Metastable states in ultrathin magnetic films

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Abstract

The equilibrium phase diagram of a two-dimensional Ising model with competing
exchange and dipolar interactions is analyzed using a Monte Carlo simulation tech-
nique. We consider the low temperature region of the (δ, T) phase diagram (δ being
the ratio between the strengths of the exchange and dipolar interactions) for the
range of values of δ where striped phases with widths $h = 1$ and $h = 2$ are present.
We show that the transition line between both phases is a first order one. We also
show that, associated with the first order phase transition, there appear metastable
states of the phase $h = 2$ in the region where the phase $h = 1$ is the thermodynam-
ically stable one and *viceversa*.

The physics of ultrathin films and quasi-two-dimensional systems is of current
interest because of its many technological applications. For instance, metal-
on-metal films are used in electronics, data storage and catalysis. In partic-
ular, the use of ultrathin magnetic films as data storage devices requires a
high degree of accuracy and spatial resolution in the magnetization control.
Hence, a lot of experimental and theoretical effort has been devoted in the last
years[1] to understand both the equlibrium and out-of-equilibrium properties
of these kind of systems. Realistic theoretical descriptions of these systems in-
clude both the exchange and the dipolar interactions between the microscopic
spins. The competition between long ranged antiferromagnetic dipolar inter-
actions and short ranged ferromagnetic exchange interactions can give rise to
a variety of unusual and interesting macroscopic phenomena. Works in two di-
mensional uniaxial spin systems, where the spins are oriented perpendicular to

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the lattice and coupled with these kind of interactions, have shown a very rich phenomenological scenario concerning both its equilibrium statistical mechanics[2,3] and non-equilibrium dynamical properties[4–6]. In particular, some of these results[5] showed the existence of different types of slow relaxation dynamics when the system is quenched from a high-temperature configuration to a subcritical temperature, depending on the relative strenghts of the dipolar and exchange interactions. This change in the relaxation properties could be an effect of a change in the low temperature equilibrium properties of the system, such as the presence of metastable states. In this work we investigate in detail the low temperature phase diagram of that system in the region where the change in the relaxation properties has been observed.

The above mentioned system is described by the Ising like Hamiltonian

$$H = -\delta \sum_{<i,j>} \sigma_i \sigma_j + \sum_{(i,j)} \frac{\sigma_i \sigma_j}{r_{ij}^3} \tag{1}$$

where the spin variable $\sigma_i = \pm 1$ is located at the site i of a square lattice, the sum $\sum_{<i,j>}$ runs over all pairs of nearest neighbor sites and the sum $\sum_{(i,j)}$ runs over all distinct pair of sites of the lattice; r_{ij} is the distance (in crystal units) between sites i and j; δ represents the quotient between the exchange J_0 and dipolar J_d coupling parameters, where the energy is measured in units of J_d, which is always assumed to be antiferromagnetic ($J_d > 0$). Hence, $\delta > 0$ means ferromagnetic exchange coupling.

The overall features of the the finite temperature phase diagram of this model were described by MacIsaac and coauthors[3] by means of Monte Carlo calculations on 16×16 lattices and analytic calculations of the ground state[7].

They found that the ground state of Hamiltonian (1) is the antiferromagnetic state for $\delta < 0.425$. For $\delta > 0.425$ the antiferromagnetic state becomes unstable with respect to the formation of striped domain structures, that is, to state configurations with spins aligned along a particular axis forming a ferromagnetic stripe of constant width h, so that spins in adjacent stripes are anti-aligned, forming a super lattice in the direction perpendicular to the stripes. They also showed that striped states of increasingly higher thickness h become more stable as δ increases from $\delta = 0.425$. Moreover, they showed that the striped states are also more stable than the ferromagnetic one for arbitrary large values of δ, suggesting such a phase to be the ground state of the model for $\delta > 0.425$. Monte Carlo calculations on finite lattices at low temperature[3,4] gave further support to this proposal, at least for intermediate values of δ. Furthermore, such simulations have shown that striped phases of increasingly higher values of h may become thermodynamically stable at finite temperatures for intermediate values of δ. These results are in agreement with other analytic ones[2,8]. For low values of δ the system presents an antifer-
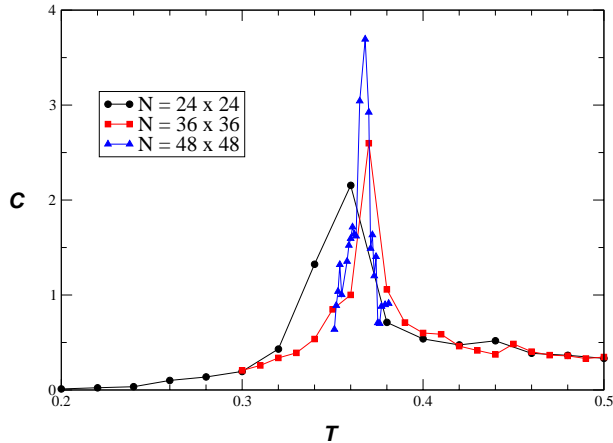
Fig. 1. Specific heat C *vs.* temperature T for $\delta = 1.1$ and different system sizes N.

