

## Spin-noise spectroscopy of randomly moving spins in the model of light scattering: Two-beam arrangement

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A strict analytical solution of the problem of spin-noise signal formation in a volume medium with randomly moving spin carriers is presented. The treatment is carried out in the model of light scattering in a medium with fluctuating inhomogeneity. Along with conventional single-beam geometry, we consider the two-beam arrangement, with the scattering field of the auxiliary (tilted) beam heterodyned on the photodetector illuminated by the main beam. It is shown that the spin-noise signal detected in the two-beam arrangement is highly sensitive to motion (diffusion) of the spin carriers within the illuminated volume and thus can provide additional information about the spin dynamics and spatial correlations of spin polarization in the volume media. Our quantitative estimates show that, under real experimental conditions, spin diffusion may strongly suppress the spin-noise signal in the two-beam geometry. The mechanism of this suppression is similar to that of the time-of-flight broadening with the critical distance determined by the period of two-beam spatial interference rather than by the beam diameter.

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### I. INTRODUCTION

Spectroscopy of spin noise rapidly developing during the past decade has been shown to be an efficient method of research with a wide range of interesting informative abilities in the field of magnetospin physics [1–3]. Spin-noise spectroscopy (SNS) has made it possible to study resonance magnetic susceptibility of nano-objects (e.g., quantum wells and quantum dots), hardly accessible for the electron spin resonance technique [4,5], to observe the dynamics of nuclear magnetization [6,7], and to investigate certain nonlinear phenomena in such systems [8]. The fact that magnetization is detected, in SNS, by optical means<sup>1</sup> provides this method with additional informative channels. Specifically, studying the spin-noise power dependence on the probe light wavelength makes it possible to identify the type of broadening (homogeneous or inhomogeneous) of optical transitions [9,10]. Temporal modulation of the probe beam (e.g., shaping the ultrashort optical pulses) allows one to extend the range of the detected noise signals up to microwave frequencies [11]. The use of tightly focused probe beams provides an opportunity to detect the noise signals with a high spatial resolution and even to perform three-dimensional tomography of magnetic properties of materials [12]. The range of objects of SNS is not restricted to solid-state systems. Nowadays, this method is widely applied to the study of atomic gases [13], from which SNS originated [14].

The magnetic state of a material (magnetization), in SNS, is monitored by polarization plane rotation of the probe beam transmitted through the sample. It is assumed, in these measurements, that the detected angle of the polarization

plane rotation is proportional to the total magnetization of the illuminated volume of the sample. This is considered to be valid even for spontaneous spatiotemporal stochastic fluctuations of the magnetization detected in SNS. This simple picture is commonly used to interpret experimental data in SNS. In a consistent analysis, however, a polarimetric signal detected in SNS should be regarded as a result of scattering of the probe light by the randomly gyrotropic medium [15]. Such an analysis performed in Ref. [16] allowed us to justify the above simple picture and, in addition, to propose a two-beam modification of the SNS that makes it possible to observe both temporal and spatial correlations in magnetization of the illuminated region of the medium. In Ref. [16] we restricted our treatment to the case of thin samples (compared to the Rayleigh length of the probe beam), typical for experiments with solid-state samples. In this paper we consider a more general case of a volume medium with moving spin carriers (more typical for atomic vapors). In the first part of the paper, which is a continuation of the work in Ref. [16], we analyze the formation of the SNS signals for the samples with the thickness exceeding the Rayleigh length of the focused light beams. We show that the noise signal ceases to increase with the sample thickness when it substantially exceeds the Rayleigh length. For the case of the two-beam arrangement, we derive the expression for the spin-noise signal which shows that this signal is defined by the fluctuations of the gyrotropy only in the region of the overlap of the beams used. We also obtain an explicit expression for the spin-noise signal in the medium with diffusion of spin particles. Our estimates show that atomic diffusion in gaseous systems may drastically suppress the noise signal created by the auxiliary beam and thus hinder its observation.

The paper is organized as follows. In Sec. I, in the single-scattering approximation, we derive the expression for the noise polarimetric signal from the sample transilluminated by

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<sup>1</sup>This is performed by measuring polarization modulation of the probe beam passing through the sample under study.

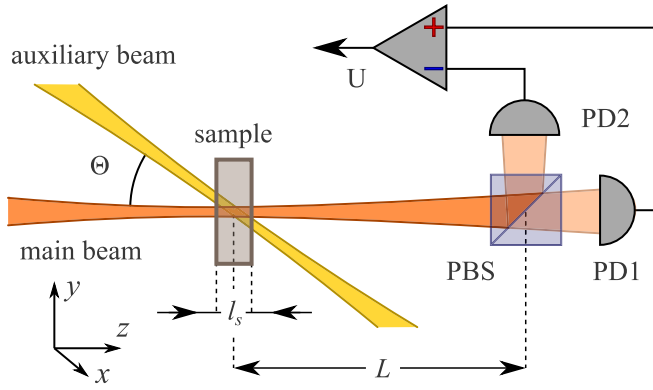


FIG. 1. Two-beam experimental arrangement. Here PBS denotes the polarization beam splitter and PD1 and PD2 are photodetectors.

two coherent laser beams (referred to as main and auxiliary), with only one of them (the main) hitting the detector [Eq. (13)]. In Sec. II we obtain relationships for the gyrotropy noise power spectrum detected using SNS. We present calculations of these spectra for samples of arbitrary thickness in the framework of the model of resting gyrotropic particles and of the diffusion model. We show that amplitude of the noise spectrum becomes independent of the sample thickness when the latter exceeds the Rayleigh length of the beam [Eq. (20)]. We also describe the effect of time-of-flight broadening of the spectrum arising in the diffusion model and present a simple experimental illustration of the conclusions using as a model object a thick cell with Cs atoms in a buffer-gas atmosphere. In Sec. III we present an analysis of signals observed in the two-beam arrangement of SNS [16]. For the contribution to the noise spectrum associated with the auxiliary beam, we obtain an expression that takes into account diffusion of the gyrotropic particles [Eq. (37)]. Recommendations are given regarding the choice of the systems where the above signal can be observed. The results of the work are summarized in the Conclusion.

## II. POLARIMETRIC SIGNAL FROM A RANDOMLY GYROTROPIC SAMPLE: THE TWO-BEAM ARRANGEMENT

In this section we present the solution of a problem typical for noise spectroscopy. Let us consider a weakly gyrotropic sample with spatial distribution of the gyration vector described by the function  $\mathbf{G}(\mathbf{R})$ , with  $|\mathbf{G}(\mathbf{R})| \ll 1$ . The sample is probed with two Gaussian beams with a frequency  $\omega$  (Fig. 1), for which the sample is transparent. One of the beams (hereafter referred to as main), after passing through the sample, hits the differential polarimetric detector comprised of a polarization beam splitter and two photodetectors. The total output signal is obtained as a difference of signals of the two detectors. The second beam (hereafter referred to as auxiliary) also passes through the sample, but does not hit the detector. Electric fields of the main and auxiliary beams will be denoted, respectively, by  $\mathbf{E}_0(\mathbf{R})$  and  $\mathbf{E}'_0(\mathbf{R})$ . We assume that the detector is initially balanced, i.e., polarization of the main beam is chosen so that, in the absence of the sample [at  $\mathbf{G}(\mathbf{R}) \equiv 0$ ], the output signal of the detector is zero. Our task is to find the gyrotropy-related increment of the output signal  $\delta U$  (in what follows, just *signal*)

in the first order of gyrotropy  $\mathbf{G}(\mathbf{R})$ . A similar problem for thin (compared to the Rayleigh length) samples was considered in Ref. [16]. Below we present the solution of this problem for samples of arbitrary length.

