Simulation of the Reorientation Transition in Ultrathin Magnetic Films with Striped and Tetragonal Phases

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(Received 12 August 1997)

We show, using a model of an ultrathin magnetic film, that when the dipole-dipole and exchange interactions are of comparable strength there exists a reorientation transition from a planar ferromagnetic phase at low temperature to either an orientationally ordered striped phase or a tetragonal phase at higher temperature. Which of these transitions occurs depends on the relative strength of the magnetic surface anisotropy. The phase diagram, as found from extensive Monte Carlo simulation, is determined as a function of temperature and magnetic surface anisotropy. [S0031-9007(97)05024-2]

PACS numbers: 75.70.–i, 75.40.Mg, 75.60.Ch

Magnetic thin films exhibit a reorientation transition from an ordered state with out-of-plane magnetization to an ordered state with in-plane magnetization both as temperature increases and as the thickness of the film increases [1–4]. The origin of the reorientation transition is believed to be the competition between the magnetic surface anisotropy (MSA), which favors an out-of-plane orientation for the magnetic moments, and the dipolar interaction which favors an in-plane orientation for the moments. Competition between the short-ranged exchange interaction and the long-ranged dipolar interactions also contributes to the richness of the phase diagram. The competition between these two interactions results in striped and tetragonal phases when the magnetic moments are oriented in the out-of-plane direction (i.e., at large MSA) [5]. Both of these phases have been observed experimentally in systems which exhibit a reorientation transition [1,3].

In this Letter the phase diagram for a magnetic monolayer with a dipolar interaction of similar strength to that of the exchange interaction is derived from extensive Monte Carlo simulation. The Hamiltonian $\mathcal{H}$ for the model system can the written as

$$\mathcal{H} = -J \sum_{\langle \vec{r}, \vec{r}' \rangle} \vec{S}(\vec{r}) \cdot \vec{S}(\vec{r}') - K \sum_{\vec{r}} |S^z(\vec{r})|^2 + g \sum_{\vec{r}, \vec{r}', \alpha, \beta} S^\alpha(\vec{r}) \Gamma^{\alpha\beta}(|\vec{r} - \vec{r}'|) S^\beta(\vec{r}') . \quad (1)$$

J, K, and g are the strengths of the exchange interaction, the magnetic surface anisotropy, and the dipolar interaction, respectively. $\vec{S}(\vec{r})$ is a classical vector of unit magnitude representing the magnetic moment at position $\vec{r}$ and $\Gamma^{\alpha\beta}(|\vec{r} - \vec{r}'|)$ is the dipolar interaction.

A number of authors have studied this Hamiltonian in the limit where $J/g \gg 1$ [6–13]. The phase diagram predicted in Ref. [8] is shown schematically in Fig. 1(a) and consists of a perpendicular ferromagnetic phase in region I, a planar ferromagnetic phase in region II, and a paramagnetic phase in region III. At the phase boundary separating the two ordered phases, a first order reorientation transition occurs. We note that the reorientation transition temperature increases with increasing K/g. While it is generally accepted that the dipolar interaction plays a critical role in determining the nature of the reorientation transition, the fact that it destabilizes the perpendicular ferromagnetic phase [14,15] has not been generally acknowledged or taken into account in these previous studies.

The other limit studied in the literature is the case $J/g = 0$ [16]. The resultant phase diagram is shown schematically in Fig. 1(b). While this phase diagram is similar in many aspects to that of Fig. 1(a), the ordered phases are antiferromagnetic and, somewhat surprisingly, the reorientation transition temperature decreases with increasing K/g. This implies that the reorientation transition is from the planar phase at low temperature to the perpendicular phase at higher temperature. A similar reorientation

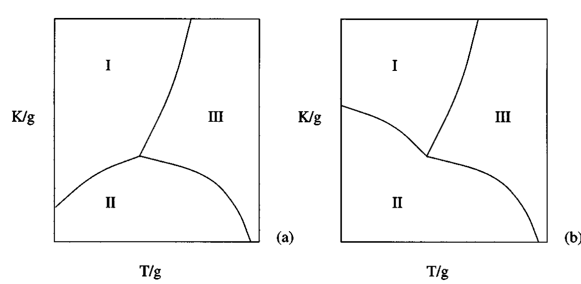

FIG. 1. Schematic phase diagrams for the previously treated cases when $J/g \gg 1$ (a) and $J/g = 0$ (b). Region I is ordered out of plane, region II is ordered in plane, and region III is paramagnetic or tetragonal.

