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**MAGNETISM  
AND FERROELECTRICITY**

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# Formation of Inhomogeneous Magnetic Structures in Weak Ferromagnets with Rhombohedral Symmetry

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**Abstract**—A model of formation of inhomogeneous magnetic structures in weak ferromagnets with rhombohedral symmetry is proposed. This model is based on the general theory of ferromagnetism in these compounds. The quantitative calculations of the dependence of the period of magnetic inhomogeneities on the parameters of the samples are presented and compared with experimental data.

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## 1. INTRODUCTION

A remarkable feature observed in weak ferromagnets with rhombohedral symmetry is that they exhibit fringe structures. These structures were revealed for the first time in iron borate  $\text{FeBO}_3$  at temperatures below 190 K [1]. Fringes have also been observed in hematite in the ferromagnetic phase. The appearance of fringes was initially explained by a specific optical effect, which can manifest itself in such crystals upon transmission of linearly polarized light through magnetic domains separated by an inclined domain wall. However, the proposed model does not allow one to explain all specific features of the observed structures and, as was shown in our previous study [2], these fringes are either magnetic structures with an inhomogeneous magnetization distribution or inhomogeneous magnetic structures. In this paper, we consider physical factors that can be responsible for the formation of inhomogeneous magnetic structures in weak ferromagnets with rhombohedral symmetry.

## 2. OBJECTS OF INVESTIGATION, EXPERIMENTAL RESULTS, AND THEIR THEORETICAL JUSTIFICATION

The objects of our investigation were  $\text{FeBO}_3$  single crystals grown using the gas-transport reaction method. From each group of grown crystals, we selected samples containing no twins or inclusions. The samples had a platelike structure with the (111) basal plane. The domain structures were visualized on a magneto-optical setup based on the Faraday effect.

The observation of the variations in inhomogeneous magnetic structures revealed the following facts.

(i) The period of inhomogeneous magnetic structures  $\lambda$  is independent of the geometric sizes of the sample in the directions parallel to its basal plane, and the value of  $\lambda$  increases with increasing thickness of the sample. As was shown earlier in [2], this behavior of the inhomogeneous magnetic structures is caused by the deviation of the ferromagnetic vector  $\mathbf{m}$  from the basal plane. In turn, this leads to an increase in the magneto-static energy, which decreases upon the formation of an inhomogeneous magnetic structure.

(ii) Fringes in an inhomogeneous magnetic structure are formed only along the three directions coinciding with the twofold symmetry axes parallel to the (111) plane. The proper choice of one system of fringes can be made by applying a magnetic field in the basal plane in the direction parallel to one of the three symmetry planes. The basal plane and the symmetry plane in the crystals under investigation are mutually perpendicular.

The appearance of the fringes can be explained in terms of crystalline anisotropy inherent in weak ferromagnets with rhombohedral symmetry. In these ferromagnets, according to the Dzyaloshinskii theory [3], three magnetic states can be distinguished. In state *I*, the ferromagnetic moment is absent. The material is an antiferromagnet with the antiferromagnetic vector  $\mathbf{I} = \mathbf{M}_1 - \mathbf{M}_2$ , where  $\mathbf{M}_1$  and  $\mathbf{M}_2$  are the magnetizations of the sublattices in the direction perpendicular to the (111) basal plane or along the  $C_3$  axis. In the weakly ferromagnetic phase, there exist two characteristic magnetic states, namely, states *II* and *III*, with due regard for the contributions made to the anisotropy by the fourth-order interactions, which depend on the orientation of the antiferromagnetic vector  $\mathbf{I}$  with respect to the preferred directions in the basal plane. The rhombohedral crystals in the basal plane have the three two-

fold symmetry axes  $L_2$  and three symmetry planes  $\sigma$  perpendicular to this plane. In state *II* (Fig. 1a), the antiferromagnetic vector  $\mathbf{I}$  lies in the symmetry plane and is inclined with respect to the basal plane by the angle  $\delta$ , whereas the vector  $\mathbf{m}$  lies in the (111) plane and is directed along the twofold axis.

