

## FARADAY EFFECT IN THE IRON GARNET $Y_3Fe_5O_{12}:Ho^{3+}$

N. F. KHARCHENKO, V. V. EREMENKO, and L. I. BELYĬ

Low-Temperature Physico-technical Institute, Academy of Sciences, Ukrainian S.S.R.

Submitted May 12, 1967

Zh. Eksp. Teor. Fiz. 53, 1505-1509 (November, 1967)

The Faraday effect was used to observe the turning over of the magnetic sublattice of holmium ions introduced into yttrium iron garnet. The value of the exchange field acting on a holmium ion was obtained from the experimental data; it is  $(2.6 \pm 0.2) \times 10^5$  Oe. A strong dependence of the Faraday rotation on the intensity of the external magnetic field, in pure yttrium garnet, was also observed.

It has been shown by Tyablikov<sup>[1]</sup> and by Gusev<sup>[2]</sup> that in ferrites with two sublattices, on attainment of a certain critical value of the magnetic field, there can be observed a process of turning of the sublattices from an antiparallel to a parallel arrangement. In this process, the magnetization of the ferrite increases with increase of the field intensity according to a linear law and is independent of temperature. Such a process was observed experimentally in gadolinium iron garnet.<sup>[3]</sup>

In the case of three-sublattice iron garnets,<sup>[4]</sup> there should first occur a turning over of the rare-earth sublattice relatively to the collinear sublattices of the iron ions; thereafter, at considerably larger fields, the antiparallel orientation of the magnetic moments of the iron ions is broken up. The values of the critical fields for the beginning and the ending of the turning-over process of a rare-earth sublattice coincide with the values of the critical fields for a two-sublattice ferrite, if the two nonequivalent sublattices of iron ions in the garnet are replaced by a single resultant sublattice. These values are

$$H_1 = \frac{M_2 - M_1}{\sqrt{M_2 M_1}} H_e, \quad H_2 = \frac{M_2 + M_1}{\sqrt{M_2 M_1}} H_e, \quad (1)$$

where  $M_1$  and  $M_2$  are the magnetizations of the sublattice of rare-earth ions and of the resultant sublattice of iron ions, respectively,  $H_e = (N_1 J_{12} N_2 J_{21} / M_1 M_2)^{1/2} S_1 S_2$ ,  $N_1$  and  $N_2$  are the numbers of ions in the sublattices,  $S_1$  and  $S_2$  are the spin moments of the ions, and  $J_{12}$  and  $J_{21}$  are exchange integrals. It is convenient to express the formulas in terms of the exchange field  $H_{\text{exch}}$  that acts on the spin moment of a rare-earth ion and that coincides with  $H_{\text{eff}} = J_{12} N_1 S_1 S_2 / M_1$  when the magnetic moment of the ion  $M_1$  is purely spin. If, however, there

is a nonvanishing orbital moment, then, according to Wolf and Van Vleck,<sup>[5]</sup>  $H_{\text{exch}} = g_J H_{\text{eff}} / 2(g_J - 1)$  and

$$H_{1,2} = \left(1 \mp \frac{M_1}{M_2}\right) \frac{2(g_J - 1)}{g_J} H_{\text{exch}}, \quad (2)$$

and the magnetic susceptibility during the turning-over of the rare-earth sublattice is

$$\chi = g_J M_2 / 2(g_J - 1) H_{\text{exch}}, \quad (3)$$

where  $g_J$  is the g-factor of the ground state of the ion.

For substituted iron garnets of the type  $Y_{3-x}Me_xFe_5O_{12}$ , in which a majority of the rare-earth ions have been replaced by nonmagnetic yttrium ions, the values of  $H_1$  and  $H_2$  are close to each other:

$$H_{1,2} = \left(1 \mp \frac{cM_1}{M_2}\right) \frac{2(g_J - 1)}{g_J} H_{\text{exch}}, \quad (4)$$

$cM_1$  is the magnetization of the rare-earth sublattice. The magnetic susceptibility due to the turning process is independent of the concentration  $c$  of the rare-earth ions.

