Anisotropy of the Faraday effect in the weak ferromagnet $YFeO_3$

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An experimental investigation was made of the magnetic-field dependence of the Faraday effect in a weak ferromagnet $YFeO_3$ at the wavelength $\lambda = 0.63\,\mu$m. Measurements were made for different orientations of the direction of light propagation **k** and of the magnetic field **H**. Changes in the Faraday effect in the $\mathbf{k}\|\mathbf{c}$, $\mathbf{H}\|\mathbf{a}$ case were not proportional to changes in the component of the magnetic moment m_z. A phenomenological description of the Faraday effect in $YFeO_3$ was used to separate the ferromagnetic, antiferromagnetic, and diamagnetic contributions to the effect. The antiferromagnetic contribution dominating the Faraday effect was strongly anisotropic. A theoretical analysis was made of the microscopic Faraday effect mechanisms in $YFeO_3$ using the example of a dipole-allowed transition ${}^6A_{1g} \rightarrow {}^6T_{1u}$. The mechanisms proposed earlier could not account for the strong anisotropy of the antiferromagnetic contribution and a satisfactory explanation required allowance for the "spin–foreign orbit" exchange-relativistic interaction.

1. INTRODUCTION

Investigations of the magnetooptic rotation of the plane of polarization of light, known as the Faraday effect, in rare-earth orthoferrites $RFeO_3$ have been going on for twenty years since these crystals were synthesized in the late sixties. The interest in the Faraday effect is due to unusual magnetooptic properties of these materials compared with another large class of magnetic insulators composed of the rare-earth iron garnets $R_3Fe_5O_{12}$. In contrast to ferrimagnetic rare-earth iron garnets, the majority of rare-earth orthoferrites are weak ferromagnets at room temperature and their spontaneous magnetic moment m is approximately an order of magnitude less than the moment of iron garnets. Nevertheless, numerous investigations have demonstrated[1–8] that the components responsible for the Faraday effect in the transparency range are an order of magnitude greater for rare-earth orthoferrites than for iron garnets. Moreover, the Faraday effect in these two types of crystal has opposite signs. Since rare-earth orthoferrites and iron garnets consist of the same types of ions (R^{3+}, Fe^{3+}, O^{2-}), and explanation of the experimental observations should be sought in microscopic Faraday effect mechanisms reflecting the differences between the magnetic and crystallographic structures of these crystals.

A phenomenological analysis of the Faraday effect in rare-earth orthoferrites[8–11] shows that the effect in these crystals and other weak ferromagnets may be associated both with the spontaneous magnetic moment **m** and with the antiferromagnetic moment **l**. The ferromagnetic and antiferromagnetic contributions to the Faraday effect in $YFeO_3$ for light propagating along the c axis are separated in Ref. 10. Assuming that the Faraday effect is governed mainly by the spin system, it is shown there that antiferromagnetic contribution in this crystal is approximately 7 times higher than the ferromagnetic component. However, the separation made in Ref. 10 requires additional confirmation by independent experiments to exclude a possible manifestation of the Faraday effect unrelated to the spin system, i.e., of the diamagnetic Faraday effect. Moreover, no data have yet been published on the anisotropy of the ferromagnetic and antiferromagnetic contributions, although such an anisotropy has been predicted phenomenologically and it is known that the Faraday effect in rare-earth orthoferrites is anisotropic. Our aim was to investigate experimentally the ferromagnetic and antiferromagnetic contributions to the Faraday effect in $YFeO_3$ and to identify the origin of these contributions on the basis of a microscopic theory.

2. PHENOMENOLOGICAL ANALYSIS

We now consider the Faraday effect in rare-earth orthoferrites employing the gyration vector **g** to describe it.[12] This axial vector, which is a dual of the antisymmetric part of the permittivity tensor ε_{ij}, governs the Faraday rotation θ_F in noncubic crystals in accordance with the relationship

$$\theta_F = A\,\mathrm{Re}(\mathbf{gn}), \tag{1}$$

where **n** is a unit vector in the direction of light propagation **k**, and A is a coefficient which depends on the direction of **k**, on the polarization of light, and on the principal values of the refractive index.

