Interplay between atomic alignment and orientation fluctuations in the spin noise spectroscopy of metastable helium

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We perform spin noise spectroscopy experiments in metastable helium atoms at room temperature, with a probe light whose frequency is blue detuned from the D0 line. Both circular birefringence fluctuations (Faraday noise) and linear birefringence fluctuations (ellipticity noise) are explored theoretically and experimentally. In particular, in the latter case, it is shown that two noise resonances are isolated at the Larmor frequency and at twice the Larmor frequency. The behavior of these resonances with the orientation of the probe field polarization is analyzed and explained for large probe detunings. Moreover, the simple structure of metastable helium allows us to probe, model and explain the changes in the behavior of these peaks when the probe detuning is reduced, inducing an optical pumping, which leads to circular and linear dichroisms.

I. INTRODUCTION

Noise is usually considered as detrimental to measurements. However, noise of fundamental origin can be of tremendous interest for physicists, as it may contain useful information. A striking historical example is the determination of star diameters using correlations in light intensity fluctuations [1]. Further, the intensity noise of a probe laser can be analyzed for spectroscopy purposes [2]. More recently, the development of quantum information protocols in the continuous variable domain fundamentally relies on quantum noise correlations between light quadratures [3], while second order (and higher order) correlators of atomic spin fluctuations were shown to give access to classical (and purely quantum) signatures of the interaction of the system with its environment [4–7].

Nevertheless, experimental measurements of spin correlations are usually challenging, owing to the fact that the signal is extremely weak. In magnetic systems, the spectroscopy of fundamental noise due to random spin fluctuations, called Spin Noise Spectroscopy (SNS), can be optically performed by measuring the associated fluctuations of the Faraday-like rotation experienced by a linearly polarized probe beam propagating through the sample [3] (see Fig. 1(a)). Although a first experimental effort was initially reported in the early 1980s [8], only recently this method has seen a renewed interest due to advances in narrow linewidth lasers and developments in low noise electronics required for spectrum analysis [9].

It was then used to probe different properties of various media such as thermal atomic vapours, semiconductors or quantum wells [5].

SNS is also proposed as a tool for precision magnetometry [10], using squeezed-light [11] or homodyne detection amplification [12] [13] to improve its sensitivity. Recent works extend the SNS technique to the study of spatiotemporal correlations by spatial phase-modulation of the optical probe [14]. Measurement of correlations beyond the second order raises a lot of interest, as they can be used to probe the limits of the linear response and fluctuation-dissipation theory and thus give access to new phenomena [15].

Contrary to alkali atoms, the |2\(^1\)S\(_1\rangle \leftrightarrow |2\(^3\)P\rangle transition of metastable helium has a fine structure only, with three absorption lines separated by a frequency difference larger than the 0.8 GHz Doppler broadening at room temperature [10]. The highest excited level is...
the \(|^{2}P_{0}\rangle\) state, which is nearly 30 GHz away from the intermediate \(|^{2}P_{1}\rangle\) one: one can then choose to blue detune the frequency of a probe beam from the corresponding \(D_{0}\) line in order to probe this transition in a purely dispersive and perturbative manner without being significantly disturbed by the influence of the two other lines. The aim of the present paper is thus to explore the properties of the SNS in this relatively pure and simple closed \(J = 1 \rightarrow J = 0\) atomic system, in order to put into evidence the peculiarities of spin noise in the case of a spin 1 system like the \(|^{2}S_{1}\rangle\) state. In particular, we find that such a spin 1 system can exhibit two spin noise resonances instead of one, corresponding to atomic alignment and orientation fluctuations. The existence of the second resonance has already been reported [17, 18], but the use of metastable helium allows us to fully explore and understand its polarization dependence. Moreover, we explore the changes in the polarization dependent behavior of both resonances when the detuning of the probe laser is varied. Based on simulations, we look for a simple physical picture for the measured results.

