Stripe Domain Structures in a Thin Ferromagnetic Film

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We present a theory of stripe domain structures in a thin ferromagnetic film with easy-axis magnetic anisotropy and long-range dipole interactions, for a wide range of temperatures below the reorientational phase transition. Spatial anisotropy generated by the exchange energy within domain walls pins the orientation of the domains. The domain structure can be described as a smectic liquid crystal with positional order destroyed by both thermal meandering of domain walls and by proliferated dislocations. The theory is in good agreement with experiment.

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Recently, the physical properties of thin ferromagnetic films have been the subject of intense investigation, in part due to the prospect of using such films as high-density storage devices. Such ultrathin films have unusual properties. Fe and Co on Au or Cu substrates have Curie temperatures exceeding room temperature. Moreover, the magnetization of Fe/Cu(100) and Fe/Ag(100) films has been found to align perpendicular to the film at low temperatures and parallel to the film at high temperatures [1].

The perpendicular magnetization at low temperatures can be understood in terms of perpendicular anisotropy. Néel [2] pointed out that such anisotropy is generated by boundaries, a conclusion confirmed by recent numerical calculations [3]. On the other hand, for a thin slice of a ferromagnet with parallel boundaries, the dipole interaction favors an orientation of the magnetization parallel to the boundaries, by an amount proportional to the width of the slice. Hence, since the perpendicular anisotropy at the boundary is independent of the thickness, one can expect a perpendicular magnetization to appear only in very thin ferromagnetic films.

Pappas *et al.* [4] have performed experimental studies on the phase transition from perpendicular to parallel magnetization (reorientation phase transition, RPT). They find, in addition to the parallel and perpendicular magnetization states, a temperature interval of about 20 K between the disappearance of perpendicular magnetization and the appearance of parallel magnetization. Pescia and Pokrovsky [5] have proposed that the RPT is the result of a competition between the surface magnetic anisotropy and the dipole forces renormalized by strong thermal fluctuations (see also the discussion [6]). Earlier we conjectured that the regime where the magnetization disappears, observed by the authors of [4], can be ascribed to a domain structure penetrating the film [7]. The important role of domains was anticipated by the authors of Ref. [4].

The present paper focuses on the phase with perpendicular magnetization. Yafet and Gyorgy have considered [8] this phase at low temperatures, finding that the long-range dipolar interaction makes it unstable with respect to the formation of a stripe domain structure. Allenspach, Stampanoni, and Bischof [9] discovered orientationally disordered domains in a film of Co on Au(111) using electron microscopy. More recently, Allenspach and Bischof have found much more ordered domain structures in a system of Fe/Cu(001), for which the RPT occurs at $T_{\mathrm{RPT}} = 288$ K. They find well-defined stripes clearly aligned along the (100) axis of the substrate, but disordered positionally [10]. Domains have been observed in an interval between 230–280 K, with a period in a range of a few μm. As the temperature is decreased, the stripe period increases, and a clear up-down asymmetry develops.

In the present paper: (1) we argue that the regime where the magnetization disappears in Ref. [4] is due to this striped phase; (2) we develop the properties of the striped phase, including the physical mechanism (exchange energy within domain walls and elastic interaction between them) that causes orientational order; (3) we show that these properties provide a natural explanation for the temperature dependence of the period of the stripe structure, and for the up-down asymmetry; and (4) we show that the striped phase can be considered to be a kind of smectic liquid crystal, for which we discuss the role of thermal fluctuations in causing spatial, but not orientational, disorder [11].

Our theory begins with the standard Hamiltonian for a planar ferromagnet:

$$H = \tfrac{1}{2}\Gamma \int [\nabla \cdot \mathbf{n}(\mathbf{x})]^2 \, d^2x - \lambda \int n_z^2(\mathbf{x}) \, d^2x + \frac{1}{8\pi}\Omega(T) \iint \frac{[\mathbf{n}(\mathbf{x}) - \mathbf{n}(\mathbf{x}')]^2 - 3\{\boldsymbol{\nu} \cdot [\mathbf{n}(\mathbf{x}) - \mathbf{n}(\mathbf{x}')]\}^2}{|\,\mathbf{x} - \mathbf{x}'\,|^3} \, d^2x \, d^2x', \tag{1}$$

where Γ is the exchange integral, λ is the effective anisotropy constant (which includes contributions both from the surface anisotropy and from the short-range part of the dipole interaction), and $\Omega = 2\pi(g\mu_B S)^2/a^4$ (g denotes the Landé factor, μ_B the Bohr magneton, and a the lattice constant) is the strength of the dipole interaction; ν is a unit vector directed along $\mathbf{x} - \mathbf{x}'$. The first two terms in the Hamiltonian, Eq. (1), correspond to local interactions. The third term represents the long-range dipole interaction, which makes a single domain configuration unstable with respect to the formation of the stripe domain structure [8].

