

FARADAY EFFECT AND REVERSAL OF MAGNETIC SUBLATTICES OF RARE-EARTH
IRON GARNET

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An investigation of the Faraday effect (FE) in holmium iron garnets near the temperature of magnetic compensation in pulse magnetic fields up to 160 kOe revealed jumplike changes of the FE; these are attributed to the reversal of the magnetic sublattices of the iron garnet. An estimate is presented of the effective field acting on the total magnetic moment of the rare-earth iron, $H_{\text{eff}} = 60 \pm 10$ kOe at $T = 136^\circ\text{K}$. In addition, a sharp increase of the FE in a field ~ 135 kOe is reported at a temperature below 56°K , and the hypothesis is advanced that it is connected with the destruction of the umbrella-like magnetic structure of the holmium iron garnet in an external magnetic field.

IN rare-earth iron garnets, the Faraday effect (FE) can be regarded as a sum of independent contributions of all the sublattices of the ferrites. In the infrared region of the spectrum ($\lambda > 4 \mu$), the contributions are due essentially to precession of the magnetic moments of the sublattices under the action of the magnetic field of the light wave, i.e., they are the consequence of exchange resonance^[1-3], and at shorter wavelengths, the FE of the sublattices is connected with electronic transitions both inside the $3d^5$ shells (Fe^{3+} ions) and the $4f^n$ shells (Re^{3+} ions), and with the stronger transitions between the different electronic configurations^[4]. At an incident-light frequency sufficiently remote from the fundamental absorption, the magnetization and the FE of rare-earth ions and transition-metal ions in the ground S-state are proportional to each other, with the proportionality coefficient that does not depend on the temperature^[5]. The distance between these frequencies, in the case of the visible part of the spectrum, is large for almost all the rare-earth ions, unlike the case of Fe^{3+} ions in iron garnets. The fundamental absorption in the latter, starting already in the vicinity of $20,000 \text{ cm}^{-1}$, and the existing intense broad absorption band at frequencies 11,000 and $16,400 \text{ cm}^{-1}$ ^[6], cause the dependence of the FE of the Fe^{3+} ions in the visible region of the spectrum on the temperature to be determined not only by their magnetization, but also to a larger degree by the change of the width and intensity of the bands, and also by the shift of the edge of the fundamental absorption upon cooling^[6]. In addition, just as in ferromagnetic d-metals^[7], a strong dependence of the FE of the iron sublattices on the magnetic field intensity is observed in yttrium iron garnet^[8], this is being apparently connected with the peculiarities of the occurrence of the EF in ions with a ground S-state¹⁾.

¹⁾The FE in these ions is determined completely by the frequency separation $\Delta\nu$ between the centers of gravity of the absorption of right- and left-hand circularly polarized light, which in turn is determined by the spin-orbit interaction in the excited state^[5]. Since in iron garnets the resultant FE of all the iron ions is the difference of the contributions of two iron sublattices, noticeable relative changes of FE are possible already at small changes of $\Delta\nu$. On the other hand, a decrease of $\Delta\nu$ is possible under the influence of an external magnetic field acting also on the orbital angular momentum of the ion in the excited state.

Nevertheless, at constant temperature and constant magnetic field intensity, the FE of the iron ions will be proportional to the projection of their magnetization on the light-propagation direction. Opposite signs of the FE of the rare-earth sublattice and the resultant FE of the sublattices of the iron ions at identical direction of their magnetization vectors, and also the different values of the FE, bring about a situation wherein the magnetic moments are cancelled but not the FE. This makes it possible, by investigating the FE, to supplement the information obtained from magnetic measurements concerning the magnetic structure of the garnet^[1]. Thus, on going through the magnetic-compensation temperature T_c , a sharp reversal of the sign of the FE is observed, connected with the change of the direction of the magnetization vectors of the sublattices^[1]. Unique changes of the FE can occur also in the case of "breaks" of the sublattices^[9-12], a detailed analysis of which can help determine the direction of the magnetic-moment vectors of the sublattices as functions of the external field.

