Spin noise of magnetically anisotropic centers

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(Received 17 October 2022; revised 13 January 2023; accepted 15 February 2023; published 28 February 2023)

Spin noise spectroscopy, as a sort of magnetic resonance technique, uses, for detection of spin precession, spontaneous fluctuations of magnetization revealed as a peak in the Faraday-rotation (FR) noise spectrum at Larmor frequency. In the model of precessing magnetization, the FR noise signal should be the greatest in the Voigt geometry (with magnetic field aligned across the light propagation), and should vanish in the Faraday geometry (with the field along the probe beam). This reasoning employs, implicitly or explicitly, the so-called Van Vleck theorem that establishes, within the limits of certain assumptions, a direct relation between the FR and magnetization of the spin system. We show that violation of these assumptions in crystals with anisotropic paramagnetic centers may qualitatively change the conventional laws of spin noise detection, making, in particular, the FR noise detectable in the Faraday geometry. These conclusions are confirmed by experimental studies of spin-noise spectra of CaF_2 crystals with tetragonal centers of Nd^{3+} ions.

DOI: 10.1103/PhysRevB.107.064427

I. INTRODUCTION

Spins in the external magnetic field exhibit spontaneous precession which is accompanied by spontaneous oscillations of transverse magnetization of the spin system. Detection of these oscillations by means of the Faraday-rotation (FR) technique provides the basis of the spin noise spectroscopy (SNS) that has been greatly developed during the last two decades [1–5]. Conceptual novelty of the SNS has manifested itself in a number of specific abilities inaccessible for the conventional electron paramagnetic resonance spectroscopy. Initially, the main peculiarity of the SNS technique was considered to be its nonperturbative character [6]. To date, however, this experimental approach has revealed many interesting and curious features of other kind that have made it useful and informative for studying both optical and spin-related properties of paramagnets. In the general sense, most specific properties of the SNS are related to the statistical, rather than coherent, summation of individual spin contributions into the total signal. Due to this fact, the SNS revealed abilities more typical for *nonlinear* optics [7], like applicability of the pump-probe technique [8], realization of optical three-dimensional (3D) tomography [9,10], sensitivity to the mechanism of broadening of optical transitions [5], etc. Today, the SNS is widely applied to atomic systems (mainly alkali metals) and semiconductor structures ([11–13]). Recently, the effect of the *giant spin-noise gain* has been discovered that allowed us to successfully apply the SNS technique to rare-earth-doped dielectric crystals [14]. Application of polarization-noise spectroscopy to intrinsic emission of the object allowed one to investigate specific features of dynamics of the exciton-polariton condensate [15,16].

Conventional arrangement of the SNS experiment [6] implies detection of the FR noise power of the laser beam transmitted through the sample in magnetic field $\mathbf{B}$ aligned across the light beam. This experimental arrangement is usually referred to as the *Voigt geometry*. The magnetic moment (magnetization $\mathbf{M}$) of each particle of the sample exhibits stochastic precession around the magnetic field with the Larmor frequency $\omega_L = g\mu B/\hbar$ [17] [Fig. 1(a)]. The gyration vector $\mathbf{g}$, which is dual with the antisymmetric part of the optical polarizability tensor α of each particle, $\alpha_{ij} \sim \varepsilon_{ijk} g_k$, and, in accordance with the Van Vleck theorem [18], is proportional to the particle magnetization $\mathbf{g} \sim \mathbf{M}$, also precesses around the magnetic field. As seen from Fig. 1(a), in the Voigt geometry, the projection $g_k = (\mathbf{g}, \mathbf{k})/k \sim M_k \equiv (\mathbf{M}, \mathbf{k})/k$ should oscillate on the Larmor frequency ω_L, thus making a spectrally localized contribution to the FR noise spectrum.

Let us rotate now the magnetic field by 90° to align it along the probe beam direction $\mathbf{B}\|\mathbf{k}$ [Fig. 1(b)]. In this configuration (usually called Faraday geometry), magnetic moments of the particles will precess as before, but projection of magnetization M_k and proportional to it projection of the gyration vector g_k upon this direction will remain constant [Fig. 1(b)]. Thus, in the Faraday geometry, the FR noise at the Larmor frequency should be absent.

In what follows, we present experimental data showing that rare-earth (RE) ions in a crystalline matrix, under conditions of resonant probing, may reveal the FR noise at the Larmor frequency even in the Faraday geometry. The goal of this paper is to explain this effect that may seem either paradoxical or trivial.

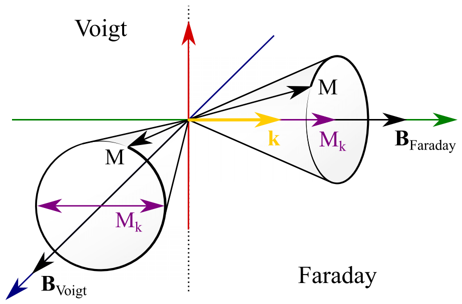

FIG. 1. Precession of magnetic moment in the Voigt (denoted by magnetic field vector $\mathbf{B}_{\text{Voigt}}$ in the left part of the figure) and Faraday ($\mathbf{B}_{\text{Faraday}}$ in the right) geometry. It is seen that projection of magnetization M_k upon the probe beam direction $\mathbf{k}$, in the Voigt geometry, oscillates. In the Faraday geometry, magnetization precesses around the direction $\mathbf{k}\|\mathbf{B}$, and its projection upon the probe beam direction $\mathbf{k}$ remains fixed.

