Magnetic resonance in the Faraday-rotation noise spectrum

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A maximum at the magnetic resonance frequency of sodium atoms in the ground state is observed near the 5896 Å absorption line in the fluctuation spectrum of the azimuth of the polarization plane of light crossing a magnetic field in sodium vapor. The experiment is a demonstration of a new EPR method which does not require in principle magnetic polarization of the investigated medium, nor the use of high-frequency or microwave fields to induce the resonance.

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INTRODUCTION

All the existing approaches to the investigation of paramagnetism are based on registration of the regular magnetization produced in a paramagnet under the influence of applied (constant or alternating) magnetic fields, or produced by other special techniques (optical pumping). In particular, electron and nuclear magnetic resonance are observed in the form of spectral singularities in the response of the magnetized medium to an applied high-frequency magnetic field. They are revealed either by the decrease of the longitudinal magnetization under the influence of the resonant alternating magnetic field, or by the appearance of precessing magnetization perpendicular to the external magnetic field. One way or another, the necessary premise for the study of a paramagnet in such an approach is the presence of initial magnetic polarization, whose variation under the influence of an external perturbation is used in fact for the diagnostics of its magnetic properties. Yet if the average populations of the magnetic sublevels of the paramagnet are equal, the magnetization of a microscopic sample should be subject to thermal fluctuations, which naturally are small to the extent that the law of large numbers is applicable. An investigation of the dynamics of these fluctuations can yield the same information as the magnetic-resonance method, but under substantially different conditions.

In the present study we have recorded, for the first time ever, magnetic resonance in the fluctuation spectrum of the transverse magnetization of an ensemble of paramagnetic atoms. The magnetization fluctuation was detected by the noise of the Faraday rotation in the spectral region near a resonant optical transition of the system. The object of the investigation were sodium-vapor atoms, which are paramagnetic in the ground state $3^2S_{1/2}$.

The present paper is related in its content to a number of works of the last decade (see the preceding paper[1] and the references in it), devoted to investigations of the information content of the noise spectrum of the intensity of light that is resonantly emitted or absorbed by a medium. An important step forward in the present paper is probing by nonresonant radiation that does not disturb the investigated dynamics of the magnetization. This permits the use of intense laser light sources that ensure so reliable a registration of the sought fluctuations that one can speak of the establishment of a methodologically new trend in magnetic-resonance research.

QUALITATIVE PICTURE AND ESTIMATE OF THE MAGNITUDE OF THE EFFECT

By virtue of the known connection between magnetic rotation of the polarization plane and the magnetization of the spin system (see, e.g., Ref. 2), the fluctuations of the magnetization of an ensemble of paramagnetic atoms should be observed in the noise of the rotation of the plane of polarization of light passing through the medium. The spectrum of this noise-induced modulation of the azimuth of the plane of polarization of the light is determined by the dynamics of the fluctuations of the elements of the density matrix of the ensemble of atoms and, as can be easily seen, contains in the simplest case ($S = \frac{1}{2}$) two components. The first, due to the fluctuations of the populations of the magnetic sublevels, reflects the fluctuations of the longitudinal magnetization of the spin system, is centered at zero frequency, and should be optimally observed in a light beam parallel to the applied magnetic field. The width of this spectral component ($\Delta\omega_1$) is determined by the time of the longitudinal spin relaxation of the paramagnetic atoms ($\Delta\omega_1 \approx 2T_1^{-1}$). The second component is connected with the fluctuations of the coherence of the magnetic sublevels and reflects the fluctuations of the transverse magnetization. This component is centered at the frequency of the magnetic splitting of the sublevels and is optimally observed in a beam perpendicular to the magnetic field. In classical terms, it is the result of precession, in an external magnetic field, of randomly produced magnetization whose transverse component turns out to be periodically (at the Larmor-precession period) oriented parallel and antiparallel to the probing light beam. The width of the second noise-spectrum component is determined by the transverse (phase) relaxation of the magnetization ($\Delta\omega_2 \approx T_2^{-1}$). In condensed media, the width of this component will, naturally, coincide with the width of the EPR line, and in atomic systems, in such a registration procedure, the resonance line can be broadened as a result of the effective phase relaxation due to the time of diffusion (or flight) of the atom through the light beam.

