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Evolution in the optical detection of magnetization

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Contents

Abstract. This paper represents an extended version of the talk given at the XVI Feofilov Symposium on the spectroscopy of crystals activated by rare-earth ions and devoted to the 100th anniversary of the birth of the outstanding physicist-spectroscopist and Corresponding Member of the USSR Academy of Sciences, Petr Petrovich Feofilov (Saint-Petersburg, November 9 – 13, 2015). In this review, we briefly elucidate the development of research on the optical detection of magnetization initiated by P P Feofilov and then carried on by his disciples and followers.

Keywords: magneto-optics, magnetic resonance, magnetic susceptibility, spin noise spectroscopy

1. Introduction

The effects of magnetic field on the optical properties of a medium may be considered from different viewpoints. The first one is based on the changes in the optical susceptibility tensor (or in its spectrum). Another one takes into account the magnetic field-induced anisotropy of the medium or the anisotropy of its luminescence characteristics. Yet another considers the violation of time-reversal symmetry. All these approaches are tightly connected with each other and are often indistinguishable. However, if we consider polarization magneto-optical effects in diluted paramagnets, then the easiest way is to start off with Zeeman structure of the optical transitions and with selection rules for different polarizations. At the early stages of development of polarization magneto-optics [Faraday rotation and magnetic circular dichroism (MCD) methods], researchers compared these techniques with the Zeeman effect spectroscopy and considered that the main advantage of the first ones is the possibility of revealing the hidden Zeeman structure of the absorption (or luminescence) spectra when it was not resolved because of large width of individual spectral components [1]. Papers written during those years analyzed the contributions to magneto-optical effects directly associated with paramagnetic magnetization, which define the possibility of applying optics for magnetic measurements.

Over the last fifty years, the capabilities and the scope of applications of polarization optics for magnetic measurements have significantly widened. These peculiar changes are partially due to the laser revolution in polarimetry and other advancements in measurement technologies, and partially due to the realization of ideologically new approaches to investigations of many-particle spin systems. Such an evolution of optical polarization methods for the detection of magnetization in transparent paramagnets (dielectrics and semiconductors) is the subject of the present paper.

This paper is an extended version of a talk which was given at the XVI Feofilov Symposium on the spectroscopy of crystals activated by rare-earth ions, which was dedicated to the 100th anniversary of the birth of Petr Petrovich Feofilov. For this reason, the main focus was on investigations which were started under the supervision of P P Feofilov at the S I Vavilov State Optical Institute. We followed the same line when choosing the material for the present historical sketches.

2. Magnetization in magneto-optical response spectroscopy

A particular feature of the influence of a magnetic field on the optical properties of a material is the fact that the optical susceptibility tensor of a magnetized medium acquires an antisymmetric part, which is known to be equivalent to an

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axial vector [2]. Therefore, in terms of symmetry, it is natural to expect the appearance of quantities with the symmetry of axial vectors in the material response in the applied magnetic field (for example, angular momentum or a gyration vector). A purely optical result of the magnetic field's influence on the medium (or the material magnetization by any other mechanism) is the manifestation of circular anisotropy effects — the Faraday rotation and magnetic circular dichroism. Based on these simple considerations, one can reach the conclusion that the circular anisotropy effects (directly linked to the gyration vector of the medium) can carry information about both the strength of the magnetic field applied to the material and the value of the medium magnetization.

Furthermore, we will mainly consider below linear in field (or in magnetization) effects of circular anisotropy, arising when the probe light direction is parallel to the magnetization, which under static conditions corresponds to the collinearity of an external magnetic field and the observation direction (Faraday geometry). However, when measuring the dynamics of processes or investigating nonequilibrium systems, the system magnetization vector direction does not match the direction of the external field vector (in the case of spin precession, for example), and the measurements can be performed in the transversal field orientation with respect to the light propagation direction (the so-called Voigt geometry). In this case, the magnetization component parallel to the light propagation direction starts to oscillate, thus resulting in the oscillation of the magneto-optical activity value.

A great deal of information about spin-system properties can be obtained from *spectroscopic* investigations of magneto-optical activity, which were of great importance in early work on magneto-optics [1].

The simplest scheme for highlighting the explanation of magneto-optical activity effects (Faraday rotation of the polarization plane and magnetic circular dichroism) is illustrated in Fig. 1 for the case of optical transition between two doubly degenerate states. According to the known selection rules, the absorption line of such a transition splits in the longitudinal magnetic field into two circularly polarized components with opposite rotation directions. Similar splitting is also experienced by the corresponding refractive index dispersion curves for circular polarizations.

It is common to distinguish three contributions to magneto-optical activity [3]. The first one (the so-called A-contribution or the diamagnetic contribution) corresponds to the frequency shift of spectral components in the magnetic field. Its spectral behavior is shown in Fig. 1a. The second one, the B-contribution (also known as polarization or Van Vleck contribution) is associated with the magnetic perturbation of the system's wave functions. Finally, the C-contribution is caused by a variation of the Boltzmann distribution of particles over the ground state magnetic sublevels. Spectral dependences of B- and C-contributions, which are often called paramagnets, are demonstrated in Fig. 1b. Their sum is proportional to the magnetization of the spin system [4], which is the basis of the optical method for magnetization detection.

In thermodynamically equilibrium systems, the B- and C-contributions behave similar to the conventional paramagnetic susceptibility and, unlike the diamagnetic contribution, they display temperature dependence. Moreover, paramagnetic magneto-optical effects are characterized by a less steep spectral dependence, which allows observing their contributions to the Faraday effect in the transparency region far from optical resonances.

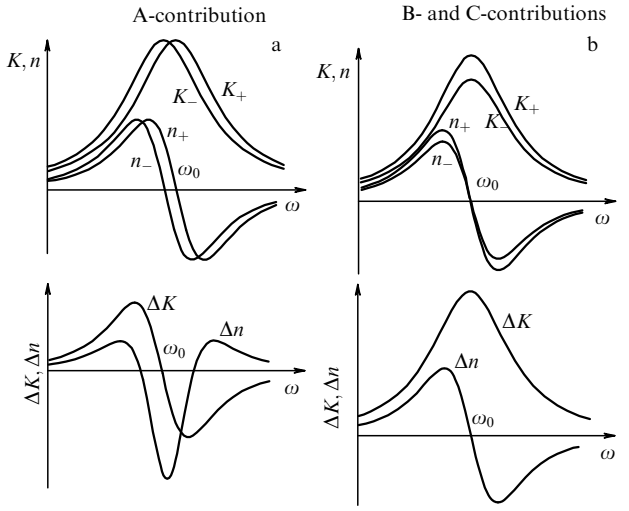

Figure 1. Schematic illustration of the formation of different contributions to the spectra of magneto-optical activity in the optical transition region. Magnetic circular dichroism and Faraday rotation spectra are defined by the difference between the absorption coefficients ($\Delta K = K_+ - K_-$) and refractive indexes ($\Delta n = n_+ - n_-$) for two circular polarizations.

The above-given simplified illustration of contributions to magneto-optical activity can in most of the cases qualitatively describe the main features of paramagnetic and diamagnetic contributions. One should keep in mind that the absolute value of the paramagnetic contribution, which is directly controlled by the system's magnetization, depends on many parameters that are not related to magnetization, such as the energy structure of the electronic states in the paramagnetic material, oscillator strength of an optical transition, spin–orbit interaction value, and light wavelength. Therefore, the optical methods for the detection of magnetization are usually used for relative magnetization measurements or to detect their variations (either induced by an external perturbation or spontaneous). Such measurements, however, appear to be very informative in many cases.

