

Faraday effect of rare-earth iron garnets in strong magnetic fields

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The Faraday effect is investigated experimentally in single-crystal samples of the rare-earth iron garnets (REIG) $R_3Fe_5O_{12}$ ($R = Y, Gd, Tb, Dy, Er, Tm, Yb, Eu, Sm, \text{ and } Ho$) and also in the mixed iron garnets $R_xY_{3-x}Fe_5O_{12}$ ($R = Tb, Dy$). The measurements are made in pulsed magnetic field up to 200 kOe, in a temperature range from 4.2 to 300 K, and a wavelength of the light $\lambda = 1.15 \mu\text{m}$. The Faraday-effect field dependence observed in REIG cannot be explained by taking into account only the usually considered "paramagnetic" contribution to the Faraday effect. A theory is developed that takes into account, besides the paramagnetic mechanism, a diamagnetic mechanism as well as the mixing of the wave functions of the ground and excited multiplets. The contribution of each of these three mechanisms to the angle of rotation of the polarization plane by the rare-earth sublattice of the iron garnet are estimated theoretically. It is concluded that the mixing mechanism contributes significantly to the field and temperature dependence of the Faraday effect in REIG.

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INTRODUCTION

In Ref. 1 we presented experimental data on the Faraday effect of terbium-yttrium iron garnets in magnetic fields up to 200 kOe. It was shown that the contribution made to the resultant rotation of the light-polarization plane from the terbium sublattice is not proportional (as is usually assumed)² to the magnetic moment of this sublattice. This manifests itself most clearly at temperatures above the magnetic compensation point: in this temperature region an increase of the external magnetic field decreases the magnetic moment of the terbium sublattice, and a field equal to the molecular field H_M^0 exerted on the terbium sublattice by the iron sublattice the magnetization of the terbium sublattice vanishes, whereas the Faraday rotation due to this sublattice differs from zero in the indicated field. It was suggested in Ref. 1 that to explain the field dependence of the Faraday effect of both terbium-yttrium and other rare-earth iron garnets (REIG) account must be taken, besides the usually considered paramagnetic mechanism of the Faraday rotation due to the population difference of the ground multiplet of the rare-earth ion and proportional to the rare-earth sublattice magnetization, also of other mechanisms, viz., the diamagnetic due to splitting, in the effective field, of the levels of the excited multiplet to which the optical transition takes place, and the mixing mechanism due to the admixture of wave functions of excited multiplets to the wave function of the ground multiplet. It must be emphasized that all these mechanisms of the Faraday effect in paramagnets were considered earlier (see, e.g., the review by Serber³), and the diamagnetic mechanism connected with the exchange splitting of the excited levels of RE ions was observed for REIG for the first time in Ref. 4 and was discussed in Refs. 5 and 6. The main point, to which attention was called in Ref. 1, was that in magnetically ordered substances (and particularly in REIG), the effective fields acting on the excited multiplet and mixing the wave functions of the ground and excited

multiplets differ from the effective field that acts on the ground multiplet. It is this which leads to the singularities of the field dependences of the Faraday effect of terbium-yttrium iron garnets, observed in Ref. 1, and in particular to the fact that when the moment of the rare-earth sublattice vanishes the Faraday effect due to this sublattice does not vanish.

We note that the conclusion that the Faraday rotation produced in REIG by the rare-earth sublattice, the diamagnetic mechanism, and the mixing mechanism were described in Ref. 1 under the assumption that the rotation due to the iron sublattice is the same in all the garnets and is equal to the Faraday effect in yttrium iron garnet. According to a different point of view the singularities of the Faraday rotation of REIG is attributed to a change in the contribution of the iron sublattice. In addition, no theoretical estimates were given in Ref. 1 of the values of the diamagnetic contribution and of the mixing contribution. Thus, the model proposed for the Faraday effect of REIG in Ref. 1 was hypothetical in many respects. The hypothetical character of the conclusions of Ref. 1 was resolved to a considerable degree by experiments on the Faraday rotation in terbium gallate garnet,⁹ which confirmed the substantial role of the mixing mechanism and (or) of the diamagnetic mechanism in the Faraday effect, at least for terbium ions in garnets.

In this paper we consider in greater detail the theory of Faraday rotation due to rare-earth ions. Theoretical estimates are presented of the contributions of various mechanisms to the rotation angle of the polarization plane. Experimental results are presented of measurements of the Faraday effect of various REIG in magnetic fields up to 200 kOe. The possibility is analyzed of describing the experimental data within the framework of the model considered.

THEORY

The rare-earth iron garnets $R_3Fe_5O_{12}$ (cubic crystal structure, space group $O_h^{10} - Ia3d$) are three-sublattice fer-

rimagnets. One of the sublattices (c sublattice) comprises the rare earth ions R^{3+} , and the other two (a and d sublattices) are made up of the Fe^{3+} ions. Since the exchange interaction between the iron ions in the a and d sublattices is considerably larger than the other exchange interactions, it is possible to combine the a and d sublattices into a single iron sublattice with magnetization M_{Fe} equal to the difference between the magnetization of the a and d sublattices. The magnetic ordering of the RE ions takes place under the action of the exchange field produced by the iron sublattice. The exchange interaction within the RE sublattice can be neglected. The RE ions in the iron garnets (IG) are located in six nonequivalent points having a surround symmetry D_2 (Ref. 10). The Hamiltonian of the interaction of the RE ions with the crystal, external, and exchange fields is of the form

$$\mathcal{H} = V_{cr} - \mu_B (\mathbf{L} + 2\mathbf{S}) \mathbf{H} - 2\mu_B \mathbf{S} \mathbf{H}_{ex}, \quad (1)$$

where V_{cr} is the crystal-field potential, and H_{ex} is the exchange field applied to the RE ion by the iron sublattice and assumed by us to be isotropic. The anisotropy of the exchange interaction could be taken into account by assuming that the quantity H_{ex} in (1) is a function of the orbital-momentum operator L of the RE ion. This dependence can be represented in the form of an expansion in irreducible tensor operators

$$H_{ex}(\mathbf{L}) = \sum_{n=0}^{\infty} \sum_{m=-n}^n G_n^m Y_n^m.$$

Recent investigations¹¹⁻¹³ have shown that $|G_n^n (n \geq 1)| \ll G_0^0$, therefore we retain in the expression for H_{ex} only the first term G_0^0 , i.e., we assume that H_{ex} is isotropic. Allowance for the terms $G_n^n Y_n^n$ with $n \geq 1$ may turn out to be important in the calculation of effects in which the decisive role is played by the anisotropy energy. It must also be emphasized that the parameter H_{ex} is the only one that determines the exchange interaction for the entire ground term; at the same time, as will be shown below, the molecular fields that determine the magnitude of the effect can have substantially different values for different multiplets of the ground term.

