Phase Diagram for a Magnetic Thin Film with Dipolar Interactions and Magnetic Surface Anisotropy

A. B. MacIsaac* and J. P. Whitehead
Department of Physics, Memorial University of Newfoundland, St. John's, Newfoundland, Canada A1B 3X7

K. De'Bell
Department of Physics, Trent University, Peterborough, Ontario, Canada K9J 7B8

P. H. Poole
Department of Applied Mathematics, University of Western Ontario, London, Ontario, Canada N6A 5B7
(Received 7 March 1996)

The phase diagram of a model thin film with long-range dipole-dipole interactions and magnetic surface anisotropy is determined as a function of temperature and the surface anisotropy. We show that the model studied here exhibits a reorientation transition in the absence of an exchange interaction. Further we show that, unexpectedly, the transition is from an in-plane phase to an out-of-plane phase with increasing temperature, which is the opposite of the ferromagnetic case. The implications of this study for the full phase diagram of a model which includes both the dipolar and exchange interactions are discussed. [S0031-9007(96)00552-2]

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

Recent advances in experimental techniques for creating and studying magnetic thin films have led to a great deal of interest in the critical properties of magnetic monolayers and thin films [1–3]. Much of the theoretical work has focused on the properties of two-dimensional, ferromagnetic systems with a dominant short-range exchange interaction and magnetic surface anisotropy (MSA), as well as, a much weaker, long-range dipolar interaction [4–6]. The general Hamiltonian for such a system is

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

where J is the exchange parameter, g is the strength of the dipolar interaction, and K is the magnetic surface anisotropy. $\Gamma^{\alpha\beta}(\vec{r} - \vec{r}')$ is the dipolar interaction given by

$$\Gamma^{\alpha\beta}(\vec{r} - \vec{r}') = \frac{\delta_{\alpha\beta}}{|\vec{r} - \vec{r}'|^3} - 3\frac{(r'^{\alpha} - r^{\alpha})(r'^{\beta} - r^{\beta})}{|\vec{r} - \vec{r}'|^5}, \quad (2)$$

and $\vec{S}(\vec{r})$ defines the magnetic dipole moment at $\vec{r}$. Repeated indices imply a summation over components. This model is expected to apply to systems such as Fe on the Cu(100) surface studied recently by Allenspach and Bischof [2]. Experimentally this system exhibits a reorientation transition in which the magnetization changes direction from perpendicular to the plane (out of plane) at low temperature to parallel to the film (in plane) at higher temperature. A similar transition has been observed in several other systems [1,3,7]. While the reorientation transition has been studied analytically using mean field theory [4,8,9], renormalization group analysis [5,6,10], and Monte Carlo simulation [6,11,12], there are a number of aspects of the reorientation transition that are poorly understood.

In this Letter we study the reorientation transition for the case of a dipolar interaction and magnetic surface anisotropy, but no exchange interaction ($J = 0$). Our reasoning for excluding the exchange interaction is that while the dipolar interaction is generally much weaker than the exchange interaction in real systems, it plays a crucial role in determining the phase behavior and morphology of the magnetic ordering. In the case of the planar system ($K \to -\infty$), the anisotropic character of the dipolar interaction serves to stabilize the magnetic order at finite temperature [13,14]. In the case of the Ising system ($K \to +\infty$), the competition between the antiferromagnetic character of the long-range, dipolar interaction and the ferromagnetic, short-range exchange interaction gives rise to striped phases [15–17]. These phases have been observed experimentally [2] and are the subject of considerable theoretical interest. In addition, there are a number of quasi-two-dimensional magnetic systems in which the exchange interaction is of comparable magnitude to the dipolar interaction [18] and which can exhibit a relatively weak magnetic anisotropy [19]. The pure dipolar model is of obvious relevance to such systems.

Our results for this pure dipolar system also provide an interesting contrast to the case of large finite values

 © 1996 The American Physical Society

of J/g and have important implications for the complete phase diagram of the Hamiltonian in Eq. (1). Previous work on systems which interact only via an exchange interaction ($g = 0$, $K = 0$) show that the spins do not order for any nonzero temperature [20]. The addition of the MSA leads to Ising-like ordering for $K > 0$ as shown by Bander and Mills [21] using renormalization group (RG) arguments and by Erickson and Mills [22] using Monte Carlo simulation. More recent RG work by Pescia and co-workers [5] and Monte Carlo work by Hucht, Moschel, and Usadel [11] have treated the combination of dipolar and exchange, but considered the limit $J/g \gg 1$. Both sets of authors predict the existence of a reorientation transition from an out-of-plane ferromagnetic state at low temperature to an in-plane ferromagnetic state as the temperature increases for some ratios of K/g. Unfortunately neither work fully considers the effect of stripe phases, which have been shown to be integral to any understanding of the perpendicular phases. The limiting cases $K = +\infty$, taking into proper account the stripe phases [17], and $K = -\infty$ [14] have led to a number of interesting observations, some implications of which will be discussed below.

