Anisotropy of linear magneto-optical effects in thulium orthoferrite

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The Faraday effect in $TmFeO_3$ is investigated for various directions of the weak ferromagnetic moment. The off-diagonal components of the tensor $\hat{\epsilon}$–$\gamma(\lambda)$ responsible for the extraordinarily large linear magneto-optical effects in orthoferrites are determined. It is shown that the contribution of Tm^{3+} ions to these effects is small. It is found that $\gamma(\lambda)$ is strongly anisotropic and varies by more than two times upon reorientation of the weak ferromagnetic moment. It is concluded that the noncubic character of the crystal fields acting on the octahedral complex FeO_4 must be taken into account in the theory of linear magneto-optical effects in orthoferrites.

Much interest has been evinced recently in experimental investigations of linear magnetooptical effects in weak ferromagnets. Studies were made of the polar Kerr effect[1], of elliptic birefringence[2,3] and the Faraday effect[4] in orthoferrites, and of the equatorial Kerr effect in hematite[5]. The unusually large values of off-diagonal components of the dielectric tensor of orthoferrite, which determine all the aforementioned linear magnetooptical effects, are customarily attributed to the noncubic character of the crystal fields acting on the iron ions[1,6]. Kahn, Pershan, and Remeika[1] have proposed a mechanism of "anisotropic quenching of the orbital angular momentum." To explain the mechanism responsible for the linear magnetooptical effects in orthoferrites, it is of interest to investigate experimentally their anisotropy. The present paper is devoted to a study of the anisotropy of the Faraday effect in thulium orthoferrite. There have been no experimental investigations of the anisotropy of the Faraday effect in orthoferrites to date. The anisotropy cannot be determined by a comparison of the off-diagonal components of the dielectric tensor for different directions in orthoferrite crystals with various compositions, since orthoferrites with different compositions but with the same direction of the weak ferromagnetic moment have values of the Faraday effect that differ by 30–40% in a broad spectral interval[4].

Investigations of the Faraday effect in orthoferrites have shown that in the visible region of the spectrum this effect is quite large and amounts to several thousand degrees per centimeter[4]. Orthoferrites are orthorhombic biaxial crystals. Their optical axes lie in the (100) plane and make angles of the order of 50° with the [001] axis. Therefore the Faraday effect, which is proportional to the thickness of the sample, can be realized when light propagates along the optical axis only in those orthoferrites, whose weak ferromagnetic moment is directed along the [001] axis. If the magnetic moment is directed along the [100] axis, then its projection in the optical axes turns out to be zero and the Faraday effect, which is proportional to the sample thickness, vanishes. Even pulsed magnetic fields are insufficient to change significantly the direction of the spontaneous magnetic moment of the orthoferrite[7]. To investigate linear magnetooptical effects in orthoferrites at different directions of the magnetic moment it is therefore more convenient to use an orthoferrite with a reorientation point. In thulium orthoferrite, for example, in the temperature region 82–92°K, a spontaneous reorientation of the weak ferromagnetic moment takes place from the

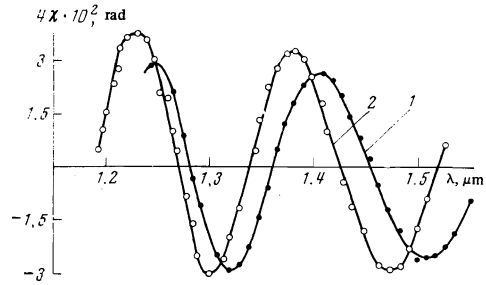

FIG. 1. Dispersion dependence of the Faraday effect in a $TmFeO_3$ plate 350 μ thick at 290°K (curve 1) and at 100°K (curve 2). The light propagates along the [001] axis.

[100] axis to the [001] axis[7]. The off-diagonal component of the dielectric tensor of the orthoferrite can be determined in these two cases from the dispersion relations for the angle of rotation of the major axis of the resultant ellipse at the exit from the crystal, by using the procedure of the preceding paper[2].

Figure 1 (curve 1) shows the dispersion dependence of $\chi(\lambda)$–the angle of rotation of the major axis of the resultant ellipse relative to the direction of polarization of the incident light in $TmFeO_3$ 350 μ thick, with the light propagating along the [001] axis at room temperature. In the investigated wavelength range from 1.25 to 1.55 μ, there are no Tm^{3+} ion absorption lines. The angle χ is given by[8]

$$\operatorname{tg} 2\chi = \frac{I_+ - I_-}{2I_0} = \frac{\gamma}{n\Delta n} \sin \frac{2\pi \Delta n d}{\lambda}. \qquad (1)$$

Here $I_\pm$ is the intensity of the light passing through the system comprising a polarizer, the sample, and an analyzer oriented 45° relative to the polarizer, at the two possible directions of the sample magnetization, d is the sample thickness, and λ is the wavelength.

