curve is symmetric with respect to $(0,0)$ depends on how well the quantity $L_z - \Omega_z/\gamma_{eff} \approx (L_y + 2 \text{ nG})$ has been zeroed.

**Zeroing of the $L_x$**

The probe light-shift is proportional to the probe photon spin $s_m$, which can be reliably measured and zeroed using the probe pumping term:

$$P_m^{pr} = \frac{R_m R'_{tot}}{R_{tot}^2 + \gamma_e [\delta B_z + L_z - \Omega_y/\gamma_{eff}]}$$  \hspace{1cm} (4.23)

For the zeroing process, the pump light is blocked in order to suppress all the terms in (3.20) except for $P_m^{pr}$ (that is, all the terms proportional to $P^e_z$). The drop in electron polarization affects the condition for compensation point ($B_c = -\lambda M^e P^e_z - \lambda M^n P^n_z$) and the applied longitudinal field is adjusted to the resonant value $B_{z0}^{pr}$ found by scanning the $B_z$ field at no pump light and finite $s_m$. A square $B_z$ modulation between the resonant field $B_{z0}^{pr}$ and $B_{z0}^{pr} + \Delta B_z$, where: \(\gamma_e^2 \Delta B_z^2 + R_{tot}^2 \gg (R_m R'_{tot})\) with $R_{tot}' = R_{tot} - R_{pu}$, is applied and the comagnetometer response is zeroed by tuning the stress plate (in the experiment the applied detuning was $\Delta B_z \approx 30 \text{ \mu G}$).

Because of the effect on polarization (the pump beam remains off), the $B_z$ scan is not performed during the experiment; instead, the resonant field $B_{z0}^{pr}$ found before the run of the experiment (but under approximately similar conditions) is used. Setting the longitudinal field to $B_{z0}^{pr}$ maximizes the comagnetometer response to $s_m$ modulation; however, the routine gives reliable, robust zeroing for a range of values close to $B_{z0}^{pr}$, so that (small) drifts in $B_{z0}^{pr}$ during the experiment do not compromise the efficiency of the routine.
Spin-exchange collisions between the alkali and noble gas atoms polarize the K atoms and a small electron polarization is maintained in the absence of the pump beam (assuming that the noble gas has been partially polarized) \( R_{se}^p P^n_z / R_{tot}^e \sim 0.4\% \) (this estimate assumes that the \(^3\)He polarization does not change appreciably). The terms of (3.26) proportional to \( P_z^e \) are suppressed (compared to normal operation), but not eliminated, and there remains (for the general comagnetometer state) a sensitivity to transverse magnetic fields. Therefore, the \( L_x \) zeroing routine is applied after the transverse fields have been zeroed; in particular for \( B_y \), the routine effectively zeroes \( q_y \approx B_y + \gamma_e B_c L_x / R_{tot}^e \), and the \( B_y \) and \( L_x \) routines should be run in iteration.

As discussed in [TK], in the actual, experimentally realized comagnetometer the \( L_x \) zeroing based on the probe pumping is not completely equivalent to the routine based on the \( L_z L_x \) term; spatial non-uniformity of the \( L_x \) (due to non-homogeneous probe intensity or photon spin) is weighted differently in the two cases (when \( L_z \) depends on position) and the zeroing of the average \( s_m \) does not eliminate first order sensitivity to \( L_z \). However, in the case of the thesis experiment the pump light-shift has small variation in the probe beam region and the two routines for zeroing \( L_x \) lead to approximately identical results.

4.2.4 Zeroing of pump-probe nonorthogonality

The pump-probe nonorthogonality \( \alpha \) can be zeroed by comparing the measured signal at high and zero pump intensities [TK]. In the limit of high pumping rate \( (R_{pu} \text{ much higher than any other rate}) \) the comagnetometer signal is

\[
P_m |_{R_{pu} \to \infty} \approx \frac{R_{pu}^e}{R_{tot}^e} \alpha + \frac{\gamma_e R_{pu}^e}{R_{tot}^e} \left( L_y \right. - \left. \frac{\Omega_y}{\gamma_{eff}} \right) \frac{R_{pu} \to \infty}{\to} \alpha
\]

and for \( R_{pu} = 0 \) the signal becomes \( P_m |_{R_{pu}=0} \approx 0 \), assuming that the probe photon spin has been zeroed. As mentioned above, in the absence of the pump beam, spin-
exchange collisions with the $^{3}$He atoms maintain a small electron polarization and subsequently the measured signal depends on the magnetic fields even when the pump beam is modulated off. To suppress this sensitivity and acquire a robust reference point of zero comagnetometer signal a large $\delta B_{z}$ field is applied while the pump beam is blocked. The angle $\alpha$ can be adjusted manually by steering the pump or probe beam.

In terms of the comagnetometer noise, the zeroing of $\alpha$ suppresses the noise associated with the fluctuations of $P_{z}^{e} \approx R_{pu}/R_{tot}^{e}$, which is estimated to be smaller than one part to 5000 for the time scales of the measurement ($\sim 10$ sec). Polarization noise $\delta P_{z}^{e}/P_{z}^{e} = 5 \times 10^{-4}$ corresponds to an effective anomalous field noise (as far as the pump-probe nonorthogonality is concerned): $\alpha R_{tot}^{e} \delta P_{z}^{e}/(\gamma_{e} P_{z}^{e}) \sim 2 \times 10^{-4} \alpha$ fT/rad; this suggests that an angle $\alpha \sim 10$ µrad would have an effective field noise $\sim 0.1$ fT, which is significantly smaller than the noise of the experiment (see Section 4.6.6). In the thesis experiment, the approximate zeroing of $\alpha$ was performed initially (before the start of the acquisition) and then only sporadically (approximately every couple of weeks) by steering the probe beam; this was enough to maintain the angle $\alpha$ smaller than 10 µrad. No automated routine for the pump-probe nonorthogonality was implemented to avoid the polarization perturbation (which results in significant signal drifts) and (appreciable) change of the probe optical path.

\footnote{As discussed in Section 1.1.5, the position of the probe beam at the cell (approximating the cell as a point) is maintained at the “sweet spot” even when the probe propagation angle changes using the variable wedge (deviator).}

\footnote{This is a rather conservative estimate. The main source of electron polarization fluctuations is the pump intensity noise. Although not directly evaluated in the cell, the pump intensity noise measured inside the enclosure (see Section 4.1.3) is less than one part to 1000, which corresponds to $\delta P_{z}^{e}/P_{z}^{e} = 5 \times 10^{-4}$ for $P_{z}^{e} = 0.5$.}
4.2.5 Field angular misalignment

So far, it was implicitly assumed that the applied fields (through the coils) were aligned with the x, y and z axes. Small misalignments (estimated to be $\sim 10^{-3}$ rad) of the field axes from the xyz system does not compromise the zeroing procedure, and after all the fields and light-shifts have been tuned the effect of the field misalignment in the comagnetometer noise is insignificant (however, there is a small correction to the calibration as will be discussed in the next section).

4.3 Calibration

No (large, known) anomalous spin coupling fields can be applied to the comagnetometer in order to calibrate the apparatus. Therefore, an indirect conversion of the signal measurement to effective magnetic units has to be used. In principle, the calibration can be performed by applying known rotation $\Omega_y$, or light-shift $L_y$, which can be modulated in order to separate the comagnetometer response from other sources of optical rotation of the probe beam. The experiment of this thesis does not provide the possibility to reliably apply an accurate, controlled rotation (though, this is not the case in [34]). The light-shift calibration requires accurate knowledge of the (not easily known) beam intensity in the measurement volume; in addition, large light-shift is associated with non-negligible optical pumping, which adds an extra term in the Bloch Eq. (3.2) and the signal solution (3.26) has to be modified. In [34] a calibration based on the comagnetometer response to a low frequency, sine wave modulation of $B_x$ field according to Eq. (3.80) was proposed and implemented.

In this thesis, the calibration was extracted from the slope (derivative) of the response to $B_y$. Assuming that $\delta B_z$ and $L_z$ are zero, and neglecting the small contri-
\[ \frac{\partial^2 P_m}{\partial B_y \partial \delta B_z} = S \approx \frac{\gamma_e P_e}{B_c R_{tot}} (1 - \mathcal{C}) \]  
\[ \mathcal{C} = C_{se}^{me} + D_{se}^{en} + F_{sd}^{m} \]  

The coefficients \( \beta_e \), \( \beta_n \) and \( \beta_\Omega \) that multiply \( b_y^e \), \( b_y^n \) and \( \Omega_y \) in Eq. (3.26) can be expressed in terms of the \( S \) and the longitudinal field at the compensation point \( B_c \):

\[ \beta_e \approx S B_c \]  
\[ \beta_n \approx -\left(1 - \frac{\delta B_z}{B_c}\right) \beta_e \]  
\[ \beta_\Omega \approx -\left(1 - \frac{\gamma_{eff} \delta B_z}{\gamma_n B_c}\right) \frac{1}{\gamma_{eff}} \]

The comagnetometer signal (projection of electron polarization along the direction of the probe beam) induces Faraday rotation in the linearly polarized light (see Section 2.8.1); the optical rotation, through a combination of a quarter-waveplate, Photoelastic Modulator and polarizer (see Section 4.1.5), is registered on the modulation frequency Fourier component (first harmonic) of the light intensity, which is detected by the photodiode. After current to voltage conversion and lock-in demodulation and amplification the signal (in Volts) is recorded by the computer. The recorded voltage \( V_{rec} \) is related to gyroscopic rotation and anomalous fields through the equation:

\[ V_{rec} \approx \kappa_c \left(b_y^e - b_y^n - \Omega_y / \gamma_{eff}\right) \]  
\[ \kappa_c = G_{lc} \times (q_e R_t) \times \left(\frac{1}{\sqrt{2}} F_{\alpha m}\right) \times \left(\frac{1}{2} |K| d_c r_c f_{osc} D(\nu)\right) \times S B_c \]

where \( G_{lc} \) is the lock-in amplification (which can be selected over a range, and in the experiment an amplification of 50 was used), \( q_e \) is the electron charge, \( R_t = 8 \times 10^6 \) \( \Omega \).
is the total transresistance of the photodiode amplifier, $F \sim 1.2 \times 10^{15}$ photons/sec is the flux of photons detected, $\alpha_m \sim 0.09$ rad is the amplitude of the PEM retardation, $[K]$ is the alkali-metal density, $d_c$ is the cell diameter (approximately equal to the path length of the probe light), $r_e$ and $c$ are the electron radius and speed of light respectively and $D(\nu)$ was defined in Eq. (2.63). The $1/\sqrt{2}$ factor comes from the fact the lock-in outputs the rms value.

In the actual experiment, $S$ is approximated from the slope in the $\delta B_z$ zeroing: $S \approx (\Delta P_{m,1} - \Delta P_{m,2}) / (2\Delta B) = S_{sl}$, and the longitudinal field $B_c$ is approximated by the applied field $B^a_c = B_c - B^e_z$ (that is, we neglect the environmental field $B^e_z$, which is estimated to be 10 $\mu$G originating mainly from magnetic shields). We then have:

$$\beta_e \approx -\beta_n \approx -\gamma_{eff} \beta_\Omega \approx S_{sl} B^a_c = \mathcal{B}$$

Taking into account the finite fields $\Delta B_z$, $\Delta B_y$ in the estimation of the derivative $(\frac{\partial^2 P_{m}}{\partial B_y \partial \delta B_y})$, the environmental field $B^e_z$, the potential misalignment of the applied field with the z axis (defined by the axis of pump propagation), the possibility of an initial (before the start of the zeroing) comagnetometer state with small but finite longitudinal field $\delta B_{z0}$ and pump light-shift $L_z$, and a finite $\delta B_z$ after the zeroing
routine, one can show that up to second order in small quantities:

\[ \beta_e \approx B (1 + \mathcal{W}) \]  
\[ \beta_n \approx -B \left( 1 + \mathcal{W} - \frac{\delta B_z}{B_z^2} \right) \]  
\[ \beta_\Omega \approx -\gamma_{\text{eff}} B \left[ 1 + \mathcal{W} - \frac{\gamma_{\text{eff}} \delta B_z}{\gamma_n B_z^2} \right] \]  
\[ \mathcal{W} \approx \frac{B_z^{\text{ev}}}{B_z^n} + \theta^2 + \frac{\psi^2}{2} + 2\phi^2 + \left( \frac{R_{\text{se}}^c}{\gamma_e \lambda M^c P_{e,c}^z} + 2 \frac{R_{\text{tot}}^n}{\gamma_n^2 \lambda M^n P_{n,z}^z} \right) \psi \]
\[ + \frac{\gamma_e^2}{R_{\text{tot}}^c} \left[ -\delta B_z^2 + 3\delta B_{z0}^2 + \Delta B_z^2 + 2\delta B_z \tilde{\Omega}_z - 2\delta B_z L_z - 4\delta B_{z0} \tilde{\Omega}_z \right] \]
\[ + 4\delta B_{z0} L_z - 2B_z^2 \phi \Omega_y/\gamma_{\text{eff}} \]  
\[ + \frac{\gamma_e}{R_{\text{tot}}^c} \left[ -2B_{z0} \phi + \left( 2B_{z0} + L_z - \tilde{\Omega}_z \right) \psi \right] \]
\[ - 2\phi \Omega_x/\gamma_n \]  
\[ + \mathcal{O} \left( 10^{-7} \right) \]  

where \( \phi, \theta, \psi \) are the Euler angles of the coil frame system with respect to the xyz axes (see Fig. 4.16). The above equations describe the functional dependence (up to second order) of the calibration slopes on various experimental parameters, but should not be taken as exact (even up to the second order). In the actual experiment the main deviation of the estimated calibration from the actual value results from gradients in the polarization and fields. Although Equations (4.33)-(4.36) predict the dependence of the measured calibration on the gradients\(^50\) (e.g. there are regions in the comagnetometer cell contributing to signal that \( \delta B_z \) is nonzero), in experiments where \( \beta_\Omega \) could be measured accurately it was found that the calibration based on \( \partial^2 P_m/\partial B_y \partial \delta B_y \) exhibits a higher sensitivity on gradients than the one predicted from Eq. (4.33)-(4.36). Typically the disagreement between \( \beta_\Omega \) (as measured from actual rotations) and \( \gamma_{\text{eff}} B \) (as derived from \( \delta B_z \) zeroing) is \( \sim 10\% \). A theoretical understanding of this behavior is still pending. As demonstrated in [34], the calibration based on the low frequency response to \( B_x \) modulation is more accurate and

\(^50\)Even if the gradients from the applied magnetic field are negligible, the K spin magnetization is not homogeneous due to pump light absorption, which could potentially result in measurable gradients, especially at high temperatures.
robust to gradients; however, this method was proposed after the end of the thesis experiment.

![Figure 4.16: Definition of the Euler angles, defined with respect to the “fixed” (xyz) and “rotated” (XYZ) frame.](image)

We stress that Equations (4.33) - (4.36) predict $\sim 5\%$ difference between the measured slope $B$ and the actual calibrating factor $\beta_e$ for the thesis experiment. Crude measurements of rotational signals from exciting rotational modes on the optical table confirmed this difference (see Section 4.6.4).

The potential systematic deviation of the estimated calibration from the actual value does not affect the nature of the data analysis; the reported anomalous field estimate and uncertainty may have to be scaled by a correction factor (not larger than 1.1, see Section 4.6.4), but the relative uncertainty remains the same. In addition, the noise in the calibration has an insignificant effect on the relative uncertainty of the anomalous field, since the implemented modulation technique (modulation of the spin source, see Section 5.1) eliminates the constant background signal from the earth rotation $\Omega_y/\gamma_{eff} \sim 271 \, \text{fT}$.

\[51 \text{In fact, if the findings of [34] also hold for the experiment of this thesis, the actual field values and uncertainties are smaller than those reported.}\]

\[52 \text{The calibration uncertainty is directly reflected on the measurement of this rotational component.}\]
4.4 Pump power optimization

The sensitivity of the comagnetometer to anomalous fields depends on the pumping rate $R_{pu}$; taking into account that $P_e \approx R_{pu} / (R_{pu} + R_{sd,t}^e)$, where $R_{sd,t}^e = R_{tot}^e - R_{pu}$ is the total loss rate of longitudinal electron polarization, it is straightforward to show that the highest sensitivity is realized when $R_{pu} = R_{sd,t}^e$ (this is the $R_{pu}$ value that maximizes $P_e / R_{tot}^e$), or equivalently when $P_z = 0.5$.

In principle, the optimum $R_{pu}$ can be found from the comagnetometer calibration (approximately proportional to $\frac{\partial^2 P_m}{\partial B_y \partial B_y} \approx \gamma_e P_e / R_{tot}^e$); however, this procedure, besides being lengthy in time, is susceptible to large drift noise arising from the changes in (noble gas) polarization. In the thesis experiment, the pump intensity is adjusted to its optimum value by maximizing the response to a sine wave modulation of the $B_y$ field. Following the procedure described in Section 3.4, one can show that for a longitudinal field detuned from the compensation point by a finite field $B_z = B_c + q\omega / \gamma_e$, for the conditions $\omega \gg (|\omega_e|, |\omega| / q^2)$, the comagnetometer response to a transverse field $B_y = B_{y0} e^{i\omega t}$ is:

$$P_m \approx \frac{\gamma_e P_e}{2 R_{tot}^e (\omega^2 - \omega_0^2)} B_y$$  \hspace{1cm} (4.37)

$$\approx \frac{\gamma_e P_e}{2 R_{tot}^e \omega^2} B_y, \quad \omega \gg R_{tot}^e / q$$ \hspace{1cm} (4.38)

From the last equation it can be seen that the maximum response is realized when $R_{pu} \approx R_{sd,t}^e$.

Before the start of the measurement, the intensity was adjusted to its optimum value using the above described routine with $\omega = 2\pi 100 \text{ rad/s}$ and $B_y = 1 \mu\text{G}$. The condition for $R_{pu} \approx R_{sd,t}^e$ was also verified by the measurement of $R_{sd,t}^e$ (see Section 5.3). In practice, $\omega$ is limited by the bandwidth of the lock-in measurement of optical rotation and the computer processing time (the demodulation of the oscillating comagnetometer response is performed by the computer). For the described experiment the finite $\omega$ affects the optimum $R_{pu}$ to less than 1%.
4.6.2). The $^3$He magnetization correspondingly to this optimum pump power was recorded, and was used as the set point of a feedback mechanism that constrained $^3$He polarization fluctuations by adjusting the pump power (and consequently the alkali polarization). During the actual experiment the pump power was determined by this feedback mechanism; for reasonably small drifts in oven temperature and probe wavelength and for negligible $^3$He leakage through the cell wall over the course of a month, the pump power that maintains the nuclear magnetization at the set point is to a good approximation equal to the optimum value.

4.5 Effect of comagnetometer noise in the tuning routines

In general, the effect of comagnetometer noise in the tuning routines is quite complex, because of the non-trivial noise spectrum (see Section 4.6.6) and the dependence of noise on the fields (which are modulated during the application of the routines). Here, we consider the simplified picture of a constant comagnetometer noise spectrum, and we drop the noise dependence on the modulated parameters. The fields are adjusted from a linear interpolation between the two points oriented (approximately) symmetrically around the center axes (the ordinate is proportional to the response to modulation and the abscissa is the step field to be zeroed), and the zero crossing (and slope) uncertainty can be found from the chi-square fitting to a straight line.[150] One can then show that the uncertainties in the fields due to the comagnetometer

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[54] This is found from the applied longitudinal field that zeroes $\delta B_z$.

[55] The optimum value, being a stationary point with respect to the ratio of pump rate to spin destruction rate, has no first order sensitivity to this ratio.
noise are (to lowest order):

\[
\begin{align*}
\delta B_z^{ns} & \approx \frac{B_c}{2\Delta B_y} \frac{\delta b_{eff}^{ns}}{\sqrt{2t_m}} \sim 0.02\ \mu G \quad (4.39) \\
\delta B_y^{ns} & \approx \frac{B_c}{2\Delta B_z} \frac{\delta b_{eff}^{ns}}{\sqrt{2t_m}} \sim 0.01\ \mu G \quad (4.40) \\
\delta B_x^{ns} & \approx \frac{B_c R_{tot}^e}{\gamma_e \Delta B_z^2} \frac{\delta b_{eff}^{ns}}{\sqrt{2t_m}} \sim 0.05\mu G \quad (4.41) \\
\end{align*}
\]

(4.42)

and in the calibration:

\[
\frac{\delta S^{ns}}{S} \approx \frac{B_c}{2\Delta B_z \Delta B_y} \frac{\delta b_{eff}^{ns}}{\sqrt{2t_m}} \sim 0.5\% \quad (4.43)
\]

where \( S \) was defined in Eq. (4.25), and \( \delta b_{eff}^{ns} \) is the effective magnetic field noise per unit bandwidth of the comagnetometer; for the numerical evaluation, it was assumed that \( \delta b_{ns} \approx \delta \tilde{b}_{ns} \sim 2\ \mu T/\sqrt{\text{Hz}}. \) As mentioned before, the above equations should only be considered as rough estimates, associated with the signal noise; in the actual experiment the uncertainty is significantly larger (relative uncertainty \( \sim 5\% \)) than the one predicted from Equations (4.39)-(4.43)\(^{56}\).

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\(^{56}\)For an elaborate discussion on the issue the interested reader is referred to [34].
4.6 Comagnetometer Characterization

We now evaluate important experimental parameters, and the performance of the comagnetometer as a sensitive probe of anomalous spin coupling fields is characterized.

4.6.1 Optical Absorption

The optical absorption of K provides information about the resonant wavelength for which the pump light-shift becomes zero; in addition, it gives a measure of the alkali density and the noble gas density assuming knowledge of the pressure broadening due to the noble gas. For the optical absorption spectrum the transmitted power of the probe laser beam through the cell is recorded as the frequency of laser is scanned through resonance. For this measurement, the quarter-wave, PEM and analyzer were removed and there was no pump light and $^3$He polarization. The $B_z$ field (perpendicular to the probe propagation axis) was increased to a few mG to eliminate any effects from electron spin polarization (created by residual probe photon spin polarization). The transmitted signal is normalized with the light power incident to the cell, monitored by a separate photodiode, and is amplified for optimum use of the computer (the recording medium) dynamic range. During the scan the probe intensity was on the order of a few $\mu$W/cm$^2$ resulting to an insignificant saturation (power) broadening ($I/I_s \sim 10^{-4}$). The wavelength is measured with a wavemeter (Agilent / HP 86120B) with resolution of 0.5 pm.

The optical absorption spectrum can be seen in Fig. 4.17. Each point is the average of the transmitted power (normalized with the incident to the cell power) over a three second interval. Small oscillations in the normalized transmitted power are due to interference of the laser (of large coherence length) in the surfaces of the optical elements. A detailed discussion of the optical absorption of alkalis in the presence of buffer gas, under similar conditions to the thesis experiment, can be found in [159].
Figure 4.17: Optical absorption spectrum of K D1 line in the comagnetometer cell at the temperature of the actual experiment. The solid red line is the fit to Eq. (4.44).

and references therein. Briefly, the laser linewidth and jitter (estimated -from the specifications- to be on the order of few tenths of MHz) are much smaller than the optical linewidth and no instrumental corrections need to be applied. Similarly, the Doppler broadening ($\sim 0.9$ GHz) has a small effect on the absorption profile and can be neglected. The experimental data are fitted to a curve given by:

$$P_T(\nu) = P_0 \exp \left[ -\frac{A}{(\nu - \nu_0)^2 + \Gamma^2} - \frac{B (\nu - \nu_0)}{(\nu - \nu_0)^2 + \Gamma^2} \right]$$ (4.44)

where $P_0$, $A$, $B$, $\nu_0$ and $\Gamma$ are allowed to vary to minimize the $\chi^2$ of the fit. The parameters $\nu_0$ and $\Gamma$ are the resonant frequency and linewidth (HWHM) respectively and the term (in the exponent) proportional to $B$ expresses the dispersive first order correction to the Lorentzian profile due to the interactions of alkali atoms with the buffer gas [159, 181]. The parameter $A$ is related to the alkali density $[K]$ through the relation [169]:

$$A = [K] r_c f_{osc} d_c \Gamma$$ (4.45)
where \( r_e \) is the electron radius, \( c \) then speed of light in vacuum and \( d_c \) is the diameter of the cell. From the nonlinear fit it was found that:

\[
\lambda_0 = \frac{c}{\nu_0} = (770.078 \pm 0.002) \text{ nm} \quad (4.46)
\]
\[
\Gamma = (100 \pm 1) \text{ GHz} \quad (4.47)
\]
\[
[K] = (3.12 \pm 0.04) \times 10^{16} \text{ cm}^{-3} \quad (4.48)
\]

To the best of our knowledge, there is no direct experimental measurement of the optical broadening of the K D1 line from \(^3\)He to verify the estimated from the filling process pressure of the cell. From pressure broadening measurements on Cs [149] and Rb [159] with \(^3\)He and \(^4\)He buffer gases, the K D1 broadening due to \(^3\)He is expected to be \(~ 5 - 10\%\) higher than the broadening due to \(^4\)He, which has been measured to be 6.65 GHz/amg [131]; based on this estimation the \(^3\)He density on the cell is evaluated to be: \(~ 14 \) amagat.

We note that the optical linewidth is large enough for the hyperfine structure (of the excited and ground electronic state) or the different K isotopes (through the different hyperfine structure) to have a significant effect; this was verified by fitting the experimental data to the sum of four functions of the form in Eq. (4.44) (two for each \(^{39}\)K and \(^{41}\)K) with appropriate weights and shifts: the results agreed with those of the simpler fit within the errors.

As mentioned before, the measured resonant frequency is used to zero the pump light-shift and uncertainty of \( \delta \lambda_0 = 2 \) pm in the resonant wavelength corresponds to a potential \( L_z \sim 0.1 \mu \text{G} \).

\footnote{We neglected the much smaller hyperfine structure of the excited state.}
4.6.2 Relaxation Rate Measurement

The relaxation rate of K can be estimated from the comagnetometer response to $B_y$ modulation; for zero $B_x$, $L_x$, $L_y$ and neglecting the small contribution from the gyroscopic signal and $R_{se}^e$, it is straightforward to show that up to second order in $\delta B_z$ and $L_z$:

$$\frac{\partial P_m}{\partial B_y} \approx \frac{\gamma_e P_e^e R_{tot}^e}{B_c} \frac{\delta B_z}{R_{tot}^e + \gamma_e^2 (L_z + \delta B_z)^2}$$  (4.49)

Experimentally, the finite difference $\Delta S/\Delta B_y$ approximates the partial derivative as described in Section 4.2.1 for the $\delta B_z$ zeroing. The comagnetometer response to $B_y$ modulation has a dispersive functional dependence on $\delta B_z$, with a half-width $R_{tot}^e/\gamma_e$ and an asymmetry with respect to zero that depends on the $L_z$. Figure 4.18 (a) shows various response curves fitted to functions in the form of Eq. (4.49) for a range of pumping intensities. In Fig. 4.18 (b) the total relaxation rate is plotted as a function of the incident pumping power. The electron spin destruction rate ($R_{sd,t}^e$) can be estimated from the extrapolation to zero pump power. Due to the nonlinear character of the pumping light propagation (see Section 4.6.3), linear extrapolation to $R_{pu} = 0$ underestimates $R_{sd,t}^e$; a simple numerical model that takes into account propagation effects estimates the total electron spin destruction rate to be: $R_{sd,t}^e \approx (160 \pm 10) \text{ sec}^{-1}$ (this corresponds to approximately 20% correction with respect to the linear extrapolation). The results of the fit were consistent with zero $L_z$ within the accuracy of the measurement ($\delta L_z < 1 \mu \text{G}$).

The longitudinal lifetime of $^3\text{He}$ can be characterized from the evolution of polarization in the dark. For this, polarized noble gas is allowed to decay in the absence of polarized noble gas is allowed to decay in the absence of

\footnote{We remind that $R_{tot}^e = R_{pu} + R_{sd,t}^e$ is the sum of the pumping rate and the total electron spin destruction rate.}

\footnote{The error in the estimation comes from the uncertainty of the model. The value is in reasonable agreement from an estimation based on the collision cross-sections recorded in literature, K and buffer gas pressures, and probe characteristics.}

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pump beam (and significant electron polarization) except for small intervals of time ($\sim 20$ sec), during which the new compensation point $B_c$ is found. Note that the compensation point is an approximate measure of the noble polarization $B_c \approx |\lambda M^n P^n_z|$. The holding longitudinal field $B_z$ remains at the last found compensation point during the evolution in the dark. This way the relaxation due to (transverse) magnetic field gradients (Equation 2.10.2) originating from the $^3$He polarization remains approximately constant at the value of the operating point. In Fig. \ref{f4.19} the $^3$He polarization decay is shown. The estimated relaxation rate\textsuperscript{61} is $R_{sd}^n = 9 \times 10^{-2}$ h$^{-1}$; this value is consistent with the measured equilibrium $^3$He polarization and the estimated $^3$He pumping rate. The exponential character of the decay indicates that the relaxation due to environmental magnetic field gradients (e.g. from the shields) is negligible, though the $^3$He spin coherence time may be limited by the gradients created by the pump beam.

\textsuperscript{60}When the pump beam is off, there is a small alkali polarization due to spin-exchange collisions; this polarization is very small and can be ignored. The probe beam has a negligible effect on the evolution of $^3$He.

\textsuperscript{61}The estimated contribution of spin exchange collisions with the unpolarized alkali metal atoms ($R_{se}^{ne} \sim 10^{-6}$ h$^{-1}$) is much smaller than the measured relaxation rate.

Figure 4.18: (a) The comagnetometer response to $B_y$ modulation as a function of $\delta B_z$ for various incident pump powers. The fitting functions are of the form in Eq. (4.49). (b) Total electron relaxation as a function of the incident pump power. The linear extrapolation to zero pump power underestimates the spin destruction by approximately 20%.
longitudinal field or (more likely) by the gradients resulting from the asphericity of $^3$He magnetization.\footnote{To evaluate the contribution of $^3$He magnetization to the magnetic field gradients that affect the spin relaxation, a decay measurement should be performed with keeping the longitudinal holding field at a fixed value, as discussed in \cite{34}.}

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{figure4.19.png}
\caption{Noble gas polarization decay in the dark with changing holding longitudinal field. The measured relaxation corresponds to $T_1 \approx 11$ hours.}
\end{figure}

### 4.6.3 Polarization distribution estimation

The characteristic diffusion time of $^3$He through the cell ($\sim 8$ sec) is significantly smaller than the characteristic relaxation rate so that the noble gas polarization can be considered homogeneous to a very good approximation.

Unlike the $^3$He, the alkali polarization distribution is not uniform. This can be seen with a very simple model for the light propagation: Neglecting diffusion (which in the environment of high noble gas density is suppressed) and assuming homogeneous alkali spin loss rate (this is only a rough approximation since the spin destruction rate from the probe beam depends on the position), the steady state distribution of...

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pumping rate along the z direction (axis of beam propagation) is given:

\[
\frac{dR_{pu}(z)}{dz} = -[K]\sigma_0 (1 - P^e_z(z)) R_{pu}(z) \tag{4.50}
\]

\[
P^e_z(z) = \frac{R_{pu}(z)}{R_{pu}(z) + R^e_{sd}} \tag{4.51}
\]

where \(\sigma_0 = r_e c f_{osc}/\Gamma\) is the absorption cross section on resonance and \(\Gamma\) the optical linewidth (HWHM). The solution to the above equations can be written with the help of the principal value of the Lambert \(W\)-function (also called Omega function, and is defined as the inverse of: \(f(W) = We^W [2]\)):

\[
R_{pu}(z) = \tilde{R}_{sd} W \left[ \frac{R_{pu}(0)}{R^e_{sd}} \exp \left( -[K]\sigma_0 z + \frac{R_{pu}}{R^e_{sd}} \right) \right] \tag{4.52}
\]

We used the student version of FlexPDE to estimate the polarization distribution in a 2D model, taking into account the cell boundary condition \((P^e_z = 0\) at the walls), the increased spin destruction rate in the probe beam path, and the diffusion through the cell; it was found that in the measuring volume the alkali polarization varies less than 5%.

### 4.6.4 Gyroscope

As mentioned before, the comagnetometer responds to rotations around the y axis (axis perpendicular to the pump and probe plane) in an equivalent way to the anomalous fields \(b^e_y\) and \(b^m_y\). The gyroscopic properties of the comagnetometer have been discussed and demonstrated in [115][34]; TK] for setups similar to this thesis. Here, we simply use the signal dependence on \(\Omega_y\) to verify (approximately) the calibration of the comagnetometer and demonstrate its sensitivity to magnetic like spin-coupling fields.
Following arguments similar to those in Section 3.4 the comagnetometer signal to a harmonically varying rotation \((\Omega_x + \Omega_y) e^{i\omega t}\) is to first order in \(\omega/\omega_0\):

\[
P_m(\omega) = \frac{\gamma_e P_e}{R_{tot}} \left[ i \omega \frac{\Omega_x}{\omega_0 \gamma_n} - \left( 1 + i Q \frac{\omega}{R_{tot}} + i \frac{\omega \omega_0}{R_{tot} \omega_0} \right) \frac{\Omega_y}{\gamma_{eff}} \right] \tag{4.53}
\]

where \(\omega_0 = \lambda \gamma_e M^e P_z^e\) and \(\omega_0 = \lambda M^n P_z^n\). In Fig. 4.20 the comagnetometer response to dynamic \(\Omega_y\) rotation is compared with an independent rotational measurement. The optical table was manually rotated \(63^\circ\), and six non-contact, inductive, linear position sensors were used to measure the orientation of the table in time. There is a satisfactory agreement on the order of a few percent between the comagnetometer response (Eq. (4.53)) calibrated with the procedure described in Section 4.3 and the rotation vector estimated from the position vector reading.

Figure 4.20: Comparison of the angular velocity estimation \(\Omega_y\) from the inductive position sensors and the comagnetometer signal calibrated through the response to magnetic fields.

\[63\] In [TK] and [47] a more controlled way for rotating the setup was realized with the use of a piezoelectric actuator; however, for the purposes of this thesis (approximate verification of the calibration and demonstration of sensitivity to effective spin coupling fields) the rough rotational perturbation was adequate.
4.6.5 Response to AC magnetic fields

The comagnetometer exhibits reduced sensitivity to magnetic fields compared with the anomalous spin coupling fields $b^e_y$ and $b^n_y$. In Fig. 4.21 the comagnetometer response to $B_x$, $B_y$ and $B_z$ harmonic modulation is shown as a function of the frequency with the curve normalized with respect to the low frequency (quasi-static) response to $b^e_y$ (or $b^n_y$). In the low frequency limit, the comagnetometer sensitivity to transverse fields can be well described by Eq. (3.80). The data in Fig. 4.21 were acquired at a cell temperature lower than the one used in the anomalous field measurement, resulting in a smaller resonance frequency ($\approx 10$ Hz instead of $\approx 21$); in the main thesis experiment the $B_x$ and $B_y$ sensitivity (in the low frequency regime) is expected to be a factor of $\approx 2$ and $\approx 4$ respectively lower. We note that for a field applied outside the magnetic shields the comagnetometer response is further suppressed by a factor $\sim 10^6$ due to the magnetic shields.

4.6.6 Noise

The measurement of anomalous spin coupling fields is compromised by the comagnetometer noise. In general, the comagnetometer sensitivity is suppressed by two sources of noise: polarimetry noise associated with the optical rotation noise, independent of the presence of spins, and fluctuations in the parameters of Equations (3.2) and (3.3) that describe the effect of spins in the optical rotation. Here, we postpone for later the discussion of the systematic noise.

Polarimetry Noise

In the actual experiment, besides the fundamental optical shot noise, polarimetry is also affected by electronic noise and fluctuations in the optical path of the probe beam.
Figure 4.21: Comagnetometer suppression of magnetic fields compared to the response to a static anomalous spin coupling field $b'_y$ (or $b'_y$). The solid and dashed line is the response predicted by Eq. (3.80) for the $B_x$ and $B_y$ field respectively. In the main thesis experiment the suppression at low frequencies is further enhanced due to the larger resonance frequency.

The electronic noise was measured to be $\approx 2.4 \mu V/\sqrt{Hz}$, approximately constant for frequencies larger than 0.02 Hz\textsuperscript{64}, this result agrees reasonably well with the electronic power spectral density estimation based on the resistive Johnson noise of the transresistance $R_f$ and the first stage amplifier noise (see Fig. 4.11)\textsuperscript{65}. The use of the photoelastic modulation scheme and lock-in amplifier carries the signal at a spectrum regime (around $f_m = 50$ kHz), where electronic noise is significantly smaller compared to low frequencies $< 10$ kHz.

\textsuperscript{64}Here, and for the rest of the chapter, the reported noise spectrum refers to the lock-in demodulated signal.

\textsuperscript{65}Other elements in the photodiode amplifier circuit, the lock-in amplifier, computer recording and the noise pick-up from cables have an insignificant effect on the total electronic noise.
The effect of probe intensity noise on polarimetry can be estimated experimentally by tuning the frequency of the lock-in amplifier off the modulation frequency, the measurement is insensitive to noise associated with the fluctuations in the light polarization and manifested at the photoelastic modulation frequency, but depends on the intensity (and electronic) noise extended over a wide spectrum. This can be seen from Equations (3.115) and (3.119); one can show that for lock-in frequency \( \omega = \omega_m + \Delta \omega, \Delta \omega \ll \omega_m \) in the limit of large \( T \):

\[
\langle S_{\Delta \omega}(T) \rangle \approx \phi \frac{\alpha |\beta|^2}{\omega_m^2 - \omega^2} \frac{\sin[\omega_m T]}{\sqrt{2T}} \approx 0, \quad \omega_m T \to \infty
\]

\[
\delta S_{\Delta \omega}^2(T) \approx \frac{3\alpha^2 |\beta|^2}{16T} \left( 1 - \frac{11\Delta \omega}{6\omega_m} \right) \approx \delta S_{\Delta \omega \rightarrow 0}^2(T)
\]

The limit in (4.54) denotes that the equation is valid when fluctuations in optical polarization angle \( \phi \) occur at time scales larger than it takes for \( \sin(\omega_m T) / (\omega_m T) \) to become close to zero.

Before proceeding, we derive the expected (theoretical) photon shot noise of the measurement. The photon shot noise (in \( V/\sqrt{Hz} \)) can be estimated from Equations (3.117) and (3.120) to be:

\[
\delta V_{PSN} \approx \sqrt{3q_e R_t V_{0,2}}
\]

where \( V_{0,2} \approx \sqrt{2} \) V is the amplitude of the second (with respect to the photoelastic modulation) harmonic signal measured by the lock-in, \( q_e \) is the electron charge (absolute value) and \( R_t = 8 \times 10^6 \) \( \Omega \) is the total transresistance of the photodiode circuit. As mentioned in Section 4.1.5 the photodiode output has a large DC offset \( V_{dc,0} \approx 0.9 \) V, which also contributes to the shot noise. In the simplest model the

\[ ^{66}\text{For this measurement the modulation frequency has to be well outside the bandwidth of the lock-in amplifier.} \]
noise in Eq. (4.56) and the shot noise from DC voltage $V_{dc,0} = \sqrt{2q_e R_t V_{dc,0}}$ are added in quadrature, which results in an estimated photon shot noise $\sim 2.8 \mu V/\sqrt{\text{Hz}}$.

It was found that the probe intensity power spectral density (taking into account the contribution of the electronic noise) was about a factor of 1.5 larger than the expected photon shot noise estimate. The source of the extra noise is unknown to the writer.