II. EXPERIMENTAL SET-UP

Figure 1(b) shows a schematics of the experimental setup. A linearly polarized laser beam at 1.083 \(\mu\)m is sent into a cell filled with 1 Torr of \(^{3}\)He at room temperature. This wavelength corresponds to the \(|^{2}S_{1}\rangle \leftrightarrow \ |^{2}P\rangle\) optical transitions, and the laser beam diameter is about 0.6 mm. A radiofrequency discharge at 27 MHz generates a plasma in the cell, so that a fraction of helium atoms are excited to their metastable state through collisions with electrons, leading to a metastable helium density of about \(2 \times 10^{11} \text{ cm}^{-3}\). The remaining He atoms then play the role of a buffer gas, which redistributes optical pumping all over the Doppler broadening by collisions [114]. While propagating into the helium cell, the laser beam experiences a random Faraday rotation induced by random fluctuations of the populations of the three Zeeman ground state sublevels. A transverse DC magnetic field is added to the atoms in order to shift the spin noise resonance around the Larmor frequency \(\omega_{L}\) [4, 5]. It is obtained using two rectangular coils, which are larger than the cell, so that it can be considered as homogeneous along the probe beam propagation. The \(|^{2}S_{1}\rangle\) ground state has three Zeeman sublevels, with a \(\pm 2.8 \text{ MHz/Gauss Zeeman shift}\) for the \(m = \pm 1\) sublevels, respectively. In our experimental conditions, the Larmor frequency lies in the MHz range.

A half-wave plate (HWP) followed by a polarizing beamsplitter (PBS) allow to get an equal average intensity on both photodiodes of a balanced detection, so that their signal difference is zero on average. The associated noise power spectral density can then be recorded using an electrical spectrum analyzer (ESA). To model the balanced detection scheme, the output field complex amplitude can be written as the sum of a mean value \(E\) and a fluctuating part \(e(t)\):

\[
E_{\text{out}}(t) = E + e(t),
\]

where \(E\) is assumed to be real, and the fluctuations \(e(t)\) can consist in amplitude, phase or polarization variations, which carry the spin noise information.

After the HWP, the field sent to the balanced detection has an average polarization at 45° of the beamsplitter axes \(x\) and \(y\):

\[
E_{\text{out}}(t) = \frac{1}{\sqrt{2}} \left[ (E + e_{\|}(t) + e_{\perp}(t)) x + (E + e_{\|}(t) - e_{\perp}(t)) y \right].
\]

where \(e_{\|}(t)\) and \(e_{\perp}(t)\) are the components of \(e(t)\), which are parallel and perpendicular to \(E\), respectively, and \(E\) is the amplitude of \(E\). The signal at the output of the balanced detection is thus proportional to:

\[
D = |E_{\text{out}}(t) \cdot x|^2 - |E_{\text{out}}(t) \cdot y|^2
= 2E \text{Re}(e_{\perp}(t)) + O(e(t)^2).
\]

The ESA thus records the power spectral density (PSD) of the field fluctuations polarized orthogonally to the mean output field, and in phase with it. This corresponds to fluctuations of the orientation of a linearly polarized field, which we can call Faraday noise. This first order calculation remains valid in the case of a mean field with a small ellipticity. Figure 1(c) shows...
examples of experimental noise spectra obtained for increasing transverse magnetic field values, in the case of a 1.5 mW probe laser blue detuned by 1.5 GHz: the spin noise resonance frequency, which is centered on the corresponding Larmor frequency, increases with the magnetic field value.

It is also possible to monitor ellipticity fluctuations by inserting a quarter-wave plate (QWP) just after the cell, as shown in Fig. 2(a). The QWP is oriented to transform an input elliptic polarization into a linear one: the ellipticity noise is then changed into an orientation noise, which can be detected with the same set-up as before. However, the QWP induces a $\pi/2$ phase shift on the fluctuations along the $y$ axis so that, when the averaged ellipticity is small and can be neglected, Eq. (2) must be replaced by:

$$E_{\text{out}}(t) = \frac{1}{\sqrt{2}} \left[ (E + e_{\parallel}(t)) \mathbf{x} + (E + e_{\parallel}(t) - i e_{\perp}(t)) \mathbf{y} \right].$$