The signal  $\delta U$  arises due to the fact that at  $\mathbf{G}(\mathbf{R}) \neq 0$  the beam hitting the detector contains not only the field of the main beam, but also the field  $\mathbf{E}_1(\mathbf{R})$  that appears as a result of scattering of the main or auxiliary beam by the sample. Since we neglect any optical nonlinearity, these two fields may be calculated independently and the signal  $\delta U$  may be represented as a sum of two contributions related to scattering of the main and auxiliary beams. Since the detector is permanently irradiated by the main beam, detection of these fields occurs in the regime of heterodyning, with the role of local oscillator played by the field of the main beam.

In what follows we will use complex electromagnetic fields with time dependence in the form  $e^{-i\omega t}$  assigning a physical sense to their real parts (which will be denoted by calligraphic letters). The calculations will be performed in the coordinate system with its  $x$  and  $y$  axes aligned along principal directions of the polarization beam splitter and  $z$  axis collinear with the main beam. The coordinate origin is located in the region of the sample, with its characteristic size  $l_s$  being much smaller than the distance from the photodetector  $L$ :  $l_s \ll L$  (Fig. 1). For the signal  $\delta U$  we are interested in, we will use the expression [16]

$$\delta U = \frac{\omega}{\pi} \text{Re} \left( \int_0^{2\pi/\omega} dt \int_{-l_x}^{l_x} dx \int_{-l_y}^{l_y} dy [\mathcal{E}_{x0}(x, y, L) E_{x1}(x, y, L) - \mathcal{E}_{y0}(x, y, L) E_{y1}(x, y, L)] \right). \quad (1)$$

Here the integration over  $x$  and  $y$ , for products of components of the complex field of scattering  $\mathbf{E}_1(x, y, L)$  and the real part of the field of the main beam  $\mathcal{E}(x, y, L) \equiv \text{Re} \mathbf{E}_0(x, y, L)$ , is performed over the effective photosensitive surface of the detector  $2l_x \times 2l_y$ , located at a distance  $L$  from the sample along the main beam propagation direction (Fig. 1). The integration over  $t$  corresponds to averaging over the period of optical oscillations.

Below, following [16], we will calculate the field of scattering produced by the auxiliary beam [we will denote it, as before, by  $\mathbf{E}_1(\mathbf{R})$ ] and the related polarimetric signal denoted by  $\delta U_t$ . As shown in Ref. [16], this field satisfies the inhomogeneous Helmholtz equation

$$\nabla^2 \mathbf{E}_1 + k^2 \mathbf{E}_1 = -4\pi k^2 \alpha(\mathbf{r}) \mathbf{E}'_0(\mathbf{r}) \equiv -4\pi k^2 \mathbf{P}'(\mathbf{r}). \quad (2)$$

Here  $k \equiv \omega/c$  ( $c$  is the speed of light),  $\alpha(\mathbf{r})$  is the polarizability tensor of the gyrotropic medium [connected with the gyration vector as  $\alpha_{ik}(\mathbf{r}) = i \varepsilon_{ikj} G_j(\mathbf{r})$ , with  $\varepsilon_{ijk}$  the unit antisymmetric tensor],  $\mathbf{P}'(\mathbf{r})$  is the sample polarization induced by the field  $\mathbf{E}'_0(\mathbf{r})$  of the auxiliary beam, and  $\nabla^2 = \partial^2/\partial x^2 + \partial^2/\partial y^2 + \partial^2/\partial z^2$  is the Laplace operator. The solution of Eq. (2) is obtained using the Green's function  $\Gamma(\mathbf{r})$  of the Helmholtz operator  $\Gamma(\mathbf{r}) = -e^{ikr}/4\pi r$  and has the form

$$\mathbf{E}_1(\mathbf{r}) = k^2 \int \frac{e^{ik|\mathbf{r}-\mathbf{R}|}}{|\mathbf{r}-\mathbf{R}|} \mathbf{P}'(\mathbf{r}) d^3\mathbf{R}. \quad (3)$$

For further calculations, it is convenient to introduce the vector function  $\Phi(\mathbf{R})$  with the components defined by the expression<sup>2</sup>

$$\Phi_i(\mathbf{R}) \equiv \int_S dx dy \mathcal{E}_{i0}(x, y, z) \frac{e^{ik|\mathbf{r}-\mathbf{R}|}}{|\mathbf{r}-\mathbf{R}|} \Big|_{z=L},$$

$$i = x, y, \mathbf{r} = (x, y, z) \quad (4)$$

and auxiliary functions  $\Phi_i^\pm(\mathbf{R})$ ,

$$\Phi_i(\mathbf{R}) \equiv \Phi_i^+(\mathbf{R})e^{-i\omega t} + \Phi_i^-(\mathbf{R})e^{i\omega t}. \quad (5)$$

Using Eqs. (1), (3), and (4), we can obtain the following equation for the contribution  $\delta U_t$  into the output signal associated with the auxiliary beam:

$$\delta U_t = k^2 \frac{\omega}{\pi} \text{Re} \int_0^{2\pi/\omega} dt \int d^3\mathbf{R} [P_x^t(\mathbf{R})\Phi_x(\mathbf{R}) - P_y^t(\mathbf{R})\Phi_y(\mathbf{R})]. \quad (6)$$

By substituting  $\Phi(\mathbf{R})$  into this equation in the form of Eq. (5) and taking into account that  $\mathbf{P}^t(\mathbf{r}) \sim e^{-i\omega t}$ , we can ensure that, after integration over time, only terms containing  $\Phi_{x,y}^-(\mathbf{R})$  survive in Eq. (6):

$$\delta U_t = 2k^2 \text{Re} \int d^3\mathbf{R} [\Phi_x^-(\mathbf{R})P_x^t(\mathbf{R}) - \Phi_y^-(\mathbf{R})P_y^t(\mathbf{R})] e^{i\omega t}. \quad (7)$$

The factor  $e^{i\omega t}$  eliminates the time dependence of the field  $\mathbf{P}^t(\mathbf{r})$ . Let us write explicit expressions for the fields of the main  $\mathbf{E}_0(\mathbf{r})$  and auxiliary  $\mathbf{E}_0^t(\mathbf{r})$  Gaussian beams [16],

$$\mathbf{E}_0(\mathbf{r}) = e^{i(kz-\omega t)} \sqrt{\frac{8W}{c}} \frac{kQ}{(2k + iQ^2z)} \exp\left[-\frac{kQ^2(x^2+y^2)}{2(2k + iQ^2z)}\right] \mathbf{d}$$

$$\equiv \mathbf{A}_0(\mathbf{r})e^{-i\omega t}, \quad (8)$$

$$\mathbf{E}_0^t(\mathbf{r}) = e^{i(kZ-\omega t+\phi_t)} \sqrt{\frac{8W_t}{c}} \frac{kQ}{(2k + iQ^2Z)} \times \exp\left[-\frac{kQ^2(X^2+Y^2)}{2(2k + iQ^2Z)}\right] \mathbf{d}_t \equiv \mathbf{A}_0^t(\mathbf{r})e^{-i\omega t}, \quad (9)$$

where

$$\mathbf{r} = (x, y, z), \quad \begin{pmatrix} X \\ Y \\ Z \end{pmatrix} \equiv \hat{R}\mathbf{r} + \delta\mathbf{r}, \quad (10)$$

$$\hat{R} \equiv \begin{pmatrix} 1 & 0 & 0 \\ 0 & \cos \Theta & \sin \Theta \\ 0 & -\sin \Theta & \cos \Theta \end{pmatrix}.$$