In state *III* (Fig. 1b), which appears when the antiferromagnetic vector  $\mathbf{I}$  is directed parallel to the twofold axis, the vector  $\mathbf{m}$  should lie in the symmetry plane and deviate from the basal plane by an angle  $\gamma$ . Therefore, the direction of the ferromagnetic vector with respect to the basal plane can vary depending on the orientation of the antiferromagnetic vector  $\mathbf{I}$  with respect to the twofold axes. According to [4], these variations in the orientation of the vector  $\mathbf{m}$  can occur upon application of a magnetic field  $h$  rotating in the (111) plane. When the magnetic field  $h$  is directed at the angle  $\varphi$  with respect to the twofold axis  $L_2$ , the angle of inclination of the vector  $\mathbf{m}$  to the basal plane should vary according to the law  $\gamma = \gamma_0 \sin 3\varphi$ . The maximum deviation of the ferromagnetic moment from the basal plane should be observed when the magnetic field is aligned parallel to one of the three symmetry planes.

A comparison of the experimental results with theory demonstrates that inhomogeneous magnetic structures are formed in state *III*, in which the antiferromagnetic vector is aligned parallel to the twofold axis  $L_2$  and the ferromagnetic vector lies in the symmetry plane  $\sigma$  at the angle  $\gamma$  with respect to the basal plane. During the formation of an inhomogeneous magnetic structure, the value of  $\gamma$  periodically changes as is shown in Fig. 2. Therefore, the formation of inhomogeneous magnetic structures in the  $\text{FeBO}_3$  compound can be caused by the crystalline magnetic anisotropy inherent in weak ferromagnets with rhombohedral symmetry.

In order to determine the equilibrium period of the inhomogeneous magnetic structure, we write the free magnetic energy  $E$  of the crystal in state *III*. In the absence of an external magnetic field, the free magnetic energy can be represented in the following form:

$$E = E_a + E_{ms} + E_{ex}, \quad (1)$$

where  $E_a$  is the effective energy of the crystalline magnetic anisotropy,  $E_{ms}$  is the energy of the demagnetizing field, and  $E_{ex}$  is the energy of inhomogeneous exchange.

In writing formula (1), it was assumed that, upon the formation of the inhomogeneous magnetic structure, the magnitude and direction of the antiferromagnetic vector remain unchanged. These assumptions are based on the fact that, at any point of the inhomogeneous magnetic structure, the magnitude of the ferromagnetic vector does not change; therefore, the magnitude of the antiferromagnetic vector  $\mathbf{I}$  remains constant. Since the inhomogeneous magnetic structure is formed in state *III*, the antiferromagnetic vector in this structure should be oriented along the twofold axis. In this case, the field

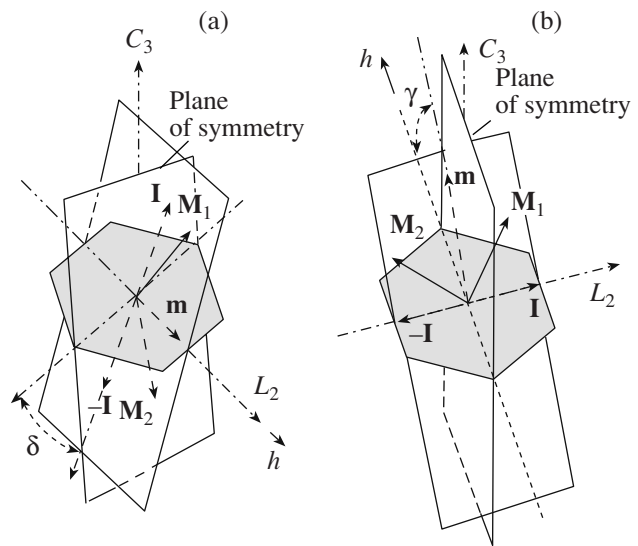


Fig. 1. Possible states of magnetic moments in weak ferromagnets with rhombohedral symmetry according to the Dzyaloshinskii theory [3].