Consequently, the signal analyzed by the ESA becomes:

$$D = 2E \text{Im} (e_{\perp}(t)) + \mathcal{O}(e(t)^2).$$

Instead of the field fluctuations that are in phase with the mean field, this setup thus probes the part of the fluctuations in quadrature with it, i.e., the ellipticity noise. In the following, we call this setup the ellipticity noise detection (END) setup, while the first one is called the rotation noise detection (RND) setup. These two setups correspond to the two limiting cases explored in [13], where the relative phase between the local oscillator and the signal can be continuously tuned: the balanced detection plays the role of a homodyne detector in which the local oscillator is provided by the mean field $E \mathbf{x}$ and the role of the signal is played by the fluctuations $e(t) \cdot \mathbf{y}$. One can choose between the two signal quadratures by adding or not a QWP as shown in Fig. 2(b). It should be noted that we always check that the wave plate is oriented so that the average output polarization is linear and the detection properly balanced.

| Frequency (MHz) | PSD (fW/Hz) |
|----------------|-------------|
| 0.18           | 0.18        |
| 0.16           | 0.16        |
| 0.13           | 0.13        |

Table 1: Examples of experimental spectra obtained with this END set-up for increasing transverse magnetic field values, in the case of a 1.5 mW probe laser blue detuned by 1.5 GHz: one can notice that there are now two spin noise resonances, at the Larmor frequency $\omega_L$ and at $2\omega_L$. The existence of the second peak is discussed in the next section.

**III. RESONANCE AT TWICE THE LARMOR FREQUENCY**

The existence of such a dual-peak noise spectrum can be understood if one remembers that the probed ground state is a spin 1 system, composed of three Zeeman sublevels. Figure 3 reproduces the eigenmodes of evolution of the density matrix of the ground state in the basis of the Zeeman sublevels $m_z = -1, 0, +1$, where $z$ is the light propagation direction. The system evolves under the influence of the magnetic field oriented along $x$.

Figure 3(c) shows examples of experimental noise spectra obtained for increasing transverse magnetic field values, in the case of a 1.5 mW probe laser blue detuned by 1.5 GHz: one can notice that there are now two spin noise resonances, at the Larmor frequency $\omega_L$ and at $2\omega_L$. The existence of the second peak is discussed in the next section.

**Figure 4.** (a,b) Experimental and (c,d) simulated power spectral densities (PSD) obtained 1.5 GHz above the $D_0$ transition for a 1.5 mW laser input power. They are plotted as a function of the input polarization angle $\theta$ from the transverse magnetic field for (a,c) the RND set-up and (b,d) the END set-up. Experimental spectra are recorded every 4°. The ESA resolution and video bandwidths are 91 kHz and 15 Hz, respectively. The simulations are obtained with a spontaneous emission decay rate $\Gamma_0/2\pi = 1.6$ MHz, an optical coherence decay rate $\Gamma/2\pi = 0.8$ GHz that takes Doppler broadening into account, and equal transit and Raman decay rates $\Gamma_1/2\pi = \Gamma_2/2\pi = 30$ kHz. The probe beam Rabi frequency is $\Omega/2\pi = 40$ MHz.

**Figure 2(c) shows examples of experimental noise spectra obtained with this END set-up for increasing transverse magnetic field values, in the case of a 1.5 mW probe laser blue detuned by 1.5 GHz: one can notice that there are now two spin noise resonances, at the Larmor frequency $\omega_L$ and at $2\omega_L$. The existence of the second peak is discussed in the next section.**
performs a complete oscillation in a time $\pi/\omega_L$.