In the present investigation we measured the FE in holmium iron garnet at temperatures close to T_c , in pulsed magnetic fields up to 160 kOe, and at a light wavelength 6,328 Å. The results allow us to conclude that the "break" of the sublattice actually does take place and at temperatures close to T_c it has the character of a reversal of the magnetic moments. In addition, the FE was measured also at lower temperatures ($T = 24^\circ\text{K}$), where both the non-collinear magnetic structure of the holmium iron garnet and its destruction in strong magnetic fields can exist.

Figure 1 shows oscillograms demonstrating the change of the intensity of the light passing through a plate of holmium garnet placed between polarizers as the magnetic field intensity increases from zero to 80 kOe within a time 3.5×10^{-3} sec. The horizontal sweep on the oscillograms is proportional to the field intensity, and the vertical one is proportional to the Faraday angle of rotation of the plane of polarization (the angle between the polarizers is 45°). The fact that the observed changes of the intensity are connected with the change of the FE is confirmed by direct measurements of the angle of rotation of the plane of polariza-

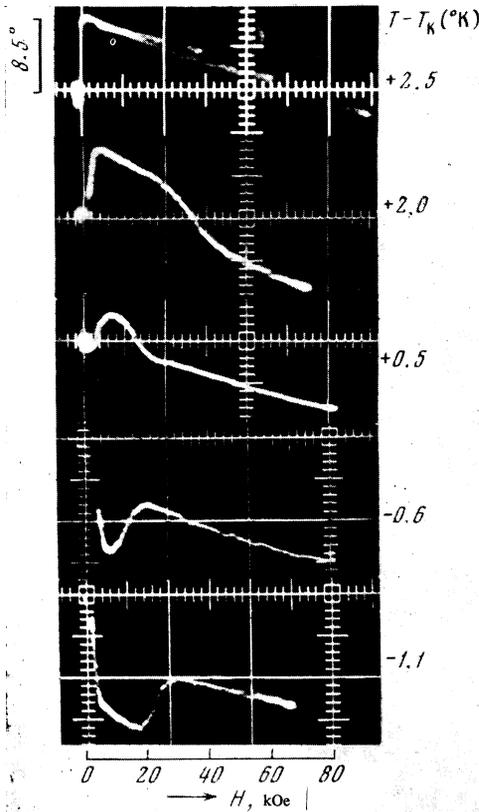


FIG. 1. Oscillograms of the dependence of the Faraday rotation on the magnetic field intensity in the garnet $\text{Ho}_3\text{Fe}_3\text{O}_{12}$ near the compensation point $T_C = 136^\circ\text{K}$; $d = 100\mu$ (110).

tion, performed in the same field interval by a method described earlier^[13]. The temperature of the sample (plate 100μ thick), cut in the (110) plane, was varied near T_C and was measured with copper-constantan thermocouples glued to the sample.

It is seen from the oscillograms that at temperatures that differ little from $T = 136^\circ\text{K}$ there is observed, besides the usual linear change of the FE with increasing magnetic field ($|d\varphi/dH| \approx 0.12 \text{ deg/kOe}$ at $|\varphi_0| \approx 8.5^\circ$), due to the additional magnetization of the rare-earth sublattices and to the change of the Faraday rotation of the iron ions in the external field, also a sharp decrease of the FE when the critical value of the magnetic field intensity H_1 is reached. The field H_1 increases rapidly with increasing difference $(T - T_C)$.²⁾ The jump-like changes in the absolute value of FE can be connected with the process of the "break" of the sublattices, which at almost equal values of the sublattice magnetization should have the character of a reversal, similar to the reversal of the magnetic moments in antiferromagnets.

