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EXCHANGE INTERACTION AND MAGNETO-OPTICAL EFFECTS IN FERRITE GARNETS

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It is shown that in the infrared region of the spectrum, along with the Faraday effect due to electronic transitions, there exists a Faraday effect due to magnetic spin resonance, that is, to a precession of the magnetization vector under the action of the magnetic field of the light wave. In ferrite garnets containing a magnetoactive rare-earth sublattice this effect is also determined by exchange resonance, that is, by precession of the magnetization vector of one sublattice in the exchange field of the other. In europium ferrite garnet the Zeeman effect on the line $^7F_0 \rightarrow {}^7F_6$ is observed in an exchange field of 2.5×10^5 oe. In this case the Faraday effect is due to exchange and not to spin-orbit splitting.

1. EXCHANGE RESONANCE IN FERRITES AND PRECESSION OF THE SPONTANEOUS MAGNETIZATION VECTOR AT OPTICAL FREQUENCIES

MEASUREMENTS of the Faraday effect in yttrium garnet[1] have shown that rotation of the plane of polarization is independent of λ in the region $4\mu < \lambda < 9\mu$. It has been assumed that this rotation is an exclusive property of ferromagnetic semiconductors. In the first part of this paper we shall show that the Faraday effect in the transparency region of a ferrite garnet is actually of purely magnetic origin. The frequency independent rotation of the plane of polarization of infrared light is due to the precession of the spontaneous magnetization vector of the ferromagnetic semiconductor under the influence of the magnetic vector of the light wave, i.e., it is a consequence of magnetic spin resonance. The characteristic frequencies of this resonance ω_0 occur in the region of significantly longer wavelengths; we shall therefore be interested in the theory of the Faraday effect for $\omega \gg \omega_0$.

The theoretical treatment of the motion of the ferromagnetic spin system in crossed constant and alternating magnetic fields is based on the well-known Landau-Lifshitz equation.[2] In particular, the Faraday effect, i.e., the rotation of the plane of polarization of the electromagnetic wave in the magnetization direction of the ferromagnet, is determined by the magnitude of the off-diagonal component of the permeability tensor $M = i\mu_{xy} = -i\mu_{yx}$, an expression for which can be obtained from the solution of the Landau-Lifshitz equation. From Eq. (38) of [2] we obtain for $\omega \gg \omega_0$ the following expression for $\tilde{M} = M_{\omega \gg \omega_0}$:

$$\tilde{M} = (4\pi\gamma/\omega) I_s, \qquad (1)$$

where I_S is the saturation magnetization of the ferromagnet and $\gamma = e/mc$.

Off-diagonal components of the permeability tensor different from zero lead to a specific Faraday rotation equal to

$$\alpha_F = \frac{\sqrt{\varepsilon}}{2c} \omega M$$

Substituting $M = \tilde{M}$ from Eq. (1), we have

$$\tilde{\alpha}_F = \frac{2\pi\sqrt{\varepsilon}}{c} \gamma I_s \text{ rad/cm}$$

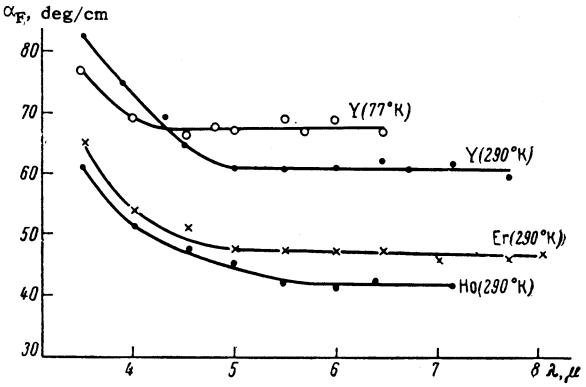

FIG. 1. Faraday effect in yttrium, erbium, and holmium garnets in infrared light at different temperatures; H_{ext} = 2400 oe.