In the 1960s, the majority of experiments on the optical detection of magnetization in magneto-diluted systems have been performed on dielectric crystals activated by paramagnetic ions (rare-earth and iron group ions), which could be utilized as perfect model objects for the solution to many problems in the field of magnetic resonance and relaxation phenomena. Among all such objects, great magneto-optical activity was particularly observed in crystals activated by some divalent rare-earth ions, in which allowed interband transitions (4f–5d) are located close to (or within) the visible range. However, spectral magneto-optical changes in the vicinity of forbidden intraband transitions of rare-earth ions (4f–4f) also allowed separating different contributions to the magneto-optical activity and identifying the ongoing magnetic processes.

A demonstrative example for the realization of such an opportunity is given in Fig. 2. The measurements were performed using a rather unique system — a fluorite crystal with Dy^{3+} trigonal centers (trigonal symmetry of the centers appears due to the local compensation of the activator excess charge by an oxygen ion). The uniqueness of the system was marked by incredibly long times (up to several hours) of spin–lattice relaxation of the centers in the ground state at liquid-

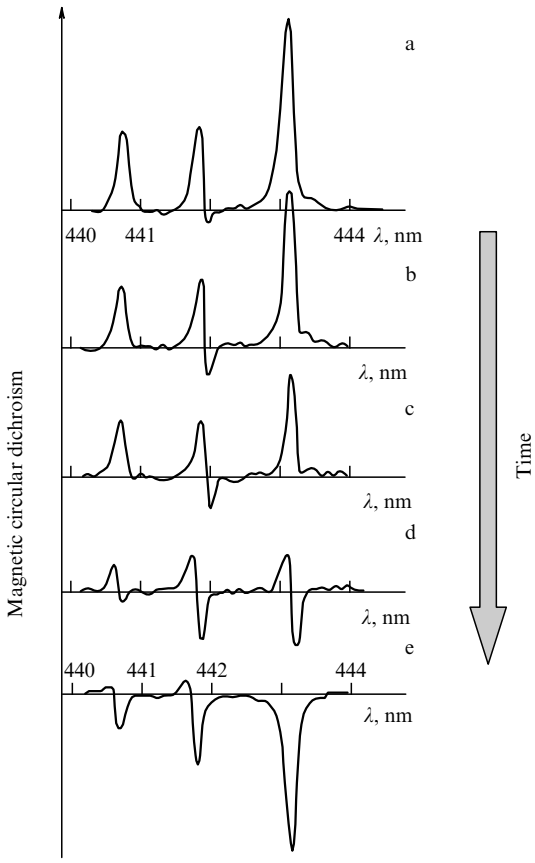

Figure 2. Relaxation dynamics of the magnetic circular dichroism spectrum measured for an ensemble of trigonal centers in $CaF_2 - Dy^{3+}$ crystal, reflecting the relaxation dynamics of the spin-system magnetization.

helium temperature. Moreover, the largest possible g-factor anisotropy of the ground-state doublet ($g_\perp = 0$) allowed achieving doublet population inversion by just switching the sign of the external magnetic field. Figure 2 illustrates the dynamics of the circular magnetic dichroism spectrum of the crystal (in the region with intraband transitions). The sample was preliminarily magnetized in a strong magnetic field and after that the field sign was rapidly changed. It is seen from the figure that during the magnetization relaxation the initially inverted spin system changes the sign of magnetization, when approaching the equilibrium state. Correspondingly, the paramagnetic contribution to MCD also changes its sign and becomes purely diamagnetic, when the system magnetization becomes zero.

Since in any case the optical method considered is based on recording the small (induced by magnetization variation) variations in the medium gyrotropy, it was mainly applied to isotropic materials and cubic crystals, which had no intrinsic birefringence. The isotropy of cubic crystal optical properties is known to be limited by the linearity of their polarizability. This also holds true for the magnetic and magneto-optical properties of a cubic crystal. At low temperatures and (or) in strong magnetic fields, the field dependence of the medium magnetization (and, correspondingly, the dependence of the paramagnetic contribution to the magneto-optical activity) stops being linear and the isotropy of cubic crystal properties can be violated. In paper [5], in particular, it was shown that the magnetic and magneto-optical properties of cubic crystals with anisotropic impurity centers become anisotropic near

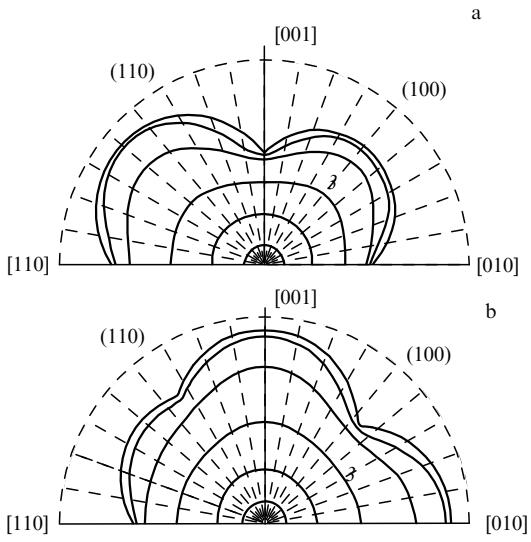

Figure 3. Directional magnetization dependences of a cubic crystal with tetragonal (a) and trigonal (b) centers, having the largest possible anisotropy of the ground-state g-factor ($g_\perp/g_\parallel = 0$). As the ratio between the magnetic splitting ΔE and the thermal energy $k_B T$ increases (solid curves), the directional magnetization dependence loses its initial isotropy. Curves indicated by the number *3* correspond to the equality condition $\Delta E = 2k_B T$.

the saturation point, and the character of this anisotropy gives information about the symmetry and the degree of anisotropy of impurity centers. This fact is illustrated in Fig. 3, which displays the directional dependences of the paramagnetic contribution to the magneto-optical activity of a cubic crystal (or the directional dependences of the crystal magnetization) for different degrees of magnetic saturation and for the cases of tetragonal and trigonal centers with the largest possible g-factor anisotropy.

The described effect can be considered as one of the methods of revealing the hidden anisotropy of cubic crystals with anisotropic centers. Such a task was first performed by P P Feofilov by exploiting the phenomenon of polarized luminescence, which cannot refer to linear response effects either [6] (i.e., it is impossible to describe the linear connection between the exciting field and the luminescence field).

3. Laser revolution in polarimetry

In the pre-laser era, magneto-optical investigations of diluted paramagnets, as was mentioned in Section 2, consisted of spectroscopic measurements, because the measurable signals could be detected only close to (or directly in) optical transitions, and the information extracted from the spectra regarded mainly the positions and intensities of Zeeman spectral components. The situation changed radically after the advent of laser polarimetry with an extremely high sensitivity (restricted only by the photocurrent shot noise).