Consider now a system of parallel stripes in an external magnetic field H perpendicular to the film. Using the Hamiltonian of Eq. (1), and including the Zeeman interaction, we have calculated the free energy per unit area F of the film, assuming a long-range structure of alternating up and down stripes, of widths $L-\delta$ and $L+\delta$, respectively. There are two terms. The first is the energy per unit length $E_s = \sqrt{2\Gamma\lambda}$ due to the creation of domain walls of thickness $l = \sqrt{\Gamma/2\lambda}$, which involves both the anisotropy and the exchange energy of Eq. (1). Except near the RPT, we may assume that $l \ll L$. The second term is the interaction of the domain walls through the isotropic part of the dipole interaction of Eq. (1):

$$E_d = -\frac{\Omega}{2\pi}\sum_{m,n}\int_{2mL+l}^{(2m+1)L+\delta-l} dx \int_{(2n-1)L+\delta-l}^{2nL-l} dx' \int_{-L_y/2}^{L_y/2} dy \int_{-L_y/2}^{L_y/2} dy' [(x-x')^2 + (y-y')^2]^{-3/2}, \tag{2}$$

where L_y is the length of the film in the y direction. Including both terms, we find that

$$F = E_s n - \frac{2\Omega}{\pi} n \ln \frac{2\cos\frac{\pi\delta n}{2}}{\pi l n} - 2H\mu_B \delta n. \tag{3}$$

Here $n = 1/L$ is the total length of domain walls per unit area. We use the renormalized magnetic field $H = S^z H_{\text{bare}}$ in Eq. (3). Note that strong thermodynamic fluctuations occur in 2D for isotropic or almost isotropic ferromagnets. They renormalize the coupling constants of the magnetic Hamiltonian in a manner similar to that of Polyakov's renormalization procedure [12]. Hence, in the above equations we must employ the renormalized couplings $\Gamma(T)$, $\lambda(T)$, $\Omega(T)$, omitting the temperature dependence for simplicity [5].

For $H = 0$, the free energy, Eq. (3), reaches its minimum for $n = 1/L$ given by

$$n^*(T) = \frac{2}{\pi l}\exp\left(-\frac{\pi E_s}{\Omega} - 1\right), \tag{4}$$

which depends on temperature via E_s, Ω, and l. The exponent in Eq. (4) is typically very large so that the size of the domain $L = 1/n^*$ appears to exceed any experimental realization at low temperature, whereas it takes a reasonable value somewhat below the reorientational phase transition, where the effective magnetic anisotropy λ is sufficiently small. However, Eq. (4) fails in the vicinity of the RPT, because the condition $l \ll L$ does not hold. In that case, instead of well-defined domains, there is simply a modulated spin structure, with characteristic dimension given by the so-called dipole length value $L_d = 4\pi\Gamma/\Omega$ [13, 14].

For $H \neq 0$, minimization with respect to δ and n leads to

$$\delta = \frac{2}{\pi n}\sin^{-1}\frac{4H\mu_B}{\Omega n^*}, \qquad n = \frac{1}{L} = n^*\sqrt{1 - \left(\frac{4H\mu_B}{\Omega n^*}\right)^2}. \tag{5}$$

From Eq. (5) it follows that, at the threshold value $H_c \equiv \Omega n^*/4\mu_B$ of the magnetic field, the "black" stripe broadens to infinity, whereas the thickness of the "white" stripe remains finite and has only a nonsingular temperature dependence $L - \delta \to 2/\pi n^*$. Such asymmetric behavior is in qualitative agreement with experiment [10]. Taking the experimental width of the "white" domain $L = 3 \times 10^4$ Å, and $\Omega a = 1$ K we estimate the threshold magnetic field to be $H_c \sim 0.3$ Oe.

