Spin Noise in Birefringent Media

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It is known that linear birefringence of the medium essentially hinders measuring the Faraday effect. For this reason, optically anisotropic materials have never been considered as objects of the Faraday-rotation-based spin noise spectroscopy. We show, both theoretically and experimentally, that strong optical anisotropy that may badly suppress the regular Faraday rotation of the medium, practically does not affect the measurement of the spatially uncorrelated spin fluctuations. We also show that the birefringent media provide additional opportunity to measure spatial spin correlations. Results of the experimental measurements of the spin-noise spectra performed on Nd^{3+} ions in the uniaxial crystal matrices well agree with the theory.

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Introduction.—In the past decade, the spin noise spectroscopy (SNS) has gained an important place in the experimental physics of magnetic resonance. Being primarily demonstrated on atomic systems [1], this technique is, at present, widely used in studies of electron and nuclear spin systems in semiconductors. The SNS was applied to GaAs electronic system both in thermal equilibrium [2] and beyond it [3], to describe the spin dynamics in quantum dots [4,5] and quantum wires [6,7], to reveal nuclear bath effect [8–12]. In the last decade, a number of works expanding the experimental boundaries of SNS were presented (i.e., optically resolved SNS [13], ultrahigh bandwidth detection [14], spatiotemporal spin fluctuation imaging [15]), as well as several fundamental reviews in this area [16,17]. Recently, SNS has been successfully applied to dielectric crystals with paramagnetic impurities [18–20]. As compared with the conventional electron paramagnetic resonance (EPR) spectroscopy, the SNS offers a number of new specific possibilities of research and reveals many features more typical for nonlinear optics, such as manifestation of Z-scan-type sensitivity [21–23] and probing the resonant optically induced spin effects [24,25].

At the same time, it might seem that the SNS, based on detection of the Faraday rotation (FR) noise, should have inherited the main features (both merits and drawbacks) of the FR method. Specifically, the FR measurements are known to be highly sensitive to linear birefringence of the medium [26–29], and the strong optical anisotropy leads to almost total canceling of static FR [30]. This circumstance, for a long time, prevented application of the SNS to optically anisotropic crystals.

In this Letter, we consider the effect of linear birefringence on polarization of the transmitted light and come to a curious result that while the polarization state of the transmitted light may be dramatically distorted by the anisotropic medium, its fluctuations, associated with spatial fluctuations of gyrotropy of the medium, remain practically intact.

The fact that the linear birefringence affects the spatially uniform and spatially fluctuating FR (gyration) in a strongly different way, can be supported by the following reasoning. Polarization of the light traveling through a birefringent medium exhibits spatial oscillations, with the σ_+ and σ_- components being periodically interchanged. As a result, the sign of the FR is periodically inverted so that the total FR appears to be close to zero. For the spatially uncorrelated gyration, this mechanism of the FR suppression does not work, because any inversion of the random FR leaves it random, and the measured FR noise remains the same.

In this work, we present a rigorous theoretical description of the effect of linear birefringence upon the FR noise detected in the SNS experiments. We also show that the FR noise signal, in a uniformly birefringent medium, may be enhanced or suppressed for spin arrangement spatially correlated at the wave vector of the intrinsic polarization beats of the birefringent medium. Applicability of the SNS to birefringent media is illustrated by experimental study of spin noise in rare-earth-doped crystals.

Theory.—We consider the light propagation along one of the principal axes (z) of a birefringent crystal, with two other principal axes x and y being characterized by the dielectric constants ε_x and ε_y, respectively. The case of the light propagation in an arbitrary direction can be considered in a similar way (see Discussion below). The Fourier component of electric field of the probe light at the frequency ω is given by

$$E_{x,y}^{(0)}(z) = E_{x,y}^{(0)}(0)\exp(ik_{x,y}z), \quad (1)$$

where $k_{x,y} = k_0\sqrt{\varepsilon_{x,y}}$ are the wave vectors of the light components polarized along the corresponding axes, $k_0 = \omega/c$, and the sample face is located at $z = 0$.