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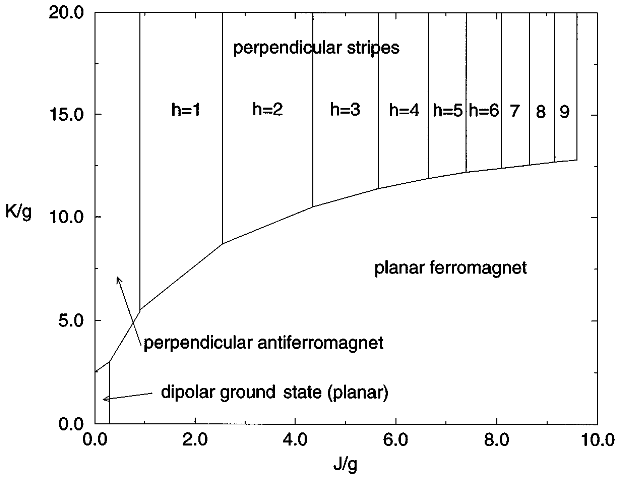

FIG. 2. The regions where the various in-plane and out-of-plane phases have the lowest energy. For regions where a perpendicular striped phase is the lowest energy phase the stripe width h has been indicated.

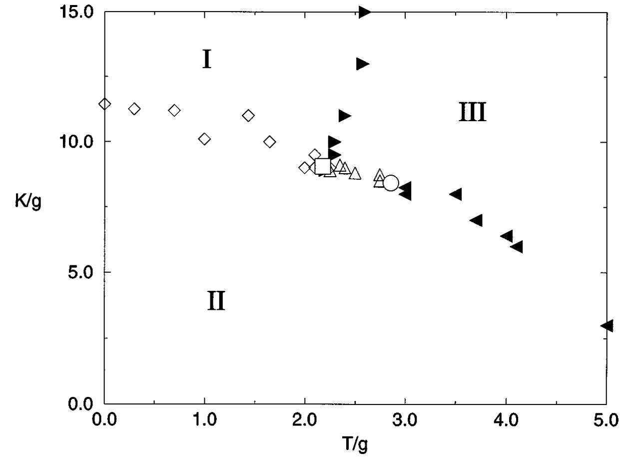

FIG. 3. The phase diagram for $J/g = 6$, on a $N = 40 \times 40$ lattice, as a function of temperature and magnetic surface anisotropy. Region I is ordered out of plane, region II is ordered in plane, and region III is paramagnetic or tetragonal. Solid symbols indicate continuous transitions, while unfilled symbols show first order transitions. The locations of the tricritical point and the triple point are shown by the opaque circle and square, respectively. The error in the points along the second order transition lines is approximately the size of the symbol, while the error associated with the points along the first order transition lines is larger due to hysteresis effects.

transition has been observed experimentally in Ni(001) on Cu(001), although the transition is believed to be the result of the temperature dependence of both the surface and volume anisotropy [17].

We consider the phase diagram for a finite value of J/g. In this case the presence of the striped and tetragonal phases, when the moments are oriented out-of-plane, is an essential feature of the phase diagram. At zero temperature the phase diagram in the $(K/g, J/g)$ plane may be determined from energy calculations based on methods previously reported in the literature [14]. This diagram is shown in Fig. 2.

To consider finite temperature effects, we use Monte Carlo simulations to determine the phase diagram in the $(K/g, J/g)$ plane for $J/g = 6$. The simulations are performed on a $N = 40 \times 40$ square lattice using Glauber dynamics. Simulations have been performed on other system sizes, both smaller and larger, but no detailed finite size analysis has been done. A typical simulation consists of 10^6 Monte Carlo steps per site, of which the first 10% are used to equilibrate the system. This system has a ground state of out-of-plane stripes of width four atomic spacings for all $K/g > 11.44 \pm 0.01$. The long-range nature of the dipole-dipole interaction has been treated using Ewald summation techniques [14].

The phase diagram is shown in Fig. 3. It consists of three distinct regions. In region I the spins are aligned perpendicular to the plane of the film in stripes of width 4. The stripes display a high degree of orientational order and are aligned along either the x or the y axis of the underlying square lattice. In region II the spins are in the planar ferromagnetic phase. Region III is somewhat more complex. Although region III does not manifest any discernible magnetic ordering, depending on the value of K/g, the spins can be aligned either parallel or perpendicular to the plane of the film. Also for large values of K/g ($K/g > 8.4 \pm 0.1$) the system exhibits some magnetic structure in the form of orientationally disordered stripes similar to those discussed by Booth *et al.* [5] for the dipolar Ising model. Figures 4–7 show the perpendicular component of the magnetic moment for typical configurations in each of these regions. The vector field (S^x, S^y) is shown in Fig. 5 for the same configuration as shown in Fig. 4(a).

