Giant spin-noise gain enables magnetic resonance spectroscopy of impurity crystals

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We apply spin-noise spectroscopy to the classical objects of electron paramagnetic resonance spectroscopy—dielectrics with paramagnetic impurities—which seemed to be unsuitable for this technique because of their low specific Faraday rotation (FR) associated with strong (allowed) optical transitions. We show, however, that by detecting the spin noise at the wavelengths of weak (forbidden) transitions (with low regular FR), the signal can be strongly enhanced, provided that the optical transition is broadened inhomogeneously. This *spin-noise gain effect*, which is controlled by the ratio of the inhomogeneous linewidth to the homogeneous one, relates primarily to the parity-forbidden intraconfigurational transitions of the impurity ions with unfilled inner electronic shells. Specifically, for the f - f transitions of rare-earth ions in crystals, this factor can reach $\sim 10^8$. In this paper, we describe the specific features of the experimental equipment required for the realization of this kind of measurements, present results of successful experiments of spin-noise spectroscopy on rare-earth ions in crystals, and briefly discuss the prospect of applying this method of magnetic resonance spectroscopy in present-day research.

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I. INTRODUCTION

The spectroscopy of magnetic resonance can be applied to virtually all quantum objects possessing angular momentum and, therefore, is one of the key techniques of contemporary physics research. Detection of the magnetic resonance spectrum usually implies observation of the response of a paramagnet (“spin system”) to an AC magnetic field with a frequency close to that of the spin precession. Since the discovery of the magnetic resonance effect by Zavoisky in 1944 [1], two main detection methods were elaborated. One of them uses detection of the changes of the AC field, as done, e.g., in the conventional optical spectroscopies of absorption or reflection. In the second method, the resonance is revealed in the AC-field-induced changes of the object’s properties as done, e.g., in optical pump-probe spectroscopy. However, the principle of resonance detection in both cases is the same: the resonance is observed only through the response to the excitation of spin precession by the external AC field.

In 1981 [2], it was demonstrated experimentally that observation of the magnetic resonance does not necessarily require application of the resonant AC field; the resonance can be observed also by detecting the excess fluctuations of the medium’s refractive index at the spin-precession frequency.

During recent years, this *spin-noise spectroscopy* (SNS) has considerably developed [3–7] and demonstrated some specific capabilities that were not foreseen initially [8]. Specifically, the SNS sensitivity was found to be sufficient to detect the spin noise (SN) of a single charge carrier in a semiconductor structure [9,10], was applied to monitor the nuclear dynamics of the host lattice in a doped semiconductor [11,12], was used as an instrument of high-resolution tomography [13] and as a spectroscopic tool to identify the origin of broadening of optical transitions and to measure their homogeneous width [14,15], etc. As a result, SNS has nowadays turned into an important method of research in atomic and semiconductor physics [16–19].

It should be noticed, however, that this method has so far not been applied to dielectrics with paramagnetic impurities—systems which served as the primary material basis for electron resonance (EPR) spectroscopy and are nowadays widely used in state-of-the-art optics and photonics [20–23]. These objects so far did not look promising for the application of SNS because of their low efficiency of conversion of “magnetization–FR.” This fact was confirmed in Ref. [24] using data on regular Faraday rotation (FR) of crystals with rare-earth (RE) ions normalized to a single spin (the so-called *Faraday-rotation cross section* [25]). We show, however, that the efficiency of the magnetization–FR conversion may drastically differ for regular and stochastic signals.

