PHYSICAL REVIEW B VOLUME 26, NUMBER 1 1 JULY 1982

Phase transitions with spontaneous modulation—the dipolar Ising ferromagnet

T. Garel* and S. Doniach
Department of Applied Physics, Stanford University, Stanford, California 94305
(Received 8 February 1982)

A uniaxial ferromagnet of finite thickness spontaneously breaks into parallel "striped" domains as a result of demagnetizing forces. In a sufficiently large applied magnetic field a transition takes place to a hexagonal or "bubble" phase. We study the phase diagram of this system with the use of a Ginzburg-Landau approach and we investigate the effects of dislocations on the two-dimensional order of the modulated phases.

I. INTRODUCTION

Modulated phases in which an order parameter has a spontaneous spatial variation may be thought of as resulting from a competition between short-range attractive forces leading to a local alignment and longer-range repulsive forces leading to a competitive antialignment. Examples[1] are charge-density-wave systems and anisotropic magnetic systems such as the axial next-nearest-neighbor Ising (ANNNI) model[1] or the helical antiferromagnet.[2] Spin-glasses, in which the competing interactions are disordered, exhibit even more complex phases.[3]

The nature of the phase transitions of models of this type has recently received renewed attention. The purpose of this paper is to discuss the phase diagram of a particular class of spontaneously modulated systems, represented by the dipolar ferromagnet with uniaxial anisotropy in the geometry of a slab of finite thickness D and infinite extent in the plane.

At low temperatures, in zero applied field, this class of magnet (much studied in connection with bubble memory applications[4]) spontaneously orders in a structure of stripe domains, whose period results from a balance between domain-wall energy and demagnetizing energy.[5] Above a critical applied field, a first-order transition takes place to a hexagonal phase, which we will refer to as the "bubble" phase. At still higher fields, a further transition takes place to a uniformly magnetized phase.

A basic distinguishing feature of this class of models relative to the anisotropic models (of the ANNNI type) is the complete degeneracy of the Hamiltonian with respect to orientation in the plane. In particular, this means that fluctuations into the striped phase at high temperatures strongly suppress the Ginzburg-Landau mean-field transition, as first discussed by Brazovskii.[6]

Another important distinction from models of the ANNNI type arises from the long-ranged nature of the dipolar interaction: For finite slab thickness, the dipolar coupling is *always* relevant, no matter how weak relative to the nearest-neighbor exchange. So the ground state of the system remains modulated at all values of the ratio $a(g\mu_B)^2/JD$. (See Sec. II for definitions.)

In this paper, we first discuss the phase diagram of the system in an applied magnetic field in the context of a Ginzburg-Landau approach, then study the effects of defects and fluctuations on the mean-field phase transitions. In Sec. II, the model is formulated as lying in a class of two-dimensional Ginzburg-Landau models with nonlocal interactions, and in Sec. III, the effects of an applied field are worked out in the Ginzburg-Landau approach. In Sec. IV, we show that dislocations in the striped phase will always lead to absence of positional order in this phase at finite temperature, but that a Kosterlitz-Thouless transition will occur in the bubble phase.

II. MODEL

We consider a slab of thickness D of a strongly uniaxial (Ising-like) material. At low temperatures and zero applied field, the system breaks into domains due to the competition between exchange and dipolar forces, with a zero net magnetization [Fig. 1(a)]. We take the system as isotropic in the xy plane. Assuming a periodic structure of period $2d$, the energy of the system per unit surface of the slab may be written[7]

$$E=\left[4\pi\xi\frac{D}{d}+\frac{16d}{r^2}\sum_{n\ \mathrm{odd}}\frac{1}{n^3}(1-e^{-n\pi D/d})\right]M_s^2 \quad . \qquad (1)$$

In Eq. (1), ξ is a parameter characterizing the domain-wall thickness.[4] The first term represents the wall energy and the second the demagnetizing contribution. The equilibrium distance is given by

$$\frac{\partial E}{\partial d}=0 \quad . \qquad (2)$$

There are a number of underlying assumptions in Eq. (1), such as the fact that the walls are taken to be

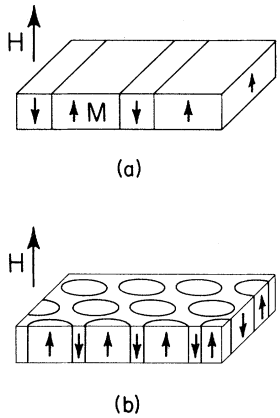

FIG. 1. (a) Low-temperature zero-field ground state; (b) bubble lattice.

