[3]Tonkie ferromagnitnye plenki (Thin Ferromagnetic Films), (translations), R. V. Telesnin, ed., Mir, 1964.
[4]Y. Murayama, J. Phys. Soc. Jap. 21, 2253 (1966).
[5]A. Holz and H. Kronmüller, Phys. Status Solidi 31, 787 (1969).
[6]V. V. Tarasenko, E. V. Chenskiĭ, and I. E. Dikshtein, Zh. Eksp. Teor. Fiz. 70, 2182 (1976) [Sov. Phys. JETP 43, 1136 (1976)].
[7]A. I. Akhiezer, V. G. Bar'yakhtar, and S. V. Peletminskiĭ, Spinovye volny (Spin Waves), Nauka, 1967 (transl., North-Holland, 1968).
[8]L. D. Landau, Zh. Eksp. Teor. Fiz. 7, 19 (1937) [Phys. Z. Sowjetunion 11, 26 (1937) (in German)] (translation in Collected Papers of L. D. Landau, Pergamon Press, 1965, p. 193 and in D. ter Haar, Men of Physics: L. D. Landau, Vol. II, Pergamon Press, 1965, p. 61).

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Investigation of ferromagnetic resonance in $Y_3Fe_5O_{12}$ by the method of optical spectroscopy

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Results are presented of an investigation of inelastic scattering of linearly polarized monochromatic light ($\lambda = 6328$ Å) by homogeneous-precession magnons excited by FMR in a thin $Y_3Fe_5O_{12}$ plate. By separating the contributions of the magneto-optical effects to the intensities of the spectral lines with combination frequencies, the amplitudes and polarizations of the precessions are reconstructed as functions of the microwave magnetic field. It is shown that when the threshold for the second-order parametric process is exceeded, besides saturation of the amplitude of the homogeneous mode, a substantial change takes place also in its polarization.

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High-frequency modulation of light in ferromagnetic resonance (FMR) in magnetically ordered crystals has been the subject of relatively few studies. Mention should be made of a paper by Hanlon and Dillon,[1] where direct spectroscopic proof was obtained that microwave satellites appear when monochromatic linearly polarized light passes through a $CrBr_3$ single crystal. High-frequency modulation of light in antiferromagnetic resonance in a $CoCO_3$ crystal was observed by Borovid-Romanov *et al.*[2]

At the same time, the study of magneto-optical phenomena due to FMR is of considerable interest, especially in crystals having comparable values of the Faraday rotation (FE) and the linear magnetic birefringence—the Cotton-Mouton effect (CME). It will be shown below that in this case optical measurements yield sufficiently complete information on the most important parameters of the homogeneous magnetization oscillations in FMR.

We consider the interaction of linearly polarized monochromatic light with a ferromagnetic crystal, in which homogeneous precession of magnetization is excited. We assume for simplicity that the crystal has cubic symmetry (for example, $Y_3Fe_5O_{12}$) and is magnetized along the [001] axis.

We choose the coordinate system such that its axes coincide with the fourfold axes of the crystal, and the constant magnetic field H_0 is directed, as usual, along the Z axis. In this case the crystal is optically uniaxial with the optical axis directed along the magnetization.[3] Inasmuch in FMR the deviation of the magnetic moment from its equilibrium position is small, it is accurate enough to state that the optical axis of the crystal precesses. Assume that the light propagates along the X axis. The vector $\mathbf{E}$ of the incident light can then be represented in the form

$$\mathrm{E}_0=\{E_{0z}, E_{0y}\}=\{\cos\psi, \sin\psi\}e^{i\omega_0 t}, \quad (1)$$

where ψ is the angle between the vector $\mathbf{E}$ and the Z axis; ω_0 is the frequency; the intensity is set equal to unity. The change of the polarization of the light passing through the crystal can be described by the corresponding Jones matrix $\hat{Q}$, the elements of which have been calculated with allowance for the principle of superposition of the FE and the CME. By a procedure similar to that used by Tron'ko[4] we obtain expressions for the matrix element that describe the light emerging from the sample

$$Q_{zz}=Q_{yy}^*=\cos\Delta l+i\delta\cos 2\theta_y\sin\Delta l/2\Delta, \quad (2)$$