A laser polarimeter of this type was first described in papers [7, 8]. In order to reach the sensitivity restricted by the shot noise, it was necessary to suppress excessive noise of the laser source, which was achieved by applying a polarization scheme with balanced detection of the signal. In such a two-channel scheme (Fig. 4), the variations of photocurrents in two photodetectors associated with the output light polarization change are summed, while the variations caused by the source intensity changes (noise) are subtracted. This allowed

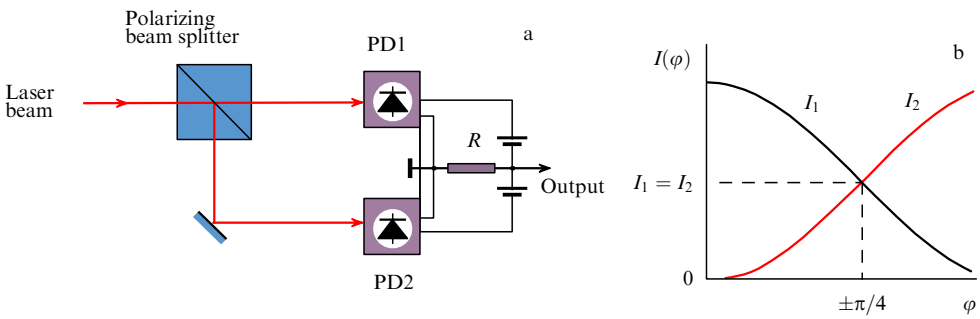

Figure 4. (a) A schematic of a balanced detector used in modern laser polarimeters in order to suppress the excessive light noise from the source. (b) Photocurrents of the detectors PD1 and PD2 versus the azimuthal angle of the polarization plane of the light incident on the polarizing beam splitter. At the point $\pm\pi/4$, two photocurrents fully compensate each other in the output load R.

suppressing excessive light noise by several orders of magnitude. The absolute sensitivity was also maximized by using light beams with maximum intensity. Later on, a number of techniques were suggested which allowed improving the restricting level of the polarimetry sensitivity by significantly increasing the beam power, while maintaining the low level of light load on the photodetector [9, 10]. As a result, the polarimetric measurement sensitivity was increased by more than three orders of magnitude with respect to the sensitivities achieved in the 1960s. This led to a radical change in our conception regarding the capabilities of methods for magnetization detection.

The sensitivity scale of the laser-polarimetry method for magnetic measurements in the case of, for example, trivalent rare-earth ions in crystals is illustrated in Fig. 5. The figure shows field dependencies of Faraday rotation in BaF_2 crystals with a 0.3 mol. % concentration of impurity ions measured at the same wavelength ($\lambda = 632.8$ nm) [11]. One can see from the figure that the value of the paramagnetic contribution (the one that saturates as the field increases) to the Faraday effect in these specimens (1 mm in thickness) is on the order of several degrees. If we take into account the fact that the sensitivity of laser polarimeters with laser powers of about 1 mW are approximately 10^{-6} degrees, then it is easy to conclude that even for a large probe light wavelength detuning from the optical resonance the laser-polarimetry methods allows detecting the smallest variations of the medium magnetization. As was discovered in recent years, one can detect not only regular changes in the paramagnetic magnetization induced by external perturbations but also its spontaneous fluctuations in the thermodynamic equilibrium state.

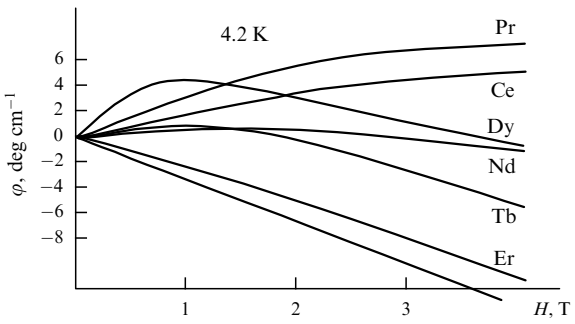

Figure 5. Field dependences of the Faraday rotation in BaF_2 crystals activated by trivalent rare-earth ions (0.3 mol.%) at the wavelength of 632.8 nm. The lowest (linear) field dependence of rotation corresponds to the diamagnetic rotation of an undoped crystal lattice. As the magnetic field increases, the contribution to the Faraday effect, for which the impurity ions are responsible, demonstrates saturation that is characteristic of the paramagnetic contribution.

4. Optical detection of regular magnetization signals

The effects of a regular magnetization response on external perturbation, which are usually the subject of any standard magneto-optical experiment, can be separated into two general types: effects caused by resonant excitation of the spin system resulting in its precession (optical registration of the magnetic resonance) and effects of response to low-frequency (nonresonant) excitation. After realization of ultimate polarimetric sensitivity, the effects of both types acquired a new practical meaning.

4.1 Optical registration of electron paramagnetic resonance

The simplest method for optical registration of electron paramagnetic resonance (EPR) in the ground state of the impurity ion is based on the Faraday effect variation under the saturation of the transition between magnetic sublevels of this state in a high-frequency magnetic field. Usually, the power of a microwave or radio frequency (RF) field acting on a specimen is modulated with the acoustic frequency and the corresponding magneto-optical signal is registered by scanning the applied magnetic field (see more details in Ref. [11]). It was shown that in such experiments the magnetic resonance is easily detected optically at a fixed probe laser beam wavelength in the visible spectrum in all trivalent rare-earth ions, despite the absence for them of allowed optical transitions in the visible range [11, 12].

More convenient objects for the optical detection of EPR among rare-earth activators are divalent ions, the most popular of which is the thulium ion. EPR spectra of this ion were easily optically detected at the He–Ne laser wavelength in low magnetic fields at frequencies down to the magnetic resonance of the order or even less than the EPR line width (Fig. 6). Series of investigations were also performed for MeF_2-Tm^{2+} crystals, regarding the spin-system relaxation dynamics [12].

The above conclusion about the relatively low sensitivity of the optical method for magnetization detection to the probe light wavelength (or, at a fixed wavelength, to the paramagnetic impurity type) was confirmed by EPR spectroscopy research on trivalent rare-earth ions, which are 'transparent' in the visible range [11]: the EPR spectra of all rare-earth ions were registered with a high signal-to-noise ratio using an He–Ne laser as a radiation source.

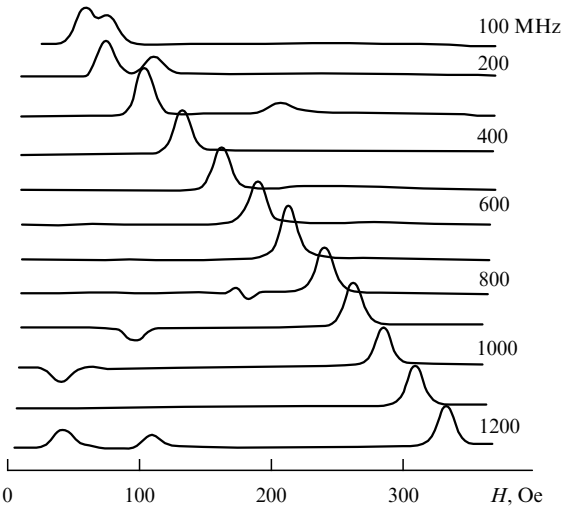

Figure 6. EPR spectra of a fluorite crystal with divalent thulium recorded using a laser EPR spectrometer. Temperature is 2 K.

Later on, the laser-polarimetry method was applied to measuring the EPR of rare-earth ions in glass [13]. The resonant signal was observed using the method described above in all rare-earth-activated glass specimens; however, it turned out later that the observed signal was caused by the heating of the spin system through a two-level system bath [14], and its connection with the spin-system magnetic resonance spectrum was indirect. The high sensitivity of the method and high efficiency of spin-system excitation by the microwave field in these experiments allowed the authors to develop a method for the detection of small paramagnetic impurity concentrations in glass [15].