Probe beam motion and thermal drifts of the optical elements result in fluctuating optical path and contribute to optical polarization noise. The overall polarimetry noise is the incoherent sum (the terms have to be added in quadrature) of all the above contributions. The resulting noise spectrum is colored (non-uniform): at high frequencies, where thermal noise and beam motion is small, the spectrum is flat and limited by electronic and photon shot noise; at low frequencies, the noise is dominated by the drifts in the beam polarization due to changing optical path and acquires the form of 1/f noise. Considerable effort was applied to reduce the low frequency noise and extend the flat, photon shot noise and electronic noise limited region to the low frequency regime.

In Fig. 4.22 the experimentally measured noise spectrum is shown. The roll off at frequencies larger than 50 Hz is due to the limited lock-in bandwidth (3 msec time constant, and 24 dB/oct digital filter). For frequencies higher than 0.1 Hz the polarimetry noise is approximately $7 \times 10^{-8}\text{ rad}/\sqrt{\text{Hz}}$.

We emphasize that the values of experimental parameters used in this thesis do not represent a global optimum in the parameter space; we feel that, given realistic experimental constraints (including time and money), the optimum should not differ significantly from the one realized here. In any case, it was enough to lead to results highly competitive with the respect to contemporary similar measurements.

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67 This probably (slightly) overestimates the noise since it neglects correlations between the DC offset and the harmonic components.

68 Here, the term 1/f is loosely used to express the trend of noise to increase significantly at low frequencies.
Figure 4.22: Noise spectrum associated with the probe beam and its detection. Overall, the sensitivity of optical rotation is mainly limited by the intensity laser noise with a small contribution from the electronic noise.

**Comagnetometer Noise**

In Fig. 4.23 the optimized comagnetometer noise spectrum is given. It can be seen that the sensitivity of the anomalous field in the frequency domain 0.1-1 Hz is determined by the polarimetry noise. Compared to the comagnetometer spectrum under conditions described in [TK], in this thesis there was an improvement in the low frequency noise, more than a factor of 4 in the timescales of the signal modulation (see Section 5.1). The resonance frequency of the K-$^3$He coupled ensemble (see Section 3.3) has been increased to $\approx 21$ Hz due to the higher noble gas and alkali density. The significant improvement in the sensitivity demonstrates the progress realized in this work.

The spectrum in Fig. 4.23 was taken before the actual run of the experiment. The average spectrum during anomalous field measurement can be seen in Fig. 5.3.
Figure 4.23: The comagnetometer sensitivity is limited by polarimetry noise in the frequency domain 0.1-1 Hz. In this thesis the low frequency performance of the comagnetometer was considerably improved. The large peak at 21 Hz is the resonance of the K-3He coupled spin ensembles.
4.7 Spin Source

The anomalous field that the comagnetometer measures is created by a separate nucleon spin source. The author of this thesis happily acknowledges the work of Justin Brown, who implemented the optical and mechanical setup, and performed the first measurements and characterization of the spin source.

4.7.1 Experimental Setup

A schematic of the experimental setup can be seen in Fig. 4.24.

![Schematic of the spin source experimental setup](image)

Figure 4.24: Schematic of the spin source experimental setup. DLA: Diode Laser Array, Pol: Polarizer, LCW: Liquid Crystal Waveplate, FG: Fluxgate, CC: Compensation Coil, EPR: Coil for EPR, BS: Beam Sampler, PD: Photodiode, IS: Integrating Sphere

The source for the anomalous field is a high pressure, atomic gas of $^3$He, polarized in a typical optical pumping scheme [9, 194]: circularly polarized light transfers angular momentum to K atoms, creating high alkali polarization (ideally close to 100%) with the help of molecular N$_2$ which suppresses the depolarizing radiation.
trapping (see Section 2.7), and the noble gas is polarized through spin exchange collisions with the alkali-metal atoms.\footnote{It has been shown \cite{157, 15, 46} that large noble gas polarization can be realized with hybrid optical pumping, where an optically polarized alkali atomic species (e.g. Rb) at low density collisionally polarizes another alkali metal (e.g. K) at high density which then transfers angular momentum to the noble gas (e.g. $^3$He) through spin-exchange collisions; in this thesis, we adopted the single alkali optical pumping because of the available resources (laser diode array at 770 nm).}

The atomic ensembles are contained in a cylindrical-like cell, made of Corning 1720 aluminosilicate glass, thickness $\approx 4$ mm, inner diameter 4.3 cm, length 12.8 cm, and slightly curved (truncated spherical) top and bottom surfaces to provide increased mechanical strength \cite{155}. By loading water the cell volume was measured to be 186 cm$^3$. The cell is filled with $\approx 12$ amg of $^3$He, $\approx 0.029$ amg N$_2$ and a droplet of K (a few mg) in natural abundance through a process identical to the one described in \ref{4.1.1} and was heated to approximately 190°C (the temperature is measured with a RTD located at the middle of the cell; we note that there is a temperature variation of a few degrees across the oven). The oven, thermally insulated from the environment with polyimide foam, is cylindrical (2.65 inches inner diameter, 8.5 inches length, 1/4 inches thickness) following the shape of the cell, made of Nema Grade G-7 and optical glass windows at the bases for the pumping light. The oven operation is based on forced air flow: hot air is input at one end and carried away through a chimney at the other end. Additional heating results from the optical pumping, since most of the laser power is ultimately converted to heat. The G-7 oven structure acts as the coil frame for the applied magnetic field, whose cartesian components are independently controlled through a triad of coils (see Fig. 4.25 (a)). Although this design provided poor thermal protection of the wires, it resulted in a compact spin source. Cosine winding for the transverse fields was employed in order to realize homogeneous transverse field, and gradient coils allow first order cancelation of the longitudinal magnetic gradients.
Figure 4.25: (a) The oven, made of Nema Grade G-7 material, is the coil frame for the longitudinal and transverse fields. (b) A solenoidal coil is wound on the cell surface along its entire length to compensate for the magnetic field generated by the spin source.

The optical power is provided by a broad-area laser diode array tuned to the D1 K resonance. An external cavity with appropriate beam shaping lenses and diffraction grating feedback\(^70\) narrows the optical spectrum of the output laser field\(^71\). A small fraction (< 4%) of the beam is input to a multimode fiber (through an integrating sphere) and guided to a spectrometer to monitor continuously the laser spectrum. An electronically controlled liquid crystal variable waveplate in combination with a quarter waveplate convert the light polarization to circular. The shape of the beam is matched to the dimensions of the cell with an appropriate set of lenses. Approximately 2.1 W of laser light is directed to the cell; a mirror placed at the output (outside the oven) reflects the unabsorbed light back to the cell to increase the pumping power. A beam pick off directs a fraction of the light transmitted through the cell to a photodiode for the EPR measurement.

The optical setup for the spin source is developed on a separate breadboard and with an appropriate geometry to allow for the desired orientation of the atomic spins.

\(^{70}\)The first order diffraction is directed to the diode array and the zero order is the output of the external cavity.

\(^{71}\)The external cavity reduces the spectrum significantly and the optical power at 768.6 nm (D\(_2\) line of K) is only a small fraction.
The spin source is placed at the (comagnetometer) probe beam detector side (see Figures 4.26 and 5.1) as close to the comagnetometer as the magnetic shields (of the comagnetometer) permit: this corresponds to approximately 50 cm away from the comagnetometer. We note that the comagnetometer shields, designed for the requirements of the experiment described in [TK], are not optimized for the measurement of (anomalous) spin coupling fields: the strong dependence of interaction on the distance favors the implementation of smaller geometries (for instance, in [34, 33] a more compact version of the magnetic shields is described).

Figure 4.26: Photo of the spin source placed close to the comagnetometer. The optical board where the setup was developed is vertical. The retro reflecting mirror, the EPR photodiode, fluxgate and enclosure are not shown. Optics for the comagnetometer probe beam detection can also be seen.

The high temperature oven and the relatively large laser power constitute a heat source, which can potentially affect the noise spectrum of the comagnetometer. Therefore, the spin source is thermally isolated with the use of polyimide foam. Furthermore, special care is devoted to block optical leakage from the spin source to either the probe beam detector or the photodiode of the comagnetometer pump intensity.
feedback; for this, the spin source setup is fully covered by a thin aluminum frame (painted black) and an extra layer of black rubberized fabric.

Contrary to the comagnetometer case, there is no requirement for low noise operation. Fluctuations in the pumping optical power or magnetic fields, to the extent that they do not significantly alter the longitudinal polarization, do not affect the performance of the spin source. The main requirement for field (and to a smaller extent pump) stability arises from the polarization measurement using the EPR signal discussed in [4.7.2]. The low value of the anomalous field (if it exists at all) being significantly smaller than the measurement error of the comagnetometer (over the timescale of a few second) imposes loose limitation on the uncertainty in the spin source $^3$He polarization; less than 1% polarization uncertainty ($\sim 6\%$ relative polarization uncertainty) is realized with current source stability of one part in $10^4$, which is readily available with most commercial power supplies. In addition, inhomogeneities of the noble gas polarization (from gradients in the pumping power and alkali-density due to absorption and nonuniform temperature respectively) are smoothed out by the diffusion (the characteristic timescale for relaxation is much larger than the diffusion time across the cell), so that to a good approximation the $^3$He magnetization can be considered uniform.

The spin source implementation is based on the requirement of a compact large nucleon (in particular neutron) spin ensemble, to generate a coherent anomalous magnetic-like field. This can be realized at high $^3$He density and noble gas polarization, and large dimension cell. In practice, the $^3$He density is limited by mechanical considerations for the cell; the cylindrically shaped cell has to endure large stresses at an elevated temperature$^{72}$ (190°C) for an extended period of time (a few months$^{73}$). The operating temperature (190°C) corresponds to alkali-metal density $\sim 10^{14}$ cm$^{-3}$

$^{72}$Besides the thermal stresses induced at high temperatures, the pressure is increased (e.g. a 12 amagat cell heated to 190°C results in $\sim 20$ atm pressure).

$^{73}$Note that the comagnetometer cell with the more compact, approximately spherical design has higher stress tolerances than the spin source cell.
and is constrained by the temperature tolerance of the cell and the magnet wires. The finite available optical power restricts the size of the cell.

Taking into account the isotropic interaction $\propto \mathbf{S} \cdot \mathbf{N}$, where $\mathbf{S}$ and $\mathbf{N}$ are the electron spin and $^3$He nuclear spin respectively, and the anisotropic interaction $\propto (3\mathbf{S} \cdot \hat{\mathbf{R}} \hat{\mathbf{R}} \cdot \mathbf{N} - \mathbf{S} \cdot \mathbf{N})$ with $\hat{\mathbf{R}}$ being the unit space vector connecting the two spins, the dynamics of the noble gas polarization can be well approximated with the rate equations:

$$
\frac{dP_n}{dt} = -R_{sd}^{n} P_n + k_a[K] (\bar{P}_e - P_n) + k_b[K] (-\bar{P}_e/2 - P_n)
$$

(4.57)

where $k_a$ and $k_b$ are the spin-exchange rate coefficient for the isotropic and anisotropic hyperfine coupling ($k_a[K] = R_{se}^{ne}$), $[K]$ is the alkali density, $R_{sd}^{n}$ the nuclear spin destruction rate, $\bar{P}_e$ is the K polarization averaged over the entire volume of the cell. The steady state solution of the above equation is:

$$
P_n^\infty = \bar{P}_e \frac{(k_a - k_b/2) [K]}{R_{sd}^{n} + (k_a + k_b) [K]}
$$

(4.58)

The noble gas polarization is approximately proportional to the alkali-metal polarization (which in turn depends on the ratio of optical pumping rate to the alkali spin destruction rate) and increases with the spin-exchange collision rate, and decreases with the nucleus spin destruction rate.

---

74 The strength of the anomalous field source depends on the total number of polarized noble gas atoms.

75 Here, we neglect the influence of magnetic or magnetic-like fields to the evolution of the longitudinal polarization. The large applied longitudinal field suppresses the transverse polarization components (except for resonant excitations) and the equations describe adequately the optical pumping.

76 This approximation holds when the diffusion time across the medium (for $^3$He atoms) is much smaller than the characteristic relaxation time scale, as is the case in our experiment.
For the conditions of the experiment, the electron spin destruction rate (at a given temperature) is determined by the fundamental spin-loss collisions (K-K, K-\(^3\)He and K-N\(_2\)) -diffusion to the walls and magnetic field gradients do not affect appreciably the K spin relaxation-. No precise measurement of the K polarization was performed. By mapping the RF resonance curve over the entire Zeeman spectrum\(^{79}\) we were confident that the alkali spin polarization around the central axis of the cell was close to unity (the non-negligible laser power transmitted through the optically thick vapor also indicates the very high electron polarization). However, the alkali atoms in the region away of the axis and close to the output side of the cell experienced reduced optical pumping due to distortion from the curved input cell window and the larger light absorption (from increased spin destruction as the atoms diffuse to the cell walls) \([34, 35]\), resulting in smaller average K polarization \(\bar{P}_e\).

Diffusion of the \(^3\)He atoms through magnetic field gradients is a major source of noble gas spin relaxation (see Section 2.10.2). In principle, to the extent that the field gradients are not generated from the longitudinal holding field, their contribution to spin destruction can be significantly suppressed at large longitudinal field \(B_0\). During the actual experiment the spin source was placed in close proximity to the co-magnetometer magnetic shields which introduced measurable relaxation due to field gradients\(^{81}\). For the most part (sidereal days 3022-3057, when the spin polarization at the spin source was in the y direction of the comagnetometer), the spin source was operated at 7.7 G longitudinal field, corresponding to the (approximate) maximum current (1.96 A) that we felt confident to run in the coil for a long time (over a

\(^{77}\)As before, electron and alkali-metal polarization are used alternatively.
\(^{78}\)We use the word fundamental to describe processes that cannot be circumvented at the given experimental conditions.
\(^{79}\)This was done in the same way as for the EPR signal, discussed later. For a discussion in alkali metal polarization measurement based on a similar concept see \([44, 81]\).
\(^{80}\)The detected light power was transmitted through a small region around the central axis of the cylindrical cell.
\(^{81}\)It is likely that the homogeneity of the applied longitudinal field is affected by the magnetic boundary of the shields.
month \(^{82}\): for spin source orientation along the \(z\) direction (sidereal days 3118-3126) the longitudinal field was increased to 12.8 G (3.2 A in the coil) to compensate for the additional relaxation introduced by the geometry (apparently due to larger magnetic field gradients). For both spin source orientations the longitudinal lifetime \(T_1 \sim 21\) \(h\) \(^{83}\).

In addition to the gradients, the noble gas spin relaxes due to diffusion to the walls \(^{84}\). As it was shown in \([14, 44]\), the relaxation on the walls depends strongly on the alkali density \([K]\), and the overall relaxation can be written as

\[
\Gamma_n = k_{se}[K] (1 + X) + \Gamma_{n,0},
\]

where \(\Gamma_{n,0}\) is the room temperature relaxation (which according to \([14, 44]\) includes the magnetic field gradient contribution, though look at footnote \(^{85}\)) and \(X\) is a dimensionless factor that depends on the surface to volume ratio \(S/V\) (for the spin source cell \(S/V \sim 1\) cm\(^{-1}\)) and the manufacturing process \([14]\); comparing the \(^3\)He relaxation rates at room and high (operating) temperature and from measured values of the K-\(^3\)He spin-exchange rate the factor \(X\) was estimated to be \(\sim 0.2\).

The steady state noble gas polarization (when no spin modulation was applied) was \(\sim 16\%\) or \(\sim 18\%\) depending on the helicity of the pump light \(^{86}\), which is appreciably smaller than those that have been reported in the literature for similar pumping schemes \([46, 15]\) (\(\sim 80\%\) for K-\(^3\)He atomic ensembles). In the spin source described here, the main limitation is the optical power (e.g. in \([15]\) 78\% polarization was

\(^{82}\)As mentioned before, the magnetic coils are rested on the outside surface of the oven and are poorly cooled. Additional heat is generated by the power dissipated in the ohmic losses, which at large current values may result in wire failure.

\(^{83}\)Away from the magnetic materials, and for longitudinal field 7.7 G, \(T_1 \sim 31\) h

\(^{84}\)The strong temperature dependence of this effect suggests that the relaxation mechanism is not due to interaction with the glass molecules (as is the case for the wall relaxation of alkali atoms) \([14, 44]\).

\(^{85}\)For the estimation of \(X\) we followed the discussion in \([14]\), which assumes that the relaxation due to diffusion through magnetic field gradients at the time of passage through resonance is small. We note that the relaxation due to gradients has a temperature dependence, since the diffusion constant scales approximately as \(T^{1/2}\). For the estimation of \(X\) we performed measurements away from magnetic materials, at a location where the gradients had a small effect on relaxation.

\(^{86}\)Due to the large cell birefringence (resulting from the manufacturing process and the pressure stress) the purity of the circular polarization is not the same for right and left polarized light.
realized using $\approx 20$ Watts of optical pumping power, which is one order of magnitude larger than the one available for the thesis experiment).

A solenoidal coil is wound on the cell surface along its entire length (see Fig. 4.25(b)) to reduce the magnetic interaction of the spin source with the comagnetometer and the environment (see Section 5.4 for a discussion of the systematic effects that may arise from this magnetic interaction). A uniformly polarized cylinder is equivalent to a surface current density in the azimuthal direction $\mathbf{j}_n\parallel \hat{n}$ and can in principle be completely canceled by an ideal surface coil. Due to polarization inhomogeneities (mainly resulting from field gradients), the non-cylindrical cell shape (curved windows), the non-ideality of the solenoidal coil (the wires have a finite thickness and the current density is not uniform) and non-overlap of the effective surface current with the current coil, the compensation is not exact: the magnetic field from the spins is reduced by approximately a factor of 15, as can be seen if Fig. 4.27. Likewise to the comagnetometer case, changes in the applied compensation coil field occur smoothly in order to reduce spin excitation.

A coil (single turn) is mounted on the side of cell (over the compensation coil) and is used to detect the electron paramagnetic resonance (EPR) of K. The EPR coil runs through the whole length of cell to provide an averaged value of the EPR signal.

A fluxgate magnetometer (Bartington Mag-03) located close to the cell (approximately 8 cm along the radial direction from its center) measures the magnetic field of the spin source. The measurement provides information on the spin orientation and is used to calibrate the compensating field.

The spin source is controlled with a computer equipped with a multi-function Data Acquisition Card (DAQ) and run under the Labview environment. This allows for fully automated digital recording and routine applications on the spin source.

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$^{87}$The magnetic field of a magnetized object of magnetization $\mathbf{M}$, which discontinuously falls to zero at the surface $S$, is equivalent to the magnetic field created from an effective surface current density $\mathbf{M} \times \hat{n}$, where $\hat{n}$ is the normal to $S$, and an effective volume current density $\nabla \times \mathbf{M}$. 

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Figure 4.27: Fluxgate measurement of the magnetic field close to the cell while the $^3$He spins are reversed. The blue dashed curve is taken when the compensation coil does not follow the spin flip, while in the red curve the current in the coil reverses with the spins. The compensation coil reduces the magnetic field from the cell by approximately a factor of 15. The residual uncompensated field is due to polarization inhomogeneities, the non-cylindrical cell shape, finite thickness of the wire coil and cell walls. The data shown are the average over many runs.

The interested reader is referred to [34] for a detailed description of the experimental setup.

### 4.7.2 Polarization Measurement

To estimate the anomalous spin coupling between nucleons, knowledge of the number of polarized $^3$He atoms (more accurately the product of polarization times the density) in the spin source is required. For this, the shift of the electron paramagnetic resonance (EPR) of K atoms due to the polarized $^3$He was used [158, 159, 44].

As discussed in Section 2.12 for a polarized K-$^3$He ensemble in the presence of a DC magnetic field $B_0$ the hermitian part of the alkali Hamiltonian can be written in
the form (neglecting light-shift and the small alkali nuclear Zeeman contribution):

\[ H = A_{Jh} \mathbf{S} \cdot \mathbf{I} + g_e \mu_B \mathbf{\ddot{B}} \cdot \mathbf{S} \tag{4.59} \]

\[ \mathbf{\ddot{B}} = \mathbf{B}_0 + \mathbf{\ddot{B}}_{\text{He}} \tag{4.60} \]

\[ \mathbf{\ddot{B}}_{\text{He}} = \frac{8\pi}{3} \kappa \frac{\langle K \rangle}{K} \mu_{\text{He}} [\text{He}] \tag{4.61} \]

where \( \mathbf{\ddot{B}}_{\text{He}} \) is the effective field from the \(^3\text{He}\) magnetization experienced by the alkali atoms, \( \mathbf{B}_0 \) is the applied magnetic field, and \( K = 1/2 \) is the spin quantum number of the noble gas. The enhancement factor \( \kappa \) includes the collisional spin-exchange frequency shift and the classical magnetic field generated by the magnetized noble gas. Here, we discriminate the enhancement factor \( \kappa \) (applicable to the cylindrical cell) from \( \kappa_0 \) defined in \([158]\) for spherical geometry. It can be shown\(^{88}\) \([144]\) that the average \( \kappa \) along the cylinder axis is:

\[ \kappa = \kappa_0 + \frac{1}{2} - \frac{3}{2} \left[ 1 + \frac{R}{l} - \sqrt{1 + \left(\frac{R}{l}\right)^2} \right] \tag{4.63} \]

where \( l \) and \( R \) are the cylinder length and radius respectively. For our cell \( R/l \approx 0.33 \) and the (fractional) correction is: \( (\kappa - \kappa_0) / \kappa_0 \approx 1% \). We note that for the estimation of \( \kappa \) the average magnetic field from \(^3\text{He}\) magnetization over the area sampled by the detected photons is required; this region was close to the cylinder axis (within a distance of \( \approx 1 \) cm) and the correction (4.63) is sufficient for accuracy < 1%.

\(^{88}\)The magnetic field of a uniformly magnetized cylinder of radius \( R \) and length \( l \) is along the axis (in Gaussian system):

\[ B_z(z) = 4\pi M - \frac{4\pi M}{2} \left[ \frac{z/d - 1/2}{\sqrt{(R/l)^2 + (z/d - 1/2)^2}} - \frac{z/d + 1/2}{\sqrt{(R/l)^2 + (z/d + 1/2)^2}} + 2 \right] \tag{4.62} \]

where \( M \) is the magnetization, the \( z \)-axis is the cylinder axis and the origin is taken to be the center of the cylinder. It is then straightforward to show that the ratio of the average field along the cylinder axis to the magnetic field inside a uniformly magnetized sphere (magnetization \( M \)) is:

\[ \kappa_g = 3/2 \left( -R/l + \sqrt{1 + (R/l)^2} \right) \]. The enhancement factor \( \kappa \) is given by: \( \kappa = \kappa_{KS} + \kappa_g = \kappa_0 - 1 + \kappa_g \).
The resonance frequencies of Zeeman transitions ($\Delta F = 0$, $\Delta m_F = \pm 1$) are given by Equations (2.10) and (2.11) to second order in the effective field. Expanding in the small quantity $\tilde{B}_{\text{He}}/B_0$ for the $F = 2$ manifold the resonance frequencies are given by:

$$f_0 = \frac{g_e \mu_B B_0}{\hbar[I]} - \frac{4(g_e \mu_B B_0)^2 (m_F - 1/2)}{[I]^3 h A_{hf}}$$

$$+ \frac{g_e \mu_B B_0}{\hbar[I]} \tilde{B}_{\text{He}} - \frac{8(g_e \mu_B)^2 B_0 (m_F - 1/2) \tilde{B}_{\text{He}}}{[I]^3 h A_{hf}}$$

(4.64)

The detection of EPR is based on the dependence of light absorption on polarization. As discussed in Section 2.8.2, the absorption cross section is (for the D1 transition where $J_e = 1/2$) proportional to $1 - P^e_\parallel$, where $P^e_\parallel$ is the longitudinal alkali polarization. When a transverse RF magnetic field is applied the longitudinal polarization is reduced (see Section 2.16.1) and the light absorption is increased. This change depends on the frequency of the RF field according to Eq. (2.150). In the language of (semi-classical) quantum mechanics, for the highly polarized alkali medium most K atoms are in the $F = 2, m_F = 2$ state (for left circularly polarized pump light, or $F = 2, m_F = -2$ for oppositely polarized pump light), where they do not absorb laser photons; application of RF field at (or around) the EPR frequency $F = 2, m_F = 2 \rightarrow 1$ (or $F = 2, m_F = -2 \rightarrow -1$) causes transitions between the Zeeman states, lowering the population in the dark $F = 2, m_F = 2$ ($F = 2, m_F = -2$) state, and the light absorption is increased.

Due to the high alkali polarization (close to 1) and the appreciable Breit-Rabi splitting of the hyperfine manifold ($\sim 60$ kHz) (much larger than the EPR width), we only need to consider the end transitions ($F = 2, m_F = 2 \rightarrow 1$ or $F = 2, m_F = -2 \rightarrow -1$); the other transitions (e.g. $F = 2, m_F = 1 \rightarrow m_F = 0$) have an insignificant effect on the observed signal. The transmitted laser power (assuming for now to be

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Figure 4.28: EPR signal as a function of the RF frequency in the vicinity of the $F = 2, m_F = 2 \rightarrow 1$ resonance. Each point is the lock-in signal to a small frequency modulation of the RF field. The curve is an average over 33 sweeps.

monochromatic on the optical D1 resonance) is given by:

$$P_L = P_L^0 \exp \left[ -[K] \sigma_0 \left[ l - \int_0^l P_e^0(\zeta) d\zeta \right] \right]$$  \hspace{1cm} (4.65)

$$P_e^0(\zeta) = P_e^{00}(\zeta) \frac{1 + (\Delta f T_2/2\pi)^2}{1 + (\Delta f T_2/2\pi)^2 + \gamma_K^2 B_{RF}^2 T_1 T_2}$$  \hspace{1cm} (4.66)

where $P_L^0$ is the light incident power, $\sigma_0$ is the absorption cross section (for unpolarized atoms), $[K]$ is the density of K atoms, $\gamma_K$ their gyromagnetic ratio, $P_e^{00}$ is the longitudinal polarization in the absence of the RF field, $T_1$ and $T_2$ are the longitudinal and transverse relaxation time respectively ($T_1 \approx T_2$), $2B_{RF}$ is the amplitude of the (linearly polarized) RF field, $\Delta f = f_0 - f$ is the detuning of the RF frequency $f$ from the EPR resonance $f_0$. For small (with respect to the EPR linewidth, which is mainly determined by the optical pumping rate) frequency modulation of the RF field ($f = f_0 + \delta f + A_{fm} \cos \omega_m t, \omega_m T_2 \ll, A_{fm} T_2 \ll 1, \delta f T_2 \ll 1$) one can show that

\footnote{Since the cell was filled with K in natural abundance there is \( \approx 6.7\% \) $^{41}$K with different EPR frequency spectrum. Because of the $^{41}$K small abundance and large difference in EPR transitions $m_F = 2 \rightarrow m_F = 1$ ($m_F = -2 \rightarrow m_F = -1$) between $^{39}$K and $^{41}$K, $^{41}$K can be neglected to a good approximation.}
to first order in small quantities:

\[
P_L = P_L^0 \exp \left[ -[K] \sigma_0 \left( l - \int_0^l P_e^0(\zeta) \big|_{f \rightarrow f_0} d\zeta \right) \right] \\
\times \left[ 1 + \frac{B_1^2[K] T_1 T_2^2 \gamma_K^2 \sigma_0 \int_0^l P_e^0(\zeta) \big|_{f \rightarrow f_0} d\zeta}{2\pi^2(1 + \gamma_K^2 B_1^2 T_1 T_2)^2} \right] \delta f A_m \cos \omega_m t
\]  

(4.67)

The measured EPR lock-in signal for the \( F = 2, m_F = 2 \rightarrow 1 \) transition can be seen in Fig. 4.28. There is a pronounced asymmetry in the response, probably associated with (magnetic field and alkali polarization) gradients. Despite the deviation from the dispersion function (Eq. (4.67)) the zero crossing point is well distinguished. The asymmetry results in an apparent shift of the feedback set point from the actual EPR signal (which is stronger at larger holding magnetic field) and a systematic error in the polarization calculation. This may explain the deviation of the measured EPR frequencies from the Breit-Rabi theoretical predictions (see Eq. (4.64)) and the \( \approx 10\% \) relative difference in the \(^3\)He polarization estimation for the \( F = 2, m_F = 2 \rightarrow 1 \) and the \( F = 2, m_F = -2 \rightarrow -1 \) transitions.

In Fig. 4.29 the schematic for the EPR measurement is shown. Using a Voltage Controlled Oscillator (VCO) -Wavetek 166 function generator-, the EPR coil is driven with a frequency modulated RF field. The sinusoidal component of the transmitted

\[90\] However, the asymmetry in the EPR curve does not affect the polarization estimation if it does not depend on the total magnetic field and remains the same as \(^3\)He is flipped.
light intensity (second term in Eq. (4.67), proportional to $\delta f$) is measured by a digital lock-in amplifier referenced to the modulation frequency; the output is directed to a PID feedback circuit, which adjusts the DC level at the VCO input to keep the lock-in signal to zero, maintaining the average (over the modulation period) RF frequency locked to the EPR resonance. Using a fraction of the VCO output, the average RF frequency is recorded by the counter in the DAQ board, and the magnetic field that corresponds to the measured EPR frequency is calculated from Eq. (4.64) (for $m_F = 2$ or $m_F = -2$ depending on the handedness of the pumping light$^{91}$). The operating parameters for the EPR system are given in Table 4.2.

![Graph showing EPR frequency over time](image)

Figure 4.30: Polarization measuring cycle for the two light helicities. The nuclear spins are flipped and the contribution of $^3$He effective magnetic field to the EPR resonance is isolated. The arrows indicate orientation of the nuclear magnetization. The large changes in the EPR frequency are due to reversals in the electron polarization (the pump photon spin changes sign), so that different resonances ($m_F = 2 \rightarrow 1$ or $m_F = -2 \rightarrow -1$) are employed.

The $^3$He polarization was estimated by comparing the EPR frequency for opposite orientation of the noble gas spins (parallel and anti-parallel to the magnetic field). This way, the frequency shift due to $^3$He is isolated and no precise determination of the holding field is required. The $^3$He spin reversal was realized by applying an appropriate (AFP) transverse field as described in the following section. The loss rate per $^3$He is very small and has a negligible effect on the polarization estimation.

---

$^{91}$In practice this is determined by the frequency: the $m_F = 2 \rightarrow 1$ has smaller frequency compared to $m_F = -2 \rightarrow -1$.
During the nuclear polarization measurement the helicity of the laser pump and the current at the compensation coil remain constant, i.e. they do not follow the spin reversal (contrary to what is happening when the comagnetometer signal is recorded). The measuring cycle can be found in Fig. 4.30.

The polarization estimation based on the EPR detection was verified by measuring the magnetic field of the spin source with the fluxgate and using electromagnetic theory to calculate the polarization. The agreement between the two polarization estimation methods was $\sim 15\%$, which is satisfactory given the uncertainty in the exact position of the fluxgate sensor and the geometry of the cell.

We stress that due to the inhomogeneities in the electron polarization, magnetic field, variations in $\kappa$, and noise in the magnetic field, the accuracy of the nuclear polarization estimation is limited to a few % (relative uncertainty $< 10\%$). The spin source nuclear polarization affects the reported strength of the spin-dependent force and its uncertainty in the same (proportional) manner, and the main result of the thesis is not altered; perhaps the value and uncertainty of the anomalous field should be corrected by the same small factor.

<table>
<thead>
<tr>
<th>EPR</th>
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</thead>
<tbody>
<tr>
<td>RF Frequency</td>
<td>5.3 / 5.6 MHz</td>
</tr>
<tr>
<td>Mod. Frequency</td>
<td>8.6 / 9.7 MHz</td>
</tr>
<tr>
<td>Mod. Amplitude</td>
<td>210 Hz</td>
</tr>
<tr>
<td>Lock-in time const.</td>
<td>10 msec</td>
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<tr>
<td>Lock-in roll off</td>
<td>24 dB/oct</td>
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</tbody>
</table>

<table>
<thead>
<tr>
<th>AFP</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Sweep Start</td>
<td>12 (31 kHz)</td>
</tr>
<tr>
<td>Sweep Stop</td>
<td>36 (55 kHz)</td>
</tr>
<tr>
<td>$\tau_s$</td>
<td>20 ms</td>
</tr>
<tr>
<td>$\tau_r$</td>
<td>30 ms</td>
</tr>
<tr>
<td>RF Amplitude</td>
<td>0.5 G</td>
</tr>
</tbody>
</table>

Table 4.2: Parameters for EPR and AFP. The values in the parentheses correspond to 12.8 G holding field.

4.7.3 Spin Flip

During the experiment frequent reversals of the $^3$He spins in the spin source were performed in order to bestow a characteristic frequency on the anomalous field. There-
fore, a low magnetization loss per flip is required to maintain high nuclear polarization. This was realized by applying a transverse RF field according to the conditions of the adiabatic fast passage \([1,158]\).

To analyze the spin behavior under the influence of a RF field, it is convenient to consider the dynamics in the rotating frame (see Section 2.16.1), which is the frame rotating around the z axis (of the longitudinal field \(B_0\)) at the frequency \(\omega\) of the RF field. Neglecting the counter-rotating component and allowing for time varying RF frequency and amplitude, in the rotating frame the effective magnetic field experienced by the spins is: \(H_{\text{eff}}(t) = (B_0 - \omega(t)/\gamma_n)\hat{z} + B_1(t)\hat{i}\), where \(\gamma_n\) is the gyromagnetic ratio of \(^3\)He and \(2B_1(t)\) is the amplitude of the RF field. As the RF frequency is swept from far from resonance (for concreteness assume far below resonance) through resonance to a value much above the resonance, \(H_{\text{eff}}\) changes direction continuously: initially parallel to the \(z\) axis, along the \(\hat{i}\) direction when on resonance \((\omega = \gamma B_0)\), and finally antiparallel to the \(z\) axis. If the sweep (relative change of the effective field) is slow enough with respect to the time scale of the system set by the (instantaneous) effective Larmor frequency \(\gamma_n B\), \(B = |H_{\text{eff}}|\) (more precisely when the Fourier components of the effective field change at frequencies on the order \(\gamma_n B\) are negligible), the spins can follow the changing field adiabatically\(^{92}\), so that by the end of the sweep their direction has been reversed. The adiabatic condition \(\dot{B}/B \ll \gamma_n B\) is typically strongest at resonance where it can be expressed as \(\dot{B}/B_1 \ll \gamma_n B\). In the presence of magnetic field gradients (resulting in a distribution of Larmor frequencies) the magnetization of the sample is still reversed by adiabatic passage (though the maximum transverse magnetization is reduced).

The spin relaxation puts a limit on how slow the sweep should be for an efficient spin reversal: the time of passage through resonance\(^{93}\) \(\sim B_1/\dot{B}\) should be negligible.

---

\(^{92}\)This is effectively an application of the adiabatic theorem \([28,89]\). In this particular case, the angle between the magnetization and the effective field \(H_{\text{eff}}\) remains constant.

\(^{93}\)The relaxation is largest around resonance where the effective field is small; therefore the fast condition refers to the passage through resonance.
compared to the relaxation time (fast condition), so that the polarization loss is insignificant. In the thesis experiment the spin relaxation is mainly determined by diffusion through magnetic field gradients and can be approximated in the rotating frame by: $D|\nabla B_z|^2/B^2$, where D is the $^3$He diffusion constant. Combining the above the adiabatic fast passage condition is:

$$\frac{D|\nabla B_z|^2}{B_1^2} \ll \frac{\dot{B}}{B_1} \ll \gamma B_1$$

(4.68)

As can be seen from (4.68) large $B_1$ are advantageous for the adiabatic fast passage.

In the experiment, the RF field employed for the adiabatic fast passage is:

$$B_{RF} = \frac{B_1}{\tau_r} \left\{ \begin{align*}
\cos \left[ \omega_0 t + \frac{1}{2} r_\omega t^2 \right], & \quad 0 \leq t < \tau_r \\
\cos \left[ \omega_0 t + \frac{1}{2} r_\omega t^2 \right], & \quad \tau_r \leq t < (\tau_r + \tau_s) \\
\left( 1 - \frac{t-\tau_r-\tau_s}{\tau_r} \right) \cos \left[ \omega_0 t + \frac{1}{2} r_\omega t^2 \right], & \quad \tau_r + \tau_s \leq t < (\tau_r + \tau_s + \tau_r)
\end{align*} \right. \quad (4.69)$$

$$r_\omega = \frac{\omega_f - \omega_0}{\tau_r + \tau_s + \tau_r} \quad (4.70)$$

The amplitude of transverse field (we used $B_z$) is varied smoothly (linearly) from zero to a maximum value ($B_1$), remains constant to this maximum value during the passage through resonance, and is smoothly lowered to zero; the frequency of the field (first derivative of the sinusoidal phase) changes linearly in time with rate $r_\omega$. The RF waveform was generated at the analog output of the computer DAQ board, smoothed through a low frequency filter with corner frequency $\sim 800$ kHz, and amplified to generate maximum transverse field $B_1 \approx 0.5$ G (restricted by the current limit of the amplifier at the resonance frequency). The optimized parameters of the waveform used are given in Table 4.2. By repeating 3000 AFP pulses every 3 sec$^{94}$, the AFP losses were estimated to be: $< 3 \times 10^{-6}$ per flip and $< 9 \times 10^{-6}$ for the spins aligned

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$^{94}$For this measurement, the pump light polarization followed the state of the noble gas magnetization.
in the y and z direction respectively; however, as the data in Fig. 5.10 seem to suggest it is likely that the spin loss rate during the actual measurement is higher than the values reported here.
Chapter 5

Anomalous Field Measurement

The optimized comagnetometer and spin source described in the previous chapter are combined to explore non-magnetic, long range, spin-dependent forces. Here, the procedure for data acquisition and analysis is described and the results are discussed. We present the constraints on anomalous fields that the work of this thesis placed.

5.1 Experimental Procedure

The anomalous field drops with the distance from the source. Therefore, the $^3$He spin source is placed as close to the comagnetometer shield as possible, the proximity being limited by the comagnetometer shields. A simplified schematic of the experiment can be seen in Fig. 5.1.

The comagnetometer is continually prepared to work in approximately its optimum (in terms of sensitivity) state, by executing automated routines that tune the magnetic field in the longitudinal and transverse direction and zero the probe light-shift; the pump wavelength was adjusted before the run of the anomalous spin coupling measurement (see Section 4.2). As can be seen from Eq. (3.26), the comagnetometer noise performance is most sensitive to $\delta B_z$ (absolute value), which is
particularly susceptible to experimental drifts. This puts stricter requirements for the zeroing of $\delta B_z$, compared to the transverse fields and light-shifts.

In the actual experiment, data are collected in records of 200 sec, after which the $B_z$ is tuned according to the routine described in 4.2.1 (minor tuning procedure). Approximately every 90 min routines for the magnetic field adjustment in the three cartesian components and the zeroing of probe light-shift are performed in the following order (major tuning procedure):

$$
\delta B_z \delta B_z B_y \delta B_z B_x \delta B_z L_x \delta B_z \delta B_z
$$

1To a good approximation, the parameter $\delta B_z$ can be thought as the difference of two large parameters: the applied environmental longitudinal field and the effective field from the $^3$He. For instance, relative fluctuations of 1% in the $^3$He polarization (of the comagnetometer) result in $\delta B_z \sim 5 \, \mu G$. 

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Note that the longitudinal field is readjusted after every zeroing routine to ensure that the most critical comagnetometer parameter \( \delta B_z \) remains small. Both the minor (\( \delta B_z \)) and the major tuning procedures are repeated three times to reliably estimate and zero the relevant parameters. The tuning processes are associated with perturbations that reduce the comagnetometer \(^3\)He polarization; to correct for this additional relaxation, the pump power, which is electronically controlled through a feedback mechanism (see Section 4.1.5), is increased for some time above the optimal level; for the magnetic field tuning the high pump power is applied for a fraction of a second, while after the probe light-shift zeroing the pump power remains high for a couple of seconds.