It is useful to remind one here that the impurities in dielectric materials that are most important for science and applications (such as Cr^{3+} and Nd^{3+} in laser media, or the RE ions in up-converters or quantum counters, in quantum memory schemes, etc.) are usually ions of the transition-metal

FIG. 1. Typical spectra of absorption (a), Faraday rotation (b), and spin-noise power (c) of RE ions in crystals. The left and right panels show the spectra of the strong interconfigurational (homogeneously broadened) $f-d$ transitions and of the weak intraconfigurational (inhomogeneously broadened) $f-f$ transitions, located in the UV and visible/IR spectral regions, respectively. The zoomed-in areas in panels (a) and (b) show schematically the inner (hidden) structure of a single inhomogeneously broadened line (absorption or FR) of the $f-f$ transition. To be precise, we show in the zoomed-in area of panel (b) spectral components with opposite signs, arising due to the smallness of the homogeneous width of the optical transitions compared to their Zeeman splitting.

groups or, in other words, ions with not completely filled inner electronic shells. The optical spectra of these ions are typically characterized by two kinds of transitions: strong, allowed interconfigurational and weak, parity-forbidden intraconfigurational transitions. The former are often characterized by strong magneto-optical activity and therefore considered as the most promising candidates for magneto-optical applications. In the trivalent RE ions, which are the subject of our present work, the strong allowed transitions ($4f^{n-1}5d$) are located in the ultraviolet (UV) spectral range, while the narrow parity-forbidden $4f^n-4f^n$ transitions are observed in the visible and near-infrared ranges (see, e.g., Ref. [26]). A schematic of typical optical absorption and FR spectra of trivalent RE ions in crystals are shown in Figs. 1(a) and 1(b), respectively.

In this paper, we report on the successful application of the spin-noise technique for detecting magnetic resonance in crystals activated by RE ions. We show theoretically and confirm experimentally that the value of the FR fluctuations of a paramagnet cannot be evaluated based on the value of

its regular FR (FR in external magnetic field). On the other side, the FR noise appears to be drastically enhanced for the resonantly probed optical transitions with a high ratio of the inhomogeneous linewidth to the homogeneous one [see Fig. 1(c)]. When applied to the forbidden intraconfigurational transitions of RE ions in crystals, this enhancement (*the spin-noise gain effect*) reaches up to seven to eight orders of magnitude. The proposed experimental approach turns RE-activated crystals into favorable objects of SNS.

II. THEORETICAL CONSIDERATIONS

It was shown in Ref. [14] that the shape of the optical spectrum (wavelength dependence) of the SN power, detected within the optical transition, is strongly affected by inhomogeneous broadening of the transition. In particular, for a homogeneously broadened line, the optical spectrum of the SN power exhibits a dip in the center of the line which gradually disappears with increasing inhomogeneous linewidth. Here, we will consider the case when the inhomogeneous width of the transition strongly exceeds its homogeneous width and will concentrate on the effect of SN gain revealed under these conditions.

Let us consider the formation of the SN signal under these conditions and derive a simple estimate of the strength of the SN signal. Figures 1(a) and 1(b) show schematically absorption and FR spectra of trivalent paramagnetic RE impurities in crystals in the range of interconfigurational ($f-d$) and intraconfigurational ($f-f$) transitions. Let the inhomogeneous and homogeneous linewidths of a single $f-f$ transition be Γ and γ , respectively, as shown in the zoomed-in insets of Figs. 1(a) and 1(b).

It is convenient for our reasoning to choose the wavelength of the probe beam such that the regular FR of the system is essentially different from zero, which is the case somewhere on the slope of the inhomogeneous absorption line. To simplify the calculation, we consider the case of total spin polarization when all spins are oriented along the light propagation direction (say, $S_i = +1/2$ for all i) and the magnitude of the FR φ_s (FR of saturation) is the greatest. Let us express the angle φ_s through the length of the sample d and impurity concentration n (spin density) using the definition of the FR cross section σ introduced in Ref. [25]: $\varphi_s = \sigma nd$. The number of impurity centers within the probe beam volume N is given by $N = ndA$, where A is the beam cross section.

Because of the inhomogeneous broadening of the line, the FR (being itself independent of the homogeneous width γ) is contributed only by the fraction of impurity centers with optical frequencies within an interval of the order of the homogeneous linewidth γ . This number N' , is approximately equal to $N\gamma/\Gamma$, and thus can drastically differ from the total number of impurity centers N .

