Physics of the Solid State, Vol. 42, No. 11, 2000, pp. 2097–2102. Translated from Fizika Tverdogo Tela, Vol. 42, No. 11, 2000, pp. 2036–2041.

MAGNETISM AND FERROELECTRICITY

Induced Magnetic Superstructure in the $FeBO_3$: Mg Weak Ferromagnet

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Received February 18, 2000

Abstract—The domain structure of a $FeBO_3$: Mg single crystal was studied with a polarizing microscope. It was found that application of a magnetic field along the hard axis in the basal plane of this weak ferromagnet gives rise, within a certain field-strength interval, to a magnetic superstructure observed against the background of the macrodomain structure of the sample. The magnetic superstructure is visually represented as a quasi-periodic system of bands oriented perpendicular to the applied field, with an alternating magneto-optic image contrast along an axis coinciding with the magnetic-field direction. The absence of sharp changes in the contrast of the magnetic superstructure image along this axis is explained as being due to the smooth variation of the sublattice magnetic-moment azimuth with spatial coordinates. The results obtained are discussed within the parameters of the instability of a uniform magnetic state of a system in the random field induced by a magnetic field. © *2000 MAIK "Nauka/Interperiodica".*

The instability of a uniform magnetic state of a ferromagnet, caused by a weak random field which makes the state with nonuniform magnetization energetically favorable, was first discussed in [1]. That publication stimulated numerous experimental and theoretical investigations aimed at studying the effect of a random field on the magnetic state of a magnet. Those studies stimulated, in particular, the discovery of the existence of a microdomain structure in dilute uniaxial antiferromagnets through the use of neutron-diffraction and magnetic measurements [2, 3]. The ambiguous behavior of such a structure in a magnetic field with respect to the instant of its application (before cooling below the magnetic-transition temperature or after it) indicated that the forming magnetic states are metastable. A microdomain structure was also observed, in addition to the uniaxial, in easy-plane antiferromagnets containing extended or point defects [4]. It was shown that, depending on the correlation length of the random anisotropic fields, there may form both a structure with domains having a uniform antiferromagnetism vector and an amorphous magnetic structure with the antiferromagnetism vector varying continually in direction in the basal plane [4].

A particular place among easy-plane antiferromagnets is occupied by weak ferromagnets; their ferromagnetic moment permits one to control their magnetic structure through the application of a weak magnetic field, which makes it possible, in principle, to follow the effects associated with the presence of a random field (induced, for instance, by the randomness of the exchange or by local variations of the competing anisotropic interactions) fairly easily. To study random-field effects, we have carried out an experimental investigation of the influence of diamagnetic impurity ions on the stability of a uniform magnetic state of a weak ferromagnet in an external magnetic field.

1. SAMPLES AND EXPERIMENTAL TECHNIQUE

The study was performed on an $FeBO_3$ single crystal (space group D_{3d}^6), in which part of the Fe^{3+} ions was replaced by Mg diamagnetic ions. Magnetic inhomogeneities were revealed by magneto-optic visualization of the domain structure of the sample. The choice of the subject for the study and of the experimental technique used therein was motivated primarily by the fact that the behavior of the $FeBO_3$ domain structure had been thoroughly investigated and, therefore, one could carefully analyze the differences in the magnetic response between nominally pure (impurity-free) crystals and crystals diluted by a diamagnetic impurity. In addition to this, the magnetic, optical, and magneto-optic properties of iron borate are well known (see, e.g., [5]). $FeBO_3$ is a two-sublattice weak ferromagnet with a Néel temperature T_N = 348 K, below which a stable domain structure is observed to exist. $FeBO_3$ is practically transparent to visible light for wavelengths $\lambda <$ 500 nm, and its magneto-optic properties are governed primarily by the Faraday effect and magnetic linear dichroism [6].

† Deceased.

