New spontaneous order-disorder phase transition in magnetic films with a periodic deformation of the domain-wall profile

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A new spontaneous phase transition has been observed in magnetic garnet films with a small uniaxial-anisotropy constant. This transition is accompanied by a disruption of the order in the initially regular stripe domain structure. It is also accompanied by a change in the temperature dependence of the spatial period of this structure.

Among the various magnetic phase transitions, one can single out so-called spontaneous transitions, which are induced by a change in only the temperature T of the magnetic material. Falling in this category are order-disorder phase transitions at the Curie point, structural magnetic phase transitions, and orientation (spin-flip) transitions, among others. Spontaneous phase transitions occur rather frequently in media having a domain structure, which can strongly influence the kinetics of the phase transition and also features observed in various thermodynamic quantities under these conditions. In this paper we are reporting the experimental observation of a spontaneous phase transition associated with a freezing of a surface soft mode in magnetically uniaxial films with a regular stripe domain structure.

Magnetically uniaxial films with a weak perpendicular anisotropy (with a quality factor $q_u = K_u/2\pi M^2 < 1$, where K_u is the uniaxial-anisotropy constant, and M the magnetization) with thicknesses in a certain interval $t_{\rm cr1} < t < t_{\rm cr2}$ have domain structure with a specific distribution of the magnetic moment. This distribution is characterized by a periodic deformation of the profile of the domain walls near the surfaces of the material. Experiments carried out on plates of magnetoplumbite have shown that, as the sample thickness t decreases, this domain structure converts into an ordinary stripe domain structure. The amplitude of the deformation of the domain-wall profile smoothly approaches zero. This behavior corresponds to a second-order phase transition over thickness.

It was shown in Refs. 4 and 5 that spontaneous and orientation second-order phase transitions involve an amorphization of the regular domain walls, because one effective elastic constant (or several of these constants) of the magnetic subsystem vanishes. The corresponding soft modes in the spectrum of elementary excitations of the magnetic material are bulk modes in this case. For the phase transition between nonuniformly magnetized states with an ordinary stripe domain structure and a "modulated" one, discussed in Ref. 3, the soft mode responsible for this transformation should be a surface mode, so it should not lead to a complete amorphization of the domain structure. Nevertheless, the vanishing of one of the effective elastic constants should have an important

effect on the behavior of the magnetic subsystem. For an experimental study of the critical behavior of the magnetic material, however, we would like to realize this phase transition by varying an external parameter (e.g., the temperature T), rather than the film thickness.

A similar phase transition has been observed in a study of the temperature dependence of the period of the domain structure, d, and the period of the modulation of the domain-wall profile, Λ , in iron garnet films with the composition $\operatorname{Lu}_{2.1}\operatorname{Bi}_{0.9}\operatorname{Fe}_{5-x}\operatorname{Mg}_x\operatorname{O}_{12}$ (x=0-0.17) with thicknesses of 5-20 μ m. These films were grown by liquid-phase epitaxy with a $\operatorname{Bi}_2\operatorname{O}_3$ solvent on $\operatorname{Gd}_3\operatorname{Ga}_5\operatorname{O}_{12}$ substrates in the (111) orientation. The films were grown under conditions similar to those used in Ref. 6. The Curie point T_C of the films of the specified composition was ≈ 560 K, the magnetization was $4\pi M \approx 1800$ G, and the effective fields of the cubic and induced uniaxial anisotropy did not exceed 100 Oe.

Curves of d(T) and $\Lambda(T)$ were constructed from diffraction patterns created by a laser beam with a wavelength $\lambda = 0.63~\mu m$. With the transmission planes of the polarizer and the analyzer perpendicular for the regular stripe domain structure, with a periodic distortion of the domain-wall profile in the absence of a magnetizing field, in the case of normal incidence of the light on the sample, we observed a series of diffraction peaks. The angular positions of these peaks are given by

$$\vartheta_{n,m} = \arcsin\frac{\lambda}{d\Lambda} \sqrt{(2n+1)^2 \Lambda^2 + m^2 d^2},\tag{1}$$

$$\varphi_{n,m} = \arctan \frac{md}{(2n+1)\Lambda} , \qquad (2)$$

where n and m are integers, and the angles $\vartheta_{n,m}$ and $\varphi_{n,m}$ are reckoned from respectively the normal to the film surface and the direction perpendicular to the planes of the domain walls. [In the case m=0, expressions (1) and (2) become the known expressions for a structure with unmodulated plane domain walls.] Fig. 1, a and b, shows the typical shape of the domain structure and a diffraction pattern from this structure, respectively, at room temperature for one of the test films (we will refer to this film below as film 1). This film had a thickness of 10 μ m and an Mg²⁺ concentration x=0.025. Fig. 1c is a photograph of a fragment of the original domain structure. This photograph was taken at a large magnification with a small depth of field in order to visualize the sinusoidal distortions of the profile of the domains walls. A graphics analysis of the photographic prints was carried out on a personal computer with the help of standard retouching programs.

