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OPTICAL PROPERTIES

Polarimetry of Regular and Stochastic Signals in Magnetooptics

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Abstract—A brief review is presented of application of the high-sensitive laser polarimetry for detecting small changes—either regular, or stochastic—of the medium magnetization. Principles and results of application of the spectroscopy of spin fluctuations to atomic and semiconductor systems are described. New possibilities of studying the radiospectroscopic and optical properties of spin systems using the methods of noise spectroscopy are considered. Prospects for expanding the circle of objects of the spin noise spectroscopy at the expense of impurity dielectrics are discussed.

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INTRODUCTION

Among optical media of the modern photonics and optoelectronics, an important place is occupied by paramagnets, namely, by materials and structures containing elementary carriers of angular momentum (spin). Complementary degrees of freedom associated with the spin system are revealed in specificities of interaction between the light and the medium, which allow one to apply optical methods for studying spin dynamics. Interconnection between the optical and magnetic properties of spin systems finds many applications not only in scientific research, but also for solving practical problems of processing and storing information [1].

In virtue of the pseudovector nature of angular momentum (as well as of the magnetization vector), the permittivity tensor of the magnetized medium also acquires properties of the axial vector. For this reason, the magnetized paramagnet becomes magnetooptically active, i.e., acquires ability to rotate polarization plane of the transmitted light. This remarkable effect, discovered by M. Faraday in 1845, not only established direct relationship between two fields of physics—optics and magnetism—but has also demonstrated specific symmetry-related features of magnetic perturbation of the medium and, eventually, opened the possibility of monitoring magnetization of a spin system by means of polarimetry.

As applied to the contemporary experimental investigations, the Faraday effect is often associated with the polarization plane rotation in any magnetized medium regardless of the external magnetic field (which is implied in the classical definition of the Faraday effect). The magnetization can be nonequilibrium or nonstationary; it can be induced by optical pumping or by a high-frequency magnetic field. In any case, the magnetization aligned along the light beam propagation will be revealed in the effect of magnetooptical rotation.

In this paper, we briefly consider evolution of using the Faraday effect as a tool for magnetic measurements. We will dwell, in more detail, on the possibilities of optical measuements of magnetization fluctuations, and will discuss conditions under which the optical measurements of the fluctuation characteristics of the magnetization can be efficient.

METHODOLOGICAL ASPECT

An important significance in the polarimetric detection of magnetization of a magnetically diluted paramagnet has the sensitivity of the measurements, namely, the scale of the smallest detectable angles of the polarization plane rotation. As is known, any polarimetric measurement, in the optical spectral range, is based on the Malus law that specifies how variations of the light polarization state are converted into changes of the light intensity, after which the task of measuring variations in the light polarization is reduced to a purely photometric problem. Sensitivity of these measurements is known to be limited by shot noise of the photodetector (or, better to say, by noise of the shot noise), which, for a given frequency bandwidth, is determined only by the value of the photocurrent.

It is noteworthy that in the prelaser times, the problem of fundamental limit of polarimetric sensitivity was not topical, since the sensitivity of the optical polarimeters and spectropolarimeters was restricted mainly by electronic noise of the instrument. For the first time, the problem of realization of the ultimate polarimetric sensitivity has been set and solved in publications [2, 3], where lasers have been used as light

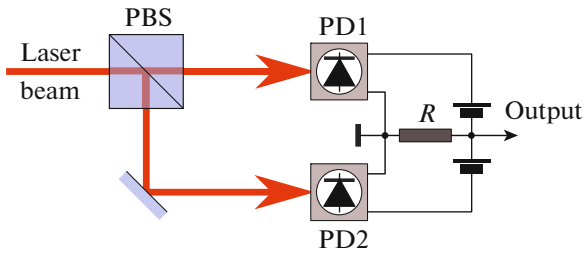

Fig. 1. Balanced detector—heart of the up-to-date laser polarimeter. PBS—polarizing beamsplitter, PD1 and PD2—photodetectors, *R*—load resistor.

sources. As a result, sensitivity of the polarimetric measurements has been increased by more than three orders of magnitude. It was found that the smallest detectable angles (for the accumulation time of 1 s and light power of about 1 mW) lied in the range of 10^{-8} rad. The main obstacle on the way to realization of this highest sensitivity was related to the excess intensity noise of the laser source. A number of tricks have been proposed to overcome this obstacle [4]. The most important component contributing to a suppression of the excess noise was the balanced detector (Fig. 1), which became an indispensable attribute of any up-to-date sensitive polarimeter. Thus, 1976 became the year of laser revolution in the polarization magnetooptics, for which the high polarimetric sensitivity signified possibility to detect extremely small variations of magnetization.

