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Inhomogeneous magnetoelectric interaction in multiferroics and related new physical effects

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1. Introduction

In this report, we consider a variety of phenomena related to inhomogeneous magnetoelectric interaction. The interconnection between the spatial modulation of the order parameter and electric polarization, known as the flexoelectric effect in liquid crystals, in the case of multiferroics manifests itself in the form of spin modulation induced by electric polarization, and as an inverse effect of formation of spin-induced electric polarization. This *flexomagnetoelectric* interaction also underlies the influence of the ferroelectric domain structure on the antiferromagnetic structure and the magnetoelectric properties of micromagnetic structures. We also consider the influence of an inhomogeneous magnetoelectric effect on the dynamic properties of multiferroics, in particular, on the magnon spectra.

The last decade has been characterized by a spate of interest in media with interrelated magnetic and electrical properties* (see reviews [1–10], and also special issues of two journals devoted to this problem [11]). The interest in this area is expressed, on the one hand, by enhanced attention paid to the fundamental aspects of magnetoelectric phenomena in multiferroics (substances in which magnetic and electric orderings coexist), and, on the other hand, by the expectation of concrete practical applications of magnetoelectrics in spin electronics (in particular, as the base for magnetic-memory devices) and in sensor technology.

The application of multiferroics will make it possible to significantly enlarge the functional possibilities of spintronics: a new degree of freedom — electric polarization — provides the additional means to tune the magnetic and magnetoresistive properties of spintronic elements [12] and to realize four-state logical units [13, 14]. Moreover, the use of magnetoelectric phenomena will make it possible in perspective to avoid using electric currents for magnetic recording [15–18], which is quite timely, since the progressively developing miniaturization of the traditionally used inductive elements is encountering the problem of excessive heat release because of an increase in the current density [19].

An enhanced interest in the fundamental mechanisms of interaction between magnetic and electric subsystems in solids has also been observed lately [10, 20–27], and the variety of types of magnetoelectric interactions has grown noticeably. In the classical review [28], which reflected the level of knowledge on multiferroics in the early 1980s, it was assumed that the polarization $\mathbf{P}$ and the magnetizations $\mathbf{M}_s$ of the sublattices (here s is the order number of a magnetic sublattice) were coupled mainly via the interaction of the form

$$F^{\mathrm{ME}} = -\frac{1}{2}\sum_{ss'} \gamma_{ss'}^{ijkl} P^i P^j M_s^k M_{s'}^l ,$$

which did not require any additional conditions except for the very existence of magnetic and electric ordering; recently, however, other interaction modes, introduced in Ref. [28], have come to the foreground, namely, those that are linear in the order parameters. In particular, the association of electric polarization with the presence of spatially modulated spin structures in the substance was established [29–31], and effects odd with respect to field, which make it possible to control the magnetic structure with the aid of an electric field, have also been discovered [32–35]. Whatever the origin of spatial modulation, i.e., whether as a result of competing exchange interactions, as in frustrated multiferroics [29–33], as a result of the magnetoelectric nature, as in bismuth ferrite [1, 36], or as a result of establishing equilibrium micromagnetic configuration [34, 35], in all cases nonzero spatial derivatives of the magnetic order parameter, $\nabla_i M_j$, existed, which created prerequisites for the manifestation of inhomogeneous *magnetoelectric interaction*.

2. Inhomogeneous magnetoelectric (flexomagnetoelectric) interaction

The inhomogeneous magnetoelectric interaction of the $P_i n_j \nabla_k n_l$ type, where $\mathbf{n}$ is the unit vector of the magnetic order parameter (magnetization or the antiferromagnetic vector), was introduced in Refs [37, 38] in connection with the possibility of the appearance, in magnetically ordered crystals, of long-period structures, and with the possibility of the manifestation of the inverse effect of the formation of electric polarization at domain boundaries [39]. The close analogy between the spatially modulated spin structures in ferroelectromagnets and the spatial modulation of the director in nematic liquid crystals has been noted in Ref. [40]: it manifests itself in the similarity of the mathematical expressions for the energy of inhomogeneous magnetoelectric interaction in ferroelectromagnets and the energy of flexoelectric interaction in liquid crystals, where the director $\mathbf{n}$ serves as the order parameter. This gives grounds to call the inhomogeneous magnetoelectric interaction the *flexomagnetoelectric* interaction.

In the isotropic case or in the case of cubic symmetry, the inhomogeneous magnetoelectric interaction, to an accuracy of the total derivative, takes on an elegant form [40]

$$F_{\mathrm{flexo}} = \gamma \mathbf{P}\big(\mathbf{n}\,\mathrm{div}\,\mathbf{n} + [\mathbf{n}\times \mathrm{rot}\,\mathbf{n}]\big) . \qquad (1)$$

* Since the mid-1990s, the number of published works devoted to magnetoelectric materials has increased twofold–threefold every five years, and has reached about 800 publications per year.