RE ions with unfilled $4f$ shell have an appreciable spin-orbit interaction. The distances to the nearest excited multiplet for the different RE ions are ≈ 1000 , ≈ 350 , ≈ 2000 , ≈ 3000 , ≈ 5000 , ≈ 6500 , ≈ 8000 , and $\approx 10000 \text{ cm}^{-1}$ for Sm^{3+} , Eu^{3+} , Tb^{3+} , Dy^{3+} , Ho^{3+} , Er^{3+} , Tm^{3+} , and Yb^{3+} , respectively.

The interactions of the RE ion with the crystal and exchange fields are smaller by one-two orders than the multiplet splitting on account of the spin-orbit interaction, so that the lower states of an RE ion in a garnet are states due to splitting of the ground multiplet (with given J) by the crystal and exchange fields. A similar situation obtains also for excited multiplets to which the electro-dipole transitions take place, but the excited states have so far not been investigated in sufficient detail.

We consider the Faraday effect due to direct transition to parity- and spin-allowed multiplets that stem from the $4f^{n-1}5d$ configuration. Since we are interested mainly in

the field dependence of the Faraday effect, we shall pay special attention in the forthcoming theoretical investigation to the aforementioned mixing mechanisms and to the diamagnetic contribution to the polarization-plane rotation.

A. Ground-state wave functions. For an RE ion in a nonequivalent point of type r the eigenfunctions of the ground multiplet, split by the crystal, exchange, and external fields, can be represented in a coordinate frame with axis $z || \langle 111 \rangle$ in the form

$$|\psi_i^r\rangle_0 = \sum_m C_{im}^r |S_0 L_0 J_0 m\rangle, \quad (2)$$

and the energy eigenvalues are

$$E_i^r = \langle \psi_i^r | V_{cr} - \mu_B g_{J_0} H_{eff}^0 J_z | \psi_i^r \rangle, \quad (3)$$

where

$$H_{eff}^0 = H \pm H_m^0 = H \pm [2(g_{J_0} - 1)/g_{J_0}] H_{ex}, \quad (3a)$$

g_{J_0} is the Landé factor of the ground multiplet, and the \pm signs pertain respectively to temperatures below and above the magnetic-compensation point T_{comp} .

We take now into account the mixing of the ground and first excited states of multiplets with $J = J_0 - 1$ for heavy RE ions and $J = J_0 + 1$ for light RE ions in the exchange and external magnetic fields. Accurate to terms of first order in the ratio of $\mu_B H$ and $\mu_B H_{ex}$ to the distance between the ground and the first-excited multiplets, we find that the corrections to the eigenfunctions (2) are of the form

$$\begin{aligned} \Delta \psi_i^r &= \sum_{mm'} C_{im'}^r \frac{\mu_B (H \pm 2H_{ex})}{\Delta} \langle L_0 S_0 J m' | S_z | L_0 S_0 J_0 m \rangle | L_0 S_0 J m' \rangle, \end{aligned} \quad (4)$$

where Δ is the distance between the ground and the first excited multiplets, and the matrix element of the operator S_z is equal to

$$\begin{aligned} \langle L_0 S_0 J m' | S_z | L_0 S_0 J_0 m \rangle &= (-1)^{J-m'} \begin{pmatrix} J & 1 & J_0 \\ -m' & 0 & m \end{pmatrix} \langle L_0 S_0 J || S_z || L_0 S_0 J_0 \rangle, \end{aligned} \quad (4a)$$

$$\begin{aligned} &\langle L_0 S_0 J || S_z || L_0 S_0 J_0 \rangle \\ &= (-1)^{J+L_0+S_0+1} [S_0(S_0+1)(2S_0+1)(2J_0+1)(2J+1)]^{1/2} \\ &\times \left\{ \begin{matrix} L_0 & S_0 & J_0 \\ 1 & J_0 & S_0 \end{matrix} \right\}. \end{aligned} \quad (4b)$$

We have used here the equality

$$\begin{aligned} \langle J_0 S_0 J m' | -\mu_B (L_z + 2S_z) H \mp 2\mu_B S_z H_{ex} | L_0 S_0 J_0 m \rangle \\ = \langle L_0 S_0 J m' | -\mu_B J_z H - \mu_B (H \pm 2H_{ex}) S_z | L_0 S_0 J_0 m \rangle \\ = \langle L_0 S_0 J m' | S_z | L_0 S_0 J_0 m \rangle (H \pm 2H_{ex}) \mu_B, \end{aligned}$$

since the matrix element $\langle L_0 S_0 J m' | J_z H | L_0 S_0 J_0 m \rangle$ vanishes at $J \neq J_0$. It is also obvious that the considered matrix element differs from zero only at $m = m'$, as follows directly from the known properties of the $3J$ symbols contained in (4a).

Using the explicit expressions for the $3J$ and $6J$ symbols

(see Ref. 14) that enter in (4a) and (4b), we find that the eigenfunctions of the ground multiplet, with account taking of the admixture of the first-excited multiplet, will take the form

$$\psi_i^r = \psi_{i0}^r + \Delta\psi_i^r = \sum_m C_{im}^r \left\{ |L_0 S_0 J_0 m\rangle + \frac{\mu_B H_{eff}^1}{\Delta} S_m |L_0 S_0 J m\rangle \right\}, \quad (5)$$

where S_m for heavy ($J_0 = L_0 + S_0$, $J = J_0 - 1$) and light ($J_0 = |L_0 - S_0|$, $J = J_0 + 1$) RE ions is of the form

$$S_m = - \left(1 - \frac{m^2}{J_0^2}\right)^{1/2} \left(\frac{S_0 L_0}{2J_0 - 1}\right)^{1/2}, \quad J_0 = L_0 + S_0, \quad J = J_0 - 1, \quad (6a)$$

$$S_m = (-1)^{2J_0} \left[1 - \frac{m^2}{(J_0 + 1)^2}\right]^{1/2} \left[\frac{(L_0 + S_0 + J_0 + 2)(L_0 + S_0 - J_0)}{4(2J_0 + 3)} \right]^{1/2},$$

$$J_0 = |L_0 - S_0|, \quad J = J_0 + 1,$$

and

$$H_{eff}^1 = H \pm 2H_{ex}. \quad (7)$$

B. Magnetization. Before we investigate the Faraday effect, we shall calculate the magnetization M_R of the RE sublattice (per ion). Averaging the operator of the magnetic moment $\mu = \mu_B(L_z + 2S_z)$ over the states (5), we find

$$M_R = \frac{1}{6} \sum_{r=1}^6 \text{Sp} \mu \rho_r = M_0(H_{eff}^0, T) + \Delta M_{VV}, \quad (8)$$

where M_0 is the magnetic moment of the RE sublattice in the absence of multiplet mixing, ΔM_{VV} is the van Vleck magnetic moment due to the mixing of the first excited (thermally unpopulated) multiplet with the ground state. The expressions for M_0 and ΔM_{VV} are

$$M_0 = \frac{1}{6} \mu_B g_{J_0} \sum_{mir} |C_{im}^r|^2 m \rho_i^r, \quad (9)$$

$$\Delta M_{VV} = \frac{\mu_B^2 H_{eff}^1}{3\Delta} \sum_{mir} |C_{im}^r|^2 S_m^2 \rho_i^r,$$

where

$$\rho_i^r = \exp(-E_i^r/T) / \sum_k \exp(-E_k^r/T).$$

We note that with rising temperature $M_0(T, H_{eff}^0) \rightarrow 0$ with increasing populations of the levels of the ground multiplet, whereas at temperatures that satisfy the relation $w \ll T \ll \Delta$ (where w is the characteristic level splitting of the ground multiplet by the crystal and exchange fields), while

$$\Delta M_{VV} \approx \frac{\mu_B^2 H_{eff}^1}{3\Delta} \sum_{mir} |C_{im}^r|^2 S_m^2 \frac{1}{2J_0 + 1} \quad (10)$$

does not depend on temperature.