Here  $W$  and  $W_t$  are the intensities of the main and auxiliary beams, respectively. The parameter  $Q$  is connected to the beam radius in the waist  $\rho_c$  by the relation  $Q \equiv 2/\rho_c$ . Polarization of the main and auxiliary beams is specified by the Jones vectors

$\mathbf{d}$  and  $\mathbf{d}_t$  lying in the planes perpendicular to the propagation directions of the beams. The sense of the angle  $\Theta$  is made clear by Fig. 1 and, as in Ref. [16], we assume that  $\Theta < 1$ . The parameters  $\delta\mathbf{r}$  and  $\phi_t$  describe, respectively, the spatial and phase shifts of the auxiliary beam with respect to the main one. In Eqs. (8) and (9) we introduced time-independent amplitudes of the fields of the main and auxiliary beams  $\mathbf{A}_0(\mathbf{r})$  and  $\mathbf{A}_0^t(\mathbf{r})$ . Using Eq. (2) to express polarization  $\mathbf{P}^t(\mathbf{r})$  through the field of the auxiliary beam (9), we obtain, with the aid of (7), the expression for the detected signal

$$\delta U_t = 2k^2 \text{Re} \int d^3\mathbf{R} [\Phi_x^-(\mathbf{R})\alpha_{xx}(\mathbf{R})A_{0x}^t(\mathbf{R}) + \Phi_x^-(\mathbf{R})\alpha_{xy}(\mathbf{R})A_{0y}^t(\mathbf{R}) - \Phi_y^-(\mathbf{R})\alpha_{yx}(\mathbf{R})A_{0x}^t(\mathbf{R}) - \Phi_y^-(\mathbf{R})\alpha_{yy}(\mathbf{R})A_{0y}^t(\mathbf{R})]. \quad (11)$$

Now we use the result of [17] showing that the function  $\Phi_i^-(\mathbf{R})$  can be expressed through the main beam amplitude  $\mathbf{A}_0(\mathbf{r})$  as follows (see footnote 2):

$$\Phi_i^-(\mathbf{R}) = -\frac{i\pi}{k} A_{0i}^*(\mathbf{R}), \quad i = x, y. \quad (12)$$

By substituting Eq. (12) into Eq. (11) and taking into account that, in the considered case of gyrotropic sample, the polarizability tensor has the form  $\alpha_{ij} = i\varepsilon_{ijk}G_k(\mathbf{R})$  ( $\varepsilon_{ijk}$  is the unit antisymmetric tensor), we obtain the following final expression for the polarimetric signal  $\delta U_t$  from the gyrotropic sample illuminated by the main and auxiliary light beams:

$$\delta U_t = 2\pi k \text{Re} \int d^3\mathbf{R} [A_{0x}^*(\mathbf{R})A_{0y}^t(\mathbf{R}) + A_{0y}^*(\mathbf{R})A_{0x}^t(\mathbf{R})] G_z(\mathbf{R}). \quad (13)$$

Equation (13) shows that the polarimetric signal associated with the auxiliary beam [ $\mathbf{A}_0^t(\mathbf{r})$ ], detected by its mixing with the wave of the main beam [ $\mathbf{A}_0(\mathbf{r})$ ], is controlled by gyrotropy of the sample only in the region of overlap of the two beams. Recall that Eq. (13) describes the contribution to the polarimetric signal arising due to scattering of the auxiliary beam. Along with this contribution, there always exists the contribution related to scattering of the main beam observed in the conventional single-beam arrangement, when the auxiliary beam is absent. To calculate this contribution, one just has to set  $\mathbf{A}_0^t(\mathbf{r}) = \mathbf{A}_0(\mathbf{r})$  in Eq. (13). The total signal in the two-beam arrangement is obtained by summation of the two contributions.

### III. NOISE POWER SPECTRUM IN THE SINGLE-BEAM ARRANGEMENT

In this section we calculate the spin-noise signal for the conventional single-beam geometry. Polarization of the main beam (which is the only one in this arrangement) is specified by the Jones vector  $\mathbf{d} = (\cos \phi, \sin \phi, 0)$  (in the coordinate system introduced above). Using Eq. (8), we can show that dependence of the beam radius  $\rho(z)$  (at 1/e-level of the field squared) on

<sup>2</sup>It is noteworthy that the function  $\Phi(\mathbf{r})$  has the sense of the field created by the source that is distributed over the surface of the detector and has the density  $\mathcal{E}_0(x, y, L)$ . For this reason, the field  $\Phi(\mathbf{r})$  is similar to that of the main beam  $\mathcal{E}_0(\mathbf{r})$ . An explicit expression for  $\Phi(\mathbf{r})$  was derived [17] and is presented below Eq. (12).

the coordinate  $z$  has the form

$$\rho(z) \equiv \sqrt{\frac{4k^2 + Q^4 z^2}{2k^2 Q^2}} = \frac{\rho_c}{\sqrt{2}} \sqrt{1 + \frac{z^2}{z_c^2}} = \frac{\lambda}{\sqrt{2\pi\rho_c}} \sqrt{z_c^2 + z^2}. \quad (14)$$

Here  $\rho_c = 2/Q$ ,  $\lambda \equiv 2\pi/k$  is the light wavelength, and  $z_c \equiv \pi\rho_c^2/\lambda$  is the Rayleigh length (half-length of the quasicylindrical region of the Gaussian beam). As was already noted, the polarimetric signal in the single-beam arrangement (denoted by  $u_1$ ) can be calculated using Eq. (13), by setting in it  $\mathbf{A}_0^t(\mathbf{r}) = \mathbf{A}_0(\mathbf{r})$ . With allowance for (8) and (14), we have

$$u_1 = \sin 2\phi \frac{8\pi kW}{c} \int \frac{d^3\mathbf{R} G_z(\mathbf{R})}{\rho^2(z)} \exp\left[-\frac{x^2 + y^2}{\rho^2(z)}\right]. \quad (15)$$

Since the gyrotropy of the sample is connected with its magnetization, temporal fluctuations of the latter give rise to fluctuations of the gyrotropy:  $G_z(\mathbf{R}) \rightarrow G_z(\mathbf{R}, t)$ . In a typical experiment on spin-noise spectroscopy, one observes the noise power spectrum of the gyrotropy  $\mathcal{N}(\nu)$ , which is determined by the Fourier transform of the correlation function of the polarimetric signal  $\mathcal{N}(\nu) = \int dt \langle u_1(t)u_1(0) \rangle e^{i\nu t}$ . Using Eq. (15), we obtain, for the gyrotropy noise power spectrum, the expression

$$\begin{aligned} \mathcal{N}(\nu) &= \sin^2 2\phi \left[\frac{8\pi kW}{c}\right]^2 \int dt e^{i\nu t} \int \frac{d^3\mathbf{R} d^3\mathbf{R}'}{\rho^2(z)\rho^2(z')} \\ &\times \exp\left[-\left(\frac{x^2 + y^2}{\rho^2(z)} + \frac{x'^2 + y'^2}{\rho^2(z')}\right)\right] \\ &\times \langle G_z(\mathbf{R}, t)G_z(\mathbf{R}', 0) \rangle. \end{aligned} \quad (16)$$

The correlation function of the gyrotropy  $\langle G_z(\mathbf{R}, t)G_z(\mathbf{R}', 0) \rangle$  entering this equation is calculated on the basis of one of the models of the sample under study. Most frequently, the gyrotropy is implied to be created by ensembles of gyrotropic particles (e.g., paramagnetic atoms) and is described by the expression

$$G_z(\mathbf{R}, t) = \sum_i g_i(t)\delta(\mathbf{R} - \mathbf{r}_i(t)), \quad (17)$$

where  $g_i(t)\delta(\mathbf{R} - \mathbf{r}_i(t))$  is the contribution of the  $i$ th particle to the total gyrotropy of the sample and  $\mathbf{r}_i(t)$  is the coordinate of the  $i$ th particle that may be time dependent. The function  $g_i(t)$  can be considered proportional to the magnetic moment of the  $i$ th particle, with the proportionality factor being generally dependent on the frequency  $\omega$  of the light beam.