From the "period" of this dependence it is possible to determine the quantity $\Delta n = n_x - n_y = (3.12 \pm 0.05) \cdot 10^{-2}$, and then also the off-diagonal component of the tensor $\hat{\epsilon} - \epsilon_{xy} = \gamma(\lambda)$. The dispersion dependence of $\chi_1(\lambda)$ obtained in accordance with (1) from the experimental curve of Fig. 1 is shown in Fig. 2 (curve 1). It pertains to the temperature 290°K, i.e., to the case when the weak ferromagnetic moment of $TmFeO_3$ is directed along the [001] axis. The Faraday effect investigated on $TmFeO_3$ samples cut perpendicular to the optical axis at 290°K confirms this dependence. At temperatures below 82°K, the weak ferromagnetic moment of $TmFeO_3$ is directed along the [100] axis.

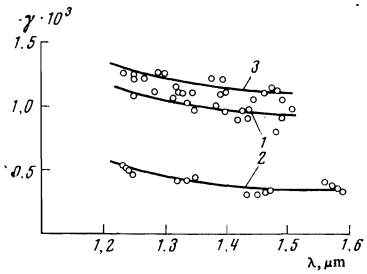

FIG. 2. Dispersion dependence of the off-diagonal component γ of the tensor ϵ for $TmFeO_3$ at 290° (curve 1), 80° (curve 2) and 100°K (curve 3).

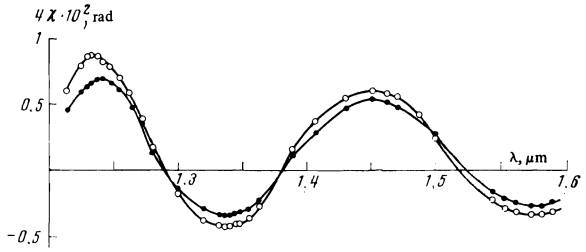

FIG. 3. Dispersion dependence of the Faraday effect in a $TmFeO_3$ plate 140 μ thick at 80°K. The light propagates along the [100] axis. The electric field of the light wave E ∥ [001], analyzer–along [011]–○, E ∥ [010], analyzer along [0$\bar{1}$1]–●.

Figure 3 shows the dispersion dependences of $(I_+-I_-)/I_0 = 4\chi$ in $TmFeO_3$ at 80°K for a plate 150 μ thick, perpendicular to the [100] axis, for the light-wave electric vector directed along the axes [001] and [010] of the crystal. Just as in the case of Fig. 1, these plots are oscillatory, but the curves are patently asymmetrical relative to the abscissa axis and are not described completely by relation (1). Curves analogous to those shown in Fig. 3 were obtained at 65°K, i.e., their shift relative to the abscissa axis is not due to the fact that the temperature 80°K is close to the reorientation region. Relation (1) is valid only for transparent crystals and does not take into account absorption or dichroism. It was shown earlier[6] that in the region of the absorption band of rare-earth ions they must be taken into account. Jastrzebski[9] obtained relations that generalize (1) to include dichroic media characterized by tensors $\hat{\epsilon}$ and $\hat{\sigma}$ of the form

$$\varepsilon=\begin{pmatrix} \varepsilon_x & i\gamma & 0 \\ -i\gamma & \varepsilon_y & 0 \\ 0 & 0 & \varepsilon_z \end{pmatrix}, \quad \sigma=\begin{pmatrix} \sigma_x & i\sigma & 0 \\ -i\sigma & \sigma_y & 0 \\ 0 & 0 & \sigma_z \end{pmatrix}$$

In this case we have for the electric vector in an incident wave parallel to the x axis

$$\operatorname{tg} 2\chi=\frac{I_+-I_-}{2I_0}=2(A\sin\varphi-B\cos\varphi)e^{-2\alpha d}+2B. \qquad (2)$$

and for an electric vector parallel to the y axis

$$\operatorname{tg} 2\chi=\frac{I_+-I_-}{2I_0}=-2(A\sin\varphi+B\cos\varphi)e^{2\alpha d}+2B. \qquad (3)$$

Here

$$A=\frac{-\gamma(\varepsilon_x-\varepsilon_y)-(4\pi/\omega)^2\sigma(\sigma_x-\sigma_y)}{(\varepsilon_x-\varepsilon_y)^2+(4\pi/\omega)^2(\sigma_x-\sigma_y)^2},$$
$$B=\frac{-\gamma 4\pi\omega^{-1}(\sigma_x-\sigma_y)+4\pi\omega^{-1}\sigma(\varepsilon_x-\varepsilon_y)}{(\varepsilon_x-\varepsilon_y)^2+(4\pi/\omega)^2(\sigma_x-\sigma_y)^2}, \qquad (4)$$
$$\alpha=\frac{1}{4c}\frac{1}{\varepsilon_0^{1/2}}(\sigma_x-\sigma_y), \quad \varepsilon_0=\frac{\varepsilon_x+\varepsilon_y}{2},$$
$$\varphi=\frac{\omega}{2c}\frac{1}{\varepsilon_0^{1/2}}(\varepsilon_y-\varepsilon_x).$$

Expressions (2) and (3) go over into (1) if we put $\sigma_x = \sigma_y = \sigma = 0$. These expressions were obtained for the case $\epsilon_x - \epsilon_y \gg \gamma$ and $\sigma_x - \sigma_y \gg \sigma$. Relation (2) can also be obtained from the more general expression of Donovan and Webster[10] to the line for tan 2χ. Both experimental curves of Fig. 3 for polarization of the incident light along the x and y axis are described by a single expression of the type (2). The question of the sign of the terms containing B in relation (3) therefore remains unclear.

