Magnetooptic properties of rare-earth orthoferrites in the region of spin reorientation transitions

E. A. Balykina, E. A. Gan'shina, and G. S. Krinchik

M. V. Lomonosov State University, Moscow

(Submitted 25 March 1987)
Zh. Eksp. Teor. Fiz. **93**, 1879–1887 (November 1987)

An investigation was made of the magnetooptic reflection spectra of $TmFeO_3$, $HoFeO_3$, and $ErFeO_3$ in the photon energy range 1.5–4.5 eV under conditions close to the region of the $G_xF_z \rightarrow G_zF_x$ spin reorientation. A strong anisotropy of resonances at 3.0, 3.9, and 4.2 eV confirmed the mechanism of anisotropic freezing of the orbital momentum, suggested earlier in order to explain the anomalously strong magnetooptic effects in orthoferrites. In the case of $ErFeO_3$ on a (010) face and of $TbFeO_3$ on a (110) face it was found that the temperature of the spin-reorientation transition T_2 was shifted strongly in the upward direction. The existence of surface magnetism in $ErFeO_3$ at temperatures in the range 100–140 K was deduced from the results. Reorientation of the magnetic spin system in $ErFeO_3$ at $T_c = 47$ K was observed for $ErFeO_3$.

The room-temperature magnetization of the rare-earth ions in rare-earth orthoferrites ($R\,FeO_3$) is very weak and practically all the features of the magnetic properties of these ferrites are governed by the subsystem of ordered Fe^{3+} ions. In particular, the magnetooptic spectra of the family of rare-earth orthoferrites obtained in Ref. 1 using the polar Kerr effect are basically similar for different compounds. Cooling enhances the role of the rare-earth sublattice and this gives rise to a number of special features of magnetic properties such as magnetic ordering of the rare-earth ions, spin-reorientation transitions, compensation points, etc. However, very few magnetooptic investigations of rare-earth orthoferrites have been made at low temperatures. These have been mainly measurements of the Faraday effect in several ferrites in the transparency range.[2–4] Among the reliably established experimental observations is the anisotropy of the Faraday effect: after the $G_xF_z \rightarrow G_zF_x$ spin-reorientation the Faraday effect decreases severalfold. This has been deduced first from the relative smallness of the Faraday effect in $SmFeO_3$ at room temperature[5] and then confirmed by investigations of the temperature dependences of the properties of $TmFeO_3$ (Ref. 2), and of $SmFeO_3$ and $YCo_{0.006}Ti_{0.006}Fe_{0.988}O_3$ (Ref. 6). The Faraday effect in $DyFeO_3$ has been studied at low temperatures.[4] The observed linear reduction in the effect on increase in the magnetic field has been attributed to an increase in the paramagnetic contribution by the rare-earth sublattice to the Faraday effect.

We investigated the magnetooptic reflection effects in erbium, holmium, thulium, ytterbium, terbium, and yttrium orthoferrites at temperatures in the range 10–300 K, where some of the orthoferrites exhibit spin-reorientation transitions of the $G_xF_z \rightarrow G_zF_x$ type as well as points of compensation of the magnetic moments of the iron and rare-earth sublattices. The magnetooptic reflection effects have the advantage that they allow us to carry out investigations at shorter wavelengths, where strong optical transitions in the Fe^{3+} ions are located and these determine the magnetooptic properties of orthoferrites in the visible and infrared parts of the spectrum; moreover, the natural linear birefringence of these crystals makes practically no contribution to these properties. Finally, the small depth of penetration of light into these crystals makes it possible to investigate independently the properties of a thin surface layer.

SAMPLES AND EXPERIMENTAL METHOD

Orthoferrite single crystals were grown by the floating zone method using radiative heating.[7] The orientation of the crystallographic axes was set using an URS-100 x-ray diffractometer. Plane-parallel plates of erbium, holmium, and thulium orthoferrites were cut at right-angles to the [010] direction and the error in the orientation of the axes did not exceed 2°. After mechanical polishing our samples were annealed in air at 1200 °C for 3 h. Moreover, measurements were made also on natural specularly reflecting faces of single crystals of $TbFeO_3$, $YbFeO_3$, and $YFeO_3$ orthoferrites grown from a molten solution. A sample was placed in a helium continuous-flow cryostat with a special insert where temperatures in the range 10–300 K could be established. Under the influence of an excess pressure created in a helium Dewar, liquid helium rose in the insert, evaporated, and cooled the sample which was thus located directly in a helium stream. The temperature of a sample was varied by controlling the rate of flow of gaseous helium. This temperature was measured with a Cu-CuFe thermocouple and the error in the determination of the temperature of a sample was 1 K.