We have investigated the Faraday effect in a crystal of yttrium iron garnet with a small addition (a few percent) of holmium ions,  $Ho^{3+}$ , for which the values of the critical fields, estimated by formula (4), do not exceed the range of fields attainable by the pulse method.<sup>[6]</sup> As is well known,<sup>[7]</sup> the Faraday effect and the magnetization are proportional to each other in compounds of rare-earth ions, if the change of the transition frequency  $\Delta\omega$  between the electronic configurations  $(4f)^n$  and  $(4f)^{n-1}5d$ , under the influence of the crystalline and magnetic fields, is negligible in comparison with the difference between the transition frequency and the frequency of the incident radiation:

$$\Delta\omega \ll \omega_0 - \omega. \quad (5)$$

It is here assumed that only the lowest multiplet is populated; nondiagonal elements of the dipole-moment matrix of the transition are neglected, and the diamagnetic Faraday effect (which can always be neglected in the low-temperature region) is not taken into account. For almost all trivalent rare-earth ions, these requirements are fulfilled, and the proportionality between  $\varphi$  and  $M$  is confirmed experimentally.<sup>[8]</sup>

Since the exchange interaction can be replaced by the action of some effective internal field on the total magnetic moment  $M_J$  of an ion, the proportionality when condition (5) is fulfilled should still hold for magnetically ordered crystals. Then, thanks to a sufficiently large specific rotation of the trivalent holmium ion, the behavior of the magnetization of the holmium sublattice in  $Y_3Fe_5O_{12}:Ho^{3+}$  can be studied through the change in the Faraday rotation. The effect of a deviation of the resultant sublattice of the iron ions from the direction of the external field can be neglected in the case under investigation, and it can be considered that the change of rotation is caused entirely by the  $Ho^{3+}$  ions.

The measurements of the Faraday effect were made with the apparatus described earlier,<sup>[6]</sup> in pulsed magnetic fields of intensity up to 170 kOe; the time of rise of the field to its maximum value was  $\sim 3.5$  msec. A specimen of thickness about 80 microns was cut in the form of a plate, oriented in the (110) plane. The specimen temperature was varied from 20 to 150°K. The light source was a helium-neon laser, with radiation wavelength 6328 Å, sufficiently distant from the nearest group of absorption lines of the  $Ho^{3+}$  ions in yttrium iron garnet (6385 Å); these make a contribution to the Faraday effect only at a distance of the order of the line-width and should not show up at  $\lambda = 6328$  Å.<sup>1)</sup>

Figure 1 shows the dependence of the Faraday effect in  $Y_3Fe_5O_{12}:Ho^{3+}$  on the intensity of the magnetic field. The rotation decreases with increase of magnetic field,

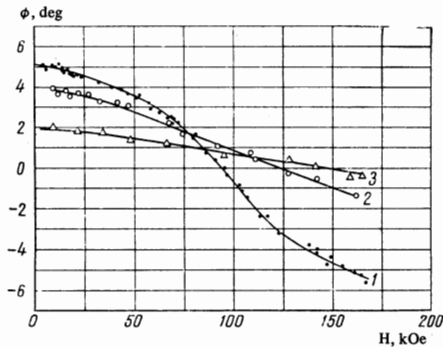


FIG. 1. Dependence of the Faraday effect on intensity of the external magnetic field in garnets: 1,  $Y_3Fe_5O_{12}:Ho^{3+}$ ,  $d = 80$  microns,  $T = 20^\circ K$ ; 2,  $Y_3Fe_5O_{12}:Ho^{3+}$ ,  $d = 80$  microns,  $T = 134^\circ K$ ; 3,  $Y_3Fe_5O_{12}$ ,  $d = 50$  microns,  $T = 20^\circ K$ .

<sup>1)</sup> Measurements of the dispersion of the Faraday effect near the absorption lines, in a constant field of 3 kOe at  $T = 20^\circ K$ , and also the Zeeman effect, which was investigated at magnetic field intensities up to 120 kOe, are in agreement with this assertion. The authors take this occasion to express their thanks to V. P. Novikov for making the Zeeman-effect measurements.

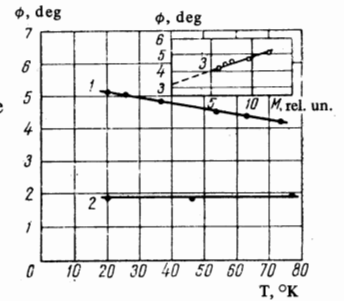


FIG. 2. Temperature dependence of the Faraday effect in iron garnets in field  $H \rightarrow 0$ : curves 1 and 3,  $Y_3Fe_5O_{12}:Ho^{3+}$ ,  $d = 80$  microns; 2,  $Y_3Fe_5O_{12}$ ,  $d = 50$  microns.