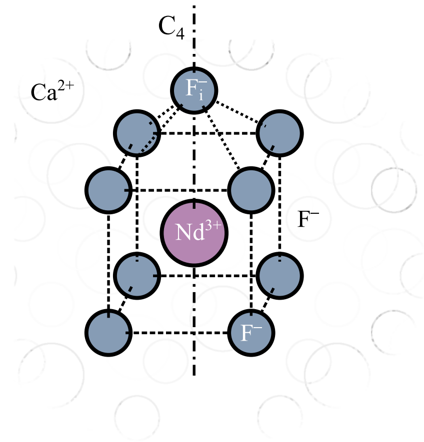

FIG. 2. Schematic representation of the Nd^{3+} center in the CaF_2 crystal. The Nd^{3+} ion replacing the Ca^{2+} ion of the crystal lattice brings an excess charge that is compensated by the interstitial fluorine ion (F_i^-). As a result, the Nd^{3+} center acquires tetragonal symmetry, with its principal axis (C_4) aligned along the line $Nd^{3+} - F_i^-$.

The paper is organized as follows. In Sec. II we describe the sample under study, the setup for measuring the FR noise, and the experimental results thus obtained. Section III contains a simplified analysis of these results showing that magnetic anisotropy of the impurity centers by itself does not allow to explain the FR noise signal at the Larmor frequency in the Faraday geometry. We also make a conclusion here about inapplicability of the Van Vleck theorem to the case of resonant probing of the paramagnet. In Sec. IV, we calculate the contribution of an axial center to the FR noise under conditions of resonant probing and show that, in the presence of magnetic and optical anisotropy, this contribution may reveal a resonant feature at the Larmor frequency even in the Faraday geometry. In the Conclusion, we briefly summarize the results of the work.

II. EXPERIMENTAL

To verify our assumptions, we have chosen the sample of CaF_2:Nd^{3+} (0.1 mol%), similar to that used in our previous work [19]. In this crystal, the neodymium ions occupy tetragonal positions, with interstitial F^- ions compensating excess charge of the impurity and being aligned along the fourfold symmetry axes of the crystal, thus forming three magnetically nonequivalent groups. Correspondingly, the spin-noise spectrum should reveal, at each magnetic field, three peaks associated with three different precession frequencies of these groups. The behavior of these peaks in rotating magnetic field was studied in [19]. The structure of the tetragonal impurity center in the CaF_2 crystal is shown schematically in Fig. 2. The probe beam was aligned along the threefold symmetry axis of the crystal which was normal to the sample surface. One can see that, in this case, the symmetry axes of all tetragonal centers appear to be tilted with respect to the light propagation direction. To perform the measurements, the magnetic field $\mathbf{B}$ of fixed magnitude was rotated around the axis normal to the probe light wave vector $\mathbf{k}$, thus passing through both basic experimental configurations (Faraday's and Voigt's). The schematic of the experimental setup is shown in Fig. 3(a). The beam of a tunable continuous-wave Ti:Sapphire laser, after passing through the sample, was directed to a conventional balanced polarimetric detector which comprises a half-wave plate, a polarizing beamsplitter (PBS), and a balanced photodetector (BPD). The digital

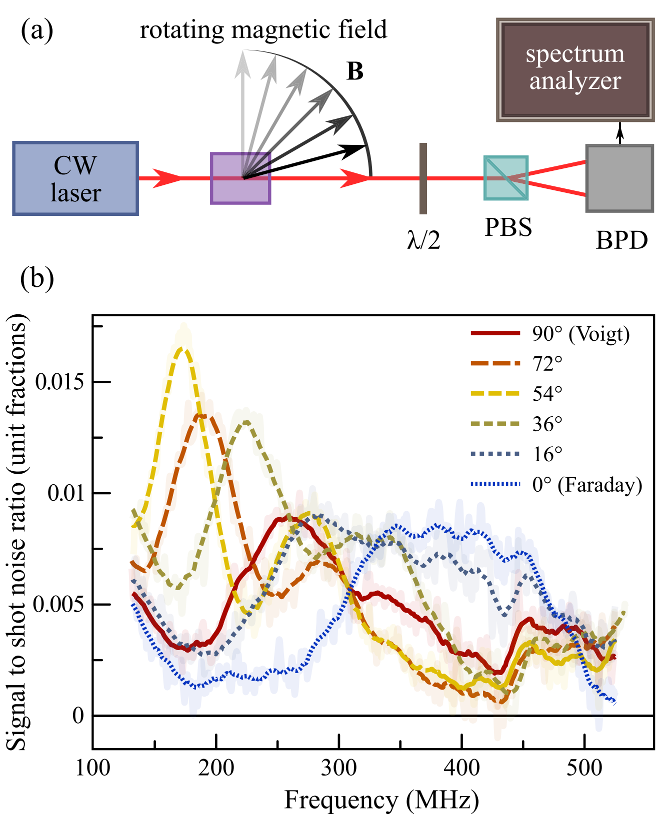

FIG. 3. (a) Schematic of the experimental setup. See notations in the text. (b) The FR noise spectra for different orientations of the magnetic field: $\phi = \pi/2$ (Voigt geometry) and $\phi = 0$ (Faraday geometry). $B \approx 10$ mT.

real-time spectrum analyzer at the output of the detector provides the FR noise spectrum of the input beam.

