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LETTER TO THE EDITOR

The temperature dependence of the domain spacing in ultrathin magnetic films

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Abstract. We study the domains in an ultrathin magnetic layer with perpendicular magnetization as a function of temperature. The domain spacing is given by the balance between the interface energy and the demagnetizing energy. The demagnetizing energy is calculated in a continuum theory which is valid provided that the domains are large compared with the lattice spacing. The temperature dependence of the interface energy arises from the entropy of the rough walls. This is obtained from the exact results for the two-dimensional Ising model. We predict that the domain size shrinks rapidly as the temperature is raised towards the transition.

Recent experiments [1] have observed magnetic domain walls in ultrathin magnetic films (of a few monolayers (ML thickness). This letter is concerned with the evolution of the domains as the temperature is raised, for films with strong anisotropy such that the direction of magnetization is perpendicular to the plane.

The theory of domains in ultrathin films has been studied by Yafet and Gyorgy [2] and Kaplan and Gehring [3] at low temperatures using a continuum model. Czech and Villain [4] have used a discrete model and also studied the 'floating phase' which will occur at high temperatures. The domain walls in a two-dimensional 2D Ising system are rough for $T \neq 0$ which means that the regular array of walls becomes destabilized and the domain lattice 'melts' at low temperatures. They considered the breakdown of the regular domain pattern and also the limiting behaviour near to the transition to the paramagnetic state.

We use an alternative approach. The temperature dependence of an isolated interface in the 2D Ising model is known exactly [5, 6]—we use this in a mean-field model based on well separated and hence non-interacting walls. At low temperatures the size of the domains is large, of the order of microns, and so a continuum theory which is used to evaluate the magnetostatic is valid. We find that as the temperature is raised the size of the domains shrinks rapidly; this means that we appear to enter a regime in which the domain size becomes microscopic. At this point our theory has broken down, however we have shown that an ordered phase is no longer stable. Hence the inclusion of dipolar effects lead to a strong reduction in the apparent transition temperature.

In bulk systems it has been shown that the domain spacing is reduced sharply in the neighbourhood of T_c [7, 8, 9] and that the critical point occurs approximately when the size of the domain spacing becomes of the order of magnitude of the correlation length (at this point the assumption of non-interacting walls has broken down). We use the same condition in the 2D case.

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In the absence of dipolar interactions the ordered phase is ferromagnetic and the ordered parameter is the average magnetization. Inclusion of the dipolar interactions means that domains form and so the average magnetization of the whole sample is zero. The order parameter at low temperatures becomes the regular arrangement of the domain walls. Thus as the temperature is raised the system loses its long range order when the domain pattern becomes disordered rather than when the magnetization vanishes within each domain.

In this letter we generalize the work of Kaplan and Gehring [3] to finite temperature. The high-temperature region is examined and the results compared with the theory of Czech and Villain [4]. The implications for experiment are also discussed.

At low temperatures the observed domain size is large—of the order of 10^3 lattice spacing—so a continuum theory is valid. In an Ising system the domain walls are sharp (one lattice spacing wide) at $T = 0$ but at higher temperatures they broaden into a continuous linear wall of width ξ (the correlation length). At finite temperature the wall is rough—this additional entropy causes the interface free energy σ to fall rapidly with temperature [5]. It actually falls almost linearly with $|(T_c - T)/T_c|$ for all $T/T_c \geqslant \frac{1}{3}$; this is exact close to T_c. Here T_c is the transition temperature of the magnetic model *excluding* the dipolar interactions.

In a real magnet the anisotropy K is smaller than the exchange interaction J, leading to a Bloch wall of width $w_0 \simeq a_0\sqrt{(J/K)}$ at low temperature (a_0 is the lattice spacing) and a linear wall at high temperature [10].

In this work we assume that the wall width w is equal to the Bloch width w_0 at low temperatures, and then becomes equal to the correlation length ξ, for $\xi \geqslant w_0$. However we take the temperature dependence of the interface energy from the Ising model calculation [5] because this includes the roughening effects correctly.

The domain structure arises because of an interplay between the interface energy (wall energy) and the dipolar energy. We use the results of the 2D Ising model to obtain the temperature dependence of the energy of an isolated interface and we consider only the situation in which the domain walls are sufficiently widely separated for the interactions between walls to be neglected. The dipolar energy depends on the value of the perpendicular magnetization in the film. As it is a long-range interaction one needs to sum over the whole film. The average perpendicular magnetization will vanish over the region of the Bloch wall and this effect causes strong changes in the dipolar energy. Since our theory is valid only for well separated walls we use the continuum model to calculate the dipolar energy throughout.