and especially large changes are observed near 100 kOe at 20°K. At higher temperatures, the rotation decreases more slowly and almost linearly over the whole field range. The decrease of the Faraday rotation is easily explained if we take into account that the external and exchange fields for the holmium ions in the garnet  $Y_3Fe_5O_{12}:Ho^{3+}$  are directed oppositely to each other. However, such a strong effect of the magnetic field on the Faraday effect in a ferrite containing a small admixture of  $Ho^{3+}$  ions is not consistent with the temperature dependence of the effect in zero magnetic field (the amount of rotation of the plane of polarization in zero field is determined by extrapolation of the  $\varphi(H)$  curves to the value  $H = 0$ ). With lowering of the temperature from 80 to 20°K, the rotation in  $Y_3Fe_5O_{12}:Ho^{3+}$  increases by 1° (Fig. 2, curve 1), and the magnetization increases by an amount equal to about half the maximum value.<sup>[9]</sup> At the same time an external field of intensity up to 150 kOe at 20°K produces a change in the rotation of the plane of polarization in the same specimen of almost 10° (Fig. 1, curve 1).

To clarify the nature of this peculiarity, the dependence of the Faraday effect on specimen temperature and on external magnetic field intensity was studied in the pure garnet  $Y_3Fe_5O_{12}$ . As was expected, the Faraday rotation in yttrium garnet remains practically unchanged on increase of the temperature from 20 to 77°K (Fig. 2, curve 2), but it depends strongly on external magnetic field intensity and can even change sign (Fig. 1, curve 3). It diminishes with increase of magnetic field intensity almost linearly, beginning at  $H = 40$  kOe, for the whole temperature range investigated (20 to 70°K).

Since the  $Fe^{3+}$  ions are in an exchange field of intensity more than  $10^6$  Oe, it would seem that an external field  $H \sim 10^5$  Oe should not have such a strong influence on the Faraday effect. In Krinchik and Gushchina's study<sup>[10]</sup> of the equatorial Kerr effect in ferromagnetic d-metals, there was also observed an increase of the effect with increase of magnetic field intensity, considerably stronger than the increase of magnetization attributable to change of the spin-orbit interaction under the influence of the external magnetic field. It is possible that the large change of the Faraday effect in yttrium garnet is also of the same origin.<sup>2)</sup>

To determine the share of the holmium sublattice in the Faraday rotation in  $Y_3Fe_5O_{12}:Ho^{3+}$ , it is necessary to subtract the rotation due to the iron ions. If we as-

<sup>2)</sup> It was G. S. Krinchik who suggested the possibility of such an explanation of the observed peculiarity, and the authors take this occasion to express their thanks to him.

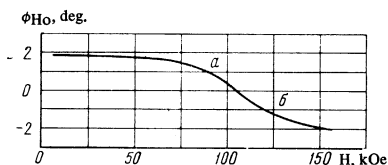


FIG. 3 Change of the Faraday rotation of the holmium ions in  $Y_3Fe_5O_{12}:Ho^{3+}$  with increase of magnetic field intensity at  $T = 20^\circ K$ .

sume that the rotation of the plane of polarization caused by the iron ions diminishes with increase of the external magnetic field intensity in  $Y_3Fe_5O_{12}:Ho^{3+}$  in the same way as in pure yttrium garnet, and that it is completely responsible for the change of the Faraday effect in  $Y_3Fe_5O_{12}:Ho^{3+}$  in a field  $H < 50$  kOe (that is, far from  $H_1$ ), then it is possible to isolate the rotation connected with the sublattice of holmium ions. The value, necessary for this, of the Faraday effect caused by the sublattice of iron ions in zero external field was determined from the temperature dependence of the rotation in  $Y_3Fe_5O_{12}:Ho^{3+}$  (Fig. 2), which can be described by the expression  $\varphi = \varphi_0 + AM_{Ho}$ , where  $\varphi_0$  is the rotation of the plane of polarization by the iron atoms,  $M_{Ho}$  is the magnetization of the holmium sublattice, and  $A$  is a coefficient of proportionality. The value of  $\varphi_0$  is easy to determine by plotting  $\varphi(M_{Ho})$  (Fig. 2, curve 3) with the aid of data on the magnetization of holmium iron garnet.<sup>[9]</sup>