Orthoferrite plates were bonded by BF-2 adhesive to nonmagnetic steel substrates and were placed in a special holder which made it possible to rotate a sample around a direction perpendicular to the plane of the sample and thus orient exactly the crystallographic axes relative to the direction of the applied magnetic field.

The magneooptic effects were measured using automatic apparatus based on a DMR-4 monochromator in which the data were acquired and processed employing a D3-28 microcomputer. A dynamic method of recording the magnetooptic effects[8] was used. The alternating component of the signal was due to magnetization reversal by a 30-Hz alternating magnetic field. The maximum amplitude of the alternating magnetic field in the gap of an electromagnet was 4 kOe. The sensitivity of the apparatus was 10^{-5}. Magnetooptic spectra were recorded in the photon energy range 1.5–4.5 eV at a fixed angle of incidence of light in the range 60–70°.

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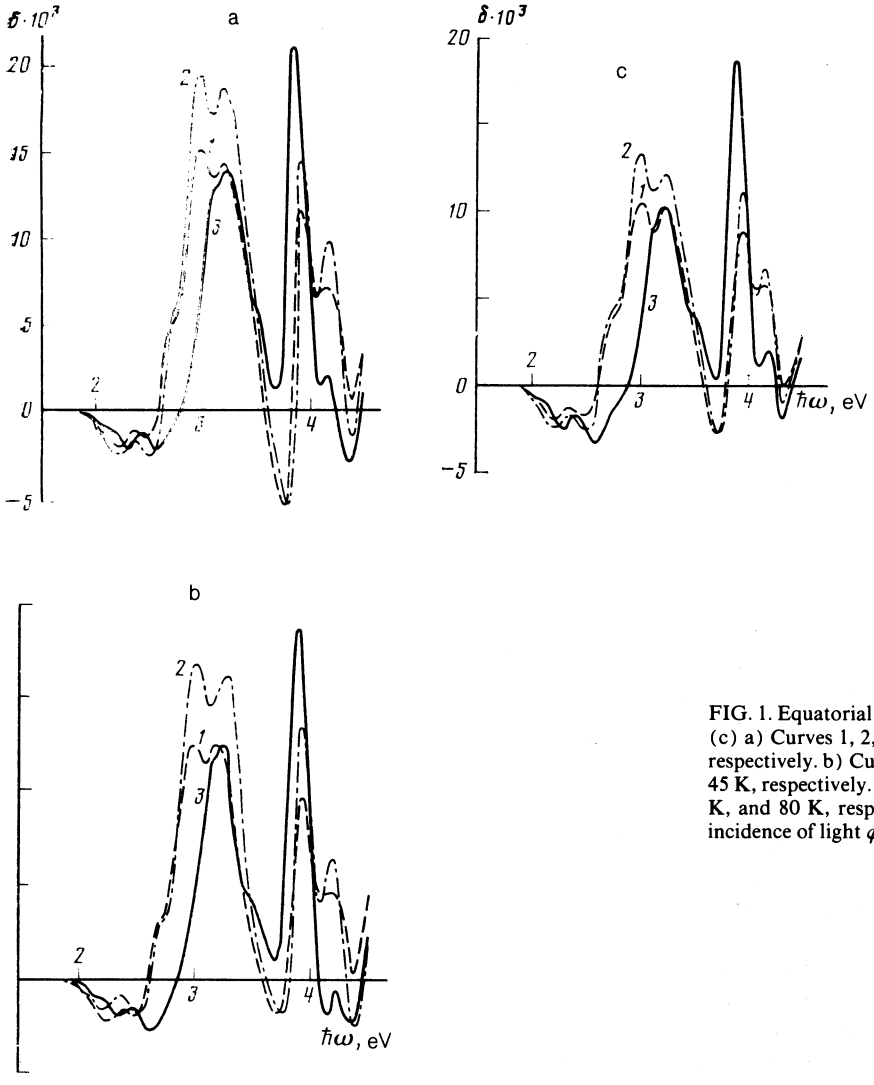