The zeroing of \( \delta B_z \) also provides the calibration of the sensor (comagnetometer) and a measurement of the comagnetometer \(^3\)He effective field on \(^2\)K. The measured value of \(^3\)He magnetization is used in the PID feedback mechanism (typical timescale on the order of an hour) that controls the noble gas polarization by adjusting the pump power. As mentioned in Section 4.1.4, the choice of \(^3\)He magnetization set point relied on the (approximately) steady state noble gas polarization for incident pump rate optimized according to the high frequency modulation routine.

In the time interval that the comagnetometer tuning routines are applied, the spin source \(^3\)He polarization measurement is performed (see Section 4.7.2): the EPR frequency of \(^2\)K atoms is measured for the two orientations of the noble gas spin, without any change in the pump light polarization or in the current of the compensation coil. The measurement is passed through a digital filter that rejects outliers and applies a four point moving average analysis to estimate the spin source \(^3\)He polarization; this value is used to adjust the current in the compensation coil.

\(^2\)The applied field that zeros \( \delta B_z \) is to a good approximation the effective field that K atoms experience from the \(^3\)He magnetization.

\(^3\)The PID parameters were not fully optimized due to time constraints; the characteristic timescales of noble gas polarization evolution were used in the controller.

\(^4\)Measurements that lie beyond three times the standard deviation of the previous ten samples are considered outliers.
The recorded signal from the lock-in demodulation of the probe beam (first harmonic, in-phase component) has contributions depending on the optical path element and from the comagnetometer K-${\text{3}}^4\text{He}$ spin ensemble (Equation \((4.13)\)). In addition, the comagnetometer power spectral density is non-white, exhibiting minimum noise around the region 0.1-1 Hz. To isolate the spin contribution (which carries the information on the anomalous interaction) from the background, and in order to take advantage of the colored noise, a modulation scheme is utilized to impart a frequency (and phase) on the anomalous field: the spins in the spin source are flipped approximately every 3 sec; this results in a characteristic field frequency of approximately 0.17 Hz, which is the point of lowest comagnetometer noise. The spin reversal itself lasts $< 100$ msec, so that the acquisition dead time is only a small fraction of the recoding time. In every $\approx 200$ sec recording interval 65 flips are performed, which are accompanied with corresponding reversals of the (spin source) pump light polarization (so that the alkali and noble gas are aligned) and current polarity in the compensation coil to reduce the magnetic (but not the anomalous) field effect from the spin source spins. Because of the odd number of spin reversals, consecutive records start with opposite spin orientation and pump light helicity; a signal average over potential systematics can thus be realized and the spin source polarization is estimated from an equally weighted average of the EPR frequencies for the two end transitions $F = 2, m_F = 2 \rightarrow 1$ and $F = 2, m_F = -2 \rightarrow -1$ (as mentioned in Section \([4.7.2]\) the two end transitions result in polarization estimation relative difference $\sim 10\%$). For the experiment with the spin source in the $z$ direction (sidereal days 3117.74-3126.55), we employed a stochastic (rather than a deterministic) approach for the initial spin state of each record: at the end of every record a random number is generated (from the stochastic routine available in Labview) uniformly distributed between zero and one; for numbers greater than 0.5 an additional spin reversal is performed, while for the other number no other spin flip is realized.
For low losses in Adiabatic Fast Passage (Section 4.7.3), the effective spin destruction rate of $^3$He in the spin source is not significantly compromised from the frequent spin flip; in particular, for AFP loss rate $3 \times 10^{-6} \ (9 \times 10^{-6})$ per flip (see Section 4.7.3), and an effective flip rate $4.2 \text{ sec/flip}$ (taking into account the time interval of the tuning routines) the spin loss rate is $\sim 1/400 \ h^{-1} \ (\sim 1/130 \ h^{-1})$, which should have only a small effect on nuclear polarization. However, as will be discussed in Section 5.5 in the thesis experiments spin reversals resulted in reduced average polarization.

Special care is taken to eliminate all the electrical connections between the comagnetometer and the spin source, to avoid cross coupling through electrical paths. The two parts of the experiment (source and detector) are powered from different phases in a three-phase power circuit provided at the building where the experiment took place; this way the cross coupling through power source circuitry is reduced significantly. Furthermore, the spin source optical board is electrically isolated from the comagnetometer optical table to rule out ground loops and ground sharing paths between the two sections of the experiment.

The synchronization of the comagnetometer and spin source is realized through an optocoupled signal. When the comagnetometer starts the acquisition of data, a voltage signal (TTL, positive logic), which remains ON (high voltage) during the (comagnetometer) signal acquisition, is sent to the optical isolator; this way the two parts of the experiment acquire a common time reference point without compromising the electrical isolation. The rising edge of the synchronizing pulse triggers the spin flip process in the spin source, while the falling edge activates the $^3$He polarization measurement at the spin source (see Fig. 5.2).

All the data (related to both the comagnetometer and the spin source) are reported in reference to the Greenwich Mean Sidereal Time (GMST), in units of sidereal days

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An optocoupler (also called photocoupler and optical isolator) is composed of a light source (typically LED) that converts electric signal to light, a dielectric channel to isolate voltage surges, and a photosensor that detects the incoming light, generating an electric signal [86, 103].
since 2000 January 1st, 12h (noon) Universal Time (UT1). Before the start of every signal acquisition section, the computer that operates on the comagnetometer (and also records the signal) acquires the GMST by connecting through the internet to the US Naval Observatory time service; for the interval in between the updates (occurring approximately every day) the clock located in the DAQ board or (when no analog input occurs) in the computer provide the time base. Similarly, the spin source computer clock is updated to the US Naval Observatory time every approximately one day, and the recording (see below) rate is set by the analog input board clock. We point out that the spin source time keeping does not need to be accurate, since the synchronization pulse provides the reference point for the time adjustment.

The comagnetometer signal is recorded at 200 Hz rate during the acquisition interval; in addition, the calibration (that provides the conversion from the voltage to the effective anomalous field) and applied magnetic fields that tune the comagnetometer-
ter are recorded. Likewise, the synchronizing pulse, the fluxgate reading, the spin state (1 or -1 depending on two $^3$He spin orientation), the flip state (1 during the AFP process and 0 otherwise), and the EPR counter reading are recorded in the spin source computer at 10 Hz rate. Specifically for the noisy fluxgate signal appropriate anti-aliasing filter is utilized.

The measurement of electron paramagnetic resonance is associated with frequency components on the order of a few MHz (see Section 4.7.2), which may introduce (through radiation) electromagnetic interference (EMI) to the recorded comagnetometer signal, changing according to the comagnetometer spin state. To avoid the EMI effect, negative logic switches controlled by the synchronization pulse are placed at the input and output of the VCO; effectively the VCO feeds the EPR coil only during the comagnetometer tuning routines, when no anomalous field measurement is performed.

The effect of systematic sources of error (see Section 5.4 for a discussion), was reduced by reversing the correspondence between electronic and spin states, and also the orientation state of the comagnetometer relative to the spin source spins. When the holding field in the spin source is reversed with a simultaneous adjustment of the $\lambda/4$ waveplate to modify the pump light handedness, the anomalous field change phase with respect to the applied electronic routines in the spin source (e.g. LCW drive voltage, AFP current). Likewise, reversing the spins in the comagnetometer (by flipping the holding field and pump helicity) changes the correlation between the anomalous field measurement and certain systematic effects (see Section 5.4). In Table 5.1 the configuration states of the spin source and comagnetometer in sidereal time are shown, where we arbitrarily defined one configuration as positive and the reverse as negative. After a reversal in the comagnetometer polarization direction, the system is left to equilibrate for a couple of days (so that the noble gas polarization settles to its equilibrium value) before resuming to the actual measurement (days 3029.80-
3031.74 and 3044.84-3049.27). When the spin source geometry was changed (from spin source orientation along the $y$ axis to spin source orientation along the $z$ axis), the initial measurements were limited by systematic effects arising from spin source pump light leakage to the comagnetometer system. Additional light blocking and more careful coverage of seams and corners reduced the effect to below the experimental resolution. The large non-acquisition time interval 3057.68-3117.4 (of approximately two months) reflects the process of identifying and resolving the systematic noise of the measurement.

<table>
<thead>
<tr>
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<tbody>
<tr>
<td>3022.62-3029.80</td>
<td>y</td>
<td>+</td>
<td>+</td>
</tr>
<tr>
<td>3031.74-3036.48</td>
<td>y</td>
<td>-</td>
<td>+</td>
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<tr>
<td>3036.57-3044.84</td>
<td>y</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>3049.27-3053.50</td>
<td>y</td>
<td>+</td>
<td>-</td>
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<tr>
<td>3053.66-3057.68</td>
<td>y</td>
<td>+</td>
<td>+</td>
</tr>
<tr>
<td>3117.74-3121.60</td>
<td>z</td>
<td>+</td>
<td>+</td>
</tr>
<tr>
<td>3122.54-3126.55</td>
<td>z</td>
<td>+</td>
<td>-</td>
</tr>
</tbody>
</table>

Table 5.1: Times indicating configuration reversals of the comagnetometer or the spin source.

The comagnetometer and spin source can be arranged in different geometries to explore different types of anomalous spin interactions (see Section 5.5). We performed measurements with the spins in the spin source aligned in the $y$ and $z$ direction (see Table 5.1).

\footnote{Unless explicitly stated otherwise, the axes are labeled with respect to the comagnetometer: the pump direction is the $z$-axis, the $x$-axis lies in the plane defined by the pump and probe beam direction and $y$-axis is perpendicular to this plane.}
5.2 Data Analysis

The complete data analysis was performed in the Matlab environment. The analysis software was proved on a simulated noisy signal to be free of biases, correctly performing the statistical estimation of the parameters.

5.2.1 Digital filtering

The approximately 200 sec comagnetometer signal record is digitally filtered in frequency space by applying an effective band pass filter in the frequency region 0.08 - 1.1 Hz (we remind that the signal frequency to be measured is approximately at 0.167 Hz). In particular, the average signal over 18 sec is subtracted from the raw signal \(X_0\) to remove slow drifts (occurring at timescales larger than 18 sec). The modified record \(X_1\) goes through a low pass filter with cutoff frequency \(f_c = 0.08\) Hz (Eq. (5.1)), and the resultant is subtracted from the originally modified signal \(X_1\), thus removing low-frequency components. The difference signal \(X_2\) is further transformed (to \(X_3\)) by applying a low pass filter with cutoff frequency \(f_c = 1.1\) Hz (see Fig. 5.4(b)). The effective band pass filter in the frequency interval 0.08-1.1 Hz with the relatively flat, low noise spectrum, removes the contribution of high noise frequency components, without appreciable reduction in the power of the actual signal frequency (\(\sim 0.17\) Hz).

In Fig. 5.3 the average FFT spectrum over all the records, before and after filtering, is seen. Compared to the spectrum in Fig. 4.23 which was acquired before the start of the anomalous field measurement, the noise spectrum of the data exhibits increased power at \(\approx 0.05\) Hz, the cycle frequency of the chiller used to cool the comagnetometer system (pump and probe laser, comagnetometer shields; see Section

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8 We used white Gaussian noise for this simulation.
9 The application of the high pass filter does not render the initial long term drift removal through averaging (modified signal \(X_1\)) unnecessary; in fact the transformation leading to \(X_1\) is important to reduce “edge” effects from the digital filtering.
10 We used a Hann window for the estimation of the power spectral density [150].
and at 1.8 and 2.8 Hz, the mechanical resonance frequency of the optical table. It is not clear what contributed to the increased comagnetometer sensitivity to the chiller (a systematic study was not performed due to time constraints); the elevated noise at the mechanical resonances is simply an effect of averaging a large number of spectra, some of which were acquired during periods of increased vibrational noise.

![Power spectral density of the comagnetometer noise](image)

Figure 5.3: Power spectral density of the comagnetometer noise (blue solid line) estimated from the actual data that were used in the measurement of anomalous spin coupling field. The filtered spectrum (red dashed line) results from the application of an effective bandpass filter.

Before proceeding, we briefly describe the filtering process: the signal in time domain to be filtered is transformed to the frequency space using the (discrete) fast Fourier algorithm\[^{11}\] (FFT), and the Fourier transform is multiplied by the filter function (based on the Fermi-Dirac distribution):

\[
F(f) = \frac{1}{e^{(f-f_c)/f_w} + 1} \tag{5.1}
\]

\[^{11}\text{More specifically we used the FFT routine provided in the Matlab environment, which is based on the FFTW library}[74].\]
where \( f_c \) and \( f_w \) are the cutoff frequency and transition frequency width respectively (see Fig. 5.4 (a)); for \( f - f_c \gg f_w \), \( F \approx 0 \), while for \( f_c - f \gg f_w \), \( F \approx 1 \). An inverse Fourier transform is then applied to give the filtered signal in the time domain. For the data analysis we used \( f_w = 0.01 \) Hz.

![Figure 5.4](image)

**Figure 5.4:** (a) Digital filter function described by Eq. (5.1) for \( f_c = 1.1 \) Hz and \( f_w = 0.01 \) Hz. (b) Comagnetometer signal before (blue curve) and after (red curve) the application of digital filtering.

The application of digital filtering to the finite (approximately 200 sec) non-periodic record distorts the signal at points where sharp transitions occur; for the thesis case, the distortion is stronger at the beginning and end of the acquired record ("edge" effect). The effect depends on the bandpass region, that is on the cutoff frequency \( f_c \) and on smoothness of the filter roll-off \( f_w \); the values of \( f_c \) and \( f_w \) used are a compromise between the suppression of high noise frequency components and

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12. Partial analysis of the data that was performed for bandpass filter around the region 0.08-1.1 Hz and different \( f_w \) showed that the final result is not a sensitive function of filter parameters.

13. Unless otherwise explicitly stated, we use the term record to describe the comagnetometer recorded signal between the application of \( \delta B_z \) zeroing routines, duration of approximately 200 sec.

14. In principle, the fast spin reversals in the spin source should cause relatively sharp transitions at the comagnetometer signal (if anomalous, long-range spin coupling fields exist at all); however, the recorded signal is dominated by noise, and the ringing during the spin flips can be ignored.

15. The edge effect results from the fact that the Fourier transform effectively multiplies an infinite run of sampled data with a rectangular window which is unity during the actual sampling interval and zero otherwise; alternatively one can think that the Fourier decomposition treats the acquired signal as if it were part of a periodical signal with period the record time length.
small signal distortion. Because of the edge effects, the data during the first two spin reversals (approximately 6 sec) and the last 8 sec of the record are neglected in the analysis; overall, out of an average $\sim 260$ sec cycle (including the tuning routines and the signal recording and dead time from the resolution of the spin reversal duration) $\approx 177$ sec are used in the estimation of the anomalous field (this corresponds to an efficiency of $\sim 65\%$).

### 5.2.2 Analysis of the data within a record

From the time record of the $^3$He spin reversals in the spin source, and adjusting with the use the synchronization pulse for the time difference between the two computers that control the comagnetometer and the spin source (which recorded the comagnetometer signal and the information relevant to the spin source state respectively), the (comagnetometer signal) data that correspond to the interval between (spin source) flips are found. The mean of the $k$th ($k = 0, 1 \ldots 60$) interval (of approximately 2.8 sec) is calculated:

$$
\bar{y}_k = \frac{1}{N_p} \sum_{i=k+1}^{N_p+k} x_i 
$$

(5.2)

where $x_i$ are the filtered comagnetometer sampled data and $N_p \approx 2.8 \text{ sec} \times 200 \text{ Hz}$ is the number of samples in this interval. The uncertainty in the mean is estimated by:

$$
\sigma_{\bar{y}_k} = g_0 \sum_{i=k+1}^{N_p+k} \frac{(x_i - \bar{y}_k)^2}{\sqrt{N_p(N_p - 1)}} 
$$

(5.3)

where the factor $g_0$ corrects for the fact that the $x_i$ data are not independent; the filtering processes (and to a smaller effect the colored noise) introduce correlations between the samples\(^{16}\). It is shown in C.1.1 that under reasonable approximations relevant to the thesis case, $g_0 \sim \sqrt{112}$. To the extent that the correction due to

---

\(^{16}\)The correlation function is the Fourier transform of the power spectral density. Uncorrelated data (correlation function proportional to the Dirac delta function) correspond to white spectral density and any deviation from the flat spectrum introduces correlations.
correlations is a multiplicative factor, it does not have any effect on the evaluation of the anomalous field: the final estimate is scaled by the reduced $\chi^2$ of the data, which accounts for possible underestimation or overestimation of the calculated errors (see below).

We clarify that unless otherwise explicitly stated we follow the one sigma convention; that is, the uncertainty term describes one standard deviation.

The standard deviation of the segments within the record $\sigma_{\bar{y}_k}$ is compared to a lower and upper threshold to determine if the data correspond respectively to saturated values of the lock-in amplifier\textsuperscript{17} or to periods of significantly increased measured noise (resulting from electromagnetic pickup of RF fields generated from welding action in a room located in proximity with the experiment); the lower and upper threshold are five times smaller and larger respectively from the typical $\sigma_{\bar{y}_k}$. The records with sections outside the acceptable region are considered outliers and are not analyzed. Over the course of the month, we had very few incidents of outliers.

**String analysis**

For each record the string analysis that was introduced in [62] is performed, which isolates the signal correlated with spin reversals from background drifts. One can write for the (mean) value of the measurement between spin flips:

$$\bar{y}_k = (-1)^k s + \sum_{n=0}^{M} p_n k^n \quad (5.4)$$

where we assumed a polynomial (with coefficients $p_n$) background\textsuperscript{18} associated with various drifts on parameters affecting the measurement, and a response $s$ to the potential anomalous field which follows the spin reversals in the spin source; the total

\textsuperscript{17}Due to drifts in the optical elements the measured first harmonic signal may exceed the lock-in scale if the quarter waveplate is not readjusted in time (see Section 4.1.5).

\textsuperscript{18}This is a very general background form, since any physical small drift around a stable equilibrium value can be written as a polynomial expansion.
number of measurements used in the analysis was \( M + 1 = 61 \) (as mentioned above, the first three and the last sections of the record, which are the most susceptible to the filter edge effect, were neglected). It can be shown that\(^{19}\) the sum:

\[
\zeta = \frac{1}{2^m} \sum_{k=0}^{m} (-1)^k C^m_k \bar{y}_k
\]

(5.5)

where \( C^m_k = \frac{m!}{k!(m-k)!} \) is the binomial coefficient, \( m \) is an integer ranging from 1 to \( M \), and the \( 1/2^m \) factor normalizes \( \zeta \) so that the coefficient of the signal \( s \) is unity; does not depend on the polynomial coefficients \( p_{n=0,1,\ldots,m-1} \) \(^{[62]}\), while retaining the sensitivity to the signal \( s \). Effectively, this processing of data is equivalent to fitting a polynomial of order \( m - 1 \) through the string points in a piece-wise fashion \(^{[62]}\). We used the three point sum (\( m = 2 \) in Eq. (5.5)) in the analysis to remove the dominant linear drift; higher order drifts were typically smaller and reduction in the single string uncertainty by using more than three points does not counterbalance the losses in the final uncertainty from abridged statistics. Overall, within a single record \( M - 2 = 59 \) strings are formed with:

\[
\zeta_k = \frac{(-1)^k}{4} \left( \bar{y}_k - 2\bar{y}_{k+1} + \bar{y}_{k+2} \right)
\]

(5.6)

Each point \( \bar{y}_k \) contributes to three consecutive strings\(^{20}\), resulting in correlations between the strings. Neglecting (small) correlations on \( \bar{y}_k \) points (from filtering and colored noise), the error of each string is given by:

\[
\sigma_{\zeta_k} = \frac{1}{4} \sqrt{\sigma_{\bar{y}_k}^2 + 4\sigma_{\bar{y}_{k+1}}^2 + \sigma_{\bar{y}_{k+2}}^2}
\]

(5.7)

\(^{19}\)From the identity \((1 - X)^m = \sum_{k=0}^{m} (-1)^k C^m_k \), and differentiating with respect to \( X \).

\(^{20}\)With the exception of the first and last point which contribute only to one string, and the second and second to last which participate in two strings. Conveniently, this further reduces the edge effect from data filtering.
The mean signal for the entire record is the weighted average over the record strings:

\[
\bar{\zeta}_r = \delta_r \frac{\sum_{k=1}^{M-1} \zeta_k (\sigma_{\zeta_k})^2}{\sum_{k=1}^{M-1} 1 (\sigma_{\zeta_k})^2}
\]

(5.8)

where we introduced the parameter \(\delta_r = \pm 1\) which, depending on the record, adjusts the global phase of the signal to the correct spin orientation at the spin source. Following [87], the uncertainty in \(\bar{\zeta}_r\) is estimated to be:

\[
\sigma_{\bar{\zeta}_r} = f_0 \sqrt{\tilde{\chi}^2_1} \sqrt{\sum_{k=1}^{M-1} \frac{1}{\sigma_{\zeta_k}^2}}
\]

(5.9)

The factor \(f_0\) (approximately) corrects for the correlations of the overlapping strings. It is shown in [C.1.2] that for large number of strings in the record and similar sample uncertainties \(\sigma_{\bar{y}_k}\), \(f_0 \approx 4/\sqrt{6}\).

In Eq. (5.9), the reduced \(\chi^2\) is defined as\(^{21}\):

\[
\tilde{\chi}^2_1 = \frac{1}{M-2} \sum_{k=1}^{M-1} \frac{(\zeta_k - \bar{\zeta}_r)^2}{\sigma_{\zeta_k}^2}
\]

(5.10)

and compensates for systematic misestimation of \(\sigma_{\zeta_k}\). In this context, the reduced \(\chi^2\) effectively introduces a scaling factor for the \(\sigma_{\zeta_k}\) to correctly (under the assumption of normal distribution) describe the variance of the sampled elements \(\zeta_k\). Typically, the \(\tilde{\chi}^2\) ranged between 1 and 2, indicating that Eq. (5.7) slightly underestimate the scattering of the strings in the record. This underestimation is mainly due to uncompensated (of higher order than linear) background drift. In Fig. 5.5 the distribution of reduced \(\tilde{\chi}^2\) for all the records (in the experiment with spin source along the y

\(^{21}\)We drop the dependence of \(\bar{\zeta}_r\) on \(\delta_r\) which plays no role for this calculation. The use of \(\bar{\zeta}_r\) for the expected value of the data reduces the degrees of freedom \(d\) by one, that is \(d = M - 2\) (though in our case of large \(M\) this reduction has an insignificant effect).
direction) for strings with $m = 2$ (three point sum) and $m = 4$ (five point sum) are shown.

Figure 5.5: Reduced $\chi^2$ distribution of all the records acquired with the spin source in the $y$ direction for three-point string (a) and five-point string (b) analysis.

5.2.3 Conversion to effective magnetic field units

The estimated value and the associated error for each record are converted to effective (anomalous) magnetic field units using the calibration average from the $\delta B_z$ zeroing routine (see Section 4.3) applied at the start and end of the record. This way the reported values refer to the anomalous spin-coupling field components $b^n_y - b^c_y = \tilde{b}_y$ (see Chapter 3). We note that the statistical uncertainty in the calibration, being appreciably smaller (relative uncertainty $\sim 5\%$, see Fig. 5.6) than the scattering in the various records, has an insignificant effect on the reported anomalous field measurement.

5.2.4 Combining all the data

The data for all records are combined and the weighted average, uncertainty in the mean and reduced $\chi^2$ are calculated as before; no correction factor (e.g. $f_0$) needs
Figure 5.6: Calibration factor of the comagnetometer estimated from $\delta B_z$ zeroing.

to be applied in the estimation of error, since there are no correlations between the records.

5.3 Raw Results

5.3.1 Spin source orientation along the y axis

The anomalous field coupling was measured during about a month ($\approx 24.5$ days) to be:

$$\tilde{b}_y = (0.025 \pm 0.57) \text{ aT}$$  \hspace{1cm} (5.11)

with a reduced $\chi^2$ of 0.99. The data taken are summarized in figures 5.7 (a) and 5.8 (a). In 5.8 the various records were grouped in approximately one day interval for plotting purposes; each point is the weighted average of the records over the particular interval with the corresponding error in the mean weighted by the reduced $\chi^2$ of the records. The histogram of values for each $\approx 200$ sec record with bin size 10 aT is shown in Fig. 5.9 (a). The data can be fit reasonably well to a Gaussian function with mean -0.3 aT and standard deviation (of the distribution of records) 46 aT (this results in uncertainty in the mean of $46/\sqrt{N_r} \approx 0.58$ aT, where $N_r = 6826$
is the number of records); this estimation is consistent with the weighted average and variance evaluation, though it differs in that it does not use the errors in the record values. We also note that the measured uncertainty agrees with the prediction of the comagnetometer power spectral density shown in Fig. 5.3: for comagnetometer noise $\sim 0.75 \text{ fT/}\sqrt{\text{Hz}}$ at the signal frequency ($\approx 0.167 \text{ Hz}$) the expected noise after integration over 24.5 days with efficiency $\approx 65\%$ is $\sim 0.6 \text{ aT}$.

![Figure 5.7](image1.png)

Figure 5.7: Collection of all the records acquired with the spin source orientated in the $y$ (a) and $z$ (b) direction.

![Figure 5.8](image2.png)

Figure 5.8: Simplified plot of the spin-correlated measurement of $\tilde{b}_y$ for spin source in the $y$ (a) and $z$ (b) direction. Each point represents an average over approximately one day. Up and down triangles indicate opposite directions of the spin source, filled and empty triangles denote opposite orientations of the comagnetometer.
Neglecting the contribution of the $b_y^e$ (in literature, there are tight constraints on nonmagnetic electron spin coupling fields, see Table 5.2), the measured uncertainty is $\tilde{b}_y \approx b_y^a$ and corresponds to an energy resolution:

$$2 \times \mu (\textsuperscript{3}\text{He}) b_y^a = 7.51 \times 10^{-26} \text{ eV} \quad (5.12)$$

where $\mu (\textsuperscript{3}\text{He}) = 1.0745 \times 10^{-26} \text{ J/T}$ is the magnetic moment of $\textsuperscript{3}\text{He}$ \cite{lit} and the factor of 2 accounts for the energy difference between the states with opposite spin.\footnote{The presence of the anomalous spin coupling shifts one $\textsuperscript{3}\text{He}$ spin eigenstate by $\mu (\textsuperscript{3}\text{He}) b_y^a$ and the other spin eigenstate by $-\mu (\textsuperscript{3}\text{He}) b_y^a$ resulting in an energy spacing of $2 \mu (\textsuperscript{3}\text{He}) b_y^a$.} Equivalently the effective frequency resolution of the measurement is:

$$\gamma (\textsuperscript{3}\text{He}) b_y^a = 18.2 \text{ pHz} \quad (5.13)$$

where $\gamma (\textsuperscript{3}\text{He}) = 32.4341 \text{ MHz/T}$ is the gyromagnetic moment of $\textsuperscript{3}\text{He}$ (denoted as $\gamma_n$ in the previous chapters). To the best of our knowledge, the absolute precision of this measurement is the highest reported in literature.

\begin{figure} [h]
\centering
\subfloat[]{
\includegraphics[width=0.45\textwidth]{histogram_a}
\caption{Counts vs. Anomalous field (aT) for spin source in the $y$ (a) and $z$ (b) direction. The data closely follow a Gaussian distribution.}
}\hspace{0.5cm}
\subfloat[]{
\includegraphics[width=0.45\textwidth]{histogram_b}
\caption{Counts vs. Anomalous field (aT) for spin source in the $y$ (a) and $z$ (b) direction. The data closely follow a Gaussian distribution.}
}\end{figure}
5.3.2 Spin source orientation along the z axis

In the geometry where the spins in the spin source are orientated along the $z$ direction, similar measurements were performed for $\approx 7.7$ days resulting in uncertainty:

$$\tilde{b}_y = (-0.4 \pm 0.83) \text{ aT}$$ (5.14)

with $\chi^2 = 0.95$. The recorded data are shown in Fig. 5.7(b) and Fig. 5.8(b), and the histogram of the records, which closely matches the Gaussian distribution with mean $-0.6 \text{ aT}$ and standard deviation $23.42 \text{ aT}$ (corresponding to uncertainty in the mean of $0.86 \text{ aT}$), in Fig. 5.9(b). The measured uncertainty is consistent (though slightly lower) with the comagnetometer noise spectrum: $0.75/\sqrt{7.7 \times 0.65 \times 86400} \approx 1.1 \text{ aT}$.

5.4 Systematic Effects

Systematic noise may have a measurable effect on the values estimated from the statistical analysis of data. Here, we discuss possible sources of systematics and their potential contribution to the reported precision.

5.4.1 Magnetic field leakage to the comagnetometer

The comagnetometer exhibits suppressed (compared to anomalous spin coupling fields) but finite sensitivity to actual magnetic fields. The comagnetometer signal due to the magnetic field of the spin source follows the spin reversals, and cannot be discriminated from the potential anomalous field signal by any combination of the comagnetometer-spin source states (see Table 5.1).}

\footnote{For both geometries of the experiment (spin source in the $y$ and $z$ direction) the data (to the extent that there is no actual anomalous spin-coupling field) should come from the same distribution.}
We estimated the magnetic field effect on the comagnetometer by measuring the response to a large amplitude\(^{24}\) (100 mA) square waveform\(^{25}\) current applied to the compensation coil (see section 4.7.1). The compensation coil generates a magnetic field that closely matches the one from the spin source, and can therefore be used to calibrate the systematic effect. The fluxgate located close to the spin source served as a reference to compare the large calibrating magnetic field from the coil and the magnetic field generated by the spin source\(^{26}\) during the anomalous spin coupling measurement. For both geometries the magnetic field leakage of the spin source into the comagnetometer signal was estimated to be smaller than \(4 \times 10^{-3}\) aT, which is significantly smaller than the precision of the experiment.

### 5.4.2 Faraday effect on the optical elements

The magnetic field from the spin source may also affect the measurement through the Faraday effect of probe beam optical rotation in the optical elements \(^{27}\). Light propagating through a transparent medium (e.g. glass) in the presence of magnetic field experiences rotation in the plane of polarization and the angle of rotation \(\phi\) is given by:

\[
\phi = VB_{\parallel}d
\]

where \(B_{\parallel}\) is the magnetic flux density in the direction of light propagation, \(d\) is the path length (in practice \(d\) is the length of the material or of the magnetic field region), and \(V\) is called the Verdet constant, which depends on the material and the light

---

\(^{24}\)We applied a large amplitude to be able to clearly discriminate the effect from noise after integration over a few hours.

\(^{25}\)The frequency of the waveform was 1/6 Hz.

\(^{26}\)We remind that the magnetic field from the spin source is the difference of the spin magnetic field from the compensation coil field (see Section 4.7.1).

\(^{27}\)The Faraday effect (also called the Faraday rotation) is different (though similar) from the Faraday rotation described in Section 2.8.1. Both effects can be viewed (within the classical framework) as originating from the difference in the refractive indices for right and left circularly polarized light (circular birefringence); in the Faraday effect described here the difference results from the presence of the magnetic field, while the Faraday rotation in 2.8.1 is due to the population difference in the atomic states.
wavelength, and is proportional to the light dispersion $dn/d\lambda$; typical optical glasses have Verdet constants smaller than 10 rad/Tm in the wavelength region around 770 nm [16]. From the calibration constants relating the measured voltage with the anomalous field and the probe light polarization rotation ($\sim 110 \text{ pT/V and } \sim 1.2 \times 10^{-2} \text{ rad/V}$) it is straightforward to show that for Verdet constant $V \sim 10 \text{ rad/Tm}$ and material thickness $\sim 2 \text{ cm}$, Faraday rotation of 1 aT corresponds to a magnetic field (the component parallel to the probe beam propagation) $B_\parallel \sim 5 \mu\text{G}$. This value is roughly equal to the magnetic field from spin source at the fluxgate location (see Fig. 4.27); since the fluxgate is significantly closer (more than a factor of 8) to the spin source than any optical element in the comagnetometer probe beam path, the Faraday effect from the (dipolar) spin source magnetic field can be safely ignored as a systematic of the experiment. In fact, the limit described in Section 5.4.1 for the spin source magnetic field effect ($< 4 \times 10^{-3} \text{ aT}$), applies also for the Faraday rotation.

The Faraday effect has reversed contributions to the measurement for opposite directions of the comagnetometer spin polarization: by flipping the comagnetometer polarization (both K and $^3\text{He}$) the anomalous field signal changes sign, contrary to Faraday rotation which does not depend on the comagnetometer spin state. Averaging over the two comagnetometer orientations further reduces the effect.

5.4.3 Electronic coupling

Although special care was devoted to electronically isolate the spin source and the comagnetometer, residual parasitic coupling might occur (mainly through radiation and ground sharing paths of the powering circuitry). The contribution of electronic interference to the anomalous field estimation is suppressed by averaging over measurements for a combination of spin source-comagnetometer polarization direction as described in Section 5.1, while the signal from an anomalous spin coupling field follows the sign reversal of the comagnetometer or spin source configuration, the elec-
Electronic coupling is insensitive to the spin state (assuming that the various voltages and currents to the electronic components remain unaffected, as was the case in the thesis experiment).

### 5.4.4 Light leakage

The spin source pump light helicity is changed (using the Liquid Crystal Waveplate) synchronously with the spins during the experiment in order to maintain high $^3\text{He}$ polarization. These changes in light polarization result in intensity modulation, which affects the measurement if light is leaking through the spin source and affects the comagnetometer system. We think that such a coupling between the two systems (spin source and comagnetometer) may possibly arise either through direct detection of scattered spin source pump light at the probe beam photodiode or through the feedback mechanism that controls the pump power level at the comagnetometer. As mentioned in Section 4.7.1, the spin source and comagnetometer were covered with light blocking elements; in particular, special care had to be devoted to concealing the seams and corners.

Likewise to the systematic noise from electronic coupling, the effect of light leakage does not follow configuration reversals of comagnetometer or spin source. If for some system configuration the systematic effect to measurement is in phase with the spin coupling field signal, after a change in the configuration of the spin source or the comagnetometer the systematic effect and the actual signal become out of phase; this way, averaging over all the configuration suppresses sensitivity to potential light leakage.
5.5 Limits on anomalous spin coupling fields

The result described in the previous section can be used to constrain theoretical models involving new (that is non-magnetic) long range spin-dependent forces. It is beyond the scope of this thesis to cover all the theoretical models that have been proposed and are relevant to this measurement. Here, following [189], we limit the discussion to models that have attracted significant interest in the literature. The results reported here were originally presented in [189].

Experiments based on induced magnetism that have been performed with electron spin polarized materials [48, 26] have constrained anomalous spin-dependent interactions between electrons at a level many orders of magnitude stronger than the limit posed by the current experiment; similarly, in [198], by nuclear magnetic resonance measurements between electron and nucleon spin polarized masses, the existence of a new force between electron and nucleon spins has been excluded within the precision of the thesis experiment. We can therefore ignore the effect due to $b_y$ in the interpretation of the thesis data and confine only the nucleon sector. Table 5.2 summarizes the anomalous field constraints normalized with respect to the magnetic coupling of the particles.

In writing the Bloch equations (3.2) and (3.3) the effect of $b^n$ on K was neglected; in view of the smallness of $b^e$ one might ask whether the anomalous field that couples to the K nuclei has a measurable effect on the dynamics. It can be shown that in this case the correction to $\tilde{b}_y$ is only of order $\mu_I(K)/\mu_B \sim 2 \times 10^{-4}$ and can therefore be neglected.

The limit on the anomalous field $\tilde{b}_y \approx b^n_y$ placed by the comagnetometer measurement refers to a field that in the low energy non-relativistic regime couples to

\footnote{Even if we are to make a thorough list, undoubtedly this would soon become obsolete as new models are being introduced.}

\footnote{The main reason for the increased sensitivity of the experiments concerning anomalous electron spin interactions is the possibility of generating strong electron spin orientated sources.}
<table>
<thead>
<tr>
<th>Bound ((\xi(A, B)))</th>
<th>Method</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\xi(e, e) \leq 2.0 \times 10^{-14})</td>
<td>Induced Magnetism</td>
<td>[48], [146]</td>
</tr>
<tr>
<td>(\xi(e, e) \leq 4.2 \times 10^{-15})</td>
<td>Induced Magnetism</td>
<td>[26]</td>
</tr>
<tr>
<td>(\xi(e, n) \leq 2.3 \times 10^{-11})</td>
<td>NMR</td>
<td>[198]</td>
</tr>
<tr>
<td>(\xi(n, n) \leq 1.1 \times 10^{-5})</td>
<td>Comagnetometer</td>
<td>[84]</td>
</tr>
<tr>
<td>(\xi(p, p) \leq 4.0 \times 10^{-4})</td>
<td>Molecular spectra</td>
<td>[156]</td>
</tr>
<tr>
<td>(\xi(n, n) \leq 9.7 \times 10^{-7})</td>
<td>Inverse Square Law</td>
<td>[3]</td>
</tr>
<tr>
<td>(\xi(p, p) \leq 5.8 \times 10^{-7})</td>
<td>Inverse Square Law</td>
<td>[3]</td>
</tr>
<tr>
<td>(\xi(n, n) \leq 2.5 \times 10^{-8})</td>
<td>Comagnetometer</td>
<td>This work</td>
</tr>
<tr>
<td>(\xi(p, p) \leq 3.0 \times 10^{-6})</td>
<td>Comagnetometer</td>
<td>This work</td>
</tr>
</tbody>
</table>

Table 5.2: Bounds at 1\(\sigma\) level (68% confidence interval) on the ratio \(\xi = g_A g_B \hbar^3 / 16 \pi m_A m_B c \mu_A \mu_B\) of anomalous spin dependent (dipole)\(^2\) coupling to magnetic coupling for particles \(A\) and \(B\) from laboratory experiments. Here, \(e\) denotes electron, \(p\) denotes proton and \(n\) denotes neutron, \(m_A, B\) and \(\mu_A, B\) are respectively the mass and magnetic moment of \(A\) or \(B\) particle. In writing the nucleon limits it was assumed that each time either the proton or neutron coupling dominates. The constraint on \(\xi(e, e)\) reported in [26] is based on a calibration that is questioned in [48].

The \(^3\)He atoms (effectively to \(^3\)He nucleus) rather than the “fundamental” nucleonic entities: proton and neutron.\(^{30}\) The conversion to units appropriate for the particle physics framework is performed by expressing the field as an energy term and taking into account the nuclear spin structure of \(^3\)He: as was shown in [72], in a polarized \(^3\)He atom (nuclei), the neutron (one neutron per nuclei) polarization is 87%, the protons (two protons per nuclei) are polarized to \(-2.7\)%, and the rest of the nuclear spin comes from the orbital angular momentum. The negative proton polarization is understood as having opposite orientation from the atomic polarization. Thus, the comagnetometer is sensitive to the field:

\[
\vec{b}^y \approx b_n^y = \frac{1}{\mu(\text{He})} \left[0.87 \tilde{V}^n - 2 \times 0.027 \tilde{V}^p\right]
\]  

\(^{30}\)Of course, nucleons are not fundamental particles (they are composed of quarks), and a complete treatment would examine the coupling of the field to quarks; in fact, because of nucleon structure there are correlations between the neutron and proton coupling to a new field. In this thesis we avoid complications arising from quantum chromodynamics and electroweak interactions in quarks, treating neutrons and protons as elementary particles.
where $\tilde{V}^n$ and $\tilde{V}^p$ are respectively the average over the comagnetometer volume interaction energy of neutrons and protons in the comagnetometer $^3$He with the spin source. The factor of 2 in front of the proton interaction originates from the fact that there are two protons per $^3$He atom.