The spin fluctuations in the crystal provide a stochastic polarizability [31] with the off-diagonal components $\chi(z) = \chi_{xy} = -\chi_{yx}$, which are odd under time reversal [35]. The stochastic polarizability produces the following additional polarization in the sample:

$$\Pi_{x,y}(z) = \pm\chi(z)E_{y,x}^{(0)}(0)e^{ik_{y,x}z}, \quad (2)$$

The polarization, in turn, produces a scatted field, which at the face of the sample of the length L takes the form [31,36]

$$E_{x,y}^{(1)}(L) = \pm\frac{2\pi i k_0^2}{k}E_{y,x}^{(0)}(L)\chi_\pm, \quad (3)$$

where we assume $|k_x - k_y| \ll k$, $k = (k_x + k_y)/2$, and

$$\chi_\pm = \int_0^L dz\,\chi(z)e^{\pm i(k_x-k_y)(L-z)} \quad (4)$$

are the two spatial harmonics of the fluctuating polarizability induced by the spin noise.

Generally, the linearly polarized light, after passing through the birefringent medium, becomes elliptically polarized. The polarization is described by the Stokes parameters ξ_1, ξ_2, and ξ_3, which fluctuate due to the spin noise in the sample. We define the generalized FR and ellipticity signals as [36]

$$\mathcal{F} = \frac{\langle\xi_3\rangle\delta\xi_1 - \langle\xi_1\rangle\delta\xi_3}{\sqrt{\langle\xi_1\rangle^2 + \langle\xi_3\rangle^2}}, \qquad \mathcal{E} = \frac{\delta\xi_2}{\sqrt{1-\langle\xi_2\rangle^2}}, \quad (5)$$

respectively, where $\langle\xi_i\rangle$ with $i = 1, 2, 3$ are the average Stokes parameters and $\delta\xi_i = \xi_i - \langle\xi_i\rangle$ are their deviations proportional to $E_{x,y}^{(1)}$ and to the stochastic spin polarization. Generally, on the Poincaré sphere, $\mathcal{F}$ and $\mathcal{E}$ represent fluctuations of the Stokes vector in the equatorial and meridional directions, respectively, as shown schematically in Fig. 1 at the point A.

To calculate the noise intensities of $\mathcal{F}$ and $\mathcal{E}$, we neglect the spatial correlations in the fluctuating $\chi(z)$. It is convenient to introduce a reduced real-valued spin polarization in the sample $m(z)$ according to $\chi(z) = Xm(z)$, where X is a complex coefficient [31] and the normalization condition for $m(z)$ reads

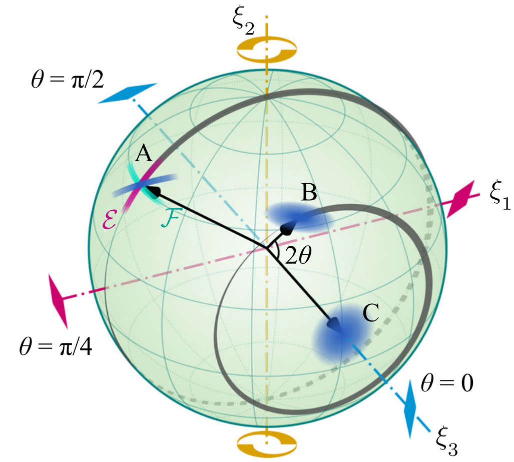

FIG. 1. Schematic presentation of the Stokes vector fluctuations for the light transmitted through a birefringent crystal. The axis ξ_3 corresponds to the polarizations along the main axes. The points A, B, and C, on the surface of the Poincaré sphere correspond to azimuths of the incident light polarization θ equal to $\pi/4$, approximately $\pi/8$, and 0. Conversion of the arc segment at A to the round spot at C indicates transformation of correlated FR and ellipticity noises to uncorrelated ones.

$$\langle m(z)m(z')\rangle = \delta(z - z'). \quad (6)$$

Typically, at the optical resonances $X \propto 1/[\gamma + i(\omega_0 - \omega)]$, where ω_0 is the resonance frequency and γ is its width [17]. Generally, one can directly express the intensities of the noise $\langle\mathcal{F}^2\rangle$ and $\langle\mathcal{E}^2\rangle$ through the coefficient X and the dimensionless parameter $(k_y - k_x)L$.

