

# Faraday effect in terbium-ytterbium iron garnets in strong magnetic fields

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The Faraday effect in terbium-yttrium iron garnets was measured at wavelengths 1.15 and 0.63  $\mu\text{m}$  in pulsed magnetic fields up to 200 kOe in the temperature interval 80–300 K. It was observed that above the magnetic-compensation temperature the Faraday rotation in fields stronger than the technical-magnetization field decreases linearly with the field, owing to the decrease of the magnetic moment of the rare-earth sublattice in the field. The field  $H_{cr}$  in which the Faraday effect in iron garnets with terbium is equal to the Faraday rotation in yttrium iron garnet is approximately equal, at nitrogen temperatures, to the molecular field  $H_M^0$  in which the magnetic moment of the terbium sublattice vanishes. When the temperature is raised, however,  $H_{cr}$  becomes considerably less than  $H_M^0$ . Various possible causes of this phenomenon are considered, viz., the nonadditivity of the contributions of the different lattices to the Faraday effect, or the difference between the Faraday rotation due to the iron sublattices in iron garnets with terbium and the Faraday rotation in yttrium iron garnet. The most probable mechanism that leads to the difference between  $H_{cr}$  and  $H_M^0$  is considered, namely: the difference between the molecular field  $H_M^0$  that acts on the ground multiplet of the rare-earth ions, on the one hand, and the molecular fields that act on the excited multiplets, on the other, as well as the difference between the intermixing wave functions of the ground and excited multiplets.

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Although magneto-optical phenomena in rare-earth iron garnets (REIG) are being intensively investigated, some essential features of these phenomena remain unclear to this day. One of the unanswered questions is that of the dependence of the Faraday effect in REIG on the magnetic field. Since the REIG are three-sublattice ferrimagnets, the Faraday rotation of the light-polarization planes in them is described usually as a sum of additive contributions of the different magnetic sublattices<sup>1</sup>:

$$\alpha_F = \pm (AM_{Fe_a} - BM_{Fe_d} + CM_R), \quad (1)$$

where  $M_{Fe_a}$  and  $M_{Fe_d}$  are the magnetic moments of the iron in the octahedral (*a*) and tetrahedral (*d*) sublattices, while  $M_R$  is the magnetic moment of the rare-earth in the dodecahedral (*c*) sublattice. Equation (1) is valid in fields stronger than the saturation field, when the magnetic moments of the sublattices are collinear with the field direction. The plus and minus signs in front of the parenthesis pertain to the temperature regions below and above the magnetic-compensation temperature  $T_{comp}$ , respectively. These signs reflect the orientations of the magnetic moments of the sublattices relative to the field direction.

According to (1), the field dependence of the Faraday effect in REIG is due to the field dependences of the magnetic moments of the sublattices. The principal contribution to  $\alpha_F(H)$  is made by the field dependence of the magnetic moment  $M_c$  of the rare-earth sublattice, since the molecular field  $H_M^0$  produced at this sublattice by the iron sublattices is weak<sup>1</sup> (of the order of  $10^5$  Oe), so that the external field influences strongly the magnetic moment of the rare-earth sublattice. Measurements on yttrium iron garnets have shown that the contribution to the Faraday effect by the magnetic moments of the iron sublattices also has a field dependence due mainly not to the magnetization of the iron by the external field, but to the field-dependent change of the spin-

orbit splitting of the iron-ion levels.<sup>2</sup>

We consider now in greater detail the field dependence of the contribution made to the Faraday effect in REIG by the rare-earth ions. Below the magnetic compensation point, the magnetic moment of the rare-earth sublattice  $M_R$  exceeds the magnetic moment  $M_{Fe_d} - M_{Fe_a}$  of the resultant iron sublattice, and is oriented along the field, i.e., the molecular field is parallel to the external field. Above  $T_{comp}$  the moment is  $M_R < M_{Fe_d} - M_{Fe_a}$  and is antiparallel to the field; consequently the molecular and external fields are likewise antiparallel. Thus, the total effective field acting on the rare-earth sublattice is equal to