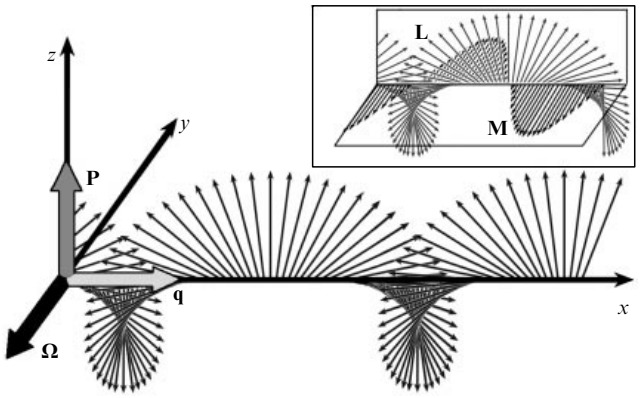

Figure 1. Spin cycloid and three mutually perpendicular vectors: rotation axis **Ω**, direction of modulation **q**, and polar vector **P**. In the inset: spin cycloid $\mathbf{L}(x)$ in the vertical plane is accompanied in bismuth ferrite by a wave of magnetization $\mathbf{M}(x)$ in the horizontal plane xy.

A very visual image, which illustrates the interconnection between the polarization **P**, the wave vector **q** of the spatially modulated structure, and the rotation axis **Ω** of spins, is given in Ref. [23]: these vectors form a trio of mutually perpendicular vectors $\mathbf{P} \sim [\mathbf{\Omega} \times \mathbf{q}]$ (Fig. 1).

It follows from this simple rule that in ferroelectrics we can expect the appearance of cycloidal ($\mathbf{q} \perp \mathbf{\Omega}$) rather than helicoidal ($\mathbf{q} \parallel \mathbf{\Omega}$) type spatially modulated structures, while for the domain boundaries to manifest electrical properties, the magnetization in them must rotate in the plane perpendicular to the domain (the Néel type domain walls).

3. Spatially modulated spin structure in bismuth ferrite $BiFeO_3$

One of first multiferroics in which a spatially modulated spin structure was discovered was bismuth ferrite ($BiFeO_3$) compound [41]. Later on, it was established that the structure has a magnetoelectric origin: bismuth ferrite represents an intrinsic ferroelectric, and the spatial modulation of spins in it is stabilized by spontaneous polarization as a result of flexomagnetoelectric interaction (1) [42, 43].

It is interesting that the detection of a spin cycloid and the establishment of its nature made it possible to explain why one more multiferroic property allowable by the magnetic symmetry — weak ferromagnetism — could not long be revealed in bismuth ferrite. The reason was that because of the presence of a spatially modulated structure in bismuth ferrite, the value of the spontaneous magnetic moment, which is proportional to the x component of the antiferromagnetic vector **L**, periodically reversed its sign and was equal to zero, on average, within the period (see inset to Fig. 1). The observation of a weak ferromagnetic moment became possible only upon the suppression of the spin cycloid and transition to a uniform antiferromagnetic state [44].

The transition between the homogeneous and incommensurate phases can be considered as a spontaneous nucleation of domain boundaries whose energy is written as

$$F_{\mathrm{DW}} = 4\sqrt{AK_u} - \pi\gamma P_{\mathrm{s}}\,, \tag{2}$$

where P_{s} is the spontaneous electric polarization, the constants A and K_u determine the energy of the inhomogeneous exchange and the uniaxial magnetic anisotropy, respectively:

$$F_{\mathrm{exch}} = A\sum_{i=x,y,z}(\nabla n_i)^2 = A\left[(\nabla\theta)^2 + \sin^2\theta\,(\nabla\varphi)^2\right], \tag{3}$$

$$F_{\mathrm{an}} = -K_u\cos^2\theta\,, \tag{4}$$

and the polar angle θ is counted from the z-axis oriented along the c-axis of the crystal (see Fig. 1). The second term in the energy (2) of domain boundaries is obtained by the integration of the flexomagnetoelectric interaction (1) over the period of the structure. Thus, the condition for the phase transition, $F_{\mathrm{DW}} = 0$, takes on the form

$$\gamma = \frac{4}{\pi P_{\mathrm{s}}}\sqrt{AK_u}\,. \tag{5}$$

Formula (5) can also be obtained from the condition of the equality of the thermodynamic energies of the homogeneous and incommensurate phases in which the distribution of the magnetic parameter is described by elliptic functions [43, 45].