THE EFFECTS OF REGULAR RESPONSE

The achieved level of polarimetric sensitivity gave a new impetus to optical methods of magnetic measurements. The most interesting results were obtained in the works on optical detection of electron paramagnetic resonance (EPR) in crystals and glasses with paramagnetic impurities. It was shown that in most cases of laser-polarimetric detection of small magnetization changes, the detuning of the probe light wavelength from optical resonance ceases to be a critical parameter of the experiment: the paramagnetic contribution to the Faraday effect is usually able to provide high sensitivity of the measurements in the whole spectral range regardless of energy structure of a particular impurity ion [5].

As a result, the range of abilities of optical technique of the magnetic resonance detection has been significantly widened. In particular, the magnetic resonance spectrum of a divalent thulium ion in fluorite-type crystals could be easily observed in the low-field and low-frequency regions (of the order of the resonance linewidth) [6], and the EPR spectra of all (paramagnetic) trivalent rare-earth ions, which are known to be deprived of allowed optical transitions in the visible spectral range, were reliably detected using one and the same source of the probe light (helium–neon laser) [7].

The high sensitivity of laser polarimeters was also successfully used to measure the "nonresonant" magnetic susceptibility of impurity paramagnets at low frequencies, when the magnitude of the optical response was controlled by the ratio between the frequency of the applied AC field and the rate of the relevant relaxation process. Such measurements made it possible to separate the van Vleck (polarization-type) contribution to the magnetic susceptibility from the population-related contribution, to detect and study inter-ion cross-relaxation processes, and to get rather specific information about anisotropy and structure of paramagnetic centers in glasses [5, 8, 9].

Nowadays, the laser polarimetry of magnetooptical response with the highest (shot-noise-limited) sensitivity has taken an important place in the arsenal of tools of modern optics and has turned into one of its routine methods.

MAGNETIC RESONANCE IN THE FARADAY ROTATION NOISE

Impressive sensitivity of the laser polarimetry to small magnetization changes of transparent paramagnets made realistic the idea to observe experimentally fluctuations of the equilibrium magnetization associated with random motion of elementary magnetic moments. For the measurements in the external magnetic field, one can consider two experimental configurations.

In the first of them—the so-called Faraday geometry (Fig. 2a)—the probe laser beam passes through the paramagnet under study along the applied magnetic field. In this case, the fluctuations of the Faraday rotation (FR) reflect fluctuations of the longitudinal magnetization of the medium with the characteristic time scale controlled by the longitudinal relaxation time T_1. The spectrum of such noise will be described by a Lorentzian centered at zero frequency with a width corresponding to the longitudinal relaxation rate T_1^{-1}.

Of much more interest is the so-caller Voigt geometry (Fig. 2b), when the probe light beam is directed across the applied magnetic field. In this case, the light (detecting rotation of the polarization plane) responds to the magnetization directed across the applied field, which, on average, is zero. However, any fluctuation component of the magnetization directed along the beam, due to the precessive character of the motion of all the elementary moments, will precess around the applied field during the transverse relaxation time T_2, after which it will be replaced by a new random realization of the fluctuation with a new amplitude and new phase. The spectrum of such a noise signal will be described by a Lorentzian centered at the Larmor frequency with the width corresponding to the transverse