$$H_{eff}^r = H_M^0 \pm H \quad (2)$$

(the plus and minus signs pertain to respective temperatures below and above  $T_{comp}$ ). Below the magnetic-compensation point, the effective field increases with increasing external field, and this leads to an increase of  $H_{eff}^0$  and, consequently, according to (1), to an increase of the Faraday rotation. Above  $T_{comp}$  (this is the region of greatest interest to us) an increase of the external field leads to a decrease of  $H_{eff}^0$ , and in an external field equal to the molecular field the effective field vanishes, and consequently the magnetic moment of the rare-earth sublattice also vanishes in such a field. Thus, if relation (1) is valid, the contribution made by the rare-earth sublattice to the Faraday rotation at  $T > T_{comp}$  decreases with increasing field, and in the critical field  $H_{cr} = H_M^0$  it is determined only by the magnetic moments of the iron sublattices. Assuming, as is done in many papers,<sup>1,3</sup> that the iron-induced rotation of the polarization plane in the REIG is equal to the Faraday rotation in the yttrium iron garnet, we find that in the field  $H_M^0$  the Faraday effect in REIG should be equal to the Faraday effect in the yttrium iron garnet.

At present there is practically no information on the Faraday effect in REIG in strong fields. Measurements

of the Faraday rotation in terbium iron garnets were made in Refs. 3 in pulsed magnetic fields up to 1200 kOe at a wavelength  $1.15 \mu\text{m}$  at room temperature, and it was observed that in the entire investigated field range the rotation of the polarization plane in terbium iron garnet is larger than in the yttrium garnet, i.e., the Faraday effect due to the terbium sublattice does not vanish up to 1200 kOe. It follows from the magnetic data, however, that the molecular field acting on the terbium sublattice in the iron garnet is 120–160 kOe at room temperature,<sup>4,5</sup> and consequently it is precisely in such fields, according to (1), that the rotation of the polarization plane in the terbium iron garnet should become equal to the Faraday effect in the yttrium iron garnet.

To clarify the cause of this contradiction and to make more precise the character of the field dependence of the rotation of the light polarization plane, we measured the Faraday effect of single-crystal terbium yttrium garnets  $\text{Tb}_x\text{Y}_{3-x}\text{Fe}_5\text{O}_{12}$  in magnetic fields up to 200 kOe in the temperature interval 80–300 K at  $1.15 \mu\text{m}$  wavelength. In addition, measurements of the Faraday effect at  $0.63 \mu\text{m}$  were made for yttrium iron garnet ( $x=0$ ), terbium iron garnet ( $x=3$ ), as well as terbium yttrium garnet with  $x=0.2$ .

The rotation of the plane of polarization was measured in pulsed magnetic fields with approximate pulse duration 15 msec by the usual intensity method,<sup>6</sup> on plates cut along the  $\{111\}$  crystal plane  $250\text{--}350 \mu\text{m}$  thick at  $\lambda = 1.15 \mu\text{m}$  and  $60\text{--}80 \mu\text{m}$  thick at  $\lambda = 0.63 \mu\text{m}$ . The light sources were LG-126 and LG-52/1 helium-neon lasers. The error in the measurement of the absolute magnitude of the rotation of the polarization plane in pulsed magnetic fields was 10–15%, and the dependences on the field and on the temperature were determined accurate to 3–5%. To determine more accurately the absolute magnitude of the Faraday effect, we measured the rotation of the plane of polarization in the same samples in constant fields up to 10 kOe, and then "fitted together" the measurements in strong fields with these data. We were thus able to decrease the Faraday-effect measurement error in pulsed fields to 5–7%.

Figure 1 shows the field dependences of the Faraday effect in terbium-yttrium iron garnets at room temperature and at the wavelength  $1.15 \mu\text{m}$ . It is seen that saturation is reached in weak fields, and with further increase of the field the rotation of the plane of polarization decreases linearly. This, as noted above, can be attributed to the decrease, in the field, of the magnetic moment of the terbium sublattice. The Faraday effect in the terbium iron garnet becomes equal to the Faraday rotation in an yttrium iron garnet in a field  $H_{\text{cr}} = 60$  kOe; approximately the same values of  $H_{\text{cr}}$  are observed for mixed terbium-yttrium iron garnets.