In the literature, another method of obtaining the conditions for phase transition also occurs, based on the harmonic approximation [36] in which it is considered that the components of the order parameter **n** change according to the law $n_i(x) \sim \sin(qx + \phi_i)$. This approach, though approximate, makes it possible to arrive at numerical estimates with an almost 10% accuracy [45].

Condition (5) can also be used for evaluating the critical magnetic field H_{c} of the induced phase transition. In this case, the uniaxial anisotropy in formula (5) in an external magnetic field H must be replaced by the effective anisotropy $K_{\mathrm{eff}}(H) = K_u - \chi_\perp H^2/2$, where $\chi_\perp$ is the magnetic susceptibility in the direction perpendicular to the antiferromagnetic vector at $H \parallel c$. In the general case of an arbitrary orientation of the magnetic field, the magnitude of H_{c} will depend on its orientation relative to the plane of the cycloid (see Appendix).

The effective magnetic anisotropy can also change under the effect of other factors, for instance, the presence of impurities of rare-earth ions, magnetostrictive contributions caused by epitaxial stresses that arise upon deposition of a substance onto a substrate with a somewhat different lattice parameter, etc. These reasons were used to explain the suppression of the spin cycloid and the appearance of weak ferromagnetism in strong magnetic fields in bismuth ferrite-based compounds doped with rare-earth ions and in thin films [1, 46]. The latter caused a wave of interest in studying bismuth ferrite and made it the most popular subject (among multiferroics) of both fundamental and applied studies (see the reviews [1, 8, 44, 47, 48]).

4. Antiferromagnetic ordering in bismuth ferrite films with a stripe ferroelectric domain structure

As was shown in numerous studies, no spatially modulated spin structure is observed in thin films (with a thickness of less than 500 nm) of bismuth ferrite [46] (see also reviews [5, 8] and the references cited therein). However, this does not mean that the inhomogeneous magnetoelectric interaction does not manifest itself in any way. Even in the homogeneous antiferromagnetic state a ferroelectric domain structure can be observed [49–51], which, as shown in Ref. [52], can modulate the antiferromagnetic vector by means of flexomagnetoelectric interaction. The inhomogeneous magnetoelectric interaction (1) manifests itself in a jump of the derivative $\nabla\theta$ at the boundary between the ferroelectric

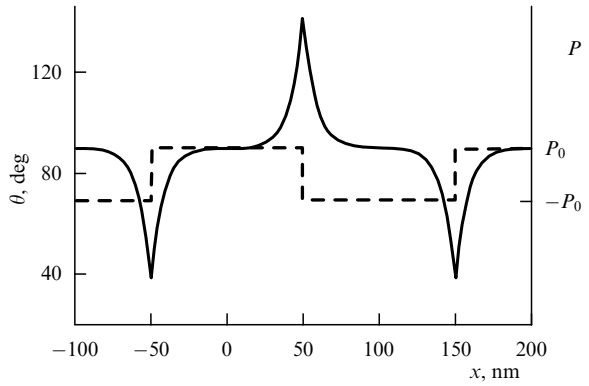

Figure 2. Modulation of the direction of the antiferromagnetic vector (solid curve, left-hand scale) under the effect of a ferroelectric domain structure (dashed curve, right-hand scale) in multiferroics with a flexomagnetoelectric interaction [52].

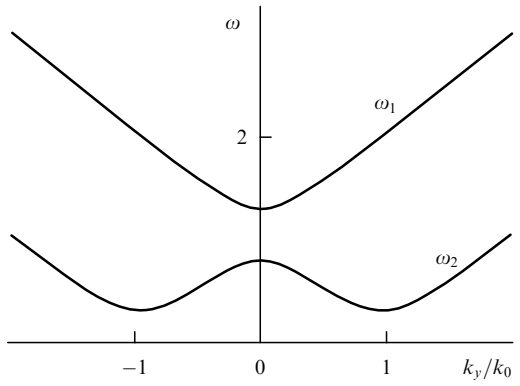

Figure 3. The appearance of a minimum in the dispersion relation for the low-frequency branch of a homogeneous antiferromagnet with a flexomagnetoelectric interaction (k_0 is the magnitude of the cycloid wave vector in the spatially modulated state [61]).

domains with the electric polarizations directed upward (+) and downward (−):

$$A(\nabla\theta)\Big|_{-}^{+} = \gamma P\Big|_{-}^{+} = 2\gamma P_{\rm s}\,. \qquad (6)$$

A graphic representation of the modulation of the direction of the antiferromagnetic vector, assigned by the angle θ, is displayed in Fig. 2.

