Investigation of inversion of the energy levels of the Tb^{3+} ion in an iron garnet in strong fields using the Faraday effect

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A jump of the Faraday effect in a terbium-yttrium iron garnet was observed in ~80-kOe fields at helium temperatures. This jump is accounted for by a model for inversion of the energy levels of the Tb^{3+} ion in a magnetic field.

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Demidov *et al.*[1] and one of the authors of this paper observed at helium temperatures in ~100-kOe fields magnetization jumps in terbium-yttrium iron garnet single crystals (TYIG) $Tb_xY_{3-x}Fe_5O_{12}$ with a small ($x \leqslant 0.65$) terbium content. The anomalies of magnetization of TYIG were explained[1] by a model of inversion of the energy levels of the rare-earth terbium ion Tb^{3+} in a magnetic field, which was proposed by Cooper[2] to explain the magnetic properties of the rare-earth RSb compounds. It is assumed in this model that the lower multiplet of the rare-earth ion (in this case Tb^{3+}) in the crystal field is split in such a way that there is a second level near the ground level. The application of the external field changes the energy of these levels and if the energy of the second level decreases with the field more sharply than that of the ground level, then the levels will cross in the H_c field: at $H > H_c$ the second level becomes the ground level. At sufficiently low temperatures when it is assumed that only the lower level is populated, the inversion of the levels must be accompanied by a jump of the magnetic moment.[2,3]

The inversion of the Tb^{3+} levels in the magnetic field was confirmed by data on the energy structure of this ion in garnets. For example, the Tb^{3+} spectrum of the paramagnetic terbium-gallium garnet has a second level situated 2 cm^{-1} from the ground level.[4] The anomalies of the ferromagnetic resonance of TYIG at low temperatures can be explained by assuming that the Tb^{3+} spectrum has energy levels located ~10 cm^{-1} from the ground level.[5–7] The inversion of levels in the ferromagnetic TYIG, however, is much more complicated than in the paramagnetic compounds examined by Cooper.[2] In ferromagnets the applied external field can produce a noncollinear magnetic structure in which the magnetic moments of the iron and rare-earth sublattices are deflected from the direction of the field, which also causes magnetization anomalies.[8] Therefore, the magnetization jump in TYIG cannot be fully accounted for on the basis of the magnetization data, since these data give no clue as to which sublattice (terbium or iron) is responsible for the observed anomalies.

The contribution of the different sublattices to the magnetization jumps of TYIG in strong fields can be determined by comparing the magnetization data with the results obtained from the measurements of the magneto-optical effects such as rotation of the polarization plane (Faraday effect). In fact, the magnetization is comprised of

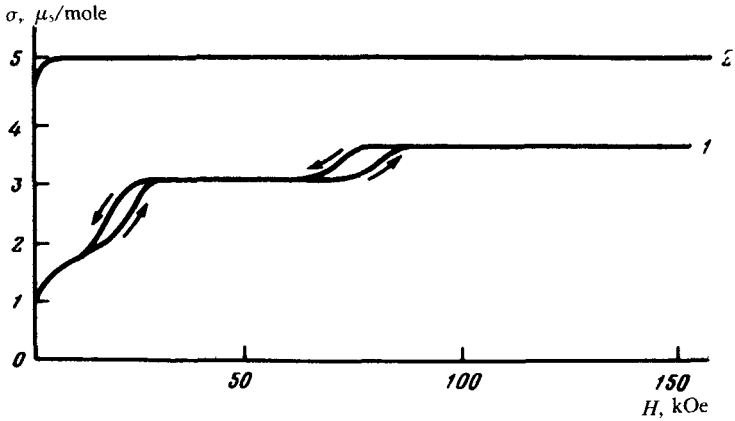

FIG. 1. Dependence of magnetization of the $Tb_{0.26}Y_{2.74}Fe_5O_{12}$ (curve 1) and $Y_3Fe_5O_{12}$ (curve 2) single crystals along the ⟨111⟩ axis.