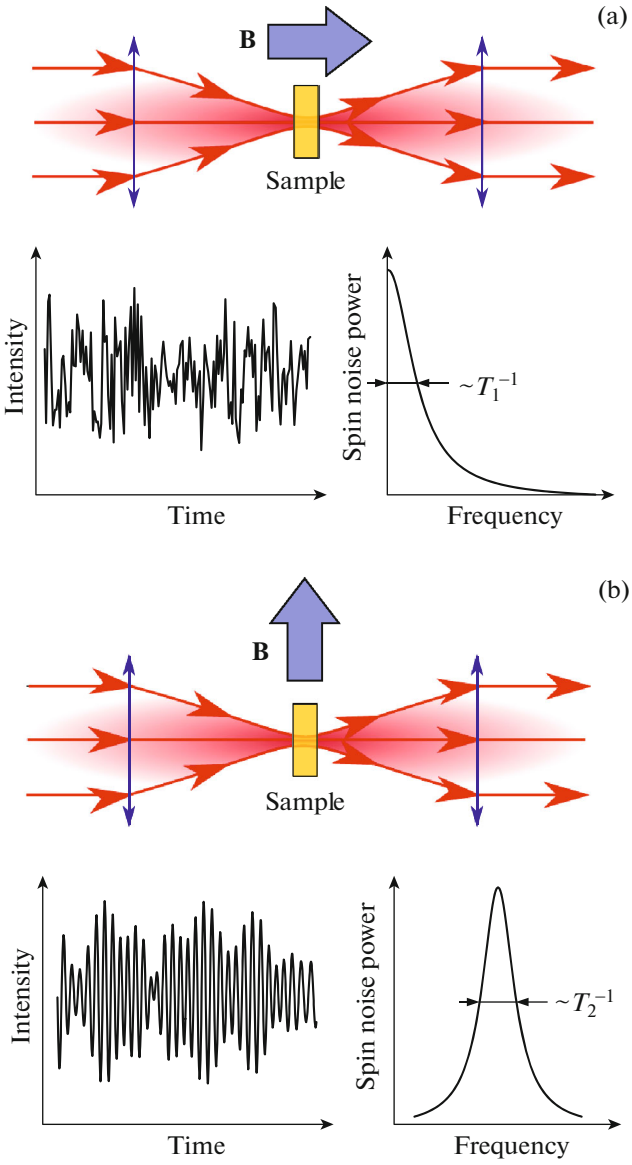

Fig. 2. The Faraday (a) and Voigt (b) geometries—the two main configurations for spin noise detection. Below each of the arrangements, we show schematically time dependences of the noise signals and their spectra.

relaxation rate T_2^{-1}. In other words, the FR noise spectrum, in this experimental geometry, will be nothing else as the magnetic resonance spectrum of the paramagnet under study.

This idea was implemented experimentally in 1981 [10]. The measurements were performed on sodium vapor at a temperature of ~50°C. The probe beam of a dye laser was tuned to close vicinity of one of the sodium *D*-lines, where the FR of the vapor strongly increased, which corresponded to extremely high efficiency of conversion of the magnetization to Faraday rotation. The experimental results of this study have confirmed correctness of the choice of the object—the magnetic resonance noise signal has been detected with a signal-to-noise ratio of about 10^2.

SPECIFIC FEATURES OF THE NOISE SPECTROSCOPY

The above experimental detection of magnetic resonance in the FR noise spectrum can be considered, from an academic viewpoint, as an illustration to the fluctuation-dissipative theorem [11], which establishes a direct connection between the spectrum of the susceptibility of the system and the spectrum of its spontaneous fluctuations. The other side of the experiment was that it offered a conceptually new method for detecting magnetic resonance, an alternative to the conventional Zavoisky's EPR technique. The new approach to the magnetic resonance detection determined its distinctive features that boiled down, first of all, to the following.

1. All conventional methods of the magnetic resonance detection involve excitation of the system by an AC magnetic field. In the spin noise spectroscopy, (SNS) the probe laser beam propagates through the medium in the region of its transparency, does not cause real optical transitions and, thus, does not perturb the system. In this sense, the SNS realizes a nonperturbative magnetic resonance technique.

2. A characteristic feature of all known methods of the magnetic resonance detection is that they imply magnetic polarization of the medium (unequal populations of the magnetic sublevels), which makes it possible to absorb the high-frequency energy. In the SNS, no magnetic polarization of the medium is required for the magnetic resonance detection. This allows one to perform measurements at high temperatures and in small magnetic fields.

3. A doubtless technical merit of the noise technique is associated with the absence of the high-frequency excitation channel, which radically simplifies the design of the magnetic resonance spectrometer and excludes the necessity to strictly match the value of the external field to the resonance frequency.

