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TRANSPARENT FERROMAGNETS

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opaque to electromagnetic radiation of any wavelength began to change in the middle forties,^[1] after sufficiently high-quality ferromagnetic dielectrics-ferrites with spinel structure-were synthesized. These compounds, and later on their analogs, turned out to be transparent in the radio and microwave bands, and their technical applications led to a rapid growth of physical and technological research on ferrodielectrics. During the last 7-10 years, successful studies were initiated of ferromagnetic dielectrics that are transparent in the literal sense of this word, i.e., they transmit infrared and visible light.^[2] Although transparency is a relative concept, it is customary to regard a substance as transparent if the penetration depth is comparable with the wavelength of the electromagnetic radiation. The list of ferromagnetic compounds having transparency windows of this type is already quite extensive: it includes both ferromagnets (EuO, EuSe, CrBr₃, CrCl₃, CrI₃) and rare-earth ferrimagnets with garnet structure $R_3Fe_5O_{12}$ (R-symbol of trivalent rare-earth ion), and also yttrium iron garnet, ferrimagnets with magnetoactive d-ions (RbNiF₃, RbFeF₃, related physical effects, and also to present a brief dis- $CdCr_2S_4$, $CdCr_2Se_4$), and weak ferromagnets-rare-earth orthferrites RFeO₃. Quite promising results were obtained with respect to practical applications of the degree of transparency: perfect yttrium iron garnet single crystals have in the near infrared an absorption coefficient on the order of 0.07 cm^{-1} .^[3]

Investigations of transparent ferromagnets by optical methods revealed interesting physical effects. Various types of collective exchange resonances in ferromagnets were observed, as well as exchange splitting of the absorption lines of individual magnetoactive ions. The anisotropy of exchange splitting was investigated for the first time and the existence of nonequivalent sites of the rare-earth ions in the iron-garnet lattice, with a local crystal-field symmetry greatly differing from cubic,^[6] has been demonstrated.^[4,5] Besides the usual mechanism of the Faraday effect in ferromagnets, due to spin-orbit interaction, an exchange mechanism was revealed, as well as a mechanism connected with the precession of the magnetic moment of the ferromagnet at optical fre-

 ${f T}_{
m HE}$ traditional notion that a ferromagnet is a substance quencies. In the transparency region of the ferromagnet, where the contribution of the electric dipole transitions is small, it was possible to measure for the first time the magnetic permeability of a ferromagnet at optical frequencies. There exists a region of wavelengths in which the contributions of the tensors $[\epsilon]$ and $[\mu]$ to the Faraday effect are comparable, i.e., the substance is bigyrotropic.^[7] It has been shown that magnetooptical methods can be used to determine the orientation of the magnetic sublattices of ferromagnets.^[8] A shift of the intrinsic absorption edge of ferromagnetic semiconductors under the influence of temperature and external magnetic fields has been observed.^[9] It has been demonstrated that the frequency of the ferromagnetic resonance changes under the influence of infrared radiation. ^[10] By introducing a small amount of rare-earth ions as an indicator, it was possible to reveal, by studying the fine structure of the absorption spectrum of these ions, the character of the exchange interactions for individual neighborhoods in a ferromagnetic crystal.^[11]

The purpose of this review is to consider these and cussion of the prospects of practical application of transparent ferromagnets. We do not touch upon problems pertaining to the optics of transparent antiferromagnets, since they are considered in a separate review.^[12]

II. TRANSPARENCY WINDOWS OF FERROMAGNETIC CRYSTALS

Porter, Spenser, and LeCrow^[2] were the first to report transparency of single-crystal yttrium iron garnet in the wavelength range from 1 to $9\,\mu$. Dillon^[13] described the near-infrared behavior of the absorption coefficient of $Y_3Fe_5O_{12}$. The minimum value of the absorption coefficient is 25 cm⁻¹. By obtaining purer and optically homogeneous crystals, this value could first be reduced to 5 cm^{-1} ,^[14] and later to 0.07 cm⁻¹,^[3,15] at room temperature (Fig. 1). We note that in [3,15], they used Bouguer's law in the form $I = I_0 \cdot 10^{-\alpha'x}$, so that to convert to the customarily employed base e it is necessary to multiply the values given there by 2.3.

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FIG. 2. Diagrams of energy levels of Fe^{3+} ions in iron garnets and orthoferrites. Calculated values of the energy levels of Fe^{3+} in tetrahedral (a) and octahedral (c) sites. Experimental values of the energy levels of Fe^{3+} in iron garnets (b) and orthoferrites (d).



FIG. 3. Absorption coefficient of $Y_3Fe_5O_{12}$ in the region of the absorption edge for different temperatures (a, solid curves) and absorption coefficient of $Y_3Fe_{4.25}Ga_{0.75}O_{12}$ (a, dashed curve). The temperature dependence of the absorption coefficient of $Y_3Fe_5O_{12}$ at 1.06 μ is shown in the insert (b).

As seen from Figs. 1 and 3, the region of maximum transparency of Y_2 Fe₅O₁₂ is $1.2-5 \mu$. The long-wave boundary of the transparency region is connected with the crystal lattice vibrations. The highest frequencies

of the natural oscillations of the tetrahedral group ν_3 in $Y_3Fe_5O_{12}$ correspond to 620 and 680 cm⁻¹. The absorption coefficient for this band is 450 cm^{-1} and gives rise to considerable absorption in the 1000 cm^{-1} region. The absorption band near 1200 cm^{-1} is apparently a combination frequency.^[15] The short-wave boundary of the maximum-transparency region is due to electric-dipole transitions of the Fe³⁺ ions, and also to very strong absorption in the vicinity of 24,000 cm⁻¹, due to the transfer of the electron from the $O^{2^{-}}$ ion to $Fe^{3^{+}}$. A diagram of the energy levels of the $Fe^{3^{+}}$ ions in $Y_3Fe_5O_{12}$ and YFeO, is shown in Fig. 2, where the theoretical calculation results are given together with the experimental results. The absorption edge of $Y_3Fe_5O_{12}$ in the region λ = 1 μ shifts with decreasing temperature towards higher energies (Fig. 3). The position of this edge is strongly influenced by various impurities,^[15] even slight ones. These effects are used to increase the transparency of garnets in the region $\lambda = 1 \mu$.

Transparency windows were also observed for rareearth iron garnets in the far infrared region of the spectrum.^[4,16] The absorption coefficient in the region of maximum transparency reaches 1 cm⁻¹. Rare-earth orthoferrites RFeO₃ with perovskite structure also have a transparency window in the near infrared.^[17,18] The absorption edge lies in the region $\lambda = 1 \mu$, corresponding to the energy level scheme of Fig. 2.

Recently-synthesized single crystals of ferromagnetic compounds turned out to be transparent in the visible region of the spectrum. The ferromagnet $CrBr_3$ ($T_c = 36^{\circ}K$) and the metamagnet $CrCl_3$ ($T_c = 16^{\circ}K$) have absorption edges in the region of 2×10^4 cm⁻¹ (for the isomorphic compound CrI_3 ($T_c = 70^{\circ}K$) the edge is shifted in the region of 10^4 cm⁻¹). ^[19 20] The energy level schemes of the indicated compounds were considered in ^[21]. Smolenskiĭ and co-workers^[22] observed transparency of the ferromagnetic crystal RbNiF₃ in the visible, ultraviolet, and infrared regions of the spectrum (Fig. 4). It was observed in ^[23,24] that the isomorphic compounds RbFeF₃ and RbNi_{1-x}Co_xF₃ are transparent. The absorption coefficient of the foregoing crystals in the region of maximum transparency is ~10 cm⁻¹. In ^[25] they investigated the optical properties of the transparent ferromagnet TiNiF₃ ($T_c = 150^{\circ}K$), and in ^[26] the optical and magneto-optical properties of Na₅Fe₃F₁₄ ($T_c = 193^{\circ}K$).