The application of the laser-polarimetry technique to EPR detection turned the optical EPR registration method from an exotic one into a quite common technique, which can also significantly broaden the capabilities of traditional EPR spectroscopy. The most important advantage of this method is a full separation of resonance excitation and detection channels, which allows performing measurements under a high resonant pump. This is important for dynamic measurements, for studies of forbidden resonance lines, and in some other cases. Among the advantages of the optical technique for EPR detection, high spatial resolution and the possibility of using a spectral degree of freedom for the identification of impurity centers can also be mentioned.

4.2 Modulation magneto-optical spectroscopy

The high sensitivity of the laser-polarimetry method to spin-system magnetization variations has significantly broadened the scope of precise measurements of frequency dependences (spectra) of spin-system magnetic susceptibility. It is quite obvious that the spectrum of the paramagnetic response to a longitudinal magnetic field variation carries information about the time of the longitudinal relaxation of magnetization (time T_1). Less obvious and more interesting is the Van Vleck contribution to magneto-optical activity, especially when the energy gap between states entangled by an external magnetic field becomes significantly smaller than the thermal energy $k_B T$, where k_B is the Boltzmann constant. Such a situation often takes place when the spin system experiences weak 'nondiagonal' perturbation. As the simplest example, we can consider a two-level spin system in a magnetic field with a Hamiltonian

$$H = \frac{1}{2}\begin{pmatrix} g\beta B & 0 \\ 0 & -g\beta B \end{pmatrix} + \frac{1}{2}\begin{pmatrix} 0 & v \\ v & 0 \end{pmatrix}, \tag{1}$$

where the first term describes the 'Zeeman' contribution depending on the magnetic field B, while the second one corresponds to an arbitrary 'nondiagonal' perturbation. Here, g stands for the g-factor, β is the Bohr magneton, and v is the value of nondiagonal perturbation. In this case, the behavior of system's energy levels in the magnetic field is described by a simple expression:

$$E_{\pm} = \pm\frac{1}{2}\sqrt{(g\beta B)^2 + v^2}\,. \tag{2}$$

It is important that for the conditions given above ($g\beta B, v \ll k_B T$) the dependence of magnetization M (and of the corresponding contribution to the magneto-optical activity) on the field is not sensitive to the perturbation v — it remains linear even if perturbation takes place:

$$\begin{aligned} M &= -\frac{\partial\langle H\rangle}{\partial B} = -\frac{\partial}{\partial B} Z^{-1}\,\mathrm{Sp}\,H \exp\left(-\frac{H}{k_B T}\right) \\ &\approx \frac{1}{k_B T\,\mathrm{Sp}1}\frac{\partial}{\partial B}\,\mathrm{Sp}\,H^2 = \frac{(g\beta)^2 B}{2k_B T}\,, \\ Z &\equiv \mathrm{Sp}\,\exp\left(-\frac{H}{k_B T}\right). \end{aligned} \tag{3}$$

If we consider a 'nondiagonal' perturbation v to be a transverse (with respect to the field B) magnetic field, it becomes clear that such a result follows directly from the validity of the superposition principle in the range of the linear relation between magnetization and field (i.e., in the range of low Zeeman energies with respect to $k_B T$). At the same time, it is obvious that the magnetization processes in a paramagnetic material in weak ($g\beta B \ll v$) and strong ($g\beta B \gg v$) magnetic fields are completely different. From the experimental point of view, this difference can easily be observed in the spin-system response dynamics. On the one hand, the paramagnetic contribution, related to changes in magnetic sublevel populations, is characterized by a specific relaxation time and, correspondingly, by some delay in the response. On the other hand, the Van Vleck contribution is connected with the mixing of the wave functions of states by the magnetic field and is inertialess. This fact is employed to separate the Van Vleck contribution to the magnetization and magneto-optical activity, and to detect the nondiagonal perturbations mentioned before.

Figure 7 plots an example with the energy level diagram of the Tm^{2+} ion ground state in a CaF_2 crystal together with the field dependence of the Faraday response of a crystal with low activator concentration to an applied AC magnetic field [16]. The frequency (above 100 Hz) of this alternating component in this case significantly exceeds the rate of ion ground-state longitudinal relaxation and, therefore, it cannot be considered high (an exception is the field range near 230 Oe, where the relaxation time decreases due to cross-relaxation processes). Accordingly, the system response to such a field reveals only the inertialess (i.e., Van Vleck) part of the magneto-optical activity. In this case, the role of 'nondiagonal' perturbation, which mixes the magnetic structure states in the ground state, is played by the hyperfine interaction. The narrow peak of this dependence for a zero field is associated

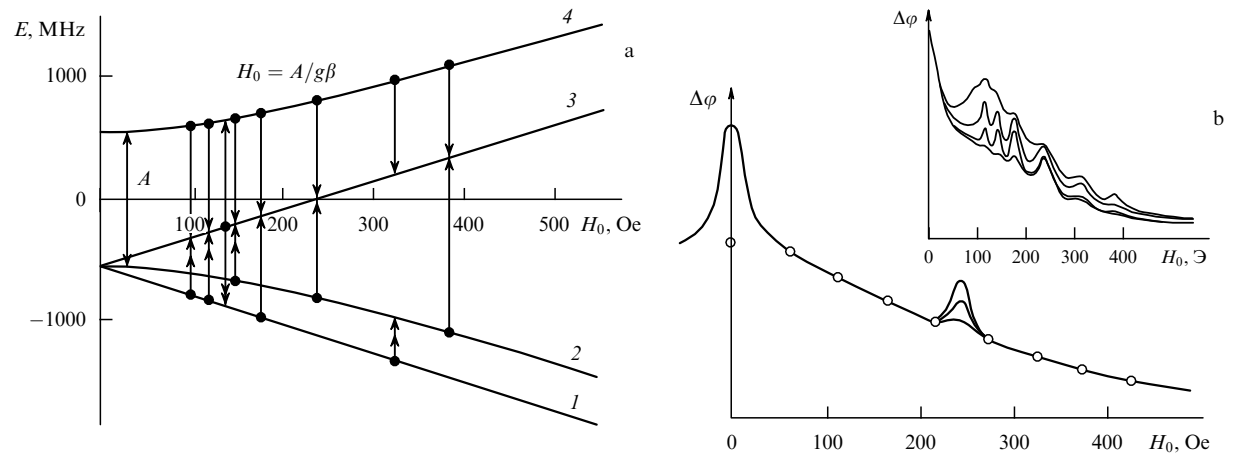

Figure 7. (a) Diagram for ground-state energy levels of a Tm^{2+} ion with indications of possible many-particle cross-relaxation processes. (b) Modulation spectrum of a CaF_2-Tm^{2+} crystal with activator concentration of 0.007 mol.%. The inset to figure b shows modulation spectra of a crystal with higher activator concentration (0.05 mol.%). Different curves correspond to different modulation frequencies (from 30 Hz to 2 kHz) [16].

with a superhyperfine interaction, which conclusively removes the degeneracy of magnetic sublevels in a zero magnetic field.

As the thulium ion concentration in the crystal is increased (see inset to Fig. 7b), the magnetization relaxation time for some field values can decrease significantly due to cross-relaxation processes, and the crystal response demonstrates a large paramagnetic contribution. Figure 7a schematically shows possible cross-relaxation transitions in the thulium ions ensemble, which manifest themselves in the given modulation spectrum.