Special attention should be paid to the question of sensitivity of the SNS as a method of magnetic resonance detection. The magnitude of the spin-noise signal is approximately equal to $1/\sqrt{N}$ of the magnetic saturation signal, and for macroscopic samples, is obviously extremely small. This is the reason why the fate of the new EPR spectroscopy proved to be hard: almost a quarter of century passed after publication [10] before the spin noise method was recognized as a practical tool of research. The first successful detection of the noise magnetic resonance in a semiconductor sample (*n*-GaAs) was performed in 2005 [12]. In that experiment, the signal accumulation time was several hours. The above mentioned absence of strict fixation of the magnetic field with respect to the AC-field frequency made it possible to radically increase

sensitivity of the measurements using a spectrum analyzer with a fast Fourier transform (instead of a scanning-type spectrum analyzer) [13–15]. After this improvement, the SNS has experienced a rapid growth—hundreds of papers and several reviews have been published since then (see, for e.g., [16–19]). A significant role in the growth of popularity of this experimental method was played by a number of specific features of the SNS not predicted at the early stages of its development and related mainly to its optical degrees of freedom.

OPTICAL ASPECTS OF THE SPIN-NOISE SIGNAL FORMATION

Variable wavelength and variable geometry of the probe laser beam provide the noise method of radiospectroscopy with an additional, rather curious, information channel. First of all, recall that the relative magnitude of the magnetization fluctuation detected in the SNS, is controlled by the factor $1/\sqrt{N}$ (N is the number of spins that form the signal) and increases with decreasing N. Therefore, unlike the regular FR signal induced by external magnetic field, the magnitude of the spontaneous noise signal depends on the light beam cross section, increasing as it decreases. For the same reason, when a bulk medium is probed by a focused laser beam, the focal region makes the main contribution to the noise signal. This allows one, by moving the focal point over the volume of the medium, to monitor spatial relief of the paramagnet and, thus, to realize a version of the 3D magnetic tomography [20].

Note that the dependence of the noise signal on the beam cross-section is nothing else than dependence of the signal on the optical power density, which is known to be more typical for the effects of nonlinear optics that imply nonlinearity in optical susceptibility of the medium. This specific feature of the SNS is also revealed in a number of other effects [21].

Another feature of the SNS that brings it closer to nonlinear optics is its ability to detect hidden structure of optical transitions and to identify the nature of broadening of the optical absorption bands. In particular, it was shown in [22] that the character of the spectral dependence of the spin-noise power on the probe light wavelength (within the absorption band of the optical transition) depends dramatically on the relative magnitude of the homogeneous broadening: if the line is broadened homogeneously, then the optical spectrum of the spin-noise power shows a dip in the center of the line, while if the main contribution to the linewidth is made by the inhomogeneous broadening, then the optical spectrum of the spin-noise power does not reveal any dip. In the intermediate cases of comparable contributions of the homogeneous and inhomogeneous broadening, the depth of the dip can be used to estimate the ratio of these two contributions [22]. As was shown in [23], in more complicated cases of inhomogeneous broadening (in the Doppler-broadened systems), the spin-noise power spectrum can be "homogenized," acquiring a dip typical for homogeneously broadened lines.

The use of correlation properties of the noise signals produced in two-beam configurations allows one to realize experiments of the pump–probe type that are usually inapplicable to the effects of linear optics. Experiments of this kind employ, in essence, the difference of summing up the correlated and uncorrelated random signals. In particular, in [24], the use of such an approach allowed the authors to elegantly measure the homogeneous width of the optical transition in an ensemble of quantum dots with a huge inhomogeneous broadening.

It should be noted that despite the apparent simplicity of the spin-noise signal formation, a number of ambiguities still remain unresolved in this issue. In work [25], it has been shown that the magnetic resonance in the FR noise spectrum of a paramagnet can be considered as a result of heterodyning of the forward-scattered Raman component of the probe light on the photodetector. Such a mechanism of the noise signal formation implies possibility to enhance the signal at the expense of a more complete utilization of the scattered light or, at least, the possibility to detect the scattered component outside the angular aperture of the probe beam. Theoretical analysis [26, 27] has shown that the aperture-related properties of the inelastically scattered field can be highly informative. Reliable experimental data on this issue, however, has not yet been obtained.

