Giant linear magnetoelectric effect in garnet ferrite films

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A large linear magnetoelectric effect in the electric field, which is forbidden in the case of the garnet structure, has been observed. The linearity of this effect suggests that the film structure does not have an inversion center and its large value indicates that the electric field has a strong effect on the magnetic anisotropy, i.e., on the orientation of the magnetization.

The magnetoelectric (ME) effect was initially detected in the antiferromagnet[1,2] Cr_2O_3. Magnetoelectric effects of various types, which differ in the nature of their dependence on the electric field (E) or magnetic field (H), have now been studied in many ferrimagnets and antiferromagnets (see, e.g., Ref. 3). The linear ME effect in the electric field is seen only in those crystals whose magnetic symmetry group does not have a spatial inversion I. In crystals with an inversion center the E dependence of the ME effect is quadratic. A quadratic ME effect is seen, in particular, in cubic rare-earth garnet ferrites which have a crystallographic space group[4] O_h^{10}. In the present letter we report the first observation of an anomalously large, linear ME effect, which is forbidden in bulk crystals, in epitaxial garnet ferrite films.

It is virtually impossible to study the ME effect in thin magnetic films by using conventional inductive methods because these methods usually require the substance to be studied to have a large volume, on the order of 1–10 mm^3. The optical polarimetric methods, which can be used on the basis of our estimates to study samples with a volume up to 10^{-6} mm^3, are considerably more capable methods.

TABLE I.

No.	Film composition	Substrate	Film thickness, μm	Faraday effect, deg.	$4\pi M_S$, G	Dev. easy axis magn., deg.
I	$(YBi)_3(FeGa)_5O_{12}$	(111),490 μm	4.7	3.7	~140	~ 0.5
II	$(YBiLaPr)_3(FeGa)_5O_{12}$	(210),605 μm	10.5	15	~100	~16

The experimental setup used for measuring the ME effect was similar to the one described in Ref. 5. We studied the rotation of the polarization plane of light at a wavelength $\lambda = 0.6328$ μm which was induced by applying a static magnetic field to the sample (the Faraday effect) and an alternating electric field E at a frequency of 1400 Hz [electromagneto-optical (EMO) effect]. The sensitivity of the measurements of the variable rotation of the polarization plane was 0.01″ and that of the steady rotation was 30″. The measurements were carried out at a temperature $T = 294$ K. The magnetic field H of up to 10 kOe was applied either in the direction of propagation of light $\mathbf{H}\|\mathbf{k}$ or at an angle γ within the range $60° < \mathbf{Hk} < 120°$. The electric field E of up to 2×10^4 V/cm was applied perpendicular to the sample's surface. We studied films of two compositions: I and II, whose principal characteristics are given in Table I.

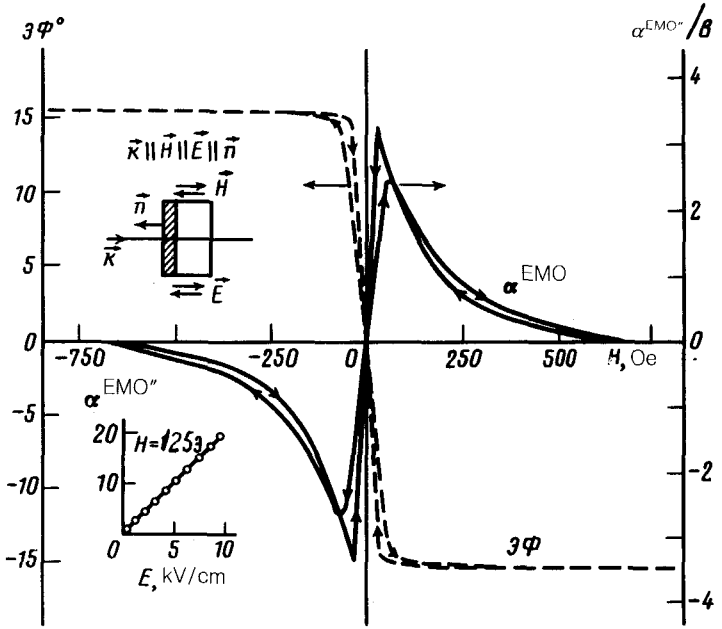

FIG. 1. Field dependence of the EMO effect and of the Faraday effect in film II in a longitudinal geometry. The inset shows a plot of the EMO effect versus the field E at $H = 125$ Oe.