Now, assuming that the total FR is the sum of contributions of the individual centers, we can write the FR per unit center φ_i as $\varphi_i = \varphi_s/N' \sim (\Gamma/\gamma)\varphi_s/N$. When detecting the FR noise (rather than the standard FR), the magnitude of the contribution of each center will remain φ_s/N' as before, but its sign will be random: $\varphi_i = \pm(\Gamma/\gamma)\varphi_s/N$.

An important quantity for SN measurements is the mean square of fluctuations of the FR $\langle \delta\varphi^2 \rangle$, where $\delta\varphi \equiv \sum_{i=1}^{N'} \varphi_i$.

The quantity $\langle \delta\varphi^2 \rangle$ can be written in the form

$$\langle \delta\varphi^2 \rangle = \left\langle \left[\sum_{i=1}^{N'} \varphi_i \right]^2 \right\rangle = [\Gamma/\gamma] \varphi_s^2 / N. \quad (1)$$

In SNS, we typically detect the spectrum of the noise power or its spectral density, with its characteristic value $W = \langle \delta\varphi^2 \rangle T_2$, where T_2 is the spin dephasing time. Using Eq. (1), we obtain

$$W \equiv \langle \delta\varphi^2 \rangle T_2 = \frac{T_2 \sigma^2 \Gamma}{\gamma} \frac{nd}{A} \equiv \eta \frac{nd}{A}. \quad (2)$$

The parameters entering the last fraction of this equation depend on the measurement conditions and, to a certain extent, can be controlled by the experimentalist. The factor $\eta = T_2 \sigma^2 \Gamma / \gamma$ can be considered as a characteristic quantity of the studied paramagnetic center or, better to say, of the particular optical transition addressed. It is this parameter that allows one to compare different spin systems from the viewpoint of applicability of SNS. We see that the factor Γ/γ (which we call the *gain factor*) can drastically enhance the noise power, leaving the FR cross section unchanged.

Note, that we have not used a specific spectral shape for the single-impurity contribution to the polarimetric signal. The only parameter we use to estimate the number of centers contributing to the signal is the homogeneous spectral width γ of the optical resonance of a single impurity. We emphasize that our estimates do not change when the homogeneous optical width becomes smaller than the Zeeman splitting ($\gamma < \omega_L$) and the spectral features of the optical susceptibility of single impurity become resolvable in opposite circular polarizations: the number of centers contributing to the polarimetric signal for strictly monochromatic probing will still be controlled by the ratio of the homogeneous and inhomogeneous widths of γ/Γ .

The above reasoning shows, in particular, that objects with a relatively large FR cross section σ may be less favorable for the SN technique than those with a smaller σ but with a larger gain factor Γ/γ . The FR in the region of the f - d transitions, which is more easily observable on divalent ions, is usually larger than the FR near the f - f transitions. At the same time, the f - d transition bands with width of $\sim 10^{13}$ Hz are broadened homogeneously, while the lines of the f - f transitions with widths around 10^{10} Hz [27], are strongly inhomogeneously broadened, as the homogeneous linewidth often is in the range of 10^2 – 10^3 Hz [26,28], thus providing spin-noise gain factors of up to 10^8 . It is this effect that allows us to solve the problem of SN detection in activated crystals.

III. EXPERIMENTAL RESULTS

Now, let us turn to the demonstration of the SN gain effect. In our experiments, we used two crystals of cubic symmetry with the RE ions possessing f - f transitions in the spectral range accessible with a Ti:sapphire laser used as the source of the probe light. Sample 1 was a SrF_2 crystal coactivated with Nd^{3+} (0.5 mol %) and Yb^{3+} (0.15 mol %). Sample 2 was a crystal of CaF_2 : Nd^{3+} (0.1 mol %). Both crystals were grown using the Stockbarger technique in graphite crucibles. In these crystals, the trivalent RE ions substitute divalent cations of the host lattice. For this reason, the impurity centers created

