Technical Physics, Vol. 46, No. 3, 2001, pp. 354–356. Translated from Zhurnal Tekhnicheskoĭ Fiziki, Vol. 71, No. 3, 2001, pp. 84–87.

BRIEF COMMUNICATIONS

The Modulated Magnetic Structure of α-Fe_2O_3 : Ga Weak Ferromagnet

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Received April 17, 2000

Abstract—The domain structure of a diluted weak α-Fe_2O_3 : Ga ferromagnet is considered. Magnetic fields of specific amplitudes and orientations applied in the basal plane of the crystal are found to produce nonuniform magnetic states due to a periodic deviation of the antiferromagnetic vector from the easy axis of crystallographic anisotropy. A phase diagram of the modulated magnetic state with the triad axis is constructed, and a dependence of the spatial period of modulation on an external magnetic field is studied. A phenomenological model and the nature of the magnetic superstructure discovered are discussed. © *2001 MAIK "Nauka/Interperiodica".*

Instability of the uniform magnetic state of magnetically ordered dielectrics is commonly attributed to exchange interactions of different sign between the closest neighboring atoms and those next to them. The arising magnetic structures have been adequately studied both experimentally and theoretically (see, for example, [1]). Under specific conditions, however, the ordered uniform state may become unstable in a weak random field [2]. Among other reasons, the random magnetic field can be produced by local spatial variations in competing anisotropic interactions of a magnetoactive ion with its surroundings. For example, as theoretically shown in [3], in easy-plane antiferromagnets, the uniform antiferromagnetic state can become energy-unfavorable because of 3D or point defects present in the crystal lattice. Nonuniform magnetic states arisen in this case are less understood.

In the recent experimental study [4] of hematite, α-Fe_2O_3, and iron borate, $FeBO_3$, which are easy-plane antiferromagnets (in the weak ferromagnetic phase), a magnetic superstructure was discovered. It appears in a particular range of magnetic fields when a small amount of diamagnetic Ga and Mg ions are incorporated. In order to obtain the parameters of the modulated magnetic states in diluted weak ferromagnets, we pursued the study of the influence of a magnetic field on the magnetic structure of α-Fe_2O_3 : Ga. One reason is that hematite is a promising material for many high-frequency devices. Therefore, mechanisms behind the nonuniformity of the macroscopic magnetic parameters of the material must be carefully investigated.

EXPERIMENTAL

Hematite was synthesized with an addition of ≈0.1 wt % of gallium oxide. About 50-μm-thick plates ~3 × 3 mm in cross section were cut from the synthesized α-Fe_2O_3 : Ga single crystals so that the principle crystallographic axis C_3 was perpendicular to the sample surface. To relieve the mechanical stress and homogenize the impurity distribution, the plates were annealed in air at T = 500°C for 10 h. Magnetic measurements indicated that in the presence of the Ga impurity, the Neel temperature of the samples (T_N = 950 K) was nearly the same as that of pure α-Fe_2O_3, while the Morin temperature decreased below the boiling temperature of nitrogen.

The domain structure of the α-Fe_2O_3 : Ga samples was visualized by a magnetooptic technique. A large body of experimental data for the domain structure of hematite that were obtained by this method [5] enables us to compare in detail the responses to an external magnetic field from pure (impurity-free) crystals and those diluted by a diamagnetic impurity. Moreover, the typical size of magnetic inhomogeneities discovered in [4] is about 50 μm, suggesting that studying the modulated magnetic states requires a considerably high spatial resolution, which is readily provided by the magnetooptic technique.

The domain structure was examined at room temperature in the transmission band of hematite (0.8–1.1 μm) using a crossed analyzer and a polarizing microscope. A light beam was aligned with the C_3 axis. A uniform magnetic field was produced by two pairs of Helmholtz coils. The field lay in the basal plane of the samples and was varied in magnitude and in direction about the magnetic anisotropy axes. The sample was oriented according to the technique described in [5]. The image of the domain structure was read out by a video camera, digitized, and visualized on a computer.