The phase boundaries separating the three different phases join at a triple point ($K/g = 9.1 \pm 0.1$ and $T/g = 2.2 \pm 0.1$ for our $N = 40 \times 40$ system) and show a number of intriguing properties. The phase boundary separating regions I and III consists of a line of second order phase transitions that are characterized by the loss of the orientational order of the stripes. This is qualitatively similar to the results presented by Booth *et al.*, and is best described in terms of an orientational order parameter

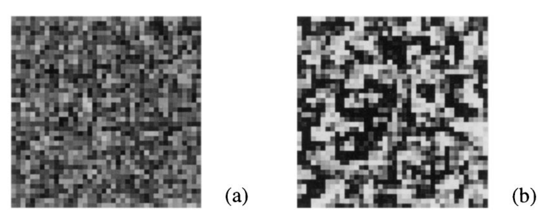

FIG. 4. A typical spin configuration for $K/g = 7.00$ with $T/g = 2.00$ (a) and $T/g = 4.00$ (b). The figure shows the S^z component of the magnetic moments using a linear gray scale where $S^z = 1$ is black and $S^z = -1$ is white.

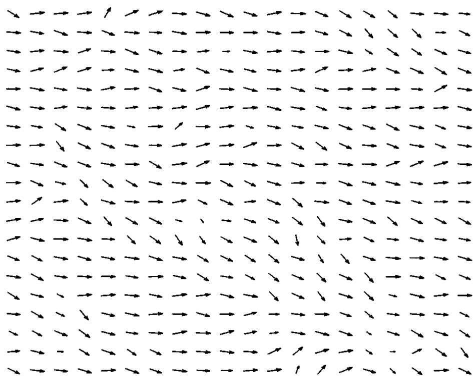

FIG. 5. The vector field (S^x, S^y) for the upper left quarter (20×20 spins) of the spin configuration shown in Fig. 4(a).

defined as

$$O_{hv} \equiv \left| \frac{n_h - n_v}{n_h + n_v} \right|, \qquad n_{h/v} \equiv \sum_{\vec{r}} |S^z(\vec{r}) - S^z(\vec{r} + \hat{x}/\hat{y})|. \tag{2}$$

This order parameter has a value of 1 when the stripes are completely oriented along the x or the y axis (smectic phase), and drops to zero in the orientationally disordered phase (tetragonal phase).

As in earlier work on the dipolar Ising model [5], we find no evidence for a nematic phase. As well, while the specific heat displays a distinct shoulder above the phase boundary for large values of K/g, there does not appear to be any evidence for a sharp transition from the tetragonal to the paramagnetic phase. However, unlike the dipolar Ising model, the orientational order parameter does exhibit a linear dependence on temperature, rather than complete saturation at low temperature. Presumably this is due to spin-wave excitations.

The phase boundary separating the two ordered phases (region I and region II), describes a reorientation transition. It consists of a line of first order transitions and like the pure dipolar case studied earlier the reorientation temperature decreases with increasing K/g. This implies that the transition is from the planar ferromagnetic phase at low temperature to the striped phase at higher temperature.

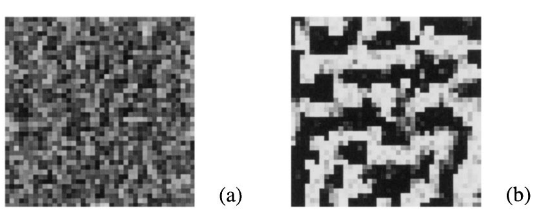

FIG. 6. A typical spin configuration for $K/g = 8.70$ with $T/g = 1.40$ (a) and $T/g = 2.60$ (b). The figure shows the S^z component of the magnetic moments using a linear gray scale where $S^z = 1$ is black and $S^z = -1$ is white.

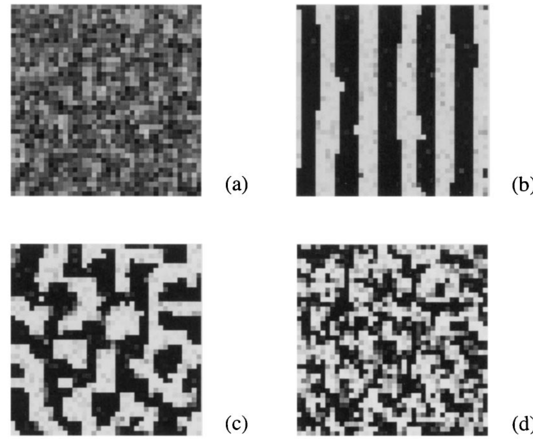

FIG. 7. A typical spin configuration for $K/g = 10.00$ with $T/g = 0.90$ (a), $T/g = 1.90$ (b), $T/g = 2.90$ (c), and $T/g = 5.90$ (d). The figure shows the S^z component of the magnetic moments using a linear gray scale where $S^z = 1$ is black and $S^z = -1$ is white.