Thus, in the presence of a ferroelectric domain structure the homogeneous antiferromagnetic state is not the ground one. If in the material there coexist antiferromagnetic and ferroelectric domain structures, then the above effect can manifest itself in the form of the pinning of the antiferromagnetic domain structure to the ferroelectric domain boundaries, which was indeed examined experimentally in manganites [53].

5. Influence of a flexomagnetoelectric interaction on the magnon spectrum in multiferroics

The presence, albeit in a latent form, of a flexomagnetoelectric interaction in the homogeneous antiferromagnetic state shows itself not only in static structures, but also in the dynamic properties of antiferromagnets, namely, in the spectra of magnon excitations which in multiferroics acquire a new quality — the property of *electromagnons* [54]. The influence of a flexomagnetoelectric interaction on the magnon spectra in multiferroics with modulated structures was considered in Refs [55–58].

In films, spatially modulated structures are absent and, therefore, the magnetoelectric interaction (1) does not seem to be urgent for them. It is precisely on this assumption that the problem of the magnon spectrum in bismuth ferrite films was solved in the recent work [59]. However, as was shown in Refs [60, 61], flexomagnetoelectric interaction (1) exerts a substantial effect on the spectrum and properties of magnons in a homogeneous state as well: a minimum appears in their dispersion relation at finite wave vectors for the waves propagating in the direction perpendicular to the electric polarization vector in the case of multiferroics [61] or in the direction perpendicular to the external electric field in the case of centrally symmetric ferromagnets [60]. Thus, the inhomogeneous magnetoelectric interaction in thin bismuth ferrite films causes the mutual interaction of two branches of spin waves propagated along the weak ferromagnetic moment $\mathbf{k} \parallel \mathbf{M}_0$ and leads to the appearance of a minimum in the low-frequency branch at the wave vector equal to the wave vector $\mathbf{k}_0$ of the cycloid in a bulk bismuth ferrite sample (Fig. 3), which reflects the development of instability in the system relative to the transition to the inhomogeneous state with increasing flexomagnetoelectric interaction. Furthermore, the presence in the substance of a homogeneous magnetoelectric interaction of the form $\alpha E_i H_j$, together with a flexomagnetoelectric interaction (1), leads to the nonequivalence (nonreciprocity) of the propagation of spin waves along and against the antiferromagnetic vector [61]. The above features can manifest themselves in light scattering (Raman and Mandelstam–Brillouin) and in neutron scattering, which can be employed for determining the parameters of magnetoelectric interactions.

Of no less interest is the practical aspect of the matter, since the magnons in multiferroics can be not only excited but also controlled by an electric field. Recently, interesting ideas have appeared concerning the creation of subminiature logic units based on magnons. The point is that in the semiconductor technology of logic elements, in the transition to nanosizes the problem arises of very strong electric fields (close to threshold) necessary for their functioning. On the other hand, modern alternative spintronic devices based on the effects of giant magnetoresistance and spin moment transfer require high current densities ($\sim 10^6$ A cm^{-2} and greater). In Refs [62, 63] it was suggested that the problem of nanominiaturization can be solved by utilizing long-lived magnons in magnetic dielectrics.

6. Surface flexomagnetoelectric effect

The specific conditions that are realized in thin films of bismuth ferrite led to the suppression of the spatially modulated spin structures existing in a bulk; however, a possible and even more natural situation is the reverse one, i.e., when the presence of the inversion center in the symmetry group of a bulk crystal forbids the flexomagnetoelectric interaction, while the conditions for the formation of a spin cycloid on the surface are created. The main distinction between the interfaces, as well as dimension-reduced systems, and bulk materials lies in the fact that the electrons and atoms of the surface are subject to the action of physical fields that are asymmetric with respect to the operation of spatial inversion. This creates prerequisites for the appear-