We assume a stripe configuration of walls (this is the equilibrium configuration for the continuum model [2, 3]). The energy per unit area is of the sum given by $\sigma(L/D)$ where L is the film thickness and D is the domain size. For sharp domain walls the dipolar energy is given in terms of a sum [11, 3]:

$$U_{\text{dip}} = \frac{16}{\pi^2} M_s^2 D \sum_{\substack{n=1 \\ n \text{ odd}}}^{\infty} \frac{1}{n^3}[1 - \exp(-n\pi L/D)]. \tag{1}$$

For finite wall width, w_0, the sum will be cut off at $n = N_0 = D/w_0$. It is convenient to write $L/D = x$. For the continuum model to be valid we need $x \ll 1$ (experimentally at low temperatures $x \simeq 10^3$ for Co on Au [1]). The free energy for the domain state is given by combining the interface and dipolar energies:

$$F(x) = \sigma x + \frac{16}{\pi^2} M_s^2 \frac{L}{x} \sum_{n \text{ odd}}^{N_0} \frac{1}{n^3}[1 - \exp(-n\pi x)]. \tag{2}$$

For small x following [3] we can write the sum in terms of a sum for small n and an integral which is non-analytic as $x \to 0$:

$$F(x) = \sigma x + 2\pi M_s^2 L x \left(\frac{\pi^2 s}{2} + \frac{\pi w_0}{2L} - \frac{\pi^2}{4} \ln \frac{w_0 x}{L} \right) + O(x^2). \tag{3}$$

The value of s is obtained from the finite series [3] and is $s = 0.634$. The equilibrium domain size D is found from minimizing equation (3) with respect to x.

$$D = w_0 \exp\left(\frac{\pi D_0}{2L}\right) - a \tag{4}$$

where

$$D_0 = \frac{\sigma}{2\pi M_s^2} \tag{5}$$

and

$$a = \frac{2w_0}{\pi L} + 0.268. \tag{6}$$

At $T = 0$ the value of $\pi D_0/2L$ is about six for a 3 ML film of cobalt on gold [3] leading to a large value of D, of the order of 10^{-6} m.

We evaluate the temperature dependence of D from equation (4) using the results of the 2D Ising model [5, 6]. For the 2D Ising model σ is almost constant for $t = (T/T_c) \leqslant 0.3$ and then falls linearly to zero at T_c. The magnetization is almost constant until $t \simeq 0.9$ when it falls abruptly to zero (as $(1-t)^{1/8}$) and the correlation length diverges as $(1-t)^{-1}$ near T_c. Over the temperature range of interest the important variation is that of σ because it is linear over such a large range and because it is in the exponential in equation (4). M_s is essentially constant over the range of interest and we can set $w \simeq w_0$ (the Bloch wall width) for $w_0 > \xi$.

A plot of reduced domain size $D(T)/D(T=0)$ as a function of reduced temperature, $k_B T/J$, obtained from equation (4) is given in figure 1, where the temperature dependence has been taken from the Onsager solution of the Ising model. A reduction of domain size has been observed by Allenspach [12] in some preliminary experiments on 3 ML FCC Fe/Cu (100) he finds that D starts to drop sharply at $T/T_c \simeq 0.6\,T_c$.

In the following we look at the high temperature or critical region more carefully. At low temperatures the dipolar interactions may be considered to be a small perturbation on the short range exchange. We have seen that experimentally the domain size may be some 103 lattice spacings for an ultrathin film [1]. Under these circumstances the domain walls are pinned and the magnetization within a domain may be measured experimentally. Technically one should regard the order parameter to be the regular array of domain walls. As the temperature is raised Czech and Villain [4] have argued that at a temperature T_R such that $k_B T_R = 2\pi M_s^2 L$ the walls cease to be pinned to the underlying lattice and there is a transition to a 'floating' array. Thus the 'ordered' phase is an incommensurate array of walls with periodicity $q = \pi/D$. Such a phase is describable in terms of an $n = 2$ order parameter corresponding to the amplitude and phase of the modulated structures. In two dimensions an ordered phase with $n = 2$ is not allowed with short-range interactions—one has a Kosterlitz–Thouless transition [13]. The effect of long range dipolar effects on this transition is an open question.

