Optical measurements of spin noise as a high resolution spectroscopic tool

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The intrinsic fluctuations of electron spins in semiconductors and atomic vapors generate a small, randomly-varying “spin noise” that can be detected by sensitive optical methods such as Faraday rotation. Recent studies have demonstrated that the frequency, linewidth, and lineshape of this spin noise directly reveals dynamical spin properties such as dephasing times, relaxation mechanisms and g-factors. Here we demonstrate that spin noise measurements using wavelength-tunable probe light forms the basis of a powerful and novel spectroscopic tool to provide unique information that is fundamentally inaccessible via conventional linear optics. In particular, the wavelength dependence of the detected spin noise power can reveal homogeneous linewidths buried within inhomogeneously-broadened optical spectra, and can resolve overlapping optical transitions belonging to different spin systems. These new possibilities are explored both theoretically and via experiments on spin systems in opposite limits of inhomogeneous broadening (alkali atom vapors and semiconductor quantum dots).

Optical probes of spin and magnetization dynamics enjoy broad applications across the many atomic, semiconductor, and metallic systems in which spin-orbit interactions allow coupling between electron spin polarization and the optical (circular) polarization of light. Experimental techniques include powerful spectroscopies based on optical Faraday/Kerr rotation, circular dichroism, or circularly-polarized photoluminescence. Using these methods, signals are typically proportional to some intentionally-induced change in net magnetization or spin polarization. Recently, however, sensitive magnetometers based on optical Faraday rotation (FR) have detected the tiny stochastic fluctuations of unperturbed spins in thermal equilibrium. In accord with the fluctuation-dissipation theorem, the radio-frequency spectrum of this intrinsic “spin noise” reveals the same dynamical spin properties as in conventional magnetic resonance techniques, but without driving the spin system away from equilibrium.

The use of FR to detect spin noise is motivated by its sensitivity to spin polarization in materials with spin-orbit coupling. This coupling leads to the well-known optical selection rules that preferentially couple optical transitions from spin-up or spin-down electron states to right- or left-circularly polarized light, respectively. Examples of such systems include band-edge transitions in III-V semiconductors, and the \( S \to P \) transitions of alkali atoms.

The energy-dependent Verdet constant \( V(E) \) of a material characterizes how much FR is elicited (per unit applied magnetic field and per unit system length) when light of photon energy \( E \) passes through the material. \( V(E) \), being sensitive to the difference between right- and left- circular indices of refraction, typically exhibits a dispersive lineshape near an isolated spin-sensitive optical transition, passing through zero at line center. When using FR to detect spin noise, it might therefore seem sensible to tune the probe wavelength to the peak of \( V(E) \), and avoid energies where \( V(E) \to 0 \). However, owing to the correlated or uncorrelated nature of spin fluctuations in different spin systems, we shall show that this is not always the case, and that in general there is no direct connection between \( V(E) \) and the magnitude of the measured spin noise.

Here we demonstrate that photon energy-dependent measurements of optical spin noise (henceforth called ‘OSN spectra’) can provide highly specific information about spin-dependent optical transitions, and can function as a novel and powerful spectroscopic tool.

In general the optical spectrum of a paramagnetic material, whose ground state represents the spin system of interest, is composed of multiple bands. Each band makes a contribution \( \theta_{i}(E) \) to the total FR \( \theta(E) \) (see Methods):

\[
\theta(E) = \sum_{i} \theta_{i}(E) \sim \sum_{i} V_{i}(E).
\]

Here, \( V_{i}(E) \) is the corresponding partial Verdet constant of the paramagnet. Note that we are considering only the paramagnetic contributions to \( \theta(E) \) connected with magnetization of the spin system.

We focus on the quantity \( \langle \delta \theta^{2}(E) \rangle \), the mean-square fluctuations of FR due to spin fluctuations. It can contain fluctuating contributions from multiple spectral bands.
When calculating its spectral dependence, however, we must distinguish between two fundamentally different cases. In the first case, all the constituent spectral bands are related to a single spin system, such that FR fluctuations from different bands occur synchronously and are correlated. Therefore \( \langle \delta \theta^2(E) \rangle \) will vary with \( E \) as the mean-square sum of all the partial contributions from which interference terms between the bands may arise:

\[
\langle \delta \theta^2(E) \rangle \sim \left( \sum_i V_i(E) \right)^2.
\] (2)

In the second case, the various spectral bands are each related to a different spin system, meaning that spin fluctuations in different bands are uncorrelated. Here, \( \langle \delta \theta^2(E) \rangle \) must be computed as the sum of the mean-square partial fluctuations, so that no interference occurs:

\[
\langle \delta \theta^2(E) \rangle \sim \sum_i \langle |V_i(E)|^2 \rangle.
\] (3)

Crucially, this distinction is significant for closely-spaced bands, and becomes most important when an overlapping multitude of nominally unresolvable spectral components exists (as for inhomogeneously broadened optical spectra).