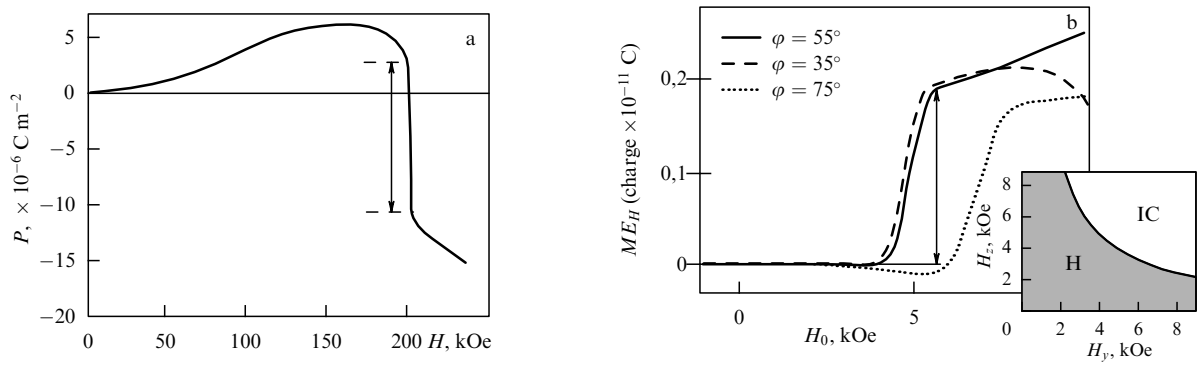

Figure 4. Magnetoelectric anomalies during phase transitions: (a) incommensurate phase–homogeneous state in bismuth ferrite [36], and (b) homogeneous state–incommensurate phase in $BaMnF_4$ [68] (in the inset to the figure, the phase diagram in the $H_y - H_z$ coordinates is given; H, homogeneous antiferromagnetic state, and IC, incommensurate phase).

ance of a flexomagnetoelectric effect in a thin magnetic film or at the interface between magnetic media.

The possibility of the formation of spatially modulated spin structures on the surface and inside thin films of magnetic materials was predicted in Ref. [64], where the condition for the occurrence of a phase transition between the homogeneous and inhomogeneous phases, which is analogous to condition (5), has also been formulated. The same formula is also valid for the boundary between the phases of a liquid crystal, with the difference being that the role of the anisotropy energy with a constant K is played there by the anchoring energy of the liquid-crystal molecules with the substrate, and the role of the exchange stiffness constant A is played by the constant of elastic interaction.

The effect of the formation of spatially modulated spin structures on the surface and inside thin films of magnetic materials was confirmed experimentally in the case of manganese monolayers [65] and in two iron monolayers epitaxially grown on a tungsten substrate with a crystallographic orientation (110) [66]. Using spin-polarized scanning tunneling microscopy, the authors of Ref. [65] observed magnetic modulation with a period of approximately 0.5 nm. The use of probes with a different orientation of the magnetic moment made it possible to establish that this structure in the magnetic monolayer corresponds to a spin cycloid [65], and in the double iron layer to Néel type domain boundaries with a definite chirality [66].

7. Flexomagnetoelectric effect as the origin of improper polarization in multiferroics

As is shown in Section 2, the flexomagnetoelectric interaction (1) in a substance with a broken spatial inversion leads to the formation of modulated spin structures, but the inverse effect is also possible: a spatial spin modulation can result in the elimination of the center of symmetry from the symmetry elements of the crystal and to the appearance of electric polarization.

It is precisely this mechanism that, it is assumed, causes the appearance of electric polarization in orthorhombic manganites $RMnO_3$ (R = Tb [29, 33], Dy [9, 32], Gd [30]), in vanadates $Ni_3V_2O_8$ [67], and in hexaferrites $Ba_2Mg_2Fe_{12}O_{22}$ [31] and explains the magnetoelectric effects observed in these substances, such as the control of the electric polarization by a magnetic field [29, 30] and the inverse effect of the transformation of spatially modulated structures under the action of an electric field [32, 33].

Indeed, as follows from Fig. 1, the reverse of the electric polarization must lead to a change in the relative alignment of the rotation axis and the direction of modulation, i.e., to a change in the chirality of the spiral, which was indeed demonstrated in Refs [32, 33].

In the case of ferroelectrics, such as $BiFeO_3$ and $BaMnF_4$, in which the electric polarization is already present, the spatial spin modulation generates an additional polarization which can be revealed only under special conditions — during phase transitions between the state with spatially modulated spin structures and the homogeneous antiferromagnetic state, i.e., when condition (5) is satisfied.

The flexoelectric polarization emerging at such phase transitions can be determined from the contribution to the thermodynamic potential:

$$\Delta P = -\frac{\partial F_{\text{flexo}}}{\partial E} = \gamma\kappa \frac{\mathrm{d}\theta}{\mathrm{d}x}\,, \tag{7}$$

where κ is the electric susceptibility of the material, $P = \kappa E$, and E is the electric field strength. By averaging over the period of the cycloid, we obtain

$$\langle \Delta P \rangle = \frac{1}{\lambda}\int_0^{2\pi} \Delta P(x) \frac{\mathrm{d}x}{\mathrm{d}\theta}\, \mathrm{d}\theta = \frac{2\pi}{\lambda}\gamma\kappa\,, \tag{8}$$

where λ is the cycloid period.

Figure 4 displays magnetoelectric anomalies manifesting themselves in the form of jumps of the electric polarization in a certain critical field, which can be explained by the phase transitions between the homogeneous and incommensurate phases.