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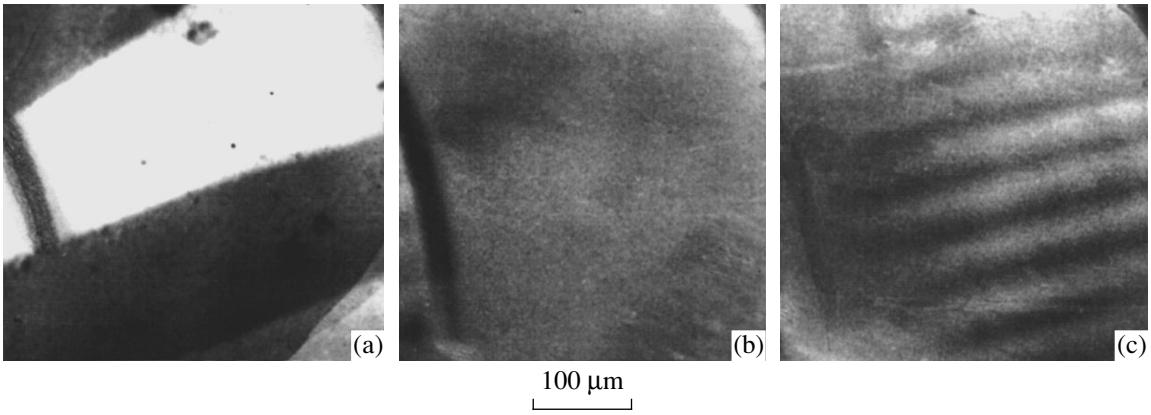

Fig. 1. Magnetooptic images of the domain structure for **H** = (a) 0, (b) 5, and (c) 7 Oe.

EXPERIMENTAL RESULTS

Figure 1 shows the evolution of the domain structure in different magnetic fields H applied perpendicular to the domain walls in the demagnetized state of the crystal. Under the applied field, the crystal first becomes single-domain; then, at $H \gtrsim 5$ Oe, there appears the quasi-periodic structure of blurred fringes with a various magnetooptic contrast. The magnetooptic contrast of the image evened up at $H \gtrsim 20$ Oe.

The fringes appeared when the direction of the applied field was close to any of the three hard magnetic axes in the basal plane (according to [5], the hard magnetic axes are perpendicular to the U_2 axes). The average period λ of the fringe pattern was H-dependent. A typical dependence of λ on the magnetic field applied perpendicular to the wave front of the arising fringes is plotted in Fig. 2. The period λ, i.e., the number of fringes per unit length, varies stepwise. Along with the pinning effect, hysteresis of λ is observed: as H decreases, the average period of the fringe pattern changes less than it does when the field increases.

The experimental magnetic state diagram in Fig. 3 depicts areas where the quasi-periodic structure of the fringes is observed according to the applied field magnitude and direction in the basal plane. In the hatched regions, the magnetooptic contrast of the image is modulated. The direction of the hatches indicates the direction of fringes with a different magnetooptic contrast.

DISCUSSION

The diffuse boundaries of the fringes suggest that the magnetization in the system is not merely alternating, which is typical of a usual domain system, but smoothly changes its direction. Such a fringe pattern was not observed in pure hematite crystals under similar magnetization conditions; therefore, one can conclude that the nonmagnetic impurity states directly affect the rotation of sublattice magnetic moments in α-Fe_2O_3 : Ga. This gives rise to the spatial-modulated ferro- and antiferromagnetic vectors in a particular range of magnetic fields. In other words, in the weak ferromagnet considered, higher-than-saturation magnetic fields cause the orientational uniform-to-modulated magnetic state transition.

To characterize the arising magnetic superstructure, let us invoke the theory of phase transitions from uniform to nonuniform magnetic states in magnetoordered structures [6, 7]. In rhombohedral antiferromagnets, including hematite, a consideration of magnetic anisotropy in the easy plane is known [6] to result in stable uniform sublattice moments in six directions, which differ from each other by an azimuth angle of $\pi/3$. Therefore, without loss in generality, the antiferromagnetic vector **l** can be assumed to be oriented close to one of these directions. In this approximation, the thermodynamic potential of the crystal can include only uniaxial anisotropy. Then, as an order parameter, we choose a small deviation β of the vector **l** from a given direction in an external magnetic field and take this direction as the X axis ($H \parallel X$). Now, following [6, 7], the thermodynamic potential of the crystal can be represented as an

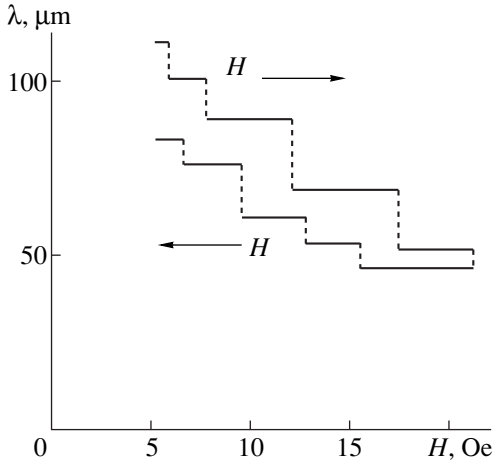

Fig. 2. Field dependence of the spatial period of the quasi-periodical fringes with different magnetooptic contrast.