It is important that measurements of the spin-noise spectra were performed under conditions of resonant probing at one of the crystal-field components of spin-orbit multiplet of the Nd^{3+} ion. The FR noise spectra obtained for different orientations of the magnetic field are shown in Fig. 3(b). We see that the resonant features of the spectra do not disappear for the magnetic field aligned along the probe beam (the Faraday geometry) that, as was mentioned above, looks paradoxical. Clarification of this paradox is the main content of the work.

III. SIMPLIFIED TREATMENT

An important role in magneto-optics of impurity ions in crystalline matrices is played by the Van Vleck theorem [18] stating that the contribution $\delta\phi$ of the impurity ions into the FR angle is proportional to the projection of magnetization $\mathbf{M}$ of the impurity system onto the light beam direction $\mathbf{k}$, i.e., $\delta\phi \sim M_k = (\mathbf{M}, \mathbf{k})/\mathrm{k}$.

In spite of the fact that the Van Vleck theorem was proved for the case of *essentially nonresonant* probing, it is often used for interpretation of experimental data in SNS, identifying the FR noise with the magnetization noise of the spin system even when the measurements are performed under conditions of optical resonance. In many cases, this approximation appears to be justified (at least for qualitative treatment), but, as will be shown below, it cannot explain the spectral feature of the FR noise observed at the Larmor frequency ω_L in the Faraday geometry.

Below we show that the noise spectrum of the impurity-ion magnetization projection M_k upon the probe beam direction $\mathbf{k}$ cannot reveal any feature at the Larmor frequency when the magnetic field is aligned along the light beam ($\mathbf{B}\|\mathbf{k}$). The operator of this projection can be presented as $\hat{M}_k = (\hat{\mathbf{M}}, \mathbf{k})/k$, where $\hat{\mathbf{M}}$ is the operator of the magnetization of the ion. The operator $\hat{M}_k$ commutes with the Zeeman Hamiltonian $H = -\hbar^{-1}(\mathbf{B}, \hat{\mathbf{M}})$ [20] of the ion, since $\mathbf{k} \sim \mathbf{B}$. We assume the temperature of the sample to be so small that only the lowest Kramers doublet of the ion appears to be populated. We denote the wave functions of these states by $|g1\rangle$ and $|g2\rangle$. Let h and m be two-dimensional matrices of the Zeeman Hamiltonian H and of the operator M_k, respectively: $h_{\alpha\beta} \equiv \langle g\alpha|H|g\beta\rangle$ and $m_{\alpha\beta} \equiv \langle g\alpha|\hat{M}_k|g\beta\rangle$, $\alpha, \beta = 1, 2$. Dynamics of magnetization of the ion is controlled, in our case, by the matrix h that plays the role of Hamiltonian. Since $H \sim M_k$, the commutator $[h, m]$ also vanishes. The noise power spectrum M_k is given by the Fourier transform of the correlation function $\langle m(t)m(0) + m(0)m(t)\rangle$, for which, using the commutativity $[\hat{h}, m] = 0$, we can write the following chain of equations:

$$\begin{aligned}\langle m(t)m(0) + m(0)m(t)\rangle &\\ = \mathrm{Sp}\ \rho_{eq}[e^{\imath ht} m e^{-\imath ht} m + m e^{\imath ht} m e^{-\imath ht}] &\\ = 2\ \mathrm{Sp}\ \rho_{eq}\ m^2. & \qquad (1)\end{aligned}$$

Here, ρ_{eq} is the equilibrium density matrix of the ion. The relationship (1) shows that the correlation function $\langle m(t)m(0) + m(0)m(t)\rangle$ does not depend on time. As a result, its Fourier transform controlling the noise power spectrum cannot reveal any feature at the Larmor frequency ω_L. Thus, we come to the conclusion that if, in the above Faraday configuration of the SNS experiment, the detected signal corresponded to the noise of the magnetization projection M_k, then no spectral feature at the Larmor frequency ω_L should have been observed. It is noteworthy that this result does not depend on possible magnetic anisotropy of the impurity ion. Since in our experiments a spectral feature of the noise spectrum at the frequency ω_L has been observed, we come to a conclusion about the inapplicability of the Van Vleck theorem even for qualitative interpretation of the experimental results.

The most substantial violation of applicability of the Van Vleck theorem, in our case, was related to resonant probing of the Nd^{3+} ions with a significant and well-optically-resolved crystal-field splitting of spin-orbit multiplets in the CaF_2 matrix. In the next section, we will calculate, in the framework of the axial crystal-field model, correlators of the polarizability tensor of the anisotropic paramagnetic center and will show that this spectrum may show a feature at the frequency of magnetic splitting (Larmor frequency ω_L) even in the Faraday geometry.

IV. OPTICAL SUSCEPTIBILITY OF THE AXIAL PARAMAGNETIC CENTER AND POLARIZATION FLUCTUATIONS OF THE PROBE BEAM

Calculation of the optical polarizability tensor of the paramagnetic center will be performed under the following simplifying assumptions:

(1) Optical susceptibility of the ion is related to transitions between two spin-orbit multiplets, ground and excited. The total momenta F of these multiplets are assumed to be the same and half-integer.

