Spin-noise spectroscopy: from proof of principle to applications

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More than 30 years ago, the feasibility of detecting magnetic resonance in the Faraday-rotation noise spectrum of transmitted light was demonstrated experimentally. However, practical applications of this experimental approach have emerged only recently thanks, in particular, to a number of crucial technical advancements. This method has now become a popular and efficient tool for studying magnetic resonance and spin dynamics in atomic and solid-state paramagnets. In this paper, we present a review of research in the field of spin-noise spectroscopy, including its physical basis, its evolution since its first experimental demonstration, and its recent experimental advances. Main attention is paid to the specific capabilities of this technique that render it unique compared to other methods of magnetic and optical spectroscopy. The paper is primarily intended for experimentalists who may wish to use this novel optical technique.

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1. Introduction

In the photonics and information science of recent years, much attention has been paid to spin-based, rather than charge-based, electronic systems. This area of research, currently referred to as spintronics, treats spin as a carrier of quantum information and is considered as a highly promising pathway to new information technologies [1,2]. This tendency has produced great interest in the dynamic properties of spin systems both in equilibrium and under different kinds of perturbations capable of controlling the spin state of the system. The new technique—spin-noise spectroscopy, developed in the last decade—refers to the experimental methods intended for studying spin systems under conditions of thermal equilibrium.

The term "spin-noise spectroscopy" (SNS) describes an experimental technique that implies spectral investigations of spontaneous fluctuations of spin-system magnetization (spin polarization). Until recently, this spectroscopy was not considered a practical instrument of experimental physics. A few experiments [3–5], in which magnetic resonance was observed experimentally in the spin noise spectrum, were primarily aimed at demonstration of the practical feasibility of such an approach. Of course, the fact that the spin system in a magnetic field should display excess noise at the frequency of magnetic resonance, as justified by the fluctuation-dissipation theorem (see, e.g., [6]), was beyond question. Still, fluctuations of this kind, fundamentally small for macroscopic systems, could be either hardly detectable or of no use in a practical sense. A certain interest in this technique was first revealed by researchers dealing with fundamental problems of quantum nondemolition measurements (see, e.g., [7,8]).

For the last several years, however, the situation has drastically changed, mainly due to remarkable advances in the digital systems of data acquisition [9–11]. At present, Faraday-rotation (FR)-based SNS, which is considered to be the most efficient approach to spin-noise detection, has found its niche among other methods of magnetic resonance spectroscopy and has gradually turned into a standard experimental technique with a wide and, in many respects, unique range of applications.

In this paper, we briefly outline the main stages in the development of this new branch of spectroscopy, starting with the first experimental observation of magnetic resonance in the Faraday-rotation noise spectrum to the most recent achievements that have revealed unique potentialities of this technique, in the spectroscopy of both the radio frequency and the optical range of paramagnets. We will consider the basic ideas underlying this experimental technique, discuss technical problems associated with its implementation, and describe specific properties of FR-based SNS that determine its rich informative potential.

We will also consider recent technical progress in this method of research that has radically changed its place in contemporary experimental physics. This paper is intended not to review the experimental measurements of SNS, but rather to review properties of noise and SNS that make these measurements possible.

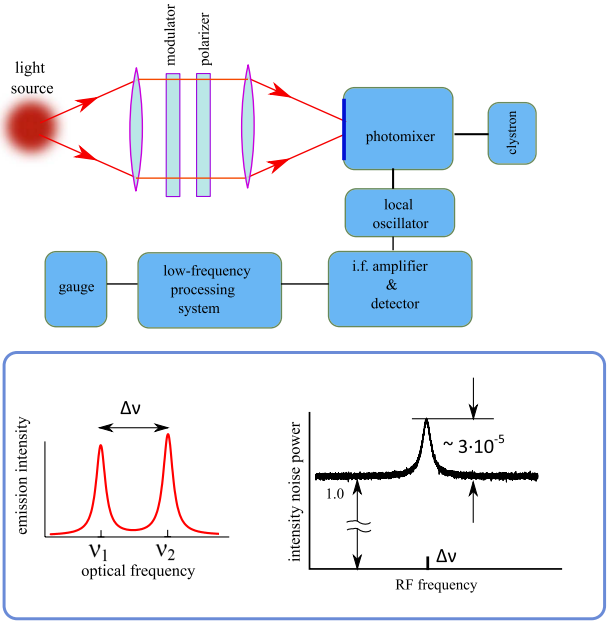
2. Historical Background

FR-based SNS combines in itself two essentially different methods of research. On the one hand, it can be considered a branch of light intensity noise (LIN) spectroscopy and, on the other, as a specific modification of the optical method of magnetization detection. To make clear the essence of FR-based SNS, it makes sense to consider these two basic methods in more detail.

2.1. Light Intensity Noise Spectroscopy

The pioneering experimental work on LIN spectroscopy that stimulated great interest was performed by Forrester *et al.* in 1955 [12] (Fig. 1). In that work, a cell with Hg vapor placed into a magnetic field was excited by an electrodeless microwave discharge, and two Zeeman components of the emission line at 546.1 nm were detected photoelectrically by means of a specially designed photodetector coupled to a microwave cavity. The detected spectrum of the LIN was found to contain a peak at the frequency of magnetic splitting of these two components. The excess noise of the light intensity related to this splitting was less than 10^{-4} of its shot-noise level. The complexity of the experiment was additionally aggravated by the high frequency of the detected signal ($\sim 10^{10}$ Hz) needed to provide a sufficiently high Q value of the noise peak. Still, the signal

Figure 1



Simplified scheme of the experiment of Forrester *et al.* [12]. Lower inset shows schematically the optical spectrum of the detected Zeeman doublet of Hg vapor (left) and the detected spectrum of the LIN.

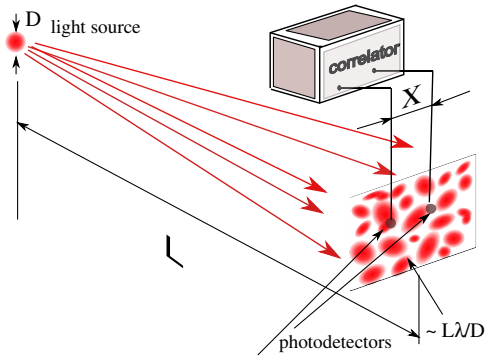
was detected, in this experiment, with a signal-to-noise ratio of around 2, for an accumulation time of 250 s.

It should be emphasized that, in spite of the fact that the frequency of the observed peak in the intensity noise spectrum exactly corresponded here to magnetic splitting of the emission line, this experiment did not have anything to do with optical detection of magnetic resonance (see [13] and references therein). Suffice it to say that the width of this peak was determined by the Doppler broadening of these Zeeman components, which should not be revealed in any way in the magnetic resonance spectrum.

The result of fundamental importance obtained in this paper was that, in the process of photoelectric conversion of the light field, the emission probability for electrons is proportional to the square of the total electric field amplitude arising due to interference between its Fourier components. In other words, this paper showed experimentally the possibility of interference between the light fields originating from different incoherent sources. This conclusion, at that time, was far from trivial, and, as the authors of [12] wrote, "many physicists found it contradictory to their ideas about the nature of interference".

The discussion raised by publication or [12] was further heated by another important experiment in the field of noise spectroscopy. The experiment in point was performed and interpreted by Hanbury-Brown and Twiss in 1956 [14]. Now, the measurements were focused on detecting spatial correlations in the intensity noise of a remote thermal source and were aimed at application of this effect to evaluating the angular dimensions of stars. The authors took advantage of the fact that emission of a remote thermal source of finite size is characterized by a certain spatial scale of light coherence controlled by the angular size of the source. Indeed, if we consider the intensity profile created by such a source with diameter D located at a distance L from the plane of observation (Fig. 2), we will see that it comprises a multitude of bright and dark spots. The brightness created at each particular point depends on whether the resultant interference of the waves from all emitters of the source is constructive or destructive. These spots are very pronounced in monochromatic laser fields and are called *speckles*. The size of a speckle is determined by the distance at which the total phase difference between the rays providing constructive or destructive interference changes by π radians. In our case, the average size of the spot is given by $L\lambda/D$, where λ is the

Figure 2



Geometry of the experiment of Hanbury-Brown and Twiss with its characteristic parameters.

light wavelength. Thus, we see that the angular size of the light source in this geometry can be directly estimated from the average size of speckles.

Hanbury-Brown and Twiss performed their experiment with a star as a light source and evidently could not examine an instantaneous profile of brightness over a large area of the illuminated surface. So, the authors made use of the intrinsic intensity noise of the light emitted by the incoherent source. These temporal fluctuations had to be correlated or not, in two spatial points of the observation plane, depending on whether these points are closer to each other than the size of the spot or not. Thus, by measuring the correlation between photocurrents of two detectors placed at a distance of X from each other, as a function of X one can estimate the average size of the speckles (of the *coherence area*) and find the angular size of the source.

The stellar interferometer created on the basis of this effect (the *intensity interferometer*) was successfully used in astrophysical measurements.

The experiments [12] and [14] were the earliest and the most famous that showed that noise in optics may be useful and informative. Note that these measurements were aimed at studying the properties of light, rather than the properties of a medium. At the same time, in the 1960s and 1970s, there were performed a considerable number of experiments in which the intensity-noise-based technique (yet without lasers) was used for studying the dynamic properties of atomic systems. Highly important results of these studies were related to the possibility of manifestation of the excited state dynamics and fine energy structure in the noise spectrum of the spontaneous emission of the system. The noise of the light transmitted by an absorbing atomic medium was employed for measuring the diffusion parameters of atoms in a gaseous medium. It should be noted that all these experiments, in the pre-laser era, were highly labor intensive. In more detail, this story is considered in the review [15].

The situation has changed dramatically, however, with the advent of lasers. Due to the extremely high luminosity of laser emission, the spectral power density of light intensity modulation in these sources at beat frequencies could exceed the background shot-noise level by many orders of magnitude, and LIN spectroscopy has gained a much wider practical sense. It became clear that LIN spectroscopy could be used not only for studying the spectral and correlation characteristics of the light source proper (like, e.g., the laser output mode structure) but also for investigating the dynamic properties of the media interacting with the light. Specifically, LIN spectroscopy, referred to as dynamic light scattering (DLS) or photon correlation spectroscopy (PCS), has found applications for studying the morphological and dynamic properties of suspensions, solutions of macromolecules and polymers, liquid crystals, biological solutions, and micro-organisms. This technique may be efficiently applied to ensembles of particles with a wide range of dimensions (from 0.001 μm to several micrometers) that is inaccessible by other methods (see, e.g., [16]).

2.2. Spectroscopy of the Light Intensity versus Spectroscopy of the Light Field

It is interesting to compare the spectroscopy of LIN with conventional optical spectroscopy (which also inevitably deals with fluctuating optical fields). Both methods are aimed at getting information about the spectral characteristics of the

optical field $E(t)$ oscillating at about 10^{15} Hz by measuring its *intensity* $|E(t)|^2$ (in reality, by measuring photocurrent or photocharge), since we cannot directly measure the field amplitude $E(t)$.

In standard optical spectroscopy, which implies measuring the spectrum of the field at optical frequencies and, therefore, may be considered spectroscopy of the optical field, the procedure of spectral decomposition of the light field precedes measurement of the light intensity. As a result, the intensity of each detected spectral component retains information about the correlation properties of the optical field that is connected with its spectrum, according to the Wiener–Khinchin theorem, through the relationship

$$I_\omega = \int \langle E(t)E^*(t+\tau)\rangle e^{i\omega\tau} \mathrm{d}\tau. \tag{1}$$

In other words, optical spectroscopy allows one, in this way, to get information about the correlation properties of the field in the range of 10^{-15} s without a photodetector with such a high temporal resolution.

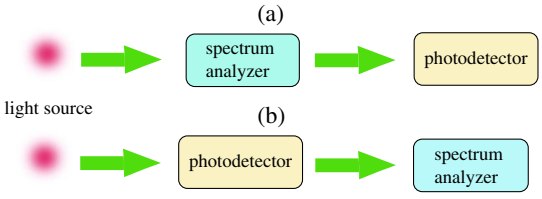
In LIN spectroscopy, the experimental arrangement is inverted: the light field is first converted into photocurrent (thus losing its carrier frequency) and then spectrally analyzed in the electronic channel of the detection system. In this case, the power spectrum of the signal under study is determined by the correlation characteristics of the *light intensity* $I(t)$ [rather than the light field $E(t)$],

$$I_\omega^2 = \int \langle I(t)I(t+\tau)\rangle e^{i\omega\tau} \mathrm{d}\tau, \tag{2}$$

and does not contain, in an explicit form, information about the correlation properties of the field at optical frequencies. In LIN spectroscopy, the scale of the frequencies of interest is limited by the bandwidth of the detection system, and the LIN spectrum, therefore, can contain information only about relatively fine features of the optical spectrum.

