Optics and Spectroscopy, 2022, Vol. 130, No. 11

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Spin Noise Spectroscopy: Three Plots

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Received September 7, 2022
Revised September 22, 2022
Accepted September 22, 2022

The effect of magnetic resonance in the Faraday-rotation noise, demonstrated for the first time on an atom system more than 40 years ago, laid the foundations of a new research direction, the spin noise spectroscopy, that has been developing extensively in the last 20 years. Currently, the most popular research objects are atom and semiconductor paramagnetic materials, while research subjects are the magnetic resonance spectroscopy, the spin dynamics, the optical spectroscopy, the spectroscopy of light polarization noises. The rapid development of this new research direction laying at the interface between optics and radio spectroscopy continuously extends the field of its applicability and opens new opportunities to use it in the experimental physics. In this article we present a few episodes that are important stages in the spin noise spectroscopy development in the last years. The article was prepared as a report on the XV Russian Conference on the Physics of Semiconductors.

Keywords: spin noise spectroscopy, magnetic resonance, optical anisotropy, inhomogeneous broadening.

DOI: 10.21883/EOS.2022.11.55098.4095-22

1. Introduction

Spin fluctuation spectroscopy or spin noise spectroscopy (SNS) is a new scientific field that combines magnetic resonance spectroscopy with optical spectroscopy. The idea of registering spontaneous fluctuations of magnetization, which underlies SFS, was first proposed by F. Bloch in 1946 [1] and was experimentally realized in 1981 on alkali metal vapor [2]. Spin noises (or magnetization noises) in the work [2] were registered by Faraday rotation noises, the high detection sensitivity of which was provided by the laser polarimetry technique [3,4]. Magnetic resonance was observed as a peak in the Faraday rotation (FR) noise power spectrum at the spin precession frequency (at the Larmor frequency). It is important here that the spin precession frequency in the SFS is measured without its excitation by an external alternating field. Such a „non-perturbing" character of the measurement procedure radically distinguishes the SFS from all known methods of magnetic resonance detection, in which the term „resonance" itself implies that the frequency of the external action coincides with the natural precession frequency of the free spin.

The discovered effect of magnetic resonance in the FR noise spectrum was originally considered only as a successful experimental illustration of the fluctuation-dissipation theorem [5] and no applied value was attached to it. Spin noise spectroscopy gained significant popularity only at the beginning of this century, after its successful application to semiconductor structures [6,7] and after a radical increase in the sensitivity of the method by replacing scanning-type spectrum analyzers with their analogs based on the fast Fourier transform [8–10]. To date, a great deal of experience has already been accumulated in the application of SFS to atomic and semiconductor systems, and a number of surprising possibilities of the method have been discovered.

The additional resource of the SFS method is realized under conditions of optical resonance, when a light beam brings the spin system out of thermodynamic equilibrium [11–13]. At the same time, an important feature of noise spectroscopy is that this method, which does not imply an optical nonlinearity of the medium, often exhibits properties that are more typical of nonlinear optics [14]. Thus, the noise signal exhibits a dependence on the optical power density of the probe beam, which was used to implement the optical version of three-dimensional tomography of bulk semiconductor structures [15]. Correlation-statistical patterns of noise signal formation make SFS sensitive to the homogeneous width of inhomogeneously broadened optical transitions [16] and make it possible not only to conduct experiments of „pump–probe" type [17,18], but also to avoid the suppression of the stochastic gyration signal typical for a regular response in birefringent media [19].

In this article, we consider three subjects that, in our opinion, have significantly influenced the development of spin noise spectroscopy in recent years. The first of them is related to the sensitivity of spin noise to the nature of the broadening of the optical transition, the second concerns the applicability of the method to optically anisotropic media, and the third relates to the violation of the generally accepted laws of observation of spin precession in media with anisotropic centers. All of the aforementioned subjects are of general interest for spin noise spectroscopy; however, they turned out to be extremely topical when applied to crystals with rare-earth (RE) ions and are illustrated by the example of these systems.

2. Formation of the spin noise signal at the transition with inhomogeneous broadening

2.1. General description of the effect

The spectral behavior of the paramagnetic Faraday effect in the region of an isolated spectral line approximately coincides with the spectrum of the corresponding line contribution to the refractive index of the medium. In the presence of several spectral lines, the full spectrum of the Faraday rotation is represented by the sum of individual contributions regardless of the origin of these lines. In the optical noise spectra of Faraday rotation, the cross-correlation characteristics of individual contributions play an important role, and the process of formation of the overall noise power spectrum is no longer unambiguous. The most expressive manifestation of this specificity of the SFS was found in resonance studies of inhomogeneously broadened transitions.