Our measurements have shown that the Faraday rotation in terbium iron garnet at wavelength  $0.63 \mu\text{m}$  has a similar field dependence. In this case  $H_{\text{cr}}$  is also close to 60 kOe.

Our data contradict the results of Refs. 3, from which,

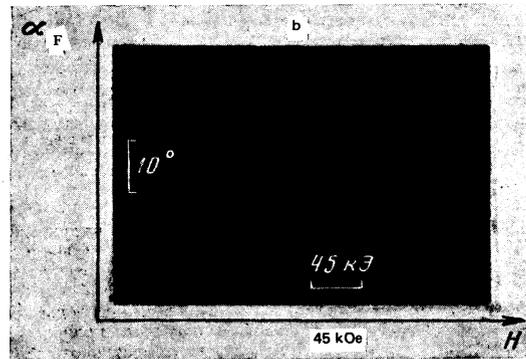
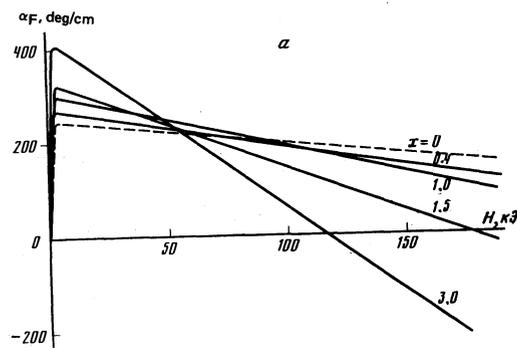


FIG. 1. a) Dependence of the Faraday effect of terbium-yttrium iron garnets on the magnetic field at 200 K and different values of  $x$ . b) Oscillogram of the dependence of the Faraday rotation of  $\text{Tb}_3\text{Fe}_5\text{O}_{12}$  on the magnetic field. Sample thickness  $d = 380 \mu\text{m}$ .

as indicated, it follows that the Faraday effect in terbium iron garnet is larger than in yttrium iron garnet in fields up to 1200 kOe. The reasons for this are not clear. It is possible that the unusual  $\alpha_F(H)$  dependence obtained in Refs. 3 is due to the experimental conditions: the pulsed fields were generated in these studies by explosion, were short in duration ( $\sim 10 \mu\text{sec}$ ), and it is possible that in this case the relaxation processes have become substantial in the course of magnetization.

We have determined  $H_{\text{cr}}$  for terbium iron garnet only in a narrow temperature interval near room temperature, since the compensation point of this ferrimagnet is high ( $\sim 250$  K). When the terbium is replaced by yttrium, the compensation point shifts towards lower temperatures and it is possible to trace the temperature dependence of  $H_{\text{cr}}$ . By way of example, Fig. 2 shows plots of  $\alpha_F(H)$  of terbium-yttrium iron garnets with  $x = 1.5$  at various temperatures. The figure shows also the field dependences of the Faraday rotation in yttrium iron garnet ( $x=0$ ). With decreasing temperature, the Faraday effect  $\alpha_{F_s}$  of the terbium-yttrium iron garnet increases, as does also the absolute value of the Faraday susceptibility  $d\alpha_F/dH$ , which characterizes the change of the Faraday rotation in the field, but not to the same degree as  $\alpha_{F_s}$ , as a result of which  $H_{\text{cr}}$  increases with decreasing temperature.

The variation of  $H_{\text{cr}}$  with temperature is approximately the same for other terbium-yttrium iron garnets and, as seen from Fig. 3, within the limits of errors, the values of  $H_{\text{cr}}(T)$  for all the investigated samples fall on one straight line.