Let us consider the limit of strong anisotropy, $q = k_x - k_y \gg 1/L$, assuming the absorption anisotropy (imaginary part of $k_x - k_y$) to be zero. From the definitions (4) and (6) we obtain the following correlators of the polarizabilities [31]: $\langle\chi_+\chi_-\rangle = X^2L$, $\langle\chi_+^*\chi_-^*\rangle = X^{*2}L$, $\langle\chi_+\chi_+^*\rangle = \langle\chi_-\chi_-^*\rangle = |X^2|L$, all the other correlators between $\chi_\pm$ and $\chi_\pm^*$ are zero. Substituting these expressions in Eqs. (3) and (5) we obtain the total noise of the FR and ellipticity [31]:

$$\langle\mathcal{F}^2\rangle + \langle\mathcal{E}^2\rangle = \frac{1}{2}\mathcal{Q}|X|^2L[3 + \cos(4\theta)], \quad (7)$$

where $\mathcal{Q} = 2(2\pi k_0^2/k)^2$ and θ is the angle between the incident light polarization plane and the x axis.

In the opposite limit of negligible birefringence, we obtain in the same way [31] $\langle\mathcal{F}^2\rangle + \langle\mathcal{E}^2\rangle = 2\mathcal{Q}|X|^2L$. Thus, the birefringence suppresses the total polarization noise only 1–2 times in contrast with the regular Faraday effect suppressed by the factor $qL \gg 1$.

For the incident light polarized at $\theta = \pi/4$, we have $\langle\mathcal{F}^2\rangle = \mathcal{Q}(\mathrm{Im}X)^2L$ and $\langle\mathcal{E}^2\rangle = \mathcal{Q}(\mathrm{Re}X)^2L$ [31], so that the FR and ellipticity noises are proportional to the regular FR and ellipticity in isotropic medium squared. Moreover, they

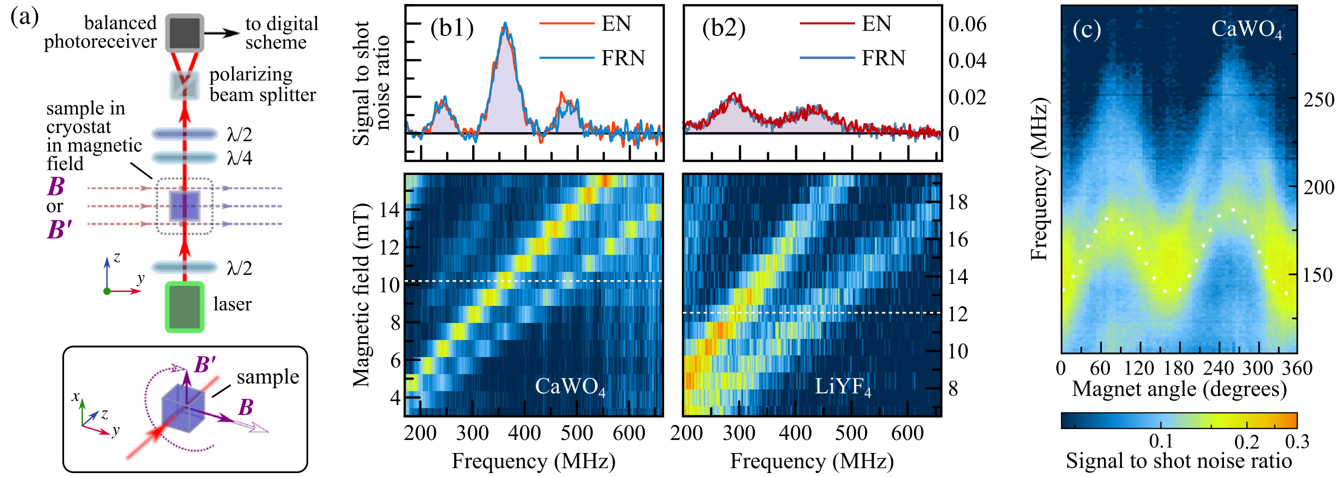

FIG. 2. (a) Schematic of the experimental setup. The inset shows two possible magnetic field geometries (fixed direction $\boldsymbol{B}$ or fixed amplitude rotating $\boldsymbol{B}'$). Panel (b) shows magnetic-field dependences of the FR noise (lower pictures) and examples of particular records of the FR and ellipticity noise spectra (upper plots) for the Nd-doped $CaWO_4$ (b1) and $LiYF_4$ (b2) crystals. (c) Orientational dependence of the FR noise spectra of the $CaWO_4:Nd^{3+}$ crystal. Magnetic field created by the permanent magnet was rotated in the plane normal to the laser beam. Both resonances follow the same orientational pattern. The low-frequency resonance presumably corresponds to the known tetragonal neodymium center (accentuated with a dotted line).

are completely correlated, $\langle \mathcal{F}\mathcal{E}\rangle^2 = \langle \mathcal{F}^2\rangle\langle \mathcal{E}^2\rangle$. On the Poincaré sphere, the FR and ellipticity fluctuations represent an arc, as shown in Fig. 1 at the point A. These results are standard for the optical SNS [17].