In the case of cubic ferromagnets and ferrimagnets, such as rare-earth iron garnets, the vector **g** is related linearly to the magnetic moment **m** or to the sublattice magnetizations $\mathbf{m}_i$ (Refs. 13–15), so that the components of the spins perpendicular to **g** make no contribution to the gyration vector. On the other hand, the orthogonal components of the ferromagnetic (**m**) and antiferromagnetic (**l**) vectors in weak ferromagnets may transform identically and the expression for g_i can be written in the form[8–11]

$$g_i = \alpha_{ij} m_j + \beta_{ij} l_j. \tag{2}$$

The components of the spins perpendicular to $\mathbf{g}_i$ may contribute to the gyration vector. For example, in the case of rare-earth othoferrites the components m_x and l_z, and also m_z and l_z transform identically,[16] and in this case the tensors α_{ij} and β_{ij} are of the form[11,17]

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$$\alpha_{ij}=\begin{pmatrix}\alpha_{xx} & 0 & 0\\ 0 & \alpha_{yy} & 0\\ 0 & 0 & \alpha_{zz}\end{pmatrix},\quad \beta_{ij}=\begin{pmatrix}0 & 0 & \beta_{xz}\\ 0 & 0 & 0\\ \beta_{zx} & 0 & 0\end{pmatrix}, \tag{3}$$

where the tensor β_{ij} is generally asymmetric. It is clear from Eq. (3) that the symmetry group of rare-earth orthoferrites imposes no restrictions on the relationships between the coefficients α_{ij} and β_{ij}, so that the Faraday effects in these crystals may in general be anisotropic.

If a crystal is subjected to a magnetic field H, we have to include in Eq. (2) also terms of the $\gamma_{ij}H_j$ type describing the diamagnetic Faraday effect, in which the structure of the tensor γ_{ij} is analogous to α_{ij}. Therefore, it follows from a phenomenological analysis that the Faraday effect in weak ferromagnets, particularly in rare-earth orthoferrites, consists of three contributions: ferromagnetic, proportional to the components m_i; antiferromagnetic, proportional to l_j; diamagnetic, proportional to H_i.

3. METHOD OF DETERMINATION OF FERROMAGNETIC, ANTIFERROMAGNETIC, AND DIAMAGNETIC CONTRIBUTIONS TO THE FARADAY EFFECT

The task of determining the ferromagnetic, antiferromagnetic, and diamagnetic contributions to the Faraday effect in $YFeO_3$ reduces to finding processes acting on a crystal which would make it possible to write down eight linearly independent equations for the unknowns α_{ii}, β_{ij}, and γ_{ii}. The problem can be partly solved by investigating the magnetic-field dependence of the Faraday effect, because the vectors **m** and **l** then vary differently.

We shall consider the case of propagation of light in a rare-earth orthoferrite in the *ac* plane at an angle α to the c axis when an external field **H** is applied in this plane at an angle ξ to the c axis (Fig. 1a). According to Ref. 18, the application of a magnetic field H causes the components m_x, m_z, l_x, and l_z to vary in the following way[1)]:

$$\begin{aligned} m_x&=[m_0+\eta\chi_\perp H\cos(\xi-\varphi)]\sin\varphi+(1-\eta)\chi_\perp H\sin\xi,\\ m_z&=[m_0+\eta\chi_\perp H\cos(\xi-\varphi)]\cos\varphi+(1-\eta)\chi_\perp H\cos\xi,\\ l_x&=l_0\cos\varphi,\quad l_z=-l_0\sin\varphi, \end{aligned} \tag{4}$$

where all the notation in Eq. (4) is taken from Ref. 18. The expression for the Faraday effect $\theta_F(H)$ can then be deduced from Eqs. (1) and (3):