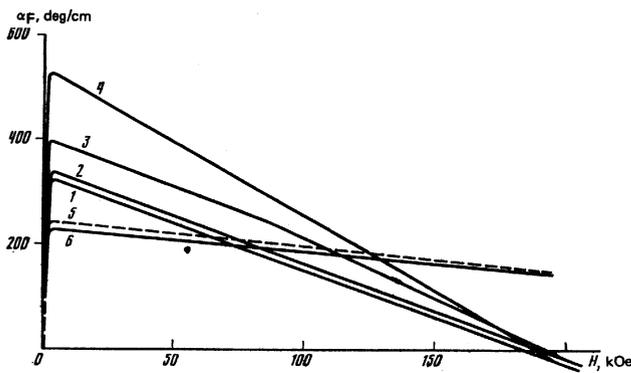


FIG. 2. Dependence of the Faraday effect of the iron garnets  $Tb_x Y_{3-x} Fe_5 O_{12}$  on the magnetic field at various temperatures  $x = 1.5$ : 1—290 K, 2—260 K, 3—230 K, 4—183 K.  $x = 0$ : 5—290 K, 6—180 K.

The value of  $H_{cr}$  of terbium iron garnet can also be estimated from measurements of the Faraday effect in weak fields. Such measurements were made in Refs. 1–3, and the field dependence of the Faraday rotation was linearly extrapolated to the strong-field region. The data obtained in this manner agree with the results of our measurements in pulsed fields (see Fig. 3).

As indicated above, if relation (1) is valid (with allowance for the change of the Faraday rotation of the iron sublattices in the field on account of the influence of the field on the spin-orbit splitting), the field  $H_{cr}$  should equal the molecular field  $H_M^0$ . Figure 3 shows the temperature dependences of  $H_M^0$  of terbium iron garnet, plotted from magnetic-measurement data.<sup>4,5</sup> It is seen that at low temperatures the field  $H_{cr}$  is close to  $H_M^0$ . However, with increasing temperature  $H_{cr}$  decreases more rapidly than the molecular field, and at room temperature  $H_{cr}$  is considerably weaker than  $H_M^0$ .

Various causes of the difference between  $H_{cr}$  and  $H_M^0$  can be proposed. First, in the analysis of the experimental data we have assumed that in iron garnets with terbium the rotation of the light-polarization plane as a result of the iron sublattices is equal to the Faraday effect in the yttrium iron garnet. Yet it is known that this contribution to the Faraday rotation can change when the yttrium is replaced by other ions. This manifests itself most strongly when diamagnetic bismuth,<sup>7</sup>

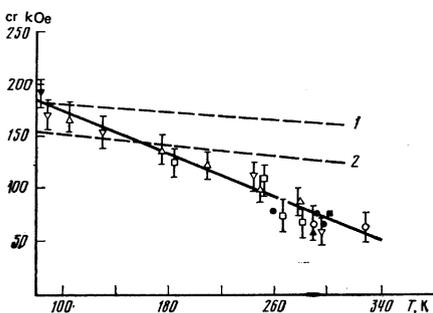


FIG. 3. Dependence of the field  $H_{cr}$  of terbium-yttrium iron garnets on the temperature. Our data:  $\blacktriangle$ ,  $\circ$  —  $x = 3, 0$ ;  $\square$  —  $x = 1, 5$ ;  $\triangle$  —  $x = 1, 0$ ;  $\nabla$  —  $x = 0, 4$ ;  $\blacktriangledown$  —  $x = 0, 2$  (light symbols— $\lambda = 1.15 \mu m$ , dark symbols— $\lambda = 0.63 \mu m$ ). Published data for composition with  $x = 3$ :  $\bullet$  — [1],  $\circ$  — [2],  $\blacksquare$  — [3]. 1, 2 temperature dependences of the molecular field  $H_M$ : 1—from Ref. 5, 2—from Ref. 4.