Busch and co-workers^[27] and Suits and co-workers^[28]



FIG. 4. Absorption coefficient α (cm⁻¹), Faraday effect α F (deg/cm), and magnetic circular dichroism MD of RbNiF₃ at 77°K.

investigated transparent ferromagnetic semiconductors containing ${\rm Eu}^{2*}$ ions (EuO, EuS, EuSe), and also the ferromagnetic semiconductors ${\rm CdCr}_2{\rm S}_4$ and ${\rm CdCr}_2{\rm Se}_4$. The position of the intrinsic absorption edge of these compounds is influenced by the temperature and by the magnetic field. A theory of this shift, based on an allowance for the magnetoelastic effects, was presented by Callen.^[29]

III. RESONANT PROPERTIES OF IRON GARNETS IN THE FAR INFRARED REGION

The energies of the exchange interactions between ions of different magnetic sublattices in ferrimagnets are of the order of $10-200 \text{ cm}^{-1}$, and the magnetic dipole transitions corresponding to the exchange splitting lie in the far infrared region of the spectrum. Investigations of the absorption spectra of ferromagnets in the far infrared region of the spectrum were carried out by Tinkham and Sievers.^[4,30,31] These investigations helped interpret to a considerable degree the near-infrared absorption spectra of $Yb_3Fe_5O_{12}$ described in Ch. VI. In magnetic systems consisting of more than one magnetic sublattice, there should take place, besides the single-ion exchange splitting of the energy levels and collective oscillations of the magnetic moment (ferromagnetic resonance), also one more type of collective oscillation, namely exchange resonance, which was predicted by Kaplan and Kittel.^[32] The frequency of exchange resonance for a ferrimagnet consisting of two magnetic sublattices was obtained from the Landau-Lifshitz equation and has the following form:

$$\omega_{\mathbf{ex}} = \lambda \left(\gamma_2 M_1 - \gamma_1 M_2 \right); \qquad (1)$$

where λ -constant of molecular field, $\gamma_1 = g_1 e/2mc$ and $\gamma_2 = g_2 e/2mc$ -gyromagnetic ratios, M_1 and M_2 -sublattice magnetizations. For most ferrimagnets, this frequency lies in the far infrared. Physically this is explained as follows. In ordinary ferrimagnetic resonance the precession of the summary magnetic moment of the ferrimagnet occurs under the influence of only the external magnetic field, and the resonant frequency lies in the microwave band. In antiferromagnetic resonance, the sublattices are acted during part of the precession period upon by a magnetic field H_m , and during part of the period only by the anisotropy field H_k ; the resonant frequency, which is proportional to $\sqrt{H_m H_k}$, falls in the submillimeter band. In the case of exchange resonance, the angle between the sublattices is not equal to zero during the entire precession period, and the resonant frequency is proportional to H_{m} = λM and shifts to the infrared region, i.e., to the region where the natural frequencies of the resonant single-ion exchange magnetic dipole transitions should be located. In the case of exchange resonance, the magnetic-susceptibility component $\kappa_{\rm X}$ should take the form $^{[32]}$

$$\kappa_{x} = \frac{M_{1x} - M_{2x}}{H_{x}} = \frac{\lambda M_{1} M_{2} (\gamma_{1} - \gamma_{2})^{2}}{\omega^{2} - \omega^{2}_{ex}} .$$
 (2)

It follows from (1) and (2) that exchange resonance can be observed in an isotropic ferrimagnet under the condition $\gamma_1 \neq \gamma_2$.

The absorption spectra of single-crystal and polycrystalline $Yb_3Fe_5O_{12}$ samples were investigated in ^[4] in the interval from 6.6 to 100 cm⁻¹ at temperatures from 2 to 70°K in a magnetic field of 11 kOe, and exchange resonances of various types were observed in



FIG. 5. Temperature dependence of the strongest absorption lines of $Yb_3Fe_5O_{12}$ in the far infrared. \bullet – splitting of the ground state of the Yb^{3+} ion at two-non-equivalent positions in the garnet lattice in the exchange field produced by the magnetic sublattices of the iron (experiment); O – exchange resonance in $Yb_3Fe_5O_{12}$ (experiment; dashed curve – exchange resonance (theory without allowance for anisotropy); solid curves – exchange resonance (theory with allowance for anisotropy). The lowest solid curve corresponds to the ferrimagnetic-resonance frequency.

ferrimagnets for the first time. Three strong absorption lines $(14.1 \pm 0.2 \text{ cm}^{-1}, 23.4 \pm 0.3 \text{ cm}^{-1}, 26.4 \pm 0.3 \text{ cm}^{-1})$ and one weak one $(24.6 \pm 0.6 \text{ cm}^{-1})$ were observed in the absorption spectrum of a polycrystalline $\text{Yb}_3\text{Fe}_5\text{O}_{12}$ sample 1.47 mm thick at 2°K. The temperature dependence of the three strongest absorption lines is shown in Fig. 5. The positions of the 26.4 and 23.4 cm⁻¹ lines do not depend on the temperature. These lines correspond to splitting of the ground state of the Yb³⁺ ion in two nonequivalent positions in the exchange field produced by the iron magnetic sublattices. The indicated values of the exchange splitting of the ground state of Yb³⁺ are close to the values obtained from investigations of the absorption spectra in the near infrared (see Ch. VI).

The third line in Fig. 5 has a temperature dependence that is quite close to the theoretical temperature dependence of the exchange-resonance frequency in the isotropic approximation (dashed curve). To obtain this curve, it was assumed that the g-factor of the Yb^{3+} ion of the rare-earth sublattice is equal to 24/7 and differs greatly from the value of the g-factor of the free ion, which equals 8/7. This difference should occur for all the rareearth ions at low temperatures. The theory of the aniso-tropy of the g factor of the Yb^{3+} ion in $Y_3Ga_5O_{12}$ and $Y_3Al_5O_{12}$ is considered in ⁽³³⁻³⁶⁾. The experimental values of the g tensors of Yb^{3+} in $Y_3Ga_5O_{12}$ and $Y_3Al_5O_{12}$ were obtained in ^[37-38]. At low temperatures, iron garnets are characterized by a very large magnetic crystalline anisotropy,^[41] allowance for which is essential in order to obtain better agreement between the theoretical and experimental $\omega_{ex}(T)$ relations. An attempt to take the anisotropy into account was made in ^[4]. In this case $\omega_{ex}(T)$ as represented by the solid curve in the central part of Fig. 5 is in much better agreement with the experimental results for $Yb_3Fe_5O_{12}$. The very lowest curve in Fig. 5 is the temperature dependence of the ferrimagneticresonance frequency of this garnet. This resonant fre-quency was observed by Richards,^[13] who obtained a higher resolution and observed also a combination resonant frequency corresponding to simultaneous excitation

of exchange and ferrimagnetic resonances. The absorption spectra of rare-earth iron garnets in the far infrared were the subject of a number of investigations, $^{(4,30,31)}$ and a temperature dependent exchange resonance was observed in samarium, holmium, and erbium garnets. The exchange-resonance frequencies in the indicated ferrites at T = 2°K are respectively equal to 33.5 ± 0.5 , 38.5 ± 0.5 , and 10 ± 0.2 cm⁻¹.

In addition to the aforementioned garnets, $Gd_3Fe_5O_{12}$ was also investigated in the far infrared region, but no exchange resonance was observed. The authors of ^[30] attribute this to the fact that the γ factors of Gd^{3+} and Fe^{3+} are close in magnitude, and the intensity of the exchange resonance becomes very small in accordance with (2).

According to Neel,^[40] rare-earth iron garnets consist, from the magnetic point of view, of three collinear magnetic sublattices. Two are made up of the Fe³⁺ ions, which occupy tetrahedral and octahedral interstices in the lattice. The magnetic moments of these sublattices are antiparallel. The third sublattice is made up of the rare-earth ions located in the dodecahedral interstices. ^[41] The magnetic moment of this sublattice is antiparallel to the total moment of the iron sublattices. In the analysis of the results of investigations with the exchange resonance in all the aforementioned garnets, both Fe³⁺ sublattices were regarded as a single sublattice, since the exchange interaction between them is large and the gfactors are close to two. The second sublattices were taken to be the rare-earth sublattice. A different situation arises apparently in $Eu_3Fe_5O_{12}$ at low temperatures. The Eu^{3+} ion is in the ground state ${}^{7}F_{0}$, has J = 0, and its entire magnetic moment is induced by the exchange interaction with the tetrahedral iron sublattice.[42] In this connection, the authors of ^[46] assume that in the analysis of the results of the investigation of the exchange resonance in $Eu_3Fe_{5-X}Ga_XO_{12}$ it is necessary to consider two magnetic sublattices of this garnet: 1) Eu^{3+} and Fe^{3+} in tetrahedral interstices, 2) Fe^{3+} in octahedral interstices. The dependence of the exchange-resonance frequency in $Eu_{3}Fe_{5-X}Ga_{X}O_{12}$ on x at $4.2^{\circ}K$ is shown in Fig. 6 and is in fair agreement with the expected theoretical values. The exchange field for the two iron sublattices is equal to 500 kOe/ μ B.