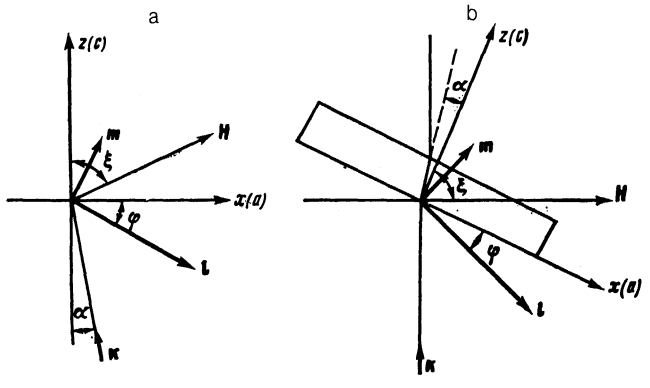

FIG. 1. Mutual orientations of the vectors **m**, **l**, **k**, and **H** and of the crystallographic axes x and z (corresponding to the a and c axes), used in the calculation of the Faraday effect (a) and the experimental geometry used in a study of the Faraday effect in $YFeO_3$ (b).

$$\begin{aligned} \theta_F(H)=A[&(\alpha_{zz}m_z+\beta_{zx}l_x)\cos\alpha-(\alpha_{xx}m_x-\beta_{xz}l_z)\sin\alpha\\ &+\gamma_{zz}H_z\cos\alpha-\gamma_{xx}H_x\sin\alpha]. \end{aligned} \tag{5}$$

The expression for the relative value of the Faraday effect $\theta_F(H)/\theta_F(0)$ obtained using Eq. (4) is

$$\begin{aligned} \theta_F(H)/\theta_F(0)=&\cos\varphi+\alpha_{zz}\chi_\perp H[\eta\cos(\xi-\varphi)\cos\varphi\\ &+(1-\eta)\cos\xi]/g_{z0}-g_{x0}\operatorname{tg}\alpha/g_{z0}-\alpha_{xx}\chi_\perp H[\eta\cos(\xi-\varphi)\sin\varphi\\ &+(1-\eta)\sin\xi]\operatorname{tg}\alpha/g_{z0}+\gamma_{zz}H\cos\xi/g_{z0}+\gamma_{xx}H\sin\xi\operatorname{tg}\alpha/g_{z0}, \end{aligned} \tag{6}$$

where

$$g_{x0}=\alpha_{xx}m_0-\beta_{xz}l_0,\quad g_{z0}=\alpha_{zz}m_0+\beta_{zx}l_0.$$

Assuming $\sin\varphi=rH$, where $r=\text{const}$ holds in the range $H\leqslant 20$ kOe (Refs. 18–20), we can rewrite Eq. (6) with accuracy up to terms quadratic in H, which gives

$$\theta_F(H)/\theta_F(0)=1+pH+qH^2,$$

where

$$\begin{aligned} p=&\alpha_{zz}\chi_\perp\cos\xi/g_{z0}-\operatorname{tg}\alpha[g_{x0}r+\alpha_{xx}\chi_\perp(1-\eta)\sin\xi]/g_{z0}\\ &+(\gamma_{zz}\cos\xi-\gamma_{xx}\sin\xi\operatorname{tg}\alpha)/g_{z0},\\ q=&-r^2/2+\chi_\perp r\eta(\alpha_{zz}\sin\xi-\alpha_{xx}\cos\xi\operatorname{tg}\alpha)/g_{z0}. \end{aligned} \tag{7}$$

For $\alpha=0$, $\xi=0$ and $\xi=90°$, Eq. (6) reduces to a system of three linearly independent equations for α_{zz}, β_{xz}, and γ_{zz}:

$$A(\alpha_{zz}m_0+\beta_{zx}l_0)={}^1/_2[\theta_F(+H)-\theta_F(-H)]|_{H\to 0},\quad \alpha=0,\ \xi=0, \tag{8}$$

$$\theta_F(H)/\theta_F(0)=1+\alpha_{zz}\chi_\perp H/g_{z0}+\gamma_{zz}H/g_{z0},\quad \alpha=0,\ \xi=0, \tag{9}$$

$$\theta_F(H)/\theta_F(0)=1+(\alpha_{zz}r\eta\chi_\perp/g_{z0}-r^2/2)H^2,\quad \alpha=0,\ \xi=90°. \tag{10}$$

Equation (8) describes a jump in the field dependence of the Faraday effect due to magnetization reversal in a crystal, Eq. (9) is the dependence $\theta_F(H)$ linear in H in the case when $\xi=0$, and Eq. (10) gives the dependence of H which is quadratic when $\xi=90°$. The procedure used to determine A is described in Ref. 8.