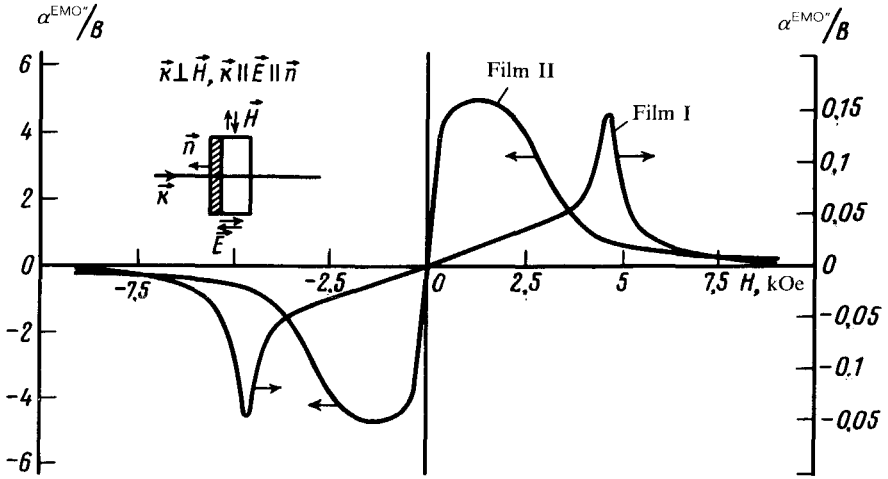

FIG. 2. Field dependence of the EMO effect in films I and II in a transverse geometry.

The first relevant and unexpected result of our experimental studies was the observation in the films of each composition of a linear EMO effect in the electric field, which is forbidden in the garnet ferrite structure. The linear effect was also observed in several other films. Figure 1 shows the results for film II in a longitudinal geometry $\mathbf{H}\|\mathbf{k}\|\mathbf{E}\|\mathbf{n}$ ($\mathbf{n}$ is the vector normal to the surface). Also shown in this figure is a plot of the Faraday effect which characterizes the change in the projection of magnetization onto $\mathbf{k}$ as a result of the application of the magnetic field $\mathbf{H}$. The EMO effect α^{EMO} peaks in the single-domain state ($H \approx 80$ Oe) and vanishes at $H > 750$ Oe, when the directions of $\mathbf{H}$ and $\mathbf{M}$ are the same. The EMO effect was not detected in film I in the given geometry. The field dependence of the linear EMO effect in a transverse geometry $\mathbf{H}\perp\mathbf{k},\mathbf{k}\|\mathbf{E}\|\mathbf{n}$ is shown in Fig. 2. This effect also vanishes when $\mathbf{M}\|\mathbf{H}$, but, in contrast with the preceding case, it is observed in the films of both compositions over broader range of magnetic fields.

The EMO effect depends strongly on the crystal rotation or on the magnetic field relative to the direction of $\mathbf{k}$. Figure 3 is a plot of the EMO effect as a function of the field in the geometry $\mathbf{kn} = 17°$ and $\mathbf{kH} = 111°$. Note the large effect at $H = 2$ kOe: $\alpha^{EMO} \approx 17.5''/V$, a value which is more than 700 times higher than that in Cr_2O_3 (Ref. 5).

The linear dependence of the EMO effect on the field E unambiguously shows that the crystal structure of the films has no inversion center. To the best of our knowledge, this conclusion has not been reached by anyone previously. A lowering of the symmetry in comparison with that of single crystals may stem either from a nonuniform deformation of the film produced by the substrate or from the selective filling of certain sites in the garnet structure by Bi^{3+}, Pr^{3+}, La^{3+} and Ga^{3+} ions.