romagnetic phase at low temperatures. At high temperatures, of course, the system always becomes paramagnetic. Specific heat calculations showed that the transition between the paramagnetic and the striped phases is a second order one[3], while the nature of the transitions between the different striped phases was not clearly determined.

We performed Monte Carlo simulations of Hamiltonian (1) on square lattices up to 48×48 sites using periodic boundary conditions and heat bath dynamics. Our calculations focused on the low temperature region of the (δ, T) phase diagram for values of δ between 0.4 and 2, which includes the transition line between the $h = 1$ (h1) and the $h = 2$ (h2) striped phases.

First we calculated through the energy fluctuations the specific heat C as a function of the temperature for different values of δ and different system sizes up to 48×48 sites. The typical behaviour of C is shown in Fig.1 for $\delta = 1.1$. By considering the peaks in the specific heat we obtained the second order critical line between the paramagnetic and the low temperature ordered phases (h1 and h2). These results (triangles in Fig.4) slightly improve those obtained by MacIsaac and coauthors[3] for 16×16 lattices, thus showing a fast convergence of the critical temperature for increasing sistem sizes, at least for small values of δ.

Next, we analized the transition between the h1 and h2 phases. To do this we introduced the staggered magnetization

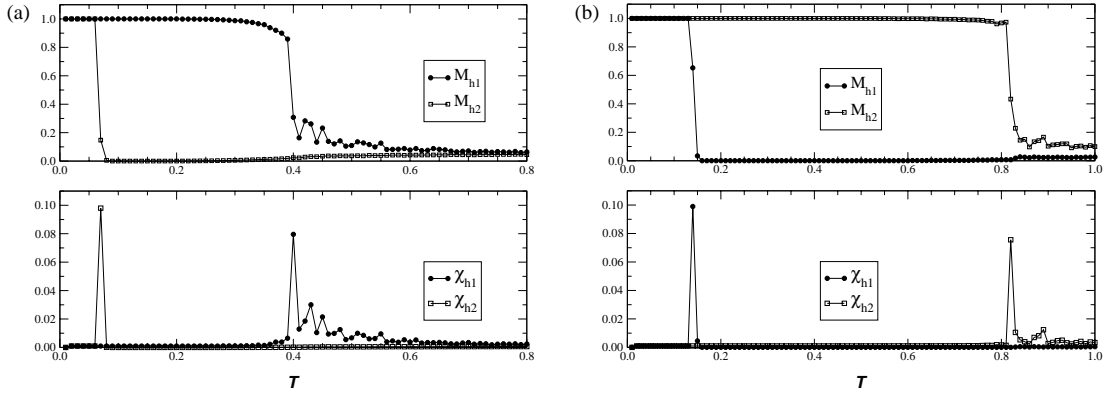$$m_{h1} = \frac{1}{N} \sum_{x,y=1}^{N} (-1)^x \sigma_{xy}, \tag{2}$$

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Fig. 2. Staggered magnetization M_{h1} (M_{h2}) and associated susceptibility χ_{h1} (χ_{h2}) *vs.* T when the system is heated starting at $T = 0$ from an initial configuration in the corresponding ground state h1 (h2), for $N = 24 \times 24$. (a) $\delta = 1$ corresponding to the stable phase h1; (b) $\delta = 2$ corresponding to the stable phase h2.

where N is the total number of lattice sites; m_{h1} equals one in the ground state corresponding to the striped phase with $h = 1$. We calculated the thermal average $M_{h1} \equiv <m_{h1}>$ as a function of T as well as the fluctuations of this quantity through the associated staggered susceptibility

$$\chi_{h1} = \left\langle m_{h1}^2 \right\rangle - \left\langle m_{h1} \right\rangle^2 . \tag{3}$$