In our experiments we investigated precisely this spectral resonant component, whose observation is

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greatly facilitated by the possibility of controlling its frequency. In particular, this makes it possible to advance into the region of sufficiently high frequencies, free of industrial noise and, by modulating the magnetic field, employ the lock-in detection technique.

A consistent theoretical analysis of the refraction noise was carried out by Golubev and Plimak.[3] We confine ourselves here to a simple estimate of the magnitude of the expected effect as applied to sodium vapor in the region of the resonant absorption lines D_1 and D_2. The refractive index n of a medium with an atom density N_0 is given in the vicinity of the absorption line with an oscillator strength f in the transparency region by the formula[4]

$$n-1=\lambda_0^3 e^2 N_0 f/4\pi mc^2\Delta\lambda \equiv kN_0/\Delta\lambda. \quad (1)$$

Here m and e are respectively the mass and charge of the electron, c is the speed of light, λ_0 is the wavelength of the center of the absorption line, and $\Delta\lambda=\lambda-\lambda_0$ is the spectral detuning. The total phase shift φ of the light over a length l, expressed in radians, is given by

$$\varphi=2\pi nl/\lambda. \quad (2)$$

Changing over to circular birefringence, we neglect the hyperfine structure of the sodium levels. The rotation θ of the plane of polarization of the light is due to the difference between the refractive indices for the orthogonal circular polarizations, which is proportional to the difference between the densities $N_{1/2}$ and $N_{-1/2}$ of the sodium atoms with spin projections $\pm\frac{1}{2}$ on the direction of the light beam:

$$\theta=\alpha k\pi l(N_{1/2}-N_{-1/2})/(\lambda\Delta\lambda). \quad (3)$$

The coefficient α is close in magnitude to unity and is determined by the angular momenta of the upper and lower states of the transition in whose vicinity the rotation is calculated. For the sodium resonant lines D_1 and D_2 this coefficient is respectively[1)] 1 and −0.5.

The fluctuations $\Delta\theta$ of the rotation of the plane of polarization are determined by the fluctuations of the difference between the numbers of atoms with opposite spin directions contained in the light beam, i.e., by the dispersion of the quantity $(N_{1/2}-N_{-1/2})lS$, where S is the cross section of the beam. When the mean values of the populations are close, $N_{1/2}\approx N_{-1/2}\approx N_0/2$, this dispersion is obviously equal to $N_0 lS\equiv N$—the total number of atoms in the beam. Thus,

$$\langle(\Delta\theta)^2\rangle^{1/2}=\frac{\pi\alpha k}{\lambda\Delta\lambda}\left(\frac{N_0 l}{S}\right)^{1/2}. \quad (4)$$

The fluctuations $\Delta\theta$ are detected with a polarimeter that transforms them into oscillations of the light intensity, and next into oscillations of the photocurrent. In the balance polarimeter used by us[6] the resultant relative fluctuation of the photocurrent i_{ph} is connected with $\Delta\theta$ by the relation $\Delta i_{ph}/i_{ph}=4\Delta\theta$. The information-containing fluctuations of the photocurrent Δi_{ph} should be compared with the shot fluctuations δi_{ph} in the given frequency band $\Delta\nu$:

$$\langle(\delta i_\Phi)^2\rangle=4ei_\Phi\Delta\nu. \quad (5)$$

Expression (5) corresponds to double the value of the Schottky noise, inasmuch as the noise of two identical independent channels are superimposed in the balance detector of the polarimeter.