IV. MAGNETIC-PERMEABILITY TENSOR OF FERRO-MAGNETIC DIELECTRICS IN THE OPTICAL FRE-QUENCY BAND

The applicability of the Landau-Lifshitz equation^[43] to the description of the dynamic properties and the magnetic permeability of ferromagnets is not limited to the far infrared region, and extends apparently up to the optical band. The proof of this statement is the result of an investigation of the Faraday effect in iron garnets in the wavelength interval from 1 to 9μ .

The Faraday effect in $Y_3Fe_5O_{12}$ was investigated in ^[44] in the wavelength range from 1 to 9 μ . With increasing wavelength, the specific rotation of the plane of polarization α_F (deg/cm) first decreases rapidly, and then stays constant from 4.5 to 9 μ . A similar result was obtained somewhat later in the wavelength region up to 6 μ in ^[45]. The quantitative value of α_F in $Y_3Fe_5O_{12}$ was made more



FIG. 6. Exchange-resonance frequency (in cm⁻¹) of $Eu_3Fe_{5-X}Ga_XO_{12}$ as a function of the Ga contents x at T = 4.2°K. \bullet , \blacktriangle – experiment, dashed line – theory with allowance for anisotropy. Observations in the shaded regions are hindered by the absorption of the radiation by the garnet lattice vibrations.



FIG. 7. Faraday effect in iron garnets of yttrium (Y), erbium (Er), and holmium (Ho) in the infrared region of the spectrum at $T \approx 290^{\circ}$ K and in yttrium iron garnet at $T = 77^{\circ}$ K.

precise in ^[7], where similar investigations were made also on $\text{Er}_5\text{Fe}_5\text{O}_{12}$ and $\text{Ho}_3\text{Fe}_5\text{O}_{12}$. It turned out that in all these garnets at room temperature, and in $\text{Y}_3\text{Fe}_5\text{O}_{12}$ also at 77°K, α F is independent of the wavelength if $\lambda > 4.5 \mu$, and amounts to several dozen degrees per centimeter (Fig. 7). It was shown in ^[7] that the frequency-independent rotation of the plane of polarization in ferrimagnets is determined by the nondiagonal component of the magnetic-permeability tensor at infrared frequencies, and is the consequence of ferromagnetic and exchange resonances.

If we denote the components of the magnetic susceptibility tensor by $\kappa_{XX} = \kappa_{yy} = \kappa$, $\kappa_{XY} = -\kappa_{yX} = iG$, and the susceptibility for the right- and left-circularly-polarized waves, for which the susceptibility tensor can be reduced to diagonal form, by $\kappa_{\pm} = \kappa \pm G$, then we obtain from the Landau-Lifshitz equations of motion of the magnetic moment^[43] in the absence of damping

$$\varkappa_{\pm} = \frac{\gamma M}{\omega_0 \mp \omega}$$

where M-saturation magnetization of the ferromagnet, γ -gyromagnetic ratio, and $\omega_0 = \gamma H$ -the proper spinprecession frequency. The values of the susceptibility components at $\omega \gg \omega_0$ are respectively

$$\mathbf{x}_{\pm} = \pm \frac{\gamma M}{\omega}, \quad \mathbf{x}_{xy} = i \frac{1}{2} \left(\mathbf{x}_{+} - \mathbf{x}_{-} \right) = -i \frac{\gamma M}{\omega}, \quad \mathbf{x}_{xx} = \frac{\mathbf{x}_{+} + \mathbf{x}_{-}}{2} = -\frac{\omega_0 \gamma M}{\omega^2}.$$

The specific rotation of the polarization plane $\alpha_{\rm F}$ is equal

(5)

to

$$\alpha_{\mathbf{F}} = \frac{2\pi \sqrt{\epsilon}\omega}{c} (\varkappa_{-} - \varkappa_{+}) = \frac{2\pi \sqrt{\epsilon}}{c} \gamma M.$$

Thus, $\alpha_{\rm F}$ does not depend on ω when $\omega \gg \omega_0$, and by determining experimentally the frequency-independent part of $\alpha_{\rm F}$ we can measure the magnetic susceptibility for right- and left-polarized waves in the optical range, which is equivalent to determining $\kappa_{\rm XY}$. It is much more difficult to measure the diagonal component of the magnetic susceptibility $\kappa_{\rm XX}$ in the optical range, since, as shown above, $\kappa_{\rm XX}$ is inversely proportional to ω^2 .

A similar situation occurs for the frequency dependence of the magnetic susceptibility for any magnetic resonance connected with spin precession.

Thus, for example, it follows from the equations of motion of the magnetic moment of a two-sublattice ferrimagnet under the condition $\omega \gg \omega_0$, ω_{ex} , ω_{rel} , where ω_0 , ω_{ex} , and ω_{rel} are the frequencies of the ferrimagnetic resonance, the exchange resonance, and the relaxation, respectively, that the off-diagonal component of the magnetic-susceptibility tensor, with allowance for the ferrimagnetic and exchange resonances, is given by

$$\varkappa_{xy} = -\frac{i}{m} \left(\gamma_1 M_1 - \gamma_2 M_2 \right). \tag{3}$$

where $\kappa_{\rm XY}$ does not depend on the magnetic crystallographic anisotropy and the shape anisotropy. The inequality employed above are satisfied at optical frequencies with a large margin, since $\omega_0 \sim 10^9 \sec^{-1}$, $\omega_{\rm eX} \sim 10^{12}$ \sec^{-1} , and $\omega_{\rm rel} \sim 10^7 - 10^{12} \sec^{-1}$. The specific rotation of the polarization plane $\alpha_{\rm F}$, with allowance for (3), is given by

$$\alpha_{\mathbf{F}^{-2}} \frac{2\pi |\mathcal{V}^{\mathbf{F}}|}{c} (\gamma_1 M_1 - \gamma_2 M_2); \tag{4}$$

here M_1 and M_2 , γ_1 and γ_2 are respectively the magnetizations and the γ -factors of the iron and rare-earth sublattices of the ferrimagnet, and $\sqrt{\epsilon} = n$ is the refractive index. The frequency dependence of n for $Y_3 Fe_5 O_{12}$ was obtained in ^[46], where n turned out to be close to 2.2 in the entire transparency region.

For yttrium, erbium, and holmium iron garnets,^[7] the experimental values of $\alpha_{\rm F}$, which do not depend on ω , are in good agreement with (4). Expression (4) was used also earlier in the microwave band, but its validity in the optical band was demonstrated in ^[7]. We note that expression (3) for a ferromagnet with one magnetic sublattice was first used for the optical band in ^[49] to calculate the Faraday effect and the polar Kerr effect in ferromagnets. Following ^[48], we can rewrite (4) in the form

where

$$M = M_1 - M_2$$
, $\gamma_{\text{eff}} = \frac{M_1 - M_2}{\frac{M_1}{\gamma_1} - \frac{M_2}{\gamma_2}}$

 $\alpha_{\rm F} = \frac{2\pi \, \sqrt{\epsilon}}{c} \left[\gamma_{\rm eff} \, M - \frac{\gamma_{\rm eff} \, M_4 M_2 \, (\gamma_1 - \gamma_2)^2}{\gamma_1 \gamma_2 M} \right] \, , \label{eq:aF}$

and the contributions of the ferrimagnetic (first term) and exchange (second term) resonances to α_F in the infrared region of the spectrum now become clear. In the iron garnets considered above, α_F is positive in both the visible and in the infrared regions of the spectrum. There exist substituted garnets for which $\alpha_F < 0$

in the visible region. Such a garnet, for example, is $Bi_{0.24}Ca_{2.76}Fe_{3.63}V_{1.38}O_{12}$. It was shown in ^[50] that, starting with 4 μ , in this garnet $\alpha_{\rm F} > 0$ and does not depend on λ , in accord with (4). Thus, the wavelength-independent $\alpha_{\rm F}$ in ferrimagnets is due to precession of the magnetic moment in the field of the light wave. The off-diagonal components of the tensor $[\mu]$ of $Y_3Fe_5O_{12}$ and of certain rare-earth iron garnets, as obtained from investigations of the Faraday effect, are given in ^[51]. μ_{XY} are equal to $10^{-4} - 10^{-5}$ when $\lambda = 5 \mu$, and vary linearly with λ . From the obtained values of μ_{XY} it can be concluded that the difference between the diagonal components μ_{XX} and μ_{YY} and unity amounts to $10^{-9} - 10^{-10}$. In the wavelength interval from 1 to 4.5μ , the iron garnets are bigyrotropic media, since the off-diagonal components of the tensors [ϵ] and [μ], and the corresponding rotations of the plane of polarization, are comparable in magnitude. It was indicated in [7] that the frequencyindependent Faraday effect should be observed also in strongly magnetized paramagnets. This effect was observed in MnF₂ in strong magnetic fields in the paramagnetic and antiferromagnetic states.^[52]