FIG. 1. Equatorial Kerr effect in $TmFeO_3$ (a), $HoFeO_3$ (b), and $ErFeO_3$ (c) a) Curves 1, 2, and 3 were obtained at $T = 295$ K, 100 K, and 80 K, respectively. b) Curves 1, 2, and 3 were obtained at $T = 295$ K, 70 K, and 45 K, respectively. c) Curves 1, 2, and 3 were obtained at $T = 295$ K, 145 K, and 80 K, respectively. 1), 2) $H \| c$; 3) $H \| a$; (010) plane; angle of incidence of light $\varphi = 65°$.

The geometry of the equatorial Kerr effect was used to record the relative change in the intensity of the reflected light $\delta = [I(H) - I(0)]/I(0)$ due to the magnetization of a sample [$I(H)$ and $I(0)$ are the intensities of the reflected light in the presence of a magnetic field and in the absence of this field, respectively]. The magnetization **m** was parallel to the surface of the sample and perpendicular to the plane of incidence of light (**m**⊥**k**, where **k** is the wave vector of light).

ANISOTROPY OF MAGNETOOPTIC SPECTRA OF Fe^{3+} IONS

Figures 1a, 1b, and 1c show the spectra of the equatorial Kerr effect obtained for $TmFeO_3$, $HoFeO_3$, and $ErFeO_3$. Curves 1 and 2 were determined at temperatures above the region of the spin-reorientation transition in a magnetic field directed along the c axis. After passing through the spin-reorientation temperature the sample was rotated so that the magnetic field was oriented along the a axis (curves denoted by 3). An analysis of the curves demonstrated that the orthoferrite spectra were strongly anisotropic and that the changes observed in the spectral dependences of the equatorial Kerr effect on transition to the G_zF_x state were common to $TmFeO_3$, $HoFeO_3$, and $ErFeO_3$. After reorientation of the magnetic moment to the a axis the spectrum of the equatorial Kerr effect became simpler: the long-wavelength component effectively disappeared from a doublet at 3.0–3.25 eV, the intensity of a spectral maximum at 4.2 eV decreased strongly, whereas a peak at 3.9 eV was greatly enhanced. In the range 3.4–3.5 eV a shoulder appeared in the frequency dependence of the effect. Moreover, the intensities of weak transitions at 2.35 and 2.7 eV changed slightly as a result of the reorientation. The positions of the maxima on the $\delta(\hbar\omega)$ curve agreed well with the identification of optical transitions in orthoferrites adopted in Ref. 1: two transitions occurred in a crystal field (a) ${}^6A_{1g}({}^6S) \rightarrow {}^4E_g({}^4G)$, ${}^4A_{1g}({}^4G)$ at 2.35 eV and (b) ${}^6A_{1g}({}^6S) \rightarrow {}^4T_{2g}({}^4D)$ at 2.7 eV; there were also four allowed electric-dipole transitions of the charge-transfer type in the $Fe^{3+}O^{2-}$ complex: (A) $t_{2u}^n(\pi) \rightarrow t_{2g}^*$ at 3.0 eV (B) $t_{1u}(\pi) \rightarrow t_{2g}^*$ at 3.2 eV, (C) $t_{1u}(\sigma) \rightarrow t_{2g}^*$ at 3.9 eV, and (D) $t_{2u}^n(\pi) \rightarrow e_g^*$ at 4.2 eV.