This fundamental distinction between the two spectroscopic techniques is schematically illustrated in Fig. 3. SNS, as a sort of LIN spectroscopy, evidently implies the second type of measurements, with photodetection preceding the spectral decomposition of the signal. At the same time, combination of the two types of spectroscopy, with preliminary spectral decomposition of the light field, as we will see below, may provide valuable additional information about the system.

Figure 3



The two experimental arrangements used in conventional optical spectroscopy (a) and in the spectroscopy of LIN (b).

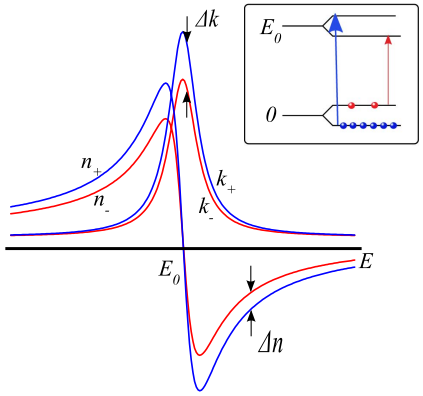
In some cases, the *light intensity spectrum* may provide information about the light field that is inaccessible in conventional optical spectroscopy. The point is that light intensity variations, as was already mentioned, are caused, in fact, by the beats between different spectral components of the optical field and, therefore, are able to reveal the phase correlation between the components not seen or not resolved in the optical spectrum. The simplest example may be given by a broadband (or "white") light with harmonically modulated intensity. This light reveals, in its intensity spectrum, a sharp peak at the modulation frequency, while its optical spectrum remains practically unperturbed by the modulation. This informative capability of light intensity spectroscopy constitutes the basis for spectroscopy of superhigh resolution. This spectroscopy, developed in the 1970s, made it possible, in particular, to realize sub-Doppler resolution in optical spectroscopy of atomic systems and to study the fundamental phenomena of interference of quantum states hidden in the inhomogeneously broadened optical transitions ([17,18]).

FR-based SNS, which we will talk about, can be regarded as a polarization version of LIN spectroscopy, with light field fluctuations provided by the spontaneous noise of spin-system magnetization. The possibility of conversion of magnetization fluctuations to those of light polarization is determined by a direct relationship between Faraday rotation and magnetization of the spin system, constituting the basis of the optical method of magnetic measurements (see, e.g., [19]).

2.3. Optical Detection of Spin-System Magnetization

Optical methods of detecting spin-system magnetization (spin polarization) rely on the fact that magnetization of a paramagnet affects its optical properties. Formation of so-called "paramagnetic" FR [20], which directly reflects magnetization of the spin system, can be schematically explained as follows (Fig. 4). The difference between populations of the ground-state magnetic sublevels of the

Figure 4



Formation of the "paramagnetic" part of the Faraday rotation in a longitudinal magnetic field for the simplest case of transition between two magnetic doublets (inset). Due to redistribution of populations over the ground-state sublevels, the absorption coefficients (k_+ and k_-) and refractive indices (n_+ and n_-) for two circular polarizations become different. These differences (Δk and Δn) give rise to magnetic circular dichroism and Faraday rotation, respectively. Magnetic splitting of the transition energies is supposed negligible compared to the linewidth.

paramagnet (of the spin-system energy levels) creates, for the light propagating along the field, a corresponding difference in the optical absorption for the two circularly polarized transitions from these unequally populated states [*magnetic circular dichroism* (MCD)]. This dichroism can be, evidently, observed only in the region of optical absorption and manifests itself in the form of ellipticity acquired by linearly polarized light transmitted through the medium. The same kind of optical anisotropy, in conformity with the Kramers–Kronig relations, is observed in the difference of the refractive indices for the two circular polarizations (*magnetic circular birefringence*) and is revealed as rotation of the polarization plane of the linearly polarized light traveling through the medium (*the Faraday effect*).

In this description, we ignored magnetic shifts of circularly polarized components of different handedness. This contribution to the magneto-optical activity of the medium (usually referred to as "diamagnetic") is not related to spin-system magnetization and is irrelevant to our consideration.

Of course, this is a highly simplified picture, and mechanisms of formation of the FR, e.g., in semiconductors cannot be exhausted by this scheme. However, it remains valid that there exists a contribution to FR (and MCD) that is directly related to spin-system magnetization. Both FR and MCD can be used to measure the magnetization. An essential difference between these two methods is that the method of dichroism implies inevitable optical excitation of the sample and, therefore, cannot be nonperturbative, whereas detection of the Faraday effect may be performed in the region of transparency without producing any real optical transitions. In addition, the MCD-based measurements of spin-system magnetization may be performed in a pure photometric way by measuring intensity of the transmitted circularly polarized light, while the Faraday rotation in the transparency region does not imply any changes in the light intensity and requires the use of a polarimetric technique.

It is also worth noting that the Faraday rotation, in the context of our treatment, is not necessarily to be observed in a longitudinal magnetic field, as implies its classical definition. It is supposed that the FR detects spin polarization along the light propagation direction regardless of the direction (and even of the presence) of the external magnetic field. In particular, the FR can be used to detect *optical orientation* of a spin system [21] or to observe oscillations of its transverse coherently precessing magnetization and, thus, to optically detect its magnetic resonance. This method was first proposed and realized in [22]. Since then, the optical technique of detecting spin-system magnetization has traversed a long path and now these methods, with their numerous and highly sophisticated modifications, are widely used not only for studying delicate properties of spin ensembles, but also for detecting and measuring external magnetic fields affecting these properties (see, e.g., [23]).

In FR-based measurements of magnetization, it is often useful to have some idea about how many spins contribute to the effect. In classical magneto-optics, the magneto-optical activity of the medium was commonly characterized by the Verdet constant corresponding to the FR angle per unit magnetic field in the sample of unit length. When measuring spin-system magnetization, we are more interested in how efficiently spin polarization is converted into FR, and the Verdet constant appears to be, for this purpose, inappropriate. A very convenient parameter, capable of characterizing this property of a paramagnet, was introduced in

[24]. It was proposed to use for this purpose the *FR cross section* σ_F numerically corresponding to the FR angle of the medium of length d with a spin density of 1 cm^{-3} and defined by the relation

$$\varphi = \sigma_F S_z d,$$

where $S_z = (n_+ - n_-)/2$ is the difference of densities of spins oriented up and down with respect to the light propagation direction. The FR cross section makes it possible to evaluate the sensitivity of the FR to magnetization changes of a particular spin system and to compare the efficiency of the FR technique with respect to different spin systems.

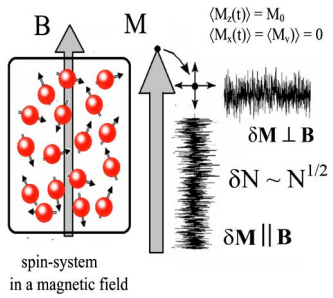
3. Basic Idea of Faraday-Rotation-Based Spin-Noise Spectroscopy

Application of magneto-optics to studying spin dynamics has been known since the 1950s. The FR and MCD effects were used, in particular, for detecting magnetic resonance in atomic systems polarized by optical pumping. In those experiments, the magnetic resonance was revealed either as a suppression of longitudinal magnetization (MCD or FR) for the light propagating along the field or as oscillations of the transverse magnetization (MCD or FR) for the light propagating across the field [25,26]. This magneto-optical technique was also used for studying the dynamic properties and energy structure of transparent paramagnets by measuring their magnetic susceptibility at subresonant frequencies [27].

It is important that, in all these experiments, the detected optical signal was a result of the *coherent response* of the spin system to a modulated (or, at least, time-dependent) perturbation (microwave pumping or external magnetic field). The idea of SNS was to detect intrinsic fluctuations of spin-system magnetization under conditions of thermal equilibrium without any regular external perturbation.

Since any real spin system consists of a finite number of spins that perform incessant random motion (maintaining thermal equilibrium of the system), its magnetization should inevitably exhibit spontaneous fluctuations due to deviations of instantaneous values of the magnetization from its mean value (Fig. 5).

Figure 5



Magnetization **M** of a spin system in a static external magnetic field **B**, due to permanent motion of individual spins, exhibits random fluctuations both in its magnitude (along the magnetization **M**) and in its direction (across the mean magnetization).

The spectrum of these random fluctuations and their correlation times should be directly connected with dynamic parameters of this motion (characteristic frequencies and correlation times). These dynamic characteristics, in principle, can be extracted from the results of noise measurements.

Of course, the notion of thermodynamic fluctuations had been known long before the late 1970s, when the experiment [3] was planned, and the problem was more of a practical, rather than a fundamental, nature: it was clear that these fluctuations in a macroscopic spin system should be extremely small, and the question was whether it was possible to detect them experimentally for a reasonable accumulation time with a sufficiently high signal-to-noise ratio and, in the final analysis, whether it had any practical purpose.

4. Experimental Geometries

Conventional magneto-optical measurements imply two main geometries of measurement, with the light beam propagating, respectively, along and across the magnetic field. These geometries are commonly referred to as the Faraday (longitudinal) and Voigt (transverse) configurations. Standard magneto-optical effects in a static magnetic field, observed in these configurations, in conformity with the symmetry of the problem, correspond to the cases of magnetic-field-induced circular and linear anisotropy. This is not the case, however, for fluctuations of the magneto-optical anisotropy, which may break the symmetry of the problem.

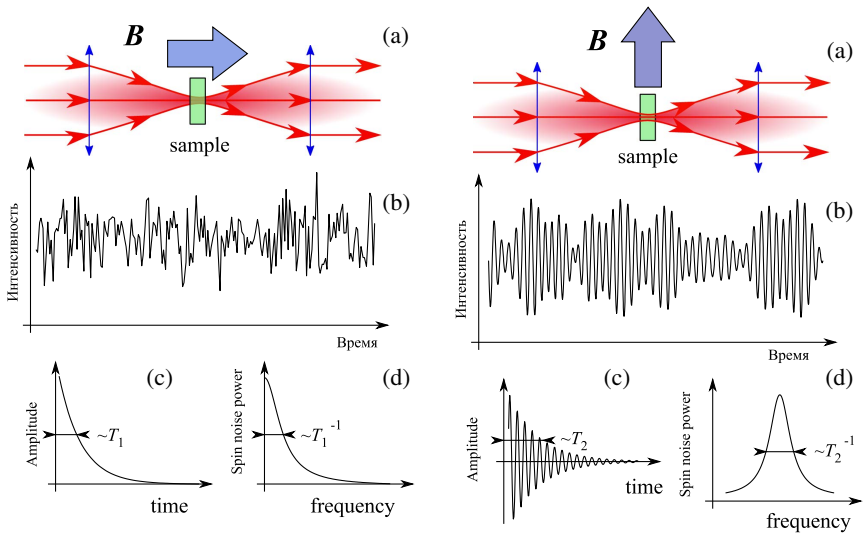
4.1. Faraday Configuration

The simplest experimental geometry for measuring FR noise, which may seem the most natural, implies detection of *longitudinal* fluctuations of the magnetization, with the probe laser beam aligned along the external magnetic field and along the equilibrium magnetization of the spin system [Faraday configuration, Fig. 6, left panel]. In this case, the detected fluctuations of the magnetization do not break axial symmetry of the system. The dynamics of these fluctuations is controlled by the only relevant characteristic time—longitudinal spin relaxation time T_1, which determines the correlation and spectral characteristics of this random process. In the time domain, the longitudinal FR noise [left panel (b) of Fig. 6] appears as a "white" noise with the removed higher frequencies (exceeding the relaxation rate T_1^{-1}). The autocorrelation function, correspondingly, acquires an exponential shape (rather than δ-wise, as for the "white" noise) with the characteristic time T_1 [left panel (c) of Fig. 6], while the power spectrum of this signal, given by Fourier transform of the autocorrelation function, is a Lorentzian centered at zero frequency with the width determined by the relaxation rate T_1^{-1} [left panel (d) of Fig. 6].

Thus, by measuring the FR noise spectrum in the Faraday geometry, we can obtain information about time T_1, and its dependence on magnetic field, temperature, and other external parameters, i.e., about the properties of the system usually obtained from the data of electron paramagnetic resonance (EPR) spectroscopy or from nonresonant magnetic measurements in oscillating magnetic fields.

It is worth noting that this experimental approach has much in common with the paramagnetic relaxation technique that was developed by the Dutch physicist

Figure 6



Measuring the spin noise (FR noise) of a spin system in an external magnetic field. The left and right panels correspond to longitudinal and transverse orientation of the magnetic field with respect to the light propagation (the Faraday and Voigt configurations, respectively). (a) Experimental arrangement, (b) a sketch of the temporal dependence of the noise signal, (c) the shape of the autocorrelation function of the noise signal, and (d) the spectrum of the signal.