as well as the rare-earth praseodymium,<sup>8</sup> is introduced in place of yttrium.<sup>2)</sup> It is possible that the introduction of terbium also leads to a change of the Faraday rotation of the iron sublattices. To explain why the difference between  $H_{cr}$  and  $H_M^0$  is largest at room temperature and decreases with decreasing temperature it is then necessary to assume that this effect becomes stronger at higher temperatures. This may be due to the influence of the  $Tb^{3+}$  ions on the magneto-optics of the  $Fe^{3+}$  ions via the crystal-lattice vibrations (phonons), which lower the spin and parity forbiddenness of the optical transitions in the  $Fe^{3+}$  ions. Since the terbium atoms have almost double the mass of the yttrium atoms, the phonon spectra of terbium and yttrium iron garnets are different, and this leads to different Faraday rotation of the iron sublattices in these garnets. However, data<sup>10</sup> on the influence exerted on the Faraday effect by replacing the yttrium in the iron garnet by the nonmagnetic lutecium whose atoms are close in mass and dimension to the terbium atoms, indicate that this effect is small.<sup>3)</sup>

The second possible cause of the field dependence of  $\alpha_F(H)$  observed in terbium-yttrium iron garnets is the non-additivity of the contributions of the individual sublattices to the Faraday effect (see Eq. (1)). It was shown recently<sup>11</sup> that the optical absorption in the near ultraviolet in yttrium iron garnet is not the sum of the processes of single-ion light-absorption by the  $Fe^{3+}$  ions in the  $a$  and  $d$  sublattices, but is determined by two-ion charge transfer from one sublattice to the other.<sup>12</sup> It is possible that the rare earth ions become involved in the multi-ion absorption processes together with the iron ions, and then the optical effects, and hence also the magneto-optical effects, cannot be combined additively in the REIG from the individual sublattices. One of the mechanisms whereby the magnetic field influences the optical rotation is then the effect of the field on the probability of charge transfer between the sublattices.

Finally, it is possible, and this seems most probable to us, that the Faraday effect due to the rare-earth ions cannot be regarded as proportional to the magnetic moment of the rare-earth sublattice, as is customarily done (see Eq. (1)). This proportionality stems from consideration of the so-called paramagnetic contribution to the Faraday effect by the difference in the populations of the levels of the ground multiplet of the rare-earth ion. In the general case it is necessary to take into account also the so-called diamagnetic Faraday effect due to the splitting of the ground and excited multiplets in the effective field. An important role is played also by the contribution made to the Faraday rotation by the mixing of the wave functions of the ground and excited multiplets under the influence of the effective field (see Refs. 13–15 as well as the reviews 16 and 17).

Taking into account all these mechanisms, we can represent the Faraday effect due to the rare-earth ions in the following form<sup>18</sup>:

$$\alpha_F^R = \pm \left( CM_R + \sum_i d_i H_{eff}^i \right), \quad (3)$$

where  $H_{\text{eff}}^i$  is the effective field acting on the  $i$ th multiplet or intermixing the wave functions of the ground and  $i$ th multiplets, while the summation is over all the multiplets. Here and below the plus and minus signs pertain to temperatures below and above the compensation point, respectively. The coefficients  $C$  and  $d_i$  are practically independent of temperature.

It is customarily assumed that the dimagnetic contribution and the contribution to the mixing of the wave functions are also proportional to the magnetic moment (see, e.g., the Pisarev's review<sup>17</sup>). This is permissible when the Faraday effect of paramagnets is considered.<sup>4</sup>) In the case of REIG the situation is more complicated because the effective fields acting on the excited multiplets of the rare earth ions and intermix the wave functions of the ground and excited multiplets differ from the effective field  $H_{\text{eff}}^0$  acting on the ground multiplet.