ABOUT OBJECTS OF THE SPIN-NOISE SPECTROSCOPY

The SNS method is, in principle, suitable for almost all transparent paramagnets, since all of them consist of a finite number of elementary magnets and, to a certain extent, are capable of revealing the Faraday effect associated with their magnetization. However, due to a difference in the efficiency of conversion of magnetization to Faraday rotation in different paramagnets, in reality this is not the case.

As is known, the first objects of SNS were alkali-metal atoms [10, 28, 29], the high magnetooptical activity of which is revealed in the vicinity of strong narrow lines of the allowed optical transitions. The next objects of the spin-noise spectroscopic were the doped semiconductor structures, whose paramagnetism was related to elementary spin carriers (electrons, holes, excitons) and manifested itself most strongly near the band edge. As applied to these systems, the spin-noise spectroscopy method turned out to be extremely fruitful. At present, it can be asserted that the circle of objects of the SNS is practically restricted to these two classes of materials.

Meanwhile, as one can easily see, the most popular objects of the EPR spectroscopy—crystals and glasses doped with paramagnetic ions—the systems that laid the foundation for the modern laser technology (ruby, neodymium glass) remain "overboard" of the new method of spin resonance spectroscopy.

Qualitatively, the reasons for this situation are not difficult to understand. The highest magnetooptical activity is generally observed in media with a high steepness of the refractive index dispersion, i.e., in media with strong and narrow features of the optical spectrum. This conclusion is implicitly confirmed by the fact that the requirements imposed upon the wavelength of the probe light in SNS are usually rather stringent. In this sense, crystals and glasses with impurity ions of transition metals (which are implied first of all) were unlucky. The allowed interconfigurational transitions of these ions in crystals are, as a rule, spectrally broad, while the narrow intraconfigurational transitions (between the levels of unfilled electronic shells) are parity-forbidden and, therefore, weak. As a result, the contributions of the transitions of both types to the refractive index of the medium appear to be rather small.

The scale of the distinction between properties of the above objects with respect to these characteristics can be easily illustrated using the concept of the FR cross-section σ_F [30], introduced to evaluate the efficiency of conversion of the spin polarization to the FR. Using the results of [31] for the FR of ^{87}Rb in the vicinity of optical resonances, according to which the angle of rotation in a cell with the length $d = 2$ cm and density of polarized atoms $N \sim 10^{11}$ cm^{-3} is $\varphi \sim 0.1$ rad, we obtain for the FR cross-section the value $\sigma_F = \varphi/Nd \sim 0.5 \times 10^{-12}$ rad cm^2.

In the semiconductor system, namely the bulk n-GaAs sample, according to [30], the FR cross-section is approximately 10^{-15} rad cm^2.

To obtain typical value of the FR cross-section for a rare-earth ion in a crystal, we can use, e.g., the results of [32], according to which the Faraday rotation of a fluorite crystal (2.5 mm thick) containing 0.1% (0.6×10^{19} cm^{-3}) of divalent thulium (one of the most magneto-optically active ions) is ~0.1 rad. Taking into account that this value was obtained under conditions close to magnetic saturation of the sample, the FR cross-section for this system is ~10^{-19} rad cm^2, which is four orders of magnitude lower than the corresponding value for the n-GaAs crystal.

The above estimates give an idea of the scale of the problems to be solved to reliably detect spin noise in the crystals with impurity paramagnetic ions by the optical method.

CONCLUSIONS

The noise method of the magnetic resonance detection, originally implemented on atomic systems, has become, in the last decade, especially valuable as a tool for studying semiconductor structures. The method is used not only for investigating the magnetic resonance spectra and spin dynamics of the charge carriers. It also provides opportunities for studying relief of the carrier density, the nature of broadening of optical transitions, specific characteristics of the kinetic motion of spins, etc. Atoms of alkali metals continue to be important objects of the SNS, in particular, playing the role of ideal models for ensembles of moving spins. Further development of the noise spectroscopy, as we see it, will go both along the lines of expanding its abilities at the expense of deeper understanding of the laws of formation of the spin-noise signal in real systems, and along the lines of expanding the circle of objects of this new technique. The latter trend will necessarily require new experimental ideas.

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