In formula (4), all the quantities with the exception of $\gamma_1 = g_1 e/2mc$ and $\gamma_2 = g_2 e/mc$ can be obtained from independent measurements, and therefore this formula can be used to determine the g-factors of rare-earth ions of ferrimagnets in a wide range of temperatures. This method of determining g-factors was proposed in ^[7] and used in ^[53-55] to determine the g-factors of the ions Dy^{3*} , Tm^{3*} , Tb^{3*} , and Eu^{3*} . It was shown in ^[53] that for a quantitative reconciliation of α_F^{exp} with (4) for terbium and dysprosium iron garnets at $T = 100^{\circ}$ K it is necessary to assume that the g-factors of the ions Tb^{3*} and Py^{3*} are equal to 0.8-0.9, whereas $g_T^{Tb} = 1.5$ and $g_D^{Ty} = 1.33$. Johnson and Teble^[54] have shown that in the tempera-

Johnson and Teble¹³⁴ have shown that in the temperature interval from 80 to 300°K the g-factor of Tm³⁺ changes from 1.36 to 1.64, and the g-factor of the Dy³⁺ ion changes from 0.67 to 1.06, thereby differing greatly, from the g-factors of the free ions ($g_J^{Tm} = 7/6$).

It was found in ^[55] by the same method that the gfactor of Tb³⁺ in Tb₃Fe₅O₁₂ changes in the interval 100– 350°K in the range from 1.0 to 1.15. It is proposed in ^[56] that the g-factors of the rare-earth ions in iron garnets should be assumed to be close to g_J, and the aforementioned differences between $\alpha_{\rm F}$ and the values calculated by formula (4) are attributed to the contribution of the single-ion exchange resonance (see Ch. III) to the frequency-independent Faraday effect. It follows from (4) that besides the reversal of the sign of $\alpha_{\rm F}$ at the compensation point^[57,53,54] there should take place also one more sign reversal of $\alpha_{\rm F}$ at a lower temperature, when $\gamma_1 M_1 = \gamma_2 M_2$, since $\gamma_1 > \gamma_2$ and M₂ increases much more strongly with decreasing temperature than M₁.

The temperature dependence of the frequency-independent Faraday effect $\alpha_{\rm F}$ in Tb₃Fe₅O₁₂ was investigated in ^[55] in the range from 25 to 350°K (Fig. 8). The Faraday effect reverses sign at the compensation point. With further decrease of temperature, $\alpha_{\rm F}$ decreases in absolute magnitude and at T = 110°K it reverses sign once more, after which it again increases. The obtained dependence agrees qualitatively with (4) if one uses for g₂ its value for the free ion (dashed curve of Fig. 8). It should be noted that (3) and (4) do not depend on the shape anisotropy and on the magnetic crystallographic aniso-



FIG. 8. Temperature dependence of the frequency-independent Faraday effect in Tb₃Fe₅O₁₂ at a wavelength 6.5 μ . \bullet – experiment, dashed line – theory, gJ^b = 1.5.

tropy. Therefore the indicated method can be used to investigate the anisotropy and the g factors of the rareearth ions in ferrimagnets. An attempt to investigate the anisotropy of the Faraday effect in Tb₃Fe₅O₁₂ was made in ^[58]. It was noted above that when $1 < \lambda < 4 \mu$ iron garnets are bigyrotropic media, i.e., the magnetooptical effect in them are determined to an approximately equal degree by the off-diagonal components of the tensors $[\epsilon]$ and $[\mu]$. A similar situation is expected to arise also in other ferromagnets. It thus becomes necessary to determine and to separate the components of these tensors. Gintsburg^[59] has shown that with the aid of the polar Kerr effect and the Faraday effect it is possible to determine the off-diagonal components of the tensors $[\epsilon]$ and $[\mu]$. The analogous formulas given in Sokolov's book^[60] for the polar Kerr effect in a bigyrotropic medium are in error. Although the procedure of [59] is beyond reproach, its implementation calls for the use of two samples of the same compound (a relatively thin one for the Faraday effect and a thick one for the Kerr effect). This apparently is the reason for the failure of the investigations [61,62] in which the authors attempted to determine by this method the off-diagonal components of the tensors $[\epsilon]$ and $[\mu]$. In this sense, it is preferable to use a procedure

that makes it possible to determine the foregoing components from investigations of the equatorial Kerr effect using s- and p-polarized light.^[63, 64, 653]

It was shown in ^[63,64] that for media characterized by tensors $[\epsilon]$ and $[\mu]$ in the form

$$\begin{split} & \varepsilon_{xx} = \varepsilon_{yy} = \varepsilon, \quad \varepsilon_{xy} = -\varepsilon_{yx} = -i\varepsilon \mathscr{M}, \quad \varepsilon_{zz} = \varepsilon_0, \\ & \mu_{xx} = \mu_{yy} = \mu, \quad \mu_{xy} = -\mu_{yx} = -i\mu \mathscr{M}', \quad \mu_{zz} = \mu_0, \end{split}$$

the reflection coefficients of s- and p-waves in the equatorial Kerr effect are equal to

$$\frac{R_p}{4_p} = \frac{\alpha n - \alpha^* \mu_0}{\alpha n + \alpha^* \mu_0} - 2i \mathscr{M} \frac{\alpha \beta \mu_0}{(\alpha n + \alpha^* \mu_0)^2} , \qquad (6a)$$

$$\frac{\dot{R}_{s}}{A_{s}} = \frac{\alpha n - \alpha^{*} \varepsilon_{0}}{\alpha n + \alpha^{*} \varepsilon_{0}} - 2i \mathscr{M}' \frac{\alpha \beta \varepsilon_{0}}{(\alpha n + \alpha^{*} \varepsilon_{0})^{2}}; \qquad (6b)$$

here α , β , and α^* are the direction cosines of the incident and refracted waves, respectively, and $n = \sqrt{\epsilon_0 \mu_0}$ is the refractive index of the unmagnetized medium.

From these formulas it is possible to obtain an expression for the change in the intensity of the reflected light upon reversal of magnetization, $\delta = (I - I_0)/I_0$, where I and I_0 is the intensity of the light reflected from the magnetized and unmagnetized ferromagnet. The value of δ is usually measured experimentally.

Thus, from the equatorial Kerr effect on s- and p-

waves it is possible to determine the off-diagonal components of the tensors [ϵ] and [μ]. In ^[65,653] the offdiagonal component of the tensor [μ] of Fe at optical frequencies was determined in accordance with (3), and it was shown that the results of ^[61,62] are in error. The relative change of the intensity of the s-component light reflected in the equatorial Kerr effect is 10⁻⁵, in accord with (6b). The scale of $\delta_{\rm S}$ in Fig. 1 of ^[65] is indicated incorrectly: it should read $\delta_{\rm S} \times 10^5$ instead of $\delta_{\rm S} \times 10^3$.