An unusual form of the optical power spectrum of the Faraday rotation noise was first described in [16] under conditions of the resonant probing of an ensemble of (In,Ga)As quantum dots. It seemed paradoxical that the FR noise power reached its maximum at the center of the transition, where the value of the regular FR turned to zero. A more rigorous theoretical consideration of the problem showed that, indeed, in the case when the homogeneous transition width turns out to be much smaller than the inhomogeneous one and the adjacent spectral packets of the line contour fluctuate independently, the noise power at each point of the contour is determined only by the density of oscillators. In the case of an optical transition with homogeneous broadening, the FR fluctuates in a correlated manner over the entire line contour, the noise power spectrum is described by the square of the spectrum of the regular Faraday effect and, obviously, vanishes at the point where the Faraday effect itself vanishes.

Of interest is the evolution of the FR noise power spectrum in the region of optical resonance as the relative homogeneous transition width decreases, i.e., as we go from a homogeneously broadened line to a line with dominant inhomogeneous broadening. The behavior of this evolution is represented graphically in Fig. 1. As can be seen from the figure, the dip in the optical noise power spectrum overgrows when the homogeneous width is about half the full width, and then the peak noise power increases, as the ratio of the inhomogeneous linewidth to the homogeneous increases, almost unlimitedly. In comparison with the noise power at the inhomogeneously broadened transition, this amplification factor is determined by the ratio of the inhomogeneous linewidth to the homogeneous one, and in some cases can be many orders of magnitude. It is this spin noise amplification effect that played the key role in the application of SFS to crystals with rare-earth impurity ions [20].

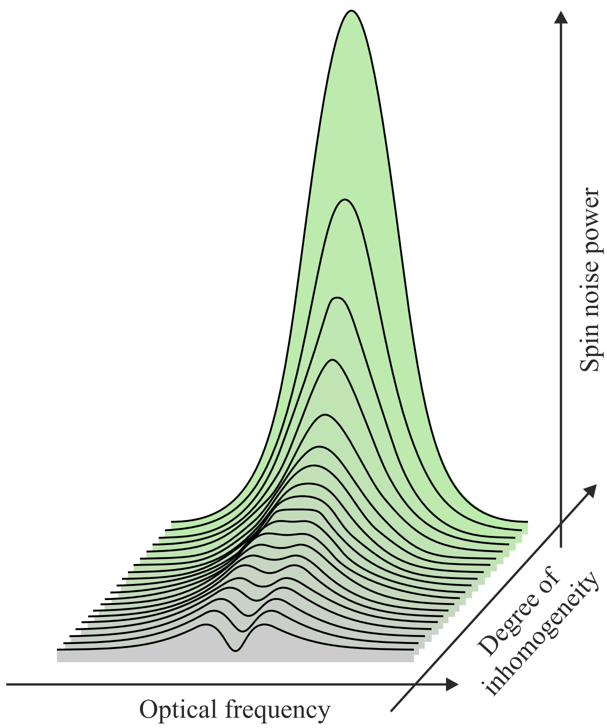

Figure 1. Schematic representation of the evolution of the optical power spectrum of spin noise as the ratio of the inhomogeneous width of the optical transition to the homogeneous one increases.

2.2. Application of SFS to crystals with RE impurities

Impurity ions of RE metals in crystals are known to exhibit two essentially different types of optical transitions: (1) allowed interconfigurational (4*f*–5*d*) transitions with broad homogeneously broadened absorption bands and strong magneto-optical activity, and (2) relatively weak, parity forbidden intraconfigurational (*f*–*f*) transitions with narrow lines and low magneto-optical activity. Initially, crystals with divalent RE ions (Eu^{2+}, Dy^{2+}, Tm^{2+}) with interconfigurational transition bands located in the visible region of the spectrum were treated as most perspective. However, numerical estimates showed that even in crystals with these ions, the contribution to the FR per ion (or, more precisely, the FR cross section [21]) turned out to be too small for the application of the SFS method. A more careful consideration, however, showed that transitions with low magneto-optical activity*f*–*f*, which at first glance are of little use, are the most promising in this sense. Indeed, the so-called narrow lines of *f*–*f* transitions have a fractional scale width of cm^{-1} (usually from 1 to 10 GHz). It is important, however, that this is a result of the inhomogeneous broadening of transitions associated with fluctuations of local crystal fields, while their homogeneous width can lie in the range of kHz and even fractions of them [22]. Therefore, despite the relatively small regular Faraday effect on *f*–*f* transitions, the spin noise signal amplification factor mentioned in the previous subsection