In later studies, the Van Vleck contribution to magneto-optical activity was utilized to solve a number of problems aimed at the investigation of the magnetic anisotropy character of impurity centers in crystals and glass. In particular, the method of a so-called artificial Van Vleck susceptibility was developed. In this method, the role of 'nondiagonal' perturbation was played by the magnetic field directed perpendicularly to the main one, which particularly allowed very distinctively visualizing the hidden anisotropy of a cubic crystal with trivalent dysprosium impurity centers and performing measurements of extremely small components of the ion ground-state g-factor [17]. Figure 8 demonstrates the experimental dependence of the CaF_2-Dy^{3+} crystal magneto-optical susceptibility on the strength of the applied longitudinal field for different values of the transverse field. The spectrum demonstrates only Van Vleck (inertialess) specimen susceptibility, which has a large value only in the field range in which the net magnetic field acting on this specific center group turns out to be oriented transversely to their symmetry axis.

Papers [18–21] reported the possibility of applying the Van Vleck (including artificial) susceptibility method to the analysis of more complicated systems, in particular, to discover the anisotropy character of impurity centers in amorphous media.

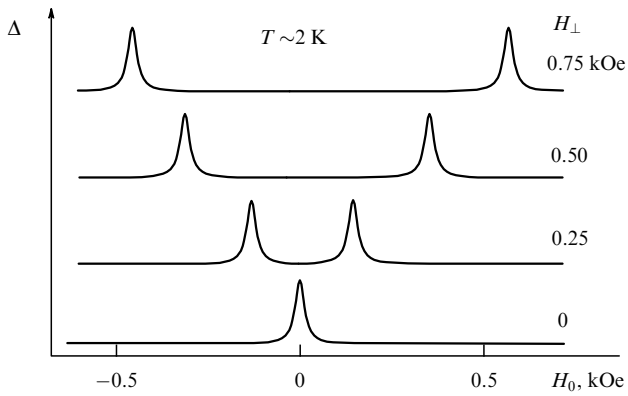

Figure 8. Spectra of artificial Van Vleck susceptibility in CaF_2 crystal with Dy^{3+} trigonal centers. Scanning of longitudinal magnetic field in the presence of a transverse one corresponds to the rotation of the net field vector. At the moment, when the net field intersects the plane perpendicular to the trigonal center axis, the magnetic splitting changes its sign and does not become zero only due to nonzero $g_\perp$. As a result, the crossing of levels in zero net field is replaced by anticrossing, which gives rise to a inertialess Van Vleck contribution to the modulation spectrum. This is registered as a narrow peak with its width carrying the information about the $g_\perp$ value. Here, $H_0 \| \langle 110 \rangle$, and $H_\perp \| \langle 100 \rangle$.

5. Optical detection of magnetization noises

5.1 Prehistory

Soon after the advent of laser polarimetry with ultimate (shot-noise limited) measurement sensitivity, it became possible to detect the magnetization spontaneous noise in an equilibrium ensemble of paramagnetic atoms. The fundamental possibility of such measurements was supported by the fluctuation–dissipation theorem and did not give rise to any doubts. The only problem was to experimentally detect these fluctuations, which should obviously be very small for macroscopic specimens.

A demonstrational experiment on magnetic resonance registration in Faraday rotation noise [22] was performed in sodium vapor placed in a transverse (with respect to the light beam direction) magnetic field with a magnitude close to Earth's field (Fig. 9). The balanced detector load was a resonant circuit tuned to the frequency of ≈ 1.3 MHz. The dye laser wavelength corresponded to the range in the vicinity of one of sodium D-lines, in which the paramagnetic contribution to the Faraday effect (as well as the measurement sensitivity) sharply increases. The precession of the fluctuation magnetization component around the vector of

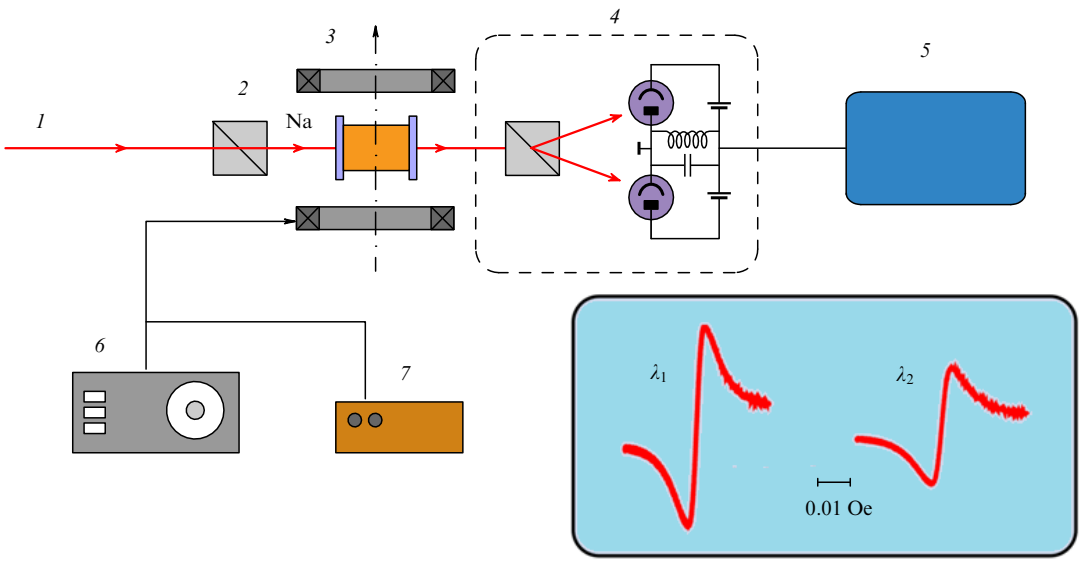

Figure 9. Schematic of the first experiment for the observation of magnetic resonance in the Faraday rotation noise spectrum: *1* — light beam of a dye laser, *2* — Glan prism, *3* — cell with sodium vapor in a magnetic field, *4* — balanced detector with a resonant load, *5* — signal detection and processing unit, *6* — acoustic generator, and *7* — sawtooth voltage generator. The inset show signals obtained for two wavelengths of the probe radiations (see details in Ref. [22]).

the applied magnetic field led to oscillations of the azimuthal angle of the polarization plane of a laser beam after passing the medium. The maximal output signal of the balanced detector was observed when the Larmor precession frequency (magnetic resonance frequency) coincided with the eigenfrequency of the circuit in the detector load. Signal registration was performed using a standard (for that time) technique of lock-in amplification under magnetic field modulation. The signal-to-noise ratio in these experiments was approximately 10^2 for an accumulation time of 1 s.

The proposed method, unlike all known methods for magnetic resonance detection based on detection of regular response signals, had a number of obvious advantages which could compensate for its expected low sensitivity. The first advantage was the nonperturbative character of the measuring procedure: a probe light at a wavelength in the paramagnetic transparency region did not induce any real transitions in the medium. The second advantage was the possibility, unusual for standard EPR spectroscopy, of detecting resonance in the absence of medium's magnetic polarization, i.e., in weak magnetic fields at high temperatures. Later on, a number of interesting features of this method were discovered (see Sections 5.2–5.5).

5.2 Semiconductor structures as the main object of spin-noise spectroscopy

Estimates have shown that in order to observe a magnetic resonance signal in Faraday rotation noise of the most popular EPR spectroscopy objects — crystals with paramagnetic ion impurities — a sensitivity higher than that of existing laser polarimeters was needed. Therefore, we considered the result obtained more an academic illustration of the fluctuation–dissipation theorem than a basis for a new EPR spectroscopy method.