If one chooses the quantization axis along $x$, i.e., along the magnetic field direction, the frequency $\omega_L$ corresponds to the transition between the levels $|m_x = 0\rangle$ and $|m_x = \pm 1\rangle$, while $2\omega_L$ corresponds to the transition between the levels $|m_x = -1\rangle$ and $|m_x = +1\rangle$. Now, if we choose the quantization axis along $z$, the system obeys the two-coupled-oscillator equations with a coupling strength $\hbar\omega_L/\sqrt{2}$:

$$ |m = -1\rangle \frac{\omega_L}{\sqrt{2}} |m = 0\rangle \frac{\omega_L}{\sqrt{2}} |m = +1\rangle. $$

Such a system has two oscillation modes: the first one at $\omega_L$ is a spin orientation mode, which oscillates between two opposite magnetizations and corresponds to Fig. 3(down); the second one at $2\omega_L$ is a spin alignment mode, which keeps the magnetization at zero and corresponds to Fig. 3(up).

**IV. POLARIZATION DEPENDENCE**

The results presented in Fig. 2(c) were obtained for a specific input polarization angle $\theta = 55^\circ$ with respect to the magnetic field orientation. More information is gained by scanning the polarization angle from $\theta = -4^\circ$ to $\theta = 94^\circ$ for both the RND and the END setups. Such SNS scans, with their spectra coded in false colors, are shown in Fig. 4(a,b). They are obtained with a 1.5 mW probe beam, which is blue detuned by 1.5 GHz from the $D_0$ line, for the (a) RND and (b) END setups. In the former case only the peak at $\omega_L$ is present, and its power does not exhibit any polarization dependence. On the contrary, both resonances can be detected in the case of the END setup, but not for the same input polarization orientations: the resonance at $\omega_L$ is visible around $\theta \approx 0$ and $\theta \approx 90^\circ$, while the peak at $2\omega_L$ exists for $\theta \approx 50^\circ$ when the peak at $\omega_L$ disappears.

We compared these measurements with numerical simulations of the time evolution of the density matrix of the system. We consider the fourth-dimensional Hilbert space generated by the three Zeeman sublevels of the $|2^3S_1\rangle$ state and the $|2^3P\rangle$ level. The evolution of $\rho$ is simulated according to the following equation:

$$ \frac{d\rho}{dt} = \frac{1}{i\hbar}[H, \rho] + \frac{1}{i\hbar}D(\rho) + f(t), $$

where $H$ is the Hamiltonian and $D(\rho)$ and $f(t)$ represent the relaxations and fluctuations of the system, respectively. For a given probe laser polarization, we can then simulate the time dependent signal obtained by the subtraction of the two photodiode currents and, by Fourier transform, obtain the corresponding spectrum. The discretization time is 50 times smaller than the smallest timescale of the system (given by the optical detuning or the Doppler broadening) and the time duration of the signal is 50 times longer than the largest timescale of the system (usually given by the transit and Zeeman coherence decay rates of 30 kHz).

The spin fluctuation matrix $f(t)$, reduced to the three Zeeman sublevels of the $|2^3S_1\rangle$ state, is generated from random numbers at each simulation step. As the model should remain consistent whatever the quantum axis, we can show that in the basis of the three ground levels $|1\rangle_z, |0\rangle_z, |1\rangle_z$ it reads:

$$ f(t) = \begin{pmatrix} -f_{-1,0} - f_{-1,1} & k^*_0 & k^*_0 \\ k_{-1,0} & -f_{-1,0} - f_{1,0} & k^*_{0,1} \\ k_{1,1} & k_{0,1} & f_{-1,1} - f_{1,0} \end{pmatrix}, $$

with $f_{i,j} \sim N(0, \sigma^2)$, $\Re(k_{i,j}) \sim N(0, 3\sigma^2/2)$, and $\Im(k_{i,j}) \sim N(0, 3\sigma^2/2)$. In these expressions, $N(x, \delta)$
is a random variable obeying the normal law of mean value $x$ and variance $\delta$, and $i, j = -1, 0, 1$. To estimate the value of $\sigma$, we consider the motion of the atoms through the laser beam, with its transit rate $\gamma$, to be the main contribution to the fluctuation terms, which can be modelled by a Poissonian law. For an average number of atoms $N$ in the interaction region, the associated variance is then $\sigma^2 = C/dt$, where $C = \gamma/3N$ and $dt$ is the discretization time. In our experimental conditions, we obtain $C \approx 5.8 \times 10^{-5} \text{s}^{-1}$.

