Optical Spectroscopy of Spin Noise

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Spontaneous fluctuations of the magnetization of a spin system in thermodynamic equilibrium (spin noise) manifest themselves as noise in the Faraday rotation of probe light. We show that the correlation properties of this noise over the optical spectrum can provide clear information about the composition of the spin system that is largely inaccessible for conventional linear optics. Such optical spectroscopy of spin noise, e.g., allows us to clearly distinguish between optical transitions associated with different spin subsystems, to resolve optical transitions that are unresolvable in the usual optical spectra, to unambiguously distinguish between homogeneously and inhomogeneously broadened optical bands, and to evaluate the degree of inhomogeneous broadening. These new possibilities are illustrated by theoretical calculations and by experiments on paramagnets with different degrees of inhomogeneous broadening of optical transitions [atomic vapors of ^{41}K and singly charged (In,Ga)As quantum dots].

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Optical probes of spin and magnetization dynamics enjoy broad applications across the many atomic, impurity-doped, semiconductor, and metallic systems in which spin-orbit interactions allow coupling between spin polarization and the optical (circular) polarization of light [1–4]. Experimental techniques include powerful polarization-based spectroscopies of optical Faraday or Kerr rotation, circular dichroism, and circularly polarized photoluminescence [5]. Typically, the signals of circular anisotropy in these experimental approaches are proportional to some intentionally induced changes in the magnetization or spin polarization of the sample. In 1981, however, it was demonstrated that the spontaneous and random fluctuations of spin polarization, which naturally occur in thermal equilibrium, can also be detected by sensitive optical Faraday-rotation methods [6]. In accord with the fluctuation-dissipation theorem, the frequency spectrum of this intrinsic "spin noise" (SN) should correspond to that of the linear susceptibility of the system [7] and, therefore, in the radio-frequency range, should contain the same information about magnetic and dynamic properties of the spin system as does its magnetic resonance spectrum. More recently, Faraday-rotation-based SN spectroscopy was considerably developed [8,9] and is becoming increasingly popular in experimental studies of atomic [10,11] and solid-state paramagnets, including semiconductors [12–14] and low-dimensional nanostructures [15–17].

Here, we demonstrate that the SN-related fluctuations of the Faraday rotation (FR) probed at different optical wavelengths may be either correlated or not, depending on whether or not they are contributed by the same spins. We show that this fact can be revealed in the optical spectra of the FR noise, which implies measuring the total spin noise power as a function of the probe light wavelength. In what follows, these spectra will be referred to as optical spin noise (OSN) spectra, which are related to conventional time-averaged FR spectra in a nontrivial way. In particular, FR fluctuations do not necessarily vanish at optical wavelengths where the conventional time-averaged FR itself is zero. The use of the probe wavelength as a tunable parameter establishes this technique as *optical spectroscopy of spin noise*. We show that this approach makes it possible, in certain cases, to reveal the structure of optical transitions hidden in the linear optical spectra and thus opens new possibilities for noise-based techniques. We consider model situations wherein OSN spectra can distinguish or resolve optical transitions associated with the same or different spin subsystems. These situations include strongly inhomogeneously broadened optical transitions, where the differences between conventional FR and fluctuating FR noise spectra are revealed in the most spectacular form.

These new capabilities are related to the stochastic nature of the optical response, which lifts the restrictions of conventional linear spectroscopy that nominally forbid penetrating into the "anatomy" of optical transitions. A related possibility of measuring homogeneous linewidths within an inhomogeneously broadened ensemble by linear Rayleigh scattering was described in Ref. [18], where the effect is caused by spatial, rather than temporal, inhomogeneities of the system.