For $\alpha\neq 0$, the field dependence of $\theta_F(H)/\theta_F(0)$ should contain not only a component quadratic in H, but also a linear one [in accordance with Eq. (7)], and the coefficient of the linear part should increase with increasing α. The expressions for the coefficients p and q in Eq. (7) are then governed not only by α_{zz}, β_{zx}, and γ_{zz}, but also by the component of the gyration vector g_{x0} and by the quantities α_{xx} and γ_{xx}. We can estimate g_{x0} also from the field dependence of the Faraday effect measured for a sheet of $YFeO_3$ cut in a plane containing the c axis and the diagonal between the a and b axes (d plane). The expression for the Faraday effect then becomes

$$\theta_F(H)=A[g_{x0}\sin\varphi+2^{-1/2}H(\gamma_{xx}-\gamma_{yy})-2^{-1/2}\alpha_{yy}\chi_\perp H]. \tag{11}$$

Therefore, it is clear from Eqs. (6)–(11) that in the case of $YFeO_3$, an investigation of the field dependence makes it possible to determine the following components of the tensors α_{ij}, β_{ij}, and γ_{ij}:

$$\alpha_{zz},\ \beta_{xz},\ \gamma_{zz},\ \alpha_{xx},\ \beta_{zx}.$$

4. APPARATUS

We investigated experimentally the magnetic-field dependence of the rotation of the plane of polarization of light at the wavelength of $\lambda = 0.6328$ μm when light was transmitted by a $YFeO_3$ plate. The apparatus used to determine the rotation of the plane of polarization was determined to within $\sim 10''$. The crystals of $YFeO_3$ were plane-parallel sheets of thickness ~ 100 μm cut in the c plane. The crystals were oriented by x-ray diffraction to within ~ 1–$2°$. We employed the experimental geometry shown in Fig. 1b. The direction of light propagation **k** was perpendicular to the direction of the magnetic field **H**, which could be varied up to 20 kOe. The crystal, located in the gap of the magnet, could be rotated about an axis orthogonal to **H** and **k**. The light was polarized parallel to the b or a axis, and the angle between **H** and the c axis in the ac plane was selected so that the condition $\delta = \pi/2 + \pi n$ was satisfied (δ is the plane shift), when the magnitude of the Faraday effect was maximal.[10] These measurements were carried out at $T = 295$ K.

5. EXPERIMENTAL RESULTS AND DISCUSSION

Figure 2 shows the magnetic-field dependence of the relative Faraday effect $\theta_F(H)/|\theta_F(0)|$ in $YFeO_3$ measured for angles $\xi = 85°$, $\alpha \approx 2°$ and $\xi = 60°$, $\alpha \approx 12°$. The quantity ξ in the former case was selected to be 85° and not 90°, so that we could reverse the magnetization of the crystal without changing its position within the magnetic gap. The deviation of ξ from 90° created a small projection of the magnetic field along the c axis which was sufficient to reverse the magnetization of the sample in a field $H = 20$ kOe. The value of $\theta_F(0)$ was deduced from the jump in the field dependence of the Faraday effect as a result of magnetization reversal; in the angles ξ given above it amounted to $\theta_F^{85} = 1.388°$, $\theta_F^{60} = 1.573°$ (**E**$\|$**b**). The dependences $\theta_F(H)/\theta_F(0)$ are of the same nature for different directions of polarization of the incident light.

The dependence $\theta_F(H)/\theta_F(0)$ is an odd function of the magnetic field H, which demonstrates that the observed rotation of the plane of polarization was due to the Faraday effect and not due to contributions quadratic in **l**, **m**, or **H**. Nevertheless, since the value of A in Eq. (1) depends on the birefringence of the crystal, we allowed for the field dependence of the birefringence[8] in analyzing the results of the measurements. In Fig. 2 we also plotted the field dependence of $\theta_F(H)/\theta_F(0)$ without allowance for the antiferromagnetic contribution, which was identical with the field dependence of the projection of the magnetization **m** along **k** in the case when $\xi = 85°$, calculated using the expressions in the system (4) and the results taken from Refs. 18–20. As in the case when $\xi = 0$, $\alpha = 0$ (Ref. 10), the Faraday effect at the angles $\alpha = 2°$, $\xi = 85°$ did not vary proportionally to m_z. Since in this experimental geometry ($\xi = 85°$, $\alpha = 2°$) the diamagnetic contribution ot the Faraday effect was practically zero, this lack of proportionality indicated that a satisfactory description of the observed behavior required allowance for the antiferromagnetic contribution proportional to β_{ij}.