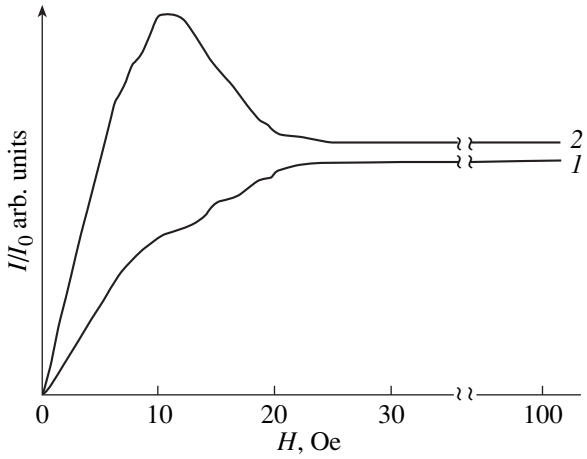

Fig. 1. Field dependences of a magneto-optic signal obtained at $T = 80$ K for different orientations of magnetization. (*1, 2*) Vector **H** is parallel and perpendicular to the domain wall direction.

The charge used to prepare the samples contained magnesium oxide and iron oxide in a weight ratio of ~0.1%. After synthesis, plates ~100 μm thick and ~3 mm wide were cut from the $FeBO_3$: Mg single crystals, such that the principal symmetry axis of the crystal, C_3, coincided with the normal to the sample plane. To relieve the mechanical stress and to make the impurity distribution over the volume more uniform, the samples thus prepared were annealed in air for 10 h at $T = 500$°C. Magnetic measurements showed that the magnesium impurity, compared with pure $FeBO_3$, did not noticeably affect the T_N of the samples.

The domain structure was studied using a polarizing microscope with a camera attachment. The measurements were carried out in transmission at the edge of the transparency window ($\lambda \approx 500$ nm), under normal light incidence, and in the geometry of nearly crossed axes of the polarizer–sample–analyzer system at 80 K (see below). In addition, the magnetic-field and orientation dependences of the intensity variation of light passed through the polarizer–sample–analyzer system, I/I_0 (where I_0 is the light intensity in zero magnetic field H and I is that at $H \neq 0$), were measured. The magnetic field was produced by two pairs of Helmholtz coils. The magnetization system permitted the orientation of the **H** vector along any direction in the basal plane of the sample at $|\mathbf{H}| = \text{const}$. The orientation of the sample, as well as the measurement of the I/I_0 ratio, was executed using the technique described in detail in [6].

2. EXPERIMENTAL RESULTS

An analysis of the field dependence of the magneto-optic signal $I(H)/I_0$, obtained at $T = 80$ K from regions of the sample comparable in size with its transverse dimensions, revealed magnetization curves of two types. The $I(H)/I_0$ dependences of the first kind (curve *1* in Fig. 1) reflect the well-known behavior of magnetic moments under increasing H, namely, an increase in the integrated ferromagnetic, **m**, and antiferromagnetic, **l**, moments through a decrease in their disorder. Curves of this kind were observed under a field application along the boundaries of the domain structure observed in the demagnetized state in the sample (Fig. 2a).

$I(H)/I_0$ dependences of the second type (curve *2* in Fig. 1) were observed under the application of a field perpendicular to the domain boundaries of the demagnetized sample. The pattern of the magneto-optic signal disagreed with the behavior of the moments **m** and **l** in a magnetic field; indeed, as H increases from zero, the I/I_0 ratio grows to a value in excess of its saturated level, to finally reach saturation at $H > 20$ Oe. As proceeds from our results, this type of $I(H)/I_0$ dependence is not observed for $T > 120$ K.

The observed anomalous pattern of the field dependence of the magneto-optic signal can be assigned to the formation, in the course of magnetization, of wedge-shaped magnetic domains, in which the rotation of the plane of polarization increases through the interference of birefringence and the Faraday effect [7]. However, this phenomenon takes place only when the direction of light propagation deviates considerably from the crystal optical axis, which was not the case in our experiment. Furthermore, it was earlier established that the $I(H)/I_0$ dependences of the second type were not observed in an identical geometry on nominally pure $FeBO_3$ crystals.