A material's Verdet constant $V(E)$ characterizes how much optical Faraday rotation is induced by the material

per unit of thickness and per unit of applied magnetic field. $V(E)$ typically depends strongly on the photon energy E of the probe light. Being sensitive to the difference between right- and left-circular indices of refraction, $V(E)$ typically exhibits a dispersive line shape near an isolated spin-sensitive optical transition, passing through zero at the line center. When using FR to detect spin noise, it might therefore seem sensible to tune the probe photon energy to the peak of $V(E)$ and to avoid energies where $V(E) \to 0$. We will show, however, that this simple reasoning, generally, does not work.

The optical spectrum of a paramagnetic material, whose ground state is the spin system of interest, is in general composed of multiple bands. Each band makes a contribution $\theta_i(E)$ to the total Faraday rotation $\theta(E)$:

$$\theta(E) = \sum_i \theta_i(E) \sim \sum_i V_i(E). \tag{1}$$

Here, $V_i(E)$ is the corresponding partial Verdet constant of the paramagnet. Note that we are considering only the paramagnetic contributions to $\theta_i(E)$ that are directly connected with the magnetization of the spin system [3,19].

We focus on the quantity $\langle \delta\theta^2(E) \rangle$, which is the mean-squared fluctuation of the FR due to intrinsic, random spin fluctuations. It may contain fluctuating contributions from multiple spectral bands. When calculating the spectral dependence of $\langle \delta\theta^2(E) \rangle$, we must distinguish between two fundamentally different situations.

In the first case, we deal with a homogeneous spin ensemble, where the ensemble's optical spectrum coincides with that of a single quantum system. Then, FR fluctuations contributed by different bands occur synchronously and are correlated, and the quantity $\langle \delta\theta^2(E) \rangle$ will vary with E as the mean square of the sum of all the partial contributions, from which interference terms between the bands may arise:

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

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

$$\langle \delta\theta^2(E) \rangle \sim \sum_i [V_i(E)]^2. \tag{3}$$

Crucially, this distinction is significant for closely spaced bands and becomes very important when an overlapping multitude of unresolved spectral components exists (such as for strongly inhomogeneously broadened optical spectra like that from quantum dot ensembles).

Figure 1 demonstrates this distinction for modeled optical spectra containing two well-resolved [Fig. 1(a)] and two unresolved [Fig. 1(b)] homogenously broadened bands [20]. Shown are the absorption spectra, the conventional time-averaged FR spectra, and the associated OSN spectra, calculated using Eqs. (2) and (3), for the cases when the two bands originate from a common spin system or from two independent spin systems. Note that the conventional FR spectra calculated using Eq. (1) remain the same whether the spin systems are common or independent—standard linear optics cannot distinguish between these two cases. In marked contrast, the corresponding OSN spectra are fundamentally different: $\langle \delta\theta^2(E) \rangle$ can be large between the two resonances for the case of independent spin systems (bottom curves), even though the conventional time-averaged FR spectrum $\theta(E)$ vanishes at this energy. If the individual bands originate from two independent spin systems, then one can have a situation in which the two transitions are not resolved in absorption or in conventional FR spectra, but are resolved in the OSN spectrum [see Fig. 1(b)]. The reason for that is that their spin fluctuations are uncorrelated and do not cancel. In this case, optical spectroscopy of spin noise can show higher resolution than conventional optical spectroscopy.

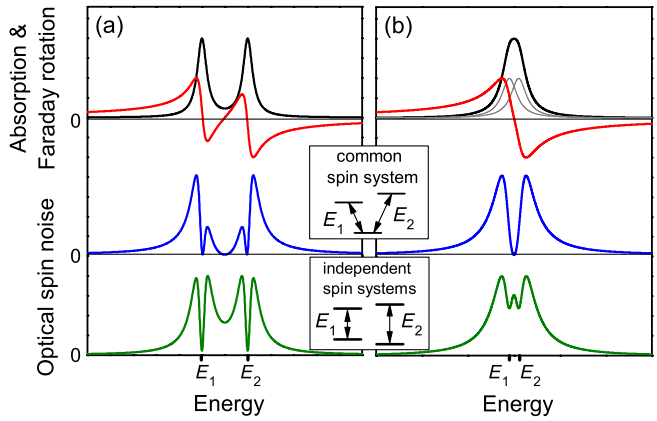

FIG. 1 (color online). Modeled optical spectra of a spin system having two (a) resolved and (b) unresolved absorption lines. The top panels show the absorption (black lines) and conventional Faraday-rotation (Verdet, red/gray lines) spectra. The middle and lower panels show the calculated optical spectra of spin noise when the two transitions originate from a common spin system (correlated spin fluctuations) or from independent spin systems (uncorrelated fluctuations), respectively.