Similar arguments also hold for the spin source: for the spin source proton and neutron contributions the interaction has to be scaled by $2 \times 0.027 = 0.054$ and 0.87 respectively. Overall, the experimental scheme described here can pose constraints on neutron-neutron, proton-proton and neutron-proton spin dependent interactions; neglecting the small mass difference between protons and neutrons, the measured interaction can be written:

\[
\tilde{b}_y \propto 0.87 \left[ 0.87(g^n)^2 - 0.054g^ng^p \right] - 0.054 \left[ 0.87g^ng^p - 0.054(g^p)^2 \right] - 0.87^2(g^n)^2 - 4 \times 0.027 \times 0.87g^ng^p + (2 \times 0.027)^2(g^n)^2
\]  
(5.17)

\[
\propto 0.87^2(g^n)^2 - 4 \times 0.027 \times 0.87g^ng^p + (2 \times 0.027)^2(g^n)^2
\]  
(5.18)

where $g^n$ and $g^p$ are the anomalous force charges of neutron and proton respectively.

Figure 5.10: Spin source polarization as estimated from EPR measurements.
We stress that a fundamental theory that predicts the existence of new spin dependent interactions should provide the correlation between the charges \( g^n \) and \( g^p \) of the neutron and proton. Here, we will concentrate on effective semi-phenomenological theories, and for simplicity (and clarity) we will neglect neutron-proton and proton-proton interactions reporting only the constrains in anomalous spin dependent forces between neutrons. The extension to the other nucleon interactions (assuming each time that only one exists) is straightforward from Eq. (5.17).

The interaction energies \( \tilde{V}^n \) and \( \tilde{V}^p \) depend on the geometry of the measurement. For all forms of interactions considered here, the comagnetometer dimensions are small enough and in the following calculations the comagnetometer will be treated as a point detector.\(^{31}\) However, the spin source cannot be considered as a point source; integration over the polarized medium has to be performed resulting in a geometrical factor that depends on the spatial profile of the force.

The potentials \( \tilde{V}^n \) and \( \tilde{V}^p \) are directly proportional to the polarization of the spin source, which for the reason discussed in Section 4.7.1 (diffusion rate much faster than relaxation rate) is considered uniform over the entire volume. Since the spin source polarization is not constant in time, the limits in (5.11) and (5.14) cannot be used directly. Instead, the anomalous field value of each record and the corresponding uncertainty are divided with the spin source polarization during the particular time interval and the (weighted) mean and uncertainty in the mean of the rescaled distribution is estimated. For the geometry with the spin source along the \( y \) direction the scaled limit is: \((0.5 \pm 4.0) \text{ aT}\), while for the \( z \) geometry we find the scaled anomalous field to be: \((-3.2 \pm 6.7) \text{ aT}\).

\(^{31}\)For the approximation to hold, the characteristic length scale of the total force spatial evolution (which depends on the position with respect to the source) has to be significantly smaller than the comagnetometer radius. This condition is fulfilled for the interactions described in this work.

\(^{32}\)The values are consistent with the limits in (5.11) and (5.14) divided by the average polarization 14.2% and 12.4% respectively.
The measured spin source polarization as a function of time is shown in Fig. 5.10. The evolution of the polarization is not thoroughly understood. The data in Fig. 5.10 indicate that during the acquisition the noble gas spin relaxation in the spin source was higher than that estimated from a separate measurement (see section 4.7.3), so that the average steady state polarization was affected by the frequent spin reversals. This is manifested in polarization increase when no measurement (and spin reversal) is performed. Drifts in the steady state polarization are possibly associated with fluctuations in the pump wavelength (which is readjusted every few days) and the oven temperature. The fast polarization drop during the time interval 3049.57-3050.26 sidereal day resulted from the spin source pump laser turning off (perhaps because of overheating); the data points in this interval represent an exponential decay based on the end points. A similar incident of pump laser turn off happened in 3039.6 sidereal day. Around sidereal days 3036.5 and 3053.5 a fraction of polarization was lost due to the spin source configuration reversal.

We note that for the estimation of the anomalous interaction knowledge of the product \([\text{He}] \times P_{\text{He}}\) is required, where \([\text{He}]\) is the \(^3\text{He}\) density in the spin source and \(P_{\text{He}}\) is the noble gas polarization. Conveniently, the EPR method employed in this work (Section 4.7.2) measures this product, so that the new force estimation does not suffer from independent uncertainties in the density and polarization.

5.5.1 Constraints on pseudoscalar boson coupling

The coupling \(g_p\) of a pseudoscalar boson \(\phi\) with mass \(m_\phi\) to a fermion \(\psi\) with mass \(M_n\) can be introduced using either a Yukawa or derivative form, with Lagrangians respectively:

\[
\mathcal{L}^{\text{Yuk}} = -ig_p \bar{\psi} \gamma^5 \psi \phi
\]  

(5.19)

\(^3\)The lower index \(p\) refers to the pseudoscalar nature of the interaction (it changes sign under parity inversion, but is not affected by proper rotations [88]); the upper index is used to denote the particle (e.g. proton or neutron).
and

\[ \mathcal{L}^{\text{Der}} = \frac{g_p}{2M_n} \bar{\psi} \gamma_\mu \gamma^5 \psi \partial^\mu \phi \]  \tag{5.20} \]

where \( \gamma_\mu, \mu = 0, 1, 2, 3 \) are the Dirac matrices, \( \gamma^5 = i\gamma^0 \gamma^1 \gamma^2 \gamma^3 \), \( \bar{\psi} = \psi^\dagger \gamma^0 \) and the Einstein summation convention is assumed \[88\].

Pseudo-Goldstone bosons (the Goldstone boson from the spontaneous breaking of an approximate symmetry)\[34\] necessarily couple with the fermion fields derivatively. It can be shown that in the low energy limit the derivative coupling can be approximated as a Yukawa type interaction (Goldberger-Treiman relation \[40\])\[35\].

In the non relativistic limit, both Yukawa and derivative forms lead to the same \( 1/r^3 \) single-boson exchange potential \[142, 65\] between identical particles\[36\].

\[ V_3 (r) = \frac{g_p^2}{4\pi} \frac{\hbar^3}{4M_n^2c} \left[ \hat{\sigma}_1 \cdot \hat{\sigma}_2 \left( \frac{m_\phi c}{\hbar r^2} + \frac{1}{r^3} \right) 
- (\hat{\sigma}_1 \cdot \hat{r}) (\hat{\sigma}_1 \cdot \hat{r}) \left( \frac{m_\phi^2 c^2}{\hbar^2 r^2} + \frac{3m_\phi c}{\hbar r^2} + \frac{3}{r^3} \right) \right] e^{-m_\phi rc/\hbar} \] \tag{5.21} \]

where \( c \) is the speed of light, \( r = r\hat{r} \) is the space vector connecting the interacting fermions, \( \hat{\sigma}_1 \) and \( \hat{\sigma}_2 \) are the unit spin vectors of the particles, and \( g_p^2/4\pi \) is a dimensionless quantity that characterizes the coupling strength. In Eq. (5.21), contact interaction terms proportional to delta function were neglected.

The interaction described in (5.21) is identical to the electromagnetic interaction between spins with:

\[ \frac{g^2}{4\pi} \frac{\hbar^3}{4M_n^2c} \rightarrow \mu_n^2 \] \tag{5.22} \]

\[ ^{34}\]Contrary to a Goldstone boson (which comes from the spontaneous breaking of a global continuous exact symmetry) a pseudo-Goldstone boson can have nonzero mass \[10\].

\[ ^{35}\]However, Yukawa and derivative couplings are not always equivalent.

\[ ^{36}\]In \[142, 65\] the potential is written in Planck units (\( \hbar = c = G = 1 \), \( G \) is the gravitational constant); to convert to CGS (SI) units the following transformation has to be performed: \( V_3 \rightarrow V_3/E_P, m \rightarrow m/M_P, M_n \rightarrow M_n/M_P, r \rightarrow r/l_P \), where \( E_P = \sqrt{\hbar c^3}/G, M_P = \sqrt{\hbar c}/G \) and \( l_P = \sqrt{\hbar G}/c \). In writing the Lagrangian, we follow the Planck system to avoid unnecessary clattering of symbols. The potential lower index follows the notation in \[61\].
where $\mu_n$ is the particle magnetic moment in CGS units.

Taking into account the finite source, the nuclear structure of $^3\text{He}$, and the scaled (with the spin source polarization) effective anomalous field limit, the spin-dependent interaction between neutrons can be constrained as a function of the mass of the pseudoscalar field that mediates the force:

$$
\frac{(g_n^p)^2}{4\pi} \leq \frac{\mu \,(^3\text{He}) \, \delta \vec{b}_P}{0.87^2 \, h^3 \,[\text{He}] \, |\int_{V_{\text{Vol}}} f(\mathbf{r} - \mathbf{x}_c) d\mathbf{r}|} \frac{4m_n^2 c}{\mathcal{F}}
$$

(5.23)

where $\delta \vec{b}_P = 4.0 \, \text{aT}$ is the polarization scaled uncertainty of the anomalous field measured with the spin source orientated in the $y$ direction, $m_n$ is the neutron mass, $[\text{He}]$ is the density of $^3\text{He}$ atoms in the spin source, $\mathbf{x}_c$ is the position vector (in some reference frame) of the comagnetometer treated as a point detector, and the integral is performed over the volume of the spin source $V_{\text{Vol}}$. In Eq. (5.24) it is understood that the two spin vectors are parallel $37$: $\hat{\sigma}_1 \cdot \hat{\sigma}_2 = 1$.

In Fig. 5.11 the one sigma limit on $(g_n^p)^2/4\pi$ is shown as a function of the boson mass. The sharp feature around mass $m_\phi \sim 10^{-3} \, \text{meV}$ is due to the geometrical factor described in (5.24) which changes sign, crossing through zero. For a massless boson we obtain $38$:

$$
\frac{(g_n^p)^2}{4\pi} \leq 6.7 \times 10^{-10}
$$

(5.25)

$37$It might appear strange that when the spins in the spin source are in the $y$ direction and the spins in the comagnetometer are mainly orientated in the $z$ (pump) direction, we take $\hat{\sigma}_1 \parallel \hat{\sigma}_2$ in Eq. (5.24). That the two spin vectors should be taken parallel is most easily understood with the magnetic field analogy (which results in an identical interaction). In general, for the interaction potentials the comagnetometer spin can be considered as aligned along the sensitive $y$ axis.

$38$In $[189]$, the spin source was treated as a purely neutron source without normalizing by the $0.87$ factor, resulting in a slightly different limit than the one reported here.
The spin-spin interaction between neutrons is constrained to be smaller than the electromagnetic interaction by a factor of: 
\[ \left( \frac{g_n}{g_p} \right)^2 \frac{h^3}{(4\pi) \times (4m_n e\mu_n)^2} \leq 2.5 \times 10^{-8} \] (Table 5.2).

Figure 5.11: Constraints at 68% confidence interval on a pseudoscalar boson coupling to neutrons as a function of the boson mass. The solid line is from this work and thin dashed line from [3] for Yukawa coupling only. The thick dashed line is from $^3$He-$^{129}$Xe maser [84], while the dotted line is the limit for protons set in [156].

The limit presented (for a massless boson) in this thesis is a factor $\sim 450$ better than a recent laboratory measurement based on a $^3$He/$^{129}$Xe maser [84]. Hydrogen molecular spectroscopy has constrained proton spin dependent interactions to be: 
\[ \left( \frac{g_p}{g_n} \right)^2 \frac{h^3}{4\pi} \leq 2.3 \times 10^{-5} \] [156]. For a Yukawa form of interaction, two-boson exchange leads to limits on $g_p$ from tests of gravitational forces: 
\[ \left( \frac{g_p}{g_n} \right)^2 \frac{h^3}{4\pi} \leq 2.6 \times 10^{-8} \] [67, 3], but these limits do not apply to the derivative form that would be expected for (pseudo) Goldstone bosons, such as the axion. There are also astrophysical bounds on $g_p$ from the observed neutrino burst of supernova 1987A [155]: 
\[ 8 \times 10^{-14} \leq \left( \frac{g_n}{g_p} \right)^2 \frac{h^3}{4\pi} \leq 8 \times 10^{-8} \]; and helioseismology constraints on exotic energy loss [154]: 
\[ \left( \frac{g_p}{g_n} \right)^2 \frac{h^3}{4\pi} \leq 3 \times 10^{-9} \]. A null search for axion emissions from the Sun at 14.4 keV M1 transition in $^{57}$Fe leads to the constraint: 
\[ \left( g_p + 0.09 g_p^2 \right)^2 \frac{h^3}{4\pi} \leq 10^{-15} \] [56]. Although
astrophysical bounds are strong, they are model dependent and there are potential loopholes, lacking the robustness of laboratory measurements [61].

5.5.2 Constraints on couplings with light (pseudo) vector bosons

Spin-dependent forces can also be mediated by spin-1 particles. It is shown in [61] that under general conditions of Lorentz invariance, a generic dimension-four, low energy coupling of a spin-1, electrically neutral field $Z'$ to nucleons can be written in the form:

$$\mathcal{L} = \bar{\psi} \gamma^\mu \left( g_N^V + \gamma_5 g_A^N \right) \psi Z'_\mu$$

(5.26)

where $g_N^V$ and $g_A^N$ are the vector and axial (or pseudo) vector coupling strength respectively. The Lagrangian in Eq. (5.26) leads to a variety of rotationally invariant interactions and the reader is referred to [61] for a detailed treatment. Here, we are interested in static (that is velocity independent) spin-spin potentials: in addition to (5.21) with $g_2^2$ replaced by $g_A^2 + g_V^2$, there are two more potentials: [61]:

$$V_2 = \frac{g_A^2}{4\pi} \frac{\hbar c}{r} (\hat{\sigma}_1 \cdot \hat{\sigma}_2) e^{-m_{1/2} r c / \hbar}$$

(5.27)

$$V_{11} = -\frac{g_V g_A}{4\pi} \frac{\hbar^2}{m_n} (\hat{\sigma}_1 \times \hat{\sigma}_2) \cdot \hat{r} \left( \frac{1}{r^2} + \frac{m_{1/2} c}{\hbar r} \right) e^{-m_{1/2} r c / \hbar}$$

(5.28)

The bounds on the spin-1 coupling can be found in a manner similar to the one described in Section 5.5.1. For the $V_2$ constraint the measurement with the spin source in the $y$ direction was used, while for $V_{11}$ the data for spin source orientation along the $z$ direction were employed.\[39\]

\[39\] As mentioned in Section 5.5.1, to evaluate $V_2$ and $V_{11}$ the comagnetometer spin (e.g. $\hat{\sigma}_1$) is considered parallel to the $y$ axis; the spin source spin is either along the $y$ ($V_2$) or the $z$ ($V_{11}$) direction.
In Fig. 5.12 (a) and (b) the constraints on the vector couplings $V_2$ and $V_{11}$ respectively are plotted as a function of the mass of the boson mediating the force. Table 5.3 summarizes the bounds from the thesis experiment in the limit of a massless spin-1 particle. For the $V_3$ form, we also included the constraint to forces induced by paraphoton exchange, a massless $U(1)$ field that couples with fermions trough dimension-six operators [61, 60]. The static potential between neutrons mediated by paraphotons is (in Planck units):

$$V_{\gamma'} = -\frac{c_n^2 m_n^2}{\pi M^{4\nu^3}} [\hat{\sigma}_1 \cdot \hat{\sigma}_2 - 3 (\hat{\sigma}_1 \cdot \hat{r}) (\hat{\sigma}_1 \cdot \hat{r})]$$  \hspace{1cm} (5.29)

where $c_n$ is a dimensionless number, and the mass $M$ sets the scale where the dimension-6 operators are generated [61]. Our measurement constrains $M/\sqrt{c_n} > 11$ GeV, higher than limits from electron spin-dependent forces (see [61] for a discussion).

<table>
<thead>
<tr>
<th>$V_2$ : $g_A^2/4\pi$</th>
<th>$V_{11}$ : $g_A g_V/4\pi$</th>
<th>$V_3$ : $(g_A^2 + g_V^2)/4\pi$</th>
<th>$V_{\gamma'}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$1.4 \times 10^{-41}$</td>
<td>$4.6 \times 10^{-26}$</td>
<td>$6.7 \times 10^{-10}$</td>
<td>$6.7 \times 10^{-10}$</td>
</tr>
</tbody>
</table>

Table 5.3: Bounds on neutron couplings to massless spin-1 bosons.
In extensions of general relativity based on the Reimann-Cartan geometry (instead of the Riemann geometry) a new nonsymmetric massless field called torsion appears, which couples to fermion spins \([171, 144]\). In particular, there are theoretical predictions that torsion couples to the fermion axial-vector current mediating a spin dependent interaction of the form \(V_2 \[144]\). The constraint on \(g_5^2/4\pi\) reported here represents 0.2% of the gravitational interaction between neutrons, for the first time confining a massless spin-1 torsion field below gravitational level.

### 5.5.3 Constraints on unparticle couplings to neutrons.

A new physical entity dubbed unparticle with unusual properties was recently suggested by Georgi \([79]\). An unparticle, that could appear as a low-energy degree of freedom from a scale invariant fundamental theory, does not have a well-defined mass, and its kinematic properties are determined from its scaling dimension \(d\) (instead from a dispersion relation). It was shown in \([129]\), that exchange of unparticles between fermions can generate long-range forces with a non integral inverse power of the distance \(r\) depending on \(d\). Theoretical considerations prefer a range for \(d \in (1, 2) \[78]\). The reader is referred to \([129]\) for a derivation of the forces mediated by unparticles. Here, we simply state the result of \([129]\) relevant to the thesis experiment: treating neutron as a fundamental particle, there is an axial coupling of unparticles to neutrons\(^{40}\):

\[
\mathcal{L}_{\text{unp}} = C_A \bar{\psi} \gamma^\mu \gamma_5 \psi \mathcal{U}_\mu \tag{5.30}
\]

\(\mathcal{U}_\mu\) is the field of axial vector unparticle and the coupling parameter can be written as \(C_A = \pm c_A \Lambda^{1-d}\), where \(c_A\) is a dimensionless positive number and \(\Lambda\) is an unknown energy scale characteristic of the effective field theory. Experiments that search for spin dependent forces in the low energy can only probe \(C_A\); here, we set \(\Lambda = 1\) TeV

\(^{40}\)There is also scalar coupling which leads to spin-independent potential, and pseudoscalar and vector couplings resulting in potentials which are suppressed by a small factor \(\hbar/m_n cr \sim 2 \times 10^{-14}/r(\text{cm})\), where \(r\) is the distance between the interacting spins.
and constrain $c_A$. Neglecting small terms $\sim \hbar/m_\eta cr \sim 2 \times 10^{-14}/r$ (cm), where $r$ is the distance between the interacting particles, the potential generated by axial unparticle exchange is:

$$U_{unp} = -E_P C_A^2 \frac{16\pi^{5/2}}{(2\pi)^{2d+2}} \left( \frac{r}{l_p} \right)^{1-2d} \left( \frac{1 \text{ TeV}}{E_P} \right)^{1-d} \frac{\Gamma(d+1/2)\Gamma(2d-2)}{\Gamma(d-1)\Gamma(2d)} \hat{\sigma}_1 \cdot \hat{\sigma}_2$$ (5.31)

where $E_P \approx 1.221 \times 10^{28}$ erg is the Planck energy and $l_p \approx 1.616 \times 10^{-33}$ cm is the Planck length. The data obtained with the spin source in the $y$ direction can be used to constrain $c_A$; the bounds in neutron couplings to axial unparticles are shown in Table 5.4 and Fig. 5.13 as a function of the scaling $d$. These limits are similar to the ones obtained from electron spin-dependent force [129] and gravitational measurements [57] and are much stronger than those from astrophysics.

<table>
<thead>
<tr>
<th>$d$</th>
<th>1</th>
<th>1.25</th>
<th>1.33</th>
<th>1.5</th>
</tr>
</thead>
<tbody>
<tr>
<td>$c_A$</td>
<td>$1.3 \times 10^{-20}$</td>
<td>$9.7 \times 10^{-16}$</td>
<td>$3.4 \times 10^{-14}$</td>
<td>$6.7 \times 10^{-11}$</td>
</tr>
</tbody>
</table>

Table 5.4: Bounds on neutron coupling to axial unparticles, setting $\Lambda = 1$ TeV.

![Figure 5.13: Constraints at 68% confidence interval on axial unparticle coupling to neutron as a function of the scaling dimension $d$ ($\Lambda = 1$ TeV).](image-url)
5.5.4 Constraints on coupling to Goldstone bosons associated with spontaneous breaking of Lorentz symmetry.

The dynamical effects of a Goldstone boson $\pi$ associated with the spontaneous breaking of Lorentz invariance down to spatial rotations has been considered in [12]. Such bosons would have an unusual quadratic dispersion relationship $\omega = k^2/M_\pi$ where $M_\pi$ is the scale of spontaneous time diffeomorphism breaking, and create an aether-like fluid ("ghost condensate" or "ether wind") introducing a preferred rest frame. The leading order coupling of the $\pi$ Goldstone to fermions is given by [12]:

$$L = \frac{M_\pi^2}{F} \bar{\psi} \gamma^0 \gamma^5 \psi + \frac{1}{F} \bar{\psi} \gamma^\mu \gamma^5 \psi \partial_\mu \pi$$  \hspace{1cm} (5.32)

where $F$ is some unknown mass scale. The first term in Lagrangian leads to an anisotropic spin interaction $\hat{\sigma} \cdot \vec{v}$ in a frame moving with velocity $\vec{v}$ with respect to the preferred frame; this kind of potential (which does not depend on the presence of other spins, i.e. of the spin source) has been considered in [120] and can be probed with a modified version of the comagnetometer described in this work [33, 117].

The second term in (5.32) gives rise to a spin-spin $1/r$ potential, which depends on the orientation of $\vec{v}$ relative to the spatial vector $\vec{r}$ connecting the interacting spins and to the orientation of spins. It is shown in [12], that the potential between a spherical (radius $R$), homogeneous spin source of total spin $S_1/2$ and a test spin $S_2/2$, moving at the same velocity with respect to the ghost condensate preferred frame is given by:

$$V(\mathbf{r}) = -\frac{M_\pi^2}{F^2} \frac{hc}{8\pi \epsilon} \left[ A(\alpha, \hat{r}\hat{v}, \gamma) (S_1 \cdot S_2) + 2B(\alpha, \hat{r}\hat{v}, \gamma) (S_1 \cdot \hat{r}) (S_2 \cdot \hat{r}) 
+ C(\alpha, \hat{r}\hat{v}, \gamma) (S_1 \cdot \hat{v}) (S_2 \cdot \hat{v}) 
+ D(\alpha, \hat{r}\hat{v}, \gamma) ((S_1 \cdot \hat{v}) (S_2 \cdot \hat{r}) + (S_1 \cdot \hat{r}) (S_2 \cdot \hat{v})) \right]$$  \hspace{1cm} (5.33)
where $\alpha = Mrv/\hbar c^2$, $\gamma = MRv/\hbar c^2$ ($M$ is expressed in energy units), $\mathbf{r}$ points from the spin source to the test spin, and

$$V \circ \mathbf{W} \equiv \mathbf{V} \cdot \mathbf{W} - (\mathbf{V} \cdot \hat{r})(\mathbf{W} \cdot \hat{r})$$  \hspace{1cm} (5.34)

$$A = F_{00} + F_{10} - F_{01} \cos \theta_v, \quad B = F_{10} + \frac{F_{20}}{2}, \quad C = F_{02}, \quad D = F_{11}$$  \hspace{1cm} (5.35)

$$F_{ij} = \alpha^i \left( \frac{\partial}{\partial \alpha} \right) \left( \frac{\partial}{\partial (\hat{r} \cdot \hat{v})} \right) f(\alpha, \hat{r} \cdot \hat{v}, \gamma)$$  \hspace{1cm} (5.36)

$$f(\alpha, \theta_v, \gamma) = \frac{8\pi}{\alpha} \int \frac{d^3k}{(2\pi)^3} \frac{e^{i\vec{k}\vec{\alpha}}}{(k\hat{v} + i\epsilon)^2 - k^4} \frac{3}{(k\gamma)^3} (\sin k\gamma - k\gamma \cos k\gamma)$$  \hspace{1cm} (5.37)

In Eq. (5.37) the use of the small number $\epsilon$ implies contour integration, and $\vec{\alpha} = M\vec{r}v/\hbar c^2$. The indices in Eq. (5.36) refer to the order of integration and $\alpha^i$ is understood as $\alpha$ to the $i$th power.

![Figure 5.14: Evolution of $\cos \theta_v$ in time during one period (one sidereal day).](image)

For spins fixed on the surface of the earth (as is the case of this work) the inner products $\hat{r} \cdot \hat{v} = \cos \theta_v$, $\mathbf{S}_{1,2} \cdot \hat{v}$ vary in time as the earth rotates around its axis and the sun, that is, the force mediated by the ghost condensate has a characteristic time dependence determined by the motion of the earth with respect to the rest frame of the condensate. For concreteness, here we take this frame to be the frame in which

\footnote{It is unnatural to assume that the preferred frame follows the earth rotation.}
the Cosmic Microwave Background (CMB) radiation is spatially isotropic\(^\text{42}\), so that \(v \approx 1.23 \times 10^{-3}c\) [22]. The estimation of the inner products as a function of time is most easily performed in the Local Horizontal Coordinate (LHC) reference frame (azimuth, altitude) [141], where sidereal time naturally appears and the laboratory directions \((x, y, z)\) are easily converted to LHC coordinates\(^\text{43}\). The conversion from equatorial to LHC coordinates (where the earth velocity with respect to the CMB frame is known to be (Right Ascension, Declination)\(= (11^h11^m, -7^\circ.06)\) [22]) is given by [141]:

\[
AZ = \cos^{-1} [\sin DEC \cdot \sin LAT + \cos DEC \cdot \cos LAT \cos HA(t)]
\]

\[
ALT = \sin^{-1} \left[ \frac{\sin DEC - \sin ALT \cdot \sin LAT}{\cos ALT \cdot \cos LAT} \right]
\]

\[
HA = \left( 24t + 12 \frac{LON}{180} - RA \right) \frac{\pi}{12}
\]

where \(AZ\) is the azimuth, \(ALT\) the altitude, \(DEC\) the declination, \(RA\) the right ascension, \(LON = -74.652\) and \(LAT = 40.345\) the Princeton longitude and latitude respectively, and \(t\) is the Greenwich mean sidereal time since epoch J2000, the unit of time for the data of the experiment. A plot of \(\cos \theta_v\) as a function of time over one period is shown in Fig. 5.14. In general, the potential effect of the ghost condensate on the detector (comagnetometer) is larger when the \(\cos \theta_v\) varies over a greater range of values during the sidereal cycle; more accurately, it is preferable to have large in absolute sense negative values (close to \(-1\)) of \(\cos \theta_v\), since the interaction is strongest in the “shadow region” of the spin source [12]. Clearly, the \(\cos \theta_v\) range of values depends on the geometry of the experiment. We find that the implemented

\(^{42}\)Likewise to the ghost condensate, the CMB provides a preferred frame. However, as the universe expands the CMB radiation redshifts away and in the far future Lorentz invariance is recovered. This is not the case for the ghost condensate, which does not dilute in the expanding universe.

\(^{43}\)The experimental \(z\) axis is orientated southeast at an angle \(\approx 30^\circ\) as calculated from the mechanical drawing of the laboratory building.

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geometry was not optimized for this type of Lorentz violation measurement; a different location of the source would have resulted in higher sensitivities for the same overall source-detector separation and experimental constraints. For low mass scale $M_\pi$ the improvement is not significant, but as the shadow region becomes smaller at high $M_\pi$ the geometry affects the result considerably.

For an accurate estimation of the potential generated by the spin source, an integration over the volume of the source has to be performed, resulting in a very intensive and time consuming numerical computation. Here, for simplicity we approximate the actual cylindrical cell with a sphere of equal volume (this sets $R$) and use Eq. (5.33) directly without any spatial integration. Also, although data acquired with both $y$ and $z$ orientation of the spin source can be used in this measurement, we limit the analysis on the data with the spin source in the $y$ direction (these data provide the strongest limit). In the following we take $S_1$ and $S_2$ to be parallel to the $y$ laboratory axis. For a particular value of $M_\pi$ we first perform an analytic integration over one Cartesian component of $k$ in Eq. (5.37), and calculate numerically the remaining integrals and the derivatives in Eq. (5.36) for a time series spanning one sidereal day with step size $0.1/24$ sidereal day using the automatic algorithms provided in Wolfram Mathematica\textsuperscript{44}. This way $A, B, C$ and $D$ are densely sampled and by spline interpolation the potential in Eq. (5.33) except for a multiplication factor $G$ can be evaluated as a function of time: $V = G \tilde{V}$; from the periodicity of $\tilde{V}$, the scaled potential is extended over the acquisition window. In Fig. 5.15 $\tilde{V}$ is plotted as a function of time for four different scales $M_\pi$ of spontaneous time diffeomorphism breaking. It can be seen that as $M_\pi$ increases the interaction between the spins remains high for shorter time. The multiplication factor is given by (neglecting the sign):

\[
G = \frac{M_\pi^2}{F^2} \frac{hc}{8\pi r} S_1 S_2 \tag{5.42}
\]

\textsuperscript{44}For the numerical integration $\epsilon$ was set to 0.01; smaller values of $\epsilon$ did not change the result within the precision of the measurement.
where \( r \approx 48.7 \text{ cm} \) is the distance of the spin source center to the comagnetometer
detector, \( S_1/2 = 0.87/2 \) the detector (comagnetometer) spin and the source spin is
\( S_2/2 = N_{\text{He}} \times 0.87/2 \), \( N_{\text{He}} \) being the number of \(^3\text{He} \) atoms in the spin source (the
polarization factor is not included here; because it changes in time it is convenient
to define \( S_1 \) without dependence on polarization, and scale the experimental record
estimate with the polarization). The record values in 5.7(a) scaled by the spin source
polarization (divided) and by the \(^3\text{He} \) magnetic moment (multiplied) are fit to effec-
tively a (weighted by the uncertainties) straight line \( \mathcal{G} \dot{V}(t) \) [150]. The constraint
of \( M_\pi/F \) is calculated from the uncertainty of the estimated slope. The bounds on
\( M_\pi/F \), shown in Table 5.5 for a few values on \( M_\pi \) reach below the strength of gravi-
tational interactions. For comparison, limits on anisotropic neutron spin interactions
[19, 20] constrain \( M_\pi/F < 4 \times 10^{-17} \) for \( M_\pi = 10^{-3} \). We note that our limits
are in the regime where other operators in the theory could be large and nonlinear
interactions in the source could be significant.

| \( M_\pi \) | \( 3 \times 10^{-4} \) | \( 1 \times 10^{-3} \) | \( 3 \times 10^{-3} \) | \( 1 \times 10^{-2} \) |
| \( M_\pi/F \) | \( 2.3 \times 10^{-20} \) | \( 2.9 \times 10^{-20} \) | \( 2.4 \times 10^{-20} \) | \( 3.1 \times 10^{-20} \) |

Table 5.5: Bounds at 1\( \sigma \) level on neutron coupling to Lorentz violating Goldstone
boson.
Figure 5.15: Scaled potential $\tilde{V}$ as a function of sidereal time.
Chapter 6

Back-action evasion in RF atomic-optical magnetometry.

In this chapter a new scheme of quantum non-demolition measurement in RF atomic-optical magnetometry is discussed. First, the sources of quantum noise and the quantum measurement in an atomic-optical magnetometry are briefly reviewed. An experiment with stroboscopic light is characterized and shown to provide back-action evasion of the quantum probe noise. A simple semiclassical stochastic theory developed is sufficient to explain the observed data. Finally, it is shown theoretically, that in a certain scenario it is possible to improve long term magnetometry sensitivity with a quantum non-demolition measurement.

It is not the purpose of this work to provide a detailed theoretical description of the quantum measurement; there are many excellent books and reviews in literature that cover the subject (see for example [180, 187, 94, 123, 18, 30]). Here, the focus is in the experimental demonstration of the back-action evasion in RF magnetometry.
6.1 Notational matters

We use capital letters to denote the collective (ensemble) observables, and small letters for the individual atom observables. The hat symbol signifies the quantum character (operator) of the variable; unit vectors are typed boldface. In the following we assume that the atomic ensemble is polarized in the $z$ direction, along which a longitudinal field may be applied, and the probe beam propagates in the $x$ direction.

6.2 Quantum Noise

Magnetometry is essentially a parameter estimation process: a quantum measurement is performed on an appropriately prepared system which evolves due to the magnetic field, and the result of measurement, stochastic in nature, conditions the estimate of the field. In particular for the atomic-optical magnetometer considered in this thesis, an oriented atomic ensemble (the quantum system) interacts with light, and the analysis (filtering) of light polarization provides information about the magnetic field.

Generally, the uncertainty in estimating the parameter $\zeta$ after a measurement of the observable $\hat{T}(\zeta)$ is given by:

$$\delta\zeta = \frac{\Delta T}{\partial \langle \hat{T}\rangle / \delta \zeta}$$

(6.1)

where $(\Delta T)^2 = \langle \hat{T}^2 \rangle - \langle \hat{T} \rangle^2$, and the averages are performed for the quantum state (which may depend on the measurement history).

Fundamentally, even in the absence of experimental imperfections, the precision of estimation is limited by the uncertainties associated with the state preparation of the quantum system, the stochastic nature of interaction between atoms and light.

\footnote{Magnetometry can also be performed with aligned atomic distributions (see [38]).}
and of the light detection. The sources of noise can be characterized as photon shot noise (PSN), atom shot noise (ASN) and light shift noise (LSN) [163].

6.2.1 Photon shot noise

The photon shot noise is associated with the detection of the light field and is present even in the absence of atomic medium. It is a manifestation of the quantum nature of light (the energy is carried in quanta); the arrival of photons at the detector is random following a distribution characteristic of the field mode.

Although recently polarization squeezed light modes have been experimentally realized and have been proved to enhance sensitivity [201], we will here consider only coherent light modes (see Section 2.17.4), potentially with additional classical noise which is not affecting the measurement. For coherent light, the time interval between photon arrivals follows a Poissonian distribution [76] and the resulting power spectral density of the photocurrent is white (flat) throughout the bandwidth of detection. It was shown in Section 3.5.2 that in a balanced polarimetry detection scheme the shot noise uncertainty in the light polarization angle is (in rad/√Hz):

$$\delta \phi = \frac{1}{\sqrt{2\Phi_m \eta}}$$

(6.2)

where \( \eta \) is the efficiency of the detector and \( \Phi_m \) is the flux of photons (Photons/sec) of the light. In deriving Eq. (6.2), the light field at the input of the photodiode was assumed to be:

$$\hat{b}(t) + \hat{a}(t)$$

(6.3)

where \( \hat{b}(t) \), the local oscillator, is a large amplitude linear polarization mode, with fluctuations much smaller that the average value, and \( \hat{a}(t) \) is the vacuum coherent polarization mode orthogonal to \( \hat{b}(t) \). In the thesis experiment, \( \hat{b}(t) \) is provided by a linearly polarized laser oscillator, while the orthogonal polarization mode remains
in its vacuum state unless through Faraday interaction with the atomic medium field quanta in this mode are created\(^2\) (see [94] for an insightful exposition of the topic).

The correlation function \( C \), the Fourier transform of the power spectral density\(^3\), is (for unlimited bandwidth detection):

\[
C(\tau) = \frac{1}{4\Phi_m \eta} \delta(\tau) \quad (6.5)
\]

The balanced polarimetry scheme is robust to intensity classical noise on \(|b\rangle\); the laser intensity noise may be well above the quantum limit without affecting the resolution of the polarization angle measurement.

We note that an imperfect detector (\( \eta < 1 \)), which can be modelled by a perfect detector with a beam splitter [76], introduces additional (to the vacuum coherent field) noise; mathematically, this is expressed by multiplying the noise spectrum with \( 1/\eta > 1 \).

### 6.2.2 Light shift noise

The quantum light field perturbs the evolution of the probed atomic ensemble in a stochastic way that may affect back the probe measurement and the magnetic estimation (quantum measurement back-action).

For the conditions of the thesis experiment, with negligible tensor light-atom coupling, the physical mechanism for this indeterministic disturbance of the atom dynamics is the light-shift noise (ac-Stark shift), caused by quantum fluctuations of the

\(^2\)In the classical framework, through Faraday rotation the electric field acquires a non-zero component in the direction transverse to the initial polarization.

\(^3\)If the power spectral density (square of the noise spectrum) \( W(f) \) is expressed for positive frequencies in the frequency (Hz) domain, the correlation function can be found from the Fourier transform:

\[
C(\tau) = \left[ \int_{-\infty}^{\infty} \frac{W(f)}{2} e^{i2\pi f \tau} df \right]
\]

where \( W(f) \) is extended through negative frequencies with \( W(-f) = W(f) \).
light polarization. As was discussed in Section 2.4.6, the light-shift can be considered as an effective magnetic field which for $J_e = 1/2$ (D1 transition) is:

$$L = \frac{r_e f_{osc} \mathcal{D}(\nu)}{A_b \gamma_e} \Phi_m \frac{e^* \times e}{i} \times e_i$$  \hfill (6.6)

where $r_e$ is the electron radius, $f_{osc}$ the oscillator strength of transition, $\gamma_e$ the electron gyromagnetic ration, $\mathcal{D}(\nu)$ the detuning defined in Eq. 2.63, $A_b$ is the area of the beam (defined explicitly later), and $e$ is the polarization dependent unit vector pointing along the direction of the electric field. For the linearly polarized light field considered in the previous section, we get:

$$\Phi_m \frac{e^* \times e}{i} \times e = \frac{1}{i} \left[ \hat{b}^\dagger(t)e_z + \hat{a}^\dagger(t)e_y \right] \times \left[ \hat{b}(t)e_z + \hat{a}(t)e_y \right] = l(t)$$  \hfill (6.7)

where we explicitly expressed the direction of the field modes, assuming propagation along the $x$ axis, and linearly polarized light along the $z$ direction. From the properties of coherent vacuum field (Eq. 2.203 and 2.203), it is straightforward to show that the light-shift points along the propagation direction:

$$l = \left( \hat{a}^\dagger \hat{b} - \hat{b}^\dagger \hat{a} \right) e_x = le_x$$  \hfill (6.8)

$$\langle l \rangle = 0$$  \hfill (6.9)

$$\langle l^\dagger(t)l(t') \rangle = \Phi_m \delta(t - t')$$  \hfill (6.10)

where we set $\Phi_m = b^\dagger b$. The light-shift of linearly polarized light is then equivalent with a stochastic magnetic field along the direction of the probe beam with flat noise.

\[\text{A semiclassical approach was used to derive the light-shift. The same formulas apply in the quantum description, with the appropriate conversion of the classical electric field vector to the quantum mechanical operator.}\]
spectrum (in magnetic field units/√Hz):

\[
\delta B_{ls} = \frac{r_{e\gamma}cD(\nu)}{A_b\gamma_e}\left[\frac{4\pi}{2\pi} \int_{-\infty}^{\infty} d\omega \langle l^\dagger(t)l(t+\tau)\rangle e^{i\omega \tau}\right]^{1/2} \tag{6.11}
\]

\[
= \frac{r_{e\gamma}cD(\nu)}{\sqrt{2\Phi_m}} \tag{6.12}
\]

Similar to the PSN in Section 3.5.2, the LSN can be derived by simple Poissonian statistics. The photon spin \(s_{ph}\) can be written as:

\[
s_{ph} = \frac{(E_+ e_+ + E_- e_-)^* \times (E_+ e_+ + E_- e_-)}{i (|E_+|^2 + |E_-|^2)} \tag{6.13}
\]

\[
= \frac{N_+ - N_-}{N_+ + N_-} e_x \tag{6.14}
\]

where \(E_+\) and \(E_-\) are respectively the positive and negative helicity (or equivalently left and right circular) polarization components of the electric field, \(e_+\) and \(e_-\) are the spherical basis vectors described in Section 2.17.2. \(N_+ \propto |E_+|^2 t\) and \(N_- \propto |E_-|^2 t\) are the number of photons with positive and negative helicity respectively crossing a surface orthogonal to the light propagation over a time interval \(t\). For linearly polarized light \(\langle N_+ \rangle = \langle N_- \rangle = \Phi_m t/2\) and for Poisson distribution the uncertainty in the number of photons is: \(\delta N_+ = \delta N_- = \sqrt{\Phi_m t/2}\). It is then straightforward to show that: \(\delta s_{ph} = 1/\sqrt{\Phi_m t}\), which combined with Eq. (2.66) results in Eq. (6.12).