C. Gorter [28] for measuring magnetic susceptibility of paramagnets in parallel fields and was later transferred to optical basis with the aid of laser polarimetry [27,29]. In Gorter's technique, the dynamics of a spin system is studied by measuring its linear response to a RF magnetic field applied along the dc field. The essential difference between these two approaches is that, in the case of noise spectroscopy, the system is not supposed to be perturbed by the external oscillating field. In terms of the fluctuation-dissipation theorem, the longitudinal magnetization noise (or the FR noise) is the counterpart of the linear magnetic (or magneto-optic) response observed in parallel fields.

In this longitudinal geometry, however, the noise is spectrally located in the vicinity of zero frequencies where measurements are often hampered by the universal $1/f$ (flicker) noise. In addition, the spectrum of longitudinal fluctuations of magnetization, bringing much data about relaxation characteristics of the system, does not contain any information about its magnetic splitting, which is highly important for characterization of the spin system.

4.2. Voigt Configuration

The above drawbacks of the Faraday configuration can be easily overcome by employing the Voigt geometry (Fig. 6, right panel). In this configuration, the light beam traveling through the paramagnet across the applied magnetic field will detect transverse magnetization of the spin system, which, by symmetry considerations, should vanish in this geometry and may arise only due to spontaneous violation of the symmetry by fluctuations.

In this configuration, any random transverse fluctuation of magnetization will precess around the magnetic field direction for approximately the transverse

relaxation time T_2 (or what is usually called T_2^*, when the spin system is subject to inhomogeneous broadening) and then will be replaced by another realization of the transverse fluctuating magnetization with another magnitude and another phase of precession. As a result, the probe light beam will exhibit randomly oscillating FR at the Larmor frequency [right panel (b) of Fig. 6]. The correlation function of this random process will now have the shape of oscillatory decay with a characteristic time $\sim T_2$ [right panel (c) of Fig. 6]. The peak of the FR noise spectrum will now be shifted away from zero frequency, and the width of the peak will be determined by the dephasing time T_2 (or T_2^*) of the spin system [right panel (d) of Fig. 6]. In other words, the FR noise spectrum will present, in this geometry, the *magnetic resonance spectrum* of the spin system.

The idea of detecting magnetic resonance in the noise of the spin-system magnetization was first mentioned by Bloch [30] in 1946. An essential contribution to the understanding of optical manifestations of spin-system magnetization, spin precession, and spin dynamics and, implicitly, to optical detection of magnetization noise, was made by Kastler in his work on optical pumping [31–33]. FR noise spectroscopy is closely related to time-resolved Faraday rotation [34] and may be considered its incoherent version.

5. Polarimetric Sensitivity

A specific feature of SNS is that it is, in fact, not a *spectroscopy of response* (to a certain extent, it may be considered a spectroscopy of response of a spin system to its stochastic perturbation by the thermal reservoir of the environment), and, therefore, the magnitude of the detected signal (*noise signal*) cannot be controlled by varying the strength of the perturbation. In addition, the magnitude of the magnetization (and FR) noise for a macroscopic sample, as was already mentioned, should be extremely small as compared with the values of "coherent" magnetization (or magnetization of saturation) induced, by the external perturbation acting in the same way upon all the spins of the ensemble. Thus, it is clear that the problem of polarimetric sensitivity may have a critical importance for SNS.

The sensitivity of polarimetric measurements in optics is known to be fundamentally limited by the so-called photon noise or shot noise of the detector photocurrent in accordance with the relationship (see, e.g., [35])

$$\Delta\varphi_{\min} \approx \sqrt{\Delta f / 2I\eta}, \tag{3}$$

where $\Delta\varphi_{\min}$ is the angle of the polarization plane rotation detected with the signal-to-noise ratio equal to unity, Δf is the bandwidth of the detection channel, I is the light intensity measured in the number of photons per second (everywhere below, when the issues of polarimetric sensitivity are discussed, the light intensity I is supposed to be given in photons per second), and η is the quantum yield of the photodetector. For instance, for light power of 20 mW, wavelength of 550 nm, and bandwidth of 1 Hz, this quantity lies in the range of 10^{-8} rad.

Such a sensitivity, as may be shown by appropriate estimates, is high enough to solve, in many cases, the problem of spin-noise detection. The most frequent reason why the shot-noise-limited sensitivity cannot be achieved with lasers (which are considered, in this context, the only suitable light sources) is related

to their excess noise, which may exceed the shot-noise level by a few orders of magnitude.

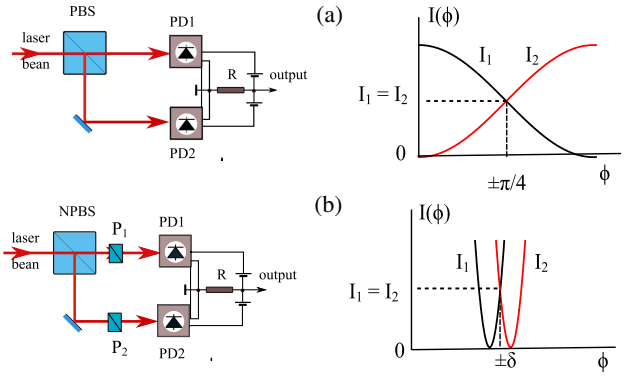
The excess noise can be suppressed using various tricks of the trade [35], the simplest and most efficient of them being *a balanced detector* (Fig. 7(a)).

The linearly polarized light beam whose polarization behavior is examined passes through a polarizing beam splitter with its two outputs coupled to photodetectors. The photodetectors are included into a differential circuit so that their photocurrents are subtracted at load resistor R. When the polarization plane of the incident light makes an angle of 45° with the polarizing directions of the beam splitter, the photocurrents of the two detectors cancel at the resistor (regardless of the light intensity and its variations). At the same time, the changes of the photocurrent related to variations of the light beam polarization plane are always anticorrelated (have opposite signs) and, as a result, add in the signal. In this way, it is possible, in practice, to suppress the excess intensity noise by approximately 3 orders of magnitude and to achieve the shot-noise limit of polarimetric sensitivity with noisy laser sources.

For the first time, as far as we know, the shot-noise-limited polarimetric sensitivity has been realized (with the use of the balanced detector), in [36,37]. Nowadays, balanced detectors are produced commercially and, in combination with polarization beam splitters, are commonly used in high-sensitivity polarimeters.

There are some other interesting methods of excess noise suppression based on the fact that shot-noise-limited polarimetric sensitivity does not substantially change when the angle between the analyzer and the polarization plane varies from $\pi/4$ to total extinction. Indeed, when moving toward total extinction (let it

Figure 7



Two schemes for measuring polarimetric signals with a balanced detector (left side) and behavior of the detector photocurrents versus azimuth of the polarization plane of the incident light φ (right side). (a) Standard 45° geometry. At $\varphi = 45°$, the photocurrents become equal, and the current flowing through the resistor R vanishes for any light intensity. (b) The scheme with variable polarization extinction. The beam is split by a nonpolarizing beam splitter (NPBS), retaining polarization of the light beam, and the level of polarization extinction is set in each arm independently (using polarizers P_1 and P_2) to equalize photocurrents. At high extinction (at small angles of detuning δ from the crossed position), the steepness of the dependence $I(\varphi)$ may become much higher. PD1 and PD2, photodetectors; P_1 and P_2, polarizers.

be $\varphi = 0$), the polarimetric response (ΔI) varies approximately linearly with φ ($\Delta I \sim I_0 \sin \varphi \cos \varphi \Delta\varphi$). At the same time, the transmitted light intensity at small φ varies as φ^2 ($I \sim I_0 \sin^2 \varphi$) making the shot noise (varying as $\sqrt{I}$) also proportional to φ. Thus, the signal-to-noise ratio remains in this range of angles φ practically the same (to within the factor $\sin \pi/4 = 1/\sqrt{2}$). In reality, however, this conclusion is violated at small φ either because of nonideality of the polarization system (the extinction ratio of the polarizers usually cannot be smaller than 10^{-5}–10^{-6}), or when the detected light intensity noise is drowned by the noise of electronics.

In the absence of excess fluctuations capable of reducing polarimetric sensitivity, the high-extinction polarization geometries can still be used for another purpose, namely, to raise the probe beam intensity (and thus to increase the shot-noise-limited polarimetric sensitivity), leaving the input light power on the detectors at a low level [38]. This is especially important for SNS, which employs broadband photodetectors with small photosensitive areas that are unable to endure high input power.

One of the methods that allows one to realize the high-extinction polarization geometry and, at the same time, to retain the opportunity to further suppress the excess intensity noise is based on the use of a polarization pile, which serves, in this case, as a dichroic medium with no birefringence. As has been shown in [39], the polarization pile makes it possible to considerably magnify the polarization plane rotation angle at the expense of reduction of beam intensity. This gadget can also be used for choosing a favorable ratio between intensity of the light passing through the sample and the input light power of the photodetector with no loss in sensitivity.

Among other high-extinction polarization geometries is a scheme with a polarization-insensitive beam splitter and independent adjustments of polarization extinction in two channels [Fig. 7(b)]. This method has all the merits of the polarization pile, but can be easier realized experimentally.

As applied to measurements of the spin-system magnetization noise, there are certain additional requirements that should be met. In particular, the wavelength of the probe laser beam should provide the most efficient conversion of magnetization noise to that of FR (this wavelength does not necessarily coincide with the region of the greatest Verdet constant or the greatest FR cross section of the system, see below). The other requirement is related to maximizing FR per a single spin, which can be achieved by reducing the cross section of the probed volume and increasing its length.

Indeed, suppose we probe a paramagnet with length l and spin density n_0 by a light beam with a cross section S (all quantum systems of the ensemble are supposed to be identical). Then, the number of spins confined in the beam will be $n = n_0 l S$. Fluctuations of the Faraday rotation at the exit of the paramagnet will be proportional to fractional fluctuations of this quantity ($n^{-1/2}$) and to the total, independent of S, Faraday rotation that goes as $\sim n_0 l$. As a result, for the FR noise, we have

$$\Delta\varphi \sim n_0 l / \sqrt{n_0 l S} = \sqrt{n_0 l} / \sqrt{S}. \tag{4}$$

So, for a given paramagnetic sample, the FR noise varies in inverse proportion with the radius of the light spot [40,41]. This is a highly important feature of

SNS, which implies that the spin noise (with all other factors being the same) *increases* with decreasing number of spins. This is why, in SNS, the probe laser beam passing through the sample should be preferably tightly focused.

Note that, using the light beam for detecting spin fluctuations, we probe a fluctuating medium (spin system) by a fluctuating agent (photon flux) and measure intensity fluctuations of the light on the background of its own shot noise [8]. It may seem, at first sight, that the spin noise can be detected only when the noise introduced by the spin system exceeds (or is comparable with) the shot noise of the probe beam. In reality, however, this is not the case. As can be shown, the mean-square level of Poissonian noise transmitted through a Lorentzian filter with bandwidth γ for the accumulation time τ can be measured with a relative accuracy of $[(1+\gamma e/i_{\mathrm{ph}})/\gamma\tau]^{1/2}$ (i_{ph} is the photocurrent and e is the electron charge), which turns into the known factor $(\gamma\tau)^{-1/2}$ at sufficiently large photocurrents ($I_{\mathrm{ph}} \gg \gamma e$) [42]. So one can easily estimate that, under real experimental conditions and for sufficiently large accumulation times, the factor $(\gamma\tau)^{1/2}$ may reach many orders of magnitude. As a result, the shot-noise power appears to be defined with fairly high accuracy and, in the form of a stable spectral background, does not preclude measuring intensity fluctuations whose power lies essentially below the shot-noise level.

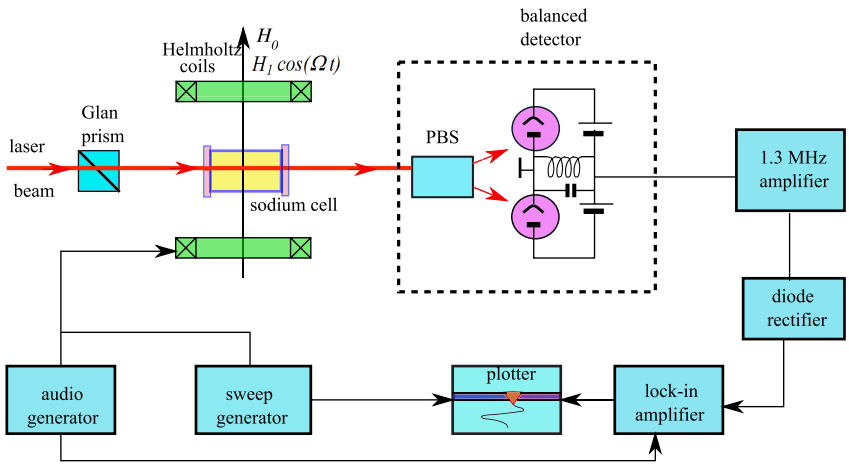
6. First Experiment

6.1. Magnetic Resonance in the FR Noise Spectrum

The first experiment on detection of magnetic resonance in the FR noise spectrum [3] was performed on sodium atoms in an atmosphere of buffer gas (neon, ~10 Torr). A schematic of the experimental setup is shown in Fig. 8.