In RE ions, $\mu_B H_{ex} \approx 10 - 20 \text{ cm}^{-1}$ and Δ is larger by two orders, so that at room temperatures and lower we have $\Delta M_{VV} \ll M_0$ and the van Vleck contribution to the magnetization of the RE lattice is therefore usually neglected (except for europium and samarium IG).

We point out also that the effective fields acting on the ground multiplet and mixing of the wave functions of the ground and first-excited multiplets [see (3a) and (7)] are different.

C. Rotation of the polarization plane. We consider now the Faraday rotation.¹⁾ The polarization-plane rotation angle for electro-dipole transitions at incident-electromagnetic-wave frequencies far from the resonance frequencies are determined by the well known expression^{2,3,15}

$$\alpha_F = \frac{\pi N e^2}{c n \hbar} \left(\frac{n^2 + 2}{3} \right)^2 \times \sum_{ab} \frac{\omega^2}{\omega_{ab}^2 - \omega^2} \{ |\langle a | P_+ | b \rangle|^2 - |\langle a | P_- | b \rangle|^2 \} \rho_a, \quad (11)$$

where $P_{\pm} = x \pm iy$; $|a\rangle$ and $|b\rangle$ are the eigenfunctions of the ground and excited states of the ion, $\hbar\omega_{ab} = E_a - E_b$, N is the number of RE ion per unit volume, n is the average refractive index, ρ^a is the population of the state $|a\rangle$. Assume that we have two groups of levels A and B (say, ground and excited multiplets). Then the energies of the excited levels can be represented in the form

$$E_b = \hbar\omega_{AB} + E_b', \quad \sum_b E_b' = 0,$$

where $\hbar\omega_{ab}$ is the distance between the gravity centers of groups A and B (it is implied here that $\sum E_a = 0$).

It was shown in Ref. 3 that α_F can be represented as three contributions, the A , B , and C terms in Serber's terminology.³ The A term is the so-called diamagnetic contribution due to the Zeeman splitting of the ground and excited groups of levels. It stems from the expansion of the frequency factor under the summation sign in (11), and is of the form

$$A_F = \frac{\pi N e^2 (n^2 + 2)^2}{9c n \hbar} \sum_{ab} \rho_a \{ |\langle a | P_- | b \rangle|^2 - |\langle a | P_+ | b \rangle|^2 \} \frac{\omega^2}{\omega_{AB}^2 - \omega^2} \frac{2\omega_{AB}(E_a - E_b')}{\hbar(\omega_{AB}^2 - \omega^2)}. \quad (12)$$

The C term is the paramagnetic contribution. It is proportional to the ion magnetization [more accurately, proportional to the mean value of the magnetic-moment operator over the functions of the ground multiplet (12)]. The B term is the result of the mixing of the states.

For RE ions, a substantial contribution to the Faraday effect is apparently made by electro-dipole transitions excited states from the configuration $4f^{n-1}5d$. At present the spectrum and the wave functions of the excited states from the configuration $4f^{n-1}5d$ are not sufficiently well known. We shall assume that these excited states are multiplets split by the crystal and exchange fields.

Consider the Faraday effect due to transitions from the states (5) to excited multiplets that arise in splitting, by the spin-orbit interaction, of terms belonging to the configuration $4f^{n-1}5d$. According to the known selection rules, electro-dipole transitions from the multiplet $L_0 S_0 J_0$ are possible to states with $J = J_0$, $J_0 \pm 1$; $L = L_0$, $L_0 \pm 1$; $S = S_0$. We shall investigate the B and C contributions. To this end we

replace ω_{ab} in (11) by ω_{LJ} ($\hbar\omega_{LJ}$ is the energy of the excited multiplet S_0LJ). In the calculation of the matrix elements contained in (11) it is convenient to transform from the operators r_{\pm} to spherical harmonics $Y_1^{\pm 1}$:

$$r_{\pm} = \mp \left(\frac{8\pi}{3} \right)^{1/2} r Y_1^{\pm 1}.$$

Using the expressions in (5), we obtain

$$\begin{aligned} \langle \psi_i^r | r_{\pm} | S_0LJm' \rangle &= \mp \left(\frac{8\pi}{3} \right)^{1/2} \langle r \rangle \langle \psi_i^r | Y_1^{\pm 1} | S_0LJm' \rangle \\ &= \mp \left(\frac{8\pi}{3} \right)^{1/2} \langle r \rangle C_{im'} \left\{ (-1)^{J_0-m} \begin{pmatrix} J_0 & 1 & J \\ -m & \pm 1 & m' \end{pmatrix} \right. \\ &\quad \times \langle L_0S_0J_0 \| Y_1 \| LS_0J \rangle + \frac{\mu_B H_{eff}^1}{\Delta} S_m \\ &\quad \left. \times \begin{pmatrix} J_0-1 & 1 & J \\ -m & \pm 1 & m' \end{pmatrix} (-1)^{J_0-1-m} \langle L_0S_0J_0-1 \| Y_1 \| LS_0J \rangle \right\}. \end{aligned} \quad (13)$$

Substituting (13) in (11) we find that, accurate to terms linear in $\mu_B H_{eff}^1/\Delta$, the polarization-plane rotation angle takes the form

$$\alpha_F = K \sum_{JL} \alpha(LJ) \frac{\omega^2}{\omega_{LJ}^2 - \omega^2}, \quad K = \frac{8\pi^2 N e^2}{3cn\hbar} \left(\frac{n^2+2}{3} \right)^2 \langle r \rangle^2, \quad (14)$$

$$\begin{aligned} \alpha(LJ) &= \sum_{rLmm'} |\langle S_0L_0J_0 \| Y_1 \| S_0LJ \rangle|^2 \left\{ \begin{pmatrix} J_0 & 1 & J \\ -m & -1 & m' \end{pmatrix}^2 \right. \\ &\quad \left. - \begin{pmatrix} J_0 & 1 & J \\ -m & 1 & m' \end{pmatrix}^2 \right. \\ &\quad \left. + \frac{2\mu_B H_{eff}^1}{\Delta} \frac{\langle S_0L_0J_0-1 \| Y_1 \| S_0LJ \rangle}{\langle S_0L_0J_0 \| Y_1 \| S_0LJ \rangle} S_m (-1)^{2(J_0-m)-1} \right. \\ &\quad \left. \times \left[\begin{pmatrix} J_0 & J & 1 \\ -m & m' & -1 \end{pmatrix} \begin{pmatrix} J & J_0-1 & 1 \\ m' & -m & -1 \end{pmatrix} \right. \right. \\ &\quad \left. \left. - \begin{pmatrix} J_0 & J & 1 \\ -m & m' & 1 \end{pmatrix} \begin{pmatrix} J & J_0-1 & 1 \\ m' & -m & 1 \end{pmatrix} \right] \right\}, \end{aligned} \quad (15)$$

where $\alpha(LJ)$ is a quantity that describes the off-diagonal susceptibility of such a transition.