the projection of the magnetic moments of the terbium $\mathbf{M}_{Tb}$ and iron $\mathbf{M}_{Fe}$ sublattices on the direction of the field

$$M = (\mathbf{M}_{Fe})_H + (\mathbf{M}_{Tb})_H \ . \tag{1}$$

On the other hand, the Faraday effect can be represented in the form[9]

$$\alpha_F = A(\mathbf{M}_{Fe})_H + B(\mathbf{M}_{Tb})_H \ . \tag{2}$$

If the coefficients A and B are known, then we can determine from the experimental data on magnetization and the Faraday effect the variation with the field of the projection of the magnetic moments of terbium $(\mathbf{M}_{Tb})_H$ and iron $(\mathbf{M}_{Fe})_H$ sublattices (these projections are denoted by M_{Tb} and M_{Fe}).

We report the results of an investigation of the anomalous magnetization of the TYIG single crystal $Tb_{0.26}Y_{2.74}Fe_5O_{12}$ along the ⟨111⟩ axis in strong magnetic fields by measuring the magnetization and the Faraday effect.

The magnetization and the Faraday effect (at the wavelength of $\lambda = 6328$ Å) where measured in pulsed magnetic fields at helium temperatures.

Figure 1 shows the field dependence of magnetization of the $Tb_{0.26}\ Y_{2.74}Fe_5O_{12}$ single crystal along the ⟨111⟩ axis. We can see that in weak fields (at $H \leqslant 30$ kOe) the magnetization increases, which is attributable to translation of the domain boundaries and rotation from the axis of slight magnetization ⟨100⟩ to the ⟨111⟩ axis. The hysteresis and spasmodic variation of magnetization in the rotation region indicate that the transition to the phase with $\mathbf{M} \| \langle 111 \rangle$ is a first-order phase transition. This type of transition to the field occurs if the second constant of the magnetic anisotropy is larger than the first.[10] Large fields produce saturation: magnetization in this case is independent of the field. However, the magnetization will increase spasmodically if the field increases to the critical value H_c. The variation of magnetization in this region is accompanied by a hysteresis ($H_c = 80$ kOe when the field increases and $H_c = 73$ kOe

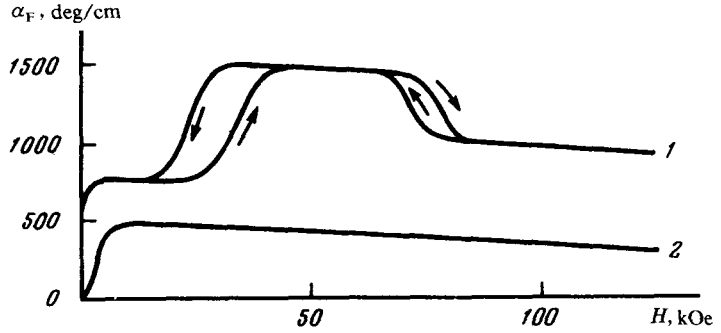

FIG. 2. Dependence of the Faraday effect for the $Tb_{0.26}Y_{2.74}Fe_5O_{12}$ (curve 1) and $Y_3Fe_5O_{12}$ (curve 2) single crystals along the ⟨111⟩ axis (the wavelength of light is 6328 Å).

when the field decreases), which indicates that this is a first-order phase transition. The magnetization remains almost constant with further increase of the field.

Figure 1 also shows the field dependence of magnetization of the yttrium iron garnet YIG $Y_3Fe_5O_{12}$. The magnetization of this ferromagnet, which becomes saturated in weak fields, is independent of the field.

The rotation of the polarization plane of TYIG $Tb_{0.26}Y_{2.74}Fe_5O_{12}$ is shown in Fig. 2 as a function of the field. It can be seen that in weak fields the Faraday effect varies sharply with the field, which is attributable to commercial magnetization.[1)] The Faraday effect has a discontinuity in the H_c field; moreover, if the magnetization increases in this field, then the rotation of the polarization plane will decrease.