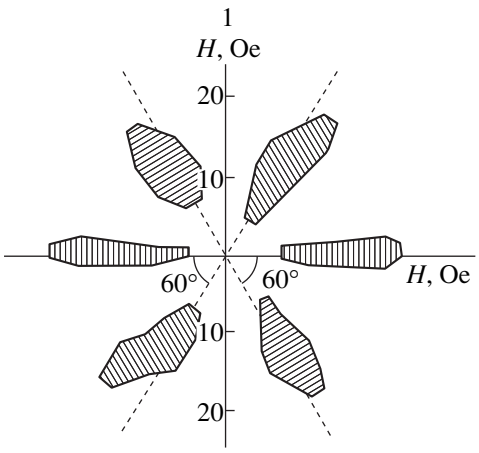

Fig. 3. Magnetic state diagram according to the external magnetic field strength **H** and azimuth for the system of fringes with different magnetooptic contrast. Hatched areas correspond to the parameters for which the modulated magnetic structure is observed. *1*, the easy magnetic axis.

expansion in the order parameter:

$$\Phi(\beta) = \int[-1/2A\beta^2 + 1/4B\beta^4 + 1/2\alpha(\beta')^2 + 1/4\gamma(\beta'')^2 + mh\beta + 1/2\mu lh(\beta')^2 + \ldots]dx, \quad (1)$$

where primes mean differentiation with respect to the corresponding argument. The external magnetic field H is taken into account through two symmetry-allowed terms: one of them, $mh\beta$ (m is the ferromagnetic vector, and $h = MH$, where M is a sublattice moment), is the Zeeman contribution into the crystal energy, and the other, $1/2\mu lh(\beta')^2$, is invariant to space–time inversion.

The functional thus constructed differs from that used in [7] only by the addition of the latter term, which accounts for the presence of the random field and renormalizes the coefficient at the first derivative, making it H-dependent.

The phase transition from the uniform to the modulated magnetic state occurs when the coefficient at the first derivative in (1) is negative; i.e., at $\mu < 0$, a field $h > \alpha/\mu l$ induces the modulated magnetic state of the crystal. In this case, functional (1) is minimized by a function like [7]

$$\beta(x) = \beta_0 + \xi\sum[C_j\exp(ik_jx) + \text{c.c.}]$$

(c.c. is the complex conjugate). Neglecting the relatively weak temperature dependence of the modulated state parameters, we can approximate the spatial distribution of the order parameter by a single harmonic:

$$\beta(x) \approx \beta_0 + \eta\cos k_0x.$$

Thus, within our model, an external magnetic field of critical value $\alpha/\mu l$ lying in the basal plane of a weak ferromagnet along the anisotropy axis initiates the orientational phase transition from the uniform to the modulated magnetic state. Modulation appears in the direction of the magnetic field, and the magnetic superstructure can be represented as a phase where the azimuth of the local antiferromagnetic (ferromagnetic) vector oscillates with a period $\lambda = 2\pi/k_0$, the vector being constantly deflected from the anisotropy axis. In accordance with the previous assumptions, there should be three directions of **l** azimuth modulation. This is consistent with the diagram of modulated magnetic states (Fig. 3).

It can be shown that under near-critical magnetic fields, the parameters of the nonuniform magnetic state are given by

$$k_0^2 = |\alpha + \mu lh|/2\gamma, \quad \beta_0 = 4\gamma h/M(\alpha + \mu lh)^2,$$

$$\eta^2 = 1/3B[A + (\alpha + \mu lh)/4\gamma - 48\gamma^2h^2B/M^2(\alpha + \mu lh)^4].$$

The above expression for the wave vector k_0 of the modulated structure gives at least a qualitative (disregarding pinning and hysteresis effects) explanation for the experimental decrease in the period of the fringes with growing H (Fig. 2).

In conclusion, let us comment on the physical interpretation of the results. Being introduced into the hematite lattice, diamagnetic Ga ions induce distortions due to a difference in the ion radii of impurity and host atoms and, possibly, due to their different charge states. This results in random anisotropy, which causes a local deviation of the ferromagnetic vector from the directions defined by crystallographic anisotropy. For **H** perpendicular to the crystallographic anisotropy axis (as in our experiment), the resulting equilibrium magnetic structure is defined by the interplay between random anisotropy, crystallographic anisotropy, and the constant magnetic field.

ACKNOWLEDGMENTS

This work was supported by INTAS, grant no. 97-0894.

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Translated by A. Sidorova