We consider now the individual contributions made to α_F by allowed transitions to the multiplets LJ . We turn first to transitions to the multiplets LJ_0 . Using the relations

$$\sum_{m'} \left[\begin{pmatrix} J_0 & 1 & J_0 \\ -m & -1 & m' \end{pmatrix}^2 - \begin{pmatrix} J_0 & 1 & J_0 \\ -m & 1 & m' \end{pmatrix}^2 \right] = - \frac{m}{J_0(J_0+1)(2J_0+1)}, \quad (16)$$

$$\begin{aligned} \sum_{m'} (-1)^{2(J_0-m)-1} &\left[\begin{pmatrix} J_0 & J_0 & 1 \\ -m & m' & -1 \end{pmatrix} \begin{pmatrix} J_0 & J_0-1 & 1 \\ m' & -m & -1 \end{pmatrix} \right. \\ &\quad \left. - \begin{pmatrix} J_0 & J_0 & 1 \\ -m & m' & 1 \end{pmatrix} \begin{pmatrix} J_0 & J_0-1 & 1 \\ m' & -m & 1 \end{pmatrix} \right] \\ &= \left(1 - \frac{m^2}{J_0^2} \right)^{1/2} \left[\frac{J_0+1}{(2J_0+1)(2J_0-1)} \right]^{1/2} \\ &= - \frac{S_m}{2J_0+1} \left(\frac{J_0+1}{L_0S_0} \right)^{1/2}, \end{aligned}$$

the validity of which can be easily verified by direct calculation, we find that the contribution made to the Faraday effect by $J_0 \rightarrow J_0$ transition is of the form

$$\alpha(LJ_0) = - \frac{6}{g_{J_0}\mu_B} \frac{|\langle S_0L_0J_0 \| Y_1 \| S_0LJ_0 \rangle|^2}{(2J_0+1)J_0(J_0+1)} [M_0(H, T) + A(LJ_0)\Delta M_{vV}]; \quad (17)$$

here

$$A(LJ_0) = g_{J_0}J_0(J_0+1) \frac{\langle S_0L_0J_0-1 \| Y_1 \| S_0LJ_0 \rangle}{\langle S_0L_0J_0 \| Y_1 \| S_0LJ_0 \rangle} \left(\frac{J_0+1}{L_0S_0} \right)^{1/2}. \quad (18)$$

To derive (17) we used Eq. (10). The ratio of the matrix elements in (18) can be expressed in terms of $6J$ symbols (Racah coefficients) in the following manner:

$$\begin{aligned} &\frac{\langle S_0L_0J_0-1 \| Y_1 \| S_0LJ_0 \rangle}{\langle S_0L_0J_0 \| Y_1 \| S_0LJ_0 \rangle} \\ &= \left(\frac{2J_0-1}{2J_0+1} \right)^{1/2} \left\{ \begin{matrix} L & S_0 & J_0 \\ J_0+1 & 1 & L_0 \end{matrix} \right\} \left\{ \begin{matrix} L & S_0 & J_0 \\ J_0 & 1 & L_0 \end{matrix} \right\}^{-1}. \end{aligned} \quad (19)$$

The first term in the brackets in (17) is the paramagnetic contribution (the C term), while the second is the B term due to mixing of the ground and first-excited multiplets. The quantity $A(LJ_0)$, as shown by direct calculation using the table of Racah coefficients,¹⁴ can have different signs, depending on J_0, S_0, L_0 and $L = L_0, L_0 + 1$, and can reach values $\sim 10^2$. For example, for the ion Tb^{3+} ($L_0 = S_0 = 3$, $L = L_0 + 1$) we have $A(L_0 + 1, J_0) = 147$, whereas $A(L_0, J_0) = -21$. For the ion Er^{3+} ($J_0 = \frac{15}{2}$, $S_0 = \frac{3}{2}$, $L_0 = 6$) we have $A(L_0 + 1, J_0) = 3 \cdot 17^2/2 = 433.5$; $A(L_0, J_0) = -3 \cdot 17/4 = -12.75$. These estimates show that the Faraday effect can contain an appreciable quasi-van-Vleck contribution (B term), which is comparable with the paramagnetic contribution proper at sufficiently high temperatures, when the quantity $M_0(H_{eff}^0, T)$ is small.

A similar analysis of the contribution of electro-dipole transitions into a state with $J = J_0 - 1$ ($L = L_0 \pm 1$) leads to the following expression for the polarization-plane rotation angle:

$$\alpha(L, J_0-1) = - \frac{6}{g_{J_0}\mu_B} \frac{|\langle S_0L_0J_0 \| Y_1 \| S_0LJ_0-1 \rangle|^2}{J_0(2J_0+1)} \times [M_0(H, T) + A(L, J_0-1)\Delta M_{vV}], \quad (20)$$

where

$$A(L, J_0-1) = g_{J_0}J_0 \left(\frac{J_0-1}{L_0S_0} \right)^{1/2} \frac{\langle L_0S_0J_0-1 \| Y_1 \| S_0LJ_0-1 \rangle}{\langle L_0S_0J_0 \| Y_1 \| S_0LJ_0-1 \rangle}.$$

For the Tb^{3+} ion we have

$$\begin{aligned} A(L_0-1, J_0-1) &= -3, \quad A(L_0, J_0-1) = 15, \\ A(L_0+1, J_0-1) &= 39. \end{aligned} \quad (21)$$

The quantity $\alpha(L_0 + 1, J_0 + 1)$ is of the form

$$\alpha(L_0+1, J_0+1) = \frac{6}{g_{J_0}\mu_B} \frac{|\langle L_0S_0J_0 \| Y_1 \| L_0+1 S_0J_0+1 \rangle|^2}{(J_0+1)(2J_0+1)} M_0(H, T). \quad (22)$$

In particular, asymptotically as $\omega \rightarrow \infty$ (i.e., when $\omega \gg \omega_{LJ}$, summing (17) and (22) over all possible L and J , we find that

for the Tb^{3+} ion the Faraday effect due to the C and B terms takes the form

$$\alpha_F = C[M_0(H_{eff}^0, T) - 3\Delta M_{VV}(H_{eff}^1)], \quad (23)$$

where

$$C = K \frac{6}{g_J \mu_B} \frac{J_0}{2J_0 + 1} \left\{ \frac{\langle L_0 \| Y_1 \| L_0 - 1 \rangle^2}{3} - \frac{\langle L_0 \| Y_1 \| L_0 + 1 \rangle^2}{4} + \frac{\langle L_0 \| Y_1 \| L_0 \rangle^2}{3 \cdot 4} \right\}. \quad (24)$$

We call attention to the fact that, as follows from the experimental data cited below, the numerical coefficient of the second term of (23) is of the order of 10 at a light wavelength $1.15 \mu m$.