The results of these simulations are given in Fig. 3 (c,d), in the case of both setups. They reproduce very well the experimental results. For example, even the fact that the noise at $\omega_L$ is stronger close to $\theta = 0^\circ$ than to $\theta = 90^\circ$. Nevertheless, one can notice a small discrepancy by about 5 to 10 between the angular positions of the peaks, and the fact that the experimental resonances are broader than the theoretical ones. The first discrepancy can be explained by the fact that finding the angle $\theta = 0$ is not easy and by some small angular shift of the DC magnetic field. The fact that the experimental peaks are broader than the simulations can arise from the fact that the Rabi frequency can be underestimated. Indeed, the simulations are based on its average value across the Gaussian beam section, while in the experiment it is larger at the center of the beam, which can broaden the SNS peaks. Possible magnetic field inhomogeneities can also contribute to this discrepancy.

Fomin & al. have already mentioned that two polarization eigenmodes can be recorded when ellipticity noise is probed. Nevertheless, these authors recorded SNS spectra only for $\theta = 0$, 45, and 90, and they observed a polarization dependence of the noise peaks that significantly differs from the one observed here (see Fig. 4) and from the predictions of these authors.

This was attributed to the fact that these experiments were performed close to the center of the D2 line of Cs atoms. The associated absorption then induces an ellipticity contribution, as already reported in semiconductors [19][20].

In the following section, we thus explore the behavior of our system when the probe detuning is reduced, and the influence of the associated optical pumping processes.

### V. DETUNING BEHAVIOR

Figure 6 shows numerical simulations of the spin noise behavior in the case of detection of (a-d) Faraday noise, i.e., circular birefringence and (e-f) ellipticity noise, i.e., linear birefringence fluctuations. In both cases, the plots from left to right correspond to decreasing values of the detuning $\Delta$ of the probe laser. It can be clearly noticed that the peak at $2\omega_L$ progressively disappears when $\Delta$ decreases in the case of the END setup, while it becomes more and more visible with the RND one. It gets also slightly shifted to smaller values of $\theta$.

At the same time, the peak at $\omega_L$ follows opposite changes for the two setups : it is nearly polarization independent at $\Delta = 300\text{ MHz}$ in the first case and at $\Delta = 1.5\text{ GHz}$ in the last one. On the contrary, this noise peak disappears in the END set-up for $\theta \approx 45$ when the detuning $\Delta$ is large, while in the case of the RND one it vanishes for small detunings.

We have tested these predictions experimentally. The corresponding data are reproduced in Fig. 6 for four different values of the probe detuning $\Delta$ with respect to the center of the Doppler profile. In all cases, the Larmor frequency is $\omega_L/2\pi \approx 2.3\text{ MHz}$ and the laser power
is about 1 mW.

Figures 6(a-d) were obtained with the RND setup for decreasing values of the detuning \( \Delta = 1.5 \text{GHz}, 3 \text{GHz}, 500 \text{MHz}, \) and 300 MHz. As predicted in Figs. 5(a-d), the peak at \( \omega_L \) becomes polarization dependent when \( \Delta \) decreases. Moreover, if one looks carefully at the RND results obtained at \( \Delta = 500 \text{MHz} \) and 300 MHz (Figs. 6(c) and (d), respectively), a quite weak SNS peak indeed appears at frequencies between 4.5 and 5 MHz and for angles \( \theta \) between 20 and 50°. The origin of the noise floor increase for angles \( \theta < 20 \) will be discussed below.

In the case of the END setup, the results of Figs. 6(e-h) also confirm the predictions of Figs. 5(e-h). Indeed, when \( \Delta \) decreases, the peak at \( 2\omega_L \) progressively disappears and the peak at \( \omega_L \) becomes less and less polarization dependent, although this effect is difficult to see because of the increase of the noise floor level for small values \( \theta \) when \( \Delta \) decreases.