### A. Model of resting paramagnetic particles

We start our treatment with the simplest model that implies that the sample consists of  $N$  identical particles at rest, randomly distributed over the volume  $V$  with the density  $\sigma$ .<sup>3</sup> In this case, the gyrotropy is given by Eq. (17) with time-independent coordinates of the particles  $\mathbf{r}_i(t) \rightarrow \mathbf{r}_i$ . The second assumption of this simple model is that the functions  $g_i(t)$  are supposed

to be random independent quantities so that  $\langle g_i(t)g_k(t') \rangle = \delta_{ik}\langle g(t-t')g(0) \rangle$ . Here the function  $\langle g(t-t')g(0) \rangle$  is the same for all particles. Under these assumptions, for the correlator entering Eq. (16), we can obtain the expression [16]  $\langle G(\mathbf{R}, t)G(\mathbf{R}', 0) \rangle = \sigma \langle g(t)g(0) \rangle \delta(\mathbf{R} - \mathbf{R}')$ . By substituting this expression into (16) and calculating the integrals with  $\delta$  functions, we obtain, for the noise power spectrum, the expression

$$\begin{aligned} \mathcal{N}(\nu) &= 32\sigma\pi^3 \sin^2 2\phi \left[\frac{kW}{c}\right]^2 \int dt e^{i\nu t} \langle g(t)g(0) \rangle \\ &\times \int \frac{dz}{\rho^2(z)}. \end{aligned} \quad (18)$$

Since  $1/\rho^2(z) \sim 1/[z^2 + z_c^2]$  [see Eq. (14)], the main contribution to the signal is made by the region of the sample in the vicinity of the beam waist, where  $|z| < z_c$ . This makes it possible to use the SNS method for tomography [12], with the spatial resolution in the longitudinal direction, as expected, being determined by the Rayleigh length  $z_c$  of the probe beam.

Let us denote the bounds of the sample along the light beam (i.e., along the  $z$  axis) by  $z_1$  and  $z_2$ . Then, using Eq. (14) for the beam radius  $\rho(z)$  and integrating over  $z$  in Eq. (18), we obtain

$$\begin{aligned} \mathcal{N}(\nu) &= 32\sigma\pi^3 k^3 \sin^2 2\phi \left[\frac{W}{c}\right]^2 \left[ \arctan \frac{z_2}{z_c} - \arctan \frac{z_1}{z_c} \right] \\ &\times \int dt e^{i\nu t} \langle g(t)g(0) \rangle. \end{aligned} \quad (19)$$

It follows from Eq. (19) that with increasing thickness of the sample ( $z_1 \rightarrow -\infty$  and  $z_2 \rightarrow \infty$ ) the noise signal is saturated approaching the limiting value

$$\begin{aligned} \mathcal{N}_\infty(\nu) &= 32\pi^4 \sigma k^3 \sin^2 2\phi \left[\frac{W}{c}\right]^2 \int dt e^{i\nu t} \langle g(t)g(0) \rangle \\ &= 32\pi^4 \sigma k^3 T_2 \left[\frac{W}{c}\right]^2 \left[ \frac{\sin^2[2\phi]\langle g^2 \rangle}{1 + [\nu - \omega_L]^2 T_2^2} \right. \\ &\quad \left. + \frac{\sin^2[2\phi]\langle g^2 \rangle}{1 + [\nu + \omega_L]^2 T_2^2} \right]. \end{aligned} \quad (20)$$

This expression corresponds to the correlator  $\langle g(t)g(0) \rangle = \langle g^2 \rangle e^{-|t|/T_2} \cos \omega_L t$ .<sup>4</sup> To illustrate the above formulas, we have measured experimentally the dependence of the noise signal area  $\int \mathcal{N}(\nu) d\nu$  of cesium vapor on the position  $z$  of the cell with respect to the beam waist (Fig. 2). The measurements were performed using a focused laser beam with the wavelength  $\lambda = 0.85 \mu\text{m}$ . The length of the cell  $2l_c$  was 2 cm. In accordance with Eq. (19), the measured dependence should have the form  $\sim \arctan[z - l_c]/z_c - \arctan[z + l_c]/z_c$ . As can be seen from Fig. 2, the experimental dependence is well approximated by this formula, with the best-fit value of the parameter  $z_c$  ( $z_c = 3.3 \times 10^{-3}$  m) well correlated with characteristics of the laser beam used. In spite of the fact that

<sup>3</sup>Such a model, for samples thin compared to the Rayleigh length, was considered in Ref. [16].

<sup>4</sup>Such a correlator corresponds to the case when gyrotropy of the sample is created by the system of paramagnetic particles in a magnetic field; the quantities  $\omega_L$  and  $T_2$  are, respectively, the Larmor frequency of the effective spin and its dephasing time.

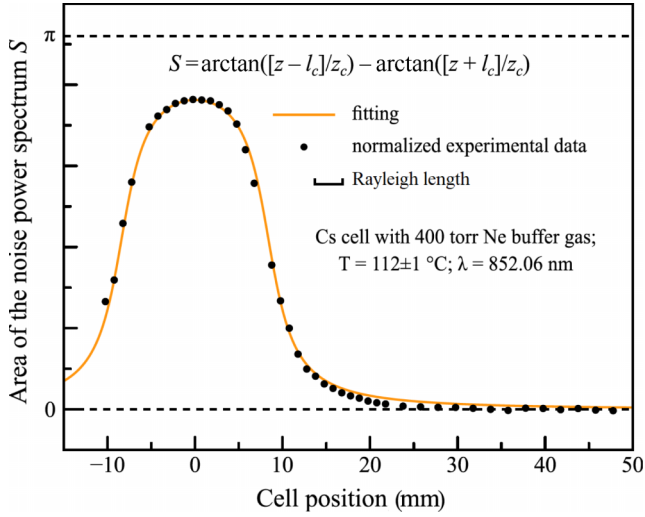


FIG. 2. Variation of area of the gyrotropy noise power spectrum  $S$  of Cs atoms in earth's magnetic field with displacement of the cell  $z$  with respect to the light beam waist. The solid curve denotes the theory and circles show the experiment. The wavelength of the light beam and its waist radius are, respectively,  $\lambda = 0.85 \text{ nm}$  and  $\rho_c = 30 \text{ }\mu\text{m}$ .

the cell thickness considerably exceeded the Rayleigh length  $z_c$  (shown in Fig. 2 by a horizontal segment), the value of the noise signal appeared to be noticeably (by  $\sim 25\%$ ) smaller than the limiting value (indicated in Fig. 2 by a horizontal line at the level  $\pi$ ). At the same time, it can be seen from the experimental illustration presented and Eq. (19) that, for the thickness of the sample  $2l_c$  exceeding the Rayleigh length by a factor of 4–5, further reduction of the beam radius  $\rho_c$  (with a corresponding decrease of the Rayleigh length  $z_c = \pi\rho_c^2/\lambda$ ) does not lead to a substantial increase of the noise signal. Thus, it makes sense to decrease the radius of the probe beam for an increasing value of the spin-noise signal only for samples that are thin compared to the Rayleigh length of the light beam.