It must be borne in mind that Eq. (14) implies summation over all the excited states of the RE ion. Obviously, therefore, some cancellation is possible of the contributions to the "mixing" effect from the transitions to different excited levels. This can be seen, in particular, from the examples given above, which demonstrate the different signs of the contributions to $A(L, J)$, corresponding to levels with different L . At the same time it can be stated that owing to the strong splitting of the excited terms by the crystal field and by the spin-orbit interaction, the known sum rules that might lead to a strong cancellation of the effect (as at $\omega \rightarrow \infty$) are not applicable here. Unfortunately we do not have the necessary data on the energy spectrum and on the wave functions of the RE ions (i.e., also on the values and on the corresponding spectroscopic coefficients) in the iron-garnet crystal; these data would permit a detailed calculation of the Faraday effect. The estimates and examples above illustrate the qualitative aspect of the situation, consisting of the fact that the Faraday effect is determined by two different effective fields, H_{eff}^0 and H_{eff}^1 ; in this case the contribution of the mixing effects to α_F can be considerably larger than the corresponding contribution to the magnetization, and the dependence of the polarization-plane rotation angle differs thus from the field dependence of the magnetization

$$M_R = M_0(H_{eff}^0) + \Delta M_{VV}(H_{eff}^1).$$

We have investigated the effect of the mixing of the first-excited multiplet with the ground one on the Faraday effect. We estimate now the influence of the diamagnetic terms and of the mixing terms of the multiplets from the configuration $4f^{n-1}5d$.

Owing to the factor $\omega_{AB}(E_a - E'_b)/\hbar(\omega_{AB}^2 - \omega^2)$ [see (12)] the diamagnetic terms contain additional smallness parameters equal to $\mu_B H_{eff}^0/\hbar\omega_{AB}$ and $\mu_B H_{eff}^1/\hbar\omega_{AB}$, where

$$H_{eff}^i = H \pm [2(g_J - 1)/g_J] H_{ex}$$

are the effective magnetic fields acting on multiplets from the configuration $4f^{n-1}5d$. The analogous parameter for the terms connected with the mixing of the first-excited multiplet and the ground one is $\mu_B H_{eff}^1/\Delta$. For RE ions, however, $\hbar\omega_{AB} \gg \Delta$. Thus, for example, for Tb^{3+} we have $\hbar\omega_{AB} = (3/4) \cdot 10^4 \text{ cm}^{-1}$, and $\Delta \approx 2 \cdot 10^3 \text{ cm}^{-1}$. Therefore at frequencies far from resonance the diamagnetic terms make a smaller contribution to the Faraday effect than the investi-

gated B term. (We note here that their contribution to the asymptotic form as $\omega \rightarrow \infty$ vanishes completely, in contrast to the B term.) In addition, as shown by analysis, the effective magnetic fields H_{eff}^J that act on multiplets in which electro-dipole transitions are allowed to differ little³⁾ from H_{eff}^0 . Thus, the field dependences of the A and C terms differ insignificantly, whereas the effective field $H_{eff}^1 = H \pm 2H_{ex}$ that mixes the multiplets differs substantially from H_{eff}^0 ($H_{eff}^1 = H \pm \frac{2}{3}H_{ex}$ for Tb^{3+}). As for the terms for the mixing of multiplets from an excited configuration, they take, owing to summation over the multiplets, a form similar to the diamagnetic terms in which H_{eff}^J is replaced by the field H_{eff}^1 . This means that they take the form

$$-A \frac{\omega^2}{\hbar(\omega_0^2 - \omega^2)^2} \mu_B H_{eff}^1, \quad (25)$$

where $\hbar\omega_0$ is the distance to the center of gravity of the term with given L , and reduces to a renormalization of the investigated B term.

Our theoretical analysis shows thus that the Faraday effect due to the rare-earth sublattices in an REIG can be represented in the form proposed in Ref. 1:

$$\alpha_F^R = CM_R(H_{eff}^0) + DH_{eff}^1. \quad (26)$$

Here H_{eff}^0 and H_{eff}^1 are given by (3) and (7), respectively; $D = C_{\chi_{VV}} \langle A \rangle$, where $\chi_{VV} = \Delta M_{VV}/H_{eff}^1 \sim \mu_B^2/\Delta$ is determined from (9), $\langle A \rangle$ is the averaged value of the quantities $A(L, J)$ whose characteristic values are given, e.g., in (21).

We note also that in Tm^{3+} and Yb^{3+} we have $\Delta \sim 10^4 \text{ cm}^{-1}$, therefore the considered mixing effects are small and for a more complete comparison with experiment the diamagnetic terms must be taken into account. Gadolinium IG is worthy of a separate theoretical examination, since $L_0 = 0$ in Gd^{3+} , therefore, in accord with the existing theory,^{3,6} its paramagnetic Faraday effect is zero. The observed rotation of the polarization plane is determined by the spin-orbit interaction of the excited terms. To analyze the field dependence of gadolinium IG we need more complete information on the splitting of the excited terms under the influence of the spin-orbit, exchange, crystal, and external-magnetic fields. We note that since the energy level of the excited terms can have a linear dependence on the fields H_{eff}^i acting on them, the analogous nonlinear contributions may turn out to be substantial also in the field dependences of the values of α_F in such IG. (The same can apply also to other ions whose diamagnetic contributions to the Faraday effect are large enough.)

SAMPLES AND MEASUREMENT PROCEDURE

In the present study we investigated the Faraday effect on single crystal samples of the REIG $R_3Fe_5O_{12}$, where $R = Y, Gd, Tb, Dy, Er, Tm, Yb, Eu, Sm$, and Ho , and also on single crystals of the mixed IG $R_x Y_{3-x} Fe_5 O_{12}$ ($R = Tb, Dy$) grown by crystallization from the molten solution. The measurements were made on plates 100–300 μm thick, oriented perpendicular to the easy-magnetization axis. For all the investigated garnets except the erbium IG the measurements were made on plates cut in the (111) plane; in the

erbium IG, whose easy magnetization axis is parallel to $\langle 100 \rangle$ below 77 K, additional measurements were made on a plate cut in the (110) plane. The measurements were made in the temperature interval 4.2–300 K in a special flow-through cryostat at a laser wavelength $1.15 \mu\text{m}$ using the standard intensity method. Exceptions were the Tb IG, whose samples were damaged below 77 K in pulsed magnetic fields, apparently because of the large magnetoelastic strains, and the Sm IG, in which the easy magnetization axis below 65 K is $\langle 110 \rangle$ and birefringence effects are substantial. Pulsed magnetic fields up to 200 kOe were used (the measurement procedure is described in greater detail in Ref. 1).