Figure 2 also shows the field dependence of the Faraday effect of YIG. It can be seen that the rotation of the polarization plane decreases with increasing field in the region of the field in which the magnetization becomes saturated. This earlier observed effect,[12] which is attributable to the influence of the field on the spin-orbit interaction of the Fe^{3+} ions,[11] determines the field dependence of the Faraday effect of TYIG in that region of the fields in which the magnetization remains constant (see Figs. 1 and 2).

The coefficients A and B, which are included in Eq. (2), can be easily determined from the data on magnetization and the Faraday effect. Since the yttrium iron garnet has only iron ions, it follows from Figs. 1 and 2 that $A = 94$ [deg-μ_B^{-1}-cm^{-1}] $- 0.32\,H$ [deg-μ_B^{-1}-cm^{-1} kOe^{-1}]. To determine B, we compare the data on magnetization and Faraday effect in the saturation region (40 kOe$\leqslant H \leqslant$70 kOe). Here, the magnetic moments of the sublattices are collinear to the field and antiparallel to each other. From the data on magnetization of YIG $M_{Fe} = 5\mu_B$; hence, $M_{Tb} = -2\mu_B$, since the magnetization of TYIG in this field interval is $M = 3\mu_B$. From the rotation in this field interval, using the value of A obtained above, we find $B = 540$ grad-μ_B^{-1}-cm^{-1}.

Thus, knowing A and B, we can determine from Eqs. (1) and (2) the magnetic moments of the terbium and iron sublattices (specifically, their projections in the direction of the field) at $H \geqslant H_c$. From our data we obtain $M_{Tb} = (1.2 \pm 0.2)\mu_B$ and $M_{Fe} = (4.8 \pm 0.3)\mu_B$.[2)]

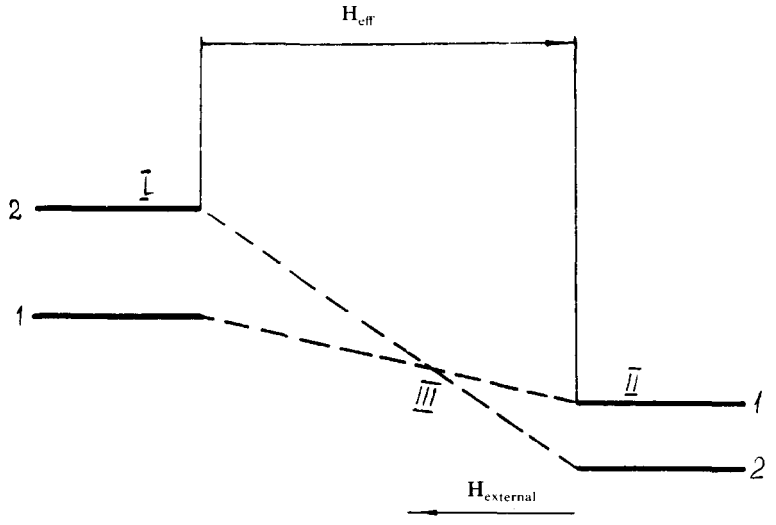

FIG. 3. The arrangement schematic of the lower energy levels of the Tb^{3+} ions in garnets: I, Splitting in the crystal field; II, splitting in the crystal and effective field; III, field of the crossing of levels.

Thus, we can see from our measurements that at $H = H_c$ the magnetic moment of the iron sublattice remains almost constant and the increase of magnetization of TYIG in this field is attributable to a large decrease of the magnetic moment of the terbium sublattice, which is oriented antiparallel to the field. This situation must exist if an inversion of the levels is to occur in this field.