As a light source, we used a cw dye laser close to the D1 or D2 absorption line of Na. The cell with sodium vapor was placed into a transverse magnetic field created by a pair of Helmholtz coils. The field was slightly modulated at a frequency of $\sim 10^2$ Hz. The differential signal of the balanced photodetector was first filtered by a resonant circuit at a frequency of 1.3 MHz, then selectively

Figure 8



Simplified schematic of the experimental setup used for detecting magnetic resonance in the Faraday rotation noise spectrum of sodium atoms [3].

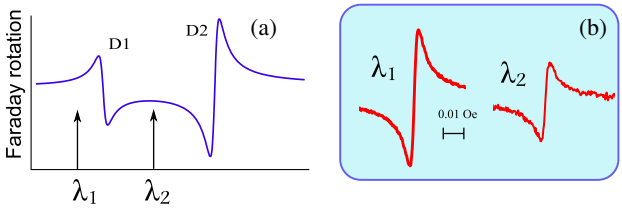
amplified, quadratically detected (rectified), lock-in amplified at the field modulation frequency, and recorded as a function of the applied magnetic field. In other words, the magnetic resonance frequency was swept with respect to the resonance frequency of the photodetection system (much like the resonance frequency is swept with respect to the frequency of the RF or microwave oscillator in conventional EPR spectroscopy).

Figure 9 shows two typical plots of the detected spin-noise resonance. The probe beam was tuned to the wing of the D1 line or to the midpoint between the two lines. The polarimetric signal of FR noise, in this experiment, substantially exceeded the shot-noise level, and the signal-to-noise ratio (or, better to say, the ratio of useful noise to harmful noise) for the accumulation time of 2 s, was close to 100:1. As far as we know, this was the first experimental demonstration of the feasibility of SNS.

6.2. Nuclear Spin Noise

In 1985, an experiment [4] on nuclear spin noise was performed that did not have anything to do with the FR technique, but ideologically was identical to the experiment in [3]. As a spin system in that experiment, an ensemble of nuclei ^{35}Cl in a $NaClO_3$ crystal was used. The experiment was performed at 4 K, and the magnetization noise was detected using a SQUID sensor. This work considerably contributed to SNS as the first observation of the resonant spin noise in a nuclear spin system predicted by Bloch [30]. Here, the number of spins contributing to the signal was much greater than in the experiment with electron spins [3], and the noise signal was expected to be extremely small. Indeed, the peak of spin noise at the frequency of nuclear quadrupole resonance of ^{35}Cl was detected with a signal-to-noise ratio of a few units for an accumulation time of 7 h. Still, further technical advances in this area made it possible to observe nuclear spin noise at room temperature [43,44] and even to develop, on its basis, an efficient method of nuclear spin-noise imaging [45]. This technique, though less sensitive than the conventional method of nuclear magnetic resonance imaging, may be indispensable, in certain cases, as an entirely noninvasive tomography that does not use any external RF or x-ray field. Spin noise acquires a particular importance for highly sensitive sensors capable of detecting just a few nuclear spins. This is exactly what provides the new, highly promising technique, based on

Figure 9



A sketch of the Faraday rotation spectrum of sodium atoms (a) and experimental plots of the EPR signal in the Faraday rotation noise (b) for two wavelengths of the probe beam (indicated by arrows). Due to modulation of the applied magnetic field, the signal, in this experiment, was proportional to the derivative of the FR noise power with respect to the magnetic field.

nitrogen-vacancy (NV)-diamond sensors, which combines high sensitivity with great spatial resolution [46].

7. Evolution of Spin-Noise Spectroscopy

The effect of magnetic resonance in the FR noise spectrum remained practically unnoticed as a possible experimental tool of magnetic resonance spectroscopy, until the end of the last century, when, with some modifications, it was reproduced on other alkaline atoms and then applied to semiconductor systems. Below, we briefly outline the main steps in the development of experimental SNS.

7.1. Application to Atomic Systems

In 2000, an interesting experiment was performed by Mitsui [5], who independently observed optically detected spontaneous spin noise of rubidium atoms at the Larmor frequency. The idea of the experiment was the same, but realized differently: the magnetization noise was detected in optical absorption, rather than in the Faraday rotation, of the atomic vapor.

A circularly polarized beam of a diode laser, tuned in resonance with the D1 absorption line of ^{85}Rb ($5\,^2S_{1/2} - {}^2P_{1/2}$) was transmitted through the cell with atomic vapor at 80°C placed into a transverse magnetic field. To simplify qualitative analysis of the results, the transition was strongly broadened (up to 6 GHz) by a buffer gas (nitrogen) at 200 Torr. Fluctuations of the spin-system magnetization in this experiment were observed in the intensity (rather than FR) noise spectrum of the transmitted light. A specific feature of this approach was that the excess intensity noise of the light source, in this case, should have been eliminated before the sample, rather than suppressed after it, because, otherwise, it could induce nonlinear effects undesirable in this experiment (they were studied independently). The laser source used for these measurements had extremely low excess intensity fluctuations, and, therefore, there was no need to employ a balanced detector for their suppression.

In that experiment, fluctuations of the spin-system magnetization were, in fact, detected by the noise of the MCD (rather than FR) of the paramagnet. From the viewpoint of linear optics, these two quantities (MCD and FR), being connected through the Kramers–Kronig relations, are equivalent and should give the same results unless the optical excitation affects the spin dynamics of the system. At the same time, in the MCD-based measurements, each act of absorption interrupts spin precession of the atom, and it may seem that the later absorption events cannot be correlated, in any way, with the previous. The author has shown, however, that this is not the case for a thermodynamically fluctuating spin system, and the MCD-based method of spin-noise detection, which looks more perturbative than the one based on the FR, is also applicable to macroscopic systems.

It is worth noting here that the question about the possibility of detecting superposition states of a quantum system (as, e.g., a precessing spin) in the noise spectrum of an electromagnetic field is not trivial. In particular, it has been shown in [47,48] that the spontaneous emission noise spectrum of an atomic medium excited randomly, in a Poissonian way, does not reveal any features related to its emission decay kinetics. This means that the intensity fluctuations of spontaneous emission excited stationary in this way cannot be used for

detecting spin noise in the excited state of the atoms. A rigorous quantum-mechanical description of the intensity-fluctuation spectrum of spontaneous emission is given in [49].

In 2004, Crooker *et al.* [40] examined in more detail the spin-noise spectra of Rb and ^{41}K atoms using the FR-based method and provided deeper insight into the capabilities of this technique. It was shown, in particular, that magnetic resonance in the FR noise spectrum can be also observed in the Faraday configuration (in the longitudinal magnetic field). This is possible, in conformity with the fluctuation-dissipation theorem, for the ESR transitions allowed in the ac magnetic field oscillating along the external dc field. An illustration of such a situation was presented in [40,50], where the resonances of this type originated from transitions with $\Delta m_F = 0$ (m_F is the projection of the total angular momentum of the state) in ^{41}K atoms.

7.2. Perturbative or Not?

One of the questions that arose after successful detection of spin noise in atomic gases, with strongly off-resonant probing of the system, was whether this technique could be considered nonperturbative or not. At first glance, it looked fairly passive or nonperturbative because it utilized Faraday rotation in the transparency region and the probe beam did not induce any real electronic transitions and, therefore, did not disturb the spin system under study. On the other hand, it could not be nonperturbative for evident reasons: the initially monochromatic probe light, after passing through the medium, became modulated and thus acquired sidebands with shifted frequencies. This means that some photons of the beam experienced inelastic interaction with the medium inevitably accompanied by some energy exchange between the light and the spin system. It is also known that the effect of a transparent medium on polarization of the light beam is accompanied by the back action of the light upon the medium even in a static magnetic field [51].

This contradiction was resolved by Gorbovitskii and Perel [52], who showed that the noise of FR at the frequency of Larmor precession can be considered a result of *coherent forward Raman scattering* of the probe laser light by the ensemble of spins randomly precessing in the transverse magnetic field. The oscillating polarization in the probe beam arises due to interference of the forward-scattered light with the light directly transmitted by the medium. This effect, according to [52], is analogous to the effect of optical mixing used in [53] for studying Brillouin scattering.

So, this process can be regarded as nonperturbative because the probe light interacts with macroscopic fluctuations of the system, and does not select any particular spin to flip. Such a conclusion is supported by the possibility of observing magnetic resonance in the MCD noise, when each absorption event explicitly destroys coherent superposition of the ground-state sublevels. The technique evidently becomes more and more perturbative as we pass from macroscopic ensembles to microscopic ones.

Note that in semiconductors, which are more complicated than pure atoms and cannot be treated in the framework of such simple models, small residual absorption may dramatically affect properties of the spin system under study even in the regions of nominal optical transparency [54].

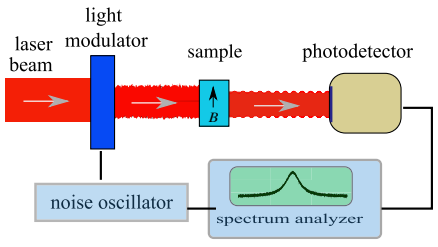
Atomic systems, mainly alkali atoms, as the most convenient model objects of SNS, have been widely used in fundamental research related to nondemolition measurements and interaction between squeezed states of light and matter. It was shown, in particular, that the spin-noise measurements performed in the region of optical transparency made it possible to surpass the standard quantum limit of phase measurements and, thus, to produce squeezed atomic spin states (see, e.g., [55,56]). This interesting topic and associated experimental results, however, lie outside the scope of this review.

7.3. "Active" Spin-Noise Spectroscopy

It is worth mentioning here another experimental approach to the optical detection of incoherent spin precession, usually also regarded as a sort of noise spectroscopy [5,57–61]. This approach is based on transformation of the modulation spectrum of the light transmitted through a paramagnet in the Voigt geometry. In this case, the light beam serves simultaneously as a probe and as a pump, and its modulation (either in intensity or in polarization) is supposed to be equivalent to modulation of the effective magnetic field applied along the light propagation. This can be achieved either by intensity modulation of a circularly polarized beam or by modulating the degree of its circular polarization. An experimental setup for these measurements may look as shown in Fig. 10.

It is well known that the coherent Larmor precession of a spin system can be induced optically by resonant pump light modulated at the appropriate frequency. The first experimental observations of optically driven spin precession were announced in [62]. The effect was detected in the vapors of alkali metals (Cs and Rb) and in metastable helium. The cell was resonantly pumped by circularly polarized light propagating across the applied magnetic field and modulated in intensity at the frequency close to that of Larmor precession of the spin system. The resonance was observed as a peak in the transmitted light intensity at the point where the modulation frequency coincided with that of the ground-state Zeeman splitting. This effect of optically driven spin precession, often referred to as "optical orientation in the rotating coordinate frame" or as a "resonance of coherence," is very close, in its physical content, to the effects of coherent population trapping [63] and electromagnetically induced transparency [64]. All these effects imply excitation of coherent superposition of two states (in our case, of two low-lying states) by superposition of two optical fields (or by a single modulated field, which is practically the same) [65].

Figure 10



Schematic of the experimental setup for active SNS. In contrast to conventional SNS, the transmitted light is not subject to polarization analysis, the beam is not necessarily focused on the sample, and no intensity noise suppression is used.

Active SNS exploits the same idea, but uses, for this purpose, a pump modulated by more or less "white" noise (rather than harmonically). Under these conditions, the system is offered, so to say, to choose on its own the frequency component capable of inducing its Larmor precession. This approach utilizes the effects of nonlinear optics and is evidently essentially perturbative.

Regularities of active SNS strongly differ from those of FR-based spectroscopy of spontaneous spin noise. At the same time, in certain respects, it can be more convenient in practice and may provide additional information about the system not related directly to its magnetic properties.

From the viewpoint of the experimentalist, this type of noise spectroscopy is, in many respects, the exact opposite of FR-based SNS: it does not imply high polarimetric sensitivity and the laser intensity noise should be well pronounced or even purposely increased, rather than suppressed. The laser beam is not supposed to be tightly focused on the sample to reduce the number of spins participating in formation of the signal. At the same time, the light power density on the sample, in these measurements, should be high enough to provide the required optical nonlinearity. As has been shown in [66,67], under sufficiently high power densities, when the Rabi frequency becomes comparable with the relaxation rates of the excited state, the intensity noise spectrum of the transmitted light becomes much more complicated, with its peaks and singularities not connected in a straightforward way with the Zeeman and Rabi frequencies of the system.