In order to study the behavior of the magnetic state of a $FeBO_3$: Mg crystal in the course of magnetization, visual representations of the evolution of the domain structure of the sample under study were analyzed. Figure 2a displays a fragment of the surface of a sample maintained at $T = 80$ K in zero magnetic field. The sample is seen to be broken down into domains with distinct boundaries. As field H was applied in the sample plane in the direction perpendicular to the domain walls, the crystal first crossed over to the single-domain structure, after which a quasi-periodic system of alternating bands with fuzzy boundaries and different contrast (Fig. 2b) was observed in fields corresponding to the anomaly in the $I(H)/I_0$ dependence. As the field was increased even more, the modulation of the magneto-optic image contrast on the sample surface disappeared. It was found that the observed band structure formed under a field applied close to the three directions in the crystal basal plane, which, judging from the domain wall orientation in the demagnetized state, are hard-magnetization axes of in-plane crystallographic anisotropy. Figure 3 illustrates the angular pattern of the above structure, relating the applied field strength to the **H** vector azimuth.

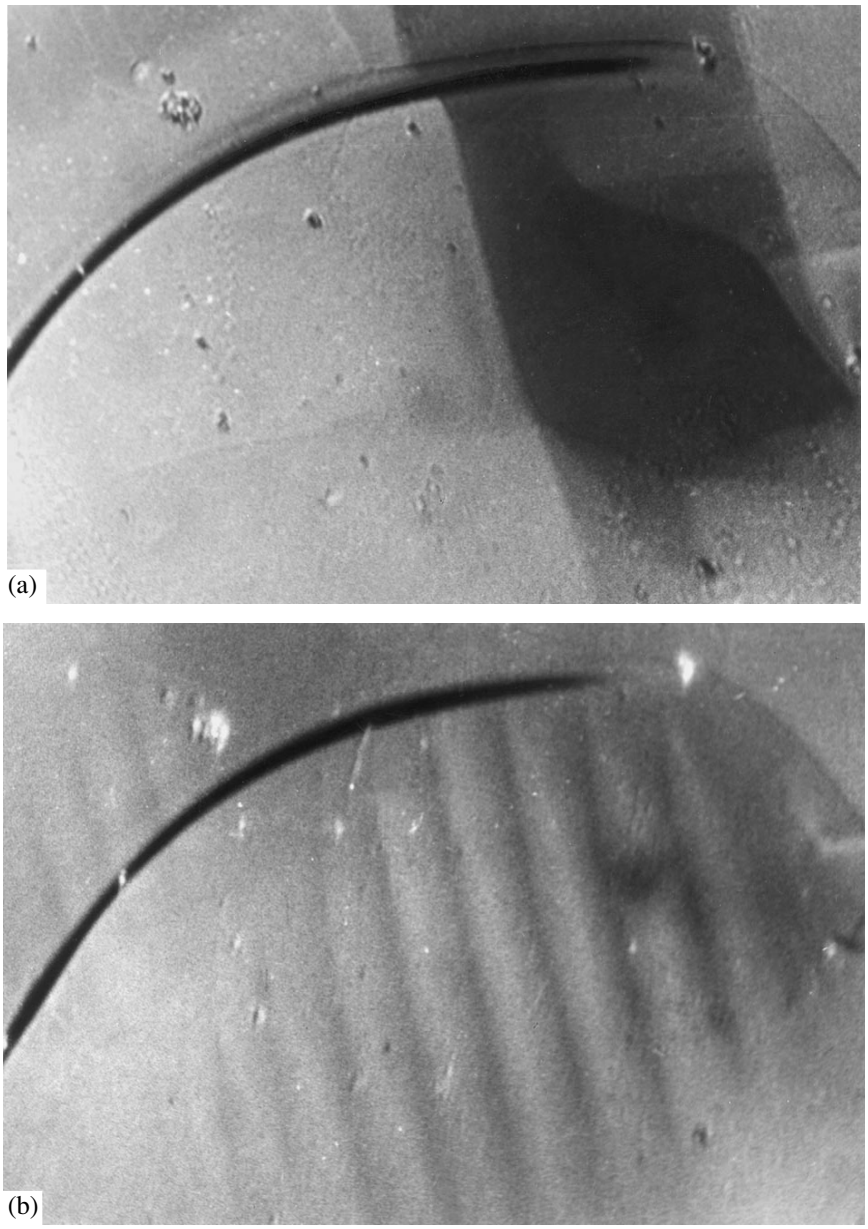

Fig. 2. Sample surface image obtained in polarized light at $T = 80$ K. H (Oe): (a) 0, (b) 12.

Studies showed that the average period of the quasi-periodic band structure with a varying magneto-optic contrast depends on H. Figure 4 presents a typical dependence of the spatial period d of the observed structure on the magnetic field applied along the normal to its wavefront. Of importance is the discontinuous change in period d in a field through a change in the number of bands fitting within the measured length, which is shown in Fig. 4 in the form of steps. In addition to this pinning effect, one observed a hysteresis in the values of d as the field H decreased (i.e., under reverse magnetization); in other words, the average period of the structure is smaller under a decreasing field than under an increasing field.