The field dependence of $|[\theta_F(0) - \theta_F(H)]/\theta_F(0)H|$ plotted for $\xi = 85°$ on the basis of the experimental data was a linear function of the magnetic field (Fig. 3), in agreement with the phenomenological description given by Eq. (9). A slight deviation of ξ from 90° should generally give rise not only to a term proportional to the square of H in Eq. (9), but also to a linear term associated with g_{x0} and with the change in m_z due the susceptibility $\chi_\perp$ [see Eq. (7)]. However, it is clear from Fig. 3 that in the case under discussion the contribution was negligible.

A simultaneous analysis of the dependence $\theta_F(H)$ for the angles $\xi = 85°$, $\alpha = 2°$ and for the case $\xi = 0$, $\alpha = 0$, considered in Ref. 10, made it psosible to determine from Eqs. (8) and (9) the coefficients α_{zz}, β_{xz}, γ_{zz}:

$$g_{z0} = \alpha_{zz} m_0 + \beta_{zz} l_0 = (4.10 \pm 0.05) \cdot 10^{-3},$$
$$\alpha_{zz} m_0 = (0.95 \pm 0.55) \cdot 10^{-3}, \quad \beta_{zz} l_0 = (3.15 \pm 0.55) \cdot 10^{-3}, \quad (12)$$
$$\gamma_{zz} = (-1.1 \pm 2.8) \cdot 10^{-6} \text{ kOe}^{-1}.$$

The wide range of intervals within which α_{zz}, β_{xz}, γ_{zz} were determined was due to the low precision of the results of the static magnetic measurements used to find the coefficient r, i.e., the angle φ. It is clear from Eq. (12) that the Faraday effect in $YFeO_3$ was governed mainly by the antiferromagnetic contribution. The magnitude of the diamagnetic contribution was less than the contributions associated with the **m** and **l** components. For example, at $H = 20$ kOe, the contribution to the gyration vector related to the tensor γ_{ij} is $\approx 2 \times 10^{-5}$, which corresponds to the Verdet constants of the diamagnetics and ferrimagnetics amounting to $\approx 1°/(\text{cm} \cdot \text{kOe})$.

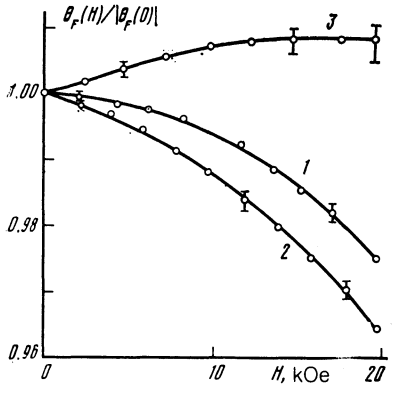

FIG. 2. Experimental field dependence of the relative Faraday effect $\theta_F(H)/|\theta_F(0)|$ for two values of the angle $\xi = 85°$ (1) and $\xi = 60°$ (2). This figure includes also the results of a calculation of the Faraday effect for $\xi = 85°$ and $\beta_{ik} = 0$ (curve 3).

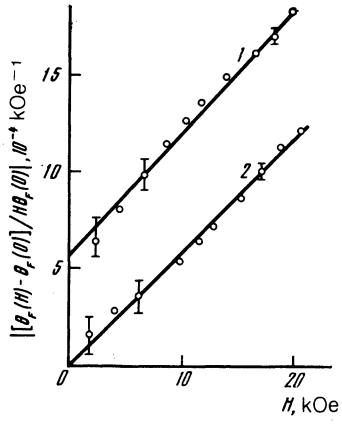

FIG. 3. Field dependences of $|[\theta_F(H) - \theta_F(0)]/H\theta_F(0)|$ for two values of the angle $\xi = 85°$ (1) and $\xi = 60°$ (2).