A stripe domain structure in a thin film is a particular case of 1D crystal ordering in two dimensions. Landau and Peierls have pointed out (see, e.g., Ref. [15]) that thermal fluctuations destroy long-range order in such systems. We will now show that in the stripe domain structure there are two main causes for disorder: first, displacements of the domain walls (domain wall meandering); second, the proliferation of dislocations in the domain system, each adding one new semi-infinite stripe. In the presence of quenched impurities, however, both domain walls and dislocations can be pinned. Thus, according to the strength and density of the pinning sites, a system of stripes can be described either as a liquid or as a glass, in both cases having a preferential orientation of the domain walls. In other words, the striped phase is a kind of smectic liquid crystal. A complete theory must explain not only the RPT as a function of temperature, but must also include the effects of thermal fluctuations in destroying long-range order.

One can take domain wall meandering fluctuations into account using classical elasticity theory. The domain wall displacement is described by a one-dimensional (scalar) field $u(x, y)$, where we assume the coordinate axes x and y to be perpendicular and parallel to the stripes, respectively. Because we are basically interested in the large scale properties of the stripe domain structure, we neglect the discrete nature of the displacement $u(x, y)$ and coordinates x and y. Since a 1D solid cannot support shear, the strain tensor has only one nonzero component.

In addition to the strain contribution, the elastic free energy also includes energies associated with domain wall bending, and with the in-plane domain wall directional anisotropy:

$$F(u(\mathbf{x})) = \frac{K}{2}\left[\frac{\partial u}{\partial x}+\frac{1}{2}\left(\frac{\partial u}{\partial y}\right)^2\right]^2+\frac{\mu}{2}\left(\frac{\partial^2 u}{\partial y^2}\right)^2+\frac{\nu}{2}\left(\frac{\partial u}{\partial y}\right)^2. \tag{6}$$

The compression constant K can be obtained from the second derivative of the free energy described in Eq. (3). The bending constant μ is less easily obtained [14]. We find that

$$K = \frac{\Omega}{\pi L}, \qquad \mu = \frac{7\zeta(3)}{64\pi^3}\Omega L. \tag{7}$$

The spatial anisotropy coefficient ν is due to the orientation dependence of the domain wall energy. As discussed below, to leading order it arises from the exchange energy expanded to higher powers in the spatial derivatives of the magnetization, which yields

$$\nu = \frac{4\mid \lambda_{xxxx} - 3\lambda_{xxyy}\mid}{Ll^3}. \tag{8}$$

For the definition of the coefficients λ_{abcd}, see below [Eq. (13)]. In order of magnitude $\lambda_{abcd} \sim \lambda/3Ll$.

Neglecting ν, which is relatively weak, and comparing the bend and shear energies to find the characteristic dimensions of a dislocation, one finds that the characteristic energy E_d of a dislocation in the shearless elastic medium is of order ΩL [16,17]. This means that dislocations exist at any temperature, and the positional order does not exist even as an algebraic order [18]. Now, taking into account the finite value of ν, we find that dislocation mediated melting (proliferation of dislocations) occurs at the melting temperature:

$$T_m = \frac{1}{2\pi}\sqrt{K_R\nu_R}L^2, \tag{9}$$

where K_R and ν_R are the elastic constant and the anisotropy constant renormalized by thermal fluctuations:

$$K_R = Z^2K, \qquad \nu_R = Z^{-1}\nu, \qquad Z = \left(\frac{64\pi\mu^{1/2}\nu}{5K^{1/2}T}\right)^{2/5}. \tag{10}$$

(The renormalization procedure, a modified version of Ref. [19], will be discussed shortly, along with domain wall meandering.) The renormalized value of the dipole constant Ω is proportional to the square of the magnetization S_z^2 [5].

If $T_m < T_{\mathrm{RPT}}$, the striped phase should be disordered at temperatures between T_m and T_{RPT}, due to dislocation-mediated melting. For a reduced magnetization $S_z^2 \sim 0.1$ [4], and weak renormalized effective anisotropy [so $Z \approx 1$ in Eq. (10)], Eq. (9) implies that the stripe period L can grow considerably with decreasing temperature, before solidification of the stripes occurs at T_m, for which we estimate that $L \sim(10\text{–}100)L_d$.