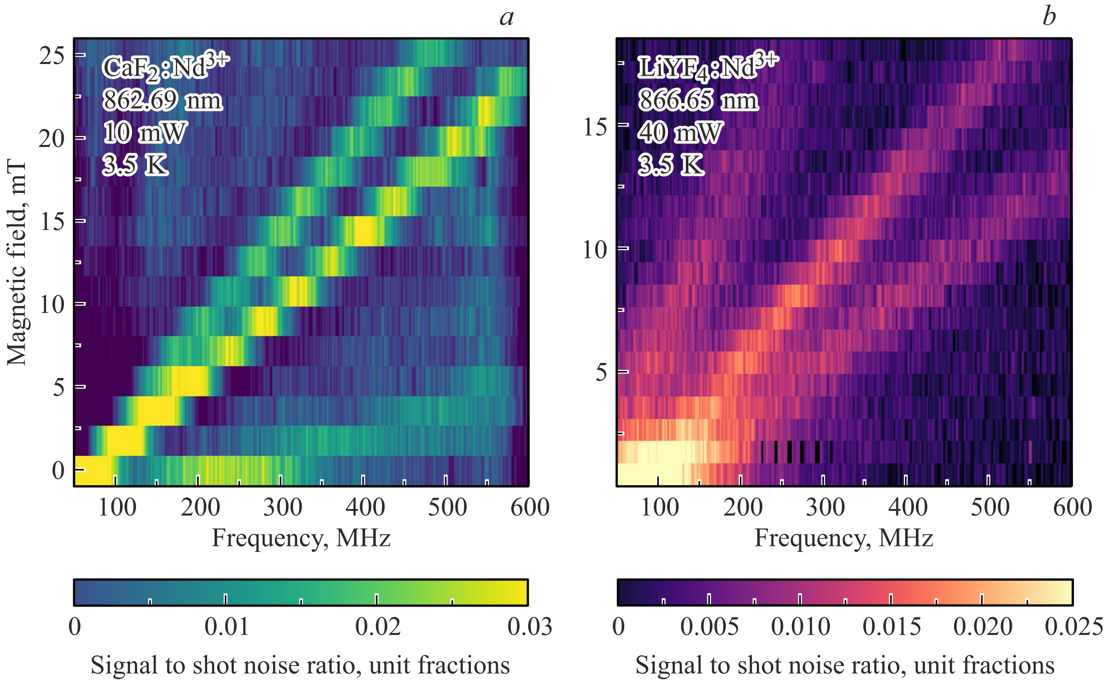

Figure 2. Field dependences of spin noise spectra of Nd^{3+} ions in a cubic CaF_2 (*a*) crystal and in a uniaxial $LiYF_4$ (*b*) crystal. The key parameters of the experiment are shown in the panels.

can reach 6–7 orders of magnitude, which makes such systems extremely favorable for the SFS method.

The result of the first successful experiments carried out on a fluorite crystal activated with a trivalent neodymium ion is shown in Fig. 2, *a*. It is essential that to realize the indicated gain in the spin noise signal, the spectral width of the probe beam must be less than the homogeneous linewidth. In our experiments [19,20,23], a tunable titanium-sapphire ring cavity laser with a generation linewidth of less than 1 MHz was used as a probe radiation source. The work [20] paved the way for the application of SFS to rare earth systems.