Figure 4a depicts the magnetoelectric dependence for bismuth ferrite. The jump of electric polarization is observed in a critical magnetic field of ~ 200 kOe upon the suppression of the spatially modulated structure. Figure 4b illustrates the different behavior of the magnetoelectric curve for $BaMnF_4$ depending on the angle of inclination of the vector of the magnetic field applied in the plane bc to the b-axis of the crystal, which was observed in the experiments [68], but has not been explained theoretically until now. As we assume, this anomaly reflects the phase transition from the homogeneous antiferromagnetic state to the incommensurate phase upon orientation of the magnetic field at an angle of 45° to the b-axis of the crystal.

Indeed, the $BaMnF_4$ symmetry (class 2, space group $A2_1am$) allows an inhomogeneous magnetoelectric interac-

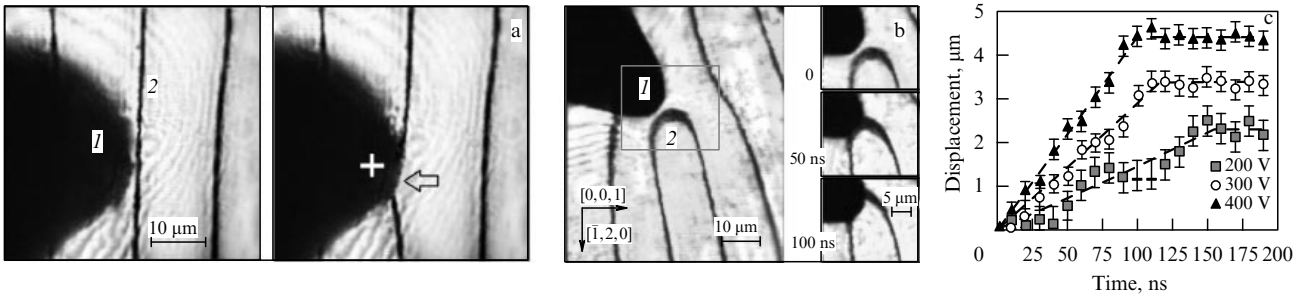

Figure 5. Electric control of the position of a domain boundary in epitaxial iron-garnet films: (a) magneto-optical image of an iron-garnet film in transmitted light (dark lines correspond to boundaries between domains): *1*, electrode; *2*, domain boundary [34]; (b) motion of the head of a magnetic domain caused by an electrostatic action [35], and (c) dependence of the displacement of a domain boundary on time at various potentials at the electrode [35].

tion in the form

$$F_{\text{flexo}} = -(\gamma_{01} + \gamma_{11} H_y H_z)\, P_x \frac{\partial \theta}{\partial x}$$

$$= -\left(\gamma_{01} + \frac{\gamma_{11} H^2}{2} \sin 2\psi\right) P_x \frac{\partial \theta}{\partial x}, \qquad (9)$$

where ψ is the angle made by the magnetic field vector with the *b*-axis of the crystal. This interaction differs from the familiar flexomagnetoelectric interaction (1) in that the coefficient γ in this case can depend on the magnitude and orientation of the external magnetic field. As can be readily seen from expression (9), the magnetoelectric coefficient reaches a maximum value at $\psi = 45^\circ$, and, indeed, the critical magnetic field in this region is a minimum, being approximately 4.5 kOe (Fig. 4b).

It is regrettable that the above-described beautiful effects are observed either at low temperatures, as in $R\text{MnO}_3$ and BaMnF_4, or in high magnetic fields, as in BiFeO_3, which hampers their practical implementation.

8. Iron-garnet films and the flexomagnetoelectric effect at room temperature

Against the background of the wave of enthusiasm caused by the creation of new materials on the base of bismuth ferrite, the prospect for the use of other materials which manifest magnetoelectric properties at room temperature, first and foremost of iron-garnet films, proved to be somewhat underestimated. The linear magnetoelectric effect in these materials, according to estimates made on the basis of measurements of the electroinduced Faraday effect, turned out to be greater by an order of magnitude than that in the classical magnetoelectric material Cr_2O_3 [69].

Furthermore, the iron-garnet films are, perhaps, the most convenient object for an analysis of micromagnetic structures by magneto-optical methods, which acquires special importance in studies of the manifestations of flexomagnetoelectric interactions in micromagnetism. Indeed, the spatial modulation of magnetization can be caused not only by the competition of exchange interactions (as in manganites) or by the inhomogeneous magnetoelectric effect (as in bismuth ferrite), but also by micromagnetic factors such as the minimization of the energy connected with the demagnetizing fields. Since a certain electric polarization can also be associated with such a modulation, the possibility appears of controlling micromagnetic structures with the aid of an electric field.