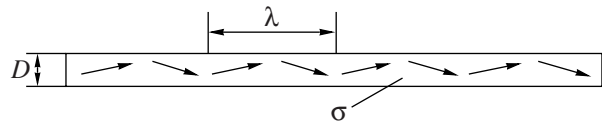


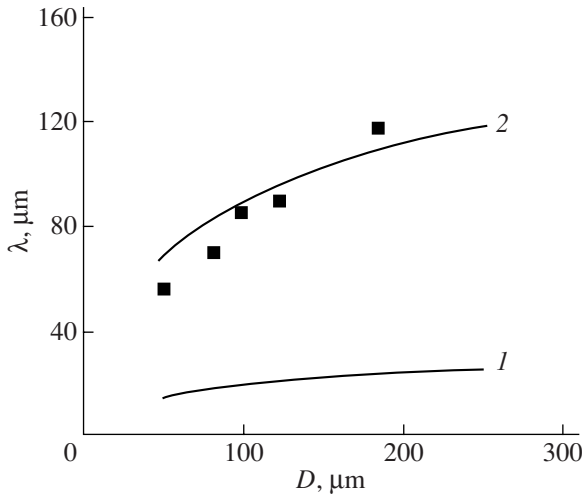
Fig. 2. Spatial variations in the orientations of the ferromagnetic vector in state *III*.

of the uniaxial anisotropy, owing to which the antiferromagnetic vector  $\mathbf{I}$  becomes aligned with the basal plane, for iron borate in the temperature range under investigation is equal to  $3 \times 10^3$  Oe [5], whereas the demagnetizing field  $H_{ms}$  responsible for the appearance of the inhomogeneous magnetic structure can be of the order of  $4\pi m \sin \gamma$ , which does not exceed 10 Oe for the  $\text{FeBO}_3$  compound at the temperature  $T = 77$  K. Therefore, the demagnetizing field cannot affect the orientation of the antiferromagnetic component of the magnetization of the sublattices.

For simplicity, we assume that the angle of deviation of the ferromagnetic vector from the basal plane  $\gamma$  in the inhomogeneous magnetic structure varies according to the harmonic law

$$\gamma = \gamma_0 \sin\left(2\pi \frac{x}{\lambda}\right), \quad (2)$$

where  $\gamma_0$  is the maximum angle of deviation of the ferromagnetic vector from the basal plane and  $\lambda$  is the period of the inhomogeneous magnetic structure.



**Fig. 3.** Dependence of the period of the inhomogeneous magnetic structure of iron borate on the thickness of the sample according to (1) the calculation from formula (9) and (2) the calculation with the correction to the magneto-static energy. Points are the experimental data.

Consequently, the magnetization component perpendicular to the basal plane in the inhomogeneous magnetic structure has the form

$$m_{\perp} = m \sin \left[ \gamma_0 \sin \left( 2\pi \frac{x}{\lambda} \right) \right]. \quad (3)$$

The maximum angle of deviation  $\gamma_0$  of the ferromagnetic vector from the basal plane for  $\alpha\text{-Fe}_2\text{O}_3$  is equal to  $10^{-3}$  rad. For the  $\text{FeBO}_3$  compound, the value of  $\gamma_0$  is smaller by a factor of 2–3 [6]. Hence, Eq. (3) can be rewritten as

$$m_{\perp} = m\gamma_0 \sin \left( 2\pi \frac{x}{\lambda} \right). \quad (4)$$

The period-averaged exchange energy per unit volume is given by

$$\begin{aligned} \langle E_{\text{ex}} \rangle &= \left\langle A \left( \frac{\partial \gamma}{\partial x} \right)^2 \right\rangle = 4\pi^2 A \gamma_0^2 \left( \frac{1}{\lambda} \right)^2 \langle \sin^2 \gamma \rangle \\ &= 2\pi^2 A \gamma_0^2 \left( \frac{1}{\lambda} \right)^2, \end{aligned} \quad (5)$$

where  $A$  is the inhomogeneous exchange interaction constant.

The energy of anisotropy can be represented in the form

$$E_a = -K_u \cos^2(\theta - \gamma), \quad (6)$$

where  $K_u$  is the effective constant of magnetic anisotropy (the easy magnetic axis is directed at the angle  $\theta$  with respect to the basal plane).