(2) The crystal field of the host matrix is assumed axially symmetric, with the axis C_∞ setting orientation of the center.

(3) The axial crystal field splits the spin-orbit multiplets into doublets $|\alpha, J, \pm M\rangle$, $M = 1/2, 3/2, ..., F$, where $\alpha = 1(2)$ for the excited (ground) multiplet, $J = F'(F)$ is the total moment of the excited (ground) multiplet, and M determines projection of the moment upon the quantization axis (chosen along the direction C_∞). Since our goal is to analyze polarization noise under the condition of resonant probing, we will assume that the probe light frequency is close to that of the transition between the doublets $|2, F, \pm 1/2\rangle \to |1, F', \pm 1/2\rangle$, with the ground doublet $|2, F, \pm 1/2\rangle$ being the only populated.

(4) The magnetic (Zeeman) splitting, in the optical spectrum, is assumed to be unresolved, i.e., $\omega_L < \delta$, where δ is the width of the optical transition.

Note that, in our magneto-optical experiments, we detect polarization fluctuations of the probe light, created by paramagnetic ions, rather than fluctuations of magnetization of these ions, calculated in the previous section. If concentration of the ions is not too high and their effect upon the probe beam polarizations can be considered in the approximation of single scattering, then the contribution of each ion into the polarization signal is additive and is described by the following relationship (see, e.g., Ref. [21]):

$$\begin{aligned}\delta u &\equiv \delta u_e + \imath \delta u_r\\ &= |A_0|^2 \pi k[\alpha_{zx} - \alpha_{xz} - (\alpha_{xz} + \alpha_{zx})\cos 2\theta + (\alpha_{zz} - \alpha_{xx})\sin 2\theta]. \qquad (2)\end{aligned}$$

The sense of the symbols entering this relationship is the following. The imaginary part δu describes the signal detected by the balanced polarimetric detector, while the real part describes the signal of ellipticity detected by the same polarimetric detector but with a quarter-wave plate placed in front of it. The axes of the plate $\lambda/4$ make the angle 45° with respect to the axes of the detector (Fig. 3); $k \equiv \omega/c$ is the wave number of the probe light; α is the tensor of polarizability of the ion, with the tensor components α_{ik} taken in the coordinate system where the components of the probe light field have the form $A_{0x} = A_0 \sin\theta$ and $A_{0z} = A_0 \cos\theta$. In this system (hereafter called the K system), the linearly polarized probe beam evidently has only x and z components and propagates along the y axis, with the angle θ specifying azimuth of its polarization plane. Variation of the probe beam polarization after passing through the paramagnet is presented by a sum of contributions (2) from each of the ions. The probe beam polarization noise observed in our experiments is related to fluctuations of the tensor α of each of the ions. Our immediate goal is to calculate these fluctuations.

Let us denote by $|1\rangle$ and $|2\rangle$ ($|11\rangle$ and $|22\rangle$) the wave functions of the excited (ground) doublet in the magnetic field and introduce special notations for these functions at $\mathbf{B} = \mathbf{0}$: $|gj\rangle \equiv |j\rangle|_{B=0}$ and $|ej\rangle \equiv |jj\rangle|_{B=0}$, $j = 1, 2$.

Since the wave functions of the excited and ground doublets ($|gj\rangle$ and $|ej\rangle$, $j = 1, 2$) are characterized by the same projections of the momentum $\pm 1/2$ (i.e., are transformed by the same representation of the point group of the crystal), the vector columns of coefficients of decomposition of the functions $|j\rangle$ and $|jj\rangle$, $j = 1, 2$ over states of the ground and excited multiplets ($|2, F, M\rangle$ and $|1, F, M\rangle$, $M = -F, ..., F$, respectively, will be the same. If for the above column vectors we retain the notations used for the corresponding wave functions, then, for the vector columns of the ground and excited doublets, in our case, we have the following relations $|j\rangle = |jj\rangle$, $j = 1, 2$, with dimensions of these columns equal to $2F + 1$.

In terms of the above definitions and with allowance for the assumptions made above, the results of the standard calculation of the polarizability tensor α, in the framework of the theory of linear response, can be presented in the form

$$\alpha_{ik}(\Delta\omega) = \frac{\hbar^{-1} d^2}{\Delta\omega + \imath\delta} \sum_{p,j,q=1}^{2} \langle p|\rho|j\rangle\langle j|J_k|q\rangle\langle q|J_i|p\rangle. \quad (3)$$

Here, $\Delta\omega$ is the detuning of the probe beam from the frequency of transition between the ground and excited doublets; δ and d are the width and dipole moment of the optical transition; $|j\rangle$, $j = 1, 2$ and J_k are the vector columns of the wave functions of the ground doublet and matrix of the kth projection of the angular momentum, respectively; and $\langle i|\rho|j\rangle$, $i, j = 1, 2$ are the elements of the density matrix of the ion on the basis of the ground doublet.