Although light-shift has a flat noise spectrum, its effect on the probe light polarization measurement (back-action noise) follows the characteristic spectrum of atomic response to magnetic fields or equivalently of the atom shot noise (see next section for a discussion on the ASN).

The light-shift noise is intimately connected with the photon shot noise: both originate from fluctuations in the polarization state of light. It is instructive to express this relationship in terms of the Stokes parameters (see Section 2.17.3): in a balanced polarimetry the measured quantity is proportional to the intensity difference
in the two orthogonal linear polarizations: \( \hat{a}_{z}^{\dagger} \hat{a}_{z}' - \hat{a}_{y}^{\dagger} \hat{a}_{y}' = \hat{\Xi}_1 \), while the light-shift operator is proportional to the intensity difference in the two circular polarization modes: \( \hat{a}_{+}^{\dagger} \hat{a}_{+} - \hat{a}_{-}^{\dagger} \hat{a}_{-} = \hat{\Xi}_2 \).

**Quantum non Demolition measurement**

Measurements where the operator observable of interest is a constant of motion in the Heisenberg picture are called quantum nondemolition (QND) measurements. Typically, a system observable \( \hat{X}_S \) is measured by detecting (possibly destroying) a probe observable \( \hat{X}_P \) which through the probe-system interaction carries information about \( \hat{X}_S \) (this means that the Hamiltonian interaction \( \hat{H}_I \) should depend on \( \hat{X}_S \) and \( [\hat{H}_I, \hat{X}_P] \neq 0 \) [167]). If \( \hat{H}_S \) is the coherent (non-dissipative) Hamiltonian of the system, then a QND measurement satisfies:

\[
\frac{\partial \hat{X}_S}{\partial t} = [\hat{X}_S, \hat{H}_S + \hat{H}_I] = 0 \quad (6.15)
\]

In the above definition, the effect of relaxation is not taken into account, which for the purposes of characterizing the measurement is neglected (it is implicitly assumed that the measurement strength is stronger than dissipation).

A concept closely related to QND is the back-action evasion (BAE) of the measurement on the system. BAE refers to a measurement that does not affect the statistics of the measured variable. Formally, the BAE condition is weaker than the QND. However, in the words of [200]: “the authors are not aware of any proposal for a BAE measurement that is not also a QND measurement”; this justifies why often the terms QND and BAE are used interchangeably.

\footnote{The definition implies that repeated measurements of the observable can be performed; clearly destructive measurements cannot be of the QND type.}
6.2.3 Atom shot noise

Atom shot noise, also called spin projection noise, characterizes the lack of information associated with the quantum state of the atomic system for the particular observable. In the atomic-optical magnetometer, the measured quantity is proportional to the projection of the \textit{collective} atomic spin along the propagation axis of the probe beam (see below for a discussion on the meaning of the atomic spin for conditions relevant to this work):

\[
\hat{F}_x = \sum_{i=1}^{N_{\text{at}}} w_i \hat{f}^{(i)}_x
\]  

(6.16)

\[
\sum_i w_i = N_{\text{at}}
\]  

(6.17)

where \( N_{\text{at}} \) is the number of atoms, \( w_i \) is the weight by which each atom contributes to the sum and is determined by the intensity distribution of the probe light\(^6\). For clarity, in the rest of the section we will assume \( w_i = 1 \) for all atoms, and only recover the intensity dependence on the interpretation of the experimental results. The spin projection variance \((\Delta F_x)^2\) and noise spectrum \(W_{\text{SN}}(f)\) are:\(^7\)

\[
(\Delta F_x)^2 = \text{Tr} \left[ \rho_{\text{en}} \hat{F}_x^2 \right] - \left( \text{Tr} \left[ \rho_{\text{en}} \hat{F}_x \right] \right)^2
\]  

(6.19)

\[
W_{\text{SN}}(f) = \frac{4\pi}{2\pi} \int_{-\infty}^{\infty} d\tau C(\tau) e^{i2\pi ft}
\]  

(6.20)

\[
C(\tau) = \frac{1}{2} \text{Tr} \left\{ \rho_{\text{en}}(t) \left[ \hat{F}_x(t)\hat{F}_x(t+\tau) + \hat{F}_x(t+\tau)\hat{F}_x(t) \right] \right\}
\]  

(6.21)

\(^6\)More accurately \( w_i \) is proportional to the light-atom interaction strength.

\(^7\)For an observable \( \hat{A} \) the physical measurable correlation function (which is also relevant to the calculation of the noise spectrum) is [118]:

\[
C(t,t') = \frac{1}{2} \left[ \langle \hat{A}^\dagger(t)\hat{A}(t') \rangle + \langle \hat{A}^\dagger(t')\hat{A}(t) \rangle \right]
\]  

(6.18)

where it was assumed that \( \langle \hat{A} \rangle = 0 \). In most of the cases considered here the two terms in Eq. (6.18) are equal so that: \( C(t,t') = \langle \hat{A}^\dagger(t)\hat{A}(t') \rangle \). In order to avoid notational cluttering we will only write the full correlation function when necessary.
where $\rho_{en}$ is the density matrix of the atomic ensemble. Equivalently, the spin correlation function $C(\tau)$ can be written in the Schrödinger representation (where observables are described by time independent operators and the time evolution is expressed in the quantum state) [76]:

$$C(\tau) = \text{Tr} \left[ \hat{F}_x \hat{V}(t + \tau, t) [F_x \rho_{en}(t)] \right]$$  

(6.22)

where $\hat{V}(t + \tau, t)$ is the evolution operator of the density matrix:

$$\rho_{en}(t) = \hat{V}(t, t_0) \rho_{en}(t_0)$$  

(6.23)

The noise spectrum depends on the spin dynamics, suggesting that dynamical parameters of the system can be deduced from the study of the noise spectrum (without applying any perturbations) [51]; this is a manifestation of the fluctuation-dissipation theorem [122].

**Standard quantum noise limit**

For a fully orientated (100% polarization) atomic vapor, prepared by absorption of circularly polarized (pump) light in a direction (z) orthogonal to the probe beam, the quantum state is the tensor product of the individual atom end states (for concreteness we use the pump direction as the quantization axis and we take for the single atom end state $|f, m_f\rangle = |ff\rangle$):

$$|F, M_F\rangle = |F, F\rangle = \prod_{i=1}^{N_{at}} |f^{(i)}, f^{(i)}\rangle$$  

(6.24)
This type of Dicke state\(^8\) is called the *coherent spin state*, for which:\(^9\)

\[
\langle F, F | \hat{F}_x | F, F \rangle = 0 \quad (6.26) \\
\langle F, F | \hat{F}_x^2 | F, F \rangle = N_{\text{at}} \langle f, f | \hat{f}_x^2 | f, f \rangle = N_{\text{at}} \frac{f}{2} \quad (6.27)
\]

The coherent spin state (CSS) satisfies the equality (minimum) in the Heisenberg uncertainty relation:

\[
(\Delta F_x)^2 (\Delta F_y)^2 \geq \frac{1}{4} |\langle [\hat{F}_x, \hat{F}_y] \rangle|^2
\]

(6.28)

since \((\Delta F_x)^2 = (\Delta F_y)^2 = N_{\text{at}} f/2\) and the commutator is \(\langle [\hat{F}_x, \hat{F}_y] \rangle = i \langle \hat{F}_z \rangle = iN_{\text{at}} f\) (we take \(\hbar = 1\) to avoid notational clutttering). The spin projection noise associated with the coherent spin state \((\Delta F_x) = \sqrt{N_{\text{at}} f/2}\) is called the standard uncertainty. This corresponds to atomic polarization uncertainty:

\[
\delta P_x = \frac{\delta F_x}{F} = \sqrt{\frac{1}{2N_{\text{at}} f}} 
\]

(6.29)

### Spin Squeezing

Other quantum states result in different values of the ASN. States (of polarized atoms) with uncertainty of a spin component transverse to the mean polarization smaller than the standard limit are called spin squeezed.

The squeezed state is characterized by the squeezing parameter \(\xi\); there are various definitions of \(\xi\) in literature \([113, 199, 200, 8]\) (see \([94]\) for a discussion). Here, we

\[\text{8A Dicke state is an eigenstate of both } \hat{J}^2 = \hat{J}_x^2 + \hat{J}_y^2 + \hat{J}_z^2 \text{ and } \hat{J}_z.\]

\[\text{9Equation (6.27) can be derived by expressing } \hat{f}_x \text{ in terms of the lowering and raising operators acting on the end spin state. A somewhat more elegant way is to take into account that the } |f, m_f\rangle \text{ is an eigenstate of } \hat{f}_x^2 = \hat{f}_x^2 + \hat{f}_y^2 + \hat{f}_z^2 \text{ and } \hat{f}_x, \text{ and that due to symmetry } \langle \hat{f}_x^2 \rangle = \langle \hat{f}_y^2 \rangle, \text{ so that } (\hbar = 1):} \]

\[
\hat{f}_x^2 + \hat{f}_y^2 + \hat{f}_z^2 = f(f+1) \Rightarrow 2\langle \hat{f}_x^2 \rangle + f^2 = f(f+1) \Rightarrow \langle \hat{f}_x^2 \rangle = f/2 
\]

(6.25)
follow [94] and define:

\[
\xi = \frac{\Delta F_\perp}{\sqrt{|\langle \hat{\vec{F}} \rangle|/2}}
\] (6.30)

which is appropriate for partially polarized states. In the coherent spin state \(\xi = 1\), and for a spin squeezed state \(\xi < 1\).

Contrary to optical states (which in principle can be infinitely squeezed; however, that would require infinite energy), there is a theoretical limit of how small \(\xi\) can be. From the Heisenberg uncertainty relation [11] Eq. (6.28): \((\Delta F_x)^2 \geq F^2/4(\Delta F_y)^2\) and taking into account that \((\Delta F_i)^2 \leq F^2\) (the uncertainty cannot be larger than the maximum/minimum value) we get:

\[
\xi^2 = \frac{2(\Delta F_x)^2}{F} \geq \frac{2F^2}{4(\Delta F_y)^2F} \geq \frac{F}{2F^2} = \frac{1}{2N_{at}}
\] (6.31)

This is the Heisenberg limit in spin-squeezing.

The noise reduction in one direction happens at the expense of elevated noise in the other (orthogonal to the mean polarization) direction. The physical mechanism that suppresses fluctuations in \(\hat{F}_x\) causes an increase in uncertainty for \(\hat{F}_y\) such that the Heisenberg uncertainty (6.28) is not violated.

Spin squeezing is associated with entanglement. It can be proven [184, 176] that for an atomic ensemble of spin-\(\frac{1}{2}\) particles [12] any state with \(\xi < 1\) is nonseparable (entangled), that is, the ensemble state \(\rho \) cannot be written as:

\[
\rho = \sum_k p_k \rho_{k}^{(1)} \otimes \rho_{k}^{(2)} \otimes \cdots \otimes \rho_{k}^{N_{at}}
\] (6.32)

\(^{10}\)All the definitions of \(\xi\) in literature are equivalent for fully polarized states.

\(^{11}\)Clearly, the squeezing becomes stronger (smaller \(\xi\)) in the fully polarized case \(\langle \hat{\vec{F}} \rangle = F\).

\(^{12}\)An \(F\)-spin system can be regarded as a collective system composed of \(2F\) spin-1/2 particles [123]. One can then state generally that squeezing implies correlations among the elementary spin-1/2 particles.
where \( p_k \) forms a classical probability distribution \( (p_k \geq 0, \sum_k p_k = 1) \), and \( \rho^{(i)} \) is the density matrix describing the state of the \( i \)th atom.

The effect of squeezing is represented in Fig. 6.1 where the spin ensemble is depicted as a semi-classical magnetization vector, with length proportional to the longitudinal spin and quantum uncertainty in the transverse components.

![Figure 6.1](image)

Figure 6.1: (a) A spin polarized ensemble is depicted as a semi-classical magnetization vector lying on a sphere of radius \( \langle |\hat{F}| \rangle \) (Bloch sphere), with quantum uncertainty in the transverse components. (b) For high polarization and large atomic ensembles the region of uncertainty in the Bloch sphere can be considered flat and depicted in a plane. The coherent and spin-squeezed along the \( x \) axis states are shown.

Various methods to prepare spin squeezed states in atomic media have been proposed and implemented \([94]\): they involve direct interactions between spins \([177, 152, 7]\), quantum state transfer from squeezed light to atoms \([93]\), application of a twisting \( \hat{H} \propto \hat{F}_x^2 \) or a countertwisting \( \hat{H} \propto \hat{F}_x^2 - \hat{F}_y^2 \) Hamiltonian \([113, 64]\), or a projective quantum non demolition measurement through Faraday interaction, which was the method used in this work.

**Squeezing from QND projective measurement.**

In the following, we sketch how the projective measurement modifies the quantum
system in the scenario of negligible back-action from the probe. For concreteness, spin-1/2 particles will be assumed.

Initially, the atomic medium is in a CSS, orientated in the $z$ direction. For the pedagogical (but unnatural, as that would require infinite energy and would be associated with large relaxation) case of negligible photon shot noise and atom relaxation, a measurement of $\hat{F}_x$ would produce any value $m = \frac{N_{\text{at}}}{2}, \frac{N_{\text{at}}}{2} - 1, \ldots, -\frac{N_{\text{at}}}{2}$ from a probability distribution $[153]$:

$$p\left(\hat{F}_x = m\right) = \frac{1}{2^{N_{\text{at}}}} \frac{N_{\text{at}}!}{\left(\frac{N_{\text{at}}}{2} + m\right)! \left(\frac{N_{\text{at}}}{2} - m\right)!}$$

and the quantum state would be a coherent superposition of all the states in the Hilbert space $\mathcal{H}$ ($\dim\mathcal{H} = 2^\otimes N_{\text{at}}$) for which $\hat{F}_x = m$, resulting in correlations between atoms. We say that the quantum state is conditioned to the measurement result; in the absence of any relaxation or Hamiltonian dynamics affecting $\hat{F}_x$, subsequent measurements reproduce the initial value and the projection noise is zero. This is a type of von Neumann measurement, where the quantum state “collapses” to an eigenfunction of the measured observable $\hat{F}_x$.

We illustrate this entanglement (and squeezing) generation by considering a system of two spin-1/2 particles. The initial state (spins polarized in the $z$ direction), written in the $z$ quantization axis as $|\uparrow\rangle_z |\uparrow\rangle_z$ is expressed in the $x$ quantization axis in the form of:

$$\frac{1}{2} (|\uparrow\rangle_x |\uparrow\rangle_x + |\downarrow\rangle_x |\uparrow\rangle_x + |\uparrow\rangle_x |\downarrow\rangle_x + |\downarrow\rangle_x |\downarrow\rangle_x).$$

(6.34)

A measurement of $\hat{F}_x$ producing $m = 0$, would result in a (conditioned to measurement) quantum state: $\frac{1}{2} [ |\uparrow\rangle_x |\downarrow\rangle_x + |\downarrow\rangle_x |\uparrow\rangle_x ]$ where the spins are entangled.

---

$^{13}$The equality between an operator $\hat{F}_x$ and a number is understood within the spin basis of quantization axis along the $x$ direction.
The projective measurement of negligible photon shot noise is an unrealistic oversimplification. In the case of finite light energy, the acquisition of information through the stochastic measurement (photocurrent difference in the balanced polarimetry scheme) and the corresponding update of the quantum state are gradual and occur continuously. The density matrix of the system that describes the state follows a stochastic master equation (see Section 6.3.1) and in the case of no relaxation the atomic system reaches diffusively in the limit of $t \to \infty$ one of the eigenstates of $\hat{F}_x$; the interested reader can find in [187, 179] a clear illustration of the gradual, continuous approach to a Dicke state. In an actual experiment, spin relaxation randomizes the spin as discussed in the following section, and the quantum state exhibits a diffusive behavior in a partially localized Hilbert phase space (determined from the outcome of measurement), without any collapse to a $\hat{F}_x$ eigenstate.

**Conditional and unconditional squeezing.**

In a QND projective measurement, the eigenstate of collapse (assuming no relaxation) cannot be deterministically predicted before the actual measurement. The squeezing realized is characterized as *conditional*, which refers to the fact that the squeezed state is conditioned to the (unknown before the experiment) measurement outcome. If we were to prepare an ensemble of identical independent systems and perform QND projective measurements on each system, the variance over all the QND trajectories would be equal to $(\Delta F_x)^2$ of the initial state [80].

By applying quantum feedback, the QND trajectory can be engineered so that the final outcome is steered to a desired eigenstate of $\hat{F}_x$ (in the absence of relaxation) [186]. Typically, in the atomic magnetometer the feedback mechanism is provided by the application of transverse magnetic field of amplitude that is adjusted according to the outcome of a light polarization measurement [180]. The squeezing to the predetermined state is called unconditional.
Spin relaxation effect on ASN

Spin relaxation modifies the quantum state in a stochastic way, contributing to spin noise; this is a demonstration of the fluctuation-dissipation theorem, which relates dissipation with noise processes \([122]\). The amount of squeezing in a continuous measurement is determined by the measurement strength (which sets how fast information is acquired from the system) compared to the relaxation rate (which expresses the rate of information loss). In the unrealistic scenario of negligible relaxation, a projective measurement would result (eventually) in a deterministic Dicke state with zero uncertainty in the measured observable \(\hat{F}_x\). For an actual measurement, relaxation mechanisms randomize (from the perspective of the observer) the atomic spin resulting in finite uncertainty. Relaxation processes determine the decay of the spin correlation function and the corresponding noise spectrum.

Mathematically, the effect of relaxation can be described in a model where the quantum system interacts with a heat bath (or reservoir). Due to the interaction the quantum system is entangled with the reservoir, so that in general the total density matrix cannot be written as a direct product of the system and bath states. The total density matrix for the system and bath satisfies \([76]\):

\[
\frac{d\rho_{\text{tot}}}{dt} = \frac{1}{i\hbar} \left[ \hat{H}_{\text{sys}} + \hat{H}_B + \hat{H}_{\text{int}}, \rho_{\text{tot}} \right]
\]

(6.35)

where \(\hat{H}_{\text{sys}}, \hat{H}_B\) and \(\hat{H}_{\text{int}}\) are respectively the system, bath and interaction hamiltonians. Since the bath remains unobserved, information is lost and dissipation for the quantum system arises. The reduced density matrix of the system is found from tracing over the bath states: \(\rho_{\text{sys}}(t) = \text{Tr}_B[\rho_{\text{tot}}]\).
ASN for an ensemble of alkali atoms with \( f > 1/2 \).

When no squeezing mechanism has been applied, so that there no correlations between atoms, and for atoms prepared in the same state (through a process which does not discriminate between atoms) the density matrix of the ensemble can be written in the separable form:

\[
\rho_{en} = \rho \otimes N_{\text{at}}
\]  

(6.36)

where \( \rho \) is the density matrix of a single atom, identical for all the atoms in the ensemble. The effect of collective summation operators can then found from considering a single atom and scaling appropriately with the number of atoms:

\[
\text{Tr} \left[ \rho_{en} \hat{F}_x \right] = N_{\text{at}} \text{Tr} \left[ \rho \hat{f}_x \right]
\]  

(6.37)

\[
\text{Tr} \left[ \rho_{en} \hat{F}_x^2 \right] = N_{\text{at}} \text{Tr} \left[ \rho \hat{f}_x^2 \right]
\]  

(6.38)

\[
\text{Tr} \left[ \rho_{en} \hat{F}_x(t) \hat{F}_x(t + \tau) \right] = N_{\text{at}} \text{Tr} \left[ \rho \hat{f}_x(t) \hat{f}_x(t + \tau) \right]
\]  

(6.39)

For the conditions of the thesis experiment (large buffer gas pressure and probe detuning) the alkali atoms of the ensemble cannot be considered as having definite spin quantum number \( f \). Depending on the longitudinal (along \( z \) direction) polarization, an alkali atom (when measured) can be in any of the two hyperfine manifolds \( (f = 2 \) or \( f = 1 \) for an alkali atom with nuclear spin \( I = 3/2 \)); similarly, relaxation mechanisms populate the two manifolds according to the dynamics described in Section 2.15.

The probe light interacts with the electron spin \( \hat{s}_x \) of the atoms, so that the observable is: \( \hat{S}_x = \sum_i N_{\text{at}} \hat{s}^{(i)}_x \) (we assumed \( w_i = 1 \)). From symmetry considerations: \( \langle \hat{s}^2_z \rangle = \langle \hat{s}^2_y \rangle = 1/4 \), so that in an oriented vapor of arbitrary polarization with no squeezing we get: \( (\Delta S_x)^2 = N_{\text{at}}/4 \). We stress that this is the total spin projection variance of a measurement of large enough bandwidth to include noise components in the Zeeman and hyperfine transition frequencies. In this work, the detection band-

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width was limited to \( \sim 1 \text{ MHz} \), so that the power appearing in the hyperfine transition frequency region \( \sim 450 \text{ MHz} \) did not contribute to the detected noise.

To estimate the spin noise variance measured in the experiment we write in the \( |f m_f \rangle \) basis\(^{14}\):

\[
\langle \hat{s}_x^2 \rangle = \text{Tr} [\rho \hat{s}_x^2] = \langle f m | \rho | f' m' \rangle \langle f' m' | \hat{s}_x | f'' m'' \rangle \langle f'' m'' | \hat{s}_x | f m \rangle
\]

(6.40)

and note that terms proportional to \( \langle f m | \rho | f' m' \rangle \) with \( f \neq f' \) result from hyperfine couplings and lie outside the detection bandwidth. Neglecting these terms we get for the measured variance:

\[
\Delta \hat{s}_x^2 = \langle a m | \rho | a m' \rangle \langle a m' | \hat{s}_x | a m'' \rangle \langle a m'' | \hat{s}_x | a m \rangle
\]

\[+ \langle b m | \rho | b m' \rangle \langle b m' | \hat{s}_x | b m'' \rangle \langle b m'' | \hat{s}_x | b m \rangle\]

(6.41)

\[= \langle a m | \rho | a m' \rangle \frac{1}{(2I + 1)^2} \langle a m' | \hat{f}_x | a m'' \rangle \langle a m'' | \hat{f}_x | a m \rangle
\]

\[+ \langle b m | \rho | b m' \rangle \frac{1}{(2I + 1)^2} \langle b m' | \hat{f}_x | b m'' \rangle \langle b m'' | \hat{f}_x | b m \rangle\]

(6.42)

\[= \frac{1}{(2I + 1)^2} \left[ \langle a m | \rho | a m' \rangle \langle a m' | \hat{f}_x^2 | a m \rangle + \langle b m | \rho | b m' \rangle \langle b m' | \hat{f}_x^2 | b m \rangle \right]\]

(6.43)

where \( a = I + 1/2 \) and \( b = I - 1/2 \) are the two hyperfine manifolds, \( I \) is the nuclear spin. To derive Eq. (6.42) we applied the Wigner-Eckart theorem for vector irreducible operator \( V_q \) [175]:

\[
\langle f m | V_q | f m' \rangle = \frac{\langle f m | \hat{V} \cdot \hat{f} | f m' \rangle}{\hat{f} (f + 1) \hbar^2} \langle f m | q | f m' \rangle
\]

(6.44)

and the identity:

\[
\vec{S} \cdot \vec{f} = \frac{\hbar^2}{2} [f(f + 1) + s(s + 1) - I(I + 1)]
\]

(6.45)

\(^{14}\)We use \( \langle \hat{s}_x \rangle = 0 \) and assume Einstein summation rule
Optical pumping at high pressure atomic media generate states that can be described by the spin temperature distribution (see Section 2.14):

$$\rho = e^{\beta f_z} / Z$$ \hspace{1cm} (6.46)

where \( Z \) is the partition function:

$$Z = \sum_{m=-a}^{a} e^{\beta m} + \sum_{m=-b}^{b} e^{\beta m}$$ \hspace{1cm} (6.47)

and the parameter \( \beta \) depends on the longitudinal polarization \( P_z \):

$$\beta = \ln \left[ \frac{(1 + P_z)}{(1 - P_z)} \right]$$ \hspace{1cm} (6.48)

One can show\(^{15}\) that the measured ASN variance of the collective spin is:

$$\Delta \tilde{S}_x^2 = \frac{N_{at}}{2(2I + 1)^2} \left\{ \sum_{m=-a}^{a} e^{\beta m} \left[ a(a + 1) - m^2 \right] \right. \left. + \sum_{m=-b}^{b} e^{\beta m} \left[ b(b + 1) - m^2 \right] \right\}$$ \hspace{1cm} (6.49)

In contrast to a spin-1/2 system, for \( I = 3/2 \) (e.g. K atoms), the ASN power is smaller for fully polarized atoms \( (P_z = 1) \) by a factor of 2/3 compared with unpolarized atoms.

Equation (6.49) was proven for the symmetric Hilbert space of independent, identically prepared particles. Under typical experimental conditions of optical pumping, \( \rho_{en} \) is not described by a symmetric density matrix \(^{[17]}\), and the validity of Eq. (6.49) becomes questionable. However, in Section 6.10, it is shown that experimental results reproduce the prediction of Eq. (6.49).

\(^{15}\)The spin temperature density matrix has only diagonal elements, and Eq. (6.43) includes only terms of \( \langle fm | \hat{f}_z^2 | fm \rangle = \left[ f(f + 1) - m^2 \right] / 2 \), with \( f = a \) or \( f = b \).
Spin noise spectrum.

The spin correlation function can be estimated from Eq. (6.21) or Eq. (6.22). For numerical calculations it is more convenient to use Eq. (6.22): the correlation function is expressed in the form Tr[\(\tilde{\rho}(t + \tau)\hat{s}_x\)], where \(\tilde{\rho}(t + \tau)\) is the solution of the density matrix equation in the Schrödinger picture (Section 2.15) with the initial condition \(\tilde{\rho}(t) = \hat{s}_x \rho_{eq}\), \(\rho_{eq}\) being the equilibrium density matrix (that is, the spin temperature density matrix assuming no diffusion) [76, 167].

The presence of spin-exchange collisions makes the density matrix evolution non-linear. For analytical estimations the average in spin-exchange Hamiltonian can be performed assuming the spin-temperature distribution, rendering equations (2.132) and (2.133) linear.

It is beyond the scope of this work to provide a detailed analytical formula of the spin noise spectrum. Here, we simply note that in the absence of diffusion the spin noise spectrum can be approximated by a sum of Lorentzian curves, with resonant frequencies and widths that correspond to the transition frequencies and widths respectively of the magnetic field response spectrum:

\[
W_{SN}(f) = \sum_{i_z} g_{i_z} \frac{\Delta \tilde{S}_x^2 r_{i_z}}{\pi (f - f_{0,i_z})^2 + r_{i_z}^2} + \sum_{i_h} g_{i_h} \frac{\left(\frac{N_{\uparrow}}{4} - \Delta \tilde{S}_x^2\right) r_{i_h}}{\pi (f - f_{0,i_h})^2 + r_{i_h}^2}, \ f \in (-\infty, \infty) \quad (6.50)
\]

where the summation is performed over the Zeeman (index \(z\)) and hyperfine transitions (index \(h\)), \(f_{0,i}\) and \(r_i\) are the resonant frequencies and width of the transition. The weights \(g_{i_z}\) and \(g_{i_h}\) are normalized so that \(\sum_{i_z} g_{i_z} = \sum_{i_h} g_{i_h} = 1\), and they depend on the matrix element of transition and the occupation probability of the states participating in the transition [139]. For Zeeman transitions, in the limit of

\footnote{This can be seen by application of the quantum regression theorem [76]: the same eigenvalues (eigenfrequencies and widths) characterize the linear dynamics of \(\langle \hat{O}_i(t) \rangle\) and \(\langle \hat{O}_i(t) \hat{O}_j(t) \rangle\). Although the system is not exactly linear due to the effect of spin-exchange collisions, for small transverse polarization fluctuations it can be well approximated by linear dynamics.}
well-resolved Zeeman resonances, we get:

\[ g_{j \rightarrow l} \propto \left| \langle j | \hat{s}_x | l \rangle \right|^2 |p_j - p_l| \]  

(6.51)

where \(|j\rangle\) and \(|l\rangle\) are states of the \(|f m_f\rangle\) form with occupation probabilities \(p_j\) and \(p_l\) respectively. When the longitudinal magnetic field is not large enough for all the Zeeman resonances to be well resolved the transition states should be expressed as eigenstates of the matrix \(\Lambda\) (transition matrix) defined in [9]; these eigenstates are in general superpositions of the \(|f, m_f\rangle\) eigenvectors and the weights \(g\) should be calculated accordingly.

We stress that the relative noise power under the Lorentzian curves is the same as the relative power response to a transverse magnetic field excitation.

For relatively small longitudinal magnetic fields the Zeeman frequencies lie close to each other, within a region smaller than the width of transition, so that the multiple Lorentzian structure of the spin noise spectrum is not distinguishable; instead a single quasi-Lorentzian curve with an effective width describes the data adequately.

Similarly, for highly polarized ensembles (where the suppression of spin-exchange relaxation results is narrow linewidths), most of the atoms reside in the end state (\(|2, 2\rangle\) or \(|2, -2\rangle\)) and a single exponential decay correlation function is a good approximation.

**Effect of diffusion**

In an atomic magnetometer operated with warm vapor (that is, no cold atoms are employed), atomic thermal motion may affect the accessible quantum state of the ensemble and the measured spin correlation. Diffusion to the walls of the cell containing the atoms randomizes the electron spin and a diffusional relaxation rate is introduced (see section 2.10.2). In experiments where the probe beam does not ex-

\[17\] This is only an approximation: the sum of Lorentzian curves is not a Lorentzian.
tend over the entire volume of the cell, atoms that contributed to the signal at a
particular instance can diffuse out of the beam, while other atoms that have been
previously unobserved and for which no information about the spin is known may en-
ter the light-atom interaction region. If the atoms diffusing out of the beam were to
remain permanently unobserved, a single exponential decay with rate determined by
the probe beam size would adequately describe the diffusion effect on the measured
ensemble spin correlation and would broaden the resonance without changing the
Lorentzian lineshape (transit time broadening). However, in reality, atoms that have
diffused out of the beam may return to the measurement region before the loss of their
spin coherence (due to a spin randomizing process). This way, the effective ensemble
characteristic correlation time is increased with respect to the one predicted from the
the lowest order diffusion mode. Furthermore, in the case (relevant to the work of
this thesis) where absorption (and subsequent spontaneous emission) of probe pho-
tons contributes significantly to spin relaxation, atoms that diffuse out of the beam
and spend considerable time of their coherence lifetime in the dark (where relaxation
is smaller) before returning inside the beam lead to an additional spectral narrowing
(Ramsey narrowing\[18\]). Since the atomic motion is Brownian, the time intervals in
the dark and bright regions are stochastic over a broad range of values, resulting
in a distinctly non-Lorentzian spectral profile. The interested reader is referred to
\[203, 204\] for a detailed discussion.

6.3 Quantum Measurement

Here, without emphasis on mathematical rigor, we sketch the quantum nature of
measurement and atomic ensemble state evolution. The interested reader should
follow the references for an in-depth discussion on the topic.

\[18\] In analogy to Ramsey spectroscopy \[55\].
The formulation of the problem is performed within the Markov approximation, which is based on the observation that the correlation times of bath operators are much shorter than the characteristic timescale of the system state evolution. Within this approximation, the correlation function for noise operators are delta functions and the knowledge of the system state $\rho(t)$ at one point in time $t = t_0$ is sufficient to determine $\rho(t)$ for all $t > t_0$ (equivalently, $\rho(t)$ satisfies a first order differential equation). The exact mathematical considerations for the Markov approximation can be found in [76] and [167].

6.3.1 Density Matrix Evolution

Formally, the evolution of the information state of the atomic system conditioned to the stochastic results of measurement (the quantum filtering equation) can be described through a quantum stochastic differential equation (see [76, 200, 29] and references therein).

The density matrix evolution of a particular atomic medium interacting with a far detuned linearly polarized probe light along the $x$ direction and conditioned to the stochastic result of a homodyne detection can be modeled by the stochastic Markovian master equation\textsuperscript{19} (neglecting for the moment the spin-exchange contribution; also diffusion is ignored):

$$
d\rho_{en} = \frac{1}{i\hbar} \left[ \hat{H}, \rho_{en} \right] dt + \sum_{l=1}^{N_{at}} \sum_{k} r_k \mathcal{D}_k [\hat{S}] \rho_{en} + M \mathcal{D}[\hat{S}_x] \rho_{en} + \sqrt{M \eta \mathcal{H}} [\hat{S}_x] \rho_{en} dW \quad (6.52)
$$

where the Hamiltonian $\hat{H}$ describes coherent dynamics (e.g. hyperfine interaction, or coupling to a magnetic field $\mathbf{B}$ -in this case $\hat{H} \approx g_e \mu_B \mathbf{B} \cdot \hat{\mathbf{S}}$), $\eta$ is the detection efficiency, $r_k$ the relaxation rate corresponding to the $k$th spin decoherence mechanism\textsuperscript{20}.

\textsuperscript{19}Equation (6.52) is a generalization of Eq. (2.143) (assuming no pump light) for the whole ensemble and conditional dynamics, expressing explicitly the quantum character of interaction.

\textsuperscript{20}In writing Eq. (6.52) we assumed that spin loss mechanisms act locally on very atom with the same rate (see [17] for a discussion).
dW are real numbers noise increments (associated with detection) with Gaussian white statistics: \( \mathbb{E}[dW] = 0, \ dW^2 = dt \) \[187\], \( M \) is the measurement strength (rate of extracting information from the system) which depends on the probe beam characteristics:

\[
M \propto \left[ \frac{r_e c f_{osc} D(\nu)}{A_b} \right]^2 \Phi \tag{6.53}
\]

with \( r_e \) being the electron radius, \( c \) the speed of light, \( f_{osc} \) the oscillator strength of transition, \( A_b \) the beam area, \( D(\nu) \) the detuning, \( \Phi \) the flux of photons (photons/sec), and the super-operators are (for an arbitrary operator \( \hat{c} \)):

\[
\mathcal{D}[\hat{c}] \rho_{en} = \hat{c} \rho_{en} \hat{c}^\dagger - \frac{1}{2} \left( \hat{c}^\dagger \hat{c} \rho_{en} + \rho_{en} \hat{c}^\dagger \hat{c} \right) \tag{6.54}
\]

\[
\mathcal{H}[\hat{c}] \rho_{en} = \hat{c} \rho_{en} + \rho_{en} \hat{c}^\dagger - \text{Tr} \left[ (\hat{c}^\dagger + \hat{c}) \rho_{en} \right] \rho_{en} \tag{6.55}
\]

The terms with the super-operator \( \mathcal{D} \) reflect dissipation\(^{21}\) and can be regarded as irreversible channels \[200\], while the super-operator \( \mathcal{H} \) term conditions the evolution to the result of measurement. In particular, the second term expresses collective spin decoherence from coupling the ensemble with the optical shot noise of the probe light \[80\].

The unconditional version of Eq. (6.52) (drop the last term) is justified on the grounds of a general theorem: for a Markovian master equation \( \dot{\rho} = \mathcal{L} \rho \) of a linear system, it can be shown \[178\] that the density matrix unconditional evolution operator must be of the Lindblad form:

\[
\mathcal{L} \rho = \frac{1}{i\hbar}[\hat{H}, \rho] + \sum_k \mathcal{D}[\hat{c}_k] \rho \tag{6.56}
\]

\(^{21}\)The theory developed in Chapter \[2\] suggests that the relaxation terms appear in the form \( \partial_{\epsilon} \rho / \partial t \sim -\tau \rho \) (in the Heisenberg picture: \( \partial_{\epsilon} A / \partial t \sim -r A \)). It can be shown \[78, 167\] that in the Markov approximation context, these forms result in the same decay as the dissipator \( \mathcal{D} \) with the appropriate choice of operator \( \hat{c} \).
where $\hat{H}$ is a Hermitian operator and $\hat{c}$ is arbitrary (the proof requires bounded operators)\cite{76}.

In a warm vapor, high density alkali ensemble the spin-exchange effect has to be included. Due to the nonlinear character of the interaction, the spin-exchange evolution cannot be written in the Lindbladian form; however, as mentioned above, for small perturbations the spin-exchange contribution can be linearized around the equilibrium point (spin temperature distribution for the conditions of the experiment) and written in the form of Eq. (6.56).

### 6.3.2 Quantum Langevin description

In many cases, it is more convenient to describe the evolution of the system in the Heisenberg-Langevin picture (rather than the density matrix formalism)\cite{76, 167}. For an operator $\hat{A}$ of an atomic ensemble, subject to Hamiltonian ($\hat{H}$) dynamics and relaxation, the equation of motion is (in the Markov approximation):

$$\frac{\partial \hat{A}}{\partial t} = i\frac{\hbar}{\hbar} [\hat{H}, \hat{A}] - r \hat{A} + \sqrt{2} r \hat{b}_A(t)$$

where $r$ is the decoherence rate associated with the relaxation mechanism and $\hat{b}(t)$ is a noise operator (quantum Langevin force) which depends on the reservoir variables and satisfies\cite{94, 167}:

$$\langle \hat{b}(t)\hat{b}(t') \rangle = D_A \delta(t - t')$$

The diffusion coefficient $D_A$ is determined from the state of the bath and can be found from the Einstein relations.

The quantum state of light is characterized with the Stokes parameters. The dynamical evolution of the light operators has been discussed in Section 2.17. For 22 Here, it was assumed that $\hat{A}$ is a physical observable, so that $\hat{A}^\dagger = \hat{A}$ and $\hat{b}^\dagger = \hat{b}$.
the experimental conditions of this thesis, retardation effects can be safely ignored and the problem can be formulated in terms of input output relations for the light field coupled with collective angular momenta of the atomic medium (see [123] for the subtleties of this approximation).

Before proceeding we clarify, that in the Heisenberg representation the average is performed for the initial state of the system, which implies that $\langle \rangle$ refers to the statistical average over different realizations of the measurement.