The Faraday effect in the microwave region has been found in semiconducting ferrites, investigated in a number of papers, and utilized in microwave technology. Up until now, however, no attention has been paid to the circumstance that magnetic spin resonance should produce the same Faraday effect for infrared and visible light as it does for electromagnetic waves in the microwave region, because $\tilde{\alpha}_F$ is independent of ω. The magnitude of the specific rotation deduced from Eq. (2) amounts to several tens of degrees per centimeter.

The investigation of the rotation of the plane of polarization of light is of interest also because by this means one can study the phenomenon of exchange resonance. As Kaplan and Kittel have shown,[3] exchange resonance, i.e., magnetic spin resonance in the effective field of the exchange interaction between the magnetic sublattices, can occur in uncompensated antiferromagnets. The intensity of the exchange resonance is proportional to $(\gamma_1-\gamma_2)^2$, where $\gamma_1 = g_1e/2mc$ and $\gamma_2 = g_2e/2mc$ are the gyromagnetic ratios of the ions on the two magnetic sublattices.

The exchange resonance frequency $\omega_0^{exch} = \lambda \times (\gamma_2 I_1 - \gamma_1 I_2)$, where λ is the molecular field coefficient, and I_1 and I_2 are the spontaneous magnetizations of the two sublattices. For values of $H_{exch} = \lambda I = 10^6$ oe, the magnitude of $\omega_0^{exch} = 3 \times 10^{12}$ cps, i.e., the resonance frequency is in the far infrared region. Because of the technical difficulties in this region, direct observation of exchange resonance has so far not been reported. Effects associated with exchange resonance have been observed only near the compensation point, since then $I_1 \approx I_2$ and the frequency ω_0^{exch} decreases and shifts toward the more convenient microwave region.

Let us consider how the exchange resonance influences the magnitude of $\tilde{\alpha}_F$. Wangsness[4] has obtained expressions for the tensor components of the magnetic susceptibility of a ferrimagnet with two magnetic sublattices. Using his Eq. (16) and assuming $\omega_0^{res} \ll \omega \ll \omega_0^{exch}$, we obtain expressions for $\tilde{M}$ and $\tilde{\alpha}_F$ valid in the absence of exchange resonance (ω_0^{res} is the characteristic frequency of ordinary ferromagnetic resonance):

$$\tilde{\alpha}_F^{res} = \frac{2\pi\sqrt{\varepsilon}}{c}\gamma_{eff} I, \qquad (3)$$

where $\gamma_{eff} = (I_1 - I_2)/(I_1/\gamma_1 - I_2/\gamma_2)$ is the ratio of the total magnetic moment of the ferrite to its mechanical moment. For frequencies $\omega \gg \omega_0^{res}$, ω_0^{exch} the applicable formula in the presence of exchange resonance is

$$\tilde{\alpha}_F^{exch} = \frac{2\pi\sqrt{\varepsilon}}{c}(\gamma_1 I_1 - \gamma_2 I_2). \qquad (4)$$

When $\gamma_1 = \gamma_2$, (3) and (4) reduce to (2). The physical interpretation of these expressions is that during ordinary ferromagnetic resonance both sublattices of the ferrimagnet, bound together by the exchange forces, precess as a single unit, whereas during the exchange type of precession each sublattice makes an independent contribution to the Faraday effect, with a sign corresponding to the orientation of the magnetic moment of the sublattice. The equations for ferrimagnets with three magnetic sublattices have a similar form. For the sake of simplicity, we shall employ Eqs.