The magnetization of the rare-earth sublattice depend strongly on the applied field and therefore even at a temperature very close to T_C , the "break" begins at appreciable deviation of the magnetic moment of the rare-earth sublattice from the resultant magnetic moment of the iron sublattices. However, as the vector

M_{Re} rotates away from the direction of H , the absolute value of M_{Re} is restored to almost the initial value $M_{\text{Re}}(H = 0) \approx M_{\text{Fe}}$, thereby accelerating the initiating rotation of the sublattice. During the process of such a reversal, the projection of the magnetic moments on the direction of H (which coincides with the light propagation direction) decreases greatly, and consequently, there should be a sharp decrease in the absolute value of FE, whereas the dependence of the magnetization on the magnetic field should experience only a small kink. In those iron garnets in which the contribution to the FE from the rare-earth sublattice predominates, during the process of the reversal, at temperatures $T > T_C$, the sign of the FE should change, and it should remain constant when $T < T_C$, although the effect may drop almost to zero (the sign of the FE is determined by the direction of the projection of the vector M_{Re} , which is reversed in the former case and remains constant in the latter). Such a regularity was indeed observed in holmium garnet (Fig. 1).

The critical reversal field for the start of the "break" of the sublattices, H_1 , can be determined by using the expression obtained by Gusev and Pakhomov for an isotropic iron garnet^[10-12]:

$$H_1 = \frac{M_2 - M_1}{\chi} + \Delta H(T), \quad (1)$$

where M_2 —resultant magnetization of the iron sublattices, M_1 —magnetization of the rare-earth sublattice (at $T < T_C$ the signs of M_1 and M_2 should be reversed), ΔH —small addition that depends on the temperature. The susceptibility χ of the "break" process, as shown in^[10,14], does not depend on the temperature and is determined by the magnitude of the exchange interaction of the rare-earth ions with the iron ions:

$$\chi = M_1 M_2 / (N_1 I_{12} N_2 I_{21})^{1/2} S_1 S_2. \quad (2)$$

Here N_1 , S_1 and N_2 , S_2 —numbers of ions and magnitudes of the spins of the ions of the rare-earth and "resultant" iron sublattices, I_{12} and I_{21} —exchange integrals, characterizing the exchange interaction of the ion of one sublattice with all the ions of the other.

In (1), the dependence of M_2 on the external field H can be neglected, since the external fields employed above are much smaller than the internal fields acting on the iron ions. The magnetization of the rare-earth sublattice prior to the start of the break can be described by the Brillouin function^[15]

$$M_1 = M_{10} B_{S_1} \left(\sum_{i=a,d,c} I_i S_i S_i \pm \mu_1 H \right), \quad (3)$$

where $M_{10} = N_1 \mu_1$ and $\mu_1 = \mu_B g_j$ —total magnetic moment of the rare-earth ion, I_i —exchange integral characterizing the interaction of the rare-earth ion with all the ions of the i -th sublattice, and the summation is over all three (a, d, c) sublattices of the iron garnet. The minus sign corresponds to temperatures larger than T_C , and the plus sign to lower temperatures. The sum in (3) can be replaced by $\mu_1 H_{\text{eff}}$, where

$$H_{\text{eff}} = \frac{1}{\mu_1} \sum_{i=a,d,c} I_i S_i S_i \quad (4)$$

is the internal effective field acting on the total magnetic moment of the rare-earth ion and produced by all the surrounding ions. It is connected with H_{exch} , which acts

²⁾ Similar changes of the FE were observed also in erbium garnet near $T_C = 80^\circ\text{K}$.

on the spin moment of the ion, by the relation $H_{\text{eff}} = 2(g_j - 1)g_j^{-1}H_{\text{exch}}^{[16]}$. The susceptibility χ can be expressed in terms of the effective field acting on μ_1 and produced by the iron ions only, which we set equal to H_{eff} , since the exchange interaction between the rare-earth ions is negligibly small, and

$$\chi = M_2 / H_{\text{eff}}. \quad (5)$$

Using (1), (3) and (5) we can calculate the values of the critical field H_1 for different temperatures. Expanding the function B_{S_1} in a series and taking the first two terms of the expansion, we obtain for H_1 the equation

$$H_1 = \pm H_{\text{eff}} \frac{M_{10}}{M_2} \left[\frac{S_1 + 1}{3S_1} x - \frac{(2S_1 + 1)^4}{45(2S_1)^4} x^3 - \frac{M_2}{M_{10}} \right] \quad (6)$$