$$Q_{zy}=-Q_{yz}^*=\Delta^{-1}(^1/_2 i\delta\sin 2\theta_y-\rho_x)\sin\Delta l, \quad (3)$$

$$\Delta=(^1/_4\delta^2+\rho_x^2)^{1/2}, \quad (4)$$

where ρ_x is the Faraday rotation per unit thickness of the crystal and is due to the component m_x of the magnetization vector (in the absence of precession we have $\rho_x\equiv 0$, for in this case the light propagates perpendicular to $\mathbf{M}_0$); δ is the specific phase shift of the CME ($\delta \sim |M|^2$); θ_y is the angle between the projection of the magnetization vector on the YZ plane and the Z axis; l is the dimension of the crystal in the X direction.

At small l (thin plate) and small $\rho_x l$ and θ_y, taking into account the fact that the observation is carried out with a crossed polarizer-analyzer system, we obtain from (1)–(4) an approximate expression for the vector $\mathbf{E}$ of the light at the exit from the analyzer:

$$\mathbf{E}_1 = \hat{P}(\psi+\pi/2)\hat{Q}(\rho_x, \theta_y, \delta, l)\mathbf{E}_0(\psi, t)$$
$$\approx \left[\rho_x l + i\delta l\left(\theta_y \cos 2\psi - \frac{1}{2}\sin 2\psi\right)\right]\begin{pmatrix} -\sin\psi \\ \cos\psi \end{pmatrix} e^{i\omega_0 t}. \quad (5)$$

Here $\hat{P}(\psi+\pi/2)$ is the matrix of a linear polarizer with a transmission axis making an angle $\psi+\pi/2$ with the Z axis.[5]

In formula (5) it is easy to change over from the quantities ρ_x and θ_y to the amplitudes of the transverse magnetization components m_{x0} and m_{y0}, using the relations

$$\rho_x = \rho_0 \frac{m_x}{M_0} = \rho_0 \frac{m_{x0}}{M_0} \cos \Omega t, \quad (6)$$

$$\theta_y = \arcsin \frac{m_y}{M_r} \approx \frac{m_{y0}}{M_r} \sin \Omega t, \quad (7)$$

where Ω is the frequency of the magnetization precession; M_0 and M_r are the saturation magnetization off resonance and at resonance, respectively; ρ_0 is the specific Faraday rotation due to the magnetization M_0. In FMR, in the course of the relaxation of homogeneous-precession magnons, spin waves are produced with wave vector $k \neq 0$, and this decreases the modulus of the magnetic moment, i.e., $M_r < M_0$. In turn, inasmuch as $\delta \sim |M|^2$, the resonant value of the CME phase shift $\delta = \delta_0(M_r/M_0)$ also decreases in comparison with the corresponding value δ_0 off resonance.

Taking the foregoing into account, we can obtain from (5)–(7) expressions for the intensities of the spectral components with combination frequencies ω_0, $\omega_0 - \Omega$, and $\omega_0 + \Omega$ at the exit from an analyzer crossed with a polarizer:

$$I_{\omega_0} \approx {}^1/_4 l^2 \tau \delta^2 \sin^2 2\psi, \quad (8a)$$

$$I_S = I_{\omega_0-\Omega} \approx \frac{l^2}{4}\tau\left(\rho_0 \frac{m_{x0}}{M_0} - \delta \frac{m_{y0}}{M_r}\cos 2\psi\right)^2, \quad (8b)$$

$$I_{AS} = I_{\omega_0+\Omega} \approx \frac{l^2}{4}\tau\left(\rho_0 \frac{m_{x0}}{M_0} + \delta \frac{m_{y0}}{M_r}\cos 2\psi\right)^2. \quad (8c)$$

We took additional account here of the sample optical transparency τ, which depends on the sample temperature. It is easily seen that the system of equations (8a)–(8c), supplemented by the dependence of τ on the pump microwave power, determines completely the amplitudes m_{x0} and m_{y0}, and consequently also the magnetization polarization. The determination of these parameters by a standard resonance method, for samples whose shape differs from spherical, entails considerable difficulties. It must be noted that the question of the contributions of the FE and the CME to the intensity of the light scattered by spin waves has been considered earlier.[8] The classical model employed here above is preferable in our case, however, since it makes it possible to determine not only the amplitude but also the polarization of the magnetization precession.