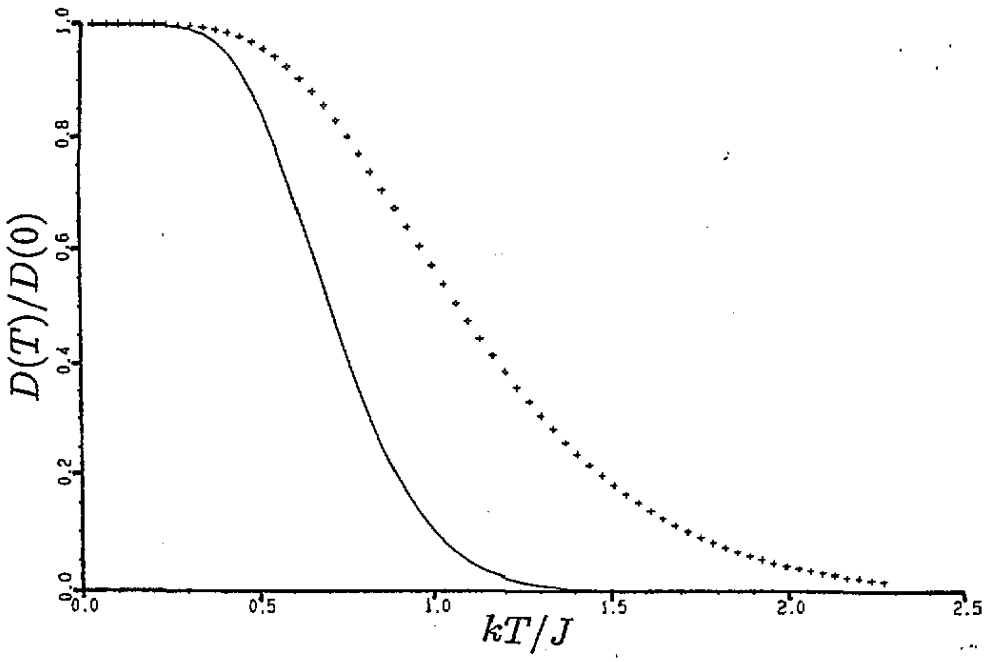

Figure 1. The temperature dependence of the domain size for a film containing one (solid line) or two (dotted line) magnetic layers.

The mean-field theory we are describing does not take account of the fluctuations of the array of domain walls which is *assumed* to remain as a regular stripe lattice. However it does include the isolated interface energy correctly as that is taken from the exact solution of the Ising model.

Near to the critical point the domain wall width is given by the correlation length $w \simeq \xi = a_0/(1-t)$ where a_0 is the lattice spacing.

The values of D may not reduce below 2ξ—at this point the size of the 'domain' is the size of the 'wall' and the concept of an ordered phase breaks down. From equation (4) we see that this occurs for

$$\frac{\pi D_0}{2L}(t^*) - a(t^*) \simeq \ln 2 \tag{8}$$

in the region where the interface energy varies as $\sqrt{2}\sigma_0(1-t)$; this is for $0.3 \leqslant t < 1$ we can solve equation (8) to find t^*. For Co on Au where at $T = 0$, $\pi D_0/2L \simeq 6$ we find that $t^* = 0.89$. Thus we see that equation (4) leads to the prediction that the domain size falls so precipitously as the temperature is raised that the apparent ordering temperature is reduced to of the order of 0.9 of the value that it would have had in the absence of dipolar interactions. The assumptions of the theory have broken down at this point because the walls will begin to interact when their separation becomes comparable with the correlation length, also our use of the continuum model to evaluate the dipolar energies becomes doubtful. However if one has reached this point when the domain separation has become almost microscopic then it would be hard to distinguish this state experimentally from one in which there were correlations but no long-range order. Our theory does predict that the phase with a large

domain size becomes unstable at a temperature considerably below the value where the transition temperature would be expected to occur if all dipolar effects were absent.

This contrasts to the predictions of Czech and Villain [4] who used a mean-field argument to predict that the domain size was reduced to $J/2\pi M_s^2$ and that the transition temperature was essentially unchanged by the dipolar interactions.

Experimentally the domain walls are not regularly ordered at low temperatures [1]. Hence on strict theoretical grounds there is no ordered phase and hence no thermodynamic phase transitions. Rather there must be a freezing transition so that the 'ordered' phase appears when the domain walls become pinned or frozen. This would make it analogous to the region near 'H_{c_2}' for a high-temperature superconductor. Experimentally both the magnetization [14] and the coercive field [15] appear at a sharp temperature (or thickness) and appear to follow power law behaviour.

We have calculated the temperature dependence of the domain size for ultrathin films magnetized perpendicularly to the plane, and shown that it reduces sharply. The inclusion of dipolar interactions in this case causes radical modification of the nature of the phase transition expected for a short-range model.

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