straight, branching in the wall structure is neglected, etc. Typically, these assumptions are valid for ratios $2 \lesssim D/\xi \lesssim 20$ (Refs. 4, 8–10) which we will consider from now on. In a perpendicular magnetic field H above a critical value, it is well known that the striped structure of Fig. 1(a) gives way to a bubble lattice structure [Fig. 1(b)]. In general, the energies of the two structures are quite close.[8,11]

If one wants to study the phase transitions between paramagnetic, striped, and bubble phases, one has to deal with dipolar forces, which, because of their anisotropic long-range character, can have subtle effects. For instance, they have been shown to be relevant close to four space dimensions.[12] In our "two-dimensional" system, their effect is contained in the infinite sum of Eq. (1), resulting from the domain structure. This function varies as $D^2/d\ln(D/d)$ for $D < d$, leading to a nonzero period (at $T=0$) for all values of the ratio $\xi D/d^2$. So the long-range character leads to a coupling which is always relevant, as discussed in Sec. I.

At nonzero temperature the walls will be smeared out by fluctuations,[13] and one may anticipate that, close to T_c, the infinite sum of Eq. (1) will be dominated by the first harmonic. This approximation is familiar in the context of modulated Ising structures where, in mean field, the amplitude of the nth harmonic grows as $(T_c - T)^{n/2}$ below T_c. We also anticipate that the stripe lattice will be broken by dislocations, leading to a fluid "smecticlike" model with no positional long-range order, but with orientational order.[14]

The key point is that in a Ginzburg-Landau (GL) free energy, the competition between the wall energy [$(\nabla\psi)^2$ term] and the dipolar term will yield, close to the critical temperature, a nonzero optimal wave vector as in Eq. (1); the direction of this wave vector is arbitrary in the xy plane, due to the isotropy of the system. The phase transition is therefore expected to be of the Brazovskii type.[6]

The above assumption, that the walls are straight in the z direction (normal to the slab plane) reduces the statistical-mechanical problem to two dimensions. In the context of a Ginzburg-Landau approach, the three-dimensional free-energy functional may be reduced to two dimensions as follows: The total free energy may be written

$$F = \int d^2x\,(F_{\rm exch} + F_D) \ , \tag{3}$$

where $F_{\rm exch}$ is the bulk ferromagnetic contribution averaged over the slab thickness, and F_D is the surface demagnetizing term. For a microscopic Ising-like model of a ferromagnet with coordination number z, lattice spacing a, and nearest-neighbor ferromagnetic exchange J, we may write

$$F_{\rm exch} = \frac{D}{2a}J(\nabla m)^2 + \frac{D}{a^3}(T - T_F)\frac{m^2}{2} + \frac{DT_F}{12a^3}m^4 - g\mu_B H\frac{mD}{a^3} \ , \tag{4}$$

where $\bar{m}(x)$ is the slab average of the magnetization oriented along the z direction, and $g\mu_B$ is the gyromagnetic coefficient. T_F is the mean-field ferromagnetic transition temperature (k_B is set equal to unity),

$$T_F = zJ \ .$$

The contribution of the demagnetizing term is[7]

$$F_D = \tfrac{1}{2}\int_S dS\,\sigma\Phi \ ,$$

where σ is the charge distribution of the surfaces created by nonzero magnetization $m(r)$ and Φ is the magnetic potential on the surface, created by σ.

For a general magnetization of this form, the demagnetizing term may be rewritten

$$F_D = \tfrac{1}{2}\int d^2x\, d^2x'\, m(x)\, g(x,x')\, m(x') \ , \tag{5}$$

where

$$g(x,x') = (g\mu_B)^2 \int dq \frac{4\pi}{q}(1 - e^{-qD}) \times \exp[iq(x - x')] \ .$$

We thus see that the dipolar magnet falls into a general class of models with competing short-range attraction and longer-range repulsion.

The Ginzburg-Landau instability condition may now be seen to occur at a finite q vector, such that

$$\frac{df_q}{dq} = \frac{d}{dq}\left[4\pi\frac{(g\mu_B)^2}{q}(1 - e^{-qD}) + \frac{Jq^2D}{a}\right] = 0 \ . \tag{6}$$

The resulting mean-field phase transition occurs at a transition temperature T_s, *above* the transition for the uniform ferromagnet T_F.