A simple quantum model

A simple approximate model that captures the main dynamical Zeeman features of the atomic-optical magnetometer considered here (longitudinal magnetic field in the $z$ direction, probe beam propagation along the $x$ axis, no pump light present) is described by the following coupled equations:

$$\frac{\partial \hat{J}_x(t)}{\partial t} = \omega_0 \hat{J}_y(t) - r \hat{J}_z(t) + \sqrt{2r} \hat{b}_x(t)$$  \hspace{1cm} (6.59)

$$\frac{\partial \hat{J}_y(t)}{\partial t} = -\omega_0 \hat{J}_x(t) - r \hat{J}_y(t) + \frac{K}{2I+1} \bar{J}_z(t) \hat{\Xi}_{1}^{\text{in}}(t) + \sqrt{2r} \hat{b}_y(t)$$  \hspace{1cm} (6.60)

$$\hat{\Xi}_{1}^{\text{out}} \equiv \eta \left[2K \hat{\Xi}_{1}^{\text{in}}(t) \hat{J}_x(t) + \hat{\Xi}_{1}^{\text{in}}(t) \right] + \eta (1 - \eta) \hat{b}_1(t)$$ \hspace{1cm} (6.61)

$$\hat{\Xi}_{2}^{\text{out}} \equiv \eta \hat{\Xi}_{2}^{\text{in}}(t) + \eta (1 - \eta) \hat{b}_2(t)$$ \hspace{1cm} (6.62)

$$\bar{J}_z(t) = \langle \hat{J}_z \rangle = \bar{J}_z(0) e^{-rt}$$ \hspace{1cm} (6.63)

where $\hat{\mathbf{J}}$ is the effective electron spin that participates in Zeeman transitions: $\hat{\mathbf{J}} = \hat{P}_a \mathbf{S} \hat{P}_a + \hat{P}_b \mathbf{S} \hat{P}_b$, $\hat{P}_a$ and $\hat{P}_b$ being projector operators in the $a = I+1/2$ and $b = I-1/2$ hyperfine manifold respectively; $\hat{\Xi}_{1,2,3}$ are the Stokes parameters defined in Section 2.17.3, $\omega_0$ is the Larmor angular frequency, $I$ is the nuclear spin of the (single) atom, $r$ and $rt$ are the respectively the transverse and longitudinal spin decoherence rates.

---

Note however that retardation effects are necessary to maintain the commutation relations [123].

The hyperfine noise spectrum is not detected.
\( \eta \) is the quantum efficiency of light detection, and \( \mathcal{K} \) is the coupling strength given by:

\[
\mathcal{K} = \frac{r_e c f_{osc} D(\nu)}{A_b} \tag{6.64}
\]

with \( A_b \) being the area of the beam. In writing the coupling of the spin with the light-shift operator, Eq. (6.6) was used taking into account that at high fields the effective gyromagnetic moment of the atoms is \( \gamma_e/(2I + 1) \) (see Section 2.16.3). The input-output terms refer to light at the input and output respectively of the atomic ensemble. The input light field is linearly polarized along the \( z \) (or \( y \)) direction and in the following vacuum coherent properties for the orthogonal polarization mode is assumed, which gives:

\[
\langle \hat{\Xi}^\text{in}_1 \rangle = 0, \quad \langle \hat{\Xi}^\text{in}_2(t)\hat{\Xi}^\text{in}_2(t') \rangle = \hat{\Xi}_0(t)\delta(t - t') \tag{6.65}
\]

The noise terms \( \hat{b}_x, \hat{b}_y \) result from spin decoherence, are Hermitian, and to a good approximation (within the Markov model)\textsuperscript{25}:

\[
\langle \hat{b}_\alpha(t)\hat{b}_\alpha(t') \rangle = D_\alpha \delta(t - t') \tag{6.66}
\]

\[
\langle \hat{b}_x(t)\hat{b}_y(t') \rangle + \langle b_y(t)b_x(t') \rangle = 0 \tag{6.67}
\]

The noise operators \( \hat{b}_x \) and \( \hat{b}_y \) are not independent. The requirement that the commutator relationship \([\hat{J}_x, \hat{J}_y] = i\hat{J}_z\) is satisfied for all times constrains the noise commutator to be: \([\hat{b}_x(t), \hat{b}_y(t')] = i\hat{J}_z(t)\delta(t - t')\). Similarly, the Langevin terms appearing in Eq. (6.61) and Eq. (6.62) originate from detection losses and satisfy vacuum coherent field properties: \( \langle \hat{b}_k(t)\hat{b}_k(t') \rangle = \hat{\Xi}_0\delta(t - t'), k = 1, 2 \). The mean of light-atom cross coupling noise operators is zero: \( \langle \hat{b}_\alpha(t)\hat{b}_k(t') \rangle = \langle \hat{b}_k(t)\hat{b}_\alpha(t') \rangle = 0, \alpha = x, y \) and \( k = 1, 2 \).

\textsuperscript{25}Equation (6.67) can be proven by expressing the noise operators \( \hat{b}_x \) and \( \hat{b}_y \) with raising and lowering noise operators and use the properties described in [167, 176].
In the model considered, dynamics associated with the tensor part of polarizability were neglected (see [123] for the effect of tensor polarizability). Furthermore, terms that have quadratic scale over fluctuations of atomic and light variables were ignored; this is the reason why in Eq. (6.60) and (6.61) the operators $\hat{J}_z$ and $\hat{\Xi}_3$ were approximated with their expectation values $\bar{J}_z$ and $\bar{\Xi}_3$ respectively. We stress that the approximation only holds in the limit of large $\bar{J}_z$ and $\bar{\Xi}_3$, so that the the Bloch and Poincaré spheres can be considered locally flat (this is related to the condition for the Holstein-Primakoff lowest order expansion to be valid). No diffusion was assumed; the spin-exchange contribution to relaxation was linearized around the equilibrium spin-temperature distribution and was included in the (total) decoherence described by the rate $r$ and the noise force $\hat{b}$.

The system of equations can be readily solved to give:

\[
\begin{align*}
\hat{J}_x(t) &= \hat{J}_x(0)e^{-rt}\cos\omega_0t + \hat{J}_y(0)e^{-rt}\sin\omega_0t \\
&\quad \frac{K}{2I+1} \int_0^t dt' \bar{\Xi}_2(t')\hat{J}_z(t')e^{-r(t-t')}\sin\omega_0(t-t') \\
&\quad + \sqrt{2r} \int_0^t dt' e^{-r(t-t')} \left[ \hat{b}_x(t') \cos\omega_0(t-t') + \hat{b}_y(t') \sin\omega_0(t-t') \right] \\
\hat{J}_y(t) &= -\hat{J}_x(0)e^{-rt}\sin\omega_0t + \hat{J}_y(0)e^{-rt}\cos\omega_0t \\
&\quad \frac{K}{2I+1} \int_0^t dt' \bar{\Xi}_2(t')\hat{J}_z(t')e^{-r(t-t')}\cos\omega_0(t-t') \\
&\quad + \sqrt{2r} \int_0^t dt' e^{-r(t-t')} \left[ \hat{b}_y(t') \cos\omega_0(t-t') - \hat{b}_x(t') \sin\omega_0(t-t') \right]
\end{align*}
\] (6.68, 6.69)

\[\text{A rigorous formulation of approximating an operator with a number can be made by performing the Holstein-Primakoff transformation [94, 101] and expanding to lowest order. The Holstein-Primakoff transformation maps the raising and lowering operator of the collective angular momentum to creation and annihilation operators of an effective oscillator mode.}\]
In the balanced polarimetry detection scheme adopted here, the Stokes parameter \( \hat{\Xi}^{\text{out}}_1 \) is measured. Using the properties of noise operators it is straightforward to show:

\[
\langle \hat{\Xi}^{\text{out}}_1(t) \rangle = 2\eta K \Xi_3(t) \langle \hat{J}_x(t) \rangle 
\]

\[
\langle \hat{\Xi}^{\text{out}}_1(t) \hat{\Xi}^{\text{out}}_1(t') \rangle = \eta \Xi_3(t) \delta(t - t') + 4\eta^2 K^2 \Xi_3(t) \Xi_3(t') \langle \hat{J}_x(t) \hat{J}_x(t') \rangle \tag{6.70}
\]

Equation (6.70) can be considered as the definition of \( K \): by equating the rotational angle \( \langle \hat{\Xi}^{\text{out}}_1(t) \rangle / (2\eta \Xi_3) \) with the Faraday rotation angle found in Section 2.8 and taking into account that \( \langle \hat{J}_x \rangle = N_{\text{at}} \langle \hat{j}_x \rangle \), Eq. (6.64) is recovered. The signal correlation function and the noise spectrum (Fourier transform of the correlation function) is the sum of a photon shot noise term (first term in the right hand side of (6.71)), and an atom shot noise possibly with light-shift contribution (last term in (6.71)).

After a bit of algebra, assuming a coherent state at \( t = 0 \) (so that \( \langle \hat{J}_x^2 \rangle = \langle \hat{J}_y^2 \rangle = (\Delta J_{x,c})^2 \) and \( \langle \hat{J}_x \hat{J}_y \rangle + \langle \hat{J}_y \hat{J}_x \rangle = 0 \)) the collective angular momentum correlation is shown to be\(^{27}\):

\[
\frac{1}{2} \left[ \langle \hat{J}_x(t) \hat{J}_x(t') \rangle + \langle \hat{J}_x(t') \hat{J}_x(t) \rangle \right] = D_{J_x} e^{-r|t-t'|} \cos \omega_0 (t - t') + \frac{K^2}{(2I + 1)^2} e^{-r(t+t')} \int_0^{\min(t,t')} d\tau e^{2r\tau} \sin [\omega_0 (t - \tau)] \sin [\omega_0 (t' - \tau)] \Xi_3(\tau) \bar{J}_x^2(\tau) \tag{6.76}
\]

\(^{27}\)The correlation estimation involves calculating integrals of the form (from the relaxation Langevin forces):

\[
\text{Int}_1 = \int_0^t \int_0^{t'} d\tau' e^{-r(t-\tau)} e^{-r(t'-\tau')} \cos [\omega_0 (t - \tau)] \cos [\omega_0 (t' - \tau')] \delta(\tau - \tau') \tag{6.72}
\]

\[
\text{Int}_2 = \int_0^t \int_0^{t'} d\tau' e^{-r(t-\tau)} e^{-r(t'-\tau')} \sin [\omega_0 (t - \tau)] \sin [\omega_0 (t' - \tau')] \delta(\tau - \tau') \tag{6.73}
\]

Their sum gives:

\[
\text{Int}_1 + \text{Int}_2 = \frac{e^{-r(t+t')} \left( -1 + e^{2r \min(t,t')} \right) \cos \omega_0 (t - t')}{2r} \tag{6.75}
\]
The term $D_J e^{-r|t-t'|} \cos \omega_0 (t - t')$ represents the atom shot noise, and the diffusion coefficient $D_J$ is the spin noise variance: $D_J = (\Delta J_{x,c})^2$ (see section 6.2.3). The last term in (6.76) describes the contribution of light-shift noise to the measurement, resulting in back-action noise (variance):

$$
B = \frac{K^2}{(2I + 1)^2} e^{-2rt} \int_0^{\min(t, t')} d\tau e^{2r\tau} (\sin [\omega_0 (t - \tau)])^2 \tilde{\Xi}_3(\tau) \bar{J}_z^2(\tau)
$$

Clearly, for $\omega_0 = 0$ the light-shift contribution, within the approximation of the model, is zero. That is, the spin measurement based on the Faraday paramagnetic interaction of light with atoms evades back-action in the limit of zero longitudinal field. For a finite longitudinal field (as in the RF magnetometry) this is not the case in general. However, the dependence of light-shift on the probe light power $\tilde{\Xi}_3$ allows the engineering of the measurement to minimize the light-shift effect (see Section 6.4).

Here, we will consider cases where the longitudinal polarization does not change significantly, and in the following the longitudinal spin will be considered approximately constant.

A semiclassical model

Operator equations can be converted to number equations. Generally, this involves defining a certain ordering of the operators and mapping products of operators with this ordering to products of complex numbers [167]. In particular for the model described in the previous section, the correspondence takes the simplest form, mapping operators to real numbers. The first and second order moments predicted from equations (6.59)-(6.61) are identical to those from the Ornstein-Uhlenbeck process.

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28 The transverse spin noise variance in the coherent state is denoted as $(\Delta J_{x,c})^2$.

29 In order to estimate the spin noise spectrum, the measurement should last longer than the transverse relaxation rate. For the experiment of this thesis, the spin-exchange contribution and the diffusion in and out of the probe beam allow the two conditions of $r/t_m \ll 1$ and $rt_m \gg 1$, $t_m$ being the measuring time, to be simultaneously satisfied.
described by the Ito differential equations:

\begin{align*}
  dJ_x &= \omega_0 J_y dt - r J_x dt + \sqrt{2r}(\Delta J_{x,c})dW_x \quad (6.78) \\
  dJ_y &= -\omega_0 J_x dt - r J_y dt + \frac{\mathcal{K}}{2t+1} J_z \sqrt{\Phi}dW_{ls} + \sqrt{2r}(\Delta J_{x,c})dW_y \\
  I_p dt &= 2\eta \Phi K J_x dt + \sqrt{\eta \Phi}dW_p \quad (6.80)
\end{align*}

where \( I_p \) is the difference photocurrent normalized with the electron charge, \( J_x, J_y \) and \( J_z \) are real numbers corresponding to the respective operators\(^{30}\), \( \Phi = \Xi_0 \) is the probe photon flux (\( \text{photons/sec} \)), \( (\Delta J_{x,c})^2 \) is the transverse collective spin uncertainty (which depends on the longitudinal polarization and is given in Eq. (6.49) for the conditions of the thesis experiment), and \( W_{x,y,ls,p}(t) \) are classical Wiener processes which satisfy:

\begin{align*}
  \langle dW_\alpha(t) dW_\beta(t') \rangle &= \delta_{\alpha\beta} dt \quad (6.81) \\
  dW_\alpha^{N+2} &= 0, \quad N > 0 \quad (6.82) \\
  dW_\alpha(t) dt &= 0 \quad (6.83)
\end{align*}

For arbitrary continuous nonanticipating\(^{31}\) functions \( G(t) \) and \( H(t) \)\(^{75}\):

\[
  \left\langle \int_{t_0}^t G(t')dW(t') \int_{t_0}^t G(t')dW(t') \right\rangle = \int_{t_0}^t \langle G(t')H(t') \rangle dt'
\quad (6.84)
\]

Equations (6.68)-(6.71) and (6.76) have their obvious correspondence in the semi-classical model.

We stress that the equivalence of the operator and real number representation results from the approximations in the quantum mechanical model: terms that have

\(^{30}\)In the quantum state diffusion language \( \mathbf{J} = \langle \hat{\mathbf{J}} \rangle_s \), the average here performed in the Schrödinger space over the pure state (characterized by \( |\psi\rangle \langle \psi| \), \( \psi \) being the Schrödinger wavefunction), which is stochastically evolving due to relaxation.

\(^{31}\)A function \( G(t) \) is called nonanticipating when \( G(t) \) does not depend on the Wiener process in the future of \( t \)\(^{75}\). Time functions of physical quantities that do not violate causality are nonanticipating.
quadratic scale over fluctuations of atomic and light variables or depend on tensor polarizability were ignored.

**Conditional dynamics**

The Heisenberg-Langevin formalism developed in the previous section describes the *unconditional* evolution of the operators characterizing the state, without any regard of the state information that is accessed through measurement. As mentioned above the averages should be interpreted statistically over the many realizations of independent measurements.

We here seek to express how the mean and variance of the observable $\hat{\Xi}^{{\text{out}}}_{1}(t)$ is conditioned on the past correlated measurements $\hat{\Xi}^{{\text{out}}}_{1}(t')$, $t' < t$. In particular, we are interested in the case of highly polarized atomic medium.

The problem can be formulated most easily in the Gaussian state approximation. For large number of atoms and longitudinal polarization and an initially coherent state, the transverse spin components follow a Gaussian probability distribution to a good approximation [31, 132]; likewise, a large photon number light field with a coherent linear polarization mode in one direction and a vacuum mode in the orthogonal direction is described approximately with Gaussian statistics. Formally, the Gaussian approximation can be derived using the Holstein-Primakoff approximation and expanding in orders of $1/N_\text{at}$ [18]. Homodyne detection and the coherent and relaxation dynamics of Eq. (6.59) - (6.60) preserve the Gaussian character within the limitations of the model [94].

A Gaussian state is fully characterized by its expectation value vector and its covariance matrix; higher moments can be calculated from the mean and covariance. Explicit formulas that describe generally the evolution of the system have been developed (see [132, 82, 68] and references therein). The formalism for an atomic-optical

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32 For the purposes of this work, a balanced polarimetry is considered equivalent to a homodyne measurement; see [29] for a comparison of homodyne and balanced polarimetry detection.
magnetometer was introduced in [132] and extended in [118]. Briefly, the combined system atomic ensemble and light field are described in a common phase space in terms of angular momentum operators (for light these are equivalent to the Stokes operators) with an operator vector \( \hat{v} \):

\[
\hat{v} = \left[ \hat{J}_x, \hat{J}_y, \hat{\xi}_1, \hat{\xi}_2 \right]
\] (6.85)

where \( \hat{J}_x, \hat{J}_y \) are the collective atomic spin operators in the directions orthogonal to the axis of polarization, and \( \hat{\xi}_1 \) and \( \hat{\xi}_2 \) are the Stokes operators of the probe beam. The covariance matrix is then defined by:

\[
\sigma_{ij} = \frac{1}{2} \left( \langle \hat{v}_i \hat{v}_j \rangle + \langle \hat{v}_j \hat{v}_i \rangle - 2 \langle \hat{v}_i \rangle \langle \hat{v}_j \rangle \right)
\] (6.86)

By partitioning the atomic ensemble and light, the model can naturally include inhomogeneities (such as non-uniform light intensity and atom relaxation) and transport (i.e. diffusion). Depending on the process involved (Hamiltonian dynamics, relaxation, diffusion or quantum-state reduction from measurements), the vector \( \hat{v} \) and covariance matrix \( \sigma \) are updated according to a well defined rule. For sufficiently small timestep, the result does not depend on the ordering of the update rules. The interested reader is referred to [132, 118] for a detailed exposition of the topic.

Alternatively, the mean and variance of a Gaussian variable \( \psi \) conditioned on the measurement of another correlated Gaussian variable \( \zeta \) with an outcome \( z \) can be calculated without evolving the full state of the system, from the following equations [94, 92]:

\[
\langle \psi \rangle|_{\zeta=z} = \langle \psi \rangle - \frac{\langle \psi \zeta \rangle z}{\langle \zeta^2 \rangle}
\] (6.87)

\[
\text{var} \left[ \psi^2 \right]|_{\zeta=z} = \text{var} \left[ \psi^2 \right] - \frac{\langle \psi \zeta \rangle^2}{\langle \zeta^2 \rangle}
\] (6.88)
The above formulas can be applied to estimate the degree of atomic spin squeezing that can be realized with a homodyne measurement. For simplicity, we consider the limit of zero longitudinal field ($\omega_0 = 0$) and use the integrated photocurrent of the balanced polarimetry measurement as the experimental outcome, assuming constant probe beam power $\Phi(t) = \Phi$. Also, the analysis will be limited to times $t \ll 1/r$. The transverse spin variance after measuring for time $t$ is:

$$ (\Delta J_x)^2|_T = (\Delta J_x)^2 - \frac{\langle J_x T \rangle^2}{\langle T^2 \rangle} \tag{6.89} $$

$$ T = \frac{1}{t} \int_0^t dt' I_p(t') \tag{6.90} $$

It is straightforward to show that to lowest order in time:

$$ \Delta J_x^2|_T = (\Delta J_{x,c})^2 \left(1 + \frac{2}{3} \eta G^2 t (\Delta J_{x,c})^2 r t \right) \tag{6.91} $$

$$ G^2 = 4\Phi K^2 \tag{6.92} $$

Assuming that at $t = 0$ the ensemble was fully polarized (along the $z$ direction), and for a probe beam with large detuning from resonance (much larger than the optical absorption width so that $D(\nu) \approx \frac{1}{\nu - \nu_0}$) the squeezing parameter is:

$$ \xi^2 = \frac{1 + \frac{9}{8} \eta OD \epsilon_p \epsilon_a \delta j_x^2}{1 - \eta rt + 4\eta OD \epsilon_p \epsilon_a \delta j_x^2} \tag{6.93} $$

where $\epsilon_p = \frac{r ec f_{osc} \Gamma}{A_0(\nu - \nu_0)} \Phi t$ ($\Gamma$ being the optical absorption HWHM in frequency units) is the number of photons absorbed by a single atom during time $t$, $\epsilon_a = rt$ is the fraction of atoms that relaxed, $\delta j_x^2$ is the single atom transverse electron spin variance, and $OD$ is the optical depth on resonance: $OD = [A] \sigma_0 l$; $[A]$ is the alkali density, $l$ is the length of the ensemble along the probe beam propagation and $\sigma_0 = r e c f_{osc}/\Gamma$ is the absorption cross section on resonance.

\[\text{\footnotesize If the spin relaxation originates from probe light absorption then } \epsilon_a = \epsilon_p.\]
We stress that the squeezing described above is based on applying an averaging filter to estimate the state and reduce the variance. This particular filter, which can be easily implemented in the laboratory, equally weights the photocurrent for the measuring time. However, the average filter is not optimal in the sense that the information from the measurement process can be processed differently to give stronger squeezing. As shown in [180, 80], for times where the spin decay is negligible, a quantum Kalman filter, which strategically (incorporating the system dynamics) underweights the photocurrent at early times, provides an optimal parameter estimator. For larger timescales where the effect of decoherence is significant, the optimal filtering has been derived in the case of spin-1/2 particles (no nuclear spin) in [[132]] using the full correlation matrix of the system. Despite the complexity, the optimal filtering can be implemented in real time with low delay, e.g. with a Field-programmable Gate Array integrated circuit.

6.4 Stroboscopic back-action

The contribution of probe back-action on the spin variance ($B$) depends on the temporal profile of the probe light power (Eq. (6.77)). The back-action noise cannot be eliminated for all times, since there is no functional form of $\Xi_3$ that gives $B(t) = 0$ for every $t$. The longitudinal magnetic field causes the spin uncertainty distribution to rotate, so that the light-shift noise in $\hat{J}_y$ evolves (through coherent dynamics) to noise in $\hat{J}_x$, and over timescales larger than the Larmor period both $\hat{J}_x$ and $\hat{J}_y$ accumulate the back-action noise.

This restriction in $(\Delta J_x)^2$ does not necessarily limit the atomic-optical magnetometer. The variable that is relevant to the measurement and RF magnetic field

\footnote{In fact the optimality of Kalman filtering can be proved from the Bayes’ rule \cite{80}.}
estimation can be written in the form:

\[
\hat{X}(t) = c_n \int_{t}^{t+T} d\tau u(\tau)\Phi(\tau)\hat{J}_x(\tau)
\]  

(6.94)

where \( T = 2\pi/\omega_0 \) is the Larmor period of oscillation, \( u(t) \) is a post-measurement filter appropriate to estimate the RF field from the measurement outcome, and \( c_n \) is a normalization constant, which is chosen to be:

\[
c_n = \frac{1}{\sqrt{2} \int d\tau u(\tau)\Phi(\tau) \cos [\omega_0 \tau - \phi]}
\]  

(6.95)

The parameter \( \phi \) depends on the power temporal profile and adjusts the phase of the cosine so that \( \hat{X} \) measures the rms amplitude of \( \hat{J}_x \) when a coherent oscillating RF field at Larmor frequency is applied. The back-action contribution to the noise of \( \hat{X} \) is:

\[
\tilde{B} = c_n^2 \frac{K^2}{(2I + 1)^2} \int_t^{t+T} d\tau d\tau' \left\{ u(\tau)\Phi(\tau)u(\tau')\Phi(\tau)e^{-r(\tau+\tau')} \right. \\
\times \left. \int_0^{\min(\tau,\tau')} d\zeta e^{-2r\zeta} \sin[\omega_0(\tau - \zeta)] \sin[\omega_0(\tau' - \zeta)] \Phi(\zeta)\hat{J}_z^2(\zeta) \right\}
\]  

(6.96)

The probe light power can be engineered to suppress back-action on \( \hat{X} \) during the measuring time. Using stroboscopic light that turns on and off at twice the Larmor frequency of the squeezed distribution.
frequency, a measurement is performed only when the squeezed distribution is aligned with the probe axis in the laboratory frame, as illustrated in Fig. 6.2.

Consider the case of probe light with:

\[ u(t)\Phi(t) \propto \sum_{k} (-1)^{k} \delta \left( t - k\frac{T}{2} \right), \quad k \in \mathbb{Z} \quad (6.97) \]

Equation (6.97) describes stroboscopic light that is instantaneously turned on every half a period; the \((-1)^{k}\) factor adjusts the sign of the measurement so that the effect of a potential RF field does not average to zero over a Larmor cycle. Combining Eq. (6.97) and (6.96), it can be seen that the probe back-action contribution to \(\hat{X}\) is expressed as a sum of terms proportional to: \(\sin k\pi \sin k'\pi, k, k' \in \mathbb{Z}\), which are all zero.

The delta functional form in Eq. (6.97) is an idealization. In actual experiments, the light remains on for a finite time, resulting in non-zero back-action on the measured variable \(\hat{X}\). In the following, the simple form of a top hat profile is assumed, with a duty cycle \(d\) defined as the fraction of stroboscopic period \(T/2\) the light is on.

We consider the limit (applicable to high Larmor frequencies) where the fraction of atoms relaxing during the Larmor period is negligible \((\tau T \ll 1)\). Also, the analysis is simplified by discretizing \(\hat{X}\) at times integer multiples of \(T\). The measured variable is then defined for integer \(k\) as:

\[ \hat{X}(k) = c_n \int_{kT}^{(k+1)T} d\tau u(\tau)\Phi(\tau)\hat{J}_x(\tau) \quad (6.98) \]

\[ = c_d \int_{kT}^{kT+d\frac{T}{2}} d\tau \hat{J}_x(\tau) - c_d \int_{kT+d\frac{T}{2}}^{(k+1)T} d\tau \hat{J}_x(\tau) \quad (6.99) \]

\[ = \hat{X}_1(k) - \hat{X}_2(k) \quad (6.100) \]
where the normalization constant $c_d$ can be written in the form:

$$c_d = \frac{\pi}{2\sqrt{2}T \sin \pi d/2} \quad (6.101)$$

After a bit of algebra, the correlation function $\langle \hat{X}(N_1)\hat{X}(N_2) \rangle$ can be shown to be:

$$\langle \hat{X}(N_1)\hat{X}(N_2) \rangle \approx \frac{(\Delta J_{x,c})^2}{2} e^{-|N_1-N_2| r T}$$

$$+ \frac{K^2}{(2I+1)^2} J_z^2 e^{-|N_1-N_2| r T} - e^{-(N_1+N_2) r T} \frac{d}{8r} \left(1 - \frac{\sin \pi d}{\pi d}\right) \quad (6.102)$$

The above equation was derived assuming negligible longitudinal polarization decay. The first term in the right hand side of Eq. (6.102) is due to the atom shot noise and the second term describes the back-action of the probe. To estimate the stationary correlation function [35] (which should be used for the calculation of the noise spectrum) we set the initial time at $-\infty$ rather than 0, taking $e^{-(N_1+N_2) r T} \approx 0$. Then, the stationary back-action variance is:

$$\text{var}[\hat{X}]_{BA} = \frac{K^2}{(2I+1)^2} J_z^2 d \left(1 - \frac{\sin \pi d}{\pi d}\right) \quad (6.103)$$

The back-action noise is not a sensitive function of the duty cycle and for small $d$ it becomes:

$$\text{var}[\hat{X}]_{BA} \approx K^2 J_z^2 \frac{\pi^2 d^3}{48r} + \mathcal{O}[d^5] \quad (6.104)$$

The conditional variance of $\hat{X}$ can be calculated from Eq. (6.88) and the correlation function $\langle \hat{X}(N_1)\hat{X}(N_2) \rangle$. The measured variable that conditions the evolution

[35] A stationary correlation function can only exist if the longitudinal polarization remains constant.
\[ \zeta(N) = \sum_{k=0}^{N} \ddot{X}(k) \]  
\[ \ddot{X}(k) = 2\mathcal{K}\eta \dot{X}_k + c_d \sqrt{\frac{\eta}{d\Phi}} \left[ \int_{kT}^{kT+\frac{d}{2}} dW_p - \int_{kT+\frac{d}{2}}^{kT+(d+1)T} dW_p \right] \]

with \( \Phi \) being the average photon flux over a period \( T \). In the limit of negligible longitudinal polarization relaxation the conditional variance after measurement time \( t = NT \) is:

\[ \Delta X^2(N) \big|_{\zeta(N-1)} = \frac{1}{2} \left[ 1 + \frac{2\eta rtG^2 \sin^2[\pi d/2]}{3\pi^2 d^2/4} \left[ 1 - \frac{j_z^2 K^2 d}{2r(\Delta J_{x,c})^2(2I+1)^2} \left( 1 - \frac{\sin \pi d}{\pi d} \right) \right] \right] \]

\[ + \frac{j_z^2 K^2 td}{4(2I+1)^2} \left( 1 - \frac{\sin \pi d}{\pi d} \right) \]  

We note that the stroboscopic technique was originally proposed in the context of mechanical harmonic oscillator \([43]\), where the conjugate quantum variables position and momentum are formally equivalent to \( \hat{J}_x \) and \( \hat{J}_y \) for constant \( J_z \) (Eq. (6.59) and (6.60) are identical to the equations of motion for a damped harmonic oscillator).

### 6.5 Experimental Measurements

Having developed a simple theory for the quantum measurement, we proceed to an experimental study of spin noise and demonstration of back-action evasion by using stroboscopic light. The work presented in the following sections was adapted from \([190]\).

The experiment is designed so that the spin projection noise is a significant fraction of the photon shot noise for the bandwidth of detection. Dropping numerical factors \(36\) Notice that the average photon flux appears in the denominator of photon shot noise term; this is due to the chosen normalization.
and physical constants, the scaling of ASN variance and PSN variance of the light optical rotation with respect to experimental parameters (atomic density \( n \), probe detuning from the optical resonance \( \Delta \nu \), length of ensemble \( l \), area of atom-light interaction \( A_b \)) is:

\[
\delta \phi_{ASN}^2 \propto \frac{nl}{A_b \Delta \nu^2} \quad (6.108)
\]

\[
\delta \phi_{PSN}^2 \propto \frac{1}{\eta \Phi} \quad (6.109)
\]

where we assumed that \( \Delta \nu \) is much larger than the optical resonance linewidth. The ASN to PSN variance ratio (for large enough detection bandwidth) increases as the probe beam area \( A_b \) becomes smaller\(^{37}\) so that a small beam size is preferable for the study of spin noise; a small sized probe beam has the additional advantage of reduced sensitivity to classical magnetic field noise. For the purposes of this experiment the wavelength of light should be tuned away from resonance so that longitudinal relaxation rate (mainly determined by the probe scattering) is small during the time of measurement (\( \sim 10 \) msec); note however that a large detuning compromises the ASN variance, see Eq. (6.108)

We stress that the ability to measure spin projection noise beyond the photon shot noise implies that squeezing can be performed. However, it is beyond the scope of this work to experimentally demonstrate large degrees of squeezing.

### 6.6 Experimental Implementation

A schematic of the experimental setup is illustrated in Fig. 6.3 and a photo is shown in Fig. 6.4.

The atomic vapor is contained in a D-shaped (truncated cylinder) glass cell, made of aluminosilicate glass (Corning 1720 and 1723), which exhibits reduced discoloring

\(^{37}\)This is due to the fact that the measurement strength increases for small probe area.
from exposure to alkali-metals at elevated temperatures. Ideally, the glass darkening should be minimal at the probe beam entry and exit surfaces of the cell and at the pump entry surface\textsuperscript{38}. The lack of availability of flat Corning 1720 surfaces led us to use Corning 1723 (which is more susceptible -rather significantly- to alkali-metal discoloring) for the probe front and back side and entry pump surface. We note that flat surfaces significantly simplify the alignment, and make the setup more robust to perturbations and drifts (see Section 4.1.5 for implications related to the polarization measurement in a curved surface).

The cell is orientated in such a way that the probe beam goes through the axis of the cylinder ($x$ axis), which is 55 mm long, limited in practice by the magnetic field gradients. We use a mixture of potassium in natural abundance, 50 Torr of N\textsubscript{2} buffer gas for suppressing radiation trapping and 400 Torr of $^4$He to slow down the

\textsuperscript{38}In fact, assuming that there are no power limitations, only the probe beam exit surface of the cell affects the quantum measurement.
Figure 6.4: Different views of the experiment.

diffusion of alkali atoms; see Fig. 6.5 for an absorption measurement). Potassium was chosen because of the light source availability in the lab; furthermore, among the alkali-metals, K experiences the lowest spin destruction rates from alkali-alkali or alkali-buffer gas collisions. The $^4$He pressure is a compromise between the requirements for low relaxation and large measurement strength given the conditions of the experiment; the measurement strength (Eq. (6.92)) becomes smaller as the pressure broadening of the optical linewidth increases, while relaxation due to thermal motion in and out of the beam and to the walls of the cell is larger for low pressures. The buffer gas pressure resulted in an optical absorption linewidth HWHM $\Gamma \approx 3.6$ GHz, see Fig. 6.5.

The cell is heated in a double-walled oven made of NEMA Grade G-7 by flowing hot air between the walls that surround the cell. Polyimide foam thermally insulates the oven, reducing the heating losses and the effect of environmental thermal perturbations. The air is heated with a resistive heater at a location away from cell (outside the shields) so that the heater magnetic field does not affect the atoms. A PID feed-

\footnote{The buffer gas pressures were estimated from the filling process and confirmed by the optical absorption measurement. The reported values here correspond to the room temperature.}

\footnote{However, in general, practical issues indicate that heavier alkali metals (i.e. Rubidium or Caesium) are preferable; high densities of Rubidium and Caesium can be realized at relatively low temperatures (compared to K), where the glass surface is not discolored by the alkali metal and optical coating is easier to apply.}

\footnote{For buffer gas densities $< 1$ amagat, spin relaxation due to collisions of alkali atoms with He can be neglected.}
Figure 6.5: Optical absorption spectrum measured at low temperature (≈ 110°C). The data acquisition and fitting procedure was described in Section 4.6.1. The measured linewidth (HWHM) is $\Gamma \approx 3.6$ GHz, which agrees well with the pressure values estimated from the filling process.

back controller adjusts the heating power so as to maintain constant temperature at the output of the heater, and a flow regulator (placed before the heater) keeps the air flow steady. By measuring the temperature inside the oven (while data were not taken), it was found that the implemented heating scheme provided temperature stability better than 0.1°C (this corresponds to < 0.5% fluctuations in K density for temperatures larger than 160°C) over the timescale of the measurements (on the order of an hour). The dimension of the inner oven was designed so that the cell is firmly rested on a polyimide rubber piece added inside the oven. To moderate condensation of K droplets on the surfaces through which light is traveling, a small opening at the oven keeps the stem of the cell at a slightly lower temperature; furthermore, cooling water is run overnight through a teflon tube that surrounds the stem cell.

The oven is rigidly held (with plastic screws) inside a NEMA grade G-10 tube, around which magnetic field and gradient coils are wrapped. Cosine winding is used to produce homogeneous magnetic fields in the transverse direction. A double layer high

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42Unless appropriately filtered, temperature probes (in fact any wire) can act as antennas allowing noisy RF fields to leak through the shields.
µ-metal and a single layer (the inner most) aluminum shield surround the assembly. We note that at RF frequencies the shielding is mainly provided by the losses at the conduction electrons, so that materials with high conductivity (e.g. aluminum or copper) provide better magnetic shielding \([147]\); the use of high µ-metal shields suppress low frequency magnetic fields that may affect the atoms adiabatically. As in the comagnetometer case, the structure is rigidly held together with a Ti frame and radial Ti rods which penetrate the shields and support the G-10 tube. The Ti body is electrically isolated from the shields in order to avoid noise-generating thermocouple and galvanic junctions, and prevent currents from the Ti inductive loops to flow to the shields. A single-point grounding scheme for each of the shield layers was used.\(^{43}\) The end caps (bases of the cylinder) of the aluminum shield are tightly screwed to the main body of the tube, and a conductive jacket provides electrical continuity across the joints. In the course of the experiment, an additional layer of copper metal shield was added to enhance magnetic shielding. The thickness of the shield was 0.01 inches, corresponding to \(\approx 1.5\) times the skin depth at frequencies around 150 kHz \([147]\).

The probe light travels through the oven and layers of magnetic shielding inside evacuated glass tubes (pressure < 1 Torr), which isolate the corresponding optical path from the turbulent air and acoustic waves. Similarly, the pump beam travels through the oven inside a vacuum tube. We note that in the experiment presented here, the surfaces of the vacuum tubes did not have optical coatings and the reflection losses were significant (\(\sim 15\%\) for each tube).

A low noise current source (Lightwave LDX-3545) generates a homogeneous DC magnetic field in the z-direction, corresponding to a Larmor frequency of 150 kHz for K atoms. First order gradients of this field along the direction of the probe beam \((dB_z/dx)\) are canceled with the use of a gradient coil.\(^{44}\) Due to the small probe beam

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\(^{43}\)In the thesis experiment, the measurement involves RF fields with wavelengths on the order of kilometers.

\(^{44}\)The gradient is adjusted by maximizing the response to a transverse RF field excitation.
size (see below) the gradient $dB_z/dy$ has a negligible effect on the measurement. In order to suppress current source noise and noise pickup of the cables, passive low pass filters are placed inside the shields. A schematic of the filtering system is shown in Fig. 6.6. Linear inductors create inhomogeneous magnetic field at the location of the atoms that has a non-negligible effect; toroidal inductors have only a small magnetic field leakage from their tori and are therefore preferred for the implementation of the filter.

\[ R_1 = 5 \, \text{k}\Omega \]
\[ R_2 = 100 \, \Omega \]
\[ L_1 = 10 \, \text{mH} \]
\[ L_2 = 1 \, \text{mH} \]
\[ C = 80 \, \mu\text{F} \]

Figure 6.6: Filter circuit diagram to reduce current source noise and noise pickup of the cables. Low pass filters are employed for the DC current fed to the $B_z$ and gradient $dB_z/dx$ coil. The $B_x$ coil is used to generate high frequency magnetic fields and noise pickup is reduced by increasing the impedance of the coil (through the $R_1$ resistors); the small $R_2$ resistance suppresses the ground loop effects.

### 6.6.1 Optical setup

Single-mode DFB lasers (GaAs semiconductor laser diode from Eagleyard) were used for the pump and probe light. These lasers feature narrow linewidth (HWHM 40
MHz), linear polarization output and with appropriate control they exhibit low intensity and wavelength noise (wavelength fluctuations are < 0.5 pm over a few hours).

**Pump beam** The pump light is steered through an optical isolator and suitable optics for mode shaping to a tapered amplifier (Sacher Lasertechnik TEC400), resulting in ≈ 500 mW power light. The amplifier output is directed to a fast, high-efficiency (∼ 85%) acousto-optic modulator (AOM), IntraAction AOM-402AF3, which allows for fast amplitude modulation of the light. The light field diffracted in the first order is used (after its polarization has been converted to circular) to pump the atoms in the z direction, while the zero order diffraction is blocked.

A combination of spherical and cylindrical lenses shapes the beam so that it illuminates all the atoms that participate in the measurement; that is, the pump light extends throughout all the cell in the direction of probe light propagation (x direction), and in the transverse y direction the pump beam size is ∼ 1 cm, more than a factor of five larger than the characteristic diffusion length during the time of the noise measurement (on the order of a few msec).

Special care is taken to shape the profile of the pump beam so that the intensity is slightly higher at the edges of cell, where the pumping requirements are stronger due to the larger spin-destruction rate from the wall relaxation. In the experiment described here, this was realized by taking advantage of the spherical aberrations of the lenses.

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45 For each of the pump and probe beam a PID controller adjusts a thermoelectric cooler (TEC) attached onto the laser diode wafer chip. For the pump laser, an additional TEC (Thorlabs laser mount TCLDM9) operated at constant current (constant cooling power) provides the power for the laser to approach the required temperature, while for the probe laser, besides the secondary TEC, water (∼ 20°C) is flown at the hot side of the second cooler (the probe laser secondary TEC is not cooling as efficiently as in the pump beam case).

46 The acousto-optic effect is the diffraction of light from sound waves [27, 161].

47 The acousto-optic modulators used in the experiment have a relatively large active aperture making the alignment easier and robust to small drifts.