In fact, the total effective field acting on the  $i$ th multiplet is made up of the molecular field acting on this multiplet and the external field, and can be represented in the same form as the effective field acting on the ground multiplet (see Eq. (2)):

$$H_{\text{eff}}^i = H_M^i \pm H. \quad (4)$$

But  $H_M^i$  can be expressed in terms of the exchange field  $E_{\text{exc}}$  exerted by the iron sublattices on the spin of the given multiplet (the exchange field can apparently be assumed to be the same for the different multiplets) in the following manner:

$$H_M^i = \frac{2S_i}{g_i J_i} H_{\text{exc}} = \frac{2(g_i - 1)}{g_i} H_{\text{exc}}, \quad (5)$$

where  $g_i$ ,  $S_i$ , and  $J_i$  are respectively the Landé factor, the spin, and the total mechanical angular momentum of the  $i$ th multiplet. (Formula (5) is valid in the Russell-Saunders approximation.) Thus, the molecular fields and consequently also the effective fields acting on the different multiplets are different.<sup>5</sup>)

In the analysis of the contribution made to the Faraday effect by the mixing of the wave functions it suffices to consider only the lowest excited multiplets belonging to the same term as the ground multiplet (i.e., to the term with the given  $S$  and  $L$ ). Then the matrix elements of the Zeeman Hamiltonian, which connect the ground and excited multiplets all contain the same factor ( $2H_{\text{exc}} \pm H$ ). In fact, the matrix elements of the Zeeman Hamiltonian

$$\begin{aligned} \mathcal{H}_z &= -\mu_B(L+2S)H - 2\mu_B H_{\text{exc}} S \\ &= -\mu_B(J+S)H - 2\mu_B H_{\text{exc}} S \end{aligned} \quad (6)$$

which are needed for the calculation of  $\alpha_F^R$ , take the form

$$\langle LSJM | \mathcal{H}_z | LSJ'M' \rangle = -\mu_B (2H_{\text{exc}} \pm H) \langle LSJM | S_z | LSJ'M' \rangle, \quad (7)$$

for if  $J \neq J'$  then  $\langle LSJM | J_z | LSJ'M' \rangle = 0$ . Expressing  $H_{\text{exc}}$  in terms of  $H_M^0$  (see Eq. (5)) we find that the contributions from the mixing of the wave functions are proportional to  $(g_0 H_M^0 / (g_0 - 1) \pm H)$ . For example, for the  $\text{Tb}^{3+}$  ion ( $g_0 = \frac{3}{2}$ ) these contributions are proportional to  $3H_M^0 \pm H$ . We note that the Faraday-effect terms due to the mixing are analogous to the known Van Vleck contribution to the equilibrium magnetic moment. However, as shown by estimates, their relative contribution to

the Faraday effect can be appreciably larger.<sup>6</sup>)

We have no sufficiently reliable data on the energy spectrum and the wave functions of the  $\text{Tb}^{3+}$  ion in the iron garnet, and shall therefore not perform a microscopic calculations, confining ourselves only to a phenomenological analysis.

We put

$$H_M^i = k_i H_M^0. \quad (8)$$

At sufficiently high temperatures the magnetic moment of the rare-earth sublattice can be represented in the form

$$M_R = \chi_R (H_M^0 \pm H), \quad (9)$$

where  $\chi_R$  is the susceptibility of the rare-earth sublattice. We then find from (3) that the field  $H_{\text{cr}}$  at which the contribution from the rare-earth sublattice to the Faraday effect vanishes is equal to

$$H_{\text{cr}} = H_M^0 \frac{C\chi_R + \sum_i d_i k_i}{C\chi_R + \sum_i d_i} \approx H_M^0 \left[ 1 + \sum_i \frac{d_i (k_i - 1)}{C\chi_R} \right]. \quad (10)$$

In the derivation of the last formula we used the inequalities  $|d_i k_i| \sim |d_i| \ll C\chi_R$ .

Thus,  $H_{\text{cr}} \neq H_M^0$  in REIG. In other words, the contribution of the rare-earth sublattice to the Faraday effect becomes equal to zero at nonzero magnetic moment of sublattice. To the contrary, when the magnetic moment of the rare-earth sublattice becomes equal to zero, the contribution from this sublattice to the Faraday rotation does not vanish. All this agrees with our experimental data on the dependence of the Faraday effect of terbium-yttrium iron garnets on the magnetic field.