Let us estimate the possibility of observing the fluctuations $\Delta\theta$ for the following real set of parameters: $N_0=10^{14}$ cm^{-3}, $\Delta\lambda=1$ Å, $f=0.33$ (the vicinity of the D_1 line), $i_{ph}=10^{-4}$ A, $l=5$ cm, $S=1$ mm^2, and $\Delta\nu=2\times10^4$ Hz. The chosen registration band $\Delta\nu$ is matched to the noise spectrum width expected from estimates of the phase relaxation time and from the inhomogeneous magnetic broadening.

At the assumed values we have $\Delta i_{ph}/i_{ph}=0.7\times10^{-5}$ and $\langle(\delta_{iph})^2\rangle^{1/2}/i_{ph}=1.1\times10^{-5}$. Thus, the flicker noise of interest to us is close in its spectral density to shot noise, leaving no doubt of the possibility of observing it. Indeed, it can be shown that after passing through a filter with half-width γ (the parameter of a Lorentz contour centered at the frequency $\omega_0\gg\gamma$), the mean squared level of the Poisson noise during the observation time τ can be determined with a relative accuracy[2)] $[(1+\gamma e/i_{ph})/\gamma\tau]^{1/2}$. At a sufficiently large photocurrent ($i_{ph}\gg\gamma e$) this factor goes over into the well known expression $(\gamma\tau)^{-1/2}$, which is valid for a Gaussian noise.[7] At $\gamma\sim10^5$ and $\tau\sim1$ sec, the aforementioned accuracy is of the order of 0.3%, i.e., within the limits of this accuracy the shot noise can be regarded as a constant background does not interfere with the measurement of the information-containing part of the noise.

EXPERIMENT

A diagram of the experimental setup is shown in Fig. 1. Linearly polarized radiation from a dye laser (rhodamine-6Zh, Spectra Physics model 375, lasing-spectrum width ~30 GHz) passed through a cell with the sodium vapor in a neon atmosphere at a pressure of 10 Torr. The lasing wavelength corresponded to the transparency region of the vapor near the absorption lines D_1 and D_2. The transverse magnetic field was produced by a system of Helmholtz coils and was calibrated beforehand against the magnetic-resonance frequency in potassium vapor under optical orientation conditions. Coaxially with the constant magnetic field, a low-frequency (~10^2 Hz) alternating magnetic field was applied to the cell. The dynamics of the azimuth

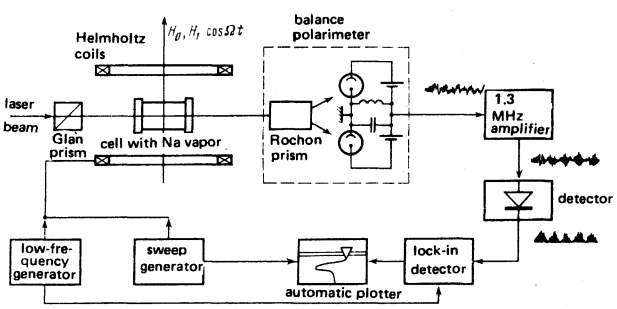

FIG. 1. Diagram of experimental setup. 1) Helmholtz coils, 2) balance polarimeter, 3) laser beam, 4) Glan prism, 5) cell with Na vapor, 6) Rochon prism, 7) 1.3 MHz amplifier, 8) detector, 9) low-frequency generator Ω, 10) sweep generator, 11) automatic plotter, 12) lock-in detector.

of the plane of polarization of the light beam passing through the cell was registered with a balanced photodetector. The photodetector signal was selectively amplified at a frequency 1.3 MHz and band width ~20 kHz, after which it was quadratically detected. The low-frequency magnetic field applied to the cell led to modulation of the position of the magnetic-resonance line relative to the registration frequency (1.3 MHz). In the presence of a signal (a singularity in the noise spectrum) this manifested itself in modulation of the noise power and in the appearance of a low-frequency signal. Afterlow-frequency amplification, the signal was again detected in phase with the modulation of the magnetic field. The signal from the output of the lock-in detector was recorded while the magnetic field H_0 was slowly scanned in the vicinity of the resonance intensity.