where $x = \mu_1(H_{\text{eff}} \pm H_1)/kT$. From (6) we can see that, starting with a certain temperature $T' = T_c + \theta$, where

$$\theta = \frac{(2S_1 + 1)^4 - 1}{15(2S_1)^4} \frac{S_1}{S_1 + 1} \left(\frac{\mu_1 H_{\text{eff}}}{kT_c} \right) T_c, \quad (7)$$

and above this temperature, $H_1 = H_{\text{eff}}$, and consequently, no jump of the FE will take place for $T > T'$, since the magnetization of the rare-earth sublattice vanishes at $H_1 = H_{\text{eff}}$, and the appearance of a magnetic moment with further increase of the field does not lead to a "reversal" of the sublattices. Assuming that the magnetic moment of the holmium iron in the garnet at $T = 136^\circ\text{K}$ is equal to the moment of the free ion $\mu_1 = 10 \mu_B$, and taking the value of the resultant magnetization of the iron sublattices, equal to its value in the yttrium garnet $\text{Y}_3\text{Fe}_5\text{O}_{12}$ at this temperature ($M_2 = 4.65 \mu_B$)^[17], we can obtain $H_{\text{eff}}(T = T_c)$, which was found to equal 63 kOe. The estimate of the value of θ at the same values of μ_1 and H_{eff} yields $\theta \approx 1.5^\circ\text{K}$. On the other hand, the jump of the FE cannot be observed at temperatures exceeding T_c by 2.2°K , this being in qualitative agreement with the calculation. It is also interesting, that the magnitude of the abrupt change of the FE does not decrease smoothly to zero when the sample is heated to $T = T'$, but vanishes at T' likewise jumpwise (Fig. 2b), as should be the case upon reversal of the sublattices (the "residual" jump should be somewhat larger than the contribution made to the FE by the iron ions at an external-field intensity $H = H_{\text{eff}}$).

The temperature dependence of the critical field H_1 , obtained from (6), is shown in Fig. 2a. The same figure shows the experimental values of H_1 , which lie somewhat lower than the calculated ones. Somewhat higher calculated values of H_1 may be due to the fact that no account was taken in the calculation of the small term ΔH in (1), which appears when $T \neq 0$ ^[10]. However, the limiting value of H_1 should equal H_{eff} . As follows from Fig. 2a, the largest observable $H_1 = 53$ kOe, and consequently H_{eff} should not exceed this value greatly. The intensity of the external field H_0 , at which the FE vanishes at higher temperatures, gives an upper limit for H_{eff} , provided only that it is assumed that the change of the contribution of the iron sublattices of the holmium iron garnet with changing field differs little from the change of the FE in yttrium iron garnet.

Figure 2 shows the values of H_0 for temperatures at which the jump of the FE is no longer observed. We see that the value of H_{eff} at these temperatures lies in the

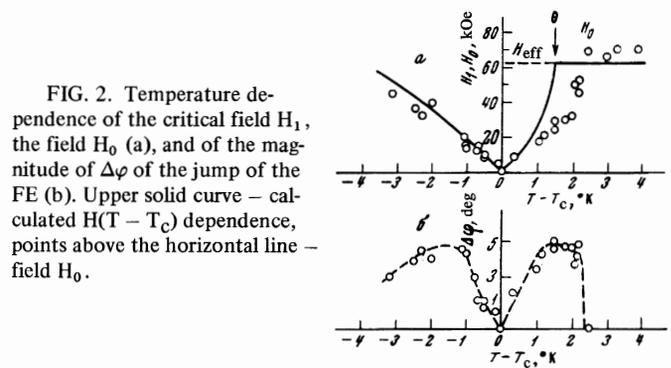


FIG. 2. Temperature dependence of the critical field H_1 , the field H_0 (a), and of the magnitude of $\Delta\varphi$ of the jump of the FE (b). Upper solid curve – calculated $H(T - T_c)$ dependence, points above the horizontal line – field H_0 .

field interval 53–70 kOe, and is closer to the lower limit (the FE of the iron ions does not equal to zero and $H = 70$ kOe). Extrapolation of the data on the susceptibility of the para process of polycrystals of holmium iron garnet^[18] to the temperature $T = 136^\circ\text{K}$ yields $\chi_n = 3.85 \times 10^{-4} \text{g}^{-1}$, which amounts to $0.068 \mu_B/\text{kOe}$ per molecule of $\text{Ho}_3\text{Fe}_5\text{O}_{12}$, and the value $H_{\text{eff}} = M_2/\chi_n = 68$ kOe obtained from this, which also agrees with our results.