3. DISCUSSION OF THE RESULTS

One could conceive of three reasons for the formation of the observed system of bands with different magneto-optic contrasts. This structure may actually represent interference bands produced by a system of domain walls inclined to the crystal basal plane, a stripe-domain structure, or again an image of a spatially modulated (incommensurate) structure in the spin system of the crystal.

The first phenomenon is observed at large angles of light incidence on the sample surface [7], a geometry which, as already mentioned, was not used in our experiment. As for the possibility of the existence of a

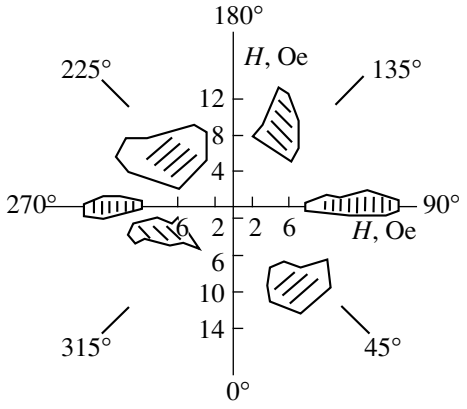

Fig. 3. Diagram illustrating the existence of contrast modulation in the sample surface image in the azimuth–applied-field plane. The hatched areas identify the regions where band structures with different magneto-optic contrast set in, and the orientation of the hatching corresponds to that of the wavefronts of the observed structure.

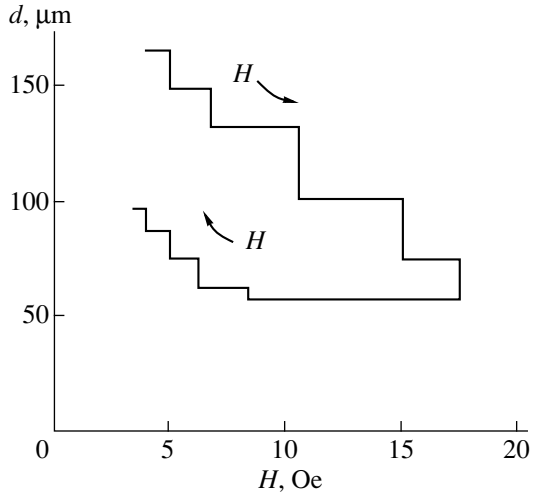

Fig. 4. Field dependence of the spatial period of the quasi-periodic structure of bands with different magneto-optic contrast.

stripe-domain structure, it can hold only if one assumes that impurity states in $FeBO_3$: Mg affect the hexagonal anisotropy constant to the extent that the ferromagnetism vector leaves the basal plane (in these conditions, in order to reduce the magnetostatic energy, the sample must break up into stripe domains). Furthermore, in a doped crystal, the sixth-order anisotropy constant must become noticeable in comparison with the second-order constant, which is unlikely, because the ions introduced into the crystal are diamagnetic. It is, in addition, known [8] that an increase of field H applied in the basal plane of such systems results in an increase in the period of the stripe structure; our experiment, however, revealed the reverse relationship (Fig. 4).