The noise method of magnetic resonance detection was first applied to solid-state systems only in 2005 [23]. The resonance peak in the Faraday rotation noise spectrum of a bulk doped n-GaAs crystal placed in a transverse magnetic field was observed at a frequency which corresponded to the conduction electron *g*-factor. The signal acquisition time regarding the signal-to-noise ratio on the order of unity in these experiments took several hours, so it was still a question whether this new method could be a new tool for experimental investigations or not.

The next important step in the evolution of noise spectroscopy, known now as spin-noise spectroscopy or spontaneous magnetic resonance spectroscopy, was the application of spectrum analyzers with a fast Fourier transform instead of scanning type spectrum analyzers. In Fourier spectrum analyzers, the broadband output signal of a balanced detector was digitized with a sampling frequency of about several GHz and transformed applying the fast Fourier transform. The resulting noise spectrum was accumulated in real time. This method of signal recording allowed increasing the measurement sensitivity by more than two orders of magnitude and the spin noise spectroscopy indeed became a new efficient tool for the study of spin systems in semiconductor structures, for which the Faraday rotation cross section [24] turned out to be several orders of magnitude larger than that of, for example, rare-earth ions in crystals. Figure 10 depicts an example of measured spin noise spectra obtained in bulk samples of n-type doped gallium arsenide [10].

5.3 Beam shape effects

The spin noise method has become quite popular over the last few years (see reviews [25–28]) and has demonstrated a number of peculiar features which were not initially predicted. These features are mainly associated with those of the optical information channel in spin noise spectroscopy. We will briefly review below some of these features.

Let us first bring attention to the fact that the value of the signal in Faraday rotation noise spectroscopy (unlike the magnitude of regular signals in Faraday rotation spectroscopy) depends on the beam cross section. As the cross section area decreases, the number of spins contributing to the noise signal decreases as well and, correspondingly, the relative noise magnitude increases. Formally, one can say that the

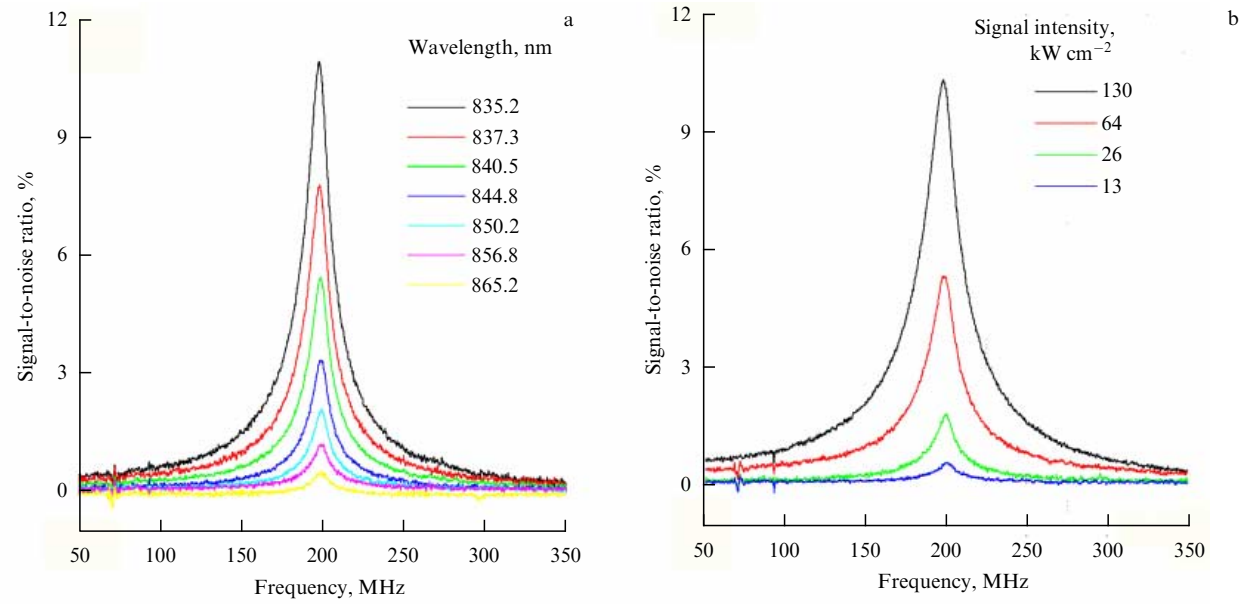

Figure 10. Spin noise spectra of a bulk specimen of n-type doped gallium arsenide (impurity concentration $C \sim 3.7 \times 10^{16}$ cm^{-3}) with $d = 170$ μm thickness for different (a) wavelengths and (b) intensities of the probe light.

spin noise signal magnitude depends on the light beam power density, which is characteristic of nonlinear, but not linear, optics.

Let us recall that there is one simple experimental technique which allows us to give a qualitative answer to the question: does the medium demonstrate nonlinearity for a given power density of an optical field? This method is known as Z-scan and its scheme consists in the following. A specimen under investigation (it needs to be transparent) is transferred through the caustic with a waist of a tightly focused laser beam (Fig. 11). A photodetector is placed behind the specimen and it registers the transmitted light intensity for one angular aperture or another. If the medium is linear, the light intensity will not depend on the specimen position. However, if the medium is nonlinear and its optical properties depend on the power density in the beam, the registered dependence will demonstrate a peak or a dip corresponding to the specimen position in the center of the waist (see Fig. 11). If in such a test experiment we register not the transmitted light intensity, but the spin noise power (Faraday rotation noise power), then it will result in a positive outcome, despite the fact that the spin noise spectroscopy does not imply any optical nonlinearity in the medium.

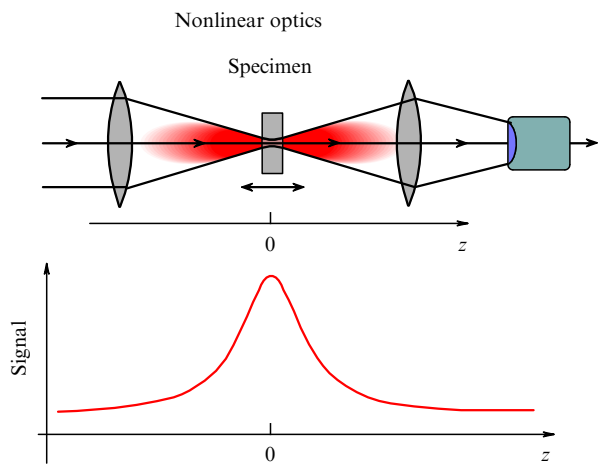

Figure 11. Illustration of the Z-scan method.

This feature of noise spectroscopy was, in particular, applied to the realization of three-dimensional tomography of semiconductor structures [29]. When probing bulk samples with a tightly focused laser beam, the main contribution to the noise signal is made by the focal region, which can be quite small. Therefore, by scanning this region along the sample, we can probe its properties and spatially resolve the morphology of its impurities. In paper [29], the capabilities of such tomography were demonstrated through the example of a model structure consisting of two semiconductor films with different doping levels, and in paper [30] spin-noise tomography was effectively applied to obtain interface morphology of the charge carrier's g-factor in a thin-film semiconductor structure. Spatial resolution of this method along the light propagation direction was defined by the Rayleigh beam waist length and was ≈ 20 μm.