### B. Diffusion model

Our assumption that the gyrotropy is created by resting particles is plausible for solid materials with embedded paramagnetic atoms giving rise to the gyrotropy. For semiconductor samples, with the gyrotropy created by the moving charge carriers, as well as for gaseous systems, this assumption may be incorrect. It is natural to take into account the motion of gyrotropic particles in such systems using a diffusion model, with  $N$  particles randomly moving in a finite volume  $V$  [18]. In this case, Eq. (17) for the gyrotropy remains valid.

Quantitative analysis and experimental study of the diffusion effects in SNS of gaseous systems were recently presented in Ref. [19]. In this section, with the aid of the relationships obtained above, we will reproduce the main results of [19], treating the spin-noise signal as a result of scattering of a Gaussian probe beam. In addition, the notions introduced in this section will be used below to calculate the signal in the two-beam arrangement, when intuitive considerations about signal formation are not as self-evident as in conventional single-beam geometries.

If one considers a semiconductor system with a relatively low electron density in the conduction band or a gaseous system with diffusion motion of gyrotropic atoms occurring in a dense medium of nongyrotropic buffer gas, then the contribution of each particle to the gyrotropy of the sample can be considered as independent of other particles. In this case, for the correlation function of gyrotropy entering Eq. (16) for the noise power spectrum, we can write the chain of equalities

$$\begin{aligned} \langle G_z(\mathbf{R}, t) G_z(\mathbf{R}', 0) \rangle &= \sum_{ik} \langle g_i(t) g_k(0) \delta(\mathbf{R} - \mathbf{r}_i(t)) \delta(\mathbf{R}' - \mathbf{r}_k(0)) \rangle \\ &= \langle g(t) g(0) \rangle \sum_i \langle \delta(\mathbf{R} - \mathbf{r}_i(t)) \delta(\mathbf{R}' - \mathbf{r}_i(0)) \rangle \\ &= N \langle g(t) g(0) \rangle \langle \delta(\mathbf{R} - \mathbf{r}_1(t)) \delta(\mathbf{R}' - \mathbf{r}_1(0)) \rangle \\ &= N \langle g(t) g(0) \rangle \langle \delta(\mathbf{R} - \mathbf{R}' - \mathbf{r}(t)) \delta(\mathbf{R}' - \mathbf{r}_1(0)) \rangle, \quad (21) \end{aligned}$$

where  $\mathbf{r}(t) \equiv \mathbf{r}_1(t) - \mathbf{r}_1(0)$  is the vector of diffusion displacement of the particle from the starting point  $\mathbf{r}_1(0)$ . Here we assume that fluctuations of gyrotropy for each particle are independent of its diffusion motion<sup>5</sup> and suppose, as before, that  $\langle g_i(t) g_k(t') \rangle = \delta_{ik} \langle g(t - t') g(0) \rangle$ . Thus, the problem is reduced to studying diffusion motion of any single particle (e.g., the first one). The coordinate  $\mathbf{r}_1(0)$  of this particle at  $t = 0$  may acquire, with equal probability, any value within the volume  $V$ . Therefore, averaging of the last  $\delta$  function over  $\mathbf{r}_1$  yields the factor  $1/V$ . By virtue of statistical uniformity of the sample, the distribution function  $P(\mathbf{r}, t)$  of the vector of diffusion displacement  $\mathbf{r}(t) \equiv \mathbf{r}_1(t) - \mathbf{r}_1(0)$  of the chosen particle does not depend on the starting point  $\mathbf{r}_1(0)$  and is defined by the diffusion equation with the initial condition  $P(\mathbf{r}, 0) = \delta(\mathbf{r})$ ,

$$\frac{\partial P}{\partial t} = D \nabla^2 P, \quad P(\mathbf{r}, 0) = \delta(\mathbf{r}), \quad (22)$$

where  $D$  is the diffusion coefficient and  $\mathbf{r} = (x, y, z)$ . Thus, the chain of equalities (21) can be continued as follows:

$$\begin{aligned} \langle G(\mathbf{R}, t) G(\mathbf{R}', 0) \rangle &= \frac{N}{V} \langle g(t) g(0) \rangle \langle \delta(\mathbf{R} - \mathbf{R}' - \mathbf{r}(t)) \rangle \\ &= \sigma \langle g(t) g(0) \rangle \langle \delta(\mathbf{R} - \mathbf{R}' - \mathbf{r}(t)) \rangle \\ &= \sigma \langle g(t) g(0) \rangle P(\mathbf{R} - \mathbf{R}', t). \quad (23) \end{aligned}$$

Here  $\sigma = N/V$  is the density of the particles. Standard solution of the problem (22) leads to the following expression for the distribution function  $P(\mathbf{r}, t)$ :

$$P(\mathbf{r}, t) = \frac{1}{8(\pi Dt)^{3/2}} \exp \left[ -\frac{r^2}{4Dt} \right]. \quad (24)$$

Substituting this function into (23), we obtain, for the gyrotropy correlator in the presence of diffusion, the final expression

$$\langle G_z(\mathbf{R}, t) G_z(\mathbf{R}', 0) \rangle = \frac{\sigma \langle g(t) g(0) \rangle}{8(\pi D|t|)^{3/2}} \exp \left[ -\frac{|\mathbf{R} - \mathbf{R}'|^2}{4D|t|} \right]. \quad (25)$$

<sup>5</sup>This assumption is arguable, but for the simplest analysis of the diffusion effects it is acceptable.

Here we took into account the parity of the correlation function. By substituting this expression into Eq. (16) for the noise power spectrum we obtain

$$\begin{aligned} \mathcal{N}(\nu) &= \sin^2 2\phi \frac{8\sqrt{\pi}\sigma}{D^{3/2}} \left[ \frac{kW}{c} \right]^2 \int dt e^{i\nu t} \frac{\langle g(t)g(0) \rangle}{|t|^{3/2}} \\ &\times \int \frac{d^3\mathbf{R} d^3\mathbf{R}'}{\rho^2(z)\rho^2(z')} \exp \left[ -\frac{x^2+y^2}{\rho^2(z)} - \frac{x'^2+y'^2}{\rho^2(z')} \right] \\ &\times \exp \left[ -\frac{|\mathbf{R}-\mathbf{R}'|^2}{4D|t|} \right]. \end{aligned} \quad (26)$$

Here  $\mathbf{R} = (x, y, z)$  and  $\mathbf{R}' = (x', y', z')$ . The integrals over  $x, y, x', y'$  are reduced to Gaussian by appropriate rotations of the coordinate system in the planes  $xy$  and  $x'y'$  that eliminate, in the exponent, the terms  $\sim xy$  and  $\sim x'y'$ . By calculating these Gaussian integrals, we arrive at the expression for the noise power spectrum

$$\begin{aligned} \mathcal{N}(\nu) &= \frac{32\sigma\pi^{5/2}}{D^{1/2}} \sin^2 2\phi \left[ \frac{kW}{c} \right]^2 \int \frac{dt}{|t|^{1/2}} e^{i\nu t} \\ &\times \int \frac{dz dz' \langle g(t)g(0) \rangle}{4D|t| + \rho^2(z) + \rho^2(z')} \exp \left[ -\frac{(z-z')^2}{4D|t|} \right], \end{aligned} \quad (27)$$

which transforms to Eq. (18) at  $D \rightarrow 0$ .