Garnet	n	Ground level of Ion: Sublattice 1	Ground level of Ion: Sublattice 2	g_1	g_2	g_{eff}	T, °K	H, oe	I_1	I_2	$\tilde{\alpha}_F^{res}$ theory	$\tilde{\alpha}_F^{exch}$ theory	$\tilde{\alpha}_F$ experiment
$Y_3Fe_5O_{12}$	2.2	$^6S_{5/2}$	—	2.0	—	2.0	290	2400	135	—	63 *	—	61
							77	2400	181	—	84	—	68
							77	2800	181	—	84	—	74
$Ho_3Fe_5O_{12}$	2.2	$^6S_{5/2}$	$^5I_8$	2.0	1.25	8.5	290	2400	135	75.8	117	41	42
$Er_3Fe_5O_{12}$	2.2	$^6S_{5/2}$	$^4I_{15/2}$	2.0	1.2	3.1	290	2400	135	46.8	63	50	47

*Values of $\tilde{\alpha}$ are given in deg/cm.

(3) and (4), since $g_1 \approx g_2 \approx 2$ for the iron sublattices in ferrite garnet, and the improvements in accuracy of the values of $\tilde{\alpha}_F$ by taking into account the difference between g_1 and g_2 exceed the limits of precision of the measurements.

Measurements of the Faraday effect in yttrium, holmium, and erbium garnets were carried out by the method described earlier[1,5] at wavelengths of from 3.5 to 8 μ. Measurements were made with an NaCl prism having a slit width of 2 mm using a vacuum thermocouple as a detector. The measurements on the yttrium garnet were made quantitatively more precise than the preliminary data,[1] and in addition were made at liquid-nitrogen temperature.

The results of the measurements are compared with theory in Fig. 1 and the table. The theoretical values for all the garnets were calculated for a dielectric constant $\epsilon = 4.48$. Data on the index of refraction of the rare-earth garnets are lacking in the literature. We measured the reflection coefficient for light of $\lambda = 0.6\mu$ and $\lambda = 1.0\mu$ from natural faces of monocrystals of yttrium and erbium garnets. The value of $n = 2.2 \pm 0.2$ was obtained for both garnets. The curves of $\tilde{\alpha}_F$ for yttrium garnet were taken in an external field H = 2400 oe. At room temperature the sample is saturated in this field, since the demagnetization field $4\pi I_S \approx 1700$ oe. At liquid-nitrogen temperature $4\pi I_S \approx 2300$ oe; hence, although the Faraday effect also increases, it still does not attain the calculated values corresponding to magnetic saturation of the sample. By forcing the electromagnet additional measurements were made at 2800 oe, which were in more satisfactory agreement with the calculated values for $\tilde{\alpha}_F$ (see table).

The data show that the experimental values of $\tilde{\alpha}_F$ for erbium and holmium garnets agree with theory only under the assumption that exchange resonance is present in these garnets. The differences between the experimental values for $\tilde{\alpha}_F$ and the theoretical values for $\tilde{\alpha}_F^{res}$ calculated from Eq. (3) for ordinary ferromagnetic resonance lie far outside the limits of possible experimental errors. A change in sign of $\tilde{\alpha}_F$ was also observed for holmium garnet upon cooling the sample to liquid-nitrogen temperature ($\lambda \approx 5\mu$), which is a further indication of the presence of exchange resonance in rare-earth garnets. Measurements of $\tilde{\alpha}_F$ at different temperatures in dysprosium and terbium garnets would be of interest, since according to Eq. (3) the specific rotations $\tilde{\alpha}_F^{exch}$ and $\tilde{\alpha}_F^{res}$ should have completely different temperature dependences in these garnets.

Thus, comparison of all the experimental results with theory leads to the conclusion that in the wavelength interval studied rotation of the plane of polarization of light by a ferromagnet is caused by its dynamic magnetic susceptibility in the alternating magnetic field of the light wave. The Faraday effect measured amounts to several tens of degrees per centimeter, i.e., it is of the same order of magnitude as the Faraday effect in ferrites at microwave frequencies. A similar effect should be observable in transparent ferromagnets and in sufficiently strongly magnetized paramagnets in a wide frequency interval, 10^9 to 10^{17} cps. It should be mentioned that the detection at very high frequencies of effects associated with the diagonal tensor components of the magnetic susceptibility tensor is practically impossible, since these components decrease in proportion to the square of the frequency.