The sensitivity of the setup was measured by imitating the signal with the aid of a Faraday cell having a known Verdet constant. The cell was fed from a high-frequency oscillator (~1.3 MHz) with low-frequency amplitude modulation of the HF power. The sensitivity threshold at the mean squared fluctuation level was $\sim 10^{-5}$ degree of angle after 0.5 sec of observation. This is worse by approximately an order of magnitude than the calculated (limiting) sensitivity.

A distinct signal of resonant increase of the noise density at 1.3 MHz was registered in the vicinity of the calculated value of the field intensity 1.85 Oe in a wide range of sodium densities $10^{12}-10^{14}$ cm^{-3}. The signal power increased when the laser frequency approached the absorption lines and passed through a maximum in the region of appreciable absorption, where the intensity of the transmitted light dropped substantially. The signal was reliably observed also in the region of undisputed transparency, particularly when the laser line was set between the resonance lines of the sodium, i.e., at a distance ~3 Å from the absorption line. In the transparency region, the signal varied linearly with the radiation intensity, as verified in the power range 10–80 mW. The signal power (likewise in the transparency region) increased when the laser beam was focused. This is natural, in view of the decreased number of atoms in the light beam, which leads to an increase of the refraction fluctuations [see expression (4)]. In all cases, the resonance width was close to the instrumental value (~35 kHz) determined by the bandwidth of the high-frequency amplifier. This observation is in accord with the estimated time of diffusion of the sodium atoms through the light-beam cross section (the remaining spin-relaxation processes are negligible under our conditions).

In the absorption region, the dependence of the signal on the beam cross section became more complicated. Focusing of the beam, which caused an increase in the density of the optical excitation, led to a considerable shortening of the time of the optical excitation cycle, to an effective increase of the rate of phase relaxation, to a broadening of the resonance line, and to a change of its contour. Simultaneously, the signal ceased to depend linearly on the beam intensity—effects of

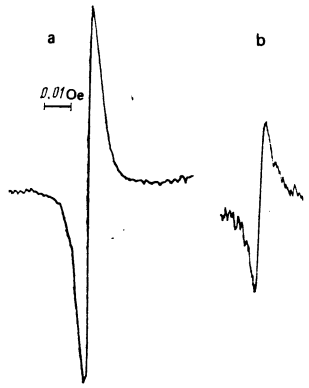

FIG. 2. Plots of EPR signal in Faraday-rotation noise: a—at the edges of the 5896 Å absorption line, b—between the 5896 and 5890 Å lines.

bleaching, distortion of the wavefront, birefringence in the cell windows, and others came into play. Therefore the optimal beam cross section at a given lasing power depended on the spectral proximity to the absorption line. When the magnetic-resonance signal was recorded, the conditions were chosen such that the effect of the light on the magnitude and shape of the signal could be neglected.

Typical plots of the noise-resonance signal, with time constant 2 sec, are shown in Fig. 2. Signal *a* was obtained with the laser tuned to the edge of the absorption line D_1 for a beam with approximate diameter 1 mm and power 20 mW. Signal *b* corresponds to the laser tuned to the midpoint of the interval between the resonant-doublet lines in a beam of the same power, focused by a lens of focal length 130 mm. In both cases the modulation amplitude was set at the maximum level that still did not broaden the resonance line.