A confirmation that the observed anisotropy of the magnetooptic spectra was due to the $G_xF_z \rightarrow G_zF_x$ rotation of the spin system was provided by the observation that in the case of $YbFeO_3$, where in the temperature range 10–300 K there was no spin-reorientation of this type, the nature of the dispersion dependence of the equatorial Kerr effect was not affected by cooling and only an increase of all the spectral maxima as well as a slight shift toward higher energies by 0.05 eV were observed (Fig. 2). Similar results were ob-

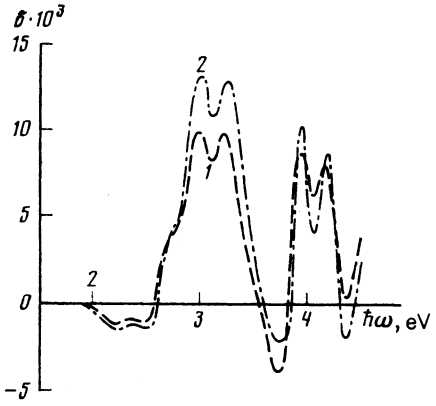

FIG. 2. Equatorial Kerr effect in $YbFeO_3$: 1) $T = 295$ K; 2) $T = 10$ K; $\varphi = 65°$; $H \| c$; (110) plane.

tained by us for $YFeO_3$ and $GdFeO_3$.

When we consider the nature of the magnetooptic anisotropy of rare-earth orthoferrites, we must remember that the extrema observed in the magnetooptic spectra were entirely due to the optical transitions in the Fe^{3+} ions. This was supported, firstly, by observation of spectra of the same type for different rare-earth orthoferrites and, secondly, by the field and temperature dependences of the equatorial Kerr effect outside the spin-reorientation region. Samples were saturated magnetically in fields of ~200 Oe and then the equatorial Kerr effect remained practically constant when H was increased up to 4 kOe. On the other hand, in the case of erbium orthoferrite the magnetization of the erbium subsystem should increase fourfold on increase in the field from 1 to 4 kOe and at T_c it should become comparable with the magnetization m_s of the iron sublattices. Therefore, if the rare-earth subsystem had made a significant contribution to the equatorial Kerr effect, we would have observed a strong $\delta(H)$ dependence in the region of 1–4 kOe. Consequently, we encountered an interesting situation: the $G_xF_z \rightarrow G_zF_x$ reorientation of the spin system occurred because of an increase in the magnetization of the rare-earth ions as a result of cooling, but the experiments revealed only the anisotropic optical properties of the subsystem of the Fe^{3+} ions of the kind which could be observed, for example, in $YFeO_3$ in a magnetic field of the order of 70 kOe, resulting in a spin-reorientation transition.

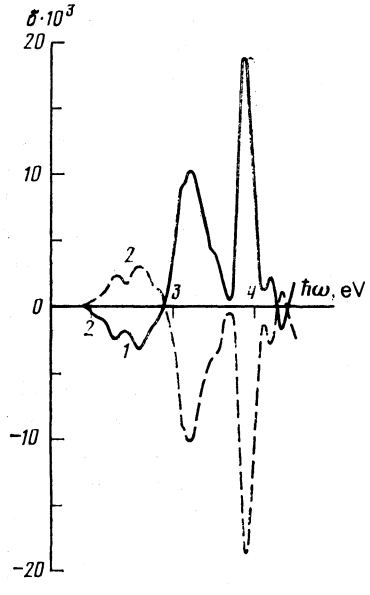

FIG. 3. Equatorial Kerr effect in $ErFeO_3$: 1) $T = 49$ K; 2) $T = 45$ K; $H \| a$; (010) plane.

The compensation point T_c, deduced as a temperature at which reversal of the sign of the effect was observed in $ErFeO_3$, amounted to 47 K, which was in agreement with the values of T_c deduced from magnetic measurements.[11] The spectra of the equatorial Kerr effect determined above and below the compensation point were symmetric (Fig. 3). Further cooling to 10 K did not alter the magnitude of the effect and the $\delta(H)$ dependences showed no changes right down to 10 K. Lack of changes in the spectra at temperatures above and below the compensation point T_c also indicated that spectra were determined by optical transitions in the Fe^{3+} ions.