The absence of sharp boundaries in the image of the forming structure suggests that **m**, rather than being an alternating quantity, as is the case with a conventional domain structure, varies only in magnitude. This means that a spatially modulated magnetic state is induced in the $FeBO_3$: Mg, dilute, weak ferromagnet within a certain magnetic-field interval. Therefore, in order to analyze the results obtained, we employ the theory of the transition of a system from a uniform magnetic state to an incommensurate structure (see, e.g., [9, 10]).

It is well known [10, 11] that magnetic anisotropy in the easy plane in rhombohedral antiferromagnets gives rise to the formation of six directions in this plane, along which the uniform sublattice moments are in a stable state. These directions are turned through the angles $\pm\pi/3$ with respect to one another and are crystallographically equivalent. Thus, without any loss of generality, one can assume that the antiferromagnetism vector in an external field H is aligned with an axis close to one of these six directions. Assuming now that the angle β by which vector **l** is canted from the given axis, chosen here as the x axis, is small, the thermodynamic potential of the crystal can be written in a form similar to that obtained in [10]

$$F(\beta) = \int [1/2A\beta^2 + 1/4B\beta^4 + 1/2\alpha(\beta')^2 + 1/4\gamma(\beta'')^2 + mh\beta + 1/2\eta lh(\beta')^2 + \dots]dx, \quad (1)$$

where the single and double primes on β denote the corresponding derivative, and the external magnetic field **H** $\parallel$ **x** is taken into account by adding two symmetry-allowed terms: the first of them, $mh\beta$ ($h = HM$, where M is the sublattice moment), is the Zeeman contribution to the crystal energy, while the second, $1/2\eta lh(\beta')^2$, is invariant under space and time inversion. Inclusion of this term into expansion in Eq. (1) merely renormalizes the coefficient of the first derivative and makes it dependent on the external field H.

Within this model, the transition to a nonuniform magnetic state takes place when the coefficient of the first derivative, $1/2(\alpha + \eta lh)$, is less than zero. In other words, for $\eta < 0$, a modulated state will be induced in the medium under an external field $h > \alpha/\eta l$. In an analysis of the effect of a field H on such a transition, the Zeeman contribution to the thermodynamic potential of the system was first taken into account in [12], where it was shown that a functional similar to Eq. (1) can be minimized using a function of the type

$$\beta(x) = \beta_0 + \xi \exp(ikx) + \text{c.c.},$$

where β_0 and $\xi$are parameters depending, in a complex way, on h and the coefficients A, B, α, γ, and η of the potential in Eq. (1), and the wave vector is defined as [10]

$$k = [|\alpha + \eta lh|/2\gamma]^{1/2}. \quad (2)$$

Expression (2) is a good description, at least qualitatively, of the experimental H dependence of the period of the structure. Indeed, as follows from Eq. (2), the period of the structure $d = 2\pi/k = 2\pi[2\gamma/|\alpha + \eta lh|]^{1/2}$ should decrease with an increasing magnetic field, exactly as was observed in the experiment (see Fig. 4).

Thus, in accordance with the above model, when a magnetic field, applied along the anisotropy axis in the basal plane of a $FeBO_3$: Mg crystal, reaches the critical level $h = \alpha/\eta l$, it induces a phase transition from a uniform to a modulated magnetic state. The axis along which the state is modulated is oriented along the **H** vector, and the magnetic superstructure can be conceived of as a ripple-on phase, where the azimuth of the local antiferromagnetism vector undergoes oscillations relative to a constant deviation angle from the anisotropy axis. The above assumptions suggest that there should be three directions along which the magnetic state of a crystal can be modulated, which is in accordance with the diagram in Fig. 3.