This technique of noise spectroscopy, as far as we know, has been applied so far only to atomic systems. In those experiments, diode lasers with frequency-modulated output emission were used as the noisy light sources. This spectral modulation was converted into intensity-modulated light in the process of its interaction with the narrow spectral features of the sample under study. Meanwhile, active noise spectroscopy, in our opinion, is a promising method of research that may find application in experimental studies of solid-state (including semiconductor) systems, which is highly important for up-to-date photonics and information science. The experimental setups, with the laser sources intentionally modulated in intensity or in polarization in a broad frequency range, may be useful in cases where the nonperturbative nature of SNS is not of primary importance. It should be noted that this method of nonlinear optics may provide information inaccessible to conventional SNS and lying far beyond the bounds of the field of magnetic resonance and spin dynamics.

7.4. Starting with Semiconductors

Atomic gases seemed to be highly favorable objects of SNS because their intense and narrow optical resonances provide strong peaks of FR in the vicinity of the lines (the so-called Macaluso–Corbino effect). Semiconductors and other solid-state systems, with their broader optical spectra, did not look so promising. Still, in 2005, Oestreich *et al.* [68] managed to successfully apply this technique to a solid semiconductor system.

The measurements were performed on a thick *n*-doped GaAs wafer at 10 K with the wavelength of the probe laser ~10 nm nm below the GaAs bandgap. The level of Si doping ($\sim 1.8 \times 10^{16}$ cm^{-3}) was chosen to correspond to the longest spin dephasing time measured using the femtosecond time-resolved FR technique [69]. The FR noise of the sample placed into a transverse magnetic field was detected with a broadband balanced photoreceiver and conventional

sweeping spectrum analyzer in the frequency range of 200–400 MHz. Behavior of the discovered peaks well correlated with the known magnetic resonance and relaxation properties of the donor-bound electrons in *n*-GaAs.

This work was, at that moment, more of fundamental than of practical significance since the accumulation time needed to reliably detect the noise signal lay in the range of several hours. Still, in these experiments, the authors successfully used, for the first time, the SNS technique for measuring the electron Lande *g*-factor and electron spin relaxation time in a semiconductor (*n*-GaAs) and demonstrated the applicability of SNS to semiconductor systems, thus laying the foundation for the semiconductor SNS (see the review [41]).

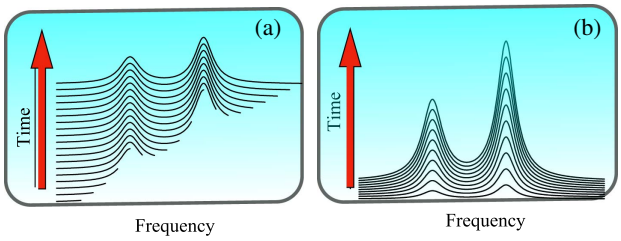
8. Technical Advances in Spin-Noise Spectroscopy

The "academic" period of SNS was completed when it became possible to perform these measurements in much shorter times in a wider frequency range on a system of greater practical importance.

8.1. Advent of the Real-Time Fast Fourier Transform Spectrum Analyzer

A real breakthrough in this field of research was made when the sweeping spectrum analyzer, in the system of data acquisition, was replaced by one with a real-time fast Fourier transform (FFT) processing system with a wide frequency range [9,11]. The conventional sweeping spectrum analyzer measures the signal only in a narrow frequency interval at a time and, therefore, constantly disregards most information contained in a time-dependent signal. The FFT spectrum analyzer, on the other hand, is capable of using the whole bulk of incoming information. It digitizes the signal in the whole bandwidth of the system (with a sampling rate of around 10^9 s^{-1}), performs FFT of the signal *in real time*, and accumulates the spectrum thus obtained. In other words, it accumulates signals in all frequency channels simultaneously, rather than only in a single channel. As a result, the accumulation time needed to achieve the same signal-to-noise ratio has decreased by a few orders of magnitude. The process of accumulation of the signal is schematically illustrated by Fig. 11.

Figure 11



The figures show schematically how the spectrum of a broadband signal is accumulated in the sweeping (a) and real-time FFT spectral analyzer (b). In the first case, the detection system successively passes through each frequency channel, ignoring, at that moment, all other channels. In the second case, the signals of all frequency channels are accumulated simultaneously. The curves are shifted for clarity.

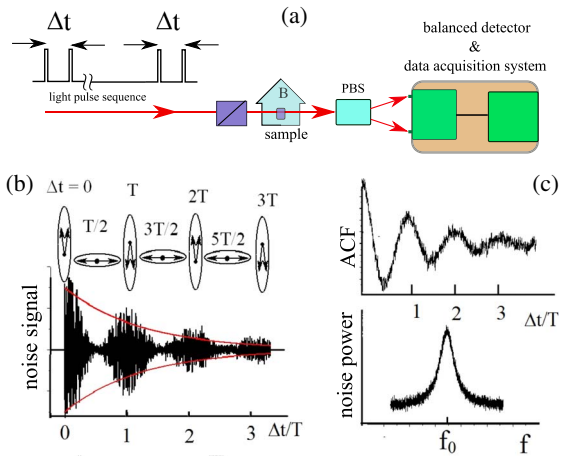
This technical advancement has turned SNS into a practical tool of magnetic spectroscopy and made it suitable, in particular, for studying the spin dynamics of low-dimensional semiconductor systems (quantum wells, quantum wires, quantum dots), which is highly important for present-day applications in photonics and optoelectronics.

8.2. Expanding the Detection Bandwidth

Another interesting idea was proposed by Müller *et al.* [10] to overcome the bandwidth limitations of optical detectors. For that purpose, they proposed using a pulsed laser with a high repetition rate (e.g., a mode-locked laser) instead of a cw laser, as a source of the probe light. In this case, the intensity spectrum of the light comprises a comb of discrete lines spaced by the pulse repetition rate f_0, and it becomes possible to observe, in the spectrum of the detected FR noise signal, not only the peak at the frequency of magnetic resonance f_R, but also the peaks at the frequencies shifted from f_R toward lower frequency by multiples of the repetition rate f_0 (provided that $f_R > f_0$). As a result, by mixing the resonance signal with the nearest peak of the probe beam intensity spectrum, the frequency of the spin-noise resonance can be transferred to the frequency range $[0, f_0/2]$. This experimental approach allowed the authors to detect spin noise at Larmor frequencies up to 16 GHz. Under these conditions, the total bandwidth of the detection system (including the spectrum analyzer) may not exceed the repetition rate f_0. At the same time, the measured spin dephasing rates are limited, in this technique, by approximately half the repetition rate. As noted in [41], the sensitivity of this technique is not reduced as compared to conventional SNS.

One more remarkable approach to the problem of bandwidth of FR-based SNS was proposed in [70]. The idea of the approach was to take advantage of an extremely broad spectrum of the femtosecond (or picosecond) light pulses emitted by a mode-locked Ti:sapphire laser and to measure the correlation characteristics of FR noise with a very high temporal resolution that is inaccessible to conventional photodetectors. For that purpose, it was suggested that the sample under study be probed by a train of pairs of ultrashort pulses with the time interval between them variable within a range comparable with the period of Larmor precession of the system [Fig. 12(a)]. In spite of the fact that the two closely spaced pulses are not resolved by the detection system, the detected FR noise will depend on the time interval between them [provided that there is some distinguished oscillation frequency, and, therefore, a distinguished characteristic time, in the FR noise, as shown in Fig. 12(b)]. In particular, when the time interval between the pulses is equal to the integer number of the oscillation periods, then contributions of this oscillating process to both pulses will have the same sign, and the total FR of this pair of pulses will fluctuate. If, however, the time interval between the pulses equals an odd number of half-periods of the oscillation, then contributions of these oscillations to the two pulses will compensate for each other, so that the total contribution of this process to FR of the pair of pulses will vanish. Thus, by scanning the time delay between the two pulses of the probe beam and detecting the FR noise power in the transmitted light, we will observe an oscillatory transient of the process directly corresponding to its correlation function, whose Fourier transform provides the corresponding spin-noise spectrum [Fig. 12(c)]. It is important that the two pulses are not supposed to be resolved by the detection system.

Figure 12



Ultrafast SNS. (a) Schematic of the experimental setup. The sample is probed by a train of pairs of ultrashort pulses with variable delay Δt. (b) Dependence of the noise signal on the time delay. Vectorial diagrams show summation of the contributions of the two pulses to the FR signal for different ratios of the time delay Δt and the Larmor precession period T. (c) Resulting dependence of the noise signal on the time delay Δt, corresponding to resonant transient of the system or to its autocorrelation function (ACF), and the spin resonance spectrum obtained by Fourier transform of the autocorrelation function.

The efficiency of this ultrafast SNS has recently been successfully demonstrated experimentally on a heavily *n*-doped bulk GaAs [71,72]. The train of picosecond pulses was produced, in that experiment, by two synchronized lasers with a repetition rate of 80 MHz.

This technique, which implies measuring the autocorrelation function instead of its Fourier image, should allow one to directly detect decay of the transverse magnetization in the time domain and to extend the accessible frequency range up to several terahertz.

The above proposals demonstrating the possibility of considerable extension of the SNS bandwidth give promise that this technique will find application in the EPR spectroscopy of standard microwave ranges for nonperturbative investigations of transparent paramagnets.

8.3. Cavity-Enhanced Spin-Noise Spectroscopy

In spite of remarkable advances in the data acquisition technique, the problem of polarimetric sensitivity retains its significance in FR-based SNS, and all suggestions that can help improve this characteristic remain of great interest.

It is well known that FR can be strongly enhanced with the aid of a Fabry–Perot cavity [73–79], due to multiple passes of the light through the magneto-optical medium. Such an approach is especially popular now for studying the dynamics of spin states of low-dimensional semiconductor structures in high-finesse microcavities, where the observed FR can be increased by a few orders of magnitude. In these studies, it is usually tacitly assumed that the measured times are much longer than the intrinsic times of the cavity (the cavity photon lifetime

and intermode beat period). This is really the case for microcavities, when the photon round-trip time over the cavity is equal or just a few times longer than the oscillation period of the light wave. When, however, this is not the case, and the FR oscillation frequency becomes comparable with or higher than that of the intermode beats, the response of the cavity becomes more complicated.

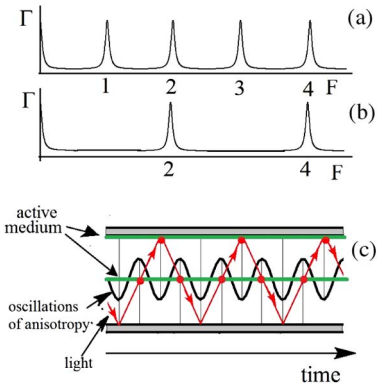
In [79], it was shown that polarization of a monochromatic light resonant to a longitudinal mode of a Fabry–Perot cavity may be highly sensitive to modulation of the intracavity anisotropy at frequencies multiple of the spacing between its longitudinal modes. This effect can be qualitatively understood in terms of the light traveling over the cavity back and forth and has much in common with the known effect of mode locking in lasers [80].

Indeed, intuitively it seems evident that enhancement of the cavity's response to weak oscillating optical anisotropy of the intracavity element will be observed when the light moving back and forth inside the cavity finds this element with the same phase of oscillation, so that the new contribution to the light polarization state is added to those already accumulated. This reasoning, though formally incorrect (being inapplicable to a monochromatic wave), proves to be useful for qualitative understanding of the resonant FR enhancement effect.

This effect can be also interpreted in terms of spectral transformation of the light passing through a polarization modulator: the resonant enhancement of FR occurs when the modulation frequency coincides with the intermodal spacing of the Fabry–Perot cavity, and the sidebands of the modulated laser light hit adjacent modes of the resonator.

Figure 13 shows the frequency dependence of the polarization modulation gain factor (Γ) for two different positions of the anisotropic element inside the cavity (for more detail, see [79]). For the sample placed in the middle of the cavity (a), the light hits the sample twice per round trip, and, correspondingly, the effect of

Figure 13



Frequency dependence of the polarization signal gain factor (Γ) in a Fabry–Perot cavity for a sample placed at the edge of the cavity (a) and in the middle of it (b). The frequency F is given in the units of intermodal spacing $f_0 = c/2L$. Panel (c) illustrates the synchronism of the light traveling over the cavity with oscillations of anisotropy of the intracavity sample (for $F = 1$). The layers in the center of the cavity and near its edge (green) depict the medium with oscillating anisotropy, the black sinusoid is the time variation of the anisotropy, and the red lines show propagation of the light beam.

enhancement can be observed at frequencies that are by a factor of 2 lower than those for the sample placed at the edge of the cavity (b).

Amplification, in this scheme, is realized in the optical (rather than electronic) channel, and the corresponding gain factor proves to be approximately equal to the cavity Q-factor ($\sim 10^2$–10^3). In other words, the Fabry–Perot cavity can be used as a *selective optical amplifier* of a polarization signal produced by the intracavity element at the frequency of intermodal spacing ($f_0 = c/2L$, L is the cavity length and c is the speed of light) or its multiples ($f = nf_0$, n is the integer). This effect can evidently be helpful in SNS for amplification of the polarization modulation arising in the probe light beam due to the random spin precession of the spin system.