3. Application of SFS to anisotropic crystals

The Faraday method of recording magnetization noise is based on the assumption that the FR is proportional to the magnetization of the medium or, more precisely, the projection of the magnetization on the direction of light propagation. Such a pattern of noise signal formation determines the natural scheme for observing magnetization noise: the probe beam, which records oscillations of the transverse magnetization component, must be directed across the applied external field (so-called Voigt geometry). It is also obvious that when the probe light propagates along the applied magnetic field (in the Faraday geometry), the projection of the magnetization precessing around the field does not oscillate, and the spin precession signal should not be observed [24]. Such a harmonious picture of the formation of the FR signal and its noise implies that the magnetization of the medium is the only source of its optical anisotropy. The situation changes significantly for optically anisotropic media, for example, low-symmetry crystals. In such media, light loses its ability to accumulate a rotation of the plane of polarization as it propagates through the crystal. The polarization evolution of light in this case is often described by the precession of the Stokes vector (or quasi-spin) over the Poincaré sphere around the effective magnetic field created by linear birefringence. In such a model, the gyration of the medium usually creates only a small orthogonal addition to this field, which practically does not affect the polarization evolution of light. This fact of the suppression of the regular Faraday rotation in optically anisotropic media seemed to indicate the impossibility of detecting FR noise in such media, and, consequently, the inapplicability of spin noise spectroscopy to them. It turned out, however, that this is not the case for spatially uncorrelated gyration fluctuations. In the work [19] this was shown theoretically and confirmed experimentally on the example of uniaxial crystals $CaWO_4$ and $LiYF_4$ doped with neodymium (Fig. 2, *b*). It is interesting that despite the significant distortion of the probe light polarization during its propagation through the crystal, the noise signal turns out to be practically unsuppressed and, moreover, depends extremely weakly on the azimuth of the probe beam polarization plane. All these patterns find their explanation in a strict theoretical description of the problem and, most importantly, significantly expand the possibilities of the SNS and the range of its potential objects. For a qualitative understanding of this result, it is necessary to pay attention to the difference in the processes of summation of stochastic and regular polarization signals in the presence of optical anisotropy.

4. Schemes for observing magnetization noise in crystals with anisotropic centers

One of the standard purposes of magnetic resonance spectroscopy is to reveal the structure and symmetry of anisotropic paramagnetic centers. In SFS, measurements of spin precession frequencies are usually carried out in magnetic fields, which can be created using permanent magnets located outside the low-temperature cryostat. The study of the orientational dependence of the spin noise spectra in this case is easily achieved by mechanical rotation of a permanent magnet. Studies of the spin noise spectra of fluorite crystals with Nd^{3+} impurity centers carried out in this way made it possible to identify the type of the center, and also to establish some invariant relationships between the frequencies of spin resonances of individual groups of centers [23]. It is significant that in this experiment the external magnetic field, as it rotated, successively passed through the Voigt geometry and the Faraday geometry, without leading to significant changes in the signal amplitude. Our further studies of crystals with anisotropic centers showed that the spin precession noise resonances in them usually do not nullify in the Faraday geometry, which, as noted at the beginning of the previous section, seems paradoxical.

The simplest explanation for this violation of the standard spin noise detection scheme can be represented as follows. Let us consider an anisotropic paramagnetic center arbitrarily oriented in an external magnetic field, along which a probe laser beam is directed. It can be assumed that due to the magnetic anisotropy of the center, the effective magnetic field acting on it turns out to be deviated from the external one and the Faraday geometry of the experiment is violated. This circumstance causes the appearance of a noise peak at the Larmor frequency for the collinear geometry of the probing beam and the magnetic field. However, a consistent solution of the model problem of polarimetric noise generated by a system of paramagnetic ions in a crystalline matrix under conditions of resonant probing shows that the described simple picture has only a qualitative meaning. Quantitative analysis involving the calculation of fluctuations of the gyrotropic part of the polarizability tensor of paramagnetic ions in a crystal field under optical resonance conditions will be given in one of our future publications.

5. Conclusion

The presented fragment of the latest achievements in spin noise spectroscopy, in our opinion, well demonstrates the dynamics of the development of this scientific field. An unexpected and paradoxical result of these studies was the successful application of FR noise spectroscopy to optically anisotropic systems, to which regular FR spectroscopy is practically inapplicable. Additional theoretical justification is needed for the fact that the studied crystals almost completely lost their anisotropy in the azimuthal dependences of the noise signals. A radical expansion of the possibilities of spin noise spectroscopy was also achieved due to the successful application of this method to dielectric crystals with RE ions. The research results described in the article, in our opinion, make a significant contribution to the development of spin noise spectroscopy and will undoubtedly find application in optical and radiospectroscopic studies of paramagnetic media.

Funding

The team of authors is grateful to St. Petersburg State University for support of this work (grant № 94030557). The experimental studies presented in Sections II.B and III were supported by a grant from the Russian Science Foundation № 21-72-10021 „Resonant laser spectroscopy of polarization fluctuations“. The work was carried out at the site of the Resource Center „Nanophotonics“ of the Research Park, St. Petersburg University.

Conflict of interest

The authors declare that they have no conflict of interest.