V. ABSORPTION LINES OF RARE-EARTH IONS IN FERROMAGNETIC CRYSTALS

It is easy to observe the absorption lines of rareearth ions in the transparency region of ferromagnetic crystals, and in the near infrared and in the visible regions these lines correspond to electron transitions within the 4f shell.^[66] For this reason, the lines are sufficiently narrow, making it possible to investigate a number of features connected with the magnetic ordering of the investigated crystal. The first to observe the fine structure of the absorption band of the Yb³⁺ ion, corresponding to the transition ${}^{2}F_{7/2} \rightarrow {}^{7}F_{5/2}$ in Yb₃Fe₅O₁₂ were Wickersheim and White in the region $\lambda = 1 \mu$. An exchange splitting of the ground and excited levels of the Yb³⁺, due to the exchange interaction of the rare-earth and iron sublattices, was observed, and the existence of non-equivalent locations of rare-earth ions in the garnet lattice was demonstrated for the first time. A relatively simple absorption spectrum, similar to the spectrum of Yb³⁺ ions which have one hole in the 4f shell, should be possessed by compounds with Ce^{3+} ions, since the 4f shell of this ion has only one electron. The cerium iron garnet does not crystallize, owing to the large ionic radius of Ce^{3+} . Wickersheim and Buchanan^[11] introduced 1% of ${\rm Ce}^{3+}$ ions into yttrium iron garnet in order to study the character of the exchange interaction between the iron and rare-earth sublattices. The exchange splitting of the 2.7- μ line of cerium is anomalously large (~0.2 μ), making it easy to investigate the fine structure of this line and even to estimate the relative intensities of the main lines and the weak satellite lines. When indium, scandium, and chromium ions are introduced into $Y_3Fe_5O_{12}$ and replace octahedral iron ions, the character of the exchange splitting of the 2.7- μ line remains unchanged. When gallium and aluminum ions are introduced in tetrahedral sites, additional satellite lines appear. The authors of [11] therefore concluded that the main contribution to the exchange field acting on the rare-earth ion in the iron garnet is made by the tetrahedral sublattice of the iron ions. Moreover, by identifying definite satellite lines with the concrete type of substitution of a tetrahedral iron ion by a nonmagnetic ion in the first coordination sphere of Ce^{3+} and estimating the intensities of these lines, it is concluded that out of the six tetrahedral Fe³⁺ ions, the two closest ones give 55% of the exchange interaction, the remaining 45% comes from the remaining four ions.

A comparatively simple absorption spectrum should be possessed also by europium iron garnets, since the ground level of the magnetoactive Eu³⁺ ions has zero angular momentum and is not split by the exchange and crystal field.

The fine structure of the absorption band of Eu^{3+} in $Eu_3Fe_5O_{12}$ was investigated in ^[7,67] for the transitions

 ${}^{7}F_{0} \rightarrow {}^{7}F_{6}$ and ${}^{7}F_{0} \rightarrow {}^{7}F_{4}$, and principal attention was paid to the polarization of the individual components of the absorption band at different orientations of the magnetizing field.

The results of the investigations of the absorption spectrum of $\text{Eu}_{3}\text{Fe}_{5}O_{12}$ in the $\lambda = 2 \mu$ region, correspond-ing to the transition ${}^{7}\text{F}_{0} \rightarrow {}^{7}\text{F}_{6}$, are shown in Fig. 9. In the case of longitudinal magnetization (infrared radiation propagates along the magnetization vector), two components of the absorption line are observed. They correspond to right-hand and left-hand circular polarization and are spaced 110 cm⁻¹ apart. In the case of transverse magnetization and linear polarization of the radiation, one component (π) is observed for radiation with electric vector e parallel to the magnetization, and two shifted components (σ) for radiation with electric vector **e** perpendicular to the magnetization. Thus, the observed picture can be interpreted as an exchange Zeeman triplet on an electric dipole transition from the ground state to one of the crystal components of the excited level. A comparison of the circular-dichroism curves at room temperature and 77°K shows that following the indicated temperature drop the splitting of the ${}^{7}F_{0} \rightarrow {}^{7}F_{4}$ line decreases by 30%, in accordance with the magnetization of the iron sublattice.^[68,69] Investigations of the temperature dependence of the absorption spectra of Eu₃Fe₅O₁₂ in the region of the ${}^{7}F_{0} \rightarrow {}^{7}F_{4}$ transition of the Eu³⁺ ion are reported in ${}^{[67]}$, where the following values were obtained for the splitting of the ${}^{7}F_{4}$ level as a function of the temperature:

$$\Delta E = 75 \text{ cm}^{-1} (T = 375^{\circ} \text{ K}), \quad \Delta E = 85 \text{ cm}^{-1} (T = 290^{\circ} \text{ K}), \quad \Delta E = 140 \text{ cm}^{-1} (T = 400^{\circ} \text{ K}).$$

The splitting of the ${}^{7}F_{4}$ level at 290°K is smaller than the corresponding splitting of ${}^{7}F_{6}$. The results agree with the assumption that the investigated splittings are due to the exchange field acting on the Eu³⁺ ion and due to the Fe³⁺ ions of the garnet.

The absolute value and sign of this field was determined in ^['i] by measuring the dependence of the splitting of the ⁷F₄ level on the external field. At H = 25 kOe, the splitting decreased from 92 to 80 cm⁻¹. Consequently, the exchange field is directed opposite to the external field and its magnitude is 220 ± 30 kOe. The measurement of the exchange field in Dy₃Fe₅O₁₂ by this method is reported in ^['1]. The value obtained was H_{ex} = 150 \pm 50 kOe.

The Faraday effect in $\operatorname{Eu}_3\operatorname{Fe}_5\operatorname{O}_{12}$ was investigated in ^[7] in the region of the ${}^7\operatorname{F}_0 \rightarrow {}^7\operatorname{F}_6$ absorption band of the Eu³⁺ ion. The rotation has a resonant character and reaches a rather large value, 10³ deg/cm (Fig. 10). The frequency dependence of $\alpha_{\rm F}$ is obtained directly from the exchange components of the circular dichroism both for transitions ${}^7\operatorname{F}_0 \rightarrow {}^7\operatorname{F}_6$ [^[72] and for ${}^7\operatorname{F}_0 \rightarrow {}^7\operatorname{F}_4$, [^{67]} so that it can be stated that an important role in the magnetooptics of rare-earth compounds is played not by the spinorbit but by the exchange splitting of the energy level. [^[7, 72] Linear dichroism in $\operatorname{Er}_3\operatorname{Fe}_5\operatorname{O}_{12}$ was used in [^[73] to determine the dimension of the spin-

Linear dichroism in $\text{Er}_3\text{Fe}_5\text{O}_{12}$ was used in ^[73] to determine the direction of the magnetic moment and the constants of the magnetic crystallographic anisotropy of this compound at 4.2° K (see also ^[12]).

An investigation of the exchange Zeeman triplet in the ${}^{7}F_{0} \rightarrow {}^{7}F_{4}$ absorption band using circularly-polarized light^[67,70] revealed rather characteristic additional ab-



FIG. 9. Absorption spectra of Eu₃Fe₅O₁₂ in the region of the absorption band ${}^{7}F_{0} \rightarrow {}^{7}F_{6}$ for circularly-polarized light ($\Delta_{right}, \Delta_{left}$) in the case of longitudinal magnetization and linearly-polarized radiation with electric vector parallel to the magnetization (π component), and with electric vector perpendicular to the magnetization (σ component).

arF, deg/cm

251

-250



FIG. 11. Form of the ${}^{7}F_{0} \rightarrow {}^{7}F_{4}$ absorption band of the Eu³⁺ ion in Eu₃Fe₅O₁₂ for right- and left-polarized (a, b) light and for longitudinal magnetization of the crystal along the [111] axis.



sorption lines. Figure 11 shows the form of the ${}^{7}F_{0} \rightarrow {}^{7}F_{4}$ absorption band for right- and left-circularly-polarized light and longitudinal magnetization of an Eu₃Fe₅O₁₂ plate along the [111] axis. The intense lines A and D in Fig. 11

correspond to the main components of the exchange Zeeman triplet, i.e., to transitions from the level A_0 to the levels $F_{\pm 1}$. The indices 0 and ± 1 denote the magnetic quantum numbers m_s of the ground level $A_0({}^{o}F_0)$ and triplet level $F_{0,\pm 1}({}^{7}F_{4})$. Thus, for example, the low-energy component A for right-polarized light corresponds to a transition to the lower level of the exchange triplet F_{+1} (state with magnetic moment oriented parallel to the magnetization of the rare-earth sublattice of the iron garnet). The hypothesis was advanced that the appearance of the remaining less intense lines is due to excitation of the spin waves in optical electronic transitions, i.e., they are combination spin-wave lines. Thus, for example, the line B was regarded as the electronic transition $A_0 \rightarrow F_+$ under the influence of left-polarized light with simultaneous excitation of a spin wave at the exchangeresonance frequency. The interval $\omega_A - \omega_B$ corresponds in order of magnitude to the Kaplan-Kittel exchangeresonance frequency.