It follows also from the theory that when the temperature is lowered  $H_{\text{cr}}$  tends to  $H_M^0$ , inasmuch as in this case the magnetic moment and susceptibility of the rare-earth sublattice increase, and consequently the second term in the square brackets of (10) decreases. Since the susceptibility of the rare-earth sublattice of most rare-earth iron garnets (including the terbium iron garnet) obey in a wide temperature range the Curie-Weiss law<sup>4,20</sup>:

$$\chi_R = C_R / (T - \theta), \quad (11)$$

and the quantities  $C$ ,  $d_i$ ,  $k_i$ , and  $H_M^0$  are practically independent of temperature, it follows from (10) that  $H_{\text{cr}}$  should increase linearly with decreasing temperature, as is in fact observed in experiment (see Fig. 3).

The considered theory explains also some other experimentally observed features of the Faraday effect in REIG. From Eq. (3), taking (8) and (9) into account, we obtain for the spontaneous Faraday effect due to the rare-earth sublattice

$$\alpha_F^R = \pm \left( CM_R + H_M^0 \sum_i d_i k_i \right). \quad (12)$$

It follows therefore from (9) that

$$\alpha_F^R / M_R = \pm \left( C + \frac{1}{\chi_R} \sum_i d_i k_i \right). \quad (13)$$

Taking into account the expression (11), as well as the independence of the coefficients  $C$ ,  $d_i$ ,  $k_i$  of the temperature, we find that according to the considered model  $\alpha_{Fs}^R/M_{Rs}$  should vary linearly with temperature. Such a dependence is indeed observed for a number of REIG, including  $Tb_3Fe_5O_{12}$  (Refs. 3 and 21), if it is assumed that the contribution of the irons sublattices to the Faraday effect in these garnets is equal to the Faraday rotation in  $Y_3Fe_5O_{12}$ .

Thus, allowance for the influence of the effective fields on the splitting of the excited multiplets and for the mixing of the wave functions of the ground and excited multiplets allows us to describe the Faraday effect in terbium-yttrium iron garnets in a wide range of fields and temperatures. There are, however, as already indicated, other possibilities of explaining the observed phenomena. For further refinement of the various mechanisms of the Faraday effect in REIG it is necessary, in our opinion, to estimate first of all more accurately the contribution made to this effect by the iron sublattices. In addition, it is important to establish the degree to which the regularities observed for terbium-yttrium iron garnets are general and satisfied for other REIG. We note in this connection that, as shown by our measurements, the Faraday rotation in dysprosium-yttrium iron garnets in strong fields behaves in analogy with the Faraday effect in the terbium-yttrium iron garnets.

<sup>1)</sup>The superscript 0 indicates that in this case is meant the molecular field acting on the ground multiplet of the rare-earth ion, since the magnetic moment is determined by the splitting, in the field, of the levels of just this multiplet.

<sup>2)</sup>It is shown in Ref. 9 that introduction of bismuth and praseodymium changes the character of the optical transitions in the iron ions in the garnets in the near-infrared.

<sup>3)</sup>There is also one other trivial reason for the difference between the Faraday effect of the iron sublattices in rare-earth iron garnets and of yttrium iron garnet. As already noted, depends on the field. But in REIG the iron ions are acted upon, besides the external field, also by the molecular field due to the rare-earth sublattice, i.e., the iron ions in REIG are in a different effective field than in an yttrium

iron garnet. Calculations show, however, that the change produced in  $H_{cr}$  by this mechanism is within the limits of the measurement error.

<sup>4)</sup>In fact, in this case  $H_{cr}^i = H$ , for all multiplets and we can express  $H_{cr}^i$  in terms of  $M$  from the usual relation  $M = \chi H$  for paramagnets.

<sup>5)</sup>The contribution proportional to the external field to the Faraday rotation of REIG, due to the influence of the field on the excited states, was introduced in Ref. 19.

<sup>6)</sup>For some rare-earth ions whose excited multiplets are close to the ground multiplet (for example the distance between the ground and first-excited multiplets of  $Tb^{3+}$  is approximately  $2000\text{ cm}^{-1}$ ) this contribution can reach 10–20% of the paramagnetic contribution to the Faraday effect.

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