EXPERIMENTAL RESULTS

Figure 1 shows the field dependences of the Faraday rotation of the investigated REIG at three temperatures: 290, 80, and 4.2 K. It can be seen that saturation of the Faraday effect is reached in technical-saturation weak fields (we recall that in all cases the measurements were made with the field oriented along the easy-magnetization axis). Further increase of the field leads to a strong change of the Faraday rotation.

We note that the basic characteristics of the Faraday rotation, namely the values α_F^s of the spontaneous Faraday effect, obtained by extrapolation to zero field from the

strong-field region, and $d\alpha_F/dH$ of the field-dependences of the Faraday effect, change greatly on going from one IG to another, and have different temperature dependences for different REIG. The temperature dependences of the spontaneous Faraday effect and of the Faraday susceptibility $d\alpha_F/dH$ for the investigated REIG are shown in Figs. 2 and 3. The literature reports extensive investigations of the spontaneous Faraday effect above 77 K (see, e.g., Refs. 2, 8, and 15–17). For all the investigated garnets, our data agree within 5–10% with the published ones. For the Dy IG a similar agreement with the results of Ref. 17 is observed in the entire temperature interval 4.2–300 K.

The field dependences of the Faraday effect of mixed Tb–Y and Dy–Y IG are similar to the field dependences of the pure REIG. In all cases the Faraday rotation changes linearly when the RE ion is replaced by the nonmagnetic yttrium.

DISCUSSION OF RESULTS

As already noted, the Faraday effect of REIG can be represented in the form

$$\alpha_F = \mp \alpha_{Fe} \pm \alpha_R, \quad (27)$$

where α_{Fe} is the Faraday rotation due to the iron sublattices, α_R is the Faraday rotation of the RE lattices and can be

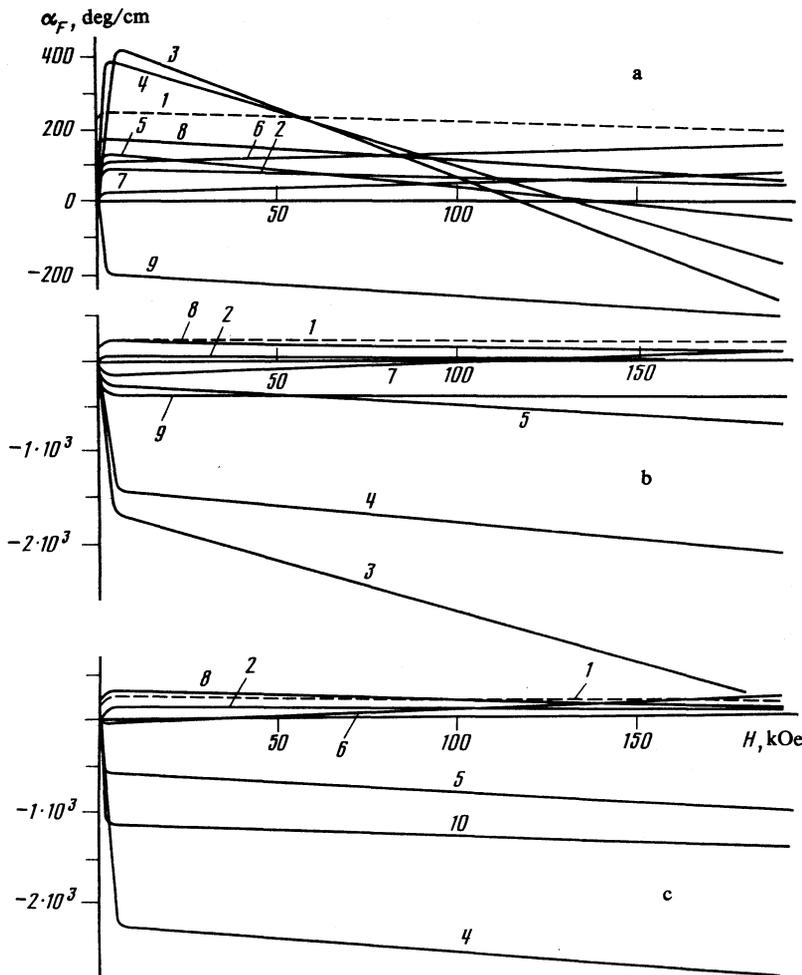


FIG. 1. Field dependences of the Faraday rotation in various REIG: 1—Y, 2—Gd, 3—Tb, 4—Dy, 5—Er, 6—Tm, 7—Yb, 8—Eu, 9—Sm, 10—Ho at different temperatures: a—290 K, b—77 K, c—4.2 K.

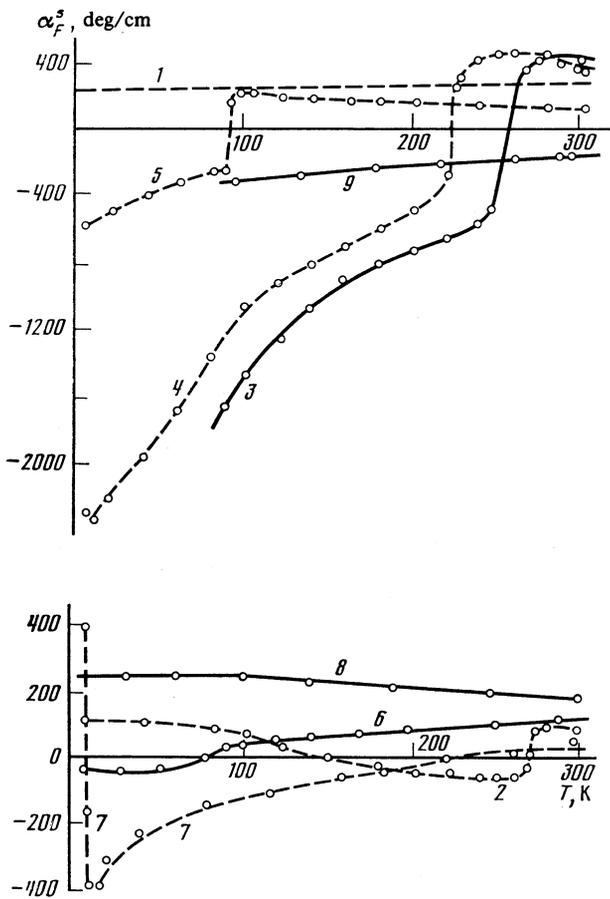


FIG. 2. Temperature dependences of the spontaneous Faraday rotation (the designations are the same as in Fig. 1).