The influence of an electric field on micromagnetic structures was predicted in many works [23, 39, 40, 70–72]. In this respect, spatially modulated structures such as domain boundaries [39, 70, 72], spin cycloids [40], magnetic vortices [23], and vertical Bloch lines [71] were considered, and it was shown that they are associated with specific distributions of electric polarization. In a recent article, I E Dzyaloshinskii [72] predicted that an electric field that exceeds a certain critical value can induce a magnetic inhomogeneity in the form of a domain wall in a uniformly magnetized medium. This phenomenon, undoubtedly, can be of interest as the prototype of a memory unit with electric recording and a magnetic readout. Unfortunately, no estimates of the threshold field at which the origin of this inhomogeneity occurs were given, but it can be expected that its strength must be significantly high.

If not nucleation, then, at least, the displacement of already existing magnetic domain walls under the action of an electric field was revealed in epitaxial iron-garnet films (about 10 μm thick) grown on the gadolinium-gallium-garnet substrates [34, 35]. Figure 5 demonstrates the effect on the magnetic structure caused by an electric field created by an electrode (pointed copper wire) that touches the dielectric surface of the film [34]. The positive (relative to the substrate) potential at the electrode tip causes an attraction of the domain wall to the electrode (Fig. 5a), while the negative potential leads to its repulsion. After the removal of the voltage, the domain boundary, like a bowed string, returns to the initial equilibrium position. However, the changes caused by the electric field by no means always have a reversible nature: if the system changed from an unstable into a more stable configuration, then the domain boundaries were frozen in the new positions.

The basic features of the effect are as follows:

(1) the effect changed sign upon the change in the electric polarity;

(2) the effect was independent of the polarity of the magnetic domain above which the electrode tip was located (T parity);

(3) the determining role in the effect belongs to the crystallographic orientation of the substrate [the effect was observed in films with the substrate orientation of (210) and (110), and was not observed in (111) films].

All these features indicate the flexomagnetoelectric origin of the effect.

Indeed, both the dependence on the electric polarity and the T parity directly follow from formula (1), and the dependence on the orientation of the substrate is connected

with the distinction between micromagnetic configurations in highly symmetric (111) films and the configurations existing in low-symmetry (110) and (210) films. In (111) films, the direction of the anisotropy axis coincides with the normal to the film, the boundaries between the domains are Bloch type domain walls ($\operatorname{div}\mathbf{M}=0$, $\mathbf{M}\times\operatorname{rot}\mathbf{M}=0$, under the assumption that $|\mathbf{M}|=\text{const}$, which is natural for micromagnetism), and no effect is observed in them, while in the (210) and (110) films the domain boundaries have a Néel component as a result of the inclination of the anisotropy axis with respect to the normal, and the effect becomes different from zero.

The possibility of motion of domain boundaries in an inhomogeneous electric field was noted in Ref. [72]; the velocity of the domain boundary must be proportional to the field gradient. As follows from experimental studies of the dynamics of domain boundaries in pulsed electric fields (Fig. 5b, c), this velocity indeed grows with increasing potential of the electrode tip. The resulting deflection of the domain boundary from the position of equilibrium also grows.

By comparing the results of measurements in an electric field with measurements in a magnetic field, it was possible to determine some quantities that characterize the effect; in particular, the voltage of 500 V (which corresponds to a field strength at the tip equal to 1 MV cm^{-1}) produces the same effect as a magnetic field of 50 Oe [35]. Hence, it is also possible to determine the constant of the effect and from it to estimate the threshold field which, according to Ref. [72], can induce a magnetic inhomogeneity: $E_t = 4\sqrt{KA}/\pi\gamma \sim$ 200 MV cm^{-1}. This magnitude of the threshold field is far from that that can be reached under ambient conditions.

However, the problematic character of the initiation of magnetic structures by an electric field does not yet preclude the prospects of creating devices for magnetic recording on the basis of the flexomagnetoelectric effect, since in the new concept of magnetic memory suggested in Ref. [73] the information recording is achieved by shifting the boundaries between magnetic domains rather than by a reversal of the polarity of a memory element. At the characteristic dimensions of a memory unit of about 100 nm and the velocity of displacement of a domain boundary at a level of 100 m s^{-1} (the order of the velocity given in Ref. [35]), the switching time of the element will be 1 ns.