In conclusion we note a peculiarity of the behavior of the FE in holmium garnet in strong fields at temperatures below 56°K . Figure 3 shows, besides the dependence of the FE on the magnetic field intensity, obtained by the direct method^[13], oscillograms similar to the preceding ones, the angle between the polarizers being chosen such that the changes of the intensity were proportional to the changes of the Faraday angle in the region of magnetic fields under consideration. The dashed line represents the value

$$\varphi = \varphi_2 + KM_{10} B_{S_1} \left(\frac{\mu_1(H_{\text{eff}} + H)}{kT} \right),$$

which reflects the behavior of the magnetization of the holmium ions subjected to the action of the internal and external fields, under the assumption that the magnetic structure is collinear and the magnetic moments are directed along the field. Here $K = 1.75 \text{deg}/\mu_B$, φ_2 —angle of the FE of the iron ions, which changes with the field in accordance with the law $\varphi_2 = \varphi_{20} + bH$, where $b = 0.02 \text{deg}/\text{kOe}$ at a sample thickness 100μ and a temperature 24°K ^[8], and the internal effective field at this temperature is $H_{\text{eff}} = 100 \text{kOe}$ ^[8, 19]. The magnetic moment of the holmium ion, the magnitude of which at low temperatures can differ from its value for the free ions

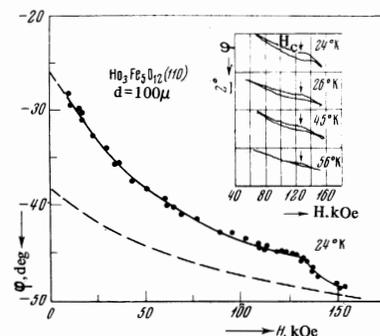


FIG. 3. Singularities in the behavior of the FE in holmium-iron garnet at temperatures below 56°K ($d = 100 \mu$, (110)).

owing to the effect of the quenching of the orbital angular momentum, was chosen equal to $8.6\mu_B$ (the angle at the vertex of the cone made up of the magnetic moments of the holmium ions equals 44° if the magnetization of the garnet per molecule of $\text{Ho}_3\text{Fe}_5\text{O}_{12}$ is $13.6\mu_B$ ^[20]). According to neutron diffraction data, this angle equals 48° ^[21].

It is seen from the figure that at a field intensity $H_c \approx 135$ kOe, starting with a temperature $\sim 56^\circ\text{K}$ and below, the FE experiences a jump which is seen more clearly with decreasing temperature. The magnitude of the critical field for this jump is practically independent of the temperature. A considerable hysteresis is also observed, which vanishes together with the jump upon heating to 56°K . The observed singularity cannot be connected with rotation of the magnetic moment of the ferrite as a whole, since, although the anisotropy field can be large, the technical-magnetization curve for this case (anisotropy constant $K_1 < 0$, $H \parallel [110]$) does not experience a jump, as is the case with iron at $H \parallel [111]$ ^[22]. Nor can it be attributed to the process of the "break" on the sublattices, for in this case there should be observed a decrease of the FE (during the rotation process, a decrease takes place in the projection of the magnetic moment of the rare-earth ion, which makes the main contribution to the FE at low temperatures, on the direction of the external magnetic field). In addition, at $T = 24^\circ\text{K}$ the value of H_1 greatly exceeds 135 kOe.