Equation (27) can be simplified assuming that the diffusion length for the characteristic decay time of the correlator  $\langle g(t)g(0) \rangle$  is smaller than the Rayleigh length. In the situation typical for SNS, when the correlator  $\langle g(t)g(0) \rangle$  decreases exponentially,  $\langle g(t)g(0) \rangle = \langle g^2 \rangle e^{-|t|/T_2} \cos \omega_L t$  (see footnote 4), the above condition can be written in the form  $\sqrt{DT_2} \ll z_c$  [see Eq. (14)]. In this case, we may set, in Eq. (27),  $\rho(z) \approx \rho(z')$ , perform integration over  $z'$ , and obtain the simplified expression for the noise power spectrum

$$\begin{aligned} \mathcal{N}(\nu) &= 32\pi^3 \sigma \sin^2 2\phi \left[ \frac{kW}{c} \right]^2 \int dt dz \frac{e^{i\nu t} \langle g(t)g(0) \rangle}{2D|t| + \rho^2(z)} \\ &\text{at } 2\sqrt{DT_2} < z_c, \end{aligned} \quad (28)$$

where  $\rho(z)$  is defined by Eq. (14). It can be seen from this relationship that, in the region of the sample where  $\rho(z) < \sqrt{2DT_2}$  (provided that such a region exists), the time dependence of the integrand deviates from  $\sim \langle g(t)g(0) \rangle$ , which is usually exponential. As a result, the shape of the noise power spectrum deviates from Lorentzian and the noise spectrum reveals the so-called time-of-flight broadening [18]. If the beam is so broad that  $\rho_c > \sqrt{DT_2}$ , then this effect proves to be suppressed and can be neglected. Estimates show that the conditions of applicability of Eq. (28) often are true in practice. Using Eq. (14) for the function  $\rho(z)$ , the integration over  $z$  in Eq. (28) can be performed analytically. Let us present the result for the case when the sample length is much larger than both the Rayleigh length and the diffusion length  $\sqrt{DT_2}$  for the time  $T_2$ :

$$\begin{aligned} \mathcal{N}(\nu) &= 32\pi^4 k^3 \rho_c \sigma \sin^2 2\phi \left( \frac{W}{c} \right)^2 \int dt \frac{e^{i\nu t} \langle g(t)g(0) \rangle}{\sqrt{4D|t| + \rho_c^2}} \\ &\text{at } 2\sqrt{DT_2} \ll z_c, l_s \gg z_c. \end{aligned} \quad (29)$$

As can be seen from Eq. (29), when the diffusion drift  $\sqrt{DT_2}$  for the time  $T_2$  is smaller than the beam radius  $\rho_c$ , the effects of diffusion can be neglected. Otherwise, the noise spectrum exhibits the time-of-flight broadening.

#### IV. TWO-BEAM NOISE SPECTROSCOPY

Above we presented calculations of the noise signals detected in the single-beam arrangement, traditional for SNS. Consider now the case when the beam that induces scattering and the beam that plays the role of local oscillator are different  $\mathbf{A}_0(\mathbf{R}) \neq \mathbf{A}'_0(\mathbf{R})$  [16] (Fig. 1). We will assume that waists of these two beams intersect in the region of the gyrotropic sample studied and we will analyze the problem under the following simplifying assumptions.

(i) Both beams propagate in the direction close to the  $z$  axis, the angle  $\Theta$  between the beams is small enough to make possible low-power approximations of its trigonometric functions, and the main components of the electric fields of the beams lie in the  $xy$  plane.

(ii) The angle  $\Theta$  is large enough not to make the length of the beam overlap larger than the Rayleigh length.

Appropriate quantitative conditions will be presented below. Let us choose the coordinate system so that both the beams (the main and auxiliary) lie in the  $yz$  plane (i.e., the beams are rotated with respect to each other around the  $x$  axis). Bearing in mind assumption (i), polarizations of the main and auxiliary beams are specified by the following two-dimensional (in the plane  $xy$ ) Jones vectors:

$$\mathbf{d} = \begin{pmatrix} \cos \phi \\ \sin \phi \end{pmatrix}, \quad \mathbf{d}' = \begin{pmatrix} \cos \eta \\ \sin \eta \end{pmatrix}. \quad (30)$$

Using assumption (ii), we can neglect, in Eqs. (8) and (9), the terms  $Q^2 z$  and  $Q^2 Z$  as compared with  $2k$ . After that, with the aid of Eq. (13), we obtain, for the polarimetric signal produced by the auxiliary beam [below referred to as  $u'_1(t)$ ], the following expression [ $\delta U_t \rightarrow u'_1(t)$ ]:

$$\begin{aligned} u'_1(t) &= \frac{16\pi k}{\rho_c^2 c} \sqrt{W W_t} \int d^3\mathbf{R} \cos \left( k\Theta y + \frac{k\Theta^2 z}{2} - \phi_t \right) \\ &\times \sin[\phi + \eta] \exp \left[ -2 \frac{x^2 + y^2 + yz\Theta + z^2\Theta^2/2}{\rho_c^2} \right] \\ &\times G_z(\mathbf{R}, t). \end{aligned} \quad (31)$$

When deriving this formula, we took into account the smallness of the angle  $\Theta$  [see the transformations (10)]. The exponential of the quadratic form of the coordinates in this formula is essentially nonzero in the region  $\sim \rho_c \times \rho_c$  (in the plane  $xy$ ) over the length  $\sim \rho_c/\Theta$  (along the  $z$  axis). Therefore, assumption (ii) can be expressed by the inequality  $\rho_c/\Theta < \pi\rho_c^2/\lambda$ . Keeping this in mind, we arrive at the conclusion that the above assumptions impose the following restrictions upon the angle  $\Theta$  between the beams:

$$\frac{\lambda}{\pi\rho_c} < \Theta < 1. \quad (32)$$

Typically,  $\rho_c \sim 30 \mu\text{m}$  at  $\lambda \approx 1 \mu\text{m}$ . Therefore, for validity of the calculations carried out in this section, the angle  $\Theta$  should meet the inequality  $10^{-2} < \Theta < 1$ , which can be easily satisfied in practice.

When calculating the noise power spectrum detected in the two-beam arrangement, one has to take into account that the total polarimetric signal  $\delta U(t)$ , in this case, is the sum  $\delta U(t) = u_1(t) + u_1'(t)$ , with  $u_1(t)$  and  $u_1'(t)$  given by Eqs. (15) and (31), respectively. Hence, the formula for the noise power spectrum  $\mathcal{N}(\nu) = \int e^{i\nu t} \langle \delta U(0) \delta U(t) \rangle dt$  will contain four contributions

$$\mathcal{N}(\nu) = \int dt [\langle u_1(t) u_1(0) \rangle + \langle u_1(t) u_1'(0) \rangle + \langle u_1'(t) u_1(0) \rangle + \langle u_1'(t) u_1'(0) \rangle] e^{i\nu t}. \quad (33)$$