The  $\varphi_{Ho}(H)$  curve obtained by this method (Fig. 3) describes, apart from a factor, the change of the projection of the magnetic moment of the sublattice of holmium ions on the direction of the external magnetic field. The section ab of the curve can be explained as a process of turning over of the sublattice, beginning at  $H \sim 85$  kOe and finishing at  $H \sim 130$  kOe. The small increase of rotation with field at  $H > 140$  kOe is apparently connected with an increase of the magnetization  $M_{Ho}$ . By using the value of magnetic field intensity  $H' = 105$  kOe at which the Faraday effect (Fig. 3) vanishes, and by supposing that  $H' = (H_1 + H_2)/2$ , it is possible to estimate the exchange field acting on the spin magnetic moment of a holmium ion. On taking the value of the g-factor for the free ion, 1.25, we get  $H_{exch} = (2.6 \pm 0.2) \times 10^5$  Oe. Such a value is close to the literature value  $H_{exch} = 2.4 \times 10^5$  Oe for holmium iron garnet.<sup>[9, 11]</sup>

The exchange field can also be estimated independently from the magnetic susceptibility during turning over of the holmium sublattice. For this purpose it is necessary to know the coefficient of proportionality between the rotation and the magnetization. Since the concentration of  $Ho^{3+}$  ions in the specimen was not known exactly, in order to determine the coefficient of proportionality the Faraday effect was measured in  $Ho_3Fe_5O_{12}$  in the temperature interval  $20$  to  $70^\circ K$ , and its dependence on the magnetization of the holmium

sublattice in the garnet (analogous to curve 3 of Fig. 2) was plotted. The coefficient of proportionality  $k$  between the Faraday effect and the magnetization of the sublattice of holmium ions, at specimen thickness  $80$  microns, was  $1.4 \pm 0.2$  deg/ $\mu_B$  (the magnetization  $M_{Ho}$  on one molecule of  $Ho_3Fe_5O_{12}$  was taken in the calculation), and the susceptibility  $\chi \approx \partial M_{Ho} / \partial H = \frac{1}{k} \frac{\partial \varphi_{Ho}}{\partial H}$  in the vicinity of the inflection point of the curve (Fig. 3) was  $0.06 \pm 0.015$   $\mu_B/kOe$ . By use of this value, we get with the aid of formula (3)  $H_{exch} = (2.00 \pm 0.50) \times 10^5$  Oe. In view of the fact that this estimate is rather rough, the agreement with the preceding value of  $H_{exch}$ , obtained by an independent method, must be considered good.

We take this occasion to thank Prof. B. I. Verkin and A. A. Shvarts for interest in the research and for support, Yu. A. Popkov for useful discussions, and A. V. Antonov for providing the specimens.

<sup>1</sup> S. V. Tyablikov, *Fiz. Metallov i Metallovedenie* **3**, 3 (1956).

<sup>2</sup> A. A. Gusev, *Kristallografiya* **4**, 695 (1959) [*Sov. Phys.-Crystallogr.* **4**, 655 (1960)].

<sup>3</sup> V. E. Rode and A. V. Vedyayev, *Zh. Eksp. Teor. Fiz.* **45**, 415 (1963) [*Sov. Phys.-JETP* **18**, 286 (1964)].

<sup>4</sup> A. A. Gusev and A. S. Pakhomov, *Izv. Akad. Nauk SSSR, ser. Fiz.* **25**, 1327 (1961) [*Bull. Acad. Sci. USSR* **25**, 1338 (1961)]; A. S. Pakhomov and A. A. Gusev, *Fiz. Metallov i Metallovedenie* **18**, 156 (1964) [*Phys. Met. Metallog.* **18**, No. 1, 149 (1964)].

<sup>5</sup> W. P. Wolf and J. H. Van Vleck, *Phys. Rev.* **118**, 1490 (1960).

<sup>6</sup> N. F. Kharchenko and V. V. Eremenko, *Zh. Priklad. Spektrosk.* **5**, 8 (1966).

<sup>7</sup> J. H. Van Vleck and M. H. Hebb, *Phys. Rev.* **46**, 17 (1934); Y. R. Shen, *Phys. Rev.* **133**, A511 (1964).

<sup>8</sup> *Low Temperature Phys. Handb. Physik N. 14-15*, 1956.

<sup>9</sup> R. Pauthenet, *Ann. Phys. (Paris)* **3**, 424 (1958); Y. Allain, M. Bichara, and A. Herpin, *J. Appl. Phys.* **37**, 1316 (1966).

<sup>10</sup> G. S. Krinchik and S. A. Gushchina, *ZhETF Pis. Red.* **4**, 244 (1966) [*JETP Lett.* **4**, 164 (1966)].

<sup>11</sup> K. P. Belov, M. A. Belyanchikova, R. Z. Levitan, and S. A. Nikitin, *Redkozemel'nye ferro- i antiferromagnetiki (Rare-Earth Ferro- and Antiferromagnets)*, Nauka, 1965, p. 229.