A. Calculation of the optical susceptibility

Let us denote by $\pm E_g$ the energy of the ground-state doublet in magnetic field (at zero field $E_g = 0$). Then, the density matrix of the ground doublet can be presented in the form

$$\rho = \begin{pmatrix} |C_1|^2 & |C_1C_2|e^{\imath[2E_g t + \phi_1 - \phi_2]} \\ |C_1C_2|e^{-\imath[2E_g t + \phi_1 - \phi_2]} & |C_2|^2 \end{pmatrix}. \quad (4)$$

Here, the phases $\phi_{1,2}$ are random and uniformly distributed over the interval $[0, 2\pi]$, while the average populations $|C_1|^2$ and $|C_2|^2$ of the states $|1\rangle$ will be considered in thermodynamic equilibrium:

$$\langle |C_1|^2\rangle = Z^{-1} e^{-E_g/k_B T},$$
$$\langle |C_2|^2\rangle = Z^{-1} e^{E_g/k_B T},$$
$$Z = e^{-E_g/k_B T} + e^{E_g/k_B T}. \quad (5)$$

The time dependence of elements of the tensor α is seen to be determined by nondiagonal elements of the density matrix (4) and represents oscillations with random phase at the frequency $2E_g$, evidently corresponding to the Larmor frequency $\omega_L = 2E_g$ mentioned in the previous section. Interaction of the paramagnetic center with phonons of the crystal lattice, which we neglected, may lead to random dephasing of oscillations of nondiagonal elements of the density matrix (4), which in turn leads to decay of the correlation functions calculated using Eq. (4). This decay will be taken into account phenomenologically at the final stage of the calculation.

Magnetic anisotropy of the paramagnetic center under consideration is determined by a specific form of the wave functions of the doublets $|g1\rangle$, $|g2\rangle$, $|e1\rangle$, $|e2\rangle$, arisen from the ground and excited multiplets under the action of the crystal field. Let us perform a corresponding calculation following Eq. [22] for the ground doublet and introduce quantities we will need later.

The Hamiltonian of interaction with the magnetic field in the representation of functions of the ground multiplet is given by the matrix $-g_L\mu(\mathbf{J}, \mathbf{B})$ [23]. The matrix of this Hamiltonian, in the representation of the functions of the ground doublet $|g1\rangle$, $|g2\rangle$ (we denote it by H^g), has the elements

$$H^g_{\alpha\beta} = -g_L\mu B_i\langle g\alpha|J_i|g\beta\rangle \quad \alpha, \beta = 1, 2. \quad (6)$$

Using orthogonality of the Pauli matrices σ^i: $\mathrm{Sp}\,\sigma^i = 0$, $\mathrm{Sp}\,\sigma^i\sigma^k = \frac{1}{2}\delta_{ik}$, $i, k = 1, 2, 3$, the matrices $g_L\langle g\alpha|J_i|g\beta\rangle$ can be represented by a linear combination of the matrices σ^k, $k = 1, 2, 3$, after which the Hamiltonian matrix H^g acquires the form

$$H^g = -\mu B_i g_{ik}\sigma^k \quad (7)$$

where the so-called g tensors [24] have elements determined by the relationship

$$g_{ik} = 2g_L \sum_{\alpha\beta} \langle g\alpha|J_i|g\beta\rangle\sigma^k_{\beta\alpha}. \quad (8)$$

For the known g tensor of the ground doublet, direct diagonalization of the two-dimensional matrix (7) yields two eigenvalues with opposite signs $\pm E_g$ [see Eq. (4)] and two two-dimensional orthonormal vector columns $a^{\pm}$, such that

$$H^g\begin{pmatrix} a_1^{\pm} \\ a_2^{\pm} \end{pmatrix} = \pm E_g\begin{pmatrix} a_1^{\pm} \\ a_2^{\pm} \end{pmatrix}. \quad (9)$$

Then the wave functions $|j\rangle$, $j = 1, 2$ of the ground doublet in the magnetic field $\mathbf{B}$ can be expressed through the functions $|gj\rangle$, $j = 1, 2$ (in zero field) using the matrix a, defined in the following way:

$$\begin{aligned}|1\rangle &= a_1^+|g1\rangle + a_2^+|g2\rangle\\ |2\rangle &= a_1^-|g1\rangle + a_2^-|g2\rangle\end{aligned} \Rightarrow \begin{pmatrix}|1\rangle\\ |2\rangle\end{pmatrix} = a\begin{pmatrix}|g1\rangle\\ |g2\rangle\end{pmatrix},$$

$$a = \begin{pmatrix}a_1^+ & a_2^+\\ a_1^- & a_2^-\end{pmatrix}. \quad (10)$$

Let us turn now to Eq. (3), which shows that to calculate the optical susceptibility tensor α_{ik} we need two-dimensional matrices of the angular-momentum projections operators in the representation of the ground doublet of the ion. By denoting these matrices by symbols J_k^c,

$$J_k^c \equiv \begin{pmatrix}\langle 1|J_k|1\rangle & \langle 1|J_k|2\rangle\\ \langle 2|J_k|1\rangle & \langle 2|J_k|2\rangle\end{pmatrix}, \quad (11)$$

relationship (3) can be presented in a compact form:

$$\alpha_{ik}(\Delta\omega) = \frac{\hbar^{-1}d^2}{\Delta\omega + \imath\delta}\,\mathrm{Sp}\,\rho J_k^c J_i^c. \quad (12)$$

Let us denote by J_k^g the matrices J_k^c in zero magnetic field $J_k^g = J_k^c|_{B=0}$. Using Eqs. (6)–(8), the matrices J_k^g can be expressed through the Pauli matrices σ^j, $j = 1, 2, 3$:

$$J_k^g \equiv \begin{pmatrix}\langle g1|J_k|g1\rangle & \langle g1|J_k|g2\rangle\\ \langle g2|J_k|g1\rangle & \langle g2|J_k|g2\rangle\end{pmatrix} = \sum_{j=1}^{3}\frac{g_{kj}}{g_{\mathrm{L}}}\sigma^j. \quad (13)$$