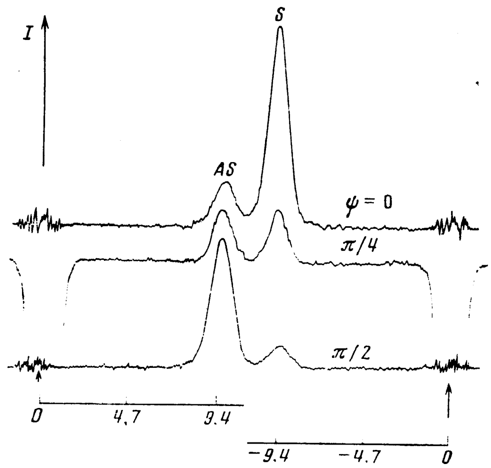

FIG. 1. Stokes (S) and anti-Stokes (AS) satellites in the spectrum of the light passing through a $Y_3Fe_5O_{12}$ plate in which FMR is excited; ψ is the angle between the vector $\mathbf{E}$ of the linearly polarized light incident on the sample and the constant magnetic field.

We have investigated the high-frequency modulation of linearly polarized light ($\lambda = 6328$ Å) by a thin $Y_3Fe_5O_{12}$ plate ($l = 60\ \mu$m) placed in the antinode of the microwave magnetic field of an reentrant 3-cm band resonator. The resonator was excited by a magnetron whose radiation was modulated with rectangular pulses with a duty factor 0.5 ("meander"). The measurements were made at rather high pump intensities (h) at which the FMR is essentially nonlinear. To separate the spectral components of the light with the combination frequencies we used a Fabry-Perot interferometer scanned by varying the air pressure between the mirrors. The light intensity at the exit from the interferometer was measured with a photomultiplier whose signal was amplified by a narrow-band amplifier and passed through a synchronous detector to an automatic recorder. All the measurements were made at room temperature.

Figure 1 shows experimental interference patterns for three characteristic polarizations of the incident light at a fixed pump power. Since we registered the alternating component of the photomultiplier signal (having the same frequency as the microwave pulse repetition), the lines corresponding to the undisplaced component of the spectrum were revealed at $\psi = 0$ and $\psi = \pi/2$ only by the increase of the noise level. The signal of the low-frequency modulation of the light of undisplaced frequency at a polarization $\psi = \pi/4$ is due to the decrease of δ during the time of the microwave pump pulses. Naturally, this signal is in anti-phase with the signals I_S and I_{AS}. The observed dependence of the intensities of the Stokes and anti-Stokes satellites on the polarization of the incident light is due, as follows from expressions (8b) and (8c), to the contribution of the magnetic birefringence. Measurement of the quantities I_S/I_0 and I_{AS}/I_0 (I_0 is the intensity of the incident light) at a fixed pump power makes it possible to determine the depth of the microwave modulation of the light from the intensity, with the aid of the relation

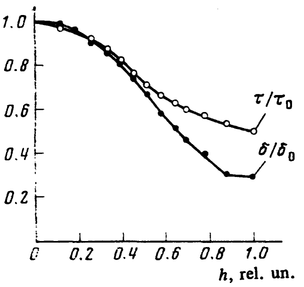

FIG. 2. Linear magnetic birefringence of the light (δ) and transparency of the crystal (τ) as functions of the microwave magnetic field; δ_0 and τ_0 are the corresponding values at room temperature (off resonance).

$$\alpha = I_\sim / I_0\tau = (I_S + I_{AS})/I_0\tau.$$

The maximal experimentally obtained value of the parameter α was 2×10^{-7}.

As shown above, to determine the transverse magnetization components we must have the dependences of the quantities δ, τ, and M_r on the microwave fields. The $\delta(h)$ dependence was determined from the change of the stationary component of the light intensity (of fundamental frequency) at the output of the analyzer crossed with the polarizer at $\psi=\pi/4$ (see Eq. (8a)). The $\tau(h)$ dependence was measured similarly, but without the analyzer. Plots of $\delta(h)$ and $\tau(h)$ normalized to the corresponding values off resonance are shown in Fig. 2. The pump-field intensity h is given in relative units. It was noted above that $\delta=\delta_0(M_r/M_0)^2$, so that the function $\delta(h)/\delta_0$ obtained by the described method reflects the growth of the number of magnons with $k\neq0$ (parametric, intermediate, and thermal) with increasing pump field h. On the other hand, the observed decrease in transparency of the sample is due to the heating of its crystal lattice by the absorbed microwave power.