The results made it possible to, firstly identify directly the source of the anisotropy of the Faraday effect in the region of the $G_xF_z \rightarrow G_zF_x$ spin-reorientation transition, which was observed in Refs. 3 and 6. In Ref. 6 this anisotropy was attributed to the influence of the transition (A) $t_{2u}^n(\pi) \rightarrow t_{2g}^*$ at 3 eV. Our results indicated that the transition (A) did indeed exhibit the required anisotropy, but we could see that a considerable role should also be played by contributions of anisotropic transitions (C) and (D) occurring at higher energies.

The existence of a strong anisotropy of many magnetooptic transitions in the Fe^{3+} ions in the region of the $G_xF_z \rightarrow G_zF_x$ spin reorientation could be understood quite readily in a qualitative sense on the basis of the symmetry approach. The symmetry of orthoferrites admitted the appearance of two independent antisymmetric components of the permittivity tensor[9,10]:

$$\varepsilon_{xy} = -\varepsilon_{yx} = i(\alpha_1 G_x + \beta_1 F_z) \text{ and } \varepsilon_{yz} = -\varepsilon_{zy} = i(\alpha_2 G_z + \beta_2 F_x),$$

the first of which governs the magnitude of the equatorial Kerr effect in the case when $k \perp c$ and the second applies to the case when $k \perp a$; α_1, β_1, α_2, and β_2 are independent coefficients. The independence of the anisotropic contributions of the components ε_{xy} and ε_{zy} is illustrated in Fig. 4. The absolute magnitude of the equatorial Kerr effect in the spin-reorientation region changed as a result of changes in the components G_i and F_i, but the nature of the magnetooptic anisotropy governed by the ratio of the intensities of the transition remained constant (at least in the first approximation).

The question why the values of ε_{xy} and ε_{yz} differ so much in the case of orthoferrites is related closely to the main problem of magnetooptics of weak ferromagnets, namely to the need to explain their anomalously strong magnetooptic properties. Weak ferromagnets $YFeO_3$ and α-Fe_2O_3 exhibit a spontaneous magnetization which is 10^2–10^3 times less than that of a ferrimagnet $Y_3Fe_5O_{12}$, whereas the magnetooptic effects in all these crystals are of approximately the same strength. Formally, this can be explained by the fact that in the expressions for ε_{xy} and ε_{yz} there are not only small components of the spins F_z and F_x, but also large components G_x and G_z which are comparable with, for example,

the magnetization of the ferrimagnet $Y_3Fe_5O_{12}$. In fact, experiments have confirmed this picture. It was shown in Ref. 11 that in the case of α-Fe_2O_3 when the expression for ε_{ij} is governed only by the component F_z of the induced magnetization (and the magnetic field is parallel to a preferred axis), the magnitude of the magnetooptic effect is at least two orders of magnitude less than in the case when ε_{ij} is governed by the linear combination of F_x and G_y (and the magnetic field lies in the basal plane).

The physical mechanism responsible for the "admixture" of the components of the antiferromagnetic vector G_i in the expression for ε_{ij} was suggested for orthoferrites in Ref. 1. It was shown there that because of the low symmetry of the local environment of the Fe^{3+} ions in the orthoferrite lattice, the quantization axes of the orbital angular momentum may not coincide with the directions of the components spin S_i of the magnetic sublattices. This mechanism, called in Ref. 1 the anisotropic freezing of the orbital momentum, in turn ensures, for example, that the expression for ε_{xy} is governed not only by the magnitude of the longitudinal components of the spin S_z and, consequently, by the components of the weak ferromagnetic vector F_z, but also by the transverse components of the spin S_x and, consequently, by the component G_x of the antiferromagnetic vector. Summing up, we can conclude that our results confirm the theory of magnetooptic properties of weak ferromagnets based on the magnetism of anisotropic freezing of the orbital momentum so that, for example, a strong reduction in the intensity of a peak at $\hbar\omega = 4.2$ eV after the $G_xF_z \to G_zF_x$ reorientation can be explained by the fact that the corresponding optical transition is characterized by $\alpha_2 \ll \alpha_1$. Naturally, the final determination of the nature of the magnetooptic properties of orthoferrites will require quantitative calculations of the magnetooptic anisotropy of the transitions A, B, C, and D, using, for example, Eq. (16) given in Ref. 12.