These examples show that optical spectroscopy of spin noise, which uses only weak optical fields, opens clear access to information lying beyond the potential of conventional linear optics. A fascinating illustration of this fact is provided by considering the OSN spectra for a collection of independent spin systems, each with homogeneous Lorentzian linewidth γ_h, distributed in energy following, e.g., a Gaussian with inhomogeneous linewidth γ_{inh}. (Note that the particular shape of the inhomogeneous distribution is of no importance here.) We consider the evolution of OSN spectra as the ratio $\varepsilon = \gamma_{\text{inh}}/\gamma_h$ varies,

that is, as the degree of inhomogeneous broadening changes. For ease of direct comparison, we fix the sum $\gamma_h + \gamma_{\text{inh}}$, which approximately fixes the width and amplitude of the total absorption spectrum. The absorption, conventional FR, and OSN spectra are calculated for $\varepsilon = 0.1$, 1, and 10 and shown in Fig. 2(a). The OSN spectrum is computed as

$$\langle \delta\theta^2(E) \rangle = \frac{1}{\sqrt{2\pi\gamma_{\text{inh}}^2}} \int \theta_h^2(E - E') e^{-E'^2/(2\gamma_{\text{inh}}^2)} dE', \quad (4)$$

where $\theta_h(E) = -E/[E^2 + \gamma_h^2]$ is the FR associated with an individual optical transition. Although the absorption and conventional FR spectra remain qualitatively unchanged as ε increases, the OSN spectrum changes dramatically [21]. When $\varepsilon \ll 1$ and the optical transition is mostly homogeneously broadened—which corresponds to the alkali vapors measured below—the OSN spectrum approximately follows the average FR squared, or $\theta_h^2(E)$. In this case, both situations (common and independent spin systems) show a pronounced dip at the band center. However, as inhomogeneous broadening increases, the dip becomes shallower, eventually disappearing when $\varepsilon > 1$. For strong inhomogeneous broadening ($\varepsilon \gg 1$), the OSN spectrum becomes similar to that of the absorption spectrum, i.e., with a maximum at the band center, even though the average FR is zero. This latter scenario corresponds well to the experimental situation in quantum dot ensembles (also shown below).

Interestingly, the area under the OSN spectrum increases rapidly for larger ε [see Fig. 2(b)], even though the absorption band's total oscillator strength remains constant. Qualitatively, this marked enhancement can be understood by noting that the probe becomes much more sensitive to fluctuations from individual resonances at small detuning when γ_h is small, since the magnitude of $\theta_h^2(E)$ diverges as $\gamma_h \to 0$ for either positive or negative detuning [see Eq. (4)]. [Note that conventional FR spectra do not show a similar dramatic enhancement, since time-averaged FR signals $\theta_h(E)$ from resonances at positive detuning are largely canceled by those at negative detuning.] We define this "enhancement factor" as the ratio of the spectrum's area normalized by the area of the corresponding spectrum for the case $\varepsilon \to 0$. Using Eq. (4), one can see that [21]

$$\int \langle \delta\theta^2(E) \rangle dE = \int \theta_h^2(E) dE = \frac{\pi}{2\gamma_h} = \frac{\pi}{2} \frac{\varepsilon + 1}{\gamma_h + \gamma_{\text{inh}}}. \quad (5)$$

Taking into account that $\gamma_h + \gamma_{\text{inh}}$ is assumed to be constant, we obtain that the enhancement factor defined above equals $1 + \varepsilon$ regardless of the type of inhomogeneous distribution. It characterizes how strongly OSN signals from an inhomogeneously broadened system can deviate from a fully homogeneous system. As shown below, this has direct bearing on real quantum dot experiments, where variations in the measured spin noise directly reveal changes in γ_h of the underlying dots within the ensemble.