It was also shown that the appearance of additional absorption lines (at least the lines B and C) is not the consequence of poor circular polarization of the light. Agreement between the frequency dependence of the Faraday effect calculated by the method of ^[72] and the experimental curve was obtained only when account was taken of additional absorption lines. Thus, this was the first experimental observation of magnetic or spin-wave absorption sidebands in a magnetically-ordered crystal -this is how the indicated additional lines were designated after they were observed in antiferromagnetic crystals^[74] (for details see the review ^[12]). It should be noted that although the possibility, in principle, of the appearance of spin-wave satellite lines in ferrimagnetic crystals is no longer subject to any doubt at present,^[75] the question of the nature of such lines and their reliable identification has been fully investigated only for antiferromagnets. This calls for a thorough theoretical study of the features of optical transitions in antiferromagnetic crystals, and for a large volume of experimental investigations. Work of this kind on ferrimagnetic crystals is still to be performed. Taking into account the experience in the study of single- and twomagnon transitions in antiferromagnets, one can attempt to identify concretely the additional lines in europium iron garnet for both longitudinal and transverse magnetization.

Assuming that the energy of the exchange magnon in the europium garnet is equal to approximately 15 cm⁻ we can interpret the line B as a two-magnon absorption line in the $A_0 \rightarrow F_{+1}$ transition, corresponding to the production of two magnons, and the line C as a sideband line in the $A_0 \rightarrow F_{-1}$ transition, corresponding to annihilation of two magnons. It is easy to see that such an identification of the transitions B and C satisfy simultaneously both the energy and the angular-momentum conservation laws. From this point of view, we can also interpret the complicated picture observed in linearly polarized light when the samples are transversely magnetized. The two middle lines, which are shifted relative to the center of the band by approximately 15 cm⁻¹, and which were at-tributed in (70) to the presence of nonequivalent sites for the rare-earth ions in the garnet sublattice, can be regarded as single-magnon satellite lines in the $A_0 \rightarrow F_0$ transition, and the splitting of the lines A and D, observed with decreasing temperature, also by approximately 15 cm⁻¹, can be regarded as a manifestation of single-magnon satellites in the transitions $\tilde{A}_0 \rightarrow F_{\pm 1}$. It should be noted that inasmuch as the interval $\hbar\omega_A - \hbar\omega_D \approx 90 \text{ cm}^{-1}$, and $\hbar\omega_e \approx 15 \text{ cm}^{-1}$, certain single-magnon and two-magnon satellite lines should coincide with frequency, which also makes their identification difficult. Thus, for example, the long-wave single-magnon satellite in the $A_0 \rightarrow F_0$ transition obviously coincides in frequency with the short-wave two-magnon satellite in the $A_0 \rightarrow F_{+1}$ transition, etc.

Hufner and co-workers^[76,77] performed a low-temperature spectroscopic investigation of rare-earth perovskites, including both weak ferromagnets (DyFeO₃, HoFeO₃, and $ErFeO_3$) and pure antiferromagnets (TbAlO₃ and $DyAlO_3$). The orientation of the magnetic moments of the rare-earth ions relative to the crystallographic axes was determined. A temperature-independent magnetic splitting of the spectral lines in the paramagnetic region (for rare-earth ions) was observed and interpreted as a result of a short-range interaction. Below the Neel point there appears an additional splitting of the order of 5 cm^{-1} , which increases with decreasing temperature and is due to long-range magnetic order. The magnetic moments of the rare-earth ions in the ground state were determined from the Zeeman effect. A somewhat unexpected conclusion was drawn that both in orthoaluminates and in orthoferrites the main role in magnetic ordering is played by the interaction of the rare earth ions with one another, this interaction being magnetic-dipole in orthoferrites and exchange in orthoaluminates. Hufner and Schmidt^[78] investigated also the exchange splitting of the energy levels of ions in erbium iron garnets and holmium iron garnets at low temperatures.

VI. OPTICAL STUDIES OF THE ANISOTROPY OF THE EXCHANGE INTERACTION IN FERROMAGNETS

Optical studies of the anisotropy of exchange interaction of the ions Yb^{3+} and Fe^{3+} in yttrium iron garnet were made by Wickersheim and White. ^[5,79,80] The splitting of the ${}^{2}F_{7/2}$ and ${}^{2}F_{5/2}$ levels by the intracrystalline field is relatively large, the ground state is located more than 500 cm⁻¹ away from the nearest excited state, as was demonstrated with nonferromagnetic ytterbium-gallium garnet as an example.

Figure 12 shows the results of an investigation of the absorption spectra of $Yb_3Fe_5O_{12}$ in the region of the ${}^{2}F_{\tau/2} \rightarrow {}^{2}F_{5/2}$ transition of the Yb^{3+} ion at different directions of the external magnetic field relative to the crystallographic axes. The experiments were performed at liquid-nitrogen temperature. The samples were plates of ytterbium iron garnet 50 μ thick, cut in the (110 plane). The spectrum were photographed with a spectrograph

FIG. 12. Absorption spectra of $Yb_3Fe_5O_{12}$ in the region of the ${}^2F_{7/2} \rightarrow {}^2F_{5/2}$ transition of the Yb^{3+} ion as a function of the direction of the external magnetic field in the (110) plane at T = 77°K.



Principal values of the exchange splittings Δv of the ground and excited states of the Yb^{*3} ion in $Yb_3Fe_5O_{12}$, principal values of the g-tensor of Yb^{3+} in $Yb_3Ga_5O_{12}$, effective field H_{eff} and exchange field H_{ex} in $Yb_3Fe_5O_{12}$

State	Prin- ciple values	$\Delta v = \frac{\Delta E}{h},$ cm ⁻¹	g eff from [37]	g g theor ³⁶	H _{eff} , kOe	H _{ex} , kOe
Ground state ${}^2F_{7/2}$	x y z	11.6 25.7 29.9	2.85 3.60 3 73	2.82 3.61 -3.84		349 611 678
Excited state ${}^{2}F_{5/2}$	x y z	$ \begin{array}{r} 15.9 \\ 5.8 \\ 29.9 \\ \end{array} $				

having a dispersion of 3.4 Å/mm in the region of 10,000 cm⁻¹. The obtained spectra turned out to be quite complicated, in spite of the fact that there are 13 electrons or one hole in the 4f shell of the Yb^{3+} ion. The complexity of the absorption spectrum is due to the fact that the Yb³⁺ ions occupy six non-equivalent positions in the garnet lattice. For certain directions of the external magnetic field, the number of non-equivalent positions decreases strongly. Thus, for a magnetic field along the [111] axis there exist only two non-equivalent positions of the Yb³⁺ ions, for which the authors obtained a groundstate exchange splitting equal to 22.1 and 25.3 cm⁻¹. These values are close to the value 25 cm⁻¹ obtained from investigations of the specific heat of $Yb_3Fe_5O_{12}$ at low temperatures.^[81]

The results of the reduction of the optical spectra of the Yb^{3+} ion are listed in the table, which contains the principal values of the splittings of the ground and first excited states (in cm^{-1}), the g-tensor components and the exchange field. The x axis was chosen to be the [100]direction, and the y and z axes are mutually perpendicular [110] directions. The components of the g-tensor of $Yb_3Ga_5O_{12}$ were obtained from paramagnetic-resonance data.^[82,83] It is seen from the table that the anisotropy of the exchange splitting is much stronger than the anisotropy of the g-factor of the Yb^{3+} ion in $Yb_3Ga_5O_{12}$. It was suggested that this disagreement may be due to the difference between the intracrystalline fields in Yb₃Ga₅O₁₂ and Yb₃Fe₅O₁₂. Experiments on the Zeeman effect at the Yb^{3+} ion in $Yb_3Fe_5O_{12}$ in magnetic fields up to 36 kOe have made it possible to determine the components of the gtensor of the ground and first-excited states, and the ground-state g-tensors of Yb₃Fe₅O₁₂ and Yb₃Ga₅O₁₂ turned orbit splitting of the energy levels of the iron-group out to be quite close.