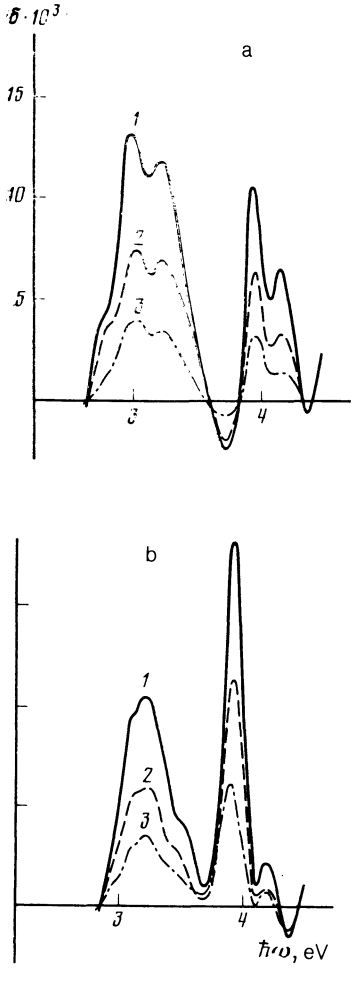

FIG. 4. Equatorial Kerr effect in $ErFeO_3$ in the region of the spin-reorientation transition. a) $H \| c$, $\mathbf{k} \perp \mathbf{c}$; curves 1, 2, and 3 are obtained at $T = 140$ K, 115 K, and 108 K, respectively; $H = 1$ kOe. b) $H \| a$, $\mathbf{k} \perp \mathbf{a}$; curve 1 was obtained at $T = 90$ K in a field $H = 1$ kOe, curve 2 was obtained at $T = 115$ K in $H = 2.5$ kOe, and curve 3 was obtained at $T = 135$ K in $H = 4$ kOe.

SURFACE MAGNETISM IN ERBIUM ORTHOFERRITE

Figure 5 shows the temperature dependence of the equatorial Kerr effect in the region of the 3.25 eV spectral maximum, obtained for two orientations of a magnetic field in thulium, holmium, and erbium orthoferrites. It is known[13] that the $G_xF_z \to G_zF_x$ spin-reorientation transition involves two second-order phase transitions at temperatures T_1 and T_2 ($T_1 < T_2$) and the nature of the phase transition at T_2 is not affected by a field $H \| c$, whereas the transition occurring at T_1 is not affected by a field $H \| a$; the other two phase transitions become broader; this is in agreement with the observation that the experimental curves obtained in a field $H \| c$ have a sharp kink of the $\delta(T)$ dependence at a certain temperature followed by a slow reduction of the effect as a result of cooling. Conversely, in $H \| a$ there is a kink after a smooth rise of the effect as a result of cooling. It is therefore natural to assume that in our figures the temperature of the kink of the $\delta(T)$ curve in $H \| a$ is T_1 and the kink in $H \| c$ is T_2. Since the magnetooptic method gives the characteristics of a surface layer of a magnetic material, whenever necessary we shall distinguish the surface critical tempera-

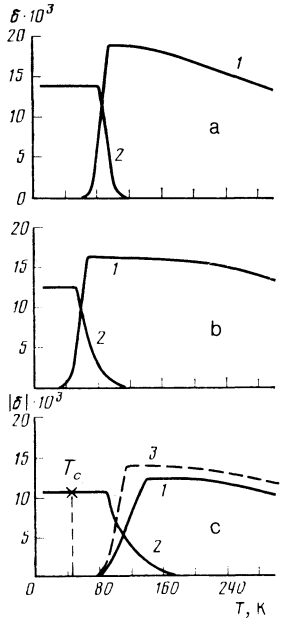

FIG. 5. Temperature dependences of the equatorial Kerr effect in $TmFeO_3$ (a), $HoFeO_3$ (b), and $ErFeO_3$ (c). $H = 1$ kOe, $\hbar\omega = 3.25$ eV; 1) $H \| c$; 2) $H \| a$, (010) plane; 3) $H \| c$, d plane. Here, T_c is the compensation temperature.