In a similar way we defined the staggered magnetization M_{h2} and its associated susceptibility χ_{h2}, corresponding to the striped phase h2. We simulated the heating of the system from $T = 0$ to a temperature higher than the critical one for different values of δ and starting from different initial configurations. To analize the stability of both phases in the different parts of the phase diagram we first calculated the staggered magnetization M_{h1} and susceptibility χ_{h1} starting from a striped configuration h1; we also calculated the staggered magnetization M_{h2} and susceptibility χ_{h2} starting in a striped configuration h2. The typical behaviours are shown in Fig.2 for $\delta = 1$ (Fig.2a), where the ground state corresponds to h1, and for $\delta = 2$ (Fig.2b), where the ground state corresponds to h2. For $\delta = 1$ we see that M_{h1} falls down to zero at the critical temperature while χ_{h1} shows a sharp peak at the same temperature, as expected for the order parameter and its conjugated susceptibility at this type of transition. On the other hand, M_{h2} remains stable up to some temperature $T_2 < T_c$, where it losses stability and falls down abrubtly to zero, while χ_{h2} is zero for almost every temperature, except near $T = T_2$. A similar behavior is observed for $\delta = 2$, but with (M_{h1}, χ_{h1}) and (M_{h2}, χ_{h2}) interchanged. In this case the phase h1 losses stability at some temperature $T_1 < T_c(\delta)$.

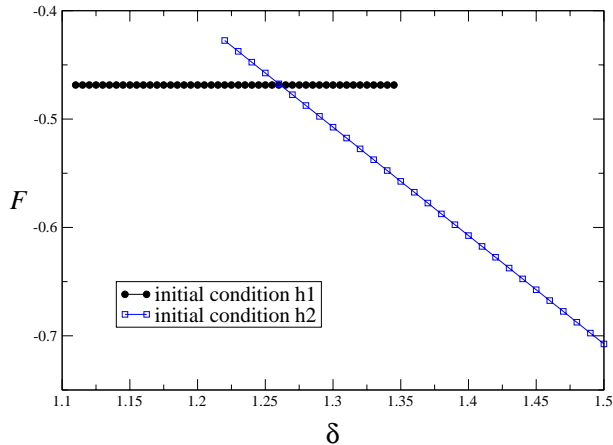All these results show the existence at low temperatures of metastable states

Fig. 3. Free energy F of the phases h1 and h2 *vs.* δ for $T = 0.2$.

h2 in the region of the phase diagram corresponding to the stable phase h1 and *viceversa*. The numerical calculation of stability lines $T_1(\delta)$ and $T_2(\delta)$ is depicted in Fig.4 by means of circles and squares respectively. We see that these lines cross at a certain value $\delta \approx 1.26$ where they join smoothly with the second order critical lines.

All these results suggest the existence of a first order phase transition line separating the phases h1 and h2. To verify this we considered the free energy

$$F = U - T \int_0^T \frac{C(T')}{T'} dT' \tag{4}$$

where $U = \langle H \rangle$. We then calculated the free energy of both phases h1 and h2 by heating the system from $T = 0$ up to a fixed value of temperature T, starting from two different initial configurations corresponding to the ground state of h1 and h2, and for different values of δ. In Fig.3 we see an example of this calculation for $T = 0.2$. The free energy of the phase h1 was calculated for increasing values of δ, starting from some small value well inside the region where this phase is stable, up to the the value of δ corresponding to the stability line of h1 at the given temperature. The same calculation was repeated a the same temperature for the free energy of the phase h2, but for values of δ ranging from the stability line up to some value well inside the region where h2 is stable. The observed continuous change of the minimal free energy from one phase to the other, with a discontinuous change in the slope as δ is varied is a clear evidence of a first order phase transition. Also the multivalued nature of the free energy gives further evidence of the metastable nature of these phases
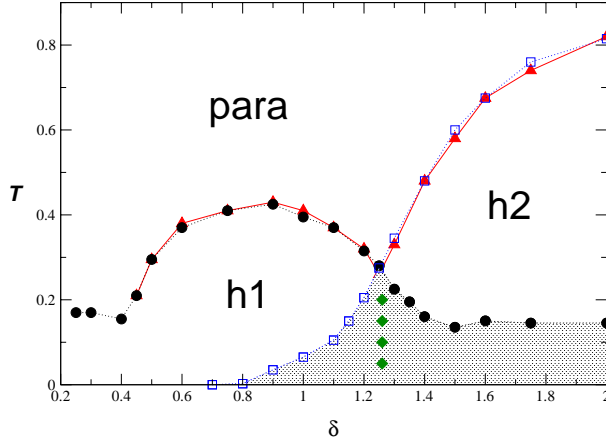
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Fig. 4. Numerical calculation of the phase diagram (δ, T) in the region of parameters under study. Filled triangles correspond to the critical temperatures $T_c(\delta)$ obtained by specific heat calculations for the phase transition between the ordered striped phases h1 and h2 and the paramagnetic one. Filled circles (open squares) correspond to the stability line of the h1 (h2) phase, obtained by analizing the staggered magnetization M_{h1} (M_{h2}). Filled diamonds correspond to the first order transition line between the h1 and h2 phases, obtained by free energy numerical calculations. The shaded region correspond to the presence of metastable states.