By contrast, if the probe beam is initially polarized along one of the principal axes (e.g., $\theta = 0$), the FR and ellipticity noise intensities are equal, $\langle \mathcal{F}^2\rangle = \langle \mathcal{E}^2\rangle$, and they are uncorrelated, $\langle \mathcal{F}\mathcal{E}\rangle = 0$ [31]. This is illustrated by the blue round spot C in Fig. 1. This is in a stark contrast with the conventional SNS, and this is related to the presence of the two spatial harmonics of the spin noise at the wave vectors $\pm q$, as defined by Eq. (4).

For the intermediate cases of the light polarization azimuth ($0 < \theta < \pi/4$), the polarization noise on the Poincaré sphere represents an elliptical spot smoothly transforming from the arc to the disk, as shown in Fig. 1 at the point B. The total intensity of the FR and ellipticity noises is generally described by Eq. (7), and changes as a function of θ no more than by a factor of 2.

The above theoretical treatment gives a general idea of the behavior of the FR noise in a birefringent medium. We see that the birefringence affects, in a certain way, behavior of the FR and ellipticity noises, but the most important result is that the total polarization noise is not suppressed in magnitude as it occurs with the regular magneto-optical effects.

Experimental.—To verify basic results of our theoretical treatment, we have chosen two uniaxial crystals, $CaWO_4$ and $LiYF_4$, activated by Nd^{3+} ions. An additional advantage of these crystals is that they contain tetragonal Nd^{3+} centers aligned along the crystal axis, that are magnetically equivalent in the external magnetic field [37,38].

A schematic of the optical arrangement with some additional explanations is presented in Fig. 2(a). The quarter-wave plate placed after the sample was used to transform ellipticity noise unto that of the polarization plane rotation detected by the standard polarimeter.

The plates of the crystals $CaWO_4:Nd^{3+}$ (1 at.%), 1.54 mm thick, and $LiYF_4:Nd^{3+}$ (~0.5 at.%), 1.22 mm thick, were placed inside a cryostat at a temperature of 3 K. The quantity qL, for these samples, was ~27 (with the birefringence $\Delta n \approx 0.016$ [39]) for calcium tungstate and ~32 (with the birefringence $\Delta n \approx 0.022$ [40]) for $LiYF_4$, respectively, that well meets approximation of the above treatment ($qL \gg 1$). Magnetic field on the sample was directed across the light beam and could be varied in magnitude [$\boldsymbol{B}$ in Fig. 2(a)]. Alternatively, for measuring angular dependencies of the spin noise spectra, we used (as in Ref. [18]) a rotating permanent magnet that could create the magnetic field $\boldsymbol{B}'$ rotating either around the laser beam [shown in the inset of Fig. 2(a)] or around the axis normal to the laser beam (not shown).

As a light source, we used a frequency-stabilized ring-cavity Ti-sapphire laser with an extremely narrow emission spectrum needed to detect spin noise of RE ions in crystals [18]. The laser beam was tuned in resonance with the low-energy component of the transition $^4I_{9/2}$–$^4F_{3/2}$ of Nd^{3+} ion characterized by a relatively small homogeneous width [41].

Note that our experiments were aimed at measuring amplitudes and positions of the spin noise peaks and practically ignored information contained in the line shape of the resonances [42–44], which deserves special consideration.

In the simplest polarization scheme, the light polarization is aligned along an anisotropy axis of the crystal, and the magnetic field is directed across the light beam. In this case, the light beam, the magnetic field, and the optic axis of the crystal are perpendicular to each other. In our measurements, we have immediately found that, indeed, optical anisotropy of the crystal had no substantial effect upon the polarization noise signal, and the spin noise spectra of Nd^{3+} ions could be detected in both crystals approximately with the same sensitivity as in the cubic CaF_2 crystal studied in [18].