FIG. 2. Experimental schematics. The narrow-band cw laser is guided through a single-mode polarization-maintaining fiber (F). It is then separated into two parts: the local oscillator (LO) and the probe. The probe light is vertically polarized and is sent focused through the sample. The interference of the horizontally polarized scattered light and the LO is measured at the balanced photodiodes. The AC component of the voltage is used to measure the spin-noise power spectrum, and the DC component is used to stabilize the phase of the interference by the PID loop. Here, PBS and nPBS, are polarizing and nonpolarizing beam splitters, respectively. Sh is a mechanical shutter, GT is a Glan-Taylor prism, and $\lambda/2$ is a half-wave plate. U_{SP} is the set point voltage for the PID loop of the piezocontrolled mirror (PZT).

thereby are mainly anisotropic due to local compensation of the excess activator charge. As a result, several groups of magnetically nonequivalent centers form and essentially complicate magnetic-resonance spectra by creating several resonances in the SN spectra at any fixed magnetic field. This situation is typical for the case of heterovalent substitution of impurity ions in cubic crystals [29,30]. Our aim, however, was not to identify and study particular centers in these crystals, but rather to convincingly demonstrate the possibility and efficiency of applying SNS to this class of objects.

To detect the spin noise, we used the homodyne detection scheme (see Fig. 2 [31,32]). It is clear from the consideration above that the SN gain effect can be observed only when the spectral width of the probe laser light is smaller than the homogeneous linewidth γ . This is why we used a tunable (cw) ring-cavity Ti:sapphire laser with a short-term (during 100 ms) spectral width of < 40 kHz. When measuring spin noise, the laser was additionally power stabilized and transmitted through a single-mode fiber to symmetrize the spatial mode. Behind the fiber, the laser was coupled into the entrance port of a Mach-Zehnder interferometer, with one beam passing through the sample and the second one bypassing the sample [local oscillator (LO)]. The light scattered by the sample had orthogonal polarization with respect to the incident beam and was then collected and combined with the LO of the interferometer. The resulting interference was measured using balanced photodiodes with 670 MHz bandwidth. The signal was digitized and fast-Fourier transformed by a field-programmable gate array [33]. The normalized difference between the LO with transmitted and blocked scattered probe light was measured for identical external conditions. The sample was placed in a cold-finger He-flow cryostat at a temperature of $T = 5.7$ K. The magnetic field was generated by an electromagnet in the Voigt configuration, i.e., it was oriented orthogonal to the probe beam direction. The laser was

FIG. 3. SN spectra of the studied SrF₂:Nd³⁺(0.5%)Yb³⁺(0.15%) crystal. (a) Transmission spectra obtained using a Cary spectrophotometer with a spectral resolution of 0.05 nm. (b) and (c) High-resolution transmission spectra obtained with a tunable Ti:sapphire laser. Laser power is 1 mW. The oscillating background signal is produced by laser etaloning at the sample surfaces. (d) and (e) are color maps demonstrating the magnetic field dependence of the SN power spectrum density (PSD) for the Nd³⁺ and Yb³⁺ ions, respectively. The spectra are detected for the same orientation of the crystal with respect to the magnetic field and light propagation directions. The white dashed lines in the color spectra mark the magnetic fields at which the spectra below are taken in shot noise units (SNU). We remind one that the multitude of lines in each figure corresponds to the several groups of magnetically nonequivalent anisotropic centers for each impurity ion.

focused on the sample to a spot of 40 μm diameter, using a 200-mm focal-length lens.

Overview measurements of transmission spectra of sample 1 [Fig. 3(a)] were performed with a Cary spectrophotometer providing a spectral resolution of 0.05 nm, in order to identify the spectral features that are suitable for the resonant SN detection. The most convenient f - f transitions were found in the spectral range 860–870 nm for Nd³⁺ and 965–980 nm for Yb³⁺, in agreement with literature data [28]. For a more detailed characterization, we performed transmission measurements with much higher spectral resolution, using the tunable Ti:sapphire laser [Figs. 3(b) and 3(c)]. Narrow spectral features are observed then in both spectral regions: the Nd³⁺ ions showed one absorption line at 863.37 nm with the FWHM $\Delta\lambda = 0.08$ nm, obtained using a Gaussian fit; the Yb³⁺ ions demonstrated a strong absorption peak at 965.15 nm with the width $\Delta\lambda = 0.02$ nm.