Figure 1 demonstrates this distinction for a simple model optical spectrum containing two well-resolved and homogeneously-broadened bands. Shown are the absorption spectrum, FR spectrum, and associated OSN spectra for the case where the two bands originate from a common spin system, and for the case of two independent spin systems, calculated using Eqs. (2) and (3) respectively. Note that the conventional FR spectrum calculated using Eq. (1) remains the same whether the two optical transitions derive from a common spin system or not – linear magneto-optics cannot distinguish between these two cases. In marked contrast, the OSN spectra are fundamentally different: \( \langle \delta \theta^2(E) \rangle \) can be large between the two components for the case of independent spin systems (bottom curve), even though the net FR vanishes.

A second example of this essential distinction and of the informative potential of OSN spectroscopy is presented in Fig. 2A, which shows absorption, FR, and OSN spectra for two overlapping spectral bands. Clearly, the two transitions are not resolved in absorption or in FR. However, they are resolved in the OSN spectrum when the bands originate from two independent spin systems, because the spin fluctuations are uncorrelated.

These examples show that OSN spectroscopy, which uses only weak optical fields, opens access to information lying beyond the potential of conventional linear optics. A fascinating illustration of this fact can be seen in Fig. 2B which considers the evolution of OSN spectra for independent spin systems as optical transitions become increasingly dominated by inhomogeneous broadening. In this limit the absorption, FR, and OSN spectra are calculated as the convolution of the corresponding homogeneous spectrum with a broader Gaussian function representing the inhomogeneous distribution of individual optical transitions (see Methods). Figure 2B shows that the shape of OSN spectra changes dramatically as the ratio of inhomogeneous to homogeneous linewidth, \( \varepsilon = \gamma_{inh}/\gamma_h \), increases. In contrast, the shape of the absorption, FR spectra and spectra of common spin system remain practically the same for all \( \varepsilon \) (not shown here). When \( \varepsilon \ll 1 \) and the optical transition is mostly homogeneously broadened, the OSN spectrum approximately follows the FR squared. In this case both situations (common and independent spin systems) show a pronounced dip at band center. As inhomogeneous broadening increases the dip becomes shallower, eventually disappearing when \( \varepsilon > 1 \), see Fig. 2B. For bands having strong inhomogeneous broadening (\( \varepsilon \gg 1 \)), the OSN spectrum becomes similar to that of the absorption spectrum (i.e., with a maximum at band center), despite the fact that the FR is zero at band center.

Additionally, the area under the OSN spectrum increases rapidly for higher \( \varepsilon \) - that is, as the homogeneous linewidths of individual resonances (within an inhomogeneously-broadened band) become smaller, Fig. 2B. Qualitatively, this behavior can be explained by the fact that for smaller \( \gamma_h \), the probe becomes much more sensitive to those resonances at small detuning \( \Delta \), since the magnitude of the FR from each individual resonance varies as \( \Delta/(\Delta^2 + \gamma_h^2) \).

Figure 2B summarizes these findings and shows the evolution of the central dip in the OSN spectrum and the development of the enhancement factor with \( \varepsilon \), calculated for an absorption band having a constant total oscillator strength. The enhancement factor represents the ratio between the areas under the OSN spectra calculated for independent and common spin systems. For very small \( \varepsilon \) these areas are equal, giving no enhancement, while with increase of \( \varepsilon \) the OSN of independent spin systems prevails.

The strong dependence of the enhancement on \( \varepsilon \), as well as the depth of the dip in the OSN spectrum, can therefore be used for evaluating the degree of inhomogeneous broadening of optical transitions associated with spin systems.

It is interesting to note that, from an experimental viewpoint, systems with strong inhomogeneous broadening can be considered as very favorable objects for OSN spectroscopy, due to the enhancement of measured spin noise power at large \( \varepsilon \).

To validate and illustrate the above considerations, we present experimental OSN data from two spin systems that correspond to the limits of predominantly homogeneous and inhomogeneous broadening (\( \varepsilon \ll 1 \) and \( \varepsilon \gg 1 \), respectively).
For the case of predominantly homogeneous broadening ($\varepsilon \ll 1$), we studied a warm (110°C) vapor of the potassium isotope $^{41}$K in nitrogen buffer gas (see Methods). Figure 3 shows how the raw spin noise data vary as the probe laser is tuned through the fundamental spin-sensitive D1 transition ($4S_{1/2} \rightarrow 4P_{1/2}$). Integrating the total noise power under these data gives the associated OSN spectrum shown in Fig. 3b. As expected (see Fig. 2a, $\varepsilon = 0.1$), it is comprised of two bumps with a well-pronounced dip at line center. This finding is supported by recent observations on $^{87}$Rb vapor.