Figure 3 shows the field dependence of $|[\theta_F(0)-\theta_F(H)]/\theta_F(0)H|$ plotted for $\xi = 60°$ on the basis of the experimental data. It is clear from this figure that the dependence $\theta_F(H)/|\theta_F(0)|$ has linear and quadratic parts, in agreement with Eq. (7). An analysis of the dependence $\theta_F(H)$ on the basis of Eq. (7) allowed us to determine the values of α_{xx} and β_{xz} on the assumption that $\gamma_{xx} \approx \gamma_{zz}$:

$$\alpha_{xx}m_0=(0.2\pm0.7)\cdot10^{-3},\quad \beta_{xz0}=(-2.1\pm1.0)\cdot10^{-3},\quad g_{x0}=(1.64\pm0.41)\cdot10^{-3}. \tag{13}$$

The tensor β_{ij} in $YFeO_3$ therefore is asymmetric and the components β_{xz} and β_{zx} have opposite signs. Hence the ratio $g_{z0}/g_{x0} \approx 2$, i.e., the Faraday effect in $YFeO_3$, is anisotropic. The absolute value of the ratio g_{z0}/g_{x0} was determined earlier[8] in measurements of the Faraday effect in samples cut in the d plane and the value obtained then agreed with that given above. The Faraday effect exhibits approximately the same anisotropy also in other rare-earth orthoferrites, as found in investigations of the magnetic orientational phase transition.[5]

6. MECHANISMS OF MAGNETOOPTIC ACTIVITY OF RARE-EARTH ORTHOFERRITES

It is generally accepted[1] that the major (if not dominant) role in the magnetooptic rotation of visible and ultraviolet light in rare-earth orthoferrites is played by allowed electric-dipole charge-transfer ${}^6A_{1g}-{}^6T_{1u}$ transitions in octahedral complexes. We shall consider the contribution of such transitions to the ferromagnetic and antiferromagnetic Faraday effects.

In the case of such S ions as Fe^{3+} the magnetooptic properties are dominated by the spin–orbit interaction in excited states $V_{so} = \lambda\,\mathbf{LS}$, where **S** is the spin and **L** is the effective orbital momentum ($L = 1$ for the ${}^6T_{1u}$ term). Thus, allowance for the spin–orbit interaction considered in the linear approximation contributes

$$\mathbf{g}=\left(\frac{n_0^2+2}{3}\right)^2\frac{2\pi e^2\lambda f}{m\omega_0}\frac{\partial F(\omega,\omega_0)}{\partial\omega_0}\sum_n\langle\mathbf{S}(n)\rangle=\alpha\mathbf{m}, \tag{14}$$

to the gyration vector, where f is the oscillator strength of the ${}^6A_{1g}\rightarrow{}^6T_{1u}$ transition; $F(\omega,\omega_0)$ is the dispersion factor; and far from the transition, we have

$$\mathrm{Re}(\partial F/\partial\omega_0)\approx4\omega\omega_0/\hbar(\omega^2-\omega_0^2)^2.$$

This magnetooptic activity mechanism accounts only for the main isotropic part of the usual ferromagnetic contribution to the gyration vector $\alpha_{xx} = \alpha_{yy} = \alpha_{zz} = \alpha$.

The antiferromagnetic contribution originates from the anisotropic coupling of the gyration vector to the spins of the Fe^{3+} ions.[1] This anisotropy may be due to the low-symmetry crystal field (LCF) acting on excited ${}^6T_{1u}$ states of FeO_6^{9-} complexes:

$$V_{\mathrm{LCF}}=\sum_{ij}B_{ij}\hat{L}_i\hat{L}_j, \tag{15}$$

where B_{ij} is the symmetric tensor of the LCF parameters such that $\mathrm{Tr}\,\hat{B}=0$.