By tuning the laser emission wavelength to the absorption peaks of the corresponding lines we could detect the strongest SN signal [14]. Figures 3(d) and 3(e) show color maps of the magnetic field dependences of the noise power spectra, detected in the Voigt geometry. Examples of these spectra at $B = 15$ mT are given below these maps for the two used wavelengths and, correspondingly, for the two ion coactivators. The multitude of lines in the SN spectra demonstrates the presence of several groups of magnetically nonequivalent, anisotropic centers, which is typical for the heterovalent substitution of impurity ions. Furthermore, these measurements show the high spectral selectivity of SN spectroscopy, especially in comparison with EPR spectroscopy, where the overall signal from the tested sample volume containing the whole variety of impurity centers is detected.

Detailed experiments were performed using the sample containing a single dopant (sample 2). Figure 4(a) shows the

laser transmission spectrum of the sample at $T = 5.7$ K. We tested the SN signal at all five absorption peaks observed in this spectral region, and managed to detect a strong SN signal only at the peak with 862.69 nm [Fig. 4(b)]. In the color map, we also observe several lines with different magnetic field dispersions. Figure 4(c) demonstrates an exemplary spectrum of the SN power density in units of the shot-noise power at $B = 12$ mT for the probe light power of 1 mW. Full information about the symmetry and structure of the impurity centers revealed in the SN spectra can be obtained from their orientational dependence, while the field dependence of the resonance linewidth can provide information about spin-relaxation rates of the impurity ion.

The method used here to demonstrate the SN gain effect implies resonant probing of the system and, in general, cannot be considered as nonperturbative. So, it is interesting to study the probe light power dependence of the magnetic resonance linewidth detected for these conditions. To that end, we measured the light power dependence of the amplitude and width of the peak at 225 MHz [see insets in Fig. 4(c)]. The amplitude power dependence (plotted in log-log scale) demonstrates an initial linear increase followed by saturation, while the linewidth in this range of light power densities remains constant at 13.6 MHz. Therefore, we can conclude that the resonant laser light, which probes forbidden electronic transitions of the RE ions, does not noticeably affect their spin dynamics for the used experimental conditions.

IV. CONCLUSIONS

In summary, we have shown that the effect of spin-noise gain, which should be observable in many paramagnetic impurities with not completely filled electronic shells, could widen the class of objects to which SN spectroscopy can be

FIG. 4. Spin-noise spectra of the $\text{CaF}_2:\text{Nd}^{3+}$ (0.1%) crystal. (a) Laser transmission spectrum of the sample at 5.7 K. The oscillations are due to the etaloning effect of the laser by the sample surfaces. (b) Color map of SN spectra detected at the absorption peak 862.69 nm versus magnetic field. Laser power is 1 mW. (c) Exemplary spectrum at $B = 12$ mT with the two insets demonstrating the power dependence of the amplitude (log-log scale) and the HWHM (semilogarithmic scale) of the peak marked in the main panel.

applied, and thus enhance the potential of EPR spectroscopy for studying impurity paramagnets. The SN technique provides the EPR spectroscopy with additional degrees of freedom hardly accessible to its classical implementation. The optical excitation and detection provide spectral selectivity of the impurity centers and high spatial resolution both in transverse and in longitudinal directions (three-dimensional tomography). It is important that the SN measurements can be performed in a wide frequency range which is not restricted by the photodetector bandwidth [34,35]. The EPR measurements in SN spectroscopy are not rigidly coupled to any fixed radio frequency, as is usually the case in conventional EPR spectroscopy, and allow one to directly obtain panoramic EPR spectra over a wide spectral range. It is important to emphasize, that the SN signal does not depend explicitly on the spin-system polarization, and the absence (or smallness) of a

population difference between spin sublevels does not prevent the SN resonance detection. We believe that these specific features of the SN spectroscopy will essentially advance the present-day magnetic resonance spectroscopy of crystals and glasses containing paramagnetic impurities.

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