Figure 2(c) summarizes these findings and shows the evolution of the central dip and the enhancement factor versus ε. Any observed central dip in OSN spectra can be used to quantify γ_h, and variations of the enhancement factor can be used to evaluate changes in γ_h, even in strongly inhomogeneously broadened systems. For comparison, the dashed line in Fig. 2(c) represents the dependence of FR squared spectra [$\langle \delta\theta(E) \rangle^2$] versus ε calculated similarly to Eq. (4) and demonstrating only a small enhancement [21].

To validate and illustrate this model, we studied OSN spectra from atomic vapors and from singly charged semiconductor quantum dots (QDs)—two spin systems corresponding to opposite limits of inhomogeneous broadening ($\varepsilon \ll 1$ and $\varepsilon \gg 1$, respectively). Following Refs. [8,16], these SN measurements utilized a weak, linearly polarized beam from a cw Ti:sapphire ring laser. FR fluctuations were detected by a balanced photoreceiver whose output voltage was amplified, digitized, and processed in real time to obtain the noise power density in units of nanoradians^2/Hz up to a few MHz for atoms and up to hundreds of MHz for QDs. The measured SN was integrated over frequency to obtain the total SN power and then recorded as a function of the probe laser's photon energy to generate the OSN spectrum.

For the case of predominantly homogeneous broadening ($\varepsilon \ll 1$), we studied a warm (110 °C) atomic vapor of ^{41}K [22]. Figure 3(a) shows how the raw spin noise data vary as

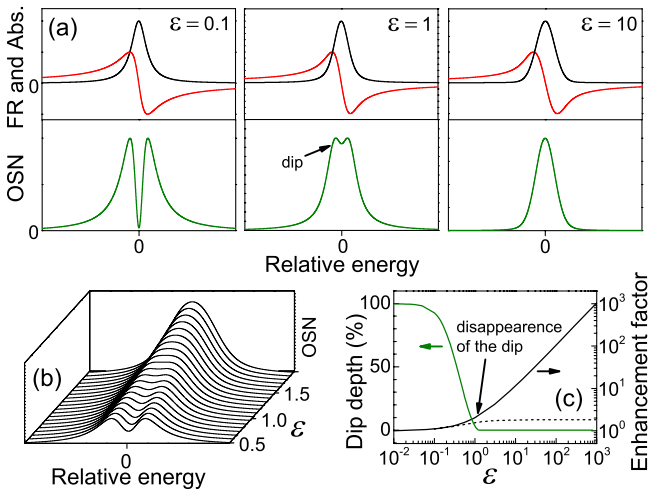

FIG. 2 (color online). (a) Calculated and normalized absorption, conventional FR, and OSN spectra for spin-dependent optical transitions exhibiting different ratios of inhomogeneous to homogeneous linewidth $\varepsilon = \gamma_{\text{inh}}/\gamma_h = 0.1$, 1, and 10. (b) Evolution of the OSN spectrum with increasing ε. For ease of comparison, $\gamma_h + \gamma_{\text{inh}}$ was fixed. (c) Enhancement factor and dip depth versus ε. The solid lines correspond to the OSN case, and the dashed line corresponds to FR squared.