By expressing $|j\rangle$ through $|gj\rangle$, $j = 1, 2$ with the aid of Eq. (10), we obtain

$$J_k^c = a^{-1T}J_k^g a^T. \quad (14)$$

By combining Eqs. (12)–(14), we can express the polarizability tensor of the ion through its density matrix (4) and g tensor (8):

$$\alpha_{ik}(\Delta\omega) = \frac{\hbar^{-1}g_{\mathrm{L}}^{-2}d^2}{\Delta\omega + \imath\delta}\sum_{jj'} g_{kj}g_{ij'}\,\mathrm{Sp}\,a^T\rho a^{-1T}\sigma^j\sigma^{j'}. \quad (15)$$

Recall again that time dependence of the tensor α arises due to the fact that generally the ion is in a superposition state with the density matrix whose nondiagonal elements oscillate at the Larmor frequency $\omega_{\mathrm{L}} = 2E_g$.

B. Calculation of the FR noise correlator for axial center

Let the axis C_∞ of the paramagnetic center be aligned along the z axis of our laboratory K system. Then, in accordance with the third assumption presented in the beginning of this section, $|g1\rangle = |2, F, 1/2\rangle$ and $|g2\rangle = |2, F, -1/2\rangle$ (here, F is the total momentum of the ground multiplet of the ion). Now, using Eq. (8) and known formulas for the matrices of angular momenta [25], we can calculate the g tensor:

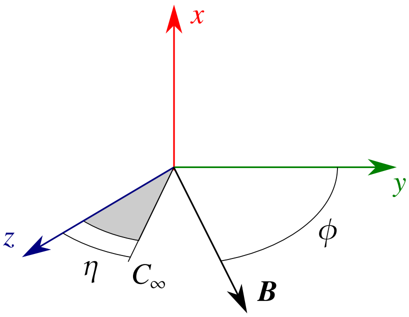

FIG. 4. On the determination of the experiment geometry which leads to vanishing of the signal at Larmor frequency for an anisotropic center. See text for details.

$$g = \begin{pmatrix}g_\perp & 0 & 0\\ 0 & g_\perp & 0\\ 0 & 0 & g_\parallel\end{pmatrix}, \quad (16)$$

where $g_\perp \equiv g_{\mathrm{L}}\sqrt{F(F+1)+1/4}$, $g_\parallel \equiv g_{\mathrm{L}}$. Further stages of calculating contribution of the paramagnetic ion into polarization fluctuations of the probe beam are as follows:

(1) Calculating the matrix of the eigenvectors a (10) and eigenvalues $\pm E_g$ of the Hamiltonian (7).

(2) Substituting the density matrix (4) into Eq. (15) and getting the time-dependent polarizability tensor α as a function of random complex amplitudes $C_{1,2}$ [see Eq. (4)].

(3) Using (2), finding the time-dependent contribution of the center under consideration into the FR $\delta u_r(t)$. Averaging over random complex amplitudes $C_{1,2}$ and calculating the correlator $\langle u_r(t)u_r(0)\rangle$, where the factor $e^{-|t/T_2|}$ takes into account phenomenologically the spin-precession decay time T_2, related to spin-phonon interaction.

(4) Under assumption of independent contributions of all N paramagnetic centers inside the probe beam, calculating the gyrotropy noise power spectrum as $\mathcal{N}(\nu) = N\int dt\,\langle u_r(t)u_r(0)\rangle e^{-\imath\nu t}$.

Consider now the case when the axis of the paramagnetic center is tilted by the angle $-\eta$ around the x axis of the laboratory K system (Fig. 4). The difference between this case and the one considered above is that the wave function of the ground doublet of the center should be "rotated" by the angle η around the x axis of the laboratory K system using the operator $e^{\imath J_x\eta}$: $|g1\rangle = e^{\imath J_x\eta}|2, F, 1/2\rangle$ and $|g2\rangle = e^{\imath J_x\eta}|2, F, -1/2\rangle$. Calculation by Eq. (8) shows that such a transformation corresponds to the following matrix of the g tensor:

$$g^\eta = \begin{pmatrix}g_\perp & 0 & 0\\ 0 & g_\perp\cos\eta & g_\parallel\sin\eta\\ 0 & -g_\perp\sin\eta & g_\parallel\cos\eta\end{pmatrix}. \quad (17)$$