In their study of smectic liquid crystals, Grinstein and Pelcovits showed [19] that in 3D space the anharmonic terms of the free energy analogous to Eq. (6) with $\nu = 0$ lead to a logarithmic renormalization of the elastic constants K and μ. In 2D, the fluctuations' correlations fall off exponentially rather than in a power-law fashion, while the effects of the anharmonic terms result in power corrections that can be obtained from an ϵ expansion in space dimension $3-\epsilon$. We have performed the calculations in the first ϵ approximation and got the exponents very close to the exact results found recently by Golubovic and Wang [20]. In particular for the correlation function of the order parameter $\phi(\mathbf{r}) = e^{ipu(\mathbf{r})}$ a minor modification of the Golubovic-Wang procedure leads to the following result:

$$\langle\phi(\mathbf{r})\phi(0)\rangle = \exp\left(-\frac{p^2}{2}Ryf(R^{1/2}x/y^{3/2})\right), \tag{11}$$

where

$$p = \frac{2\pi}{L}, \qquad R = \frac{T}{\sqrt{K\mu}} \tag{12}$$

and $f(x)$ is a dimensionless function with the asymptotics $f(x) \to$ const at $x \to 0$ and $f(x) \to x^{2/3}$ at $x \to \infty$. The renormalized anisotropy constant $\nu(T)$ of Eq. (10) has been found in a similar way. Because of rotational symmetry the Hamiltonian preserves the form of Eq. (6) in any order of perturbation by anharmonicity; only the effective elastic coefficients change. Note that the correlation function of Eq. (12) displays an anisotropy in the x, y plane. Under the conditions of the experiment [10] we estimate the coefficient R in the exponent to be of the order of unity. This means that correlations along the x direction decay within a few lateral periods, thus justifying the use of the continuum approximation. Beyond the distance scale $\sqrt{\mu/\nu}$ the correlation reduction stops, and positional order persists up to the scale determined by thermally generated dislocations. This picture of two distinct scales works only for $L \gg L_d$. The correlation (10),(11) can be checked experimentally by polarized electron diffraction and by detailed study of the electron microscopy photographs.

Finally, we indicate how the exchange energy within the domain walls causes the domain walls to have a preferential orientation. One must expand the exchange energy up to terms of the fourth order in space derivatives, thus obtaining

$$\delta H_{\mathrm{exch}} = \tfrac{1}{2}\int \lambda_{abcd}\frac{\partial^2 S^i}{\partial x_a \partial x_b}\frac{\partial^2 S^i}{\partial x_c \partial x_d}\, d^2x, \tag{13}$$

where $a, b, c, d = x, y$; $i = 1, 2, 3$; and S^i is the local value of the spin. The λ_{abcd} are coefficients of order of magnitude Γa^2, and are symmetric under any permutation of indices. For tetragonal symmetry there are two classes of nonzero coefficients, λ_{xxxx} and λ_{xxyy}. Domain walls will

be oriented along the x or y axis if $\lambda_{xxxx} < 3\lambda_{xxyy}$, and along the bisector otherwise. Specifically, for a Heisenberg model on a square lattice with nearest neighbor interactions we find $\lambda_{xxyy} = 0$ and $\lambda_{xxxx} = -\Gamma a^2/12 < 0$. The maximal gain of domain wall energy is achieved when it is parallel to the x or y axis, and calculation shows that its orientation dependence goes as $\cos 4\theta$. Since $\theta \approx \partial u/\partial y$ for small θ, expansion of this energy for small θ permits the stiffness ν in Eq. (8) to be determined. Note that the preferential orientation is not destroyed by thermal fluctuations. Indeed, the mean square of the angular fluctuations $\langle(\frac{\partial u}{\partial y})^2\rangle$ can be shown to converge even for $\nu = 0$ [18]. The contribution of next-nearest neighbors to the coefficients λ_{abcd} is much more substantial than that to the effective exchange constant Γ. The orientation of domain walls is attracted due to this contribution to the direction of a corresponding bond. On the plane (001) of an fcc crystal it is the direction (100) which has been observed experimentally.

In conclusion, we have presented a coherent picture of the major properties of the stripe domain structure in ferromagnetic thin films possessing a reorientational phase transition. The rapid growth of stripe size and the development of stripe asymmetry with decrease in temperature, and the preferential stripe orientation with respect to the lattice, have all been considered. Moreover, we have shown that over a wide range of temperatures below the reorientational phase transition, the stripe domain structure can be described as a smectic liquid crystal. It has no long-range positional order, due to dislocation mediated melting, although orientational long-range order persists. The minimal domain size near the RPT is the dipole length $L_d \sim 1$ μm. A rather weak magnetic field $H < 1$ Oe leads to a dramatic, asymmetric, collapse of the minority stripes. An interesting problem beyond the scope of our considerations is the glassy state of the domain liquid, caused by quenched defects. One expects that in 2D strong thermal fluctuations will obscure the difference between liquid and glass. Only for measurements over a short time scale can this difference occur. In any case, on the time scale of the experiment [10] (approximately one minute), the results are consistent with a picture based on the assumption of reversible behavior.

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