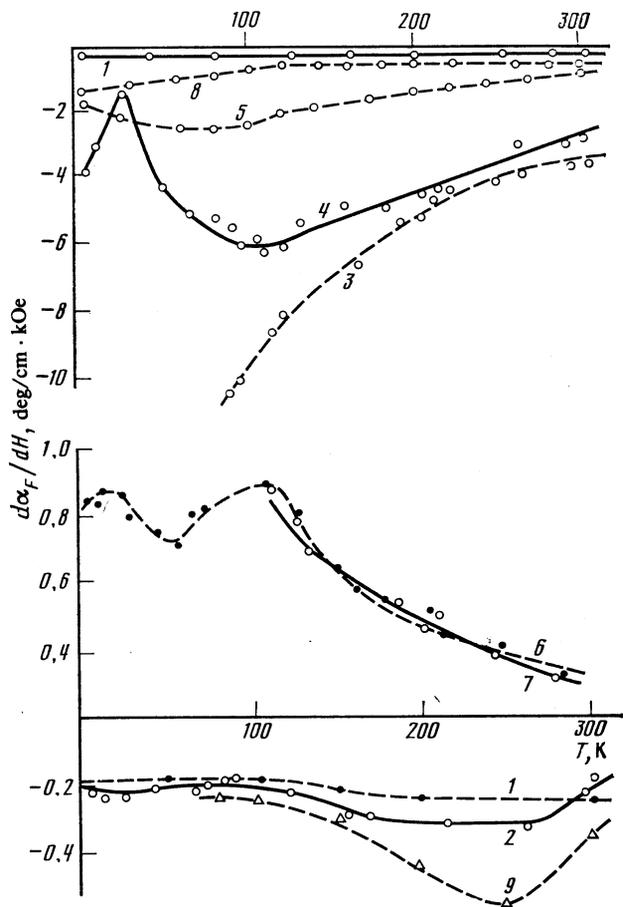


FIG. 3. Temperature dependence of the Faraday susceptibility in various REIG (the designations are the same as in Fig. 1).

described within the framework of the model developed above by Eq. (26). Above the magnetic-compensation point (this situation obtains for all the investigated REIG at room temperature) the magnetization of the RE sublattice is oriented antiparallel to the field, and the minus sign must be used in (27). As already noted, at these temperatures an increase of the external field leads to a decrease of the magnetic moment of the RE sublattice, and if the external field is equal to the molecular field H_M^0 that acts on the ground multiplet, the magnetic moment of the RE sublattices vanishes. Table I lists the values of the molecular fields for the REIG at room temperature. It lists also the critical fields H_{cr} at which the values of the REIG Faraday effect become equal to the rotation of the polarization plane in yttrium IG. In other words, H_{cr} is that external field at which the Faraday effect due to the RE sublattice is equal to zero (if it is assumed that the contribution of the iron ions to the Faraday rotation is equal to the rotation in yttrium IG).

It follows thus from the obtained experimental data that if α_{Fe} is assumed to be equal to the Faraday effect in yttrium IG, the contribution to the Faraday effect from the RE sublattice, in all the investigated REIG, just as in the Tb IG,¹ differs from zero in an external field equal to H_M^0 , when the magnetization of this sublattice vanishes. This points to a significant role, in the Faraday rotation of the RE sublattice,

of the mixing mechanism and of the diamagnetic term [the second term in the right-hand side of (26)]. We emphasize once more that this mechanism was observed by us directly in an investigation of Tb gallium garnet, where the absence of an iron sublattice permits a more accurate analysis of the situation.

We can derive from the experimental data the quantities that determine the paramagnetic mechanism of the REIG Faraday effect and the mixing mechanism. From (26) we have for the spontaneous Faraday effect of the RE sublattice

$$\frac{\alpha_{R^s}}{H_M^0} = \pm \left(C \frac{M_R^s}{H_M^0} + D \frac{g_J}{g_J - 1} \right) \quad (28)$$

TABLE I. Molecular H_M^0 and critical (H_{cr}) fields of REIG at 290 K.

Garnet $R_3Fe_5O_{12}$	H_M^0 , kOe [17]	H_{cr} , kOe (exp)	H_{cr} , kOe (theor. from Eq. (31))
R=Gd	300	~1000 *	400
Tb	160	55	59
Dy	120	55	57
Er	80	-150 *	100
Tm	90	~270 *	170
Yb	105	~400 *	440

*—values obtained by linear extrapolation of the experimental plots of $\alpha_F(H)$ to $H = H_M^0$.

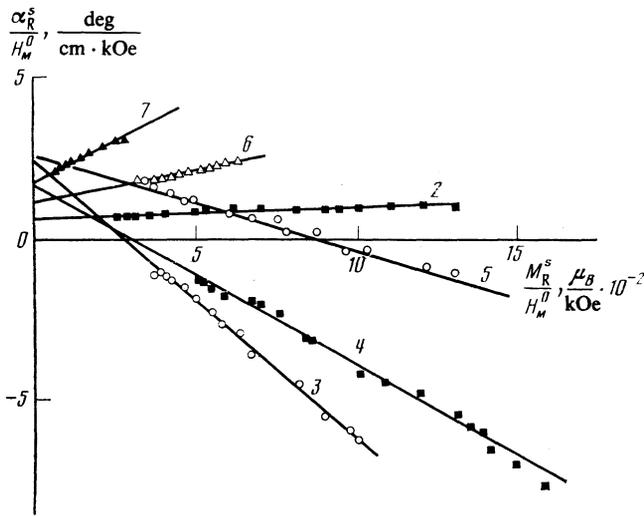


FIG. 4. Dependence of α_R^s/H_M^0 on M_R^s/H_M^0 (where α_R^s and M_R^s are the spontaneous Faraday rotation and the magnetization of the rare-earth sublattice, respectively, and H_M^0 is the molecular field acting on the ground level of the RE ion and produced by the iron sublattice). The designations are the same as in Fig. 1.

(the \mp signs pertain respectively to temperatures above and below the compensation point). Thus, by plotting α_R^s/H_M^0 vs M_R^s/H_M^0 we can find C and D (if g_J is known). We have plotted these functions for the investigated⁴⁾ REIG (Fig. 4). We used in the calculations the RE sublattice magnetic moments from Refs. 17–19 and the molecular fields at room temperature from Ref. 20. The temperature dependence of H_M^0 was used in the form

$$H_M^0 = \lambda M_{Fe}(T), \quad (29)$$

where M_{Fe} is the iron-sublattice magnetization (we have used the data of Ref. 21 for the yttrium IG). It can be seen that for all the investigated REIG, in a sufficiently wide temperature range, α_R^s/H_M^0 is linear in M_R^s/H_M^0 . Table II lists the values of the constants C and D and the temperature regions where the relation (28) holds.⁵⁾ We note that, as can be seen from Table II, the values of the constants C and D are close to the values of these constants for Tb gallium magnet. It must be recognized that a contribution to the paramagnetic term is made also by the gyromagnetic Faraday effect.⁴ Its magnitude is

$$\alpha_m^s = C_m M_R = (\pi n e / m c^2) g_J M_R^s, \quad (30)$$

TABLE II.