III. GINZBURG-LANDAU PHASE DIAGRAM

Following Brazovskii,[6] we study two basic modulated solutions:

(i) the stripe solution,

$$m = m_{s0} + m_s \cos(q_0 x) \quad ; \tag{7a}$$

(ii) the bubble solution,

$$m = m_{B0} + \sum_{i=1}^{3} m_B \cos(\vec{k}\cdot\vec{r}_i) \quad , \tag{7b}$$

with

$$\sum_{i=1}^{3} \vec{k}_i = 0, \quad |\vec{k}| = q_0 \ .$$

Note that since we are dealing with a magnetic problem, the bubble solution is not present in a zero magnetic field.[15] The solutions (i) and (ii) do not include, as mentioned before, higher-order harmonics, which in our approximation grow more slowly at temperatures just below the phase transition. Inserting Eqs. (7a) and (7b) in Eq. (6), we have

$$\text{(i)}\quad f_s = \frac{F_s}{L_x L_y} = \tfrac{1}{2} f_0 m_{s0}^2 + \tfrac{1}{4} u m_{s0}^4 - h m_{s0} + m_s^2\left(\tfrac{1}{4} f_{q_0} + \tfrac{3}{4} u m_{s0}^2\right) + \tfrac{3}{32} u m_s^4 \ , \tag{8}$$

$$\text{(ii)}\quad f_B = \frac{F_B}{L_x L_y} = \tfrac{1}{2} f_0 m_{B0}^2 + \tfrac{1}{4} u m_{B0}^4 - h m_{B0} + 3 m_B^2\left(\tfrac{1}{4} f_{q_0} + \tfrac{3}{4} u m_{B0}^2\right) + \tfrac{9}{32} u m_B^4 + \left(\tfrac{3}{2} u m_{B0} m_B^3 + \tfrac{9}{8} u m_B^4\right) \ , \tag{9}$$

where

$$\tfrac{1}{2} f_0 = \tfrac{1}{2}(T - T_F)\frac{D}{2a^3} + (g\mu_B)^2 2\pi D = \tfrac{1}{2}(T - T_0)\frac{D}{2a^3} \ ,$$

$$\tfrac{1}{4} f_{q_0} = \frac{1}{4}\left[(T - T_F)\frac{D}{2a^3} + q_0^2 \frac{JD}{a} + \frac{4\pi (g\mu_B)^2}{q_0}(1 - e^{-q_0 D})\right] = \tfrac{1}{4}(T - T_{q_0})\frac{D}{2a^3} \ ,$$

$$\tfrac{1}{4} u = \frac{T_F D}{12 a^3}, \quad h = (HD/a^3) g\mu_B \ ,$$

and q_0, obtained by minimizing F_{q_0} is given by

$$q_0 = [2\pi a (g\mu_B)^2 / JD]^{1/3} \ ,$$

assuming $q_0 D \gg 1$. As seen from Eq. (9), the bubble phase is stabilized by a suitable choice of phase relations between the three simultaneous modulations [Eq. (7b)], such that the term in $m_{B0} m_B^3$ becomes negative and sufficiently large.[16]

On minimizing Eqs. (8) or (9), one finds a stability condition for the applied field beyond which a real solution no longer exists, given by

$$3 u m_{s0}^2 > -f_{q_0}\colon\ m_s = 0 \tag{10}$$

for the striped phase;

$$\tfrac{12}{5} u m_{B0}^2 > -f_{q0}\colon\ m_B = 0 \tag{11}$$

for the bubble phase. Equations (10) and (11) correspond to critical fields

$$h_c^s = \frac{-f_{q_0}}{3}\left[f_0 - \frac{f_{q_0}}{3}\right] \ , \tag{12a}$$

$$h_c^B = \frac{-5 f_{q_0}}{12}\left[f_0 - \tfrac{203}{300} f_{q_0}\right] \ . \tag{12b}$$

A numerical study of f_s and f_B shows that these stability fields are never reached and the system undergoes a first-order transition at critical fields $0 < h_s$, $h_B < h_c^s$, h_c^B. The phase diagram is shown in Fig. 2 with the particular choice ($T_0 = T_{q_0}$), since we want to focus on the phase transition at T_{q_0}. For different values of the parameters the phase boundaries change, but the topology of Fig. 2 is preserved.