Plots of the amplitudes of the transverse components of the magnetization m_{x0} (curve 1) and m_{y0} (curve 2) against h, obtained by the described method, are shown in Fig. 3. The same figure shows a plot of the ellipticity of the magnetization polarization (m_{x0}/m_{y0}) in the transverse plane (curve 3). The measurements were made in the maximum of the FMR absorption line ($H_0 = H_{0r}(h)$) with the aid of light polarized perpendicular to the magnetization field ($\psi=\pi/2$). With increasing microwave pump power, a shift of the resonant magnetic field was observed, so that tuning to resonance was carried out at each value of h. We used in the calculations the magneto-optical constants $\rho_0=1000$ deg/cm [7] and $\delta_0 = -400$ deg/cm (our measurements).

The presented plots reveal mainly two characteristic regions of the variation of m_{x0} and m_{y0}. The interval $0<h<h_c$ corresponds to linear FMR. The ellipticity is constant in this region of h, and its value is determined approximately by the expression

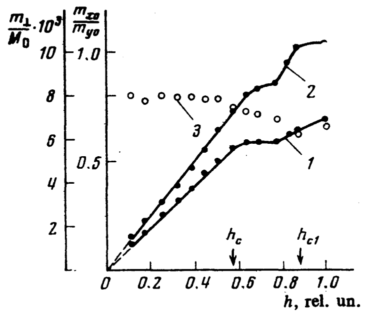

FIG. 3. Maximum amplitudes of the transverse components of the magnetization m_{x0} (curve 1) and m_{y0} (curve 2) and of the ellipticity (curve 3), obtained by the optical method, as functions of the alternating magnetic field. The plane of the ferrite plate coincides with the YZ plane. The linearly polarized microwave field is directed along X and the constant magnetic field along Z.

$$\frac{m_{x0}}{m_{y0}} \approx \left(\frac{H_0}{H_0+4\pi M_0}\right)^{1/2}.$$

In fields $h>h_c$, a partial saturation of the precession amplitude is observed, with simultaneous change of the ellipticity. The observed change of the precession polarization has a pronounced threshold and can therefore not be connected with the usual nonlinear frequency doubling. It can therefore be assumed that h_c is the threshold field for a second-order parametric processes. Estimates yield $h_c\approx0.3$ Oe. At $h>h_{c1}$, slow oscillations of the crystal magnetization are observed, with frequency $F\approx1$ MHz (self-modulation).

We have thus observed inelastic scattering of monochromatic light ($\lambda=6328$ Å) by homogeneous-precession magnons excited by FMR in a thin $Y_3Fe_5O_{12}$ plate. By separating the contributions of the magneto-optical effects (FE and CME) to the intensities of the combination-frequency spectral lines we reconstructed the dependence of the amplitude and of the precession ellipticity on the microwave pump magnetic field. It is shown that not only saturation of the homogeneous-mode amplitude, but also a change of its polarization are observed above the threshold of the second-order parametric process.

[1]J. T. Hanlon and J. F. Dillon, J. Appl. Phys. 36, 1269 (1965).
[2]A. S. Borovik-Romanov, V. G. Zhotikov, N. M. Kreĭnes, and A. A. Pankov, Pis'ma Zh. Eksp. Teor. Fiz. 23, 705 (1976) [JETP Lett. 23, 649 (1976)].
[3]G. A. Smolenskiĭ, R. V. Pisarev, I. G. Siniĭ, N. N. Kolpakova, and A. G. Titova, Izv. Akad. Nauk SSSR Ser. Fiz. 36, 1219 (1972).
[4]V. D. Tron'ko, Opt. Spektrosk. 29, 354 (1970).
[5]W. A. Shurcliff and S. S. Ballard, Polarized Light, Van Nostrand Reinhold, 1964 [Russ. transl., Mir, 1965].
[6]H. LeGall, Tran Khanh Vien, and B. Desormiere, Phys. Status Solidi 47, 591 (1971).
[7]J. F. Dillon, J. Appl. Phys. 29, 539 (1958).

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