If no special measures are taken to stabilize the relative phase  $\phi_t$  of the main and auxiliary beams, then it is natural to perform averaging over this phase, which will be below implied. Typically, both beams can be obtained by splitting the beam of the same laser. In this case, the averaging over the relative phase can be performed by means of the mirror, attached to the audio vibrator, placed in the channel of the auxiliary beam. As a result of this averaging, the cross correlators  $\langle u_1'(0) u_1(t) \rangle$  and  $\langle u_1'(t) u_1(0) \rangle$  will vanish. The first correlator  $\langle u_1(t) u_1(0) \rangle$  in Eq. (33) has been already calculated above [Eq. (16)]. It gives the noise spectrum observed in the single-beam arrangement. For this reason, in what follows we will consider the contribution to the noise spectrum related only to the auxiliary beam and controlled by the correlator  $\langle u_1'(t) u_1'(0) \rangle$ . Let us define this contribution by  $\mathcal{N}_i(\nu) \equiv \int dt \langle u_1'(t) u_1'(0) \rangle e^{i\nu t}$ . If the sample gyrotropy represents a random field statistically stationary in space and in time, then its correlation function depends only on the difference between its spatiotemporal arguments and can be represented in the form  $\mathcal{K}(\mathbf{R} - \mathbf{R}', t) \equiv \langle G(\mathbf{R}, t) G(\mathbf{R}', 0) \rangle$ . Using Eq. (31), we obtain for the correlator  $\langle u_1'(t) u_1'(0) \rangle$  the relation

$$\begin{aligned} \langle u_1'(t) u_1'(0) \rangle &= \frac{128\pi^2 k^2}{\rho_c^4 c^2} W W_t \sin^2[\phi + \eta] \int d^3\mathbf{R} d^3\mathbf{R}' \\ &\times \exp\left[-2 \frac{x^2 + y^2 + yz\Theta + z^2\Theta^2/2}{\rho_c^2}\right] \\ &\times \exp\left[-2 \frac{x'^2 + y'^2 + y'z'\Theta + z'^2\Theta^2/2}{\rho_c^2}\right] \\ &\times \cos\left[k\Theta(y - y') + \frac{k\Theta^2(z - z')}{2}\right] \\ &\times \mathcal{K}(\mathbf{R} - \mathbf{R}', t). \end{aligned} \quad (34)$$

Here the averaging over the relative phase of the beams  $\phi_t$  is performed.

Using Eq. (34) as a starting point, we can obtain a simpler approximate formula, suitable for estimating the SNS signals under experimental conditions typical for this method. Note that the exponential factors in Eq. (34) in fact shrink the integration region to the region of overlap between the main and auxiliary beams. The volume of this region  $V_o$  can be

evaluated in the following way:

$$\begin{aligned} V_o &\approx \int d^3\mathbf{R} \exp\left[-2 \frac{x^2 + y^2 + yz\Theta + z^2\Theta^2/2}{\rho_c^2}\right] \\ &= \frac{\pi^{3/2} \rho_c^3}{\sqrt{2}\Theta}. \end{aligned} \quad (35)$$

For this reason, in Eq. (34) we may restrict the region of integration over  $d^3\mathbf{R}$  and  $d^3\mathbf{R}'$  with the volume  $V_o$  and set the exponential factors to be equal to unity. After that, the integrand will appear to be dependent on the difference  $\mathbf{R} - \mathbf{R}'$ . Now let us pass to new variables  $\mathbf{r} \equiv \mathbf{R} - \mathbf{R}'$  and  $\mathbf{g} \equiv \mathbf{R} + \mathbf{R}'$ . The integral over  $\mathbf{g}$  will give the volume of integration  $V_o$ , and for the correlator (34) we can write the approximate formula

$$\begin{aligned} \langle u_1'(t) u_1'(0) \rangle &\approx \frac{\pi^{7/2} k^2}{\rho_c c^2} W W_t \sin^2[\phi + \eta] \\ &\times \int_{V_o} d^3\mathbf{r} \cos(\Delta\mathbf{k} \cdot \mathbf{r}) \mathcal{K}(\mathbf{r}, t), \end{aligned} \quad (36)$$

where

$$\Delta\mathbf{k} \equiv k\Theta \begin{pmatrix} 0 \\ 1 \\ \Theta/2 \end{pmatrix}$$

is the difference wave vector of the main and tilted beams, while the numerical factor is  $\varkappa = 16/\sqrt{2}$ . When the region of overlap of the beams is large compared to the gyrotropy correlation radius and spatial periods of cosine in (34),  $\lambda/2\pi\Theta$  (in the  $y$  direction) and  $\lambda/2\pi\Theta^2$  (in the  $z$  direction), then the integral in Eq. (36) coincides with the Fourier transform of the gyrotropy correlation function.

Using the relation (36), we can calculate the contribution to the gyrotropy noise power spectrum associated with the auxiliary beam in the presence of diffusion. For the correlation function of the gyrotropy, we use Eq. (25), in which we set  $\langle g(t)g(0) \rangle = \langle g^2 \rangle e^{-|t|/T_2} \cos \omega_L t$  (see footnote 4). Calculating the Fourier transform of Eq. (25) and substituting it into Eq. (36), we obtain

$$\begin{aligned} \mathcal{N}_i(\nu) &\approx \varkappa \frac{T_2^* \sigma \pi^{7/2} k^2}{\Theta \rho_c c^2} W W_t \left[ \frac{\sin^2[\phi + \eta] \langle g^2 \rangle}{1 + [\nu - \omega_L]^2 T_2^{*2}} \right. \\ &\left. + \frac{\sin^2[\phi + \eta] \langle g^2 \rangle}{1 + [\nu + \omega_L]^2 T_2^{*2}} \right], \end{aligned} \quad (37)$$

where

$$T_2^* \equiv \frac{T_2}{1 + k^2 \Theta^2 D T_2}. \quad (38)$$

As can be seen from this formula, diffusion leads to broadening of the noise spectrum and reduction of its amplitude, provided the diffusion length for the dephasing time  $\sqrt{D T_2}$  exceeds the spatial period of interference between the main and auxiliary beams  $1/k\Theta = \lambda/2\pi\Theta$ . In the opposite case (i.e., at  $\sqrt{D T_2} < \lambda/2\pi\Theta$ ), the contributions  $\mathcal{N}_i(\nu)$  [Eq. (37)] and  $\mathcal{N}(\nu)$  [Eq. (27)] have comparable amplitudes and spectral widths.

Now let us present arguments that allow us to believe that Eq. (37) works well even when the basic conditions of

its derivation are poorly satisfied. For this we present the result of consistent computation of the integral (34) with the correlation function of gyrotropy in the form (25). In this case, the integrand represents an exponential of some quadratic form of the integration variables. Such a form can be diagonalized with the proper orthogonal transformation of the coordinate system. After this, the integral (34) is reduced to a product of Gaussian integrals. Omitting cumbersome manipulations, we present the final result of such calculations

$$\begin{aligned} \langle u_1^t(t)u_1^t(0) \rangle &= \frac{8\pi^{7/2}k^2}{\rho_c^2 c^2} W W_t \sin^2[\phi + \eta] \frac{\sigma \langle g(t)g(0) \rangle}{(Dt)^{3/2}} \\ &\times \left[ 1 + \frac{\rho_c^2}{4Dt} \right]^{-1/2} \frac{\exp[-(M^{-1}h, h)/4]}{\sqrt{\det M}}, \end{aligned} \quad (39)$$

where the vector column  $h$  and the matrix  $M$  are defined by the relations

$$\begin{aligned} h &= k\Theta \begin{pmatrix} 1 \\ \Theta/2 \\ -1 \\ -\Theta/2 \end{pmatrix}, \quad M \equiv \begin{pmatrix} \alpha & \delta & \gamma & 0 \\ \delta & \beta & 0 & \gamma \\ \gamma & 0 & \alpha & \delta \\ 0 & \gamma & \delta & \beta \end{pmatrix}, \\ \alpha &\equiv \frac{2}{\rho_c^2} + \frac{1}{4Dt}, \quad \beta \equiv \frac{\Theta^2}{\rho_c^2} + \frac{1}{4Dt}, \quad \delta \equiv \frac{\Theta}{\rho_c^2}, \quad \gamma \equiv -\frac{1}{4Dt}. \end{aligned} \quad (40)$$

Calculations of the correlation functions of the polarimetric signal show that the results obtained using (37) and (39) at  $\rho_c > 3\lambda$  and  $0.05 < \Theta < 0.3$  practically coincide if we set in Eq. (37)  $\varkappa = 32$ .