The open symbols in Fig. 2 show the temperature dependence of the geometric parameters of the equilibrium domain structure¹⁾ in film 1, found by a diffraction method as the temperature was slowly raised. At $T = T_{\rm cr} \approx 460$ K, there is a peak on the d(T) curves. This temperature corresponds approximately to the upper limit of the region in which a modulation of the domain-wall profile exists. As $T_{\rm cr}$ is crossed, satellite peaks disappear from the diffraction pattern, while the main peaks become blurred and transform into a ring (Fig. 1e). These changes are evidence of a disruption of the long-range orientation order in the domain structure, which is typical of a second-order phase transition.^{4,5} However, direct visual inspection showed that a short-range orientational

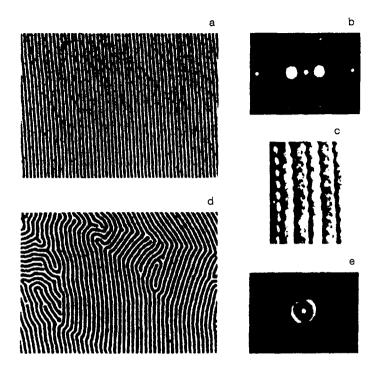


FIG. 1. Typical appearance of the domain structures in film 1 at room temperature, along with corresponding diffraction patterns (b and e).

order persists, and that the amorphization occurs through a breakup of the regular domain structure into many randomly positioned blocks. Within each block, the domain structure remains ordered, except near points belonging to three (or more) blocks. In this case the domain structure has a tendency to transform into a labyrinth (Fig. 1d). In other words, the energy of the fluctuations associated with the soft surface mode is insufficient to form magnetic dislocations and disclinations in the number required for a complete amorphization of the originally ordered domain structure. When these blocks form, the magnetic part U of the free energy F increases because interblock boundaries appear. However, the increase in the entropy S due to the disruption of the order cancels that effect and lowers the total free energy of the system, F = U - TS.

The reason for the existence of the observed phase transition is the temperature dependence of the first critical thickness of the magnetic film, which is described by²

$$t_{\rm cr1}(T) = 0.9\pi^3 K_u(T)^{-3/2} A(T)^{1/2} M(T)^2, \tag{3}$$

where A is the constant of the nonuniform exchange interaction. Since the constant of the induced uniaxial anisotropy, K_u , decreases rapidly with increasing temperature in epitaxial magnetic garnet films far from the Curie point, and since the spontaneous magnetization M (in the absence of a compensation point) and also the exchange constant A are

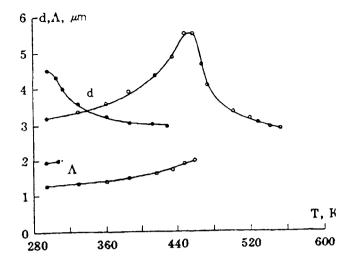


FIG. 2. Temperature dependence of geometric parameters of the domain structures in film 1 (the open circles) and in film 2 (the filled circles).

essentially independent of the temperature (Ref. 7, for example), the value of $T_{\rm cr1}$ increases with increasing temperature and becomes equal to the geometric thickness of the film at $T = T_{\rm cr}$. This hypothesis is supported by the following facts.

- (1) With increasing concentration of Mg^{2+} ions, which tend to increase the uniaxial-anisotropy constant K_u , the transition temperature T_{cr} rises and approaches the Curie point. This behavior is illustrated by the curves of d(T) and $\Lambda(T)$ for film 2 shown in Fig. 2 (the experimental points are shown by the filled symbols). This film has the same thickness as film 1 but a lower Mg^{2+} concentration (x=0.01).
- (2) In films of the same nominal composition (in the starting material), but grown from a $PbO-Bi_2O_3$ solvent, the implantation of Pb^{2+} ions results in a pronounced induced "perpendicular" uniaxial anisotropy (with an effective field ~ 1 kOe). In this case the distortions of the domain-wall profile persist over essentially the entire temperature range in which the magnetically ordered phase exists. The phase transition associated with the disappearance of these distortions occurs in the immediate vicinity of the Curie point.²⁾
- (3) As the thickness of the films of a given composition decreases, the transition temperature falls off monotonically.

The increase in the period of the distortions of the domain-wall profile, Λ , with increasing temperature seen experimentally agrees with the theoretical predictions. It correlates well with the known behavior $\Lambda(d)$ (Ref. 1, for example). There is, on the other hand, an unexpected result: The change in the sign of the derivative of the function d(T) after the modulation of the domain-wall profile disappears. This change in sign may be due to a modification of the type of magnetic-flux closure in the course of the phase transition: At $T < T_{cr}$ the flux closure is two-dimensional, while at $T > T_{cr}$ it is one-dimensional.

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²⁾In films with a very strong induced anisotropy, this phase transition may not occur at all, since near T_C not only the uniaxial-anisotropy constant but also the magnetization and the exchange constant depend strongly on the temperature. Accordingly, the critical thickness of the film is not reached at all [see Eq. (3)].

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¹⁾Approximately equilibrium domain structures were formed at any temperature by "magnetic shaking." For this purpose, we used a coil which set up an alternating magnetic field with a frequency ~ 10 kHz and an amplitude ~ 100 Oe directed parallel to the large plane of the film. In the absence of magnetic shaking, phenomena similar to those described below were observed, but they were not as pronounced [the maximum on the d(T) curve was more rounded, there was a slight amorphization, etc.].

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