The $(S) \leftrightarrow (B)$ transition is also first order but apparently with a continuous magnetization $m_{s0} = m_{B0}$ at the phase boundary.

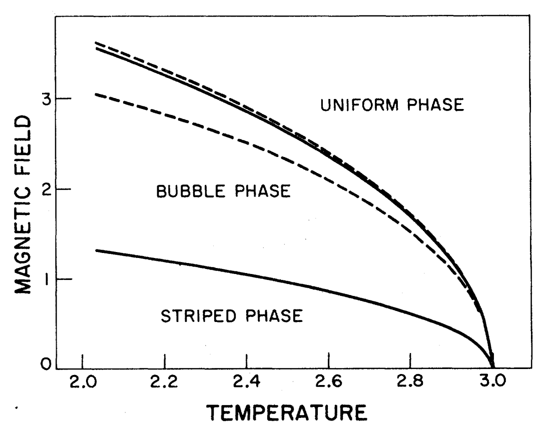

FIG. 2. Ginzburg-Landau phase diagram: solid lines, first-order phase boundaries; dashed lines, critical fields.

IV. FLUCTUATION EFFECTS

The phase diagram of Fig. 2 did not take fluctuations into account, which in the stripe or bubble solutions appears to be essential. Brazovskii, using first-order perturbation theory, found that the phase transition for $h=0$ was driven first order by fluctuations, with a shift of transitions temperature[6],

$$(T_c - T_{q_0}) \cong (q_0 u)^{2/3} \ .$$

On the other hand, the absence of long-range order in systems with a one-dimensional modulation raises questions about the validity of a perturbation analysis.[13,17,18] We now show that the absence of rigidity for shear distortions of the stripe lattice means that dislocation pairs in the stripe lattice will always be unbound at finite temperature, leading to an absence of positional long-range order in this phase. To see this, we retain the first term of Eq. (6) beyond the GL approximation of Sec. III to give

$$F_S = \frac{1}{2}\int m_q m_{-q}\left[f_{q0} + \frac{(q-q_0)^2}{2} f^4_{q_0} + (T-T_F)\frac{D}{2a^3}\right] d\vec{q} \ , \qquad (13)$$

where the integration is carried out in the xy plane. The elastic contribution of Eq. (13) may be rewritten as

$$\delta F_s \simeq \frac{1}{2}\int m_q m_{-q} \frac{(q^2-q_0^2)^2}{2(2q_0)^2} f''_{q_0} d\vec{q}$$
$$\simeq \frac{1}{2}\left[\frac{1}{8q_0^2} f''_{q_0}\right]\int [(\nabla^2 m)^2 - 2q_0^2(\nabla m)^2 + q_0^4 m^2] d^2\vec{x} \ . \qquad (14)$$

The peculiarity of the Brazovskii phase transition shows up in the elasticity. In a phase-only approximation, one has

$$m(\vec{r}) = m\,\mathrm{Re}(\exp\{iq_0[x+u(x,y)]\}) = m\,\mathrm{Re}(e^{i\phi}) \ , \qquad (15)$$

where x is the direction of the one-dimensional (1D) modulation, of amplitude m [Eq. (8) with $h=0$]. One has then, in the harmonic approximation,

$$\delta F^{(s)}_{el} = \frac{1}{32} f''_{q_0} m_s^2 \int d^2\vec{q}\, u_q u_{-q}(4q_0^2 q_x^2 + q_y^4) \ . \qquad (16)$$

At finite temperatures, the interaction energy of a pair of dislocations in an elastic medium, characterized by Eq. (16), falls off more rapidly than the logarithm of the isotropic case. Consequently, dislocation pairs are always unbound at finite temperature and the striped phase does not have positional long-range order. In general, it will be expected to have orientational order up to some temperature above which disclination pairs become unbound.[19] However, the Brazovskii perturbational analysis leads to a first-order transition. As in other melting transitions, one cannot tell *a priori* whether the disclination unbinding temperature comes above or below the Brazovskii first-order transition temperature.

For the bubble phase, we show that dislocation unbinding will occur at a finite temperature Kosterlitz-Thouless transition.