To investigate OSN in the opposite regime of predominantly inhomogeneous broadening, we measured the spin noise from an ensemble of hole-doped (In,Ga)As/GaAs quantum dots (QDs). At low temperatures, individual QDs exhibit sharp atomic-like optical transitions as narrow as tens of $\mu$eV; however, the distribution of QD sizes and compositions leads, in ensemble measurements, to a broad ~20 meV inhomogeneous linewidth. The optical transition from a single resident hole to a positively charged trion is right- or left circularly polarized depending on the hole’s initial spin orientation. Hole spin fluctuations therefore generate a measurable spin noise [7, 17].

Figure 2h shows the OSN spectrum from these holes as the probe laser was tuned through the absorption band of the QD ensemble. Also shown for reference is the usual FR spectrum of the QD ensemble, obtained by intentionally polarizing the hole spins with circularly polarized light at 1.579 eV, and simultaneously detecting the induced FR as a function of probe photon energy. As predicted for the case of an inhomogeneously-broadened line (see Fig. 2a, $\varepsilon = 10$), the OSN spectrum does not exhibit any dip at line center, but rather achieves a maximum, even though the induced FR passes through zero.

Interestingly, additional confirmation of the model is obtained from the temperature dependence of the total spin noise from these QDs. As predicted above in Fig. 2a, the detected spin noise from an inhomogeneously-broadened system will change if the ratio $\gamma_{inh}/\gamma_h$ varies. In these (In,Ga)As QDs, $\gamma_h$ is expected to be strongly temperature dependent, even at low temperatures [18]. This change is clearly apparent in the noise measurements: Fig. 4b shows the total spin noise power measured from 4 to 30 K, using a fixed probe energy (1.406 eV) corresponding to the maximum of the OSN spectrum. The total noise decreases as $\gamma_h$ of the underlying QDs grows with increasing temperature, approximately in accord with that measured by sophisticated four-wave mixing techniques in similar (In,Ga)As QDs [18]. Thus, spin noise measurements alone provide a powerful experimental tool for penetrating the internal structure of inhomogeneously broadened systems and for revealing variations of homogeneous linewidths that may otherwise be obscured by strong inhomogeneous broadening. It is especially noteworthy that this sensitivity to the hidden fine structure of an inhomogeneously broadened band works even when $\gamma_{inh} \gg \gamma_h$, making it possible to realize an effectively high (sub-linewidth) spectral resolution by means of a linear optical technique.

In summary, we have shown that an optical spectroscopy based on measuring spin fluctuations can provide important information about spin systems that is generally regarded as inaccessible for conventional linear optical spectroscopic techniques. The capabilities of this technique are illustrated by experimental OSN spectra of homogeneously and inhomogeneously broadened spin systems – electrons in the 4S ground state of $^{41}$K, and resident holes in (In,Ga)As quantum dot ensembles, respectively. These unique properties of OSN spectroscopy that make it possible to penetrate inside the optical bands while remaining within the bounds of linear optical polarizability make it a powerful method of research applicable to many materials important for present-day photonics and information science.

**METHODS**

**Optical spin noise measurements.** Spin noise measurements were performed using a weak, linearly-polarized beam from a continuous-wave Ti:sapphire ring laser. The intrinsic spin fluctuations of the medium along the laser direction imparted Faraday rotation fluctuations on the transmitted laser, which were detected by a balanced photoreceiver. The output voltage was amplified, digitized, and fast-Fourier-transformed in real time to obtain the power density of the measured spin noise (in units of nanoradians$^2$/Hz), typically from 0 Hz up to a few MHz for atoms and up to a hundreds of MHz for QDs. A full description of the experimental setup can be found in [2, 3, 17]. To obtain the OSN spectrum, the measured spin noise was integrated over frequency (total spin noise power) and then recorded as a function of the photon energy of the probe beam.

**$^{41}$K vapor.** We used $^{41}$K in a 1 cm thick glass cell containing a significant 200 Torr background of nitrogen buffer gas. Collisional broadening of the fundamental $4S_{1/2} \rightarrow 4P_{1/2}$ (D1 line) and $4S_{1/2} \rightarrow 4P_{3/2}$ (D2 line) optical transitions increased their linewidths to of order 10 GHz, which greatly exceeds their underlying hyperfine structure (~254 MHz) or Doppler width. In this regime these transitions can be considered homogeneously broadened. Spin fluctuations of the 4S electrons generate measurable spin noise when the probe laser is tuned near the D1 or D2 resonance.