48 The increased relaxation rate near the edges may also result to higher absorption if the pump intensity is not strong enough to saturate the alkali polarization to 100%.
A gradient imaging is used to evaluate the polarization non-uniformity of the vapor. A large linear magnetic field gradient \( dB_z/dx \) is superimposed to the holding field\(^{49}\), so that atoms at different parts of the cell have different resonance Larmor frequencies. A large amplitude square pulse triggered at the end of the pump period excites the atomic spins. For this measurement it is important that the atoms from all the parts of the cell (more accurately from the measurement volume) should be excited equally, that is, the Fourier transform of the transverse magnetic field pulse should be approximately constant for the frequency region spanning all the atomic Larmor frequencies. In practice, this is realized with a very short pulse (in our case the pulse duration was \( \sim 500 \) nsec). The collective spin motion is recorded and Fourier transformed. The power in every frequency component is proportional to the number of polarized atoms in the measurement volume that precess around the local magnetic field with the corresponding Larmor frequency; this way the spin polarization distribution along the probe direction can be estimated. In Fig. 6.7 the Fourier spectrum for uniform (a) and nonuniform (b) atomic polarization across the cell are shown. The red dashed line indicates the cell boundaries, found by pumping with a localized (focussed) beam; the increased power at the cell edges in Fig. 6.7(a) is due to the effect diffusion (see [151] for a discussion on the edge enhancement by diffusion in gradient imaging).

Ideally, the photon spin of the pump light should be parallel to the holding field in the measurement volume. Otherwise optical pumping, when abruptly turned off, results in a spin state that coherently precesses (even in the absence of RF transverse fields) around the magnetic field, potentially masking the quantum noise. For the case of the thesis experiment, where the longitudinal magnetic field exhibits very small distortion in the light-atom interaction region, the requirement for minimum misalignment effectively corresponds to the pump light rays propagating with small

\(^{49}\)We used a gradient of \( \sim 35 \) mG/cm.
Figure 6.7: Distribution of spin polarization across the cell as measured with a gradient imaging method. The red dashed lines denote the cell boundaries. In (a) the polarization is uniform (the increased power at the edges is due to diffusion) whereas in (b) there is a clear drop of polarization close to the cell edges. The profile in (b) was acquired at a higher atomic density compared to (a). The pumping power at high densities is not sufficient to polarize the edges of the cell.

deviation from the z direction. The optical setup was designed so that the pump beam does not expand significantly after the last lens; however, spherical aberrations cause a small misalignment of the marginal light rays from the z axis. We note that scattering at the edge of the cell or vacuum tube can also create misaligned spin state.

Due to the limited power available, we found that at high atomic densities the maximum average polarization in the measurement region (which lied \( \sim 1 \text{ cm} \) away from the pump entry cell surface) was realized with the pump light tuned off resonance. This is due to the increased optical absorption (and therefore power loss) of the pump beam in the atomic region before the measurement volume.\(^{50}\)

Overall, pump power \( \sim 120 \text{ mW} \) can be delivered to the cell. For the highest atomic polarization measurements, we use a \( \sim 10 \text{ msec} \) pumping pulse,\(^{51}\) which is turned off smoothly (decay time \( \sim 1 \text{ msec} \)) so that it does not excite significant

\(^{50}\)This behavior is predicted by the simple propagation model of Section 4.6.3.

\(^{51}\)The pumping time is determined by the pumping rate, which even at highest atomic densities of the experiment is larger than 3000 \( \text{ sec}^{-1} \), as measured by mapping the RF resonance with continuous pump light.
transverse oscillation\textsuperscript{52} (that is, the Fourier components of the pump temporal profile should have negligible power in the resonance frequency, which for the thesis experiment is 150 kHz). The applied drop-off modulation has the form \( \propto \frac{1-\tanh 4(t-t_0)}{2} \), where \( t \) is the time expressed in msec, and \( t_0 \) adjusts the phase.

**Probe beam**  The probe beam is directed to a tapered optical amplifier (Optica TA 100) for amplification. A small fraction of the power (sampled with a pellicle beam-splitter) is used in a feedback loop, where a relatively slow integral controller (timescale \( \sim 20 \) Hz) adjusts the current in the amplifier to suppress drifts in the probe power. Before entering the AOM the beam size is reduced to \(< 1 \) mm diameter, resulting in a modulation bandwidth\textsuperscript{53} \( > 2.5 \) MHz. The applied modulation is periodic with period \( T_m \) (for back-action evasion the period of modulation is half the Larmor period); and within a single modulation period can be written in the form (neglecting irrelevant time phase):

\[
\frac{1 - \tanh [50 (t - d/2)]}{2} \times \frac{1 + \tanh [50 (t + d/2)]}{2} \quad (6.111)
\]

with \( d \) being the duty cycle of the pulse.

The first order diffraction of the AOM is expanded and then mildly focused with a weak lens (the focus is located inside the cell); we stress that for the purpose of this work (study of quantum noise) it is preferable to use a small sized beam (small number of atoms). A high quality linear polarizer (Casix Glan-Laser Prism), located after the lenses and mirrors, cancels light birefringence acquired from the optical

\textsuperscript{52}In the ideal (unrealistic) case that the pump direction coincided with the longitudinal field, the rate of pump power change would not affect the atomic spins. In fact, abrupt changes in the pump power were used in order to align the pump beam with the holding field.

\textsuperscript{53}The modulation bandwidth can be expressed in the form:

\[
M_{BW} = b \frac{v}{w_b} \quad (6.110)
\]

where \( b \) is determined by the beam profile (\( b \approx 1.4 \) in the case of TEM\textsubscript{00} profile), \( w_b \) is the beam diameter and \( v \) is the acoustic velocity in the AOM medium [161].
elements, and a glass stress plate (as in the comagnetometer experiment) after the polarizer compensates to first order for any residual circular polarization of the probe beam. The stress plate is adjusted by minimizing the response to a transverse RF field when no pump light is used and the atomic polarization is created by the probe beam (for this measurement the probe beam is tuned closer to resonance than when the actual data are acquired).

Figure 6.8: Profile of the beam, measured with the knife edge scanning technique. The time derivative of the scanning output is plotted here. The red dashed line is the Gaussian fit.

The light can be directed to a beam profiler (based on knife edge scanning) for the measurement of the beam size at a distance equivalent to that of the atoms in the cell. In Fig. 6.8 the beam profile in one transverse (to light propagation) direction is plotted. Fitting a Gaussian function of the form $P = P_0 e^{\xi^2/w^2}$ (fitting parameters $P_0$ and $w$) to the knife edge scanning result, the beam size as a function of the distance from the cell was estimated to the values shown in Table 6.1. For the estimation of the spin noise, the exact beam profile was used.

---

54 The vacuum tube and the cell have some birefringence.
55 More precisely, to the time derivative of the knife edge outcome; the derivative is performed in real time with an analog differentiator.
Table 6.1: Beam size as a function of the position in the cell.

<table>
<thead>
<tr>
<th>position (cm)</th>
<th>-2</th>
<th>-1</th>
<th>0</th>
<th>1</th>
<th>2</th>
</tr>
</thead>
<tbody>
<tr>
<td>( w_y ) (µm)</td>
<td>88</td>
<td>82</td>
<td>82</td>
<td>87</td>
<td>94</td>
</tr>
<tr>
<td>( w_z ) (µm)</td>
<td>63</td>
<td>52</td>
<td>59</td>
<td>69</td>
<td>89</td>
</tr>
</tbody>
</table>

The measurements presented here were performed with the probe beam detuned from the D1 line of K by 397 GHz to the red (\( \lambda_{pr} \approx 770.890 \text{ nm} \)). For this wavelength, the tensor light-shift is less than 1% of the probe photon scattering rate and can be safely ignored\(^{56}\), therefore, the analysis of section 6.3.2 is applicable to the experiment.

### 6.6.2 Detection

The Faraday paramagnetic rotation experienced by the probe beam is measured with balanced polarimetry. Special care is taken to design a high bandwidth, low noise detection scheme. We used identical, small area PIN photodiodes (OSI Optoelectronics PIN-5D) in reverse bias (30 V) and connected to the amplifiers with very short wires. The feedback resistor and effective capacitance in the current to voltage amplifier was \( R_f = 2 \text{ kΩ} \) and \( C_f \sim 20 \text{ pF} \) resulting in an effective bandwidth \( \sim 4 \text{ MHz} \). A variable capacitor is used in one of the amplifiers to correct for small bandwidth differences between the two amplifiers\(^{57}\). The difference output in time domain is illustrated in Fig. 6.9 for a probe duty cycle of 20%.

The difference signal is digitized with a fast, low noise A/D card (NI-PCI 59222) and recorded with a computer. The function generators that control the pump and probe modulation waveforms, and the data acquisition card share the same clock in order to avoid artifacts from clock drifts and secure similar recording conditions.

\(^{56}\) As was discussed in [119], the tensor light-shift cannot be ignored even at large detunings when the excited state hyperfine structure can be clearly resolved. Here, however, the optical linewidth is significantly larger than the hyperfine splitting of the excited states and the tensor light-shift is negligible.

\(^{57}\) Bandwidth differences in the two current to voltage amplifiers are most easily observed in the shape of the difference signal.
Figure 6.9: Difference signal (blue solid line) and single photodiode output (red dashed line, multiplied by $10^{-1}$ to fit in scale) in time domain. The probe beam duty cycle was 20%.

between repeated measurements\textsuperscript{58}. The end of pumping cycle triggers (with a delay $\sim 1$ msec so that the pump intensity has fallen to zero) the recording. The Labview environment was used for the data acquisition, which also allows for real time display and evaluation of the data.

6.6.3 Atomic density estimation

To study the spin noise and compare experimental measurements with theoretical predictions the atomic density $n$ should be known.

We find $n$ by mapping the RF resonance curve and associating the measured linewidth with the spin-exchange rate between alkali atoms. For this, continuous pump and probe light at low intensity are used, so that the light contribution to linewidth is small, and a low longitudinal atomic polarization is maintained. A transverse RF magnetic field is applied at various frequencies around the Larmor frequency, and the in-phase ($y_{ip}$) and out of phase ($y_{op}$) components of the balanced polarimetry

\textsuperscript{58}This is particular important if classical systematic noise is present.
outcome, recorded with a lock-in amplifier, are fitted to the model:

\[
y_{mp}^M = A \frac{(f - f_0)}{(f - f_0)^2 + \Delta f^2} \cos \theta \\
y_{op}^M = A \frac{\Delta f}{(f - f_0)^2 + \Delta f^2} \sin \theta
\]

where \( \theta, f_0, A \) and \( \Delta f \) are allowed to vary to minimize the \( \chi^2 \) of the fit. In the above model, \( \Delta f \) is the linewidth, \( f_0 \) the Larmor frequency, \( A \) is the amplitude (which depends on the experimental conditions, i.e. magnitude of the RF field, atomic density, wavelength of the laser, length of the cell), and the parameter \( \theta \) adjusts the phase of the model.

We stress that the system of Eq. (6.112) and (6.113) describe only approximately the RF resonance curve. A more accurate theory was developed in [9], where the response to a transverse RF magnetic field is expressed in terms of the eigenvectors and eigenvalues of the relaxation operator \( \Lambda \). We performed numerical simulations and found an agreement better than 1% between the simple model (Eq. (6.112) and (6.113)) and the more complete theory of [9] for the conditions of our experiment. In particular, to a good approximation: \( \Delta f + if_0 \approx \lambda_0 \), where \( \lambda_0 \) is the eigenvalue of the relaxation matrix with the smallest real part.

The estimated linewidth is proportional to the spin-exchange rate \( R_{se} \) between alkali atoms: \( \Delta f = \kappa R_{se}/2\pi \), where \( \kappa \) depends on the density, polarization and the holding field. For the conditions of the density measurement (low longitudinal polarization, holding field resulting in \( \sim \) 85 kHz Larmor frequency and densities typically \( 3 > 10^{13} \) \( \text{cm}^{-3} \)) \( \kappa \approx 1/8 \) [99] (see Eq. (2.171)). The alkali-density is calculated from the spin-exchange rate using: \( R_{se} = n \bar{\nu} \sigma_{se} \), where \( \bar{\nu} \) is the mean velocity.
of the colliding alkali pair in the center of mass frame defined in Eq. (2.9) and
\[ \sigma_{se} \approx (1.8 \pm 0.1) \times 10^{-14} \text{ cm}^{-2} \] is the spin-exchange rate cross section. 59

6.6.4 Experimental limitations

In order to characterize the experimental constraints on the spin noise measurement, the beam was expanded to \( 2w > 3.7 \) mm and measurements of the magnetic sensitivity were performed at an alkali density \( n \approx 10^{20} \text{ cm}^{-3} \) with a 15\% duty cycle probe beam. 60 The system features a magnetic field detection limit \( > 0.4 \text{ fT}/\sqrt{\text{Hz}}, \) which is more than a factor of two larger than the spin projection limit of highly polarized atoms at the measuring conditions. 61 A preliminary investigation for the experimental, classical noise source suggests that magnetic field noise inside the shields is most likely to be the limiting factor (though further investigation is required). It is shown in Fig. 6.10 (b) that the magnetic field sensitivity has a functional dependence on \( t_d \) (time for evolution in the dark) similar to the response to white noise transverse magnetic field, but different to the response to noisy pump light. It was also found that the presence of the outermost copper shield had no effect on the sensitivity; likewise, adding white noise (more than a factor of five larger than the typical noise) to the pump and probe beam (wavelength and intensity) and to the current source driving the longitudinal field did not change the effective magnetic field sensitivity; also, larger probe beam sizes and different atomic densities demonstrated similar sensitivity.

59 The spin-exchange cross section has only a small dependence on temperature, which for the temperature range of the experiment can be neglected.

60 The measurement procedure is illustrated in Fig. 6.10 (a). After the end of the pump pulse the spins are left to evolve in the dark for time \( t_d \) and the probe beam measures the collective spin for time \( t_m \) large enough that the photon shot noise is a fraction of the spin projection noise. A known sinusoidal transverse \( B_x \) field was applied to calibrate the system. The phase of the probe beam modulation with respect to the \( B_x \) field is adjusted in order to maximize the response. The reported sensitivity is an extrapolation to zero pumping time (which in principle can be performed with very high pump power).

61 In fact, since this a projected sensitivity in the limiting case of infinite pump power and negligible pump time, the actual magnetic field sensitivity is worse than 0.4 fT/\( \sqrt{\text{Hz}} \).
Figure 6.10: Characterization of experimental magnetic field sensitivity. (a) Schematic of the measuring procedure. The atoms are polarized with a strong pump pulse, and evolve in the dark for time interval $t_d$. A probe pulse measures the collective transverse spin at the end of this interval. The pump and probe power are not plotted in scale. (b) Comparison of the atomic noise dependence on $t_d$ for the case of applied $B_x$ and pump light white noise and for typical magnetometer operation when no transverse fields are applied.

6.7 Power Spectral Density

The noise correlation function and unconditional variance can most easily be calculated from the power spectral density (PSD). Besides the simplicity in the numerical computation, the PSD provides a platform for noise estimation robust to forms of experimental noise that have spectra distinctive from quantum noise (e.g. large noise peaks resulting from electromagnetic interference to detection electronics can be easily eliminated even when appearing at a frequency within the magnetometer bandwidth).

Very efficient algorithms have been developed to estimate discrete Fourier transforms; in fact, a modern computer can Fourier process a large number of data and display the Fourier transform with small delay in real time.
The PSD frequency resolution $\delta f$ is determined by the time length of the measuring record $t_r$: $\delta f = 1/t_r$. In order to accurately estimate the noise spectrum the recording time should considerably exceed the transverse spin relaxation time $T_2$. The small beam size (diameter $\sim 160 \mu m$) results in relaxation high enough that the measurement noise spectrum can be calculated with a recording of a few milliseconds.

The power spectral density (PSD) of a 3.6 msec recording of the polarimeter output is shown in Fig. 6.11 for both unpolarized and highly polarized atoms. The

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63 This can also be seen in the time domain: for the estimation of variance uncorrelated data should be considered.

64 For unpolarized atoms the linewidth is dominated by the spin-exchange relaxation and the beam size has a small only effect; however, for highly polarized atoms diffusion through the beam is the limiting relaxation mechanism.

65 All the PSDs presented here were estimated with the algorithm provided by Labview, using a Hanning window.
resonance linewidth is significantly reduced for high spin polarization due to suppression of the spin-exchange relaxation. The conversion to (light polarization rotation) angular units was performed by dividing the recorded difference voltage signal by twice the total voltage of the photodiodes averaged over the modulation period (that is, there is no duty cycle dependence); this way, the noise reported here can be directly compared with the sensitivity realized with a continuous probe beam of the same average power (and therefore identical spin relaxation). As discussed in Section 6.3.2 the PSD can be described by the sum of a constant photon shot noise (PSN) background and a Lorentzian-like atomic noise contribution. The deviation from the Lorentzian profile is notable in our experiment due to the effect of diffusion in and out of the probe beam.

We stress that the longitudinal spin polarization does not change significantly on the time scale of the PSD recording (∼ 5 msec). The longitudinal spin coherence was measured to be ∼ 120 msec for the probe beam characteristics of this thesis, so that during the PSD recording on average only a small fraction (∼ 4%) of spins has decayed.

To account for technical imperfections that generate false signal, we take differences between consecutive recordings (with appropriate normalization). The cancellation of the classical, experimental artifacts only applies to systematic signal sources producing identical effects between successive recordings (e.g. environmental magnetic field noise cannot be suppressed by taking differences between recordings).

\[ S = \sum_{i=1}^{N-1} \left( S(\phi_{i+1} - \phi_i) \right) \]  

For this measurement a large \( B_z \) magnetic field is superimposed to the \( B_z \), so that the holding field and consequently the atomic spin (after transverse transients have decayed) have a large component in the direction of the probe beam propagation.

\[ \text{For instance, atomic excitation due to pump-longitudinal field misalignment.} \]

\[ \text{For } N \text{ series of recording } \phi_i, i = 1, 2, \ldots N, \text{ the reported spectrum is:} \]

\[ \frac{1}{2(N-1)} \sum_{i=1}^{N-1} [S(\phi_{i+1} - \phi_i)] \]  

where \( S \) denotes the PSD.
high repetition pump-measurement cycle (100 Hz for the data presented here) ensures small drift in the parameters that determine the classical signal.

The total noise var $[\phi]$ is given by the area under the PSD curve. The constant PSN background is estimated from frequencies well beyond the magnetometer bandwidth by fitting a straight line without slope; frequencies in the 20-60 kHz and 240-280 kHz region, symmetric with respect to Larmor frequency to avoid biases, were used. The spin-related variance is evaluated from numerical integration (using the Simpson method) of the measured PSD after subtracting the constant PSN background. The frequency range of integration is a compromise between the requirements for low statistical uncertainty and accurate estimation of the noise. The variance of the PSD at a specific frequency is equal to the square of the its expectation value at that frequency $[150]$; for a PSD of the form: $P(f) = g_A(f) + c_P$, $g_A(f)$ and $c_P$ are the atomic and photon noise contribution respectively, the ratio of the atomic noise integration over some frequency region to the statistical uncertainty (standard deviation) of this estimation is ($\delta f$ is the bin size):

$$\frac{\sum_k g_A(k\delta f)\delta f}{\sqrt{\sum_k (g_A(k\delta f) + c_P)^2 \delta f^2}}$$ (6.116)

For frequencies with $g_A(f) \ll c_P$, the uncertainty (denominator in Eq. (6.116)) is increasing faster than the signal (numerator in Eq. (6.116)). The range of numerical integration should also be large enough so that only a small fraction of the spin noise is outside the estimation bandwidth$^{69}$. We typically use the frequency range of 60-240 kHz for the estimation of atom noise.

$^{69}$For a Lorentzian PSD profile a frequency range 15 times the linewidth contains $\approx 96\%$ of the total noise.
6.8 Theoretical derivation of noise

The measured spectrum can be derived from a first-principle calculation. The spectrum is the Fourier transform of the correlation function: \( C_\phi(t, t') = \langle V(t)V(t') \rangle / 4\mathcal{V}_0^2 \), where \( V(t) \) is the difference voltage signal from the balanced polarimetry and \( \mathcal{V}_0 \) is the sum of the two photodiode outputs averaged over the modulation period. To account for spatial inhomogeneities we partition the photodetector and the profile of the atomic ensemble orthogonal to probe beam propagation into small segments of area \( \Delta y_i \Delta z_i \). Using Eq. (2.84), the correlation function \( C_\phi \) can be written in the form:

\[
C_\phi(t, t') = \sum_{i,j} \Delta V_i(t, z_i, y_i) \Delta V_j(t', z_j, y_j) = \frac{q_e R_f \tilde{\mathcal{V}}_0(t)}{4\mathcal{V}_0^2} \delta(t - t') + \left[ \sum_{i,j} \langle \hat{J}_x(t, z_i, y_i)\hat{J}_x(t', z_j, y_j) \rangle \left( r_e c \tilde{f}_{osc} \tilde{D}(\nu) \right)^2 \right] \times 4\mathcal{V}_0(t)\mathcal{V}_0(t') h(z_i, y_i) h(z_j, y_j) \Delta z_i \Delta y_i \Delta z_j \Delta y_j
\]

(6.120)

In the above equations \( \Delta V_i(t, z_i, y_i) \) is the voltage difference contribution to the balanced polarimetry measurement at time \( t \) of the \( i \) segment characterized by the coordinates \((z_i, y_i)\), \( \hat{J}_x \) is the segment collective electron spin (the fraction that participates in Zeeman transitions) in the probe beam propagation direction, \( q_e \) is

\(^{70}\)This section is closely related to Section 6.4. Here, we estimate the variance with respect to experimentally accessible variables, taking into account the probe beam intensity profile.

\(^{71}\)The stroboscopic modulation of light defines a preferred origin of time (a preferred phase). A correlation appropriate for the estimation of the Fourier spectrum can be defined more appropriately as:

\[
C_\phi(\tau) = \lim_{t \to \infty} \frac{1}{8\mathcal{V}_0^2 t} \int_{-t}^{t} d\xi \langle V(\xi)V(\xi + \tau) \rangle
\]

(6.117)

\(^{72}\)To derive the Eq. (6.120) the optical rotation angle \( \phi_{FR} \) from a segment was written in the form (see Eq. (2.84), (3.110)):

\[
\phi_{FR} = \frac{r_e c}{A_b} \left[ D^1_{osc} D(\nu) \right]_{D1} - \frac{1}{2} \left[ D^2_{osc} D(\nu) \right]_{D2} \langle \hat{J}_x \rangle,
\]

(6.118)

\(A_b\) being the area of the segment.
the electron charge (absolute value), $r_e$ the electron radius, $c$ the speed of light,

$$f_{osc} \tilde{D} (\nu) = f_{osc}^{D1} (\nu) \bigg|_{D1} - \frac{1}{2} f_{osc}^{D2} (\nu) \bigg|_{D2}$$

is the product of oscillator strength times the detuning taking into account both the D1 and D2 lines, $h(z,y)$ describes the power distribution of the probe beam in the plane orthogonal to the propagation axis, normalized so that $\int_{-\infty}^{\infty} dz dy h(z,y) = 1$, and $\tilde{V}_0(t)$ is the sum of the voltages recorded by two photodetectors at time $t$, proportional to the probe photon flux $\Phi(t)$:

$$\tilde{V}_0(t) = \frac{\Phi(t)}{\eta q_e R_f} \tag{6.121}$$

with $\eta$ being the detection efficiency. In the experiment the time dependence of the probe photon flux can be approximated with the pulsed function:

$$\Phi(t) = \frac{\tilde{\Phi}}{d} g(t) \tag{6.122}$$

$$g(t) = \begin{cases} 
1, & kT_m - dT_m/2 \leq t < kT_m + dT_m/2 \\
0, & kT_m + dT_m/2 \leq t < (k+1)T_m - dT_m/2 
\end{cases} \tag{6.123}$$

where $T_m$ is the period of modulation, $d$ is the duty cycle, $k$ is an integer and $\tilde{\Phi}$ is the average photon flux over the modulation period. For the voltage $\tilde{V}_0(t)$ we get:

$$\tilde{V}_0(t) = \frac{\nu_0}{d} g(t) \tag{6.124}$$

For an infinite bandwidth of detection, the total variance can be estimated by setting $t = t'$ in Eq. (6.120). However, here we are interested in the noise power that lies in the vicinity of the Larmor precession frequency.
Spatial diffusion of atoms creates correlations between the different segments and complicates the analysis significantly. A considerable simplification that leads to an analytic solution is to assume no atomic diffusion; in this case different segments remain uncorrelated and the simple model of spin dynamics described in Section 6.3.2 is valid. Combining the results of Section 6.3.2 applied to the single atom case with Eq. (6.120)-(6.124) and taking the limit of infinitesimal segments, after a bit of algebra it can be shown:

\[ C_\phi(t, t') = C_P^\phi(t, t') + C_A^\phi(t, t') + C_L^\phi(t, t') \]  

\[ C_P^\phi(t, t') = \frac{q_e R_f}{4dV_0} g(t) \delta(t - t') \]  

\[ C_A^\phi(t, t') = \frac{g(t)g(t')}{d^2} \frac{nl^2 c^2 \left[ f \tilde{D}(\nu) \right]^2}{\left( \int_{-\infty}^{\infty} h(z, y) dz dy \right)^2} (\delta j_{x,c})^2 \times \cos [\omega_0 (t - t')] \int_{-\infty}^{\infty} dz dy h(z, y)^2 e^{-r(z,y)|t-t'|} \]  

\[ C_L^\phi(t, t') = \frac{g(t)g(t')}{d^2} \frac{r_4^4 c^4 \left[ f \tilde{D}(\nu) \right]^4}{(2I + 1)^2 \left( \int_{-\infty}^{\infty} h(z, y) dz dy \right)^3} \times \left\{ \int_{-\infty}^{\infty} dz dy h(z, y)^3 e^{-r(z,y)(t+t')} \times \int_0^{\min(t, t')} ds e^{2\nu(z,y)s} \Phi(s) \sin [\omega_0(t - s)] \sin [\omega_0(t' - s)] \right\} \]  

where \( I \) is the nuclear spin, \( j_z \) the single atom longitudinal electron spin (for fully polarized atoms \( j_z = 1/2 \)), \((\delta j_{x,c})^2\) is the single atom transverse variance in the coherent state and can be found from Eq. (6.49) by setting \( N_{at} = 1 \), \( n \) is the density of

\(^{73}\)It is interesting to note that the outcome of detection (more accurately, the information extracted from the balanced polarimetry measurement) can be used to create conditional correlations between the segments.

\(^{74}\)To the best of our knowledge there are no analytical solutions; see however [203, 118] for attempts to describe mathematically the effect of diffusion.

\(^{75}\)We note that the noise terms \( \hat{b}_x \), \( \hat{b}_y \) and \( \hat{b}_1 \) appearing in Eq. (6.68) and (6.61) at different segments are uncorrelated (e.g. \( \langle \hat{b}_x(r_\perp) \hat{b}_y(r'_\perp) \rangle = 0 \) for \( r_\perp \neq r'_\perp \)). Likewise, \( \langle \hat{\Xi}_2(r_\perp) \hat{\Xi}_2(r'_\perp) \rangle = 0 \) and for \( r_\perp \neq r'_\perp \).
the atoms, $l$ the length of the cell, and $r$ is the relaxation rate written as a function of position to account for the position dependent relaxation due to probe photon absorption. We stress that $r$ is the average spin relaxation rate over the probe modulation period and Eq. (6.127)-(6.128) are correct when the fraction of spins that have decayed in a Larmor period is small.

**Photon shot noise** The photon shot noise contribution to the spectrum is ($\mathcal{F}$ denotes the Fourier transform operator):

$$\mathcal{F} [C^P_\phi (t,t')] (\omega) = \frac{q_e R_f}{4dV_0} \frac{1}{2\pi} \frac{1}{2t} \int_{-t}^{t} \int_{-\infty}^{\infty} d\tau g(t) \delta(\tau) e^{-i\omega \tau}$$

$$= \frac{q_e R_f}{8\pi V_0}, \quad \omega \in (-\infty, \infty) \quad (6.129)$$

or expressed in the positive frequency (Hz) space:

$$W^P_\phi (f) = \frac{q_e R_f}{2V_0}$$

(6.131)

Clearly, the pulsing of the probe beam does not change the photon shot noise spectrum compared to the continuous light of the same average power.

**Spin projection noise** The atom-shot noise spectrum can be calculated most easily from the convolution: $\mathcal{F} [g(t)g(t')/d^2] * \mathcal{F} [e^{-r|t-t'|} \cos \omega_0 (t - t')]$. For the Fourier transform of the probe power temporal profile we have:

$$\mathcal{F} [g/d](\omega) = \frac{1}{2\pi} \sum_k \int_{kT_m-dT_m/2}^{kT_m+dT_m/2} dt e^{-\omega t}$$

$$= \frac{\sin \omega T_m d/2}{\omega T_m d/2} \sum_k \delta \left( \omega - \frac{2\pi k}{T_m} \right)$$

(6.132)

(6.133)
from which we get:

\[
\mathcal{F}[g(t)g(t')/d^2] = \mathcal{F}[g(t)/d] * \mathcal{F}[g(t)/d]
\]

\[
= \sum_k \left( \frac{\sin \omega T_m d/2}{\omega T_m d/2} \right)^2 \delta \left( \omega - \frac{2\pi k}{T_m} \right)
\]  

(6.134)

The Fourier transform of the spin correlation function is:

\[
\mathcal{F}[e^{-r|\tau|} \cos(\omega_0 \tau)](\omega) = \frac{r}{2\pi} \left[ \frac{1}{(\omega - \omega_0)^2 + r^2} + \frac{1}{(\omega + \omega_0)^2 + r^2} \right]
\]  

(6.135)

The convolution of (6.135) with (6.136) for strobing frequency close to twice the Larmor frequency \((\omega_0 = 2\omega_0 + \chi, \chi \ll \omega_0)\) results in a spectrum with power in the vicinity of integer multiples of \(\omega_0 (\pm \omega, \pm 3\omega \ldots)\). The spectrum in the region around Larmor frequency has contributions from the \(k = 0, \pm 1\) terms of Eq. (6.135) and can be written as:

\[
\mathcal{F} * \mathcal{F} = \frac{r}{2\pi} \left[ \frac{1}{(\omega - \omega_0)^2 + r^2} + \frac{1}{(\omega + \omega_0)^2 + r^2} \right]
\]

\[+ \left( \frac{\sin \pi d}{\pi d} \right)^2 \frac{r}{2\pi} \left[ \frac{1}{(\omega - \omega_0 - \chi)^2 + r^2} + \frac{1}{(\omega + \omega_0 + \chi)^2 + r^2} \right]
\]  

(6.137)

The integrated noise is independent of relaxation and equal to: \(1 + \left[ \frac{\sin \pi d}{\pi d} \right]^2\).

We then find that the spin projection noise (variance) (for a bandwidth much larger than the maximum relaxation rate, but small enough so that the effect of noise appearing around \(\pm 3\omega\) is negligible) is:

\[
\delta \phi_A^2 = nlr_e^2 e^2 \left[ \tilde{f} \hat{D}(\nu) \right]^2 \frac{\int_{-\infty}^{\infty} h(z,y)^2 dz dy}{\left( \int_{-\infty}^{\infty} h(z,y) dz dy \right)^2} \left[ 1 + \left( \frac{\sin \pi d}{\pi d} \right)^2 \right] (\delta j_{x,c})^2
\]  

(6.138)

As discussed in Section 6.2.3, atomic diffusion changes the shape of the spectrum, but does not affect the variance. Likewise, spin decay dynamics more compli-
cated than the simple model considered here do not modify the variance. Therefore, Eq. (6.138) is directly applicable to the experiment of the thesis.

The nonuniform beam size can also be taken into account by slicing the ensemble along the axis of probe beam propagation into segments of approximately constant beam dimension (so that Eq. (6.138) –with \( l \) corresponding to the length of the segment– can be used), and summing over the variances of all the segments.

It is interesting to note that probing with stroboscopic light results in an effective increase of the measured spin projection noise by a factor \( 1 + \left( \frac{\sin \pi d}{\pi d} \right)^2 \). This increase in noise compared to the continuous probe beam does not compromise sensitivity of the magnetometer, even in the absence of light-shift. The signal from sinusoidally oscillating atomic spins monitored with stroboscopic light modulated at twice the oscillation frequency (and appropriately phase adjusted so that the light is on symmetrically around the sine wave peaks) is (see Section 6.12):

\[
\frac{1}{2} \left( 1 + \frac{\sin \pi d}{\pi d} \right)
\]

that is, a factor of \( 1 + \frac{\sin \pi d}{\pi d} \) larger than the continuous probe case. Clearly, \( (1 + \frac{\sin \pi d}{\pi d})^2 \geq 1 + \left( \frac{\sin \pi d}{\pi d} \right)^2 \), with the equality holding only for \( d = 1 \).

**Back-action noise** A similar analysis can be performed for the light-shift (back-action) noise. As in the case of spin-projection noise, the effect of the \( g(t)g(t')/d^2 \) factor in Eq. (6.128) is equivalent to multiplying the spectrum (in the region around the Larmor frequency) with \( 1 + \left( \frac{\sin \pi d}{\pi d} \right)^2 \). After a bit of algebra, one can show that in the limit of \( r \ll f_0 \) and \( f_m = 2f_0 \), where \( f_0 \) and \( f_m \) are the Larmor and modulation frequencies respectively, the light-shift contribution to the measured variance is (to
lowest order in $r/f_0$ and assuming no diffusion and 100% longitudinal polarization):

$$
\delta \phi^2_L (d) = A \left(1 - \frac{\sin \pi d}{\pi d}\right) \int_{-\infty}^{\infty} dz dy \frac{h(z, y)^3}{4r(z, y)} \\
A = \left[1 + \left(\frac{\sin \pi d}{\pi d}\right)^2\right] \frac{r^4 e^4 [\bar{f} \bar{D}(\nu)]^4 n^2 l^2}{(2I + 1)^2 \left(\int_{-\infty}^{\infty} h(z, y) dz dy\right)^3} \bar{\Phi} \tag{6.140}
$$

For small deviation of $f_m$ from $2f_0$ ($|f_m - 2f_0| \ll f_0$) and small duty cycle, it can be shown that to lowest order in $|f_m - 2f_0|/f_0$ and $d$:

$$
\delta \phi^2_L (f_m) = \frac{A}{4} \int_{-\infty}^{\infty} dz dy h(z, y)^3 \left[\frac{1}{r(z, y)} - \frac{r(z, y)}{(f_m - 2f_0)^2 + \pi^2 r(z, y)^2}\right] \tag{6.142}
$$

We stress that in the above derivation of light-shift noise, the effect of diffusion was neglected. Therefore, deviations of the experimentally measured light-shift noise from the predictions of Eq. (6.140)–(6.142) are expected; however, Eq. (6.140)–(6.142) capture the general dependence of light-shift noise on duty cycle and probe modulation frequency.

### 6.9 Spin projection noise for unpolarized atoms

The prediction of Eq. (6.138) and (6.49) for unpolarized atoms\textsuperscript{76} ($\langle \delta j_{x,c} \rangle^2 = 3/32$) was compared with the experimentally measured noise area; an agreement of better than 10% was demonstrated for various conditions of duty cycle, beam size and density.

Figure 6.12 illustrates this agreement. In Fig. 6.12 (a) the spin noise variance calculated from numerical integration of the spectrum and the prediction of Eq. (6.138) with no free parameters (all parameters are experimentally determined) are plotted.

\textsuperscript{76}From Eq. (6.49) the single atom spin variance is:

$$
\langle \delta j_{x,c} \rangle^2 = \frac{3 + 4I(I + 1)}{12(2I + 1)^2} \tag{6.143}
$$

where $I$ is the nuclear spin.
Figure 6.12: (a) (originally appeared in ICAP 2010) Spin projection noise for unpolarized atoms as a function of alkali density. Data were acquired with continuous probe beam. The measurement of atomic density is limited by the uncertainty in the spin-exchange cross section (\(\sim 5\%\)). The theoretical prediction for the noise is described by Eq. (6.138). (b) Dependence of spin projection noise for unpolarized atoms on the duty cycle of the probe beam. The values have been normalized to the noise for continuous probe.

as a function of the atomic density (continuous probe light was used). The accuracy of the first principle estimation of the variance is limited by the uncertainty in the determination of atomic density \(n\). Figure 6.12(b) shows that the normalized (to 100% duty cycle) spin noise dependence on the probe beam duty cycle can be described well with the function: \(1 + \left(\frac{\sin \pi d}{\pi d}\right)^2\).

Having demonstrated the validity of Eq. (6.138), we note that the spin projection noise can potentially be used to measure the density of atoms; all the parameters appearing in Eq. (6.138) (except for \(n\)) can be readily found with small uncertainties. This method could prove particularly valuable at high densities.

The noise area for unpolarized atoms is a good measure of fundamental atomic shot noise (ASN), since it is not affected by light-shift or stray magnetic field noise, and the scattering of photons has an insignificant effect on the quantum noise properties [119].
It therefore offers an excellent, simple and robust reference for the characterization of the atomic spin noise with polarized atoms, unambiguously calibrating the system.

## 6.10 Spin projection noise dependence on polarization

In Fig. 6.13 the spin noise ratio for (partially) polarized to unpolarized atomic ensembles is plotted as a function of the longitudinal polarization averaged over the measuring time for three different alkali densities and 10% probe beam duty cycle.

To find the longitudinal polarization a small dc magnetic field in the probe direction is applied ($B_x \ll B_z$ -typically $B_x/B_z \sim 0.2\%$); this way optical pumping creates a small DC polarization in the $x$ axis given by (after transients have decayed): $P_x \approx P_z B_x/B_z$, where $P_z$ is the longitudinal polarization to be measured.\(^{77}\) We note that the polarization is not constant, but decays during the measurement time. In the following we suppress the time dependence, though it is understood that the relevant quantities evolve in time. The optical rotation $\phi_B$ induced in the probe beam due to $P_x$ is used to estimate the polarization from the following equation (see Eq. (2.85)):

$$P_z = \frac{2\phi_B B_z}{n l r_e f_{osc} D(\nu) B_x}$$ \hspace{1cm} (6.144)

In the actual experiment, the balanced polarimetry outcome cannot be directly applied to (6.144); drifts in the incident light linear polarization or uncontrolled environmental magnetic field in the $x$ axis may result to a finite polarimetry signal (offset) even in the absence of the (known) $B_x$ field. In order to discriminate $\phi_B$ from the offset, the measurement is repeated with a $-B_x$ field, so that: $\phi_B = (\phi_1 - \phi_2)/2$,

---

\(^{77}\)The small magnetic field $B_x$ has a negligible effect on the optical pumping dynamics, and the longitudinal polarization is not altered within the precision of the measurement by the small pump-holding field misalignment.
where $\phi_1$ and $\phi_2$ are the polarimetry outcomes of the first and second measurement respectively. The error in the atomic density (found by mapping the coherent RF resonance) limits the accuracy of the longitudinal polarization\textsuperscript{78} (relative uncertainty $\sim 5\%$).

The ensemble polarization at large polarization values can also be estimated from the transverse relaxation rate \textsuperscript{[163]}. For K atoms, $P_z$ close to unity and intermediate longitudinal magnetic field (the case of the thesis work), the polarization satisfies the following equation (see Eq. \textsuperscript{(2.172)}):

$$P_z = 1 - 5 \frac{R - R_{sd}/4}{R_{se}}$$

(6.145)

where $R_{sd}$ and $R$ are respectively the longitudinal and transverse spin destruction rates. The spin exchange rate $R_{se}$ can be directly estimated from the coherent RF curve, while $R_{sd}$ is found as described in footnote\textsuperscript{66}. To evaluate $R$, a short RF pulse at the end of the pumping interval excites spin oscillation, and the decaying precession is recorded\textsuperscript{70}; using the peaks of the sinusoidal signal, the spin decoherence rate is numerically evaluated at discrete times. This method of polarization estimation is not limited by the accuracy with which spin-exchange cross section is known; however, it assumes homogenous atomic polarization distribution throughout the measurement volume, a condition that is not satisfied at high densities.