An example of pump–probe spectroscopy (called by the authors of Ref. [30] two-color spin-noise spectroscopy) using spin noise as a response of the system was demonstrated in Ref. [31], where the spin noise signal of a quantum dot array was registered using two monochromatic laser beams with slightly different frequencies. Due to the large value of inhomogeneous broadening in the quantum dot array, the two beams probed different quantum dot subarrays, until their wavelengths were as close as the homogeneous line width of the resonance. At this moment, the noise contributions from both beams became correlated and the resulting noise power increased. This effect was used to measure the homogeneous line width of a single quantum dot, resulting in a value of 6.6 μeV.

5.4 Optical spectroscopy of spin noise

Another interesting property of magnetization noise spectroscopy appears when it is applied to the study of *optical* spectra of noise power, i.e., variations of noise power with a change in

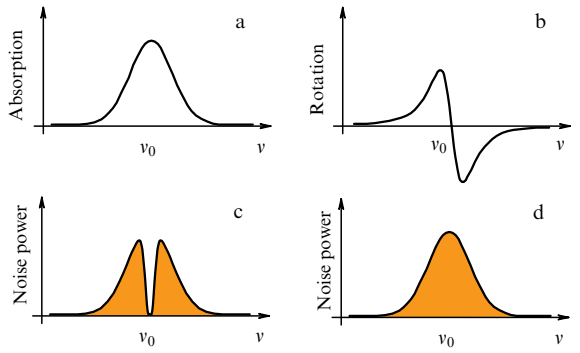

Figure 12. Illustration of the (a) absorption and (b) Faraday rotation spectra in the optical transition region. Spectra of Faraday rotation noise power for (c) homogeneous and (d) inhomogeneous broadening (in the latter case, the inhomogeneous broadening is assumed to be significantly larger than the homogeneous one).

the probe light wavelength [32]. Here one should keep in mind that the noise of the polarization plane rotation at a specific probe light wavelength is formed by the contributions from different optical spectrum components, which can be either correlated or noncorrelated. Depending on this property, the spin noise optical spectra will look different. Naturally, in conventional linear response optics, this fact does not have any influence (contributions of different spectral components are always correlated), and the corresponding information cannot be obtained.

The above-mentioned fact is revealed in especially spectacular way in optical spectra of spin noise power in the region of bands with homogeneous or inhomogeneous broadening. In the first case, the contributions from all spectral components of the band are, obviously, correlated, and the optical spectrum of spin noise power acts just like a spectrum squared of steady Faraday rotation. In the second case, the homogeneously broadened components of an optical spectrum fluctuate independently and sum up statistically at any wavelength.

Figure 12 schematically illustrates this difference with a very characteristic feature: in the center of the band, where steady Faraday rotation is zero, the Faraday rotation noises become either zero or maximal. The depth of a dip changes together with the ratio between the homogeneous and inhomogeneous line widths, so this characteristic can be used to estimate the homogeneous line width hidden in an inhomogeneously broadened spectrum.

These considerations were experimentally confirmed by measuring the optical noise spectrum in the region of a homogeneously broadened line in atomic potassium and by measuring the optical noise spectrum in the region of an inhomogeneously broadened line of a quantum dot ensemble [32]. It should be noted that information about the size of inhomogeneous spectrum broadening can usually be obtained by means of nonlinear optics (electromagnetically induced transparency, four-wave mixing, etc.), while linear susceptibility optics is not suitable for such applications.

5.5 Nonlinear spin-noise spectroscopy

As mentioned in Section 5.1, one of the main features of spin noise spectroscopy which makes it different from all known methods for magnetic resonance and magnetic susceptibility detection is the nonperturbation character of measurements. This statement, of course, should not be idealized: noise modulation of probe light polarization is a result of energy and angular momentum exchange between light and the medium during Raman scattering of the probe light [33]. Such a perturbation at low optical powers does not influence in any way the measured properties of the spin system.

The specifics of spin noise spectroscopy, however, lie in the fact that the noises of the studied system are detected using a photon flux, which is inevitably itself fluctuating, making the 'signal' noise separation difficult. This fundamental limitation of measurement sensitivity can be commonly overcome by increasing the probe beam power density. In order to increase the beam power, while maintaining the moderate illumination regime for the detector, two approaches are involved. The first one is based on the effect of Faraday rotation accumulation in a medium placed inside Fabry–Perot resonator (microcavity) [34–37]. The second approach uses the method of high polarization extinction, in which the low level of optical power on the photodetector is obtained by means of orthogonal polarizers [9, 10, 38]. In both cases, the increase in the measurement sensitivity is achieved by increasing the probe light power on the sample and, correspondingly, by lowering the relative value of quantum (shot) noise in the probe light flux.

Experimental investigations of spin noise spectra under the condition of high optical field density have, on the one hand, demonstrated that the nonperturbation concept of the method is very limited and, on the other hand, shown the new potential of this method, which showed up under the conditions of significant optical excitation of the spin system. An example of this type of investigations can be found in papers [39, 40], where elliptically polarized probe light acted in the transparency region of a semiconductor crystal (n-GaAs) located inside a microcavity. The probe light created an effective magnetic field in the sample, which significantly changed the spin noise spectrum of the system. Moreover, as was shown in these experiments, the high-intensity elliptically (or circularly) polarized light optically oriented the nuclear spin system, which also influenced the measured electron spin noise spectrum. As the optical pump was turned off (by replacing circular with linear polarization and decreasing the light intensity), the spin noise spectrum demonstrated slow relaxation towards the equilibrium state, connected with the nuclear spin system relaxation and, correspondingly, with the relaxation of the field acting on the electron subsystem (Overhauser field). The manifestation of nuclear relaxation in the spin noise spectrum of an n-GaAs specimen inside a microcavity is illustrated in Fig. 13.

6. Conclusion

The present review of studies regarding magnetization detection, which is one of most important fields of modern magneto-optics, demonstrates significant progress in this area of experimental physics over the last fifty years. It is enough to note that in the 1960s the magnet was quite seldom used in an optical laboratory. Now, the magnet is an experimental equipment component of the same importance as, for example, a low-temperature cryostat or laser. At the same time, a significant part of magneto-optical investigations is directly or indirectly connected with the measurement or variations of magnetization. Also, magneto-optical measurements, due to the specific symmetry of magnetic excitations, almost always include polarization measurements. This