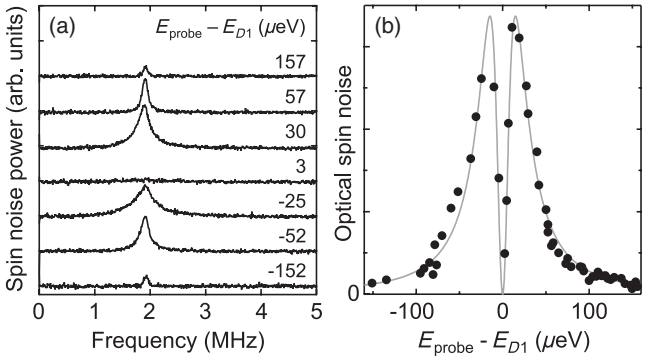

FIG. 3. (a) The raw spin noise power spectral density from fluctuations of $4S$ electron spins in ^{41}K, at different detunings of the probe laser from the $D1$ optical transition (1.6106 eV). $T = 110\,^{\circ}$C (~25% absorption on resonance). A 3 G transverse magnetic field shifts the spin noise to ~2 MHz. Essentially, no noise is observed on resonance, as is expected for a homogeneously broadened transition. (b) The OSN spectrum: the total spin noise power versus probe photon energy. The line is a fit to a single squared FR spectrum $\theta_h^2(E)$ with $\gamma_h = 14.7$ μeV.

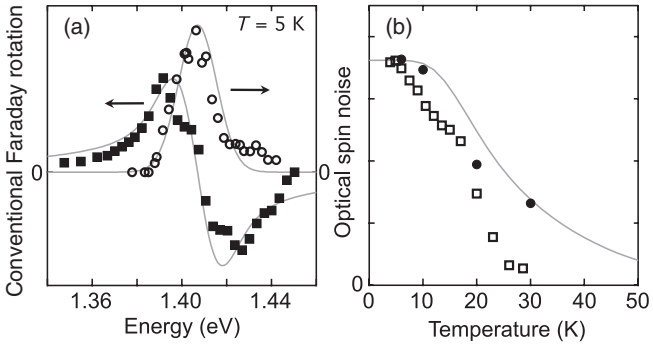

FIG. 4. (a) Squares and circles show the conventional FR spectrum and the OSN spectrum from an inhomogeneously broadened ensemble of (In,Ga)As QDs that are singly charged by holes. The lines are fits using $\theta_h(E)$ and $\theta_h^2(E)$, respectively, convolved with an inhomogeneous Gaussian broadening of $\gamma_{inh} = 8.5$ meV [see Eq. (4)]. (b) The total spin noise power (measured at 1.406 eV) increases dramatically as temperature drops and the homogeneous linewidth γ_h of the individual QDs in the ensemble decreases. The squares and circles are two data sets taken at different locations on the sample. The line is a simulation of Eq. (6).

the probe laser is tuned through the spin-sensitive $D1$ ($4S_{1/2} \rightarrow 4P_{1/2}$) optical transition. Integrating the total noise power gives the associated OSN spectrum shown in Fig. 3(b). As expected [see Fig. 2(a)], it exhibits a well-pronounced dip at the line center. This was also seen in recent studies of ^{87}Rb [23].

For the opposite regime of strong inhomogeneous broadening, we measured OSN spectra from ensembles of hole-doped (In,Ga)As QDs [24]. At low temperatures, individual QDs exhibit sharp optical transitions as narrow as several μeV; however, the distribution of QD sizes and compositions leads to a broad ~20 meV inhomogeneous linewidth in ensemble measurements. Optical transitions from single resident holes to positively charged trions are right or left-circularly polarized, depending on the hole's initial spin orientation. Intrinsic spin fluctuations of holes are therefore measurable [16,17].

Figure 4(a) shows the OSN spectrum of these holes as the probe laser was tuned through the absorption band of the QD ensemble. Also shown for comparison is the conventional FR spectrum, obtained by intentionally polarizing the hole spins with circularly polarized pump light (at 1.579 eV) and detecting the induced time-averaged FR as a function of probe photon energy. As predicted for inhomogeneously broadened transitions [see Fig. 2(a)], the OSN spectrum does not exhibit any dip at the line center but rather achieves a maximum, even though the conventional FR $\theta(E)$ passes through zero.