**(In,Ga)As QD sample.** We used a 20-layer structure of MBE-grown (In,Ga)As QDs separated by 60 nm GaAs barriers with the QD density of $10^{10}$ cm$^{-2}$ per layer. Thermal annealing for 30 s at 960°C shifted the emission to the sensitivity range of silicon photodetectors. The sample had a low level of background p-type doping. It was mounted on the cold finger of an optical
Simulations. To simulate the single band contribution we use the following functions: $f_i(E) = \gamma_h/((E - E_i)^2 + \gamma^2_{inh})$ for absorption and $\theta_i(E) = -(E - E_i)/((E - E_i)^2 + \gamma^2_{inh})$ for FR (Verdet constant), with $E_i$ the resonance energy and $\gamma_h$ the homogeneous linewidth. For inhomogeneous spectra we convolute the single contribution numerically with a Gaussian having an inhomogeneous spectra $\gamma_{inh}$. So, for OSN spectra in the case of independent spin systems we obtain:

$$\langle \delta^2(E) \rangle = \frac{1}{\sqrt{2\pi\gamma_{inh}}} \int \delta^2_i(E)e^{-E_i^2/(2\gamma_{inh}^2)}dE_i.$$ (4)

Using a temperature dependent $\gamma_h$ in Eq. (4) we calculate the temperature dependence of the total spin noise power at single energy position. Here we use Ref. [18], where the very similar QD structures were studied to define the functional dependence of homogeneous linewidth on temperature. The QD sample provides the confinement energy of $E_c = 89$ meV, defined by energy distance between the QD emission from the ground state and wetting layer. It leads us to the following parameters: $\gamma_0 = 2.6 \mu$eV, $b_1 = 30 \mu$eV and $b_2 = 4.2$ meV in the equation:

$$\gamma_h(T) = \gamma_0 + \frac{b_1}{e^{E_1/k_BT} - 1} + \frac{b_2}{e^{E_2/k_BT} - 1},$$ (5)

where $E_1 = 6$ meV, and $E_2 = 28$ meV.

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FIG. 1: Optical spin noise of common and independent spin systems. Modeled optical spectra of a spin system having a, resolved and b, unresolved spectral lines in absorption. The top panels show the absorption (black) and Faraday rotation (Verdet) spectra (red). The middle and lower panels show the expected optical spin noise (OSN) spectra for the case where the two optical transitions originate from a common spin system (correlated spin fluctuations), or from independent spin systems (uncorrelated fluctuations), respectively.
FIG. 2: Effect of inhomogeneous to homogeneous linewidth relation on optical spin noise of independent spin systems. \(a\), Optically-detected spin noise spectra for spin-dependent optical transitions exhibiting different ratios of inhomogeneous to homogeneous linewidth, \(\varepsilon = \gamma_{\text{inh}}/\gamma_{\text{h}} = 0.1, 1, \text{ and } 10\). \(b\), Evolution of the OSN spectrum with increasing \(\varepsilon\). \(c\), OSN enhancement factor and evolution of the dip depth vs. \(\varepsilon\).
FIG. 3: Optical spin noise from a homogeneously-broadened system: D1 line of $^{41}$K vapor. 

a, The raw spin noise data (power spectral density) from spin fluctuations of the $4S$ electrons in $^{41}$K, acquired as the photon energy of the probe laser was tuned through the D1 resonance at 1.60995868 eV. $T = 110^\circ C$ ($\sim 25\%$ of the probe light is absorbed on resonance). Note that essentially no noise is observed on resonance, as expected for a homogeneously-broadened line. A small transverse magnetic field was applied ($\sim 3$ Gauss) to shift the spin noise to finite frequency.

b, OSN spectrum: the total measured spin noise power as a function of the probe photon energy. The red solid line is the fitting using a single squared FR spectra $\theta_\gamma^2(E)$ with $\gamma = 14.74 \, \mu eV$ (see Methods).
FIG. 4: Optical spin noise from an inhomogeneously-broadened system: ensemble of (In,Ga)As quantum dots containing single hole spins. a, Optical spectra of the (intentionally) induced Faraday rotation, and of the detected total spin noise power, as a function of probe photon energy. Solid lines are the fits using $\theta_i(E)$ (for FR) and $\theta^2_i(E)$ (for OSN) convoluted with Gaussian representing an inhomogeneous broadening of $\gamma_{inh} = 8.5$ meV (see Methods). (As long as the homogeneous linewidth is much narrower than the inhomogeneous broadening, it has negligible effect on the final result.) b, Temperature dependence of the total spin noise power measured at the maximum of absorption band (1.406 eV), which decreases with temperature as the homogeneous linewidth $\gamma_h$ of the individual quantum dots grows. Squares and points represent two sets of data taken on the same sample at different points. Solid line is a plot using the modeling procedure presented in the Methods section.