In the approximation linear in V_{LCF} and V_{so} the coupling of the gyration vector to the spins of the Fe^{3+} ions can be represented as follows:

$$g_i=\left(\frac{n_0^2+2}{3}\right)^2\frac{\pi e^2\lambda f}{m\omega_0\hbar}\frac{\partial^2F(\omega,\omega_0)}{\partial\omega_0^2}\sum_n B_{ij}(n)\langle S_j(n)\rangle. \tag{16}$$

Going over from the spins to the vectors **m** and **l**, and allowing for the ratio of the signs of the LCF parameters for the different positions of the Fe^{3+} ions in a unit cell of $YFeO_3$, we can represent Eq. (16) in the form of Eq. (2):

$$\Delta\alpha_{ii}=\left(\frac{n_0^2+2}{3}\right)^2\frac{\pi e^2\lambda f}{m\omega_0\hbar}\frac{\partial^2F(\omega,\omega_0)}{\partial\omega_0^2}B_{ii}, \tag{17}$$

$$\beta_{zx}=\beta_{xz}=\left(\frac{n_0^2+2}{3}\right)^2\frac{\pi e^2\lambda f}{m\omega_0\hbar}\frac{\partial^2F(\omega,\omega_0)}{\partial\omega_0^2}B_{xz}, \tag{18}$$

where the parameters apply to the position (1/2, 0, 0). Far from the transition, we have

$$\mathrm{Re}(\partial^2F/\partial\omega_0^2)\approx4\omega(\omega^2+\omega_0^2)/(\omega_0^2-\omega^2)^3\hbar,$$

so that in estimating the ratio of the anisotropic to the isotropic contribution we can use

$$\frac{\Delta\alpha_{ii}}{\alpha}\approx\frac{\omega^2+\omega_0^2}{2(\omega_0^2-\omega^2)}\frac{B_{ii}}{\hbar\omega_0},\qquad \frac{\beta_{zx}}{\alpha}\approx\frac{\omega^2+\omega_0^2}{2(\omega^2-\omega_0^2)}\frac{B_{xz}}{\hbar\omega_0}, \tag{19}$$

which for $\hbar\omega \approx 2$ eV, $\hbar\omega_0 \approx 4$ eV, $|B_{ij}| \sim 0.1$ eV corresponds to values of the order of 0.01. Therefore, allowance for V_{LCF} leads to a weak anisotropy of the tensor α_{ij} and of the corresponding ferromagnetic Faraday effect, and gives rise to an antiferromagnetic contribution to the gyration vector generally comparable with the ferromagnetic contribution if $m/l \sim 0.01$. It should be noted that the frequency dependence of the conventional ferromagnetic contribution differs from that of the antiferromagnetic contribution.

The proposed mechanism of the antiferromagnetic Faraday effect is in principle equivalent to the "anisotropic orbital freezing" mechanism proposed qualitatively in Ref. 1. However, this mechanism does not account for the experimentally observed strong asymmetry of the tensor β_{ij}. We can explain this observation allowing for the fact that the FeO_6^{9-} complexes in rare-earth orthoferrites are subject to strong effective magnetic fields of exchange and exchange-relativistic origin. This is allowed for implicitly also in the mechanisms discussed above. In fact, the spin or exchange polarization of the ground state of the FeO_6^{9-} complexes is due to a strong exchange field H_E and a weaker Dzyaloshinskii field H_D. In the case of $YFeO_3$ at 4.2 K, we have $H_E = 640$ T and $H_D = 14$ T. It would seem that the presence of such strong fields should result in very large values of the usual diamagnetic contribution to the Faraday effect, which is due to the Zeeman splitting of the excited states. In fact, this does not occur. This is because the conventional diamagnetic contribution to the Faraday effect is solely due to the orbital splitting in an external magnetic field, i.e., it is associated only with the orbital part of the Zeeman interaction $V_z = -\mu_B(\mathbf{L}+2\mathbf{S})\mathbf{H}$, whereas the effective fields H_E and H_D are of purely exchange origin and—in contrast to the intrinsic magnetic field—can only create spin splitting.

It therefore follows that the effective exchange magnetic field, which could be responsible for the diamagnetic contribution to the Faraday effect, is associated with the interaction which is linear in the orbital momentum ${}^6T_{1u}$ of the state of the FeO_6^{9-} complexes. The interaction is known and is an exchange-relativistic interaction of the "spin–foreign

orbit" type. In the case of rare-earth orthoferrites, as with all weak ferromagnets, it can be represented by a sum of three contributions:

$$V'_{so} = \sum_n \{\lambda^{(0)}(n)\mathbf{LS}(n) + \lambda^{(1)}(n)[\mathbf{LS}(n) + \mathbf{L}\hat{\lambda}^{(2)}(n)\mathbf{S}(n)\}, \tag{20}$$