The adsorbed resonance corresponds to a magnetic splitting 1.3 MHz of adjacent sodium magnetic sublevels. Since a real sodium atom has a hyperfine structure (hfs) and accordingly has not two but an entire system of almost equidistant sublevels, a noise signal can be produced in principle by interference of every other (odd or even) level produced at the same registration frequency in a field of half the intensity (the noise-alignment signal). Observation of this effect calls for registration of the linear-birefringence noise for longitudinal or transverse propagation of the light relative to the magnetic field. This signal was not observed in experiment, since it is weak in the ratio of the hyperfine splitting of the excited state to the larger of the two quantities—the deviation $\Delta\lambda$ of the probing radiation from resonance and the absorption line width.[8]

As indicated, the main results were obtained for a resonance frequency 1.3 MHz. In supplementary experiments, we varied the amplifier frequency, and this shifted correspondingly the position of the resonance relative to the magnetic field. With increasing registration frequency, the signal began to broaden and then, starting with ~8 MHz, it split into a number of components as a result of violation of the equidistance of the Zeeman sublevels of the sodium (the Paschen-Back effect in hfs).

CONCLUSION

In the present study, to our knowledge, we have demonstrated for the first time ever the possibility of optically observing magnetic resonance in the magnetization-noise spectrum of an ensemble of paramagnetic atoms. The possibility of induction registration of magnetization noise due to a disordered precession of the nuclear moments was theoretically indicated in a paper by Skrotskii and Kokin.[9]

From the fundamental point of view, the validity of the described classical picture of the phenomenon and hence the possibility of obtaining information on the dynamics of a free spin system from the noise spectrum of light passing through it were not obvious to us beforehand. The classical interpretation of the effect, in which the influence of the probing light on the investigated system is neglected, neglects simultaneously the fundamental properties of the quantum-mechanical measurement procedure. For this reason we have regarded a rigorous quantum-electrodynamic confirmation of the effect as important.[3]

From the methological point of view, the technique developed can, in our opinion, be of considerable interest for certain investigations of paramagnetic centers in solids. The magnitude of the effect in solids decreases by approximately three orders on account of the increased width of the absorption lines, but this loss can be offset by an increase in the concentration of the paramagnetic centers. A distinct sensitivity margin, due to the absence of diffuse motion of the paramagnetic centers, is provided by the possibility of greatly decreasing the diameter of the probing light beam. In addition, regardless of the aggregate state of the system, it is possible to increase greatly the sensitivity of the registration by increasing the number of beam interactions with one and the same volume element—by placing the investigated object in a Fabry-Perot resonator.

Without dwelling in detail on these questions, we note certain peculiarities of the noise technique, which can make it preferable in a number of cases. The most distinguishing feature of the noise procedure of EPR registration is the absence of temperature and field dependences of the signal in the classical limit $g\beta H_0 \ll kT$, where in the case of ordinary registration the EPR signal, proportional to the population difference, decreases with temperature like $g\beta H_0/kT$. The noise procedure, which can be used equally successfully when the average populations of the magnetic sublevels are exactly equal, may turn out to be preferable for high-temperature investigations of relaxation processes. The noise-resonance technique is substantially different also because of the absence of inducing alternating magnetic fields. This excludes field broadenings of the level and simplifies the experimental procedure.

We note, finally, that in principle one can consider a modified variant of the noise procedure of EPR registration, based on registration of circular-dichroism noise and appearing in the ellipticity noise of the (initially linearly polarized) light passing through the medium in the absorption region. Without dwelling on the sensitivity of this variant of the method, which depends in a more complicated manner on the characteristics of the real optical transtion,[3] we note only that for a solid, just as in ordinary optical registration of EPR,[10] the Faraday procedure is preferable, since solids with paramagnetic centers exhibit as a rule an appreciable paramagnetic Faraday effect in the entire transparency region. This promises to permit the use of highly sensitive fixed-wavelength polarimeters to register the EPR in the noise spectra of the Faraday rotation in various condensed media.

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[1)]The picture of the spectral behavior of circular birefringence of optically oriented sodium atoms in the vicinity of a resonant doublet, with allowance for the hyperfine structure, was considered in detail in Ref. 5.

[2)]The results of this calculation were kindly supplied to us by V. P. Kozlov.

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