Our efforts to observe the noise signal from cesium atoms (see the end of Sec. II A) associated with the auxiliary beam (Fig. 1) have failed. The reason for this failure is likely to be the following. Let us compare the amplitude of the noise signal (37) related to the auxiliary beam with that of the signal (20) detected in the single-beam arrangement. Using Eqs. (37) and (20), at  $\omega_L T_2 \gg 1$ , we obtain the relationship

$$\frac{\mathcal{N}_f(\omega_L)}{\mathcal{N}_\infty(\omega_L)} = \frac{1}{1 + k^2 \Theta^2 D T_2} \frac{\lambda}{2\pi^{3/2} \rho_c \Theta} \frac{W_t \sin^2[\phi + \eta]}{W \sin^2 2\phi}. \quad (41)$$

The two last factors can be made  $\sim 1$  by tuning the polarization and intensities of the main and auxiliary beams. The second factor describes the decrease of the noise signal in the two-beam arrangement resulting from incomplete overlap of the two beams. At  $\lambda \sim 1 \mu\text{m}$ ,  $\rho_c \sim 30 \mu\text{m}$ , and  $\Theta \sim 0.1$  rad, this factor is  $\sim 1/30$ . Finally, the first factor describes the decrease of the noise spectrum amplitude  $\mathcal{N}_f(\nu)$  associated with diffusion of the gyrotropic particles. Let us estimate this factor for our particular case of cesium atoms. Taking for the diffusion coefficient of Cs atoms in the buffer gas atmosphere the value  $D = 2 \times 10^{-5} \text{ m}^2/\text{s}$  [20] and for the dephasing time

of Cs spins the value  $T_2 \sim 10^{-3} - 10^{-4} \text{ s}$ ,<sup>6</sup> we obtain that, at  $\Theta = 0.1$  and  $\lambda = 1 \mu\text{m}$ , the quantity  $k^2 \Theta^2 D T_2$  is  $\sim 10^3$ . Thus, in our case, the noise signal associated with the tilted beam appears to be suppressed by a factor of  $\sim 3 \times 10^4$  that substantially hampers its detection. It seems that observation of this signal may appear possible for systems with weak diffusion (like quantum dots) or for semiconductor systems with shorter dephasing time  $T_2$ , when the noise signal from quasifree electrons, in the single-beam arrangement, can still be reliably detected.

As it was shown above, the observation of the noise signal associated with the auxiliary beam can be difficult, due to possible smallness of this signal. At the same time, its observation is of interest because it expands the informative potential of the noise spectroscopy technique, allowing, in principle, one to measure the total spatiotemporal correlation function of the sample's gyrotropy. It makes sense to point out here one methodological possibility that appears in the two-beam experiment, which can facilitate observation of this signal. Since the auxiliary beam does not hit the polarimetric receiver, one can observe its contribution to the noise signal (detected in the main beam channel) by means of modulation (amplitude or phase) of the auxiliary beam with subsequent lock-in amplification of the noise signal. In the simplest case, the effect of the auxiliary beam can be detected as the difference of the noise signals observed in the main beam channel with the auxiliary beam turned on and off.

## V. CONCLUSION

In this paper we presented, in the single-scattering approximation, consistent calculations of polarimetric signal detected in spin-noise spectroscopy. The expressions derived can be applied to samples with the length exceeding that of Rayleigh of the probe laser beams. The calculations are performed for model systems comprised of gyrotropic particles with allowance for their possible diffusion. Analysis of two-beam arrangement of SNS was presented that makes it possible to study not only temporal but also spatial correlations of the gyrotropy. It was shown that diffusion of gyrotropic particles may broaden the noise spectra observed in the two-beam arrangement, with this broadening substantially exceeding the time-of-flight broadening observed in the conventional single-beam arrangement. Altogether, the two-beam arrangement considered in this paper provides SNS with degrees of freedom that may, in certain cases, considerably widen the potential of this tool of research as applied to systems with randomly moving spins.

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<sup>6</sup>This value is obtained from our observations of the noise spectrum of Cs atoms.



- [1] V. S. Zapasskii, *Adv. Opt. Photon.* **5**, 131 (2013).
- [2] G. M. Müller, M. Oestreich, M. Römer, and J. Hubner, *Physics E* **43**, 569 (2010).
- [3] N. A. Sinitsyn and Y. V. Pershin, *Rep. Prog. Phys.* **79**, 106501 (2016).
- [4] S. V. Poltavtsev, I. I. Ryzhov, M. M. Glazov, G. G. Kozlov, V. S. Zapasskii, A. V. Kavokin, P. G. Lagoudakis, D. S. Smirnov, and E. L. Ivchenko, *Phys. Rev. B* **89**, 081304(R) (2014).
- [5] R. Dabhashi, J. Hübner, F. Berski, K. Pierz, and M. Oestreich, *Phys. Rev. Lett.* **112**, 156601 (2014).
- [6] I. I. Ryzhov, S. V. Poltavtsev, K. V. Kavokin, M. M. Glazov, G. G. Kozlov, M. Vladimirova, D. Scalbert, S. Cronenberger, A. V. Kavokin, A. Lemaître, J. Bloch, and V. S. Zapasskii, *Appl. Phys. Lett.* **106**, 242405 (2015).
- [7] I. I. Ryzhov, G. G. Kozlov, D. S. Smirnov, M. M. Glazov, Y. P. Efimov, S. A. Eliseev, V. A. Lovtcius, V. V. Petrov, K. V. Kavokin, A. V. Kavokin, and V. S. Zapasskii, *Sci. Rep.* **6**, 21062 (2016).
- [8] I. I. Ryzhov, S. V. Poltavtsev, G. G. Kozlov, A. V. Kavokin, P. V. Lagoudakis, and V. S. Zapasskii, *J. Appl. Phys.* **117**, 224305 (2015).
- [9] V. S. Zapasskii, A. Greilich, S. A. Crooker, Y. Li, G. G. Kozlov, D. R. Yakovlev, D. Reuter, A. D. Wieck, and M. Bayer, *Phys. Rev. Lett.* **110**, 176601 (2013).
- [10] L. Yang, P. Glasenapp, A. Greilich, D. Reuter, A. D. Wieck, D. R. Yakovlev, M. Bayer, and S. A. Crooker, *Nat. Commun.* **5**, 4949 (2014).
- [11] G. M. Müller, M. Römer, J. Hübner, and M. Oestreich, *Phys. Rev. B* **81**, 121202(R) (2010).
- [12] M. Römer, J. Hübner, and M. Oestreich, *Appl. Phys. Lett.* **94**, 112105 (2009).
- [13] T. Mitsui, *Phys. Rev. Lett.* **84**, 5292 (2000).
- [14] E. B. Aleksandrov and V. S. Zapasskii, *Sov. Phys. JETP* **54**, 64 (1981).
- [15] B. M. Gorbovitskii and V. I. Perel, *Opt. Spektrosk.* **54**, 388 (1983).
- [16] G. G. Kozlov, I. I. Ryzhov, and V. S. Zapasskii, *Phys. Rev. A* **95**, 043810 (2017).
- [17] G. G. Kozlov, [arXiv:1706.04511](https://arxiv.org/abs/1706.04511).
- [18] G. M. Müller, M. Römer, D. Schuh, W. Wegscheider, J. Hubner, and M. Oestreich, *Phys. Rev. Lett.* **101**, 206601 (2008).
- [19] V. G. Lucivero, N. D. McDonough, N. Dural, and M. V. Romalis, *Phys. Rev. A* **96**, 062702 (2017).
- [20] D. Giel, G. Hinz, D. Nettels, and A. Weis, *Opt. Express* **6**, 251 (2000).