Finally, the utility of the enhancement factor—meaning the sensitivity of spin noise to the underlying homogeneous linewidth of the QDs in the ensemble—is demonstrated by the temperature dependence of the measured noise. It is known from nonlinear four-wave mixing studies [25] that γ_h is strongly temperature dependent in epitaxial QDs, even at low temperatures (whereas γ_{inh} does not change in this range). At a fixed probe energy (1.406 eV, the maximum of the OSN spectrum), Fig. 4(b) shows that the measured noise power increases dramatically as temperature drops from 30 to 4 K and γ_h decreases, even though no significant changes occur in the conventional absorption or FR spectra. This confirms the prediction that spin noise from an inhomogeneously broadened system is enhanced as γ_{inh}/γ_h increases [see Fig. 2(c)]. Following Ref. [25],

$$\gamma_h(T) = \gamma_0 + \frac{b_1}{e^{E_1/k_BT} - 1} + \frac{b_2}{e^{E_2/k_BT} - 1}, \qquad (6)$$

where we consider QDs with confinement energy 89 meV, a lowest-temperature homogeneous linewidth $\gamma_0 = 2.6$ μeV, activation energies $E_1 = 6$ meV and $E_2 = 28$ meV, and coefficients $b_1 = 30$ μeV and $b_2 = 4.2$ meV. Using $\gamma_h(T)$ in Eq. (4), we calculate the temperature dependence of the total SN power, which is shown by a line in Fig. 4(b). Here, $\gamma_h(T)$ is changing from 5.9 μeV at 30 K down to 2.6 μeV at 4 K. The initially calculated amplitude at the lowest temperature was scaled to the measured OSN amplitude. A good agreement with experiment is achieved without further fitting parameters.

Thus, spin noise measurements provide a powerful experimental tool for penetrating into the internal structure of strongly inhomogeneously broadened systems (like QD ensembles) and for revealing variations of γ_h that may otherwise be obscured. It is especially noteworthy that this sensitivity exists even when $\gamma_{inh} \gg \gamma_h$, making it possible to realize an effectively high (sublinewidth)

spectral resolution by means of the linear optical technique of spin noise.

In summary, we attract attention to the fact that mutual correlations of the FR noise at different probe wavelengths may serve as a specific source of information about the composition of complex spin systems. These correlations are not revealed in conventional optical absorption or FR spectra but can reveal themselves in the OSN spectra and are sensitive to the correlation characteristics of spin fluctuations. As a result, optical spectroscopy of spin noise can provide important information that is otherwise difficult to access via conventional linear optical spectroscopy. We illustrate capabilities of this technique by modeling OSN spectra and by experimental measurements of spin systems with either negligibly small or with very large inhomogeneous broadening of optical transitions—4S electrons in ^{41}K atoms and resident holes in (In,Ga)As QD ensembles, respectively. We believe that the proposed approach considerably widens the area of applicability of spin noise spectroscopy.

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[20] To simulate the single band contribution, we used $f_i(E) = \gamma_h/[(E - E_i)^2 + \gamma_h^2]$ for absorption and $\theta_i(E) = -(E - E_i)/[(E - E_i)^2 + \gamma_h^2]$ for FR (Verdet) spectra, with E_i being the resonance energy and γ_h the homogeneous linewidth.
[21] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.110.176601 for analytical solution for enhancement factor in spin noise and comparison with Faraday rotation spectroscopy.
[22] We used ^{41}K in a 1 cm thick glass cell with 200 Torr of the 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 around 10 GHz, which greatly exceeded their underlying hyperfine structure (~254 MHz) and Doppler width. In this regime, these transitions can be considered to be homogeneously broadened. Spin fluctuations of the 4S electrons generate measurable spin noise when the probe laser is tuned near the $D1$ or $D2$ resonance.
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