A Faraday effect due to electronic transitions under the influence of the electric vector of the light wave becomes dominant in the transition from infrared to visible light. It is possible to isolate the wavelength region where the rotation of the plane of polarization evoked by the tensor character of the permeability approximately equals the rotation brought about by the tensor character of its dielectric constant, i.e., the garnet ferrite is a bigyrotropic medium. For some wavelengths these rotations will have the same sign and the total Faraday effect will increase; for others they will be of different sign and the net effect reduces to zero.[5]

The good quantitative agreement of the experimental and theoretical values of $\tilde{\alpha}_F$ indicates that measurements of the Faraday effect in the infrared region can be utilized for the determination of the magnitude, anisotropy, and temperature dependence of the g-factor of rare-earth ions contained in garnet ferrites. All the remaining quantities in Eq. (4) can be determined fairly easily from independent measurements.

2. THE ZEEMAN EFFECT IN THE EXCHANGE FIELD OF THE FERROMAGNET

In this section the results of an investigation of the fine structure of one of the infrared absorption lines of Eu^{3+} ion in the garnet ferrite $Eu_3Fe_5O_{12}$ will be stated. The europium ion was selected for study because its ground level $^7F_0$ is not split by the crystalline field and the fine structure of the line arises only from the splitting of the higher level. According to the energy-level scheme of Eu^{3+},[6] a number of lines corresponding to elec-

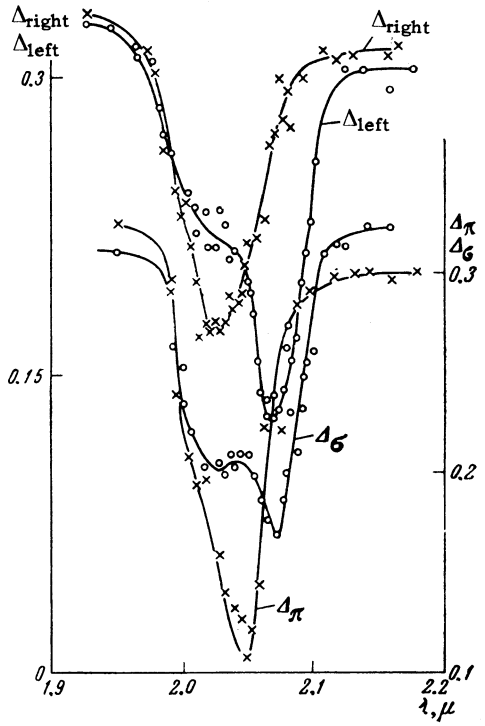

FIG. 2. Transparency of europium garnet plates 100 μ thick in a magnetic field H = 2000 oe (Δ is the ratio of the intensities of transmitted and incident light). Δ_{right} – the case of right-circularly polarized infrared light (electric vector **e** of the light wave rotates clockwise, looking along the light ray), magnetic field directed along the light beam; Δ_{left} – the same for left-circularly polarized light. Δ_{π} – linearly polarized light, **e** $\parallel$ **H**, magnetic field parallel to surface of plate; Δ_{σ} – the same for **e** $\perp$ **H**.