tures T_{1s} and T_{2s} and the volume temperatures T_{1v} and T_{2v}. We can see that in the case of $TmFeO_3$ ($T_{1s} = 85$ K, $T_{2s} = 95$ K) and $HoFeO_3$ ($T_{1s} = 53$ K, $T_{2s} = 65$ K) the temperatures T_{1s} and T_{2s} are practically the same as the corresponding temperatures of the spin-reorientation transition in the bulk, determined by a variety of methods.[13] In the case of $ErFeO_3$ there is a strong upward shift of the temperature T_{2s} compared with T_{2v}: $T_{2v} = 100$ K (Ref. 14) and $T_{2s} = 140$ K.[1)] This means that, according to the phenomenological theory, the region of reversal of the sign of the effective anisotropy constant K_1 is raised on the temperature scale by several tens of kelvin. It should be noted the upward shift of the temperature T_{2s} by 40 K, compared with T_{2v}, indicates the existence of surface magnetism in erbium orthoferrite on the (010) face. On the basis of the results obtained we can therefore say that in the temperature range from $T_{2s} = 140$ to $T_{2v} = 100$ K when the magnetic field is oriented along the c axis the magnetic moment b in the bulk of the sample is oriented exactly along the c axis, whereas on the surface it is tilted relative to the c axis by an angle which increases as a result of cooling and which reaches 90° at the temperature T_{2v}. The conclusion that, as in the case of hematite,[15] the physical reason for the appearance of surface magnetism in rare-earth orthoferrites is the lowering of the symmetry of the environment of surface magnetically ions (and not random factors such as mechanical cold working during polishing or surface contamination) was supported to some extent by the following experiment. An $ErFeO_3$ single crystal belonging to the same growth batch was cut (by the same technology as before) to form a plate oriented so that its large face was rotated by about 40° about the [001] axis relative to the initial (010) orientation (we shall call the new plane d). For this plane the temperature T_{2s} shifted downward to $T_{2s} = 113$ K [in Fig. 5c the $\delta(T)$ curve obtained for this sample in a field $H \| c$ is shown dashed].

According to the current ideas, spin-reorientations of the $G_xF_z \rightarrow G_zF_x$ type occur not because of the temperature dependence of the anisotropy constant of the iron sublattices, but because the influence of the rare-earth sublattice increases as a result of cooling. The Fe-Fe magnetic-dipole interaction does not participate in this spin-reorientation transition, because it stabilizes the G_xF_z phase. The energy responsible for the change in the sign of the effective anisotropy constant K_1 is due to the Zeeman or the Van Vleck mechanisms in the case of rare-earth ions. The transfer of this interaction to the iron sublattice occurs because of the antisymmetric Fe-Fe exchange interaction. The spin-reorientation transition in $ErFeO_3$ can be explained by allowing simultaneously for the influence of the Zeeman and Van Vleck mechanisms.[13]