It is possible that the singularities connected with the destruction of the non-collinear magnetic structure of the holmium garnet, and that there should take place an irreversible process of motion of the magnetic moments of the ions of holmium, which are in non-equivalent positions. As shown by neutron-diffraction^[21] and magnetic^[20] investigations, in the holmium iron garnet at a temperature 4.2°K there is realized a non-collinear umbrella-like magnetic structure. An explanation of the anomalously large magnetostriction of the holmium iron garnet in the region of low temperatures presupposes its presence also at higher temperatures^[23]. If it is assumed that the jump of the FE in a field $H \approx 135$ kOe is due to the "collapse" of the cone of the magnetic moment, then from its presence at $T < 56^\circ\text{K}$ it follows that the umbrella-like structure appears already near 50°K .

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¹G. S. Krinchik and M. V. Chetkin, Zh. Eksp. Teor. Fiz. 38, 1643 (1960); 40, 729 (1961); 41, 673 (1961)

[Sov. Phys.-JETP 11, 1184 (1960); 13, 509 (1961); 14, 485 (1961)].

²V. M. Chetkin, Fiz. Tverd. Tela 6, 3753 (1964) [Sov. Phys.-Solid State 6, 3013 (1965)].

³B. Johnson and R. Tebble, Proc. Phys. Soc. 87, 935 (1966).

⁴A. M. Clogston, J. Appl. Phys. 31, 1985 (1960).

⁵J. H. Van Vleck and W. G. Penney, Phil. Mag. 17, 115, 961 (1934).

⁶J. F. Dillon, J. Appl. Phys. 29, 1286 (1958); J. Phys. Rad. 20, 374 (1959).

⁷G. S. Krinchik and S. A. Gushchina, ZhETF Pis. Red. 4, 244 (1966) [JETP Lett. 4, 164 (1966)].

⁸N. F. Kharchenko, V. V. Eremenko, and L. I. Belyĭ, Zh. Eksp. Teor. Fiz. 53, 1505 (1967) [Sov. Phys.-JETP 26, 869 (1968)].

⁹S. V. Tyablikov, Fiz. Met. Metallov. 7, 3 (1956).

¹⁰A. A. Gusev, Kristallografiya 4, 595 (1959) [Sov. Phys.-Crystallogr. 4, 655 (1959)].

¹¹A. A. Gusev and A. S. Pakhomov, Izv. AN SSSR, ser. fiz. 25, 1327 (1961).

¹²A. S. Pakhomov and A. A. Gusev, Fiz. Metal. Metallov. 18, 156 (1964).

¹³N. F. Kharchenko and V. V. Eremenko, Zh. Prikl. Spekt. 5, 8 (1966); Fiz. Tverd. Tela 9, 1655 (1967) [Sov. Phys.-Solid State 9, 1302 (1967)].

¹⁴V. E. Rode, A. V. Vedyayev, and B. N. Kraĭnov, in: Fizicheskie i fiziko-khimicheskie svoistva ferritov (Physical and Physicochemical Properties of Ferrites), Minsk, 1966.

¹⁵A. A. Gusev and A. S. Pakhomov, Kristallografiya 8, 63 (1963) [Sov. Phys.-Crystallogr. 8, 45 (1963)].

¹⁶W. P. Wolf and J. H. Van Vleck, Phys. Rev. 118, 1490 (1960).

¹⁷E. E. Anderson, Phys. Rev. 134, 1581 (1964).

¹⁸K. P. Belov and V. I. Sokolov, Izv. AN SSSR ser. fiz. 30, 1079 (1966).

¹⁹K. P. Belov and S. A. Nikitin, Phys. St. Solidi 12, 1 (1965).

²⁰Y. Allain, M. Bichara, and A. Herpin, J. Appl. Phys. 37, 1316 (1966).

²¹A. Herpin, W. C. Kochler, and P. Meried, Compt. Rend. 251, 1359 (1960).

²²N. S. Akulov, Ferromagnetizm (Ferromagnetism), ONTI, 1939.

²³V. I. Sokolov and Than Dyc Hien, Zh. Eksp. Teor. Fiz. 52, 1485 (1967) [Sov. Phys.-JETP 25, 986 (1967)].

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