The two phase boundaries combine to produce the temperature dependence of the orientational order parameter shown in Fig. 8 for various values of K/g. For $K/g > 11.44 \pm 0.01$ we see O_{hv} dropping continuously with increasing temperature, to (almost) zero above the phase boundary. For $8.4 \pm 0.1 < K/g < 11.44 \pm 0.01$, we see O_{hv} initially zero at low temperature, rising to finite values as the system undergoes a reorientation transition from the planar ferromagnetic phase to the striped phase, and then dropping continuously to (almost) zero above a critical temperature, were the system enters the tetragonal phase. To the best of our knowledge these are the first simulations which display a reorientation transition to a striped phase.

The phase boundary separating the planar ferromagnetic phase in region II from the paramagnetic phase in region III is also of interest. The results of our simulations

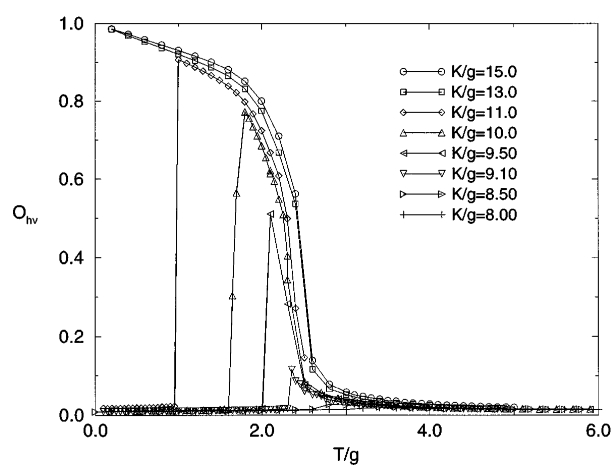

FIG. 8. The orientational order parameter for various values of K/g as found by slowly warming the system from low temperature. The lines connect successive points.

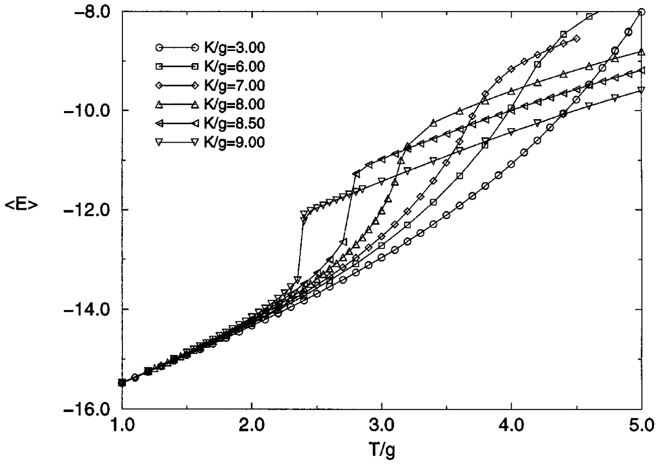

FIG. 9. The average energy for various values of K/g as found by slowly warming the system from low temperature. The lines connect successive points.

indicate that, along the portion of the phase boundary just above the triple point, the phase transition from the planar ferromagnetic phase to the paramagnetic (or tetragonal phase) is first order. However, the latent heat associated with the transition appears to decrease as T increases, dropping to zero at $T = 2.9 \pm 0.2$, $K/g = 8.4 \pm 0.1$. This suggests the existence of a tricritical point along the phase boundary. This is most clearly seen in Fig. 9, which shows the energy as a function of temperature for several values of K/g, and which shows evidence of a discontinuity in the average energy when $9.1 \pm 0.1 < K/g < 8.4 \pm 0.1$.

Visual inspection of spin configurations at temperatures just to the right of the first order line (indicated in the phase diagram by open triangles), reveals that the spin configurations are predominately out of plane and tetragonal in this region. Thus, the transitions at this critical line are first order reorientation transitions, directly from the ferromagnetic, in-plane phase to the tetragonal, out-of-plane phase.

In summary, our present Letter shows that in systems which exhibit striped and tetragonal phases, a reorientation transition can occur from the ferromagnetic in-plane phase to the out-of-plane striped phase. A further increase in temperature eventually results in a transition from the striped phase to the tetragonal phase as occurs in uniaxial out-of-plane systems. However there also exists a range of MSA parameter values for which the reorientation transition is directly from the ferromagnetic in-plane phase to the tetragonal out-of-plane phase.

Although the precise location of the phase boundaries shown in Fig. 3 will vary with system size, we expect that quantitative features will be correct for larger systems with the underlying square lattice. In systems with dipolar interactions, the lattice structure plays a significant role in determining the effective anisotropy. In particular for the planar orientation the existence of a low temperature ordered phase may depend on the symmetry of the lattice [18,19].

The authors thank Memorial University of Newfoundland, HPCNET, and Digital Equipment Corporation for the use of computational resources. The authors also acknowledge useful conversations with Dr. P. H. Poole and Dr. T. Lookman. This research is supported, in part, by the Natural Science and Engineering Research Council of Canada.

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