It follows from the above discussion that a common feature of the appearance of surface magnetism in hematite and in rare-earth orthoferrites is its observation in the region of the spin-reorientation transition, i.e., in the region where the anisotropy constant K_1 exhibits a reversal of its sign as a small value so that the surface anisotropy K_s is competitive with the anisotropy K_1 when an equilibrium orientation of the vector $\mathbf{m}_s$ is established. On the other hand, a satisfactory explanation of surface magnetism in hematite can only be obtained if we allow for the change in the Fe-Fe magnetic-dipole interaction in a surface layer[15] and in the case of rare-earth orthoferrites we need to allow for the different types of the R-Fe interactions, which is a more difficult task. The qualitative difference between the results obtained, on the one hand, for $TmFeO_3$ and $HoFeO_3$ and, on the other, for $ErFeO_3$, can be understood if we bear in mind that the appearance of surface magnetism is in a sense a threshold process[15] which occurs if the gain resulting from the surface anisotropy energy K_s exceeds the characteristic energy of formation of a domain wall $\sigma_0 = (AK)^{1/2}$ in a surface layer (in the case of $ErFeO_3$). If $K_s < \sigma_0$, then the magnetization $\mathbf{m}_s$ on the surface is oriented in the same way as in the bulk (in the case of $TmFeO_3$ and $HoFeO_3$). The angle θ governing the orientation of the vector $\mathbf{m}_s$ relative to the c axis in the surface layer of erbium orthoferrite can be determined in the temperature range 140–100 K from the relationship $\cos\theta = \delta_T/\delta_s$, where δ_T is the magnitude of the equatorial Kerr effect at a given temperature T in a field $H \| c$, whereas δ_s is the magnitude of the effect at a temperature $T > T_s$. Our experiments indicated that the value of θ was practically independent of the magnetic field when the latter was varied from 2 to 40 kOe. Figure 6 shows the field dependences of the equatorial Kerr effect in $ErFeO_3$ at temperatures in the range $T_{1s} - T_{2s}$ and of $HoFeO_3$ in the region of the spin-reorientation transition. Clearly, in the case of $HoFeO_3$ the equatorial Kerr effect changed considerably on increase in H. On the other hand, the same effect in $ErFeO_3$ was independent of H after a single-domain state was reached. This result could be explained as follows. An equilibrium value of the angle θ is established because of the surface anisotropy localized in the first few atomic layers on the surface. The structure of a transition layer can be modified if its energy in a magnetic field is comparable with the energy of a volume (bulk) anisotropy which rises considerably at temperatures $T > T_2$. For example, in the case of erbium orthoferrite at $T = 130$ K we have $K_1 = 3.2 \times 10^5$ erg/cm^3, i.e., the effective anisotropy field is $H_c = 2K_1/m_s = 64$ kOe (Ref. 14), which is considerably greater than the external magnetic field H.

We conclude by noting that the observed features of the spin-reorientation transition in the surface layer of erbium orthoferrite are not exclusive to the family of rare-earth orthoferrites. For example, preliminary experiments carried

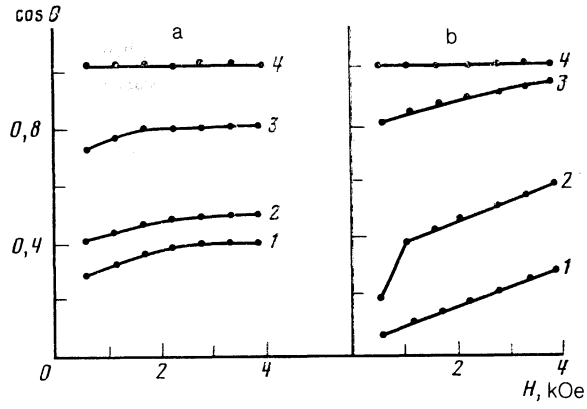

FIG. 6. Field dependences of cos θ for $ErFeO_3$ (a) and $HoFeO_3$ (b); $H \| c$, (010) plane. a) Curves 1, 2, 3, and 4 were obtained at temperatures $T = 95$ K, 100 K, 120 K, and 140 K, respectively. b) Curves 1, 2, 3, and 4 were obtained at temperatures $T = 50$ K, 55 K, 60 K, and 65 K, respectively.

out on single crystals of terbium orthoferrite have shown that on a natural (110) face the temperature T_{2s} of the spin-reorientation transition is also shifted strongly in the upward direction (it is reported in Refs. 16 and 17 that $T_{2v} = 6.5$ K, whereas the surface temperature is $T_{2s} = 50$ K). Moreover it is possible that even the nature of the spin-orientation transition changes in this orthoferrite. It is known[16,17] that the $G_xF_z \rightarrow G_zF_z$ spin-reorientation transition occurs in the bulk of $TbFeO_3$, whereas our magnetooptic measurements indicate the occurrence of spin-reorientation transition of the Morin type in the surface layer, i.e., the transition takes place to a pure antiferromagnetic state (at $T < T_{2s}$ there is no magnetooptic effect due to the components F_x and G_z).

The authors are deeply grateful to A. M. Balbashov for supplying orthoferrite single crystals, to N. I. Shpin'kov for magnetic measurements, and to V. N. Milov for his help in the construction of a helium optical cryostat.

[1)]The temperatures T_{1v} and T_{2v}, determined for our $ErFeO_3$ sample from the temperature dependences of the magnetization by N. I. Shpin'kov, were 93 and 103 K.

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Translated by A. Tybulewicz