Results of these measurements are shown in Figs. 2(b) and 2(c). Panels (b1) and (b2) show magnetic-field dependences of the FR noise spectra (lower pictures) and examples of the FR and ellipticity noise spectra (upper plots) for the Nd-doped $CaWO_4$ and $LiYF_4$ crystals, respectively. Figure 2(c) shows the orientational dependence of the FR noise spectra of the $CaWO_4$-Nd^{3+} crystal obtained in the fixed amplitude magnetic field which rotated in the plane orthogonal to the light beam [as shown in Fig. 2(a)]. The above results allowed us to ascribe the strongest peaks of the FR noise spectrum in the $CaWO_4$ and $LiYF_4$ crystals to the tetragonal Nd^{3+} centers with g-tensor components $g_\parallel = 2.03$, $g_\perp = 2.54$ [37] and $g_\parallel = 1.987$, $g_\perp = 2.554$ [45], respectively.

Discussion.—As seen from Fig. 2(b) (upper plots), the FR and ellipticity noise signals, for the probe beam polarized along the optic axis of the crystal (at $\theta = 0°$), are the same in magnitude in full agreement with the theoretical prediction. Moreover, our measurements performed at $\theta = 0°$, 45°, and 90° have shown that, within the experimental accuracy, the signals of the FR and ellipticity noise are universally equal to each other. This is related to strong inhomogeneous broadening of the optical resonance.

Indeed, in the cases of both Pauli-blocking SNS [36] and optical resonance shift SNS [46], when $X \propto 1/(\omega - \omega_0 + i\gamma)$ and $X \propto 1/(\omega - \omega_0 + i\gamma)^2$, respectively, we find that $(\mathrm{Re}X)^2$ and $(\mathrm{Im}X)^2$ averaged over ω_0 coincide. This conclusion is additionally supported by the non-Lorentzian line shapes of the spin noise spectra [42–44], which reveal the spread of the g factors.

Importantly, the main results of the treatment remain valid for arbitrary light propagation direction, with the dielectric constants along principal axes of the crystal being replaced by those along the normal-mode polarizations. Thus, by rotating the crystal, one can effectively change its birefringence varying in this way the wave vector q. This gives access to the parameters of the spin system that may reveal themselves in its spatial correlations, like, e.g., the spin diffusion coefficients and the spin-orbit coupling constants. A similar idea was considered in [47] where the wave vectors of the two waves differed by their directions rather than by magnitudes. By the analogy between the linear birefringence, which leads to precession of the Stokes vector, and a magnetic field, one can say that the measurement of the spin noise at the wave vector q is analogous to the measurement of the spin noise component at the Larmor frequency in the electron spin resonance [48].

Since the polarization noise in a birefringent medium is practically insensitive to the magnitude and direction of the birefringence, we may conclude that this noise should not be affected by a spatially nonuniform birefringence, and the SNS may be applicable to materials with spatially inhomogeneous birefringence. This conclusion is correct when magnetic anisotropy of the impurity centers is not coupled to medium anisotropy or when the impurity centers are magnetically isotropic. In most cases, however, inhomogeneous birefringence dramatically affects the spin-noise resonance linewidth, with the conservation law acting only with respect to the integral spin noise power. We will consider the laws of spin-noise power conservation in birefringent media more rigorously elsewhere.

One more effect of linear birefringence on the measured polarization noise can be related to nonmonochromaticity of the probe light. After passing through the sample, this light becomes partially depolarized, but it preserves its ξ_3 Stokes parameter (provided that the medium is homogeneous). In this case, the ellipticity noise is suppressed (for $|\xi_3| \neq 1$), while the FR noise intensity remains the same. This result is interesting, but does not have much practical sense for conventional laser sources with extremely narrow emission spectra.

Thus, we can conclude that the optical spin noise measurements are feasible for a broad class of birefringent media. Importantly, the gyration noise in the birefringent medium does not reveal any pronounced dependence on the polarization plane azimuth.

Conclusions.—In this Letter, we show, both theoretically and experimentally, that the spatially uncorrelated fluctuations of magneto-optical activity, in contrast to the spatially uniform magneto-optical effects, are not affected essentially by linear birefringence of the medium and appear to be practically isotropic with respect to the probe beam polarization. In the case of spatially correlated gyration noise, the detected noise signal may depend on the ratio of its correlation length and the length of polarization beats of the probe light ($1/q$). This fact can be used for studying correlation properties of spin systems. We believe that results of this work considerably strengthen the potential of the SNS.

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