Acknowledgments

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Appendix

Phase transition induced by a magnetic field

Let us find the dependence of the critical field on its orientation with respect to the plane of the cycloid in a harmonic approximation, assuming that the anisotropy does not affect the shape of the cycloid and that the dependence of the polar angle is described by the linear law

$$\theta = qx\,. \tag{A.1}$$

The wave vector q_0 of the modulated structure, which corresponds to the minimum of the cycloid energy, is found by minimizing contributions to the free energy that depend on the spatial derivatives with respect to the wave vector q [42]. Using Eqns (1) and (2), we obtain

$$\frac{\partial(F_{\text{exch}}+F_L)}{\partial q} = \frac{\partial(Aq^2-\gamma P_z q)}{\partial q} = 0\,. \tag{A.2}$$

For q corresponding to the energy minimum, we have

$$q_0 = \frac{\gamma P_z}{2A}\,. \tag{A.3}$$

In an external field $H=(0,H_y,H_z)$, the contributions to the free energy that correspond to magnetic ordering in a spatially modulated state are written out as

$$F_{\text{cycloid}} = Aq_0^2 - \gamma P_z q_0 - K_u\cos^2\theta - \frac{\chi_\perp H_z^2}{2}\sin^2\theta - \frac{\chi_\perp H_y^2}{2}\,, \tag{A.4}$$

where $\chi_\perp$ is the magnetic susceptibility in the direction perpendicular to the antiferromagnetic vector.

Using (A.3) and averaging the energy over the period λ, we arrive at

$$\langle F_{\text{cycloid}}\rangle_\lambda = -Aq_0^2 - \frac{K_u}{2} - \frac{\chi_\perp H_z^2}{4} - \frac{\chi_\perp H_y^2}{2}\,. \tag{A.5}$$

For a homogeneous state in which the antiferromagnetic vector is perpendicular to the magnetic field and the c-axis ($\theta=90^\circ$, $\varphi=0$), the free energy acquires the form

$$F_{\text{hom}} = -m_s H_x - \frac{\chi_\perp H_z^2}{2} - \frac{\chi_\perp H_y^2}{2}\,, \tag{A.6}$$

where m_s is the magnetization due to weak ferromagnetism.

The critical field of the phase transition, H_c, is found from the condition of the equality of (A.5) with (A.6):

$$H_c = \frac{-2m_s\sin\psi + 2\sqrt{m_s^2\sin^2\psi + \chi_\perp\cos^2\psi\,(Aq_0^2+K_u/2)}}{\chi_\perp\cos^2\psi}\,, \tag{A.7}$$

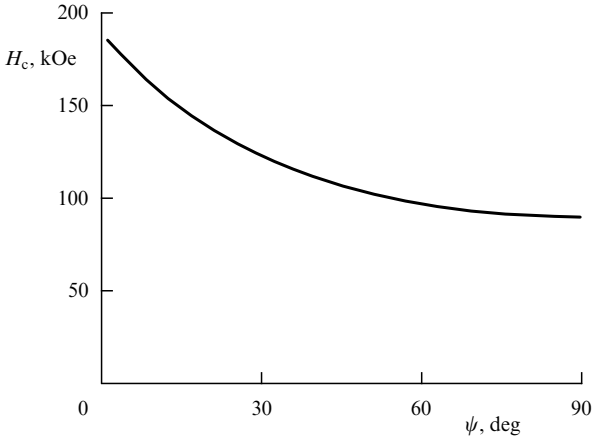

Figure 6. Dependence of the critical field of the phase transition from a spatially modulated state to a homogeneous antiferromagnetic state in bismuth ferrite on the angle of orientation of the field vector with respect to the c-axis.

where ψ assigns the angle of orientation of the field relative to the crystallographic axes: $H_c = (0, H_c \sin\psi, H_c \cos\psi)$.

Figure 6 shows the dependence of the critical field on the angle for the following parameters of the material: $A = 3\times10^{-7}$ erg cm^{-1}, $q_0 = 10^6$ cm^{-1} (corresponding to the cycloid period of 62 nm), $\chi_\perp = 4.7\times10^{-5}$, $m_s \sim 5$ G [45], and $K_u = 3\times10^5$ erg cm^{-3} (including the magnetic anisotropy $K_u^0 = 6\times10^5$ erg cm^{-3} and also the contribution caused by weak ferromagnetism, $K_{DM} = -m_s^2/(2\chi_\perp) \approx -3\times10^5$ erg cm^{-3} [74]). Let us again emphasize that these results were obtained neglecting the deformation of the cycloid shape in an external magnetic field, i.e., on the assumption that the cycloid remains harmonic and is described by dependence (A.1) and by the wave vector q_0 (A.3).

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Terahertz spectroscopy and the magnetoelectric properties of manganite-based multiferroics

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In this report, we present the results of magnetic, magnetoelectric, and terahertz (3–40 cm^{-1}) spectroscopic studies of several manganese multiferroics ($TbMnO_3$, $Eu_{1-x}Y_xMnO_3$, $0 < x \leqslant 0.5$) possessing a spatially modulated spin structure