This increase of the noise floor at small probe detunings, and its polarization dependence (see Figs. 5(c,d,g,h)), can be explained by the fact that the optical pumping effects make the probe absorption polarization dependent. Figures 7(a) and (d) show measurements of the evolution of the cell absorption versus \( \theta \) for \( \Delta = 1.5 \text{GHz} \) and \( \Delta = 300 \text{MHz} \), respectively. One can see that the polarization dependence of the cell absorption, which is already visible at \( \Delta = 1.5 \text{GHz} \), becomes strong at \( \Delta = 300 \text{MHz} \). The fact that the absorption exhibits a minimum at \( \theta = 0 \) is due to the fact that only the \( |m_z = 0\rangle \leftrightarrow |m_z = 0\rangle \) transition is probed in this case, leading to a strong optical pumping of the atoms to the \( |m_z = \pm 1\rangle \) sublevels.

This reduction of the absorption around \( \theta = 0 \) leads to an increase of the power reaching the balanced detection and thus to an increase of the associated shot noise. We thus took this effect into account in the simulations, leading to the results shown in Figs. 7(b,c,e,f). The agreement with the measurements of Fig. 7 is significantly improved.

Nevertheless, we notice that in spite of this improvement, some discrepancies still remain when \( \Delta \) is small: the peak at \( 2\omega_L \) in the RND configuration is weaker than expected, and the peak at \( \omega_L \), which is expected to exhibit a maximum at \( \theta = 0 \) and 90°, is shifted towards \( \theta = 20° \) and 70°. These discrepancies can originate from the fact that our simulations consider only an optically thin medium, and model the Doppler broadening by a simple increase of the homogeneous linewidth. Also, they might be due to the fact that we model the detection setups up to first order in fluctuations only. Finally, fluctuation sources other than the transit of the atoms through the laser beam could also be considered.

Finally, this exchange of behaviors between the RND and END setups when the detuning \( \Delta \) is decreased (see Fig. 5 and 6) is due to the fact that the optical pumping occurring at small detunings induces dichroism in the vapor. In the case of the RND setup, while the experiment is only sensitive to Faraday noise, i.e., circular birefringence, at large detunings, some linear dichroism occurs when \( \Delta \) is reduced, leading to the appearance of the peak at \( 2\omega_L \). Conversely, in the case of the END setup, while only linear birefringence, i.e., alignment noise, is observed at large detunings, optical pumping effects at smaller detunings induce circular dichroism and affect the polarization dependence of the system.
VI. CONCLUSION

We have investigated the spin noise spectra of a thermal vapor of metastable $^4$He atoms at room temperature using a probe beam, which is tuned in the vicinity of the D0 line. Using two different setups, we could record the signature of spin fluctuations using either circular or linear birefringence of the atomic ensemble. In the first case, i.e. for Faraday noise, the well-known and polarization independent SNS resonance was observed at the Larmor frequency $\omega_L$. In the second case, corresponding to ellipticity noise, we could confirm that a spin 1 system can give rise to a second resonance peak at twice the Larmor frequency for large optical detunings [17, 18]. We performed a full scan of the dependence of the intensities of the noise peaks as a function of the angle between the probe beam linear polarization and the transverse magnetic field. Moreover, we achieved numerical simulations of the system, with a spin noise due to the transit of the atoms through the laser beam, which are in good agreement with the experimental results obtained far from the center of the Doppler broadened optical resonance.

These numerical simulations predict that at smaller optical detunings, the behaviors of the two types of experiments should be exchanged, which means that the correlations are modified by the optical pumping: linear and circular dichroism effects become progressively dominant compared to circular and linear birefringences. We have recorded data in such conditions and demonstrated that this interplay is indeed visible. These results, which are due to the optical pumping induced by the probe absorption, were discussed in details, together with the remaining discrepancies between theory and experiments. It was possible thanks to the simple structure of metastable helium, which allows to consider only the effect of the D0 transition. Future theoretical work should better take into account the effect of the Doppler broadening and the fact that the medium becomes optically thick when the probe is close to resonance. Various fluctuation sources should also be considered: the well separated transitions of metastable helium can again be an asset for a deep understanding of the contributions of the effects affecting the spin noise spectra of a spin 1.

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