Taking it into account and moving over stages 1–3 presented above, we can obtain the following expression for the correlation function of the FR noise (ellipticity):

$$\langle\delta u_{\mathrm{L}}^r(t)\delta u_{\mathrm{L}}^r(0)\rangle = |A_0|^4\langle|C_1C_2|^2\rangle g_\perp^2 g_\parallel^2[\mathrm{Re}\ (\ \mathrm{Im}\ )\mathcal{P}(\Delta\omega)]^2\mathcal{C}(t), \quad (18)$$

where

$$\mathcal{C}(t) \equiv \frac{e^{-|t|/T_2}\cos[\omega_{\rm L}t]}{2}\{\cos^2\eta\cos^2\epsilon + + [\xi\cos\eta\sin\epsilon + æ\sqrt{1-\xi^2}\sin\eta]^2\},$$

$$\tan\epsilon \equiv -\frac{\tilde{B}_y}{\tilde{B}_x}, \quad \xi \equiv \frac{\tilde{B}_z}{\sqrt{\tilde{B}_z^2 + æ^2[\tilde{B}_x^2 + \tilde{B}_y^2]}}, \quad æ \equiv \frac{g_\perp}{g_\parallel},$$

$$\omega_{\rm L} \equiv \mu g_\parallel\sqrt{\tilde{B}_z^2 + [\tilde{B}_y^2 + \tilde{B}_x^2]æ^2}$$

$$\begin{pmatrix}\tilde{B}_x\\ \tilde{B}_y\\ \tilde{B}_z\end{pmatrix} = \begin{pmatrix}1 & 0 & 0\\ 0 & \cos\eta & -\sin\eta\\ 0 & \sin\eta & \cos\eta\end{pmatrix}\begin{pmatrix}B_x\\ B_y\\ B_z\end{pmatrix},$$

$$\mathcal{P}(\Delta\omega) \equiv \pi k\frac{\hbar^{-1}g_L^{-2}d^2}{\Delta\omega + \imath\delta}. \quad (19)$$

To calculate the correlation function of the ellipticity noise, in Eq. (18) one should replace Re with Im. The mean values $\langle|C_1C_2|^2\rangle$ can be evaluated using Eq. (5) as $\langle|C_1C_2|^2\rangle \sim Z^{-2}$. The frequency spectrum of the noise power $\mathcal{N}(\nu)$ can be obtained from the above expressions by the replacement $e^{-|t|/T_2}\cos[\omega_L t] \to 1/T_2\,[T_2^{-2} + (\nu-\omega_L)^2]^{-1}$ in (19). Note that the correlators thus obtained do not depend on the polarization azimuth θ of the probe beam.

C. Discussion

Expressions (18) and (19) allow one to calculate the polarization noise spectrum for arbitrary relative orientations of the axis of paramagnetic center, magnetic field, and probe beam direction. For this purpose, (i) the y axis of the laboratory K system is chosen parallel to the probe beam direction, (ii) the z axis is chosen orthogonal to the y axis and lying in the plane containing the axis of the center and the axis y, and (iii) the x axis is chosen orthogonal to the axes y and z. In the coordinate system obtained in this way we find components of the magnetic field $B_{x,y,z}$ and substitute them into Eq. (19).

Now we can make sure that in the Faraday geometry ($B_x = B_z = 0, B_y \neq 0, \epsilon = -\pi/2$), in the absence of the magnetic anisotropy ($æ = 1$), $\mathcal{C}(t)$ is always zero. In addition, Eqs. (18) and (19) allow us to explain the FR noise at the Larmor frequency observed in our experiment in the Faraday geometry. As is seen from these expressions, this spectral feature arises in the noise spectrum only in the presence of magnetic anisotropy ($æ \neq 1$) at $\eta \neq 0, \pi/2$. In our experiments, these conditions were satisfied for all three groups of the Nd^{3+} centers.

It is interesting that the considered simplest model of an anisotropic paramagnetic center predicts vanishing of the FR noise signal upon rotation of the magnetic field in the zy plane (i.e., in the plane of the probe beam and of the axis of the center, when $B_z = B\sin\phi, B_y = B\cos\phi$, Fig. 3), in some intermediate (neither Voigt's nor Faraday's) geometry at $\phi = \phi_c$, where

$$\tan\phi_c = \frac{B_z}{B_y} = \frac{(æ^2-1)\tan\eta}{1+(æ\tan\eta)^2}. \quad (20)$$

As an example of application of the relationships obtained in the previous section, we present in Fig. 5 the calculated angular dependencies of the noise spectrum for the system comprised of three groups of anisotropic centers with their axes aligned along the fourth-order axis of a cube. The probe beam is assumed to propagate along the third-order axis of the cube. Amplitude of the noise signal is indicated by the line brightness. The dependencies were calculated for the magnetic field rotating around an axis perpendicular to the probe beam direction. Under this condition, the Faraday geometry is necessarily realized at some moment, denoted in the figure by the angle 0 (or π). Figure 5(a) shows the angular dependence of the noise spectrum for the magnetic field rotating in the plane containing the light beam and the axis of centers of one of the groups. As was shown above, the noise signal of this group, at certain orientation of the magnetic field, would vanish [see Eq. (20)]. This point is indicated in the figure by a red arrow. The spectrum in Fig. 5(a) shows only two curves of three, because in this orientation two of them coincide. An