Expressions (2)–(4) contain four unknown quantities $\epsilon_x - \epsilon_y$, $\sigma_x - \sigma_y$, γ, and σ; to determine them it suffices to obtain from the experimental curves the values of B, φ, α, and $A^2 + B^2$. From the shifts of the elliptic-birefringence curve relative to the abscissa axis, it is possible to determine $4B = (1.5 \pm 0.5) \times 10^{-3}$, and then from the "period" $\Delta n = (5.96 \pm 0.01) \times 10^{-2}$, and finally the quantity $4\pi\omega^{-1}(\sigma_y - \sigma_z) = 6 \times 10^{-3}$. From (4) one obtains the dispersion relation of $\gamma_2(\lambda)$ and $4\pi\sigma/\omega = 10^{-4}$ for the case of a magnetic moment directed along the [100] axis. This last quantity does not contradict Jung's data obtained at a wavelength 0.7 μ[11]. A plot of $\gamma_2(\lambda)$ is shown in Fig. 2 (curve 2). From a comparison of the plots of $\gamma_1(\lambda)$ and $\gamma_2(\lambda)$ we see that in the entire investigated wavelength interval $\gamma_1(\lambda)$ is approximately half as large as $\gamma_1(\lambda)$. The conclusion from this comparison would be ambiguous without determining the contribution of the Tm^{3+} ions to $\gamma(\lambda)$. It was for this purpose that an experiment, analogous to the preceding one was performed at 100°K. At this temperature, the weak ferromagnetic moment of $TmFeO_3$ is doubled in comparison with its value at 290°K.

The dispersion dependence of 4χ in $TmFeO_3$ at 100°K, in a plane perpendicular to the [001] axis, is shown in Fig. 1 (curve 2). From the period of this dependence it is possible to determine $\Delta n = (3.2 \pm 0.1) \times 10^{-2}$. The off-diagonal component of the tensor $\hat{\epsilon} - \gamma_3(\lambda)$ in $TmFeO_3$ at 100°K is shown in Fig. 2. It is seen from this figure that $\gamma_3(\lambda)$ differs little in value from $\gamma_1(\lambda)$. The magnetic moment of $TmFeO_3$ is doubled when the temperature decreases from 290 to 100°K[12]. This leads to a small change of the o-f-diagonal component of the tensor $\gamma_3(\lambda)$ (Fig. 2, curve 3). Thus, the magnetic moment of Tm^{3+} has little effect on the value of $\gamma(\lambda)$. This conclusion agrees with the data of[3] for $ErFeO_3$. It should be noted, however, that Δn does not vary with temperature. At 290°K, the birefringence is $\Delta n = 3.12 \times 10^{-2}$, and at 100°K we have $\Delta n = 3.2 \times 10^{-2}$, as indicated above. Therefore curves 1 and 2 of Fig. 1, which correspond to room temperature and 100°K, are shifted along the abscissa axis. This was not taken into account in[3], where investigations analogous to those described were carried out at the different temperatures at one wavelength, and it was assumed that Δn does not vary with temperature. Thus, the anisotropy of the off-diagonal component of the tensor $\hat{\epsilon}$ of orthoferrites is quite large and should find its explanation within the theory of unusually large linear magnetooptical effects in weak ferromagnets.

The model of "anisotropic quenching of the orbital angular momentum" in the same form as in[1] does not explain the anisotropy of the linear magnetooptical effects. Indeed, in this case the linear magnetooptical effects are proportional to the quantity

$$\cos(\beta-\alpha)\sin\beta \qquad (5)$$

Here α is the small cant angle of the sublattices of the iron and β is the angle between the principal axis of the orbital-angular momentum tensor and the [100] axis. In expression (3) we have $\beta \gg \alpha$. In the case of reorientation, the angle β should not change, and consequently (5) should also remain unchanged.

To explain the experimentally observed strong anisotropy of the linear magnetooptical effects in orthoferrites, it is of interest to take into account the noncubic character of the crystal field acting on the entire octahedral FeO_4 complex. Indeed, in such a complex, transitions take place from molecular orbitals corresponding principally to the orbitals of O^{2-} to orbitals corresponding principally to the orbitals of Fe^{3+}, and this entire complex is in a surrounding-ions field of symmetry certainly lower than octahedral. The level scheme and the selection rules for the transitions should in this case be no longer those for the case of pure octahedral symmetry. It is necessary to analyze the spin-orbit splitting of such levels. It is of interest to determine experimentally the optical and magnetooptical anisotropy of the transitions within the molecular complex FeO_4 in weak ferromagnets.

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