The two polarization measurements give similar results for low atomic density, but differ by 10% at the highest density. We believe this discrepancy results from the nonuniform polarization profile of the atomic ensemble, which becomes more pronounced at high densities due to the limited pumping power (see Fig. \textsuperscript{6.7}).

\textsuperscript{78}Each reported value of polarization is the average over $> 10^4$ measurements, and the statistical uncertainty is small.

\textsuperscript{70}For the polarization estimation a continuous probe beam was used with the same average power as the stroboscopic light that was employed for the spin noise evaluation.
Figure 6.13: (Taken from [190]) Ratio of polarized to unpolarized ASN variance as a function of the average longitudinal polarization of the ensemble for three different densities. The theoretical model is described by Eq. (6.49). The probe beam duty cycle was 10%, and the error bars represent statistical uncertainty.

The measured noise ratio is well described by the simple theoretical model developed in Section 6.2.3. To the best of our knowledge, these data (originally appeared in [190]) represent the first systematic experimental study of collective spin measurements on partially polarized atomic states (see [17] for a theoretical discussion).

6.11 Back-action evasion

The agreement between the measured spin noise for polarized atoms with the theoretical spin projection prediction manifests the back-action evasion of the probe. Here, the back-action evasion of the stroboscopic measurement is directly demonstrated by considering the noise dependence on the duty cycle and the frequency of probe modulation.
As was discussed in Section 6.8, the back-action of the probe on the balanced polarimetry output is suppressed for small duty cycle \( d \) of the stroboscopic light, and increases with \( d \) reaching the maximum value at \( d = 1 \) (continuous probe beam). In Fig. 6.14 the atom noise is plotted as a function of the duty cycle of the stroboscopic probe. Each point has been normalized with the corresponding unpolarized ASN (for the particular duty cycle) to cancel the \( 1 + \left( \frac{\sin \pi d}{\pi d} \right)^2 \) factor that originates solely from the pulsed detection (it is not related to the spin dynamics). If the effect of diffusion on the variance can be neglected, the normalized variance \( (\delta \tilde{\phi})^2 \) is:

\[
(\delta \tilde{\phi})^2 = \kappa(P_z) + \left( 1 - \frac{\sin \pi d}{\pi d} \right) \zeta \quad (6.146)
\]

where \( \kappa(P_z) \) is the ratio of polarized to unpolarized spin projection noise variance that is determined by the longitudinal polarization \( P_z \) (\( \kappa = 2/3 \) for \( P_z = 100\% \)) and \( \zeta \) is independent of the duty cycle. It can be seen in Fig. 6.14 that the experimental data satisfy Eq. (6.146) to a good approximation.

The back-action evasion of the stroboscopic measurement is also demonstrated in Fig. 6.15, where the measured atom noise is plotted as a function of the frequency of probe modulation. The contribution of spin projection noise to the measurement (see Eq. (6.138)) does not depend on the stroboscopic frequency \( f_m \), and remains at the value of the ASN. This is manifested in the total noise for unpolarized atoms, where there is no light-shift noise. In the case for polarized atoms, as the strobe frequency departs from the resonance condition of twice the Larmor frequency, light-shift noise is added to the ASN, and the total noise increases until it reaches a maximum plateau. The difference of the maximum and minimum values is a measure of the back-action noise of the probe. The dependence of light-shift noise variance

\[
\zeta = \frac{r^2 c^2 \left[ \int \hat{D}(\nu) n \Phi \right]^2 f_{\infty} d\nu}{(2I + 1)^2 \left[ \int_{-\infty}^{\infty} h(z, y) d\nu \right] f_{\infty} d\nu \int_{\infty}^{\infty} d\nu \int_{\infty}^{\infty} d\nu \frac{h(z,y)^3}{4s(z,y)^3}} \quad (6.147)
\]
Figure 6.14: (Taken from [190]) Ratio of polarized to unpolarized atom noise (not including PSN) as a function of the duty cycle of the stroboscopic light. All data points were acquired with the same average intensity. The solid red curve is described by Eq. (6.146) with $\kappa(P_z)$ and $\zeta$ adjusted to match the experimental values at $d \approx 0$ and $d = 1$ respectively.

The noise on the probe modulation frequency for small duty cycle is described by Eq. (6.141) and (6.142) for the simplified case of no atomic diffusion: the noise can be written as the difference of a term independent of $f_m$ and a term with a Lorentzian-like functional dependence on $f_m$ (linewidth of $2r$ in angular frequency units). Although diffusion modifies the exact line-shape and complicates the estimate of the effective relaxation rate, the area $\mathcal{J}$ defined by the inverted quasi-Lorentzian curve (shaded area in Fig. 6.15) is independent of the relaxation and is not affected by diffusion. It is straightforward to show that (for high longitudinal polarization):

$$\mathcal{J} = \frac{A}{16} \int_{-\infty}^{\infty} dz \, dy \, h(z, y)^3$$

$$= \left[ 1 + \left( \frac{\sin \pi d}{\pi d} \right)^2 \right] \frac{r_c^4 e^4 [\tilde{f} \tilde{D}(\nu)]^4 n^2 l^2 \Phi}{16 (2I + 1)^2} \frac{\int_{-\infty}^{\infty} dz \, dy \, h(z, y)^3}{\left( \int_{-\infty}^{\infty} h(z, y) \, dz \, dy \right)^3}$$

(6.148)
and \( J \) can be calculated from measured experimental parameters. An agreement of \( \sim 10\% \) was found by comparing the shaded area\(^{81}\) in Fig. 6.15 with the prediction of Eq. (6.149).

Figure 6.15: (Taken from [190]) Measurement of atomic noise (variance) for unpolarized and highly polarized (P\( \approx 85\% \)) atomic ensembles as a function of stroboscopic frequency. The Larmor frequency of the ensemble is 150kHz, and the duty cycle of the probe is 10\%. All data were acquired with the same probe intensity. For the unpolarized case the noise does not depend on the frequency, while for polarized atoms extra light shift noise appears at detunings from the resonant condition. The shaded area is independent of the spin relaxation and diffusion, and can be calculated from Eq. (6.149). A small linear drift in the noise for polarized atoms is not understood.

### 6.12 Response to a coherent RF field

The stroboscopically modulated probe beam, that has been appropriately synchronized with a coherent spin precession, strategically distributes the available power to emphasize time intervals where the spin signal is large (peaks of the oscillations).

\(^{81}\)In order to calculate the area, numerical integration with the Simpson method was performed, assuming a background plateau estimated by fitting a straight line with slope on the first four and last four plotted data (for polarized atoms).
The lock-in signal for a probe beam modulated at twice the spin precession frequency \((\omega_m = 2\omega)\) is proportional to:

\[
\frac{\omega}{2\pi} \int_0^{2\pi/\omega} dt \left( \cos \omega t \right)^2 \frac{g(t)}{d} = \frac{1}{2} \left[ 1 + \frac{\sin \pi d}{\pi d} \right]
\]

where the probe power temporal profile \(g(t)\) is described in Eq. (6.123). Figure 6.16 demonstrates the validity of Eq. (6.150).

Figure 6.16: (originally appeared in ICAP 2010) Normalized response to coherent RF excitation as a function of the probe duty cycle. The lock-in and RF-field phases with respect to the probe modulation waveform were adjusted to maximize the response.

### 6.13 QND measurement with time-varying relaxation

High density atomic systems feature large spin-exchange rates, whose contribution to spin relaxation depends on the longitudinal polarization. In a measuring scheme where the pump beam (after polarizing the atoms) is turned off (so that it does not
cause any relaxation, the initial (transverse) spin relaxation is small, but increases in time as the longitudinal polarization drops and the effect of spin-exchange to relaxation becomes more important. This is manifested by a non-exponential decay of the transverse spin polarization, shown in Fig. 6.17.

Figure 6.17: Decay of the transverse atomic spin at high density \( n \approx 6 \times 10^{13} \text{cm}^{-3} \). The spin precession is excited by a short RF pulse at the end of the pumping interval. Initially, when the longitudinal polarization is high and the spin-exchange relaxation is suppressed, the decay is slow. At later times, as \( P_z \) drops the decay rate increases.

The advantages of suppressed initial spin relaxation rate can be harnessed most efficiently in the context of quantum non demolition measurements, which preserve temporal spin correlations and increase the bandwidth of the measurement \[170\]. In particular, the squeezing realized with a QND measurement (which in general improves the sensitivity at short time scales) can improve the long-term performance in this case of time-varying relaxation rate.

\[82\] Fundamentally (that is, assuming that there are no technical limitations), the use a pulsed pump beam results in smaller effective linewidths compared to the continuous pump case.
To study this effect we consider the measuring scheme using two short pulses of probe light as illustrated in Fig. 6.18 [8]; the first pulse is applied immediately after turning off the pump beam and creates a squeezed atomic state, while the second pulse measures the squeezed distribution after evolving in the dark for time $t_m$.

For simplicity, we analyze an ensemble with spin-1/2 atoms (no nuclear spin) and neglect complications from diffusion and spatially varying probe beam. We also consider a system with no longitudinal field, which experiences spin-exchange relaxation as in the RF case [83]. It is assumed that the pumping, squeezing and measuring pulse duration is negligible, so that the measurement repetition time is $\approx t_m$; the spin relaxation and longitudinal polarization loss that occur during the two pulses are taken into account.

The spin relaxation rate is a function of time given by [84]:

$$R(t) = R_{sd} + (1 - P_z(t))R_{se}$$

(6.151)

where the end of the pump pulse was taken as $t = 0$ and $P_z(t) = e^{-R_{sd}t}$ is the longitudinal polarization.

Let $\tilde{J}_x(0)$ and $\tilde{J}_x(t_m)$ be the measurement outcome (in terms of the ensemble spin) of the first and second pulse respectively. The variance of $\tilde{J}_x$ can be written as a sum of a PSN and an ASN term (see Eq. (6.71)):

$$\text{var}[\tilde{J}_x] = \text{var}[\tilde{J}_x]_{PSN} + \text{var}[\tilde{J}_x]_{ASN}$$

(6.152)

$$\text{var}[\tilde{J}_x]_{ASN} = \frac{N_{at}}{4}$$

(6.153)

$$\text{var}[\tilde{J}_x]_{PSN} = \frac{N_{at}}{4ODN_{sc}}$$

(6.154)

[83] In fact at low longitudinal fields the spin-exchange relaxation is significantly suppressed (see Section 2.16.2). The analysis described here is appropriate for the rotating frame in RF magnetometry; or equivalently for the in-phase component of the lock-in measurement.

[84] Notice that we dropped numerical factors in the definition of the relaxation rate (this can be thought as a rescaling of $R_{se}$, which does not change the result).
Figure 6.18: Measuring scheme with squeezing. A high power pump laser fully polarizes the atomic ensemble. A short probe pulse squeezes the spin distribution and a second pulse measures the squeezed distribution after evolving in the dark for time $t_m$.

with $N_{at}$ being the number of atoms in the measurement volume, $OD = n_r c f_{osc} / \Gamma$ is the optical density on resonance and $N_{sc}$ is the product of pulse duration $\tau_p$ and photon scattering rate $R_{pr} = \Phi r_e c f_{osc} \Gamma / (A_b \Delta \nu^2)$ ($\Gamma$ is the HWHM linewidth of optical absorption, $\Delta \nu$ is the detuning from resonance and is much larger than the optical linewidth so that $D(\nu) \approx 1 / \Delta \nu$, $\Phi$ is the photon flux in photons/sec, and $A_b$ is the area of the probe beam). To derive Eq. (6.154) the PSN optical rotation variance $1/4 \Phi \tau_p$ was converted to spin variance using Eq. (6.118). In writing Eq. (6.154), the efficiency of detection was assumed to be unity. Although in principle the experimental limitations of detection can be overcome, one needs to examine whether absorption in the atomic vapor affects the result. The fraction of photons that are absorbed by the atomic
vapors is $e^{-OD(\Gamma/\Delta \nu)^2}$, so that by going to appropriately large detunings the fractional absorption can be negligible at any optical density\textsuperscript{85}.

The best estimate of the magnetic field is obtained using the conditional measurement:

$$\tilde{J}_x(t_m) - \frac{\text{cov} \left[ \tilde{J}_x(0), \tilde{J}_x(t_m) \right]}{\text{var} \left[ \tilde{J}_x(0) \right]} \tilde{J}_x(0) \quad (6.155)$$

where the covariance of the two measurements (taking into account the relaxation in the pulses) is\textsuperscript{86}:

$$\text{cov} \left[ \tilde{J}_x(0), \tilde{J}_x(t_m) \right] = \frac{N_{at}}{4} \exp \left[ - \int_{0}^{t_m} R(t') dt' \right] \quad (6.156)$$

$$= \frac{N_{at}}{4} \exp \left[ - N_{sc,1} - N_{sc,2} - \int_{0+}^{t_m-} R(t') dt' \right] \quad (6.157)$$

$$= \frac{N_{at}}{4} \exp \left[ - (N_{sc,1} + N_{sc,2}) - (R_{sd} + R_{se}) t - (e^{-R_{sd} t} - 1) e^{-N_{sc,1} R_{se}/R_{sd}} \right] \quad (6.158)$$

The corresponding conditional variance $S$ of the measurement can then be found from Eq. (6.71) and (6.158).

A transverse magnetic field $B_y$ causes the collective spin (gyromagnetic ratio $\gamma$) to evolve according to the (average) dynamics:

$$\frac{d\langle J_x(t) \rangle}{dt} = -R(t)\langle J_x(t) \rangle + \frac{N_{at}}{2} \gamma P_z(t) B_y \quad (6.159)$$

\textsuperscript{85}Note also that $N_{sc}$ can be adjusted independently of the detuning by varying the probe power or area.

\textsuperscript{86}We denote as $0+$ the time right after the end of the first probe pulse and as $t_m$—the time just before the second probe pulse.
with the initial conditions $\langle J_x(0+) \rangle = 0$, $P_z(0+) = e^{-N_{sc,1}}$. After $t_m$ the response to $B_y$ is:

$$
\langle J_x(t_m) \rangle = \frac{N_{at}}{2} \exp \left[ -\frac{R_{se}}{R_{sd}} e^{-N_{sc,1} - R_{sd} t_m} - (R_{se} + R_{sd}) t_m \right]
\times \int_0^{t_m} dt \exp \left[ -N_{sc,1} + \frac{R_{se}}{R_{sd}} e^{-N_{sc,1} - R_{sd} t} + R_{se} t \right]
$$

(6.160)

The signal to noise ratio after total measurement time $T$ (corresponding to $T/t_m$ independent estimations of the magnetic field) is:

$$
\text{SNR} = \frac{\langle J_x(t_m) \rangle}{\sqrt{S/(T/t_m)}}
$$

(6.161)

Using this model we numerically optimize (that is, maximize the SNR) the measurement procedure with respect to the strength $N_{sc}$ of the first and second probe pulses and the measurement time $t_m$. The resulting minimum sensitivity is plotted as a function of OD in Fig. 6.19 for varying spin-exchange rates. For comparison, we also plot the variance of an optimized single-pulse measurement which does not rely on the conditional evolution. The results are scaled relative to the standard quantum limit (SQL) (the variance due to spin projection noise of a continuous measurement for time much larger than the correlation time)

$$
\delta B_{SQL}^2 = \frac{2}{N_{at}}\frac{R_{sd}}{2} (N_{at} T \gamma^2)
$$

(6.164)

\footnote{For this, we consider the variance of a continuous spin measurement assuming no light-shift and photon noise. Since measurements of $J_x$ at different times are not independent (time correlation function $C(\tau) = \frac{N_{at}}{4} e^{-R_{sd} \tau}$), we write for the variance \cite{77, 103}:

$$
\text{var} \left[ \int_0^T dt J_x(t) \right] = \frac{2}{T} \int_0^T d\tau \left( 1 - \frac{\tau}{T} \right) C(\tau)
\approx \frac{N_{at}}{4} \frac{2}{R_{sd} T}, \quad R_{sd} T \gg 1
$$

(6.162)

(6.163)}
Figure 6.19: (taken from [190]) Variance in the estimate of the magnetic field relative to SQL as a function of the optical density. The red dashed lines correspond to single-pulse measurement, while the solid blue lines two pulse measurement utilizing spin-squeezing.

It is interesting to compare the results of the simple model with those of [105]. In the absence of spin-squeezing and spin-exchange collisions, the smallest possible magnetic field variance is given by $e\delta B^2_{SQL}$, in agreement with [105]; using the two-pulse measurement (with squeezing) one can reduce the variance by a factor of $e$, the same factor as obtained in [105] with partially entangled states. In the presence of spin-exchange collisions, the sensitivity in the one-pulse scheme is reduced by the spin-exchange relaxation, while in the measurement with squeezing the sensitivity is affected less by spin-exchange relaxation and asymptotically at large optical depths reaches $\delta B^2_{SQL}$ (independent of $R_{se}$). Therefore, squeezing from QND techniques can eliminate the effects of spin-exchange relaxation, but cannot significantly enhance the sensitivity limited by a constant spin relaxation rate.
We stress that the result above, is not limited to atomic magnetometry. Similar arguments also apply to hyperfine transitions (of particular importance for atomic clocks) broadened by spin-exchange collisions \[109\] and in general to relaxation effects of nonlinear interactions, such as dipolar spin couplings \[1\].
Chapter 7

Summary and Outlook

We developed an atomic K-$^3$He comagnetometer which was used in the search of new, spin-dependent, long range forces generated by a separate optically polarized $^3$He spin source equivalent to $9 \times 10^{21}$ fully polarized $^3$He atoms. Running automated routines the comagnetometer can be tuned to the compensation point, where the sensitivity to non-magnetic spin coupling fields is high, while the response to magnetic field and light-shift perturbations is strongly suppressed. We demonstrated a sensitivity of $< 1 \text{ fT} / \sqrt{\text{Hz}}$ in the frequency region of $0.1 - 1 \text{ Hz}$, a factor of 4 improvement compared to a previous implementation of the comagnetometer [TK]. Over the course of a month we measured the $^3$He spin precession frequency with a resolution of 18 pHz, corresponding to an energy resolution of $10^{-25} \text{ eV}$. At a length scale of 50 cm, the distance of the spin source to the comagnetometer, we set limits on the anomalous spin-dependent force between neutrons to be less than $2.5 \times 10^{-8}$ of their magnetic or less than $2 \times 10^{-3}$ of their gravitational interaction. We constrained the coupling strength of light pseudoscalar, vector and pseudovector particles to neutrons, providing the strongest laboratory bounds in the case of massless boson fields. Similar limits were obtained for couplings to unparticles and Goldstone bosons associated with spontaneous breaking of Lorentz symmetry down to spatial rotations.
The anomalous spin-dependent measurement can be improved by building a more compact comagnetometer apparatus, which will increase the proximity of the comagnetometer detector to the field source. In fact, a smaller comagnetometer setup has already been implemented and used in fundamental physics research \[34\]. Furthermore, tighter constraints can by imposed by polarizing larger number of atoms in the spin source; the relatively small polarization that was realized in this work indicates that with the use of a hybrid optical pumping and a higher power pump light a factor of four improvement can be readily achieved.

In this thesis we also studied a new technique for performing quantum-non demolition (QND) measurement in a Radio Frequency atomic-optical magnetometer. It was experimentally demonstrated that by using stroboscopic probe light the back-action of the probe on the observable is suppressed; the suppression is a function of the detuning of the modulation frequency from twice the Larmor frequency and the duty cycle of the probe. A simple model based on a system of first order stochastic differential equations was developed that gave good agreement with the measurements. We explored how spin projection noise depends on the polarization in the presence of various spin relaxation mechanisms, including spin-exchange relaxation, and found that the spin temperature distribution describes adequately the data. Finally, we showed theoretically that for systems with spin relaxation changing in time, QND measurements can enhance the long-term sensitivity of atomic spectroscopy.

We plan to further investigate the classical experimental noise that currently limits the sensitivity of the magnetometer; hopefully, we will be able to extend the experimental sensitivity to a level that will allow us to explore atom shot noise and squeezing for larger optical depths and interaction volumes, where diffusion relaxation is reduced. We aim to demonstrate experimentally the long-term sensitivity improvement of a QND measurement in the warm, dense atomic vapor system, and apply the stroboscopic technique to the detection of nuclear magnetic and nuclear quadrupole effects.
resonance signals. We are currently working on a multi-pass arrangement; unlike a cavity, it has a large active volume and is robust and easy to implement. With the multi-pass approach enhanced magnetic field sensitivity and stronger squeezing can be realized.
Appendix A

A.1 Irreducible Representation of the Polarizability

Here, we define the functions and tensors that appear in Eq. (2.31):

\[ \alpha = \sum_{F_g F'_g F_e L M} A^L(F_g F'_g)(-1)^M Q^L_{-M} T^L_M(F_g F'_g) \quad (A.1) \]

The coefficients \( A^L(F_g F'_g) \) are:

\[ A^L(F_g F'_g) = \sum_{F_e} Z(F_e F'_g) \xi^L(F_g F'_g; F_e) \quad (A.2) \]

and \( \xi^L \) can be expressed with the help of Wigner 6j symbols:

\[ \xi^L(F_g F'_g; F_e) = 3G(-1)^{-F_e-3F'_g-2F_g-2J_e}[S][J_e][F_e][F'_g][F_g] \]

\[ \times \begin{aligned} &1 \ 1 \ L \\ &F'_g \ F_g \ F_e \end{aligned} \begin{aligned} &J_e \ F_e \ I \\ &F'_g \ S \ 1 \end{aligned} \begin{aligned} &J_e \ F_e \ I \\ &F'_g \ S \ 1 \end{aligned} \quad (A.3) \]

\(^1\)We follow the notation of [96], except for the fact that we use the more symmetrical 6j symbols instead for the Racah W coefficients.
The factor $G$ is defined by:

$$G = \frac{e^2 f_{osc}}{2m_e \omega^2} \sqrt{\frac{M_\alpha c^2}{2k_BT}} \quad (A.4)$$

Here, $m_e$ is the electron mass, $f_{osc}$ is the oscillator strength of the transition and depends on the reduced matrix element of the transition in the uncoupled basis [50, 175]:

$$f_{osc} = \frac{2m_e \omega_{S\rightarrow J_e}}{3\hbar e^2} |\langle S||D||J_e\rangle|^2 \quad (A.5)$$

The oscillator strength is also related to the spontaneous emission rate (in free space) through the relationship:

$$\gamma_{sp} = \frac{2e^2 \omega^2[S]}{m_e c^2 |J_e|} f_{osc} \quad (A.6)$$

In Eq. (A.1) $T^L_M$ is the spherical tensor of rank L in the coupled basis:

$$T^L_M(F,F') = \sum_{mm'} |F_m\rangle\langle F'_m|C^{LM}_{FmF'm'} \quad (A.7)$$

with $C^{LM}_{FmF'm'}$ referring to the standard definition of the Clebsch-Gordan coefficient [188]. In the polarizability equation we used $Q^L_M$ for the basis dyadics which can be defined through the unit vectors $e_m$ in the spherical coordinates:

$$Q^L_M = \sum_{mm'} i_m i^*_m C^{LM}_{1m1m'} \quad (A.8)$$

### A.2 Alkali atoms with non-negligible hyperfine excited state structure

The coefficients of $A^L(F_g F'_g)$ depend on the structure of the alkali atom, the detuning and the optical linewidth. Here, we are focusing on the conditions of the experiments performed and calculate the various coefficients for $^{39}$K (nuclear spin $I = 3/2$) at relatively high buffer gas pressures. For the D1 transition ($S \rightarrow J_e = 1/2$) there are
two hyperfine manifolds for both the ground and excited state ($F_g = 1, 2$ and $F_e = 1, 2$). Using (A.3) we find the coefficients $A^L(F_gF'_g)$: [98]:

\[ A^0(11) = \frac{G}{2}[5Z(21) + Z(11)] \quad (A.9) \]

\[ A^0(22) = \frac{G\sqrt{15}}{2}[Z(22) + Z(12)] \quad (A.10) \]

\[ A^0(12) = A^0(21) = 0 \quad (A.11) \]

\[ A^1(11) = \frac{G}{4}[5Z(21) - Z(11)] \quad (A.12) \]

\[ A^1(22) = -\frac{G\sqrt{5}}{4}[3Z(12) + Z(22)] \quad (A.13) \]

\[ A^1(12) = -\frac{G\sqrt{5}}{4}[Z(12) + 3Z(22)] \quad (A.14) \]

\[ A^1(21) = \frac{G\sqrt{5}}{4}[3Z(21) + Z(11)] \quad (A.15) \]

\[ A^2(11) = \frac{G}{4}[Z(21) - Z(11)] \quad (A.16) \]

\[ A^2(22) = \frac{G\sqrt{21}}{4}[Z(12) - Z(22)] \quad (A.17) \]

\[ A^2(21) = \frac{3G}{4}[Z(21) - Z(11)] \quad (A.18) \]

\[ A^2(12) = \frac{3G}{4}[Z(12) - Z(22)] \quad (A.19) \]

From the above equations it is clear that when $Z(F_eF_g) = Z(F_g)$ for all $F_e$ then $A^2(F_gF'_g)$ is identically zero. For clarity we write again the profile function as defined in equations (2.21)-(2.23) with the assumption of negligible dependence of the transition frequency on the magnetic quantum number:

\[ Z(F_eF_g) = \frac{1}{\sqrt\pi} \int_{-\infty}^{\infty} du \frac{e^{-u^2}}{u - \zeta(F_eF_g)} \]

\[ \zeta(F_eF_g) = \frac{1}{k} \sqrt{\frac{M_a}{2k_BT}} [(\omega - \omega_{F_e:F_g}) + i\gamma_2] \quad (A.20) \]
It is more convenient for numerical calculations to express the plasma dispersion function with the help of the complementary error function \( \text{erfc} \):

\[
Z(\zeta) = i\sqrt{\pi} e^{-\zeta^2} \text{erfc}(-i\zeta)
\]  

(Figures A.1 and A.2 show the dispersive part of \( A^L(F_gF'_g) \) as a function of light frequency for the conditions pertinent to the two experiments of the thesis; Fig. A.1 refers to buffer gas pressure 12 amagat of \(^3\)He and Fig. A.2 refers to 50 Torr of \( \text{N}_2 \) and 250 Torr of \(^4\)He). In the numerical evaluation of the profile factors we used the \(^{39}\)K values from Tables B.1 and B.1 and made the assumption (which is only an approximation) that \(^3\)He and \(^4\)He affect alkali K the same way. We see that in both cases the alignment \((L=2)\) contribution is much smaller than the scalar \((L=0)\) and vector \((L=1)\) contributions, so that it can safely be ignored. However there are many experiments, especially with cold atoms (where the collisional and Doppler optical broadening is very small) and with heavier alkali atoms (e.g. Rb and Cs that have larger hyperfine separation), where the alignment component has a significant effect \([94, 196, 64]\).

### A.3 Irreducible Tensors

An irreducible tensor operator of rank \( L \) is a set of \( 2L + 1 \) functions \( T^L_M \) that transforms like the spherical harmonic of order \( L \) under the rotation group. Equivalently, \([188]\) they can be defined with respect to the commutation relationships with angular momentum operator \( J \):

\[
[J_{\pm}, T^L_M] = \sqrt{(L \mp M)(L \pm M + 1)} T^L_{M\pm 1}
\]  

\[
[J_0, T^L_M] = MT^L_M
\]
The significance of the irreducible representation of physical quantities lies in the fact that it captures most effectively the geometrical symmetry of the processes. From two irreducible tensor $T^k_i$ and $U^j_l$ we can construct the irreducible tensor $Q^s_i$ of rank $s$ according to the rule:

$$Q^s_i = \sum_{m,\lambda} T^k_i U^j_l C^{s\sigma}_{k\lambda m j \lambda}$$  \hspace{1cm} (A.24)

Based on the above equation we can construct a spherical tensor of rank 0 and of rank 1 from two vectors (tensors of rank 1). These are the familiar inner (scalar) and outer (vector) product. The inner product of vectors $\mathbf{A}$ and $\mathbf{B}$ is:

$$\mathbf{A} \cdot \mathbf{B} = \sum_m (-1)^m A_m B_{-m}$$  \hspace{1cm} (A.25)

The outer product can be put in the simple form \cite{98}:

$$\mathbf{A} \times \mathbf{B} = -i\sqrt{2} \sum_m (-1)^m A_m Q^1_m \cdot B$$  \hspace{1cm} (A.26)

where we used the definition of the basis dyadic (Equation \cite{8}). The following identity can be shown\cite{8}:

$$\mathbf{A} \cdot (\mathbf{B} \times \mathbf{C}) = -\mathbf{B} \cdot (\mathbf{A} \times \mathbf{C})$$  \hspace{1cm} (A.27)

\footnote{This is more easily done in the Cartesian basis rather than the spherical irreducible basis.}
Figure A.1: Dispersive parts of the polarizability tensor for $^{39}$K vapor at 12 amagats of $^3$He.
Figure A.2: Dispersive parts of the polarizability tensor for $^{39}$K vapor at 250 Torr of $^4$He and 50 Torr of N$_2$
Appendix B

B.1 Properties of K atoms

In the following table we summarize the properties of K atoms that are relevant to the atomic-optical magnetometer. For all the numerical calculations performed in this thesis we used the values from this table. See also [169] for a more detailed table of the alkali-metal properties.

<table>
<thead>
<tr>
<th>Alkali Isotope</th>
<th>$^{39}$K</th>
<th>$^{41}$K</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>Natural Abundance (%)</td>
<td>93.3</td>
<td>6.7</td>
<td></td>
</tr>
<tr>
<td>Nuclear Spin</td>
<td>3/2</td>
<td>3/2</td>
<td></td>
</tr>
<tr>
<td>D1 Transition in vacuum (nm)</td>
<td>770.108</td>
<td>770.108</td>
<td>[121]</td>
</tr>
<tr>
<td>D2 Transition in vacuum (nm)</td>
<td>766.701</td>
<td>766.701</td>
<td></td>
</tr>
<tr>
<td>Oscillator Strength D1</td>
<td>0.324</td>
<td>0.324</td>
<td>[138]</td>
</tr>
<tr>
<td>Oscillator Strength D2</td>
<td>0.652</td>
<td>0.652</td>
<td></td>
</tr>
<tr>
<td>Hyperfine Splitting $^2S_{1/2}$ (MHz)</td>
<td>461.7</td>
<td>254.0</td>
<td>[11]</td>
</tr>
<tr>
<td>Hyperfine Splitting $^2P_{1/2}$ (MHz)</td>
<td>57.7</td>
<td>30.4</td>
<td></td>
</tr>
<tr>
<td>Natural Lifetime $^2P_{1/2}$ (ns)</td>
<td>26.8</td>
<td>26.8</td>
<td>[191]</td>
</tr>
<tr>
<td>Natural Lifetime $^2P_{1/2}$ (ns)</td>
<td>26.5</td>
<td>26.5</td>
<td></td>
</tr>
</tbody>
</table>

Table B.1: Properties of K alkali-metal.
Table B.2: Parameters that characterize interaction properties of K with $^4$He and molecular $N_2$. The cross sections are expressed in units of cm$^2$, pressure broadening widths are in GHz/amg and refer to the full width at half maximum (FWHM), and pressure shifts are in GHz/amg. There is a small temperature dependence of the cross sections, widths and shifts, which to first order can be ignored. The diffusion constants are in units of cm$^2$/s amg and are given at 273K; the diffusion constant temperature dependence can be approximated by $D \propto T^{3/2}$ [174].

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\sigma_{K}^{se}$</td>
<td>$1.8 \times 10^{-14}$</td>
<td>[5]</td>
</tr>
<tr>
<td>$\sigma_{K}^{sd}$</td>
<td>$1.0 \times 10^{-18}$</td>
<td>[111]</td>
</tr>
<tr>
<td>$\sigma_{K}^{He}$</td>
<td>$8.0 \times 10^{-25}$</td>
<td>[71, 21]</td>
</tr>
<tr>
<td>$\sigma_{K}^{N_2}$</td>
<td>$7.9 \times 10^{-23}$</td>
<td>[111]</td>
</tr>
<tr>
<td>$D^{He}$</td>
<td>0.35</td>
<td>[71]</td>
</tr>
<tr>
<td>$D^{N_2}$</td>
<td>0.20</td>
<td>[174]</td>
</tr>
<tr>
<td>$\Gamma^{He}$ D1</td>
<td>13.3</td>
<td></td>
</tr>
<tr>
<td>$\Gamma^{He}$ D2</td>
<td>17.7</td>
<td></td>
</tr>
<tr>
<td>$\Delta \nu_0^{He}$ D1</td>
<td>3.9</td>
<td></td>
</tr>
<tr>
<td>$\Delta \nu_0^{He}$ D2</td>
<td>2.1</td>
<td></td>
</tr>
<tr>
<td>$\Gamma^{N_2}$ D1</td>
<td>21.0</td>
<td>[131]</td>
</tr>
<tr>
<td>$\Gamma^{N_2}$ D2</td>
<td>21.0</td>
<td></td>
</tr>
<tr>
<td>$\Delta \nu_0^{N_2}$ D1</td>
<td>-15.7</td>
<td></td>
</tr>
<tr>
<td>$\Delta \nu_0^{N_2}$ D2</td>
<td>-11.3</td>
<td></td>
</tr>
</tbody>
</table>
B.1.1 K vapor density

<table>
<thead>
<tr>
<th>Melting Point</th>
<th>Solid</th>
<th>Liquid</th>
</tr>
</thead>
<tbody>
<tr>
<td>63.5°C</td>
<td>4.961</td>
<td>4646</td>
</tr>
<tr>
<td></td>
<td>4.402</td>
<td>4453</td>
</tr>
</tbody>
</table>

Table B.3: Parameters for Eq. (B.1) that determine the vapor density of alkali K [4]. The temperature $T$ is expressed in Kelvin.

The saturated density of alkali vapor at temperature $T$ in Kelvin is given in units of cm$^3$ by [4]:

$$[K] = \frac{1}{T} 10^{21.866 + A - B/T} \tag{B.1}$$

where the parameters $A$ and $B$ can be found in Table B.1.1.
Appendix C

C.1 Correction factor for estimating uncertainty in correlated data

In this section we seek to find the factor that needs to be applied in the estimation of the uncertainty in the mean to correctly account for correlations between data.

For a series of data \( x_i, i = 1, 2 \ldots N \), taken at discrete, equally spaced times \( idt \) the mean estimator is:

\[
\bar{x} = \frac{1}{N} \sum_{i=1}^{N} x_i \tag{C.1}
\]

and the variance of the mean estimator is \([7]\):

\[
\text{Var} [\bar{x}] = \frac{1}{N^2} \sum_{i=1}^{N} \sum_{j=1}^{N} K [(i - j) dt] \tag{C.2}
\]

\[
= \frac{\sigma^2}{N} + 2 \frac{\sigma}{N} \sum_{i=1}^{N} \left(1 - \frac{k}{N}\right) K [idt] \tag{C.3}
\]

where \( K [(i - j) dt] = \langle x_i x_j \rangle - \langle x_i \rangle \langle x_j \rangle \) is the correlation function between the \( i \) and \( j \) element, and \( \sigma^2 \) is the variance of the samples \( x_i \) (we assume that all the samples have the same variance).
C.1.1 Effect of filter

The application of filter changes the correlation function of the sampled data. For simplicity we assume a bandpass filter with sharp cutoffs operating on a flat noise spectrum (the spectrum can be flat only in the bandpass region). It is then straightforward to show that the correlation function (Fourier transform of the power spectral density):

\[ K(\tau) = \frac{\sin \omega_l \tau - \sin \omega_h \tau}{(\omega_l - \omega_h) \tau} \sigma^2 \]  

(C.4)

where \( \sigma^2 \) is the variance of the sampled data, \( \omega_l \) and \( \omega_h \) are the upper and lower respectively (angular) frequency limits of the bandpass region. For the parameters of the thesis experiment \( (\omega_l = 2\pi \times 1.1 \, \text{sec}^{-1}, \omega_h = 2\pi \times 0.08 \, \text{sec}^{-1}, N \sim 560 \) and \( dt = 1/200 \, \text{sec} \)\), the summation in Eq. (C.3) can be performed numerically resulting in: \( \text{Var}[\bar{x}] \approx 112 \times \sigma^2/N \), or equivalently a correction factor \( g_0 = \sqrt{112} \) defined in Eq. (5.3). We note that the correlations between the data are mainly due to the effect of the low pass filtering, which effectively renders data sampled much faster than the cutoff frequency equivalent.

The approximate scaling of \( g_0 \) with respect to the cutoff frequency can most easily be seen in the limit \( N \times dt \gg \tau_c \), where \( \tau_c = 1/2\pi f_l \) is the characteristic correlation time introduced from the low pass filter. In [77], it is shown that the effective number of uncorrelated samples in a dense set of samples is:

\[ N_{\text{eff}} \sim \frac{Ndt}{\tau_c} \]  

(C.5)

which agrees with the intuition on the physical meaning of \( \tau_c \). From the above equation, it can be seen that \( g_0 \sim 1/\sqrt{f_l dt} \)

---

\(^1\)The indices \( h \) and \( l \) refer to the filter (high pass and low pass) rather than the frequency.
C.1.2 String analysis

The notation here follows the one in Section 5.2. For the calculation of the correction factor $f_0$ (equation (5.9)) we assume that $\sigma_{\bar{y}_k} = \sigma$ for all the samples; it is then straightforward to show that neglecting the small correlation between the $\bar{y}_k$ samples the variance in each string is: $\sum_{k=0}^{m} (C^m_k)^2/4^m$. From Eq. (C.3) the uncertainty in the mean estimator $\bar{\zeta}_r$ is given by:

$$\text{Var} [\bar{\zeta}_r] = \frac{\sum_{k=0}^{m} (C^m_k)^2}{N4^m} + \frac{2}{N} \sum_{j=1}^{N} \left(1 - \frac{j}{N}\right) K(j) \tag{C.6}$$

where $\sum_{k=0}^{m} (C^m_k)^2/N4^m$ is the variance of uncorrelated data, $N$ is the number of strings in the record, and $K(j)$ is the correlation function between strings that differ in their indexing by $j$. For the three sample string (Eq. (5.6)) the correlation function is:

$$K(j) = \begin{cases} \sigma^2, & j = 1 \\ \frac{\sigma^2}{16}, & j = 2 \\ 0, & j \geq 3 \end{cases} \tag{C.7}$$

Substituting Eq. (C.7) into (C.6) and neglecting terms $\propto 1/N^2$ (equivalently taking $1 - 1/N \approx 1 - 2/N \approx 1$) we find:

$$\text{Var} [\bar{\zeta}_r] = \frac{\sigma^2}{N} \tag{C.8}$$

In fact, this is a general result: to the extent that the approximations mentioned above hold, the variance in $\bar{\zeta}_r$ is given by Eq. (C.8). Then the correction factor $f_0$, found from the ratio of actual uncertainty (standard deviation) to uncertainty $\bar{\zeta}_r$ is given by:

$$f_0 = \frac{\sigma}{\text{Var} [\bar{\zeta}_r]} \tag{C.9}$$

Because of the equal sample uncertainties $\sigma_{y_k}$ the weighted mean is identical to the simple mean estimator.
assuming uncorrelated data, is:

\[ f_0 = \frac{2^m}{\sqrt{\sum_{k=0}^{m} (C_k^m)^2}} \]  \hspace{1cm} (C.9)

where \( m + 1 \) is the number of points per string; for \( m = 2 \), \( f_0 = 4/\sqrt{6} \).
Bibliography


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[112] A. Kastler. Some suggestions concerning the production and detection by optical means of inequalities in the populations of levels of spatial quantization


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