In analyzing this effect in TYIG, we must take into account that, in addition to the external field, the effective exchange field from the iron sublattice (200 kOe) affects the Tb^{3+} ion in these ferrimagnets; moreover, for TYIG with a small terbium content, when $M_{Tb} < M_{Fe}$, the external field is antiparallel to the effective field. If there are two similar singlets in the crystal field (Fig. 3, state I) and level 2 has a larger g factor than level 1, then the effective field changes the position of the levels due to the Zeeman effect and leads to a state denoted by Roman numeral II in Fig. 3. In this case the magnetic moment of the Tb^{3+} ion increases. An applied external field, which is antiparallel to the effective field, causes the levels to cross and reverses the transition to the state I with a smaller magnetic moment of the Tb^{3+} ion.

Note that a slight decrease of the projection of the magnetic moment of the iron sublattice in the direction of the field can also be explained by this model, since the inversion of the levels must be accompanied by a distortion of the magnetic structure, which eliminates degeneration of the levels at $H = H_c$ (Jahn-Teller magnetic effect[13]).

[1]A slight difference between the $\alpha_F(H)$ curve and the $\sigma(H)$ curve is attributed to different demagnetization factors of the samples [magnetization was measured in a long rod and the Faraday effect was measured in a thin (50 μm) plate] and apparently to the large effect of mechanical orientations primarily of the surface, on magnetization of the plate.

[2]In a field parallel to the $\langle 111 \rangle$ direction, the garnet structure has two nonequivalent locations of rare-earth ions.[6] The inversion of the levels of the Tb^{3+} ions in the H_c field most likely occurs only in one of these

locations. If we assume that in weak fields the total magnetic moments in the nonequivalent locations are identical ($M_{Tb1} = M_{Tb2} = M_{Tb}/2 = 1\mu_B$), then it follows from our experimental data that at H_c the total magnetic moment in one of the locations decreases to $0.2 \pm 0.2\ \mu_B$, i.e., it equals to almost zero.

[1]V.G. Demidov, R.Z. Levitin, and Yu. F. Popov, Fiz. Tverd. Tela **18**, 596 (1976) [Sov. Phys. Solid State **18**, 347 (1976)].
[2]B. Cooper, Phys.Lett. **22**, 244 (1966).
[3]F. Varret, J. Phys. Chem. Sol. **37**, 257 (1976).
[4]D. Boal, P. Grunberg, and J. Koningstein, Phys. Rev. **B7**, 4757 (1973).
[5]J. Dillon and L. Walker, Phys. Rev. **124**, 1401 (1961).
[6]L. Walker, J. Appl. Phys. **33**, 1243 (1963).
[7]A.G. Gurevich, Magnitnyiĭ rezonans v ferritakh i antiferromagnetikakh (Magnetic Resonance in Ferrites and Anteferromagnets), Nauka, M., 1973.
[8]K.P. Belov, Ferrity v sil'nykh polyakh (Ferrites in Strong Fields), Nauka, M., 1975.
[9]J.R. Shen, Phys. Rev. **133**, 2A, 511 (1964).
[10]V.G. Bar'yakhtar, V.A. Borodin, V.D. Doroshev, N.M. Kovtun, R.Z. Levitin, and E.P. Stevanovskiĭ, Zh. Eksp. Teor. Fiz. **74**, 600 (1978) [Sov. Phys. JETP **47**, 315 (1978)].
[11]G.S. Krinchik and S.A. Gushchina, Zh. Eksp. Teor. Fiz. **55**, 490 (1968) [Sov. Phys. JETP **28**, 257 (1968)].
[12]N.F. Kharchenko, V.V. Eremenko, and L.I. Belyiĭ, Zh. Eksp. Teor. Fiz. **55**, 419 (1968) [Sov. Phys. JETP **28**, 219 (1968)].
[13]A.K. Zvezdin, A.A. Mukhin, and A.I. Popov, Pis'ma Zh. Eksp. Teor. Fiz. **23**, 240 (1976) [JETP Letters **23**, 267 (1976)].