IG $R_3Fe_5O_{12}$	C , deg cm · μB	C_m , deg cm · μB	C_e , deg cm · μB	g_J	$\frac{g_J D}{g_J - 1}$, deg cm · μB	D , deg cm · kOe	Tem- perature interval, K	Magne- tization data from Refs.
R=Gd	5	9.1	-4.1	2	0.5	0.25	300-70	[18]
Tb	-89	6.9	-95.9	$3/2$	2.5	0.83	300-90	[19]
Tb	-90	6.9	-96.9		2.1	0.7	300-90	[18]
Dy	-56	6.1	-62.1	$4/3$	1.6	0.4	300-70	[17]
Dy	-60	6.1	-66.1		1.8	0.45	300-70	[18]
Er	-28	5.5	-33.5	$6/5$	2.6	0.43	300-30	[18]
Tm	20	5.3	14.7	$7/6$	1.1	0.16	300-30	[18]
Yb	40	5.2	34.8	$8/7$	1.7	0.21	300-70	[18]
Tb ₃ Ga ₅ O ₁₂ [9]	-94	5.7	-99.7	$3/2$	-	0.47	300-77	-

where n is the average refractive index, e and m are the charge and mass of the electron, and c is the speed of light in vacuum. The values of the constants, C_m , which describe this contribution, and also of the constants $C_e = C - C_m$, which are responsible for the electro-dipole (gyroelectric) contribution to the Faraday effect, are given in Table II.

Knowing the values of C and D we can obtain the critical field H_{cr} in which the Faraday effect due to the RE sublattice vanishes. From (3a), (7), and (26) it follows that

$$H_{cr} = H_M^0 \left(1 + \frac{D}{C} \frac{H_M^0}{M_R^s} \frac{g_J}{g_J - 1} \right) \left(1 + \frac{D}{C} \frac{H_M^0}{M_R^s} \right). \quad (31)$$

We use here the molecular field approximation and assume that at sufficiently high temperatures the susceptibility of the RE sublattice does not depend on the field and is equal to

$$\chi = M_R^s / H_M^0. \quad (32)$$

The theoretical values of H_{cr} were calculated from (31) for 290 K and are given in Table I. The experimental and theoretical H_{cr} plots for the investigated REIG are compared in Fig. 5. In other words, we are dealing with comparison of results of two independent experiments—of the temperature and field dependences of the Faraday effect within the framework of the proposed theoretical model. It can be seen from Fig. 5 that for certain REIG (Tb, Dy, Er) allowance for the mixing makes it possible to describe sufficiently well the experimental situation. For other REIG (Gd, Tm, Yb) allowance for the mixing does not lead to quantitative agreement between the experimental and theoretical and $H_{cr}(T)$ plots. Thus, for example, the values of H_{cr} calculated theoretically for the Gd IG in the temperature interval 100–300 K are 450–500 kOe, whereas from the experimental data it follows that they exceed 1000 kOe. For the Yb IG, the field $H_{cr}^{theor} = 440$ kOe at room temperature and remains practically constant when the temperature is raised to 100 K, while H_{cr}^{exp} in this temperature interval drops from 400 to 300 kOe. For the Tm IG the field H_{cr}^{theor} is approximately 1.5–2 times larger than H_{cr}^{exp} . We note that this occurs mainly for those REIG whose RE sublattice contribution to the Faraday rotation is small and is comparable with the Faraday effect of the iron sublattice. In this case even small changes of the Faraday rotation of the iron ions compared with the Faraday rotation in yttrium IG will strongly influence the values of H_{cr} . Thus, for example, the assumption that the contribution of the iron ions to the Faraday effect of a gadolinium IG

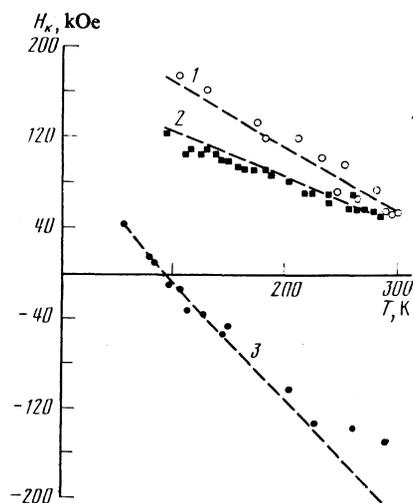


FIG. 5. Temperature dependence of the critical field in various REIG: 1—Tb—Y, 2—Dy—Y, 3—Er. Points—experiment, curves—theory.

exceeds by 20–25% leads to a decrease of the experimental value of H_{cr} from 1000 kOe to a theoretical value 500 kOe. At the same time we can exclude such a change in principle, since experimental data exist from which it follows that replacement of Y by Lu changes the polarization-plane-rotation of the iron sublattices by 10–20% (Ref. 8).

In conclusion, we dwell once more on the physical meaning of the results. We have shown experimentally and theoretically that the contribution of the RE sublattice to the Faraday effect of REIG (regardless of the degree of the influence of the RE sublattice on the contribution of the iron ions to the Faraday effect of the REIG) is determined by two mechanisms of different physical origin: paramagnetic proportional to the RE magnetization M_R of the RE sublattice, and a mixing mechanism proportional to the corresponding effective field (the mixing field H_{eff}^1). The coefficients C_e and D , which characterize the paramagnetic contribution and the contribution from the mixing of the RE sublattice in the iron garnet, also agree in sign and magnitude with the corresponding coefficients of the Faraday rotation of the RE ion in paramagnetic terbium gallate. Attempts to describe the contribution of the RE sublattice by using one term proportional to M_R seem to us therefore not to be correct enough. If it is assumed in this procedure that the contribution of the iron ions remains unchanged, then the coefficient C_e is found (in contradiction to the known physical premise that the properties of the RE ion are constant) to vary greatly in magnitude and even to reverse sign as a function of temperature.²² If, however, C_e is assumed to be independent of temperature,^{8,15} then the contribution from the RE-sublattice mixing mechanism is transformed seemingly into a change of the contribution of the iron ions to the Faraday effect of the REIG. Unfortunately, an experimental comparison of the Faraday properties of RE ions in a paramagnetic crystal and a magnetically-ordered one with structure of garnet was

realized so far only for terbium ions. We regard it therefore as very desirable to carry out similar investigations of crystals with other RE ions.

- ¹The conclusions of this subsection are fully applicable (apart from the frequency dependence) also to magnetic circular dichroism, which is determined by the real part of the off-diagonal component of the tensor ϵ_{ik} .
- ²When account is taken of the diamagnetic contributions to the polarization-plane rotation, the number of different effective fields that determine the phenomenon can be even larger.
- ³This statement is valid under the assumption that the exchange fields acting on the ground and excited multiplets from $4f^n - 15d$ are equal.
- ⁴We do not consider here the Sm and Eu IG, since the $M_R(T)$ dependence is not reliably known for the former, and for the latter the RE contribution to the Faraday rotation is small.
- ⁵The values of the parameters C and D differ somewhat when data from different papers on REIG magnetization are used. It is shown in Table II for the Tb and Dy IG how the parameters C and D change when different data on the magnetization are used.

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