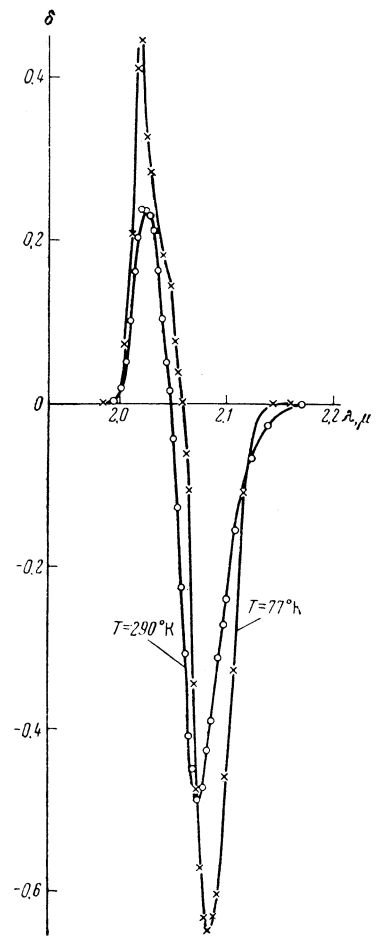

FIG. 3. Circular dichroism $\delta = (\Delta_{right} - \Delta_{left})/\Delta_{H=0}$ of europium garnet in the absorption band region.

tronic transitions between ground term levels should occur in the infrared region. The absorption line corresponding to the transition $^7F_0 \rightarrow {}^7F_6$ should lie close to $\lambda = 2\,\mu$ according to this scheme. We have actually found this line; the polarization of its components turns out to be directly connected with the orientation of the magnetization vector of the garnet ferrite.

The measurements were made in an IKS-11 spectrograph with a LiF prism having a slit width of 0.2 mm, which corresponds to a spectral interval of ~ 40 cm^{-1}. A Fresnel rhomb was used to obtain light of circular polarization. Just as in the remaining cases, the sample had the form of a polished monocrystalline plate 100 μ thick. A lead-sulfide cell cooled to liquid nitrogen temperature was used as the light detector.

Figure 2 shows the results of these measurements. With longitudinal magnetization of europium garnet in a magnetic field of 2000 oe (light directed parallel to the magnetization vector and perpendicular to the surface of the sample), one observes two absorption line components split by 110 cm^{-1}, corresponding to the right and left circular polarizations of the infrared light.

A change in the sign of the magnetization vector of the sample is equivalent to a change in sign of the circular polarization of the light. This assertion is illustrated in Fig. 3 by the curves of the circular dichroism of europium garnet, which were obtained by reversing the magnetizing field. With transverse magnetization of the sample (light directed perpendicular to the sample surface, magnetization vector lies in the plane of the sample) and linear polarization, one observes one component (π) for light with the electric vector parallel to the magnetization of the sample, and two shifted components (σ) of different intensity for light of the other polarization.

Thus, in the resolution obtained, the observed spectrum has the characteristics of a Zeeman triplet with a splitting corresponding to a magnetic field of several hundred thousand oersteds. It is natural to compare this field with the exchange field acting on a europium ion, arising from the iron magnetic sublattices. According to Wolf and

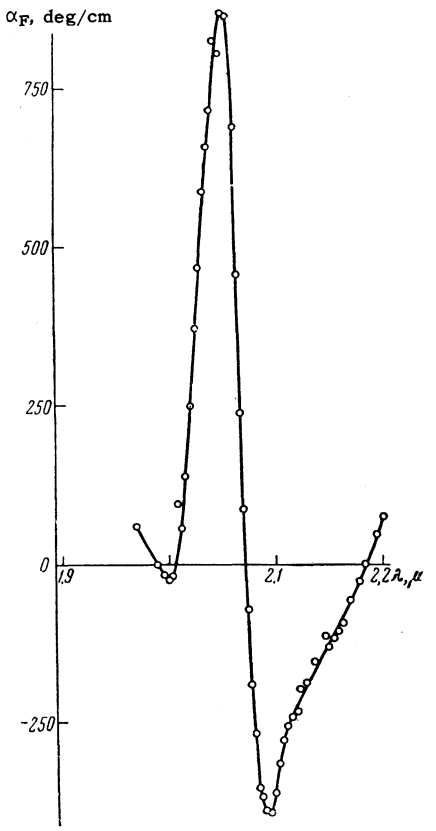

FIG. 4. Faraday effect in europium garnet in the absorption band region.