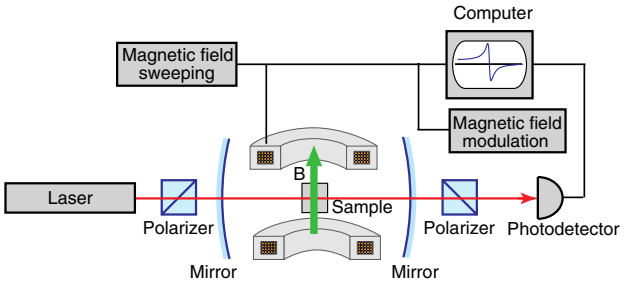
It is noteworthy that the increase in the FR signal in this method is achieved, exactly as in the case of high-extinction polarimetric measurements, at the expense of strongly increasing power density on the sample, which should be taken into account in the experiments.

One more curious possibility of application of this effect is related to prospects of creating an all-optical spin-noise spectrometer [79]. The idea of this proposal combines the effect of amplification of the polarization signal with its optical quadratic detection. Let the studied transparent paramagnetic sample be placed inside an optical cavity (Fig. 14). A monochromatic linearly polarized laser beam incident upon the cavity coincides in frequency with one of its longitudinal modes and thus passes through the cavity with no loss. At the exit of the cavity, we place a linear polarizer in a crossed position, so that no light passes through the system. Now, we start sweeping the transverse magnetic field applied to the sample. At the moment when the Larmor precession frequency of the spins becomes equal to (or a multiple of) the double intermodal frequency of the cavity ($2f_0 = c/L$), the oscillations of FR appear to be strongly enhanced by the cavity, and the light intensity at the exit (after the output polarizer) becomes nonzero.

A unique property of this system is that it allows one to detect magnetic resonance in the FR noise spectrum at any frequency without any broadband electronics and broadband photodetectors—all the needed information is contained in the dc optical signal. What is observed here is just a result of resonant coupling of the spin system with the Fabry–Perot cavity.

It is clear that such a design of an all-optical spin spectrometer imposes heavy demands both on the Q-value of the cavity and on the extinction ratio of the

Figure 14



Schematic of an all-optical spin-noise spectrometer.

polarization system. Still, it seems feasible and may be useful, in particular, for applied purposes, as a basic system for magnetometers of a new generation.

9. Optical Spectroscopy of Spin Noise

Considerable interest is currently attracted to the particularities of SNS under conditions of resonant or near-resonant optical probing of the spin system [11,81,82]. It is clear that, generally speaking, the SNS in this case loses its important property of being perturbation free when the probe laser beam induces real optical transitions. At the same time, the degree of perturbation of the spin system by the probe beam depends on how large is the optical excitation rate compared with the dephasing rate of the spin system, and, in most cases, the light power density can be decreased to the level at which optical perturbation of the system may be neglected (see, e.g., [83]). Under these conditions, SNS acquires additional remarkable properties [84].

Interesting possibilities of SNS stem from the question about mutual correlations of the FR noise at different wavelengths of the probe beam. In [84], it was pointed out that these correlations depend on whether the appropriate fluctuations are contributed by the same spin ensemble (ensemble of identical quantum systems) or not. It is important that this fact can be revealed not only in a straightforward way in the cross-correlation spectral characteristics of the FR noise (which may be thought of) but also, much easier, in the optical spectra of the FR noise power, which, generally, appear to be related to the conventional FR spectra in a nontrivial way.

The use of the probe beam wavelength as a tunable parameter of standard SNS allows one to look at this technique as a sort of *optical spectroscopy*. Such an approach makes it possible, in certain cases, to detect the structure of optical transitions hidden in the linear optical or magneto-optical spectra and, thus, opens new possibilities for the SNS technique.

Let us consider dependence of the *integrated FR-noise* (spin noise) power on the probe beam frequency. As was already mentioned, the FR-based SNS exploits the paramagnetic part of the FR to monitor magnetization of the spin system. It seems evident that, to make conversion of spin-system magnetization to FR most efficient, one has to select the wavelength of the probe light in the region of the greatest paramagnetic FR (greatest FR cross section [79]), and, vice versa, the FR noise cannot be detected at wavelengths where the FR proper turns into zero. It was shown, however, that this is not the case.

Magnetization of a macroscopic spin system is created by an ensemble of individual spins whose optical spectra may be either identical or different. As a result, the magnetization noise of the spin system may be transformed into that of the FR in different ways.

If the optical spectrum of a paramagnet comprises several spectral features, then the FR angle φ at each frequency ω will be given by the sum of partial contributions of all these features:

$$\varphi(\omega) = \sum \varphi_i(\omega). \tag{5}$$

When all the spins of the system are identical, and the optical spectrum of the whole ensemble just reproduces the one corresponding to an individual spin

system, then fluctuations of the FR will be evidently *coherent* or *correlated* over the whole spectrum, and spectral dependence of the mean-square fluctuation of the FR $\langle \delta\varphi^2(\omega)\rangle$ will be proportional to the square of the total FR (square of sum of partial contributions):

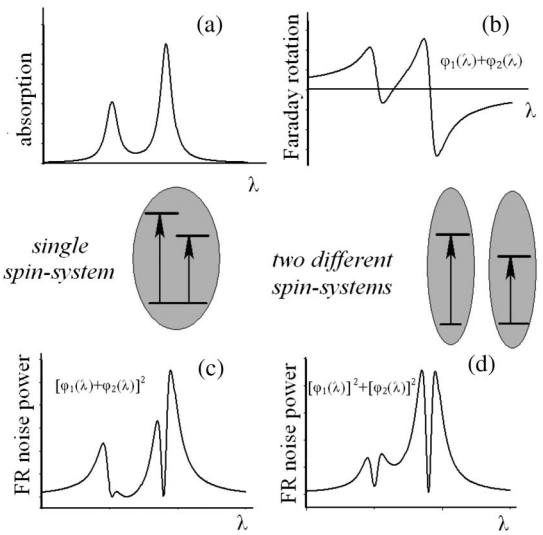
$$\langle \delta\varphi^2(\omega)\rangle \sim \left\langle \left(\sum \varphi_i(\omega)\right)^2 \right\rangle. \tag{6}$$

When, however, the paramagnet under study contains several spin subsystems with different optical spectra, the contributions of these subsystems to the FR noise will fluctuate *independently*, in an uncorrelated way, and the optical spectrum of the FR noise will be described by the *sum of squares* of the individual contributions rather than by their sum squared:

$$\langle \delta\varphi^2(\omega)\rangle \sim \sum \langle \varphi_i(\omega)^2\rangle. \tag{7}$$

As a result, the optical spectra of spin noise in these two cases, due to different interference of partial contributions in the region of their overlap, may be essentially different. Figure 15 illustrates this difference for two closely spaced absorption lines associated either with the same spin system or with different (independent) ones characterized by different optical spectra. The spectral dependence of the FR is taken here in the form of a dispersion-like curve characteristic of the paramagnetic part of the FR for the band with a width that substantially exceeds magnetic splitting of the optical transition. One can see that the distinction between these two spectra [Figs. 15(c) and 15(d)] is most pronounced in the region between the lines, where contributions of two optical transitions either compensate for each other [for a single spin system; Fig. 15(c)] or sum up [in statistical sense, for two different spin systems; Fig. 15(d)].

Figure 15



Optical spectra of absorption (a), FR (b), and FR noise power (c) and (d) of a hypothetical paramagnet with two closely spaced optical transitions. Spectra (c) and (d) correspond to the cases when optical transitions are associated with the same spin system or with two different spin systems, respectively.

One more manifestation of the correlation characteristics of spin noise in the optical spectrum, as noted in [84], is that the optical spectroscopy of spin noise may show, in some cases, spectral resolution higher than conventional optical or magneto-optical spectroscopy and, thus, may be helpful in resolving the hidden structures of optical spectra.

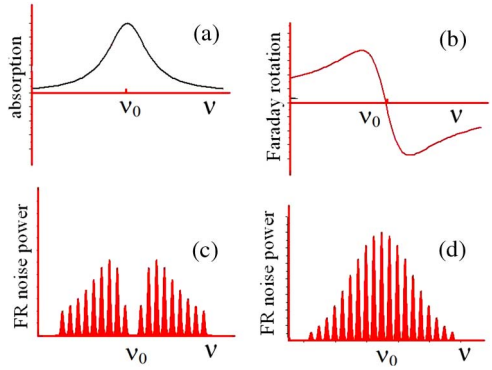
This effect is revealed in a highly spectacular form in optical spectra with inhomogeneous broadening [84]. The absorption and FR spectra in the vicinity of an isolated absorption band [Figs. 16(a) and 16(b)] are known to be the same regardless of whether the band is broadened homogeneously or inhomogeneously. Linear magneto-optics (as well as linear optics in general) cannot distinguish between these two cases. The results of calculations presented in [84] show that the optical spectra of spin noise in these two cases are drastically different [Figs. 16(c) and 16(d)].

For the homogeneously broadened band, the FR noise, which is proportional to the Verdet constant (or, better to say, to the FR cross section) squared, vanishes at the center of the band (where the Faraday rotation proper turns into zero), whereas for the band with a strong inhomogeneous broadening, this central dip disappears, and the FR noise proves to be the greatest at the band center. Qualitatively, it can be easily understood. The Faraday rotation at the center of the inhomogeneously broadened band vanishes because positive and negative contributions of the higher- and lower-lying spectral components compensate for each other, whereas their fluctuations, being uncorrelated, are summed up statistically and attain the greatest value at the point where the density of the spectral components is the greatest, i.e., at the center of the band.

When the inhomogeneous broadening is comparable with the homogeneous, the dip in the optical spectrum of the FR noise power acquires some intermediate depth, which can be used to estimate the ratio of these two contributions to the line broadening.

It was also established in [84] that the magnitude of the spin noise considerably increases with an increasing ratio of the inhomogeneous bandwidth to the homogeneous (approximately in direct proportion with it). This results from the fact

Figure 16



Typical spectra of optical absorption (a), Faraday rotation (b), and FR noise for the cases of homogeneously (c) and inhomogeneously (d) broadened bands. The two lower plots show schematically how the spin-noise spectra (red peaks) vary with the optical frequency of the probe beam (ν).

that, in the case of a strongly inhomogeneously broadened band, the main contribution to the FR noise of the probe beam is made by the spectral components lying near the laser wavelength (within the range of the homogeneous width). This relatively small number of narrow spectral components with relatively large partial contribution of each of them may provide strong enhancement of the spin-noise power for inhomogeneously broadened bands. This fact, on the one hand, makes easier SNS experiments with inhomogeneously broadened systems (like quantum dots) and, on the other, can be used for measuring the homogeneous linewidths of optical transitions.

The above properties of optical spin-noise (OSN) spectroscopy have been confirmed experimentally in [84]. The OSN spectra of the homogeneously broadened D1 line of potassium atoms and inhomogeneously broadened band of the InGaAs/GaAs quantum dots were strongly different in accordance with the above results. The effect of enhancement of the spin-noise power in inhomogeneously broadened systems was also confirmed experimentally.

Thus, optical spectroscopy of FR noise may be considered the other side of FR-based SNS that may provide interesting additional information about a spin system under study.

10. Unique Properties of Spin-Noise Spectroscopy

SNS, as a method of the ESR spectroscopy, was primarily intended to provide standard information about g-factors and relaxation rates of the spin system. As an optical technique, FR-based SNS has much in common with the conventional optical methods of ESR detection [19,85,86], with an essential difference that the detected spin precession in SNS is spontaneous (stochastic) rather than coherently excited by an external field, and the spin system is supposed to remain in the state of thermal equilibrium. What is highly significant, in our opinion, is that the use of magnetization *noise* in the capacity of signal, in combination with the optical (laser-assisted) technique, allows one to get additional information fundamentally inaccessible for conventional methods of spectroscopy and makes SNS unique in many respects.

One of the most important features of FR-based SNS, as was already mentioned, is related to its nonperturbative character: the light interacting with the paramagnet in the region of its transparency produces virtually no real excitation of the system. This property may be highly important, e.g., in studies of ultracold atoms [87] or semiconductor systems, when appearance of even a small amount of photoinduced charge carriers may substantially distort dynamics of the spin system.

Another property of SNS that essentially distinguishes it from other (fundamentally perturbative) methods of the magnetic-resonance spectroscopy is that it does not require a population difference between magnetic sublevels of the spin system to detect the resonance. This technique remains equally efficient at high temperatures and low magnetic fields (including a zero field), when the population difference becomes negligibly small (see, e.g., [3,11,54]).