Note that the one-dimensional spatial orientation of vector **l** is an oversimplified case, and that, in actual fact, there is probably also modulation along the axis perpendicular to the basal plane of the crystal. However, because of the small thickness of the sample studied and the small amplitude of the oscillations, this relation will manifest itself in the form of an effective decrease in the antiferromagnetism vector compared with its magnitude in the uniform state.

Now consider the physical meaning of the gradient terms in potential Eq. (1). The introduction of Mg ions into the iron borate lattice gives rise to distortions associated with the difference between the ionic radii of Fe and Mg (and, possibly, with the difference between their charge states). This may produce a random anisotropy, which will induce local canting of vector **l**(**m**) from the directions determined by the crystallographic anisotropy. On the whole, the equilibrium magnetic structure of the crystal will be determined by the competition between the random anisotropy, on the one hand, and the crystallographic anisotropy and the dc magnetic field, on the other. A similar situation was considered for thin polycrystalline magnetic films [13], where the role of the random anisotropy was played by the crystallographic anisotropy in crystallites whose axes were randomly oriented with respect to one another. The factors governing the orientation were the induced anisotropy and the external field H. Assuming these interactions to be also essential for $FeBO_3$: Mg and using the results obtained in [13], one can recast the expression for the modulation period in the form

$$d = 2\pi[2J/(h - K)]^{1/2},$$

where J is the exchange constant and K is the crystallographic-anisotropy energy density. A similar relation can be obtained from Eq. (2) by setting $J = \gamma/\eta l$ and $K = \alpha/\eta l$. We readily see that the coefficients of expansion in Eq. (1) do not have a simple physical meaning and that they represent, rather, some combinations of the exchange and anisotropy constants.

In conclusion, we turn back to the question of the reason for the anomalous field dependence of the I/I_0 ratio (Fig. 1). Using Jones' matrix obtained for rhombohedral weak ferromagnets in [6], one can write a column matrix describing the polarization state of the light at the exit from the crystal. On multiplying it by a conjugate row matrix and making fairly cumbersome manipulations, we obtain the following relation (to within terms linear in magneto-optic coefficients) for the relative intensity of the light passing through a polarizer–sample–analyzer system:

$$I/I_0 = 1 + Q(\delta)\sin\varphi + R\cos 3(\varphi + \varphi_1) + S(\vartheta, \Psi)\sin 2(\varphi + \varphi_2), \quad (3)$$

where ϑ and Ψ are the azimuths of the polarizer and analyzer, respectively, reckoned from the direction of the C_2 axis in the basal plane of the crystal; φ is the azimuth of the ferromagnetism vector relative to the same axis; δ is the angle by which the direction of light propagation deviates from the optical axis; φ_1 and φ_2 are constants; $Q(\delta)$ and R are magneto-optic coefficients, which determine the Faraday rotation of the plane of light polarization induced by the components of vector **m**, transverse and longitudinal with respect to the light propagation direction; and $S(\vartheta, \Psi)$ is the magneto-optic coefficient accounting for the contribution of the magnetic linear dichroism to the magneto-optic rotation. When the direction of light propagation exactly coincides with the optical axis of the crystal ($Q = 0$ for $\delta = 0$), the magnitude of the magneto-optic signal is determined only by the last two terms in Eq. (3).

As is evident from the structure of the above relation, in the case of the field oriented along the hard-magnetization axis, the anomaly in the $I(H)/I_0$ dependence may be due to the fact that the vector **m** in the modulated state cants away from the direction of the applied field, which should increase the contribution of the magnetic linear dichroism to the magneto-optic signal. In saturating fields, vector **m** aligns with the field ($\varphi = \pi/2$), and the contribution due to the dichroism term in Eq. (3) decreases.

ACKNOWLEDGMENTS

This study was supported in part by INTAS, grant no. 97-0366.

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Translated by G. Skrebtsov