namely the isotropic, antisymmetric, and anisotropic symmetric (the tensor $\hat{\lambda}^{(2)}$ in the above expression is symmetric and its trace is zero). The "spin–foreign orbit" interaction is of similar origin to the antisymmetric Dzyaloshinskii–Moriya interaction:

$$V_{as} = \sum_n \mathbf{d}(n)[\mathbf{SS}(n)], \tag{21}$$

and is obtained if we allow for the simultaneous effects of the spin–orbit interaction in the case of the FeO_6^{9-} complexes and of the exchange interaction with the neighboring complexes. Therefore, in estimating the parameters $\lambda^{(0)}, \lambda^{(1)}, \hat{\lambda}^{(2)}$ we can use an approximate relationship $\lambda(n) \propto \lambda' I'/\Delta E$, where λ' and I' are the off-diagonal parameters of the spin–orbit interaction and of the exchange, whereas ΔE is the separation from the ${}^6T_{1u}$ level to the nearest levels which are mixed by V_{so} and V_{exch}.

Therefore, the effective field due to the "spin–foreign orbit" interaction, corresponding specifically to the orbital Zeeman interaction, can reach values in excess of 10 T ($\lambda' \lesssim 10^2$ cm^{-1}, $I' \gtrsim 10^2$ cm^{-1}, $\Delta E \lesssim 10^4$ cm^{-1}). We can easily see that allowance for the isotropic "spin–foreign orbit" interaction gives rise to an additional isotropic ferromagnetic contribution to the gyration vector, whereas the second and third terms in Eq. (20) contribute to the tensor β_{ij} and the antisymmetric part of the tensor β_{ij} is governed entirely by the antisymmetric "spin–foreign orbit" interaction:

$$\beta_{zx} = -\beta_{xz} = \left(\frac{n_0^2+2}{3}\right)^2 \frac{2\pi e^2 f}{m\omega_0} \frac{\partial F(\omega, \omega_0)}{\partial \omega_0} \sum_n \lambda^{(1)}(n). \tag{22}$$

The antisymmetric "spin–foreign orbit" interaction gives rise to a characteristic "orbital" Dzyaloshinskii field H'_D for each of the excited states of the FeO_6^{9-} complexes with a nonzero orbital momentum. Clearly, such a situation occurs only in weak ferromagnets in which a conventional Dzyaloshinskii field may exist.

It follows that whereas the existence of this tensor β_{ij}, i.e., the antiferromagnetic contribution to the gyration vector, is typical of a large number of multisublattice magnetic materials, the asymmetry of the tensor β_{ij} is a specific feature of weak ferromagnets alone. In the case of rhombohedral weak ferromagnets such as $FeBO_3$, FeF_3, or α-Fe_2O_3, the tensor β_{ij} governing the antiferromagnetic contribution to the Faraday effect is entirely due to the antisymmetric contribution, in view of the requirements imposed by the crystal symmetry. In crystals of this kind the appearance of the antiferromagnetic contribution to the gyration vector is entirely due to allowance for the antisymmetric "spin–foreign orbit" interaction.

7. CONCLUSIONS

The main result of the present study is separation of the mechanisms governing the Faraday effect in a weak ferromagnet $YFeO_3$ and a theoretical description of the microscopic nature of these mechanisms. The dominant contribution to the gyration vector **g** is the antiferromagnetic Faraday effect associated with the components of the spins perpendicular to the direction of the vector **g** and characterized by a strong anisotropy. A theoretical analysis shows that in describing the magnetooptic effects in $YFeO_3$ and also in other weak ferromagnets we need to allow for a new exchange-relativistic interaction of the "spin–foreign orbit" type, which gives rise to symmetric and antisymmetric contributions to the antiferromagnetic Faraday effect. The strong anisotropy of the antiferromagnetic Faraday effect in $YFeO_3$ demonstrates the predominant role played in this crystal by the antisymmetric "spin–foreign orbit" interaction.

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[1)] In this geometry, when we consider rotations of the plane of polarization, we need not allow for quadratic terms of the $\varepsilon_{xz} \sim l_x l_z$ type, because they do not rotate the principal directions in the section of the indicatrix perpendicular to **k**.

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