For ferromagnetic and paramagnetic garnets of other rare-earth elements no such agreement of the g-factor is obtained as a rule.^[82] The local symmetry of the rareearth ions in the garnet lattice is D_2 and is far from cubic. In this connection, the potential of the crystal field con-tains nine non-vanishing parameters.^[84] A similar number of terms is contained also in the anisotropic exchange potential.^[85] The crystal field acting on the rare-earth ion in garnets is strongly influenced not only by the eight nearest O^{2-} ions, but also by other neighbors. This excludes the possibility of extrapolating the parameters of the crystal field to other isomorphic garnets. The nine parameters of the crystal field of this garnet can be determined at least from nine experimental data.^[84] It may

happen that in a strong crystal field there is only a small number of levels having energies comparable with kT. In this case the system can be described by an effective spin Hamiltonian which, while containing a smaller number of parameters than the total Hamiltonian, still reflects the entire anisotropy. By obtaining experimentally the effective spin-Hamiltonian parameters, it is possible to predict reliably also the other properties of the system. The simplest example of such a parameter is the g-tensor, the experimental determination of which for $Yb_3Fe_5O_{12}$ was described above.

The effective spin Hamiltonian of a garnet with spin S'_{i} in an external magnetic field H_{0} , in the case of a completely magnetized iron sublattice is given by^[84]

$$\mathcal{H} = \mu_{B} \sum_{i} (\mathbf{H}_{0}g_{i}S_{i} + \mathbf{H}_{E}G_{i}S_{i} - \mathbf{H}_{0}\alpha_{1}\mathbf{H}_{0} - \mathbf{H}_{0}\alpha_{1}^{'}\mathbf{H}_{E}) + \sum_{i>j} S_{i}^{'}k_{ij}S_{j}^{'} - \mathbf{H}_{0}\mathbf{M}_{Fe} + O(H^{3}),$$
⁽⁷⁾

where H_E is a unit vector in the direction of M_{Fe} , and the sum over i and j is taken for all the rare-earth ions. As a result of the work by Wickersheim and White, it is well known that the tensor g and the exchange-field tensor G are not proportional. From the Hamiltonian (7) we can predict an inclined structure of the rare-earth ion sublattice, which have not yet been observed experimentally. For systems that cannot be described by a relatively simple Hamiltonian (7), it is necessary to have a very large number of experimental data. The anisotropy of the exchange splitting in the ${}^{4}I_{15/2} \rightarrow {}^{4}S_{3/2}$ transition in erbium iron garnet was investigated by Eremenko and co-workers.^[73] The anisotropy of the fine structure of the absorption band ${}^{7}F_{0} \rightarrow {}^{7}F_{4}$ of Eu³⁺ in europium iron garnet was investigated in [67].

VII. FARADAY EFFECT IN THE VISIBLE REGION OF THE SPECTRUM

As follows from the experimental and theoretical investigations devoted to the optical Faraday effect, it is possible to separate four basic mechanisms of the rotation of the plane of polarization of light in iron garnets:

1) Gyromagnetic, connected with ferromagnetic and exchange resonances the natural frequencies of which lie in the far infrared and even in the microwave band.

2) Gyroelectric-exchange, connected with the exchange splitting of the energy levels of the rare-earth ions.

3) Gyroelectric-spin-orbit, connected with the spinions.

4) Gyroelectric, connected with intense electronic transitions of the rare-earth ions, whose natural frequencies lie in the ultraviolet region of the spectrum.

The contribution made to $\alpha_{\rm F}$ by the first mechanism plays the principal role in the infrared region of the spectrum, and it can be calculated quantitatively (see Ch. IV). The second mechanism is clear from the fundamental point of view, and plays the principal role in a narrow spectral region near the absorption lines of the rare-earth ions. Its contribution to $\alpha_{\rm F}$ can be calculated if one knows the splitting of the energy levels of the rare-earth ions under the joint action of the crystal and exchange fields (see Ch. V). A similar exchange mechanism of the Faraday effect for the d-group ions in

the ferromagnets $RbNiF_3$ and $RbNi_{1-X}Co_XF_3$ was considered by Smolenskiĭ and co-workers.^[22]

The third and fourth mechanisms, which will be considered in the present chapter, make the main contribution to the Faraday effect in the visible and ultraviolet regions of the spectrum. A theoretical analysis of the contribution of the third mechanism to the Faraday effect due to the spin-orbit splitting of electronic transitions in Fe^{3+} ions of an octahedral sublattice was made by Clogston.^[86,87] The ratio of the specific rotation of the plane of polarization α to the absorption coefficient σ at the center of the line is given by

$$\frac{\alpha}{\sigma} = 0.2 \, V \, \overline{\epsilon_0} \left(1 + \frac{2}{\epsilon_0} \right) \frac{n}{\Delta} \frac{w}{\Delta - w} \,, \tag{8}$$

where $u = 100 \text{ cm}^{-1}$ is the spin-orbit splitting of the ${}^6P_{5/2}$ level of the Fe³⁺ ion. An estimate of the value of α/σ in accordance with formula (8) at $\Delta = 2 \times 10^4 \text{ cm}^{-1}$, w = 1.6 $\times 10^4 \text{ cm}^{-1}$, and $\sqrt{\epsilon_0} = 2.5$ gave a value that agreed with experiment.

A similar mechanism of the Faraday effect in CrBr₃ in the visible region of the spectrum was considered in ^[19], but the theory of the third mechanism calls for further development. The situation can be illustrated using as an example the change of the magnetooptic effects under the influence of an external magnetic field in the saturation region. It was observed in [88] that the influence of the magnetic field on the magnetooptic Kerr effect in ferromagnetic metals is anomalously large. The increase of the equatorial Kerr effect, by a factor 10-40, exceeds the corresponding increase of the magnetization of the sample due to the susceptibility of the paraprocess. Later, Eremenko et al.^[89] have shown that the Faraday effect in yttrium iron garnet at $\lambda = 6328$ Å changes in an external magnetic field to approximately the same degree, but decreases in absolute magnitude. A detailed study of the effect of the influence of a magnetic field on the Faraday effect in yttrium iron garnets and holmium iron garnets in the transition region between 1 and 4.5 μ , where the effect changes from gyroelectric to gyromagnetic, is presented in ^[90]. The experimental dependences of $\alpha_{\rm F}$ on H in the saturation region for yttrium iron garnet are shown in Fig. 13. It is seen from them that the influence of the magnetic field on α_F decreases with increasing wavelength and vanishes in the gyromagnetic region at $\lambda = 4.5\mu$, within the limits of experimental accuracy. In the gyroelectric region

$$\xi = \frac{1}{\alpha_{\rm F}} \frac{\Delta \alpha_{\rm F}}{\Delta H}$$

is approximately 20 times larger than the relative change of the magnetization due to the paraprocess. The influence of the magnetic field of the Faraday effect, which is directly connected with the change of the magnetization in the saturation region, was observed in measurements of α F on holmium iron garnet in the gyromagnetic region.^[90] In iron garnets with rare-earth sublattice, the susceptibility of the paraprocess increases sharply and the value of ξ becomes amenable to measurement.

The origin of anomalously large change of α_F of yttrium iron garnet in a magnetic field is still not clear even from the qualitative point of view. A similar character of this anomalous change can be noted both in ferromagnetic d-metals and in yttrium iron garnet. In both



FIG. 13. Experimental dependences of the specific rotation of the plane of polarization $\alpha_{\rm F}$ in Y₃Fe₅O₁₂ on the external magnetic field at different infrared wavelengths. $1 - \lambda = 1 \pm 0.2 \mu$; $2 - \lambda = 1.5 \pm 0.2 \mu$; $3 - \lambda = 2.1 \pm 0.4 \mu$; $4 - \lambda = 4.5 \pm 0.8 \mu$.

cases, ξ has the same order of magnitude. The negative sign of ξ in the yttrium garnet (ξ is positive in ferromagnetic d-metals) can be attributed to the fact that the magnetization of the sublattice of the Fe³⁺ ions located in the octahedral sites and making the main contribution to $\alpha_{\rm F}$ ^[91] is oriented opposite to the external magnetic field in the wavelength region under consideration. Starting from the indicated analogy, we can separate two common factors in the magnetooptics of ferromagnetic d-metals and yttrium iron garnets: 1) the magnetooptic effects have a spin-orbit origin; 2) the difference between the oscillator strengths for electrons with right-hand and left-hand spins plays a decisive role. The influence of the magnetic field on any of these factors could lead to an observable change in the rotation of the plane of polarization of the light.