in some parts of the phase diagram. Repeating these calculations for different values of T we obtained the almost vertical first order transition line between the two phases, showed in Fig.4 by means of diamonds. The shaded region in Fig.4 indicates the presence of metastable states.

Finally, we considered the relaxation of the system in the metastable region starting from a non-uniform initial configuration. We prepared the system in a configuration with one half of the system in the h1 phase and the other half in the h2 phase, as shown in Fig.5. This particular configuration facilitates the nucleation of the stable phase whatever it be (h1 or h2). In the same figure we show also the time evolution of the energy for two different values of δ located at both sides of the transition line $\delta = 1.26$. We see that the energy evolves towards the mean energy of h1 (E_{h1}) at the corresponding temperature when $\delta < 1.26$, while it relaxes to the mean energy of h2 (E_{h2}) when $\delta > 1.26$. This shows that indeed the metastable configurations are unstable agains the nucleation of the corresponding stable phase.
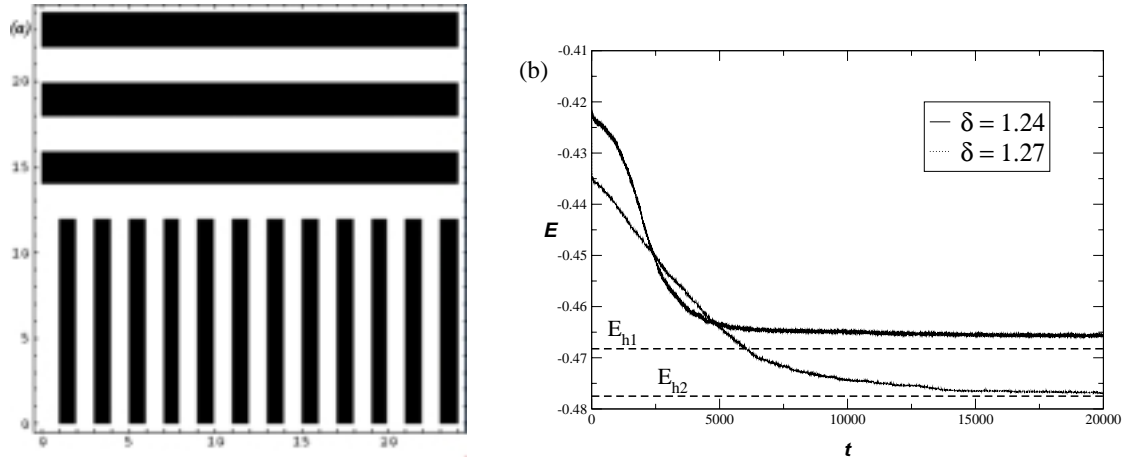
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Fig. 5. (a) Initial configuration with 50% in the ground state corresponding to h1 and 50% in the ground state corresponding to h2. (b) Time evolution of the energy of the system E starting from the initial configuration of figure (a), for $T = 0.2$ and $N = 24 \times 24$.

Summarizing, we presented numerical evidence of the existence of metastable states in the model described by Hamiltonian (1) in the low temperature region of the (δ, T) phase diagram where the transition between h1 and h2 phases takes place. We also showed that these metastable states are associated with a first order phase transition line between both phases. The presence of these metastable states may alter the normal domain growth process when the system is quenched from high temperatures into the ordered phase, by pinning the walls of the stable phase domains. Such process could slow down the normal coarsening dynamics, depending on whether the quench drops the system into the metastable region or not, thus explaining the observed change in the relaxation dynamics in this region of the phase space[5]. Some work along this line is in progress and will be published elsewhere. Another point that deserves further investigation is the possible presence of more complex metastable states for higher values of δ, as observed in a related three-dimensional model with competing nearest neighbors ferromagnetic interactions and long-range antiferromagnetic Coulomb-like interactions[9].

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