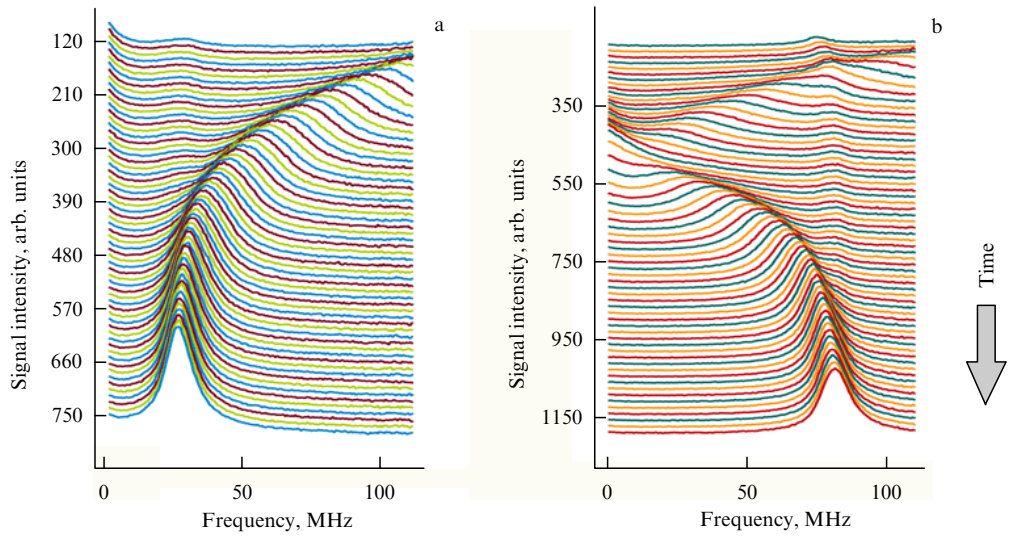

Figure 13. Manifestation of nuclear spin system dynamics in the electron spin noise spectrum of bulk n-GaAs in a microcavity. Initial spin orientation of the nuclear system was prepared by specimen pumping with circularly polarized light in the crystal transparency band. Signal intensity for (a) parallel and (b) antiparallel directions of the nuclear polarization with respect to the applied field. Time of one curve acquisition is ≈ 1 s. (See details in Refs [39, 40].)

is the reason why the evolution of optical methods for magnetic measurements so greatly depended on success in the field of high-sensitivity polarimetry achieved over the last decades. This success resulted in the development of spin noise spectroscopy. Currently, optical methods for magnetic measurements are finding a significantly large number of applications in semiconductor structure physics for the diagnostics of the dynamics and energy structure of electron and nuclear spin systems. These methods now play an important role in the modern optics of paramagnetic systems.

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References

1. Zapasskii V S, Feofilov P P *Sov. Phys. Usp.* **18** 323 (1975); *Usp. Fiz. Nauk* **116** 41 (1975)
2. Landau L D, Lifshitz E M *Quantum Mechanics. Non-Relativistic Theory* (Oxford: Pergamon Press, 1977); Translated from Russian: *Kvantovaya Mekhanika. Nerelyativistskaya Teoriya* (Moscow: Nauka, 1974)
3. Buckingham A D, Stephens P J *Annu. Rev. Phys. Chem.* **17** 399 (1966)
4. Van Vleck J H, Hebb M H *Phys. Rev.* **46** 17 (1934)
5. Zapasskii V S *Sov. Phys. Solid State* **15** 2367 (1973); *Fiz. Tverd. Tela* **15** 3544 (1973)
6. Feofilov P P *The Physical Basis of Polarized Emission* (New York: Consultants Bureau, 1961); Translated from Russian: *Polyarizovannaya Lyuminestsentsiya Atomov, Molekul i Kristallov* (Moscow: Fizmatgiz, 1959)
7. Aleksandrov E B, Zapasskii V S *Opt. Spectrosc.* **41** 502 (1976); *Opt. Spektrosk.* **41** 855 (1976)
8. Jones R V *Proc. R. Soc. London A* **349** 423 (1976)
9. Zapasskii V S *J. Appl. Spectrosc.* **37** 857 (1982); *Zh. Priklad. Spektrosk.* **37** 181 (1982)
10. Glasenapp P et al. *Phys. Rev. B* **88** 165314 (2013)
11. Antipin A A, Zapasskii V S *Opt. Spectrosc.* **50** 263 (1981); *Opt. Spektrosk.* **50** 486 (1981)
12. Zapasskii V S, in *Spectroscopy of Solids Containing Rare Earth Ions* (Modern Problems in Condensed Matter Sciences, Vol. 21, Eds A A Kaplyanskii, M F Macfarlane) (Amsterdam: Elsevier, 1987) p. 674
13. Antipin A A, Zapasskii V S, Lunter S G *Sov. Phys. Solid State* **24** 1843 (1982); *Fiz. Tverd. Tela* **24** 3248 (1982)
14. Zapasskii V S, Kozlov G G, Fedorov Yu K *Sov. Phys. Solid State* **29** 378 (1987); *Fiz. Tverd. Tela.* **29** 659 (1987)
15. Zapasskii V S, Kozlov G G, "Sposob opredeleniya soderzhaniya paramagnitnykh primesei v steklakh" ("Method for measurement of paramagnetic impurity content in glass"), Author's Certificate 1414096, Appl. 4054713 10.04.1986 (1986)
16. Aleksandrov E B, Zapasskii V S *Sov. Phys. Solid State* **20** 679 (1978); *Fiz. Tverd. Tela* **20** 1180 (1978)
17. Zapasskii V S *Sov. Phys. Solid State* **29** 514 (1987); *Fiz. Tverd. Tela* **29** 899 (1987)
18. Zapasskii V S, Kozlov G G, Malyshev V A *Sov. Phys. Solid State* **27** 1645 (1985); *Fiz. Tverd. Tela* **27** 2742 (1985)
19. Zapasskii V S, Kozlov G G, Malyshev V A *Sov. Phys. Solid State* **27** 64 (1986); *Fiz. Tverd. Tela* **28** 119 (1986)
20. Zapasskii V S, Kozlov G G, Malyshev V A *Sov. Phys. Solid State* **27** 74 (1986); *Fiz. Tverd. Tela* **28** 138 (1986)
21. Zapasskii V S, Zarubina T V, Kozlov G G *Opt. Spectrosc.* **68** 427 (1990); *Opt. Spektrosk.* **68** 733 (1990)
22. Aleksandrov E B, Zapasskii V S *Sov. Phys. JETP* **54** 64 (1981); *Zh. Eksp. Teor. Fiz.* **81** 132 (1981)
23. Oestreich M et al. *Phys. Rev. Lett.* **95** 216603 (2005)
24. Giri R et al. *Phys. Rev. B* **85** 195313 (2012)
25. Römer M, Hübner J, Oestreich M *Rev. Sci. Instrum.* **78** 103903 (2007)
26. Aleksandrov E B, Zapasskii V S *J. Phys. Conf. Ser.* **397** 012030 (2012)
27. Zapasskii V S *Adv. Opt. Photon.* **5** 131 (2013)
28. Hübner J et al. *Phys. Status Solidi B* **251** 1824 (2014)
29. Römer M, Hübner J, Oestreich M *Appl. Phys. Lett.* **94** 112105 (2009)
30. Müller G M et al. *Phys. Rev. B* **81** 121202(R) (2010)
31. Yang L et al. *Nature Commun.* **5** 4949 (2014)
32. Zapasskii V S et al. *Phys. Rev. Lett.* **110** 176601 (2013)
33. Gorbovitskii B M, Perel V I *Opt. Spectrosc.* **54** 229 (1983); *Opt. Spektrosk.* **54** 388 (1983)
34. Kavokin A V et al. *Phys. Rev. B* **56** 1087 (1997)
35. Li Y Q et al. *Appl. Phys. Lett.* **88** 193126 (2006)
36. Dong L et al. *J. Appl. Phys.* **107** 093101 (2010)
37. Zapasskii V S, Przhibelskii S G *Opt. Spectrosc.* **110** 917 (2011); *Opt. Spektrosk.* **110** 970 (2011)
38. Zapasskii V S *Opt. Spectrosc.* **47** 450 (1979); *Opt. Spektrosk.* **47** 810 (1979)
39. Ryzhov I I et al. *Appl. Phys. Lett.* **106** 242405 (2015)
40. Ryzhov I I et al. *Sci. Rep.* **6** 21062 (2016)