In order to determine the energy of the magneto-static field, we use the expression for this energy in the case of the formation of periodic stripe domain structures in thin ferromagnetic films with “supercritical” thicknesses [7, 8]. The formation of stripe domains in these films is associated with the presence of the magnetization component perpendicular to the plane of the film. By using distribution (2) in the calculations performed in [7] for the energy of the demagnetizing field  $E_{ms}$ , it is easy to show that the expression for the period-averaged energy of the demagnetizing field per volume unit takes the form

$$E_{ms} = \frac{\lambda m^2 \gamma_0^2}{4D} \left[ 1 - \exp \left( -\frac{2\pi D}{\lambda} \right) \right], \quad (7)$$

where  $D$  is the thickness of the sample.

With due regard for relationships (5)–(7), the average value of the total energy can be represented in the form

$$\begin{aligned} \langle E \rangle &= \frac{2\pi^2 A}{\lambda^2} \gamma_0^2 + \langle -K \cos^2(\theta - \gamma) \rangle \\ &+ \frac{m^2 \gamma_0^2 \lambda}{4D} \left[ 1 - \exp \left( -\frac{2\pi D}{\lambda} \right) \right]. \end{aligned} \quad (8)$$

The equilibrium value of the period  $\lambda$  can be found from the condition  $dE/d\lambda = 0$  or from the equation

$$\begin{aligned} -\frac{4\pi^2 A^2}{\lambda^3} + \frac{m^2}{4D} \left[ 1 - \exp \left( -\frac{2\pi D}{\lambda} \right) \right] \\ - \frac{\pi m^2}{2\lambda} \exp \left( -\frac{2\pi D}{\lambda} \right) = 0. \end{aligned} \quad (9)$$

The solution of Eq. (9) with respect to the period of the inhomogeneous magnetic structure  $\lambda$  at different thicknesses  $D$  of the samples is shown in Fig. 3 (curve 1). The constant of inhomogeneous exchange is determined from the expression  $A = kT_C/a$ , where  $k$  is the Boltzmann constant,  $a$  is the lattice constant, and  $T_C$  is the Curie temperature. For iron borate, we have the Curie temperature  $T_C = 348$  K and the lattice constant  $a = 5.0$  Å. Hence, the constant of inhomogeneous exchange is determined to be  $A = 10^{-6}$  erg/cm<sup>2</sup>. The value of  $m$  at a temperature of 77 K for  $\text{FeBO}_3$  is equal to 15–17 G.

The experimental dependence of the period of the inhomogeneous magnetic structure on the thickness  $D$  of the sample is also shown in Fig. 3. The observed difference between the experimental and calculated dependences  $\lambda(D)$  can be explained by the fact that, at small deviations of the ferromagnetic vector from the plane, the spins located in the vicinity of the surface deviate from the easy magnetization direction [8]. Consequently, instead of the open poles near the surface, there appears a spatial distribution of magnetic charges.

This leads to a decrease in the magnetostatic energy by the factor  $\beta$ , which can be estimated from the following expression [8]:

$$\beta = \frac{1}{1 + \mu^*}, \quad (10)$$

where  $\mu^*$  is the magnetic permeability of the ferromagnet due to the rotation of the magnetization vector. Physically, this means that the higher the magnetic permeability, the easier the deviation of the spins from the easy magnetization axis. This correction to the magnetostatic energy was termed the  $\mu$  correction. The value of  $\mu^*$  can be estimated from the calculation of the magnetic permeability  $\chi^*$  caused by the rotation of the magnetization vector; that is,

$$\chi^* = \frac{m^2}{2K_u}, \quad (11)$$

where  $K_u = mH_u$  and  $H_u$  is the effective field of magnetic anisotropy. For the  $\text{FeBO}_3$  compound, the effective field of magnetic anisotropy is estimated as  $H_u \sim 1$  Oe. Hence, we obtain  $\chi^* \sim 10$ . The calculated dependence of the period of the inhomogeneous magnetic structure on the thickness of the sample with the correction to the magnetostatic energy is presented in Fig. 3 (curve 2). The calculated curve is in good agreement with the observed experimental dependence.

It should be noted that, in the described model of the formation of inhomogeneous magnetic structures in crystals with rhombohedral symmetry, it remains unclear why fringes are absent in intermediate directions between the three preferred directions, i.e., in the

directions where the deviation of the ferromagnetic vector from the basal plane is theoretically possible. The elucidation of this phenomenon requires further theoretical and experimental investigations.

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