Van Vleck,[7] this field amounts to $\sim 2.5 \times 10^5$ oe at room temperature. Substituting the values $g = 1.5$, $J = 6$, $H = 2.5 \times 10^5$ into the formula for the total Zeeman splitting of the level of a free ion with given J ($2\Delta E = 2gJ\mu_B H$), we obtain $2\Delta E \approx 210$ cm^{-1}.

Considering that the observed absorption lines reflect a neutralized picture of the Zeeman components displaced to the right and left, the experimental data agrees completely with the quoted estimate. One should not expect too much from such an estimate, since in the development of the theory of the exchange Zeeman effect in ferromagnetic crystals it is necessary to take into account the Stark splitting of the levels, the effects that eliminate the forbiddenness of electronic transitions between f-shell levels, and the selection rules with respect to the crystalline quantum number. A comparison of the curves of circular dichroism at room temperatures (Fig. 3) shows that the splitting is 30% greater at 77° K than it is at 290° K. This fact is consistent with the assumption that the splitting arises from the exchange field, since, according to Pauthenet's data,[8] the magnetization of the iron sublattices in garnet ferrites increases by just this amount in going from room to nitrogen temperature.

On the basis of these results it is possible to reveal one of the physical mechanisms responsible for the magneto-optical properties of ferromagnets. In fact, Fig. 4 shows the results of measurements of the Faraday effect in europium garnet in the vicinity of the absorption line under study. The curve has the typical resonance form, as in other garnet ferrites near the absorption bands.[5] However, in the present case it is possible to point directly to the physical cause of the rotation of the plane of polarization, namely the exchange splitting of an excited energy level of a rare-earth ion. The exchange splitting causes the circular dichroism and double circular refraction, i.e., the difference in the absorption coefficients and refractive indices for right- and left-circularly polarized light, and consequently, also the Faraday effect. In the development of the microscopic theory of magneto-optical effects in ferromagnets it has been clear from the very beginning that the anomalously large magnitude of the magneto-optical effects in ferromagnets was connected with the action of a huge "internal field" of the order of 10^5 to 10^7 oe. However, in all quantitative theories of magneto-optical effects in ferromagnetic metals and semiconductors[9] it has been assumed that this field originates from a spin-orbit interaction. It appears to us that the fundamental role is played not by spin-orbit, but by exchange splitting of the energy levels, at least in the magneto-optics of rare-earth garnet ferrites.* The spin-orbit splitting in this case determines only the natural frequency of the resonance curve of the Faraday effect, since the splitting of the ground term in rare-earth ions is determined by the spin-orbit interaction. It is possible that the suggested mechanism of exchange splitting explains the magneto-optical properties of ferromagnets in other cases as well.

It is necessary to pay attention to one other peculiarity of the phenomenon. In essence, we have the possibility of controlling the appearance of polarized absorption lines in the spectrum of a solid by means of a weak external magnetic field. Because the orbital magnetic moment of the f-shell of a rare-earth ion is bound to the spin, a constant external magnetic field, by changing the orientation of the spontaneous mag-

*We have also detected circular dichroism in the vicinity of the absorption bands in erbium and holmium garnets. The separations between the maxima of the effects of different sign, which depend on the magnitude of the splitting, have the same order of magnitude as in the case of europium garnet. The absolute values of the effect were smaller (~ 3 to 10%) than in the europium garnet, apparently owing to the lack of resolution of the separate components of the absorption bands.

netization vector, alters the "population" of some of the energy levels that play an important role in electrical dipole transitions. Hence, one cannot exclude the possibility that the exchange Zeeman effect will find application in the construction of a kind of molecular amplifier in the optical region or of quantum light indicators.[10] Of importance here is the circumstance that the exchange splitting of the levels is very great, permitting the attainment of energy level "populations" close to 100% at relatively high temperatures.

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