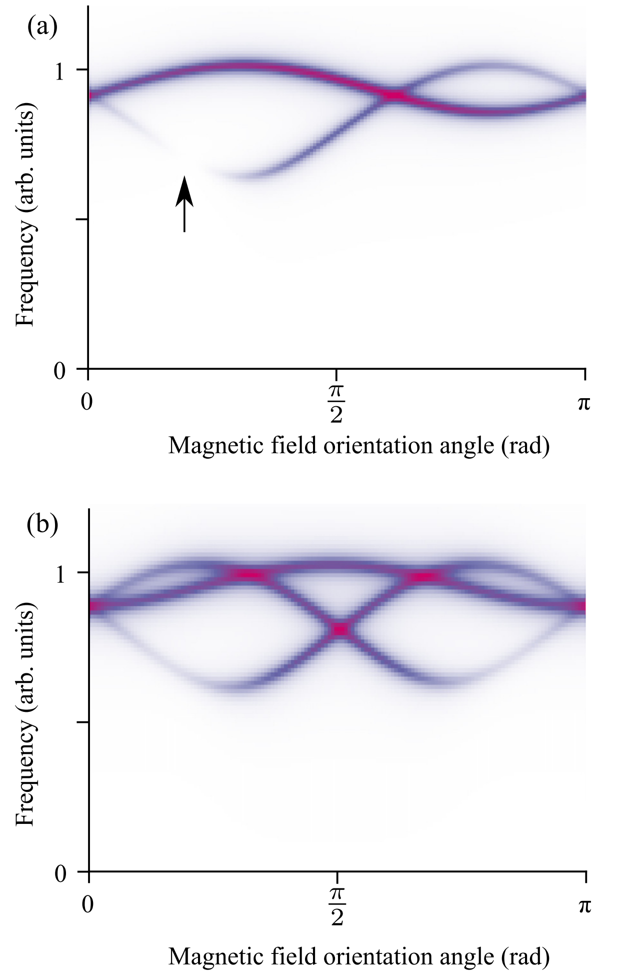

FIG. 5. The calculated angular dependencies of the noise spectrum for the considered system of three groups of anisotropic centers. Amplitude of the noise signal is indicated by the line brightness. The dependencies were calculated for the magnetic field rotating around an axis perpendicular to the probe beam direction. The Faraday geometry corresponds to the angles 0 (or π). (a) A special case when the noise signal of one of the groups, at some point (shown by red arrow), vanishes (see Eq.(20)). (b) Angular dependence of the noise spectrum for a more general case.

example of the noise spectrum for a more general case of the rotating field orientation is shown in Fig. 5(b). The calculations were performed for æ = 2. The relationship between the Larmor frequencies and the dephasing time was chosen sufficiently large to provide good resolution of the resonances of different groups.

The above theoretical treatment, as a whole, even for the simplest crystal-field model, is rather cumbersome. So, to make physical content of the results more transparent, it makes sense to present the following semiclassical comment. The FR $\delta\phi$ of the probe beam propagating along the axis 2 (y axis in Fig. 3) is controlled mainly by the antisymmetric part of the tensor α (2): $\delta\phi \sim \alpha_{13} - \alpha_{31}$. It is seen from Eq. (15) that $\alpha_{13} - \alpha_{31} \sim \langle [m_1, m_3] \rangle$, where $m_k \equiv g_{kj}\sigma^j$ are the two-dimensional matrices of the operator of the kth component of magnetic moment in the representation of the ground crystal-field doublet [see Eq. (7)]. In the case of the magnetically isotropic sample $g_{kj} \sim \delta_{kj}$, the commutator $[m_1, m_3] \sim [\sigma^1, \sigma^3] = -\imath\sigma^2 \sim m_2$, and we come to the result of Van Vleck $\delta\phi \sim \langle m_2 \rangle$, when, in accordance with the results of Sec. III, observation of the noise signal at the Larmor frequency ω_L, in the Faraday geometry, appears to be forbidden. In the presence of magnetic anisotropy (as in our case), the above commutation relations are violated, and observation of the FR noise resonance in the Faraday geometry becomes possible. This conclusion agrees well with our experimental results.

V. CONCLUSIONS

We studied the FR noise of the CaF_2:Nd^{3+} crystal in the Faraday geometry. We have found the FR noise localized at the Larmor frequency forbidden from the viewpoint of conventional SNS, which identifies the FR noise with the noise of magnetization. Calculation of the FR noise of the crystal in the framework of the axial crystal field shows that the presence of magnetic and optical anisotropy of the paramagnetic centers in the crystal matrix lifts the above restriction.

At first glance, this result may look trivial. Indeed, it seems evident that a magnetic field that "sees" the anisotropic paramagnetic center differs by its direction from the external field, and therefore the "effective" magnetic field in the Faraday geometry may contain transverse components. But this reasoning contains two errors. First, the spin precession of the anisotropic center occurs around the external (rather than effective) magnetic field, and longitudinal magnetization of the anisotropic center does not reveal any oscillation at the Larmor frequency. Second, when measuring the FR noise, we detect fluctuations of optical susceptibility of the medium, rather than direct fluctuations of spin magnetization, as is often implied in literature. In this paper, we attract attention to the fact that identification of the FR noise with the noise of magnetization is not universally correct. Such a relation between the FR and magnetization, which was initially formulated by Van Vleck [18], was based on certain assumptions that are often ignored in the present-day magneto-optics. In our case in particular, violation of the Van Vleck theorem was related to smallness of detuning of the probe beam from the optical resonance. In this paper, we show that, generally, the assumptions underlying the Van Vleck theorem are not necessarily fulfilled, and ignoring this fact may be essential even for qualitative interpretation of magneto-optical experiments.

ACKNOWLEDGMENTS

We highly appreciate financial support of the experimental work by the Russian Science Foundation Grant No. 21-72-10021. G.G.K. acknowledges support of the theoretical work from the SPbU Grant No. 94030557 and RFBR Grant No. 19-52-12054. The work was fulfilled using equipment of the Nanophotonics Research Center of the SPbU Science Park.

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