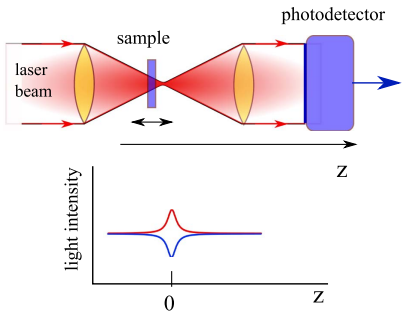
By detecting spin noise as described above, we, in fact, monitor random microscopic motion of a macroscopic object confined spatially by the probe laser beam. This allows us to get information barely accessible for conventional radio or linear optical spectroscopy. In particular, SNS was used to identify

Fermi–Dirac statistics of a degenerate electron gas in heavily doped *n*-GaAs, to distinguish between localized and delocalized conduction-band electrons, and to detect the effect of Brownian motion of the electrons in the conduction band on the spin-noise linewidth. Essential information can be also extracted from the absolute value of the spin-noise power, which is known to be directly related to the total number of spins probed by the beam [41,54,88].

A widely known method of nonlinear optics is the so-called "*Z*-scan technique," which allows one to identify optical nonlinearity and to measure, in a simple way, coefficients of nonlinear absorption or refraction of the medium [89,90]. In all modifications of this technique, the sample is drawn through the waist of a tightly focused laser beam (along the *Z* axis; Fig. 17), and intensity of the transmitted light is measured as a function of the *Z* coordinate. When the sample is optically linear and its optical properties do not depend on the light power density, the transmitted light intensity does not show any dependence on *Z*. The presence of such a dependence with a peculiarity at $Z = 0$ usually serves as an indicator of nonlinearity of the medium. If we apply this *Z*-scan technique to the measurements of spin noise, we will evidently obtain a *Z*-dependent noise signal (as if the medium were nonlinear), because, when moving the sample through the waist of the beam, the number of probed spins controls the magnitude of the spin-noise power. In other words, the tightly focused laser beam traveling through a bulk paramagnet probes the spin system mainly by a small spatial region in the vicinity of its waist. This situation has evidently much in common with the case of a nonlinear medium when the main response is provided by the region of the beam with the greatest light power density.

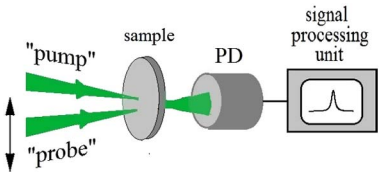
Another curious illustration of this effect may be provided by a simple two-beam experiment, which can be considered a sort of pump–probe spectroscopy of the light intensity noise (Fig. 18). Let a light beam pass through a layer of transparent medium and let us ask the question: Can we find the spot illuminated by this beam on the layer with the aid of another light beam? The usual reply is: Yes, we can do this provided that the first beam (usually called "pump") changes in some way the optical properties of the layer in the illuminated spot. In other words, this is possible (and is a typical story) in *nonlinear* optics. In the light intensity noise, however, this is also possible, in spite of the fact that the probe beam is not supposed to modify, in any way, the properties of the layer.

Figure 17



Schematic of the *Z*-scan arrangement (a) and characteristic dependence of the transmitted light intensity on position of the sample *Z* for the sample with optical nonlinearity.

Figure 18



Schematic of a two-beam intensity-noise-based experiment demonstrating detection of a spot illuminated by a pump beam with the other beam (probe) scanning over the sample layer. The noise modulating the light beam is supposed to be caused by microscopic dynamics of the illuminated spot.

Indeed, if we detect total intensity noise of the two beams (pump and probe) transmitted by the layer (Fig. 18), then the measured signal will depend on whether they pass through the same spot of the layer or not. In the first case, their fluctuations will be correlated, and the total noise power will be given by their sum squared, while, in the second case, they will be uncorrelated, and the measured noise power will be given by the sum of their squares. As a result, the spot pump can be easily detected by the probe. We can say that the illuminated spot is coded in a unique way, and the key to its code is provided by the noise of the pump.

It is appropriate to mention here the idea of two-beam spin-noise spectroscopy, which was put forward in [91] and may be promising for studying the spatial characteristics of spin systems.

These features of SNS have been used in [92] to realize three-dimensional SNS-based tomography. The efficiency of this technique was demonstrated with a pair of *n*-doped GaAs wafers, around 350 μm thick each, probed, as in the conventional *Z*-scan technique, by a focused laser beam. The wavelength of the probe beam was chosen well below the bandgap of GaAs (849 nm). The measurements were performed with no external magnetic field, so that the spin noise spectrum was centered at zero frequency. Due to the different doping concentrations of the two plates, the corresponding spin relaxation times (and spin-noise spectra) were different, which made it possible to distinguish them in the SNS experiment. In this proof-of-principle experiment, a spatial in-depth resolution of 50 μm was demonstrated. The method may allow, in the authors' opinion [93], realization of three-dimensional doping measurements with submicrometer spatial resolution even at low doping concentrations. This is one of the features of SNS that, along with its ability to penetrate inside the hidden structure of optical transitions, mentioned above, makes it close to the methods of nonlinear optics.

11. Conclusions

We have briefly outlined development of SNS over the last several years and described its main achievements and potentialities as applied to scientific research. It is curious that the idea of detecting magnetic resonance in the FR noise spectrum, which looked initially more like an academic trick useful primarily for tutorial purposes, gave birth to a highly efficient and, in many respects, unique experimental tool. This technique keeps certain properties of conventional experimental methods, like magnetic-resonance spectroscopy, optical and Raman spectroscopy, spectroscopy of double RF-optical resonance, and Gorter's paramagnetic

relaxation method, but essentially differs from any of them and acquires thereby qualitatively new features, some of which were considered in this paper.

Nowadays, as we believe, FR-based SNS, as a new experimental tool, is only at the beginning of its life in experimental research, and its potential is far from being exhausted. Especially promising is its application to studies of microsamples and nanostructures, bearing in mind that smallness of the optically probed volume of the sample (smallness of the number of spins) may be considered, under certain conditions, as a favorable factor from the viewpoint of relative magnitude of the signal. An important degree of freedom of SNS, which may considerably widen its informative capacity, is provided by the wavelength of the probe light. Nontrivial shapes of the optical spectra of the spin-noise power may contain information about hidden structure of optical transitions and thus may reveal apparent features of nonlinear optics. Interesting possibilities may be provided by different modifications of the intentionally perturbative (resonant) methods of SNS. These methods, generally, do not have much to do with polarization measurements and do not need high polarimetric sensitivity. Their main advantage is similar to the advantage of the FFT spectrum analyzer compared to the sweeping one: the broadband (rather than monochromatic) intensity modulation of the acting light substantially improves sensitivity of the technique. These methods of active noise spectroscopy, however, pertaining more to nonlinear optics, may provide the information related, to a greater extent, to the optical (rather than spin) dynamics of the system.

At present, we have every reason to believe that the novel technique of SNS will soon turn into a standard method of experimental research. Now, this is a unique case when a pure noise, usually considered a nuisance factor, turns into a basic source of scientific information. The situation, however, is rapidly changing. As the objects under study are getting smaller and smaller, spontaneous thermodynamic fluctuations are becoming more and more important, and, at the end, in the limit of smallest number of particles, the noise cannot be anything other than a signal. This trend may be illustrated by numerous investigations of single quantum dots [93], by studies of single spins [94], and by the latest spectacular advances in the field of magnetometry achieved with the use of the NV-diamond magnetic sensors [95]. All the measurements detecting spontaneous fluctuations of individual spins can reveal unperturbed dynamics of the system that are highly important for understanding the physics of quantum phenomena underlying the experimental observations.

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References

1. S. A. Wolf, D. D. Awschalom, R. A. Buhrman, J. M. Daughton, S. von Molnar, M. L. Roukes, A. Y. Chtchelkanova, and D. M. Treger, "Spintronics: a spin-based electronics vision for the future," Science **294**, 1488–1495 (2001).

2. I. I. Žutić, J. Fabian, and S. Das Sarma, "Spintronics: fundamentals and applications," Rev. Mod. Phys. **76**, 323–410 (2004).
3. E. B. Aleksandrov and V. S. Zapasskii, "Magnetic resonance in the Faraday rotation noise spectrum," J. Exp. Theor. Phys. **54**, 64–67 (1981).
4. T. Sleator, E. L. Hahn, C. Hilbert, and J. Clarke, "Nuclear-spin noise," Phys. Rev. Lett. **55**, 1742–1745 (1985).
5. T. Mitsui, "Spontaneous noise spectroscopy of an atomic magnetic resonance," Phys. Rev. Lett. **84**, 5292–5295 (2000).
6. L. D. Landau and E. M. Lifshitz, *Statistical Physics* (Pergamon, 1980).
7. A. Kuzmich, K. Molmer, and E. S. Polzik, "Spin squeezing in an ensemble of atoms illuminated with squeezed light," Phys. Rev. Lett. **79**, 4782–4785 (1997).
8. J. L. Sorensen, J. Hald, and E. S. Polzik, "Quantum noise of an atomic spin polarization measurement," Phys. Rev. Lett. **80**, 3487–3490 (1998).
9. M. Römer, J. H. Hubner, and M. Oestreich, "Spin noise spectroscopy in semiconductors," Rev. Sci. Instrum. **78**, 103903 (2007).
10. G. Müller, D. Schuh, J. Hubner, and M. Oestreich, "Electron-spin relaxation in bulk GaAs for doping densities close to the metal-to-insulator transition," Phys. Rev. B **81**, 075216 (2010).
11. S. A. Crooker, J. Brandt, S. Sandfort, A. Greilich, D. R. Yakovlev, D. Reuter, A. D. Wieck, and M. Bayer, "Spin noise of electrons and holes in self-assembled quantum dots," Phys. Rev. Lett. **104**, 036601 (2010).
12. A. T. Forrester, R. A. Gudmundsen, and P. O. Johnson, "Photoelectric mixing of incoherent light," Phys. Rev. **99**, 1691–1700 (1955).
13. V. S. Zapasskii and P. P. Feofilov, "Development of polarization magneto-optics of paramagnetic crystals," Sov. Phys. Usp. **18**, 323–342 (1975).
14. R. Hanbury-Brown and R. Q. Twiss, "Correlation between photons in two coherent beams of light," Nature **177**, 27–29 (1956).
15. E. B. Aleksandrov, Yu. M. Golubev, A. V. Lomakin, and V. A. Noskin, "Intensity-fluctuation spectroscopy of optical fields with non-Gaussian statistics," Sov. Phys. Usp. **26**, 643–663 (1983).
16. B. J. Berne and R. Pecora, *Dynamic Light Scattering with Applications to Chemistry, Biology, and Physics* (Wiley Interscience, 1976).
17. E. B. Aleksandrov, "Optical manifestations of the interference of nondegenerate atomic states," Sov. Phys. Usp. **15**, 436–451 (1973).
18. E. B. Aleksandrov, N. T. Kaliteevskii, and M. P. Chaika, "Superhigh-resolution spectroscopy based on interference of states," Sov. Phys. Usp. **22**, 760–767 (1979).
19. V. S. Zapasskii, "Optical detection of spin-system magnetization in rare-earth-activated crystals and glasses," in *Spectroscopy of Solids Containing Rare Earth Ions*, A. A. Kaplyanskii and M. F. Macfarlane, eds. (Elsevier, 1987), pp. 674–711.
20. A. D. Buckingham and P. J. Stephens, "Magnetic optical activity," Annu. Rev. Phys. Chem. **17**, 399–432 (1966).
21. F. Meier and B. Zakharchenya, *Optical Orientation*, Vol. **8** of Modern Problems in Condensed Matter Science Series (North-Holland, 1984).
22. F. Bitter, "The optical detection of radiofrequency resonance," Phys. Rev. **76**, 833–835 (1949).
23. D. Budker and D. F. Jackson Kimball, eds., *Optical Magnetometry* (Cambridge University, 2013).