References

[1] F. Bloch. Phys. Rev., **70**, 460 (1946). DOI: 10.1103/PhysRev.70.460
[2] E. B. Alexandrov, V. S. Zapassky. ZhETF, **81** (1), 132 (1981) (in Russian)
[3] E. B. Alexandrov, V. S. Zapassky.. Optika i spektroskopiya, **41** (5), 855 (1976) (in Russian).
V. S. Zapassky. Zhurnal prikladnoy spektroskopii, **37** (2), 184 (1982) (in Russian).
[5] R. Kubo. Reports on Progress in Physics, **29** (1), 255 (1966).
[6] M. Oestreich, M. Römer, R. J. Haug, D. Hägele. Phys. Rev. Lett., **95**, 216603 (2005). DOI: 10.1103/PhysRevLett.95.216603
[7] G. M. Müller, M. Oestreich, M. Römer, J. Hübner. Physica E: Low-dimensional Systems and Nanostructures, **43** (2), 569 (2010). DOI: 10.1016/j.physe.2010.08.010
[8] N. Thrane. Technical Review, **1–1979** (1979).
[9] G. M. Müller, M. Römer, J. H.bner, M. Oestreich. Applied Physics Letters, **97** (19), 192109 (2010). DOI: 10.1063/1.3505342
[10] S. A. Crooker, J. Brandt, C. Sandfort, A. Greilich, D. R. Yakovlev, D. Reuter, A. D. Wieck, M. Bayer. Phys. Rev. Lett., **104**, 036601 (2010). DOI: 10.1103/PhysRevLett.104.036601
[11] P. Glasenapp, N. A. Sinitsyn, L. Yang, D. G. Rickel, D. Roy, A. Greilich, M. Bayer, S. A. Crooker. Phys. Rev. Lett., **113**, 156601 (2014). DOI: 10.1103/PhysRevLett.113.156601

[12] Q. Huang, D. S. Steel. Phys. Rev. B, **83**, 155204 (2011). DOI: 10.1103/PhysRevB.83.155204
[13] S. A. Crooker, L. Cheng, D. L. Smith. Phys. Rev. B, **79**, 035208 (2009). DOI: 10.1103/PhysRevB.79.035208
[14] M. M. Glazov, V. S. Zapasskii. Opt. Express, **23** (9), 11713 (2015). DOI: 10.1364/OE.23.011713
[15] M. Römer, J. Hübner, M. Oestreich. Applied Physics Letters, **94** (11), 112105 (2009). DOI: 10.1063/1.3098074
[16] V. S. Zapasskii, A. Greilich, S. A. Crooker, Y. Li, G. G. Kozlov, D. R. Yakovlev, D. Reuter, A. D. Wieck, M. Bayer. Phys. Rev. Lett., **110**, 176601 (2013). DOI: 10.1103/PhysRevLett.110.176601
[17] L. Yang, P. Glasenapp, A. Greilich, D. Reuter, A. D. Wieck, D. R. Yakovlev, M. Bayer, S. A. Crooker. Nature Communications, **5**, (2014).
[18] D. Roy, L. Yang, S. A. Crooker, N. A. Sinitsyn. Scientific Reports, **5** (1), 9573 (2015). DOI: 10.1038/srep09573
[19] V. O. Kozlov, N. S. Kuznetsov, D. S. Smirnov, I. I. Ryzhov, G. G. Kozlov, V. S. Zapasskii. Phys. Rev. Lett., **129**, 077401 (2022). DOI: 10.1103/PhysRevLett.129.077401
[20] A. N. Kamenskii, A. Greilich, I. I. Ryzhov, G. G. Kozlov, M. Bayer, V. S. Zapasskii. Physical Review Research, **2** (2), 023317 (2020). DOI: 10.1103/physrevresearch.2.023317
[21] R. Giri, S. Cronenberger, M. Vladimirova, D. Scalbert, K. V. Kavokin, M. M. Glazov, M. Nawrocki, A. Lemaître, J. Bloch. Phys. Rev. B, **85**, 195313 (2012). DOI: 10.1103/PhysRevB.85.195313
[22] V. S. Zapasskii. *Optical detection of spin-system magnetization in rare-earth-activated crystals and glasses*, chap. 12, pp. 673–709, (Elsevier1987).
[23] A. N. Kamenskii, V. O. Kozlov, N. S. Kuznetsov, I. I. Ryzhov, G. G. Kozlov, M. Bayer, A. Greilich, V. S. Zapasskii. Phys. Rev. B, **105**, 014416 (2022). DOI: 10.1103/PhysRevB.105.014416
[24] V. S. Zapasskii. Advances in Optics and Photonics, **5, issue 2**, 131 (2013). DOI: 10.1364/AOP.5.000131