A similar analysis of the elastic behavior in the bubble case leads to

$$\delta F^B_{el} = \frac{1}{2}\int d^2 r(2\mu u_{ij}^2 + \lambda u_{ii}^2) \ ,$$

with

$$u_{ij} = \frac{1}{2}(\partial_i u_j + \partial_j u_i), \quad i,j \equiv x,y \ ,$$

where the Lamé coefficients are[20]

$$\mu = \lambda = \frac{3}{32} f''_{q_0} m_B^2 q_0^2 \ ,$$

where m_B is the same as in Eq. (9). Neglecting temporarily the term $(1-e^{-qD})$ in f_q [Eq. (6)], we find

$$\lambda = \mu \frac{9\pi}{8}\frac{m_B^2}{q_0}(g\mu_B)^2 \ . \qquad (17)$$

With these Lamé coefficients, we can estimate the dislocation unbinding temperature T^0_m for the bubble lattice[19] using

$$\frac{1}{\mu} + \frac{1}{\mu+\lambda} = \frac{a_0^2}{4\pi k_B T^0_m} \ . \qquad (18)$$

In general, we expect this to provide an overestimate of the melting temperature. In Eq. (18), a_0 is the bubble lattice spacing, related to q_0 by

$$a_0 = \left[\frac{4\pi}{q_0\sqrt{3}}\right] \ . \qquad (19)$$

Inserting values for μ and λ from Eq. (17) we find $k_B T^0_m = \nu m_B^2 a_0^3$, leading to

$$k_B T^0_m = \frac{\pi}{2}\frac{JD}{a} m_B^2(T^0_m, H) \ , \qquad (20)$$

where ν is a numerical coefficient given by $3\sqrt{3}/64\pi \cong 2.6\times 10^{-2}$. We note that in this approximation, T^0_m scales like a_0^3, i.e., with the slab thickness D. In the region of T_s, m_B varies, in the GL approximation, as

$$m_B^2 = \frac{C}{J}[T^0_B(H) - T] \ ,$$

where $T^0_B(H)$ is a bubble-ferromagnetic instability temperature at fixed H, and C is a number of order unity. Since for practical samples, D/a may be several hundred, this leads to a bubble lattice melting temperature very close to the bubble-ferromagnetic instability boundary

$$\frac{k_B T_m^0}{J} = \frac{\pi D/2aC}{1+\pi D/2aC} \frac{k_B T_B(H)}{J} \approx \left(1 - \frac{2aC}{\pi D}\right) \frac{k_B T_B(H)}{J} .$$

Since the actual bubble-ferromagnetic phase boundary is first order in the GL approximation and is likely to be displaced by corrections of the Brazovskii type, it is not possible to give an accurate numerical prediction of the location of the bubble melting transition. So our analysis serves to indicate where to look for the transition: Experimental observation would provide a stimulus for more detailed study of models of the Brazovskii class.

V. SUMMARY AND CONCLUSIONS

Our analysis shows that there are two types of fluctuation effects which cause melting of the stripe and bubble structures. The first type are the directional fluctuations discussed by Brazovskii which are expected to lead to first-order transitions. The second type are dislocations leading to second-order melting of the Kosterlitz-Thouless type. Our analysis, which neglects higher harmonics of the spontaneous modulation, will be expected to be valid only at temperatures close to the paramagnetic transition. We expect the transition between the stripe and bubble phases to remain first order even in the presence of fluctuation effects.

Our discussion has been based on an Ising type of model. For non-Ising systems with large enough uniaxial anisotropy, we expect the same behavior.[21] For vectorial spins, one expects a critical field below which the spins are canted.[22] The same situation is to be found whenever demagnetizing effects are important.[23]

Experiments have shown that the bubble-stripe phase transition is strongly first order at low temperatures, but, to our knowledge, there is no experimental report of stripe or bubble melting. It now seems possible to prepare amorphous ferromagnets with low in-plane anisotropy,[4] to which the theory developed here could apply. Since the bubble melting temperature scales with sample thickness, this could provide an interesting laboratory system in which to study two-dimensional melting.

ACKNOWLEDGMENTS

The work of T. Garel was partly supported by a NATO Fellowship; he also wishes to acknowledge the hospitality of the Department of Applied Physics of Stanford University and of S. Doniach. Part of the work was carried out at the Aspen Center for Physics. The authors thank the staff for their support. This research was partly supported by NSF Grant No. DMR-07934 and NIH Grant No. GM-25217.

*On leave from Laboratoire de Physique des Solides, associé au CNRS, Université de Paris-Sud, Orsay, France.

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