The absence of the EMO effect in the saturation, when $\mathbf{M}\|\mathbf{H}$, in all geometries of the experiment shows that the electric field does not change the magnetization or the

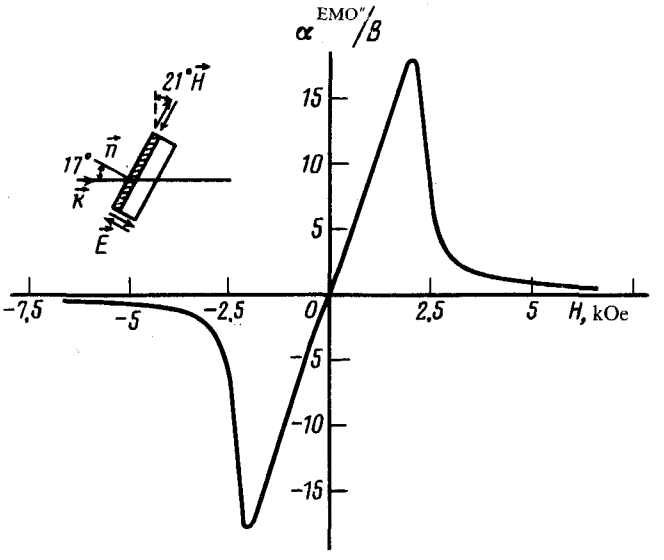

FIG. 3. Field dependence of the EMO effect in film II in the geometry $\mathbf{kn} = 17°$ and $\mathbf{kH} = 111°$.

values of the magneto-optical coefficients. To explain this effect, therefore, it is necessary to take into account the change in the spatial orientation of the vector **M** as a result of the application of the electric field **E**. In the single-domain state, the equilibrium direction of **M** in a film with an orthorhombic magnetic anisotropy is determined by the potential minimum:

$$W = K_u(\mathbf{um})^2 + K_p(\mathbf{pm})^2 - \mathbf{HM} + 2\pi(\mathbf{Mn})^2 \quad , \tag{1}$$

where $\mathbf{m} = \mathbf{M}/M$, K_u, and K_p are the uniaxial and orthorhombic anisotropy constants, and **u** and **p** are the vectors which characterize the directions of the anisotropy axes. The electric field E can manifest itself in terms of the change in the constants K_u and K_p and in terms of the change in the directions of **u** and **p**. Calculations showed that in film I the EMO effect can be explained in terms of the change in the anisotropy field $H_A \approx 4$ kOe by an amount ~ 0.15 Oe and in terms of the rotation of the axis of easy magnetization by $\sim 0.24''$ in a field $E = 1.2 \times 10^4$ V/cm. The field dependences of α^{EMO} in films I and II are different because of the difference in the parameters which characterize their magnetic anisotropy. The presence of α^{EMO} in film I and its absence in film II in a longitudinal geometry is a consequence of the difference in the slopes of the axis of easy magnetization, since in film I in this geometry there is virtually no rotation of **M**. The magnetic fields in which the maximum of the EMO effect is observed in the transverse geometry are different because of the difference in H_A, i.e., because of the different dependences of the angle of rotation of **M** and **H**.

The magnetoelectric coefficient which corresponds to the maximum EMO effect is very large: $\alpha^{\mathrm{ME}} = 10^{-2}$. This value is an order of magnitude higher than that of

Cr_2O_3 and three orders of magnitude higher than the ME coefficient for the quadratic effect which is allowed in garnet ferrite single crystals.[4] At room temperature the ME effect of this magnitude has not, to the best of our knowledge, been observed until now. The large effect is apparently linked with the fact that because of the absence of an inversion center, the electric field in garnet ferrite films can compete effectively with relatively weak relativistic forces ($H_A \sim 10^3$ Oe), rather than with the exchange forces ($H_{exch} \sim 10^6$ Oe), as is the case in Cr_2O_3.

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