A theoretical analysis of the indicated effects may turn out to be useful not only for the construction of the theory of magnetooptic phenomena, but also for a verification of the validity of exciting concepts concerning the character of the electronic states and electronic transitions in ferromagnetic 3d-dielectrics and metals.

Finally, in the visible region of the spectrum, allowed transitions in the rare-earth sublattice, whose natural frequencies lie in the ultraviolet region of the spectrum, make an appreciable contribution to the Faraday effect of iron garnets (the fourth mechanism); this contribution decreases with increasing wavelength. Since the frequency dependence of this contribution is analogous to the frequency dependence of paramagnetic rotation of the plane of polarization in rare-earth compounds, there is at present a tendency to consider this effect in ferromagnets as a sum of paramagnetic rotations of individual rare-earth ions. For example, it is shown in [92] that the change of the Faraday effect in terbium iron garnet at λ = 1.15 μ under the influence of an external magnetic field corresponds to the Verdet constant of the isomorphic paramagnetic compounds $Tb_3Ga_5O_{12}$ and $Tb_3Al_5O_{12}$.

The theory of the Faraday effect in the ferromagnetic semiconductor EuSe, based on the energy level scheme of this compound,^[93] was developed in ^[94]. The large

magnetooptic effect is due to the transition from the localized 4f levels of the Eu²⁺ ions, which lie above the valence band of Se^{2-} , to the europium-iron opticallyactive exciton levels 5d, which are split by the spinorbital interaction.

VIII. PRACTICAL APPLICATIONS OF TRANSPARENT FERROMAGNETS

Synthesis of single-crystal ferromagnets having sufficiently high transparency in the visible and near infrared regions of the spectrum makes it possible to use them in controlled devices such as gyrators, modulators, and optical gates, similar to microwave devices based on ferrites.

There is a rather large number of lasers operating in the region of maximum transparency of $Y_3Fe_5O_{12}$, and the Faraday effect in $Y_3Fe_5O_{12}$ can be used to modulate the intensity of the laser emission. Magnetooptic modulation of infrared radiation from an He-Ne laser operating at 3.39 μ was investigated in ^[95]. 40% modulation was obtained at frequencies up to 2 MHz. This modulator differs from modulators based on the use of electrooptical effects in that its modulation coefficient in the interval from 4.5 to 9 μ is independent of the frequency. In $^{[96]}$ a $Y_3Fe_5O_{12}$ sample was placed directly in the resonator of an He-Ne laser. The sample was magnetized at a frequency 6.6 MHz. Radiation pulses of several nanosecond duration were obtained. Modulation of infrared radiation using the Faraday effect in $Y_3Fe_5O_{12}$ can be obtained at frequencies up to 200 MHz. By choosing the shape of the sample such as to obtain a minimal demagnetizing factor and the weak magnetic-anisotropy fields it is possible to use for the modulation powers that are comparable with or even smaller than the corresponding powers required by electrooptical effects.^[97] The modulation of 0.9μ infrared radiation at the frequency of ferromagnetic resonance in $Y_3Fe_5O_{12}$ was realized in ^[98]. A similar modulator for the visible part of the spectrum was constructed using CrBr₃.^[19]

It was proposed to use rare-earth iron garnets in the region of the compensation point for the construction of three dimensional memory matrices with maximum coercive force.^[99] The use of focused light beams was proposed^[100] to change the temperature for the purpose of reversing the magnetization of a small heated garnet volume. Papers devoted to the practical realization of this idea have also appeared.^[101] A promising idea is to use in memory devices ferromagnets that have unprecedented large Kerr effects upon reflection. Thus, for example, in EuO at T = 10° K and at a wavelength 0.8 μ , magnetization changes the intensity of the reflected light by 30%.^[102]

The use of a narrow light beam of a gas laser to investigate diffraction of light in Y_3 Fe₅O₁₂ by hypersound in the region of intersection of the phonon and magnon branches of the energy spectrum has made it possible to establish that the rotation of the plane of polarization of hypersound in the indicated region is $\sim 220 \text{ rad/cm}$,^[103] which is higher by two orders of magnitude than was pre- the Faraday effect in ferromagnetic dielectrics, the nonviously assumed on the basis of nonlocal measurements.

Recently, pulsed coherent radiation was obtained from

eration was observed at several wavelengths near $2.09 \,\mu$, corresponding to the transition from the lower levels of the multiplet ${}^{5}I_{7}$ to the ground level ${}^{5}I_{8}$ of the Ho³⁺ ion. The ions Er³⁺ and Tm³⁺ were used to absorb the pump radiation with subsequent transfer of the radiation to the Ho³⁺ ions. A change in the direction of the external magnetic field caused a change in the radiation frequencies. The change of the radiation frequency is connected with the splitting of the multiplets of the rare-earth ions in the intracrystalline and exchange fields. The splitting of the multiplets also changed with changing orientation of the exchange field. These effects are known in general outline.^(79,80,67) Many details, however, must still be clarified. The theory of a laser using a gyrotropic active medium was considered in [105].

The Faraday effect in ferromagnetic $CdCr_2Se_4$ was investigated in ^[106] The Curie temperature of this ferromagnet is 130°K. The absorption coefficient of singlecrystal CdCr₂Se₄ on the $6-17 \mu$ region is 15 cm⁻¹. In the entire transparency region, CdCr₂Se₄ has a rather large rotation of the polarization plane. At 10.6 μ , the value of $\alpha_{\rm F}$ is about 100 deg/cm. At wavelengths larger than 14 μ , the value of α_F is 150 deg/cm and is independent of the frequency (see Ch. IV). It seems promising to use this single crystal for the construction of magnetooptical modulators for infrared radiation, operating at wavelengths up to $17-20 \mu$.

IX. CONCLUSION

Let us list the most promising trends in further studies of transparent ferromagnets.

Foremost among them is the improvement of the quality and the synthesis of new transparent ferromagnetic crystals. In principle there are no obstacles to the synthesis of large and sufficiently perfect ferromagnetic single crystals that are transparent, say, in the entire visible region of the spectrum and have a sufficiently high Curie temperature. This would open the way for many physical investigations and practical applications.

The main trend in the study of transparent ferromagnets should be the investigation of the fine structure of the absorption bands of magnetoactive ions at low temperatures. It should be borne in mind here that the exchange splitting of the absorption lines of magnetoactive ions has been observed and reliably identified in a number of cases; consequently, the number of different ions and different compounds that must be investigated in this respect will only expand in the future. On the other hand, the although the presence of magnon satellite lines in iron garnets is assumed, a reliable identification of these lines, the establishment of single- and two-magnon transitions, the determination of the wave vectors of the magnon excitations, the establishment of a connection with the exciton character of the absorption bands, and many other aspects of this interesting physical problem call for further study.

When it comes to research on the physical nature of resonant mechanisms, which are connected with absorption bands of transition ions and with the intense transisingle-crystal $Y_3Fe_5O_{12}$ doped with small amounts of Ho³⁺, tions in rare-earth ions in the far ultraviolet, still re-Er³⁺, and Tm³⁺.^[104] At liquid-nitrogen temperature, gen- main unclear both from the qualitative and from the qualitative points of view. It should be noted that even the additivity of the contributions of the different sublattices of the ferrimagnet has not yet been proved for these mechanisms, and the absence of such additivity can greatly hinder the interpretation of the experimental results. The clarification of the physical nature of the Faraday effect in ferromagnets should be greatly helped by a theoretical analysis of the anisotropy and the influence of the magnetic field on the magnitude of the rotation of the plane of polarization. The interesting problem of scattering of light by magnetically ordered crystals is likewise directly connected with the Faraday effect.

Closely connected with the last two problems is the problem of quantitatively calculating the energy spectrum and S. Singh, Phys. Rev. Lett. 19, 948 (1967). of magnetoactive ions under the simultaneous action of the intercrystalline and exchange fields. Optical experiments will apparently be among the main sources of data on the parameters of the crystal field and the effective spin Hamiltonian. Magnetooptical experiments are particularly important in cases when the system cannot be described by an effective spin Hamiltonian of a sufficiently simple form.

In any case, it can be stated with assurance that the combination in one crystal of two such interesting physical properties as optical transparency and a spontaneous magnetic moment should lead to new fundamentally important results both from the scientific and from the practical points of view.

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