24. R. Giri, S. Cronenberger, M. Vladimirova, D. Scalbert, K. V. Kavokin, M. M. Glazov, M. Nawrocki, A. Lematre, and J. Bloch, "Giant photoinduced Faraday rotation due to the spin-polarized electron gas in an *n*-GaAs microcavity," Phys. Rev. B **85**, 195313 (2012).
25. H. G. Dehmelt, "Modulation of a light beam by precessing absorbing atoms," Phys. Rev. **105**, 1924–1925 (1957).
26. W. E. Bell and A. L. Bloom, "Optical detection of magnetic resonance in alkali metal vapor," Phys. Rev. **107**, 1559–1565 (1957).
27. V. S. Zapasskii, G. G. Kozlov, and V. A. Malyshev, "Optical analogue of Gorter's paramagnetic relaxation method," Proc. SPIE **1274**, 193–199 (1990).
28. C. J. Gorter, *Paramagnetic Relaxation* (Elsevier, 1947).
29. E. B. Aleksandrov and V. S. Zapasskii, "Modulation magneto- optical spectroscopy of cross-relaxation resonances," V. S. Sov. Phys. Solid State **20**, 679–683 (1978).
30. F. Bloch, "Nuclear induction," Phys. Rev. **70**, 460–474 (1946).
31. A. Kastler, "Displacement of energy levels of atoms by light," J. Opt. Soc. Am. **53**, 902–906 (1963).
32. A. Kastler, "Optical methods for studying Hertzian resonances," Science **158**, 214–221 (1967).
33. W. Happer, "Optical pumping," Rev. Mod. Phys. **44**, 169–249 (1972).
34. S. A. Crooker, D. D. Awschalom, J. J. Baumberg, F. Flack, and N. Samarth, "Optical spin resonance and transverse spin relaxation in magnetic semiconductor quantum wells," Phys. Rev. B **56**, 7574–7588 (1997).
35. V. S. Zapasskii, "Highly sensitive polarimetric measurements," J. Appl. Spectrosc. **37**, 857–869 (1982).
36. E. B. Aleksandrov and V. S. Zapasskii, "Millisecond sensitivity in polarimetric measurements," Opt. Spectrosc. **41**, 502–504 (1976).
37. R. V. Jones, "Rotary 'aether drag'," Proc. R. Soc. A **349**, 423–439 (1976).
38. P. Glasenapp, A. Greilich, I. I. Ryzhov, V. S. Zapasskii, D. R. Yakovlev, A. V. Kavokin, G. G. Kozlov, and M. Bayer, "Resources of polarimetric sensitivity in spin noise spectroscopy," arXiv:1305.7337v1 [cond-mat.mes-hall] (31 May 2013).
39. V. S. Zapasskii, "Depression of excess light noise in polarimetric measurements," Opt. Spectrosc. **47**, 450–451 (1979).
40. S. A. Crooker, D. G. Rickel, A. V. Balatsky, and D. Smith, "Spectroscopy of spontaneous spin noise as a probe of spin dynamics and magnetic resonance," Nature **431**, 49–52 (2004).
41. G. M. Müller, M. Oestreich, M. Römer, and J. Hubner, "Semiconductor spin noise spectroscopy: fundamentals, accomplishments, and challenges," Physica E **43**, 569–587 (2010).
42. A. A. Kharkevich, *Spectra and Analysis* (Springer, 1995).
43. M. A. McCoy and R. R. Ernst, "Nuclear spin noise at room temperature," Chem. Phys. Lett. **159**, 587–593 (1989).
44. M. Gueron and J. L. Leroy, "NMR of water protons: the detection of their nuclear-spin noise, and a simple determination of absolute probe sensitivity based on radiation damping," J. Magn. Reson. **85**, 209–215 (1989).
45. N. Müller and A. Jerschow, "Nuclear spin noise imaging," Proc. Natl. Acad. Sci. USA **103**, 6790–6792 (2006).
46. T. Staudacher, F. Shi, S. Pezzagna, J. Meijer, J. Du, C. A. Meriles, F. Reinhard, and J. Wrachtrup, "Nuclear magnetic resonance spectroscopy on a (5-nanometer)^3 sample volume," Science **339**, 561–563 (2013).

47. E. B. Aleksandrov, V. P. Kozlov, and V. N. Kulyasov, "Noise spectrum of spontaneous emission," Sov. Phys. JETP **39**, 620–626 (1974).
48. E. B. Aleksandrov and V. N. Kulyasov, "Spectra of fluctuations of spontaneous emission," Opt. Spectrosc **40**, 449–452 (1976).
49. D. F. Smirnov and I. V. Sokolov, "Intensity fluctuation spectrum of the nonlinear resonance fluorescence of an atomic system," Sov. Phys. JETP **43**, 1079–1083 (1976).
50. B. Mihaila, S. A. Crooker, D. G. Rickel, K. B. Blagoev, P. B. Littlewood, and D. L. Smith, "Quantitative study of spin noise spectroscopy in a classical gas of 41 K," Phys. Rev. A **74**, 043819 (2006).
51. W. Happer and B. S. Mathur, "Off-resonant light as a probe of optically pumped alkali vapors," Phys. Rev. Lett. **18**, 577–580 (1967).
52. B. M. Gorbovitskii and V. I. Perel, "Aleksandrov and Zapasskii experiment and the Raman effect," Opt. Spectrosc. **54**, 229–230 (1983).
53. D. Eden and H. Swinney, "Optical heterodyne studies of Brillouin scattering in xenon near the critical point," Opt. Commun. **10**, 191–194 (1974).
54. S. A. Crooker, L. Cheng, and D. L. Smith, "Spin noise of conduction electrons in *n*-type bulk GaAs," Phys. Rev. B **79**, 035208 (2009).
55. A. Kuzmich, N. P. Bigelow, and L. Mandel, "Atomic quantum nondemolition measurements and squeezing," Europhys. Lett. **42**, 481–486 (1998).
56. A. Kuzmich, L. Mandel, and N. P. Bigelow, "Generation of spin squeezing via continuous quantum nondemolition measurement," Phys. Rev. Lett. **85**, 1594–1597 (2000).
57. T. Yabuzaki, T. Mitsui, and U. Tanaka, "New type of high-resolution spectroscopy with a diode laser," Phys. Rev. Lett. **67**, 2453–2456 (1991).
58. D. H. McIntyre, C. E. Fairchild, J. Cooper, and F. Walser, "Diode laser noise spectroscopy of rubidium," Opt. Lett. **18**, 1816–1818 (1993).
59. T. Ito, N. Shimomura, and T. Yabuzaki, "Noise spectroscopy of K atoms with a diode laser," J. Phys. Soc. Jpn. **72**, 962–963 (2003).
60. M. Martinelly, P. Valente, H. Failache, D. Felinto, L. S. Cruz, P. Nussenzveig, and A. Lezama, "Noise spectroscopy of nonlinear magneto-optical resonances in Rb vapor," Phys. Rev. A **69**, 043809 (2004).
61. P. Valente, H. Failache, and A. Lezama, "Diode laser noise-spectroscopy of low-frequency atomic fluctuations in rubidium vapor," Eur. Phys. J. D **50**, 133–140 (2008).
62. W. E. Bell and A. L. Bloom, "Optically driven spin precession," Phys. Rev. Lett. **6**, 280–281 (1961).
63. G. Alzetta, A. Gozzini, L. Moi, and G. Orriols, "An experimental method for observation of RF transitions and laser beat resonances in oriented Na vapor," Nuovo Cimento **36**, 5–20 (1976).
64. S. Harris, "Electromagnetically induced transparency," Phys. Today **50**(7), 36–42 (1997).
65. E. B. Aleksandrov, M. Auzinsh, D. Budker, D. F. Kimball, S. M. Rochester, and V. V. Yashchuk, "Dynamic effects in nonlinear magneto-optics of atoms and molecules," J. Opt. Soc. Am. B **22**, 7–20 (2005).
66. R. Walser and P. Zoller, "Laser-noise-induced polarization fluctuations as a spectroscopic tool," Phys. Rev. A **49**, 5067–5077 (1994).
67. G. G. Kozlov and V. S. Zapasskii, "Light-intensity susceptibility and 'active' noise spectroscopy," arXiv: 1206.1921v1 [physics.optics] (9 June 2012).

68. M. Oestreich, M. Römer, R. G. Haug, and D. Hagele, "Spin noise spectroscopy in GaAs," Phys. Rev. Lett. **95**, 216603 (2005).
69. J. N. Kikkawa and D. D. Awschalom, "Resonant spin amplification in *n*-type GaAs," Phys. Rev. Lett. **80**, 4313–4316 (1998).
70. S. Starosielec and D. Hogele, "Ultrafast spin noise spectroscopy," Appl. Phys. Lett. **93**, 051116 (2008).
71. F. Berski, Y. Kuhn, J. G. Lonnemann, J. Hubner, and M. Oestreich, "Ultrahigh bandwidth spin noise spectroscopy: detection of large *g*-factor fluctuations in highly *n*-doped GaAs," arXiv:1207.0081 [cond-mat.mes-hall] (30 June 2012).
72. J. Hubner, J. G. Lonnemann, P. Zell, H. Kuhn, F. Berski, and M. Oestreich, "Rapid scanning of spin noise with two free running ultrafast oscillators," Opt. Express **21**, 5872–5878 (2013).
73. R. Rosenberg, C. B. Rubinstein, and D. R. Herriott, "Resonant optical Faraday rotator," Appl. Opt. **3**, 1079–1083 (1964).
74. H. Y. Ling, "Theoretical investigation of transmission through a Faraday-active Fabry–Perot etalon," J. Opt. Soc. Am. A **11**, 754–758 (1994).
75. A. V. Kavokin, M. R. Vladimirova, M. A. Kaliteevski, O. Lyngnes, J. D. Berger, H. M. Gibbs, and G. Khitrova, "Resonant Faraday rotation in a semiconductor microcavity," Phys. Rev. B **56**, 1087–1090 (1997).
76. G. Salis and M. Moser, "Faraday-rotation spectrum of electron spins in microcavity-embedded GaAs quantum wells," Phys. Rev. B **72**, 115325 (2005).
77. Y. Q. Li, D. W. Steuerman, J. Berezovsky, D. S. Seferos, G. Bazan, and D. D. Awschalom, "Cavity enhanced Faraday rotation of semiconductor quantum dots," Appl. Phys. Lett. **88**, 193126 (2006).
78. L. Dong, Y. Jiang, H. Chen, and Y. Shi, "Enhancement of Faraday rotation effect in heterostructures with magneto-optical metals," J. Appl. Phys. **107**, 093101 (2010).
79. V. S. Zapasskii and S. G. Przhibelskii, "Cavity-enhanced optical anisotropy and all-optical spin noise spectrometer," Opt. Spectrosc. **110**, 917–924 (2011).
80. W. E. Lamb, Jr., "Theory of an optical maser," Phys. Rev. **134**, A1429–A1450 (1964).
81. W. Chalupczak and R. M. Godun, "Near-resonance spin noise spectroscopy," Phys. Rev. A **83**, 032512 (2011).
82. H. Horn, G. M. Müller, E. M. Rasel, L. Santos, J. Hubner, and M. Oestreich, "Spin-noise spectroscopy under resonant optical probing conditions: coherent and nonlinear effects," Phys. Rev. A **84**, 043851 (2011).
83. Q. Huang and D. S. Steel, "Optical excitation effects on spin-noise spectroscopy in semiconductors," Phys. Rev. B **83**, 155204 (2011).
84. V. S. Zapasskii, A. Greilich, S. A. Crooker, Y. Li, G. G. Kozlov, D. R. Yakovlev, D. Reuter, A. D. Wieck, and M. Bayer, "Optical spectroscopy of spin noise," Phys. Rev. Lett. **110**, 176601 (2013).
85. J. Brossel and F. Bitter, "A new 'double resonance' method for investigating atomic energy levels. Application to Hg $^3P_1$," Phys. Rev. **86**, 308–316 (1952).
86. E. B. Aleksandrov and V. S. Zapasskii, "Use of lasers to study magnetic resonance and magnetic relaxation," J. Sov. Laser Res. **6**, 291–296 (1985).
87. B. Mihaila, S. A. Crooker, K. B. Blagoev, D. G. Rickel, P. B. Littlewood, and D. L. Smith, "Spin noise spectroscopy to probe quantum states of ultracold fermionic atom gases," Phys. Rev. A **74**, 63608 (2006).

88. G. M. Müller, M. Römer, D. Schuh, W. Wegscheider, J. Hubner, and M. Oestreich, "Spin noise spectroscopy in GaAs (110) quantum wells: access to intrinsic spin lifetimes and equilibrium electron dynamics," Phys. Rev. Lett. **101**, 206601 (2008).
89. M. Sheik-Bahae, A. A. Said, and E. W. Van Stryland, "High-sensitivity, single-beam n_2 measurements," Opt. Lett. **14**, 955–957 (1989).
90. M. Sheik-Bahae, A. A. Said, T.-H. Wei, D. J. Hagan, and E. W. Van Stryland, "Sensitive measurement of optical nonlinearities using a single beam," IEEE J. Quantum Electron. **26**, 760–769 (1990).
91. Yu. V. Pershin, V. A. Slipko, D. Roy, and N. A. Sinitsyn, "Two-beam spin noise spectroscopy," Appl. Phys. Lett. **102**, 202405 (2013).
92. M. Römer, J. Hubner, and M. Oestreich, "Spatially resolved doping concentration measurement in semiconductors via spin noise spectroscopy," Appl. Phys. Lett. **94**, 112105 (2009).
93. P. Michler, ed., *Single Quantum Dots, Fundamentals, Applications and New Concepts* (Springer, 2003).
94. Z. Nussinov, M. F. Crommie, and A. V. Balatsky, "Noise spectroscopy of a single spin with spin-polarized STM," Phys. Rev. B **68**, 085402 (2003).
95. J. Miller, "Diamond defects enable nanoscale nuclear magnetic resonance," Phys. Today **66**(4), 12–14 (2013).

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