We study the Hamiltonian in Eq. (1) with $J = 0$ on a square lattice as a function of the ratio K/g. We assume that the $\hat{z}$ direction is perpendicular to the thin film, and that $\hat{x}$ and $\hat{y}$ denote the unit lattice vectors that define the square lattice of the system. From previous studies we know that the ground state for $K/g = 0$ corresponds to an antiferromagnetic state in which the spins lie in plane [14]. For the square lattice this state is highly degenerate and defines a continuous manifold of states characterized in terms of the orientation of the order parameter defined as

$$\vec{M}_{xy} = \frac{1}{N}\sum_{\vec{r}}[(-1)^{r^y} S^x(\vec{r})\hat{x} + (-1)^{r^x} S^y(\vec{r})\hat{y}]. \qquad (3)$$

This degeneracy is lifted at finite temperature and the dependence of the free energy on the orientation of the order parameter exhibits the fourfold symmetry of the underlying square lattice [14]. As the anisotropy is increased the energy of the in-plane phase increases relative to the phase in which the spins order antiferromagnetically perpendicular to the plane, and at $K/g = 2.44 \pm 0.01$ the ground state switches to this out-of-plane phase. This ground state is characterized by the order parameter M_z, defined as

$$M_z = \frac{1}{N}\sum_{\vec{r}}(-1)^{r^x + r^y} S^z(\vec{r}). \qquad (4)$$

The finite temperature phase diagram, found using Monte Carlo simulation, is given in Fig. 1. In region I the magnetic order parameter is characterized by a nonzero value for the out-of-plane, sublattice magnetization, M_z. The magnetic order in this phase may be described by the ensemble average of the magnitude of M_z, $M_\perp \equiv \langle |M_z| \rangle$.

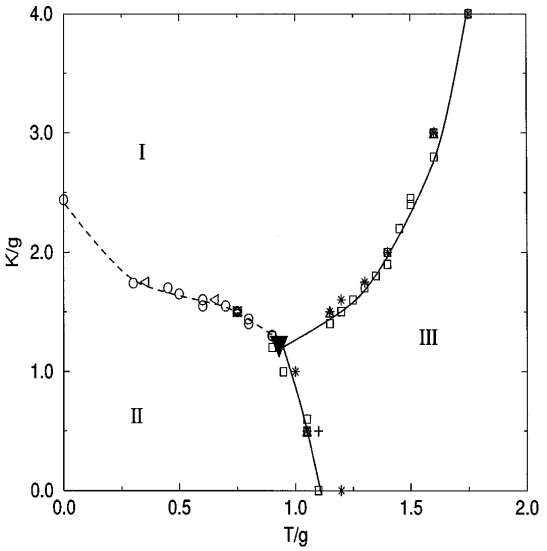

FIG. 1. Phase diagram obtained from Monte Carlo simulation. Region I is ordered out of plane, region II is ordered in plane, and region III is paramagnetic. The dashed line is a guide to the eye highlighting the line of first-order reorientation transitions between the two ordered states. The solid lines are guides to the eye highlighting the two lines of second-order transitions from the paramagnetic state to one of the two ordered states. Points on each line are coded based on system size: $N = 16^2$ ($\bigcirc, \square$), 24^2 ($\diamond, \triangle$), 32^2 ($\triangleleft, *$), and 40^2 ($\triangleright, +$).

In region II the magnetic order is characterized by a nonzero value for the in-plane, sublattice magnetization, $\vec{M}_{xy}$. The magnetic order may be described by the ensemble average of the magnitude of $\vec{M}_{xy}$, $M_\parallel \equiv \langle |\vec{M}_{xy}| \rangle$.

The phase boundary separating region I and region II from the high temperature paramagnetic phase, region III, is shown in Fig. 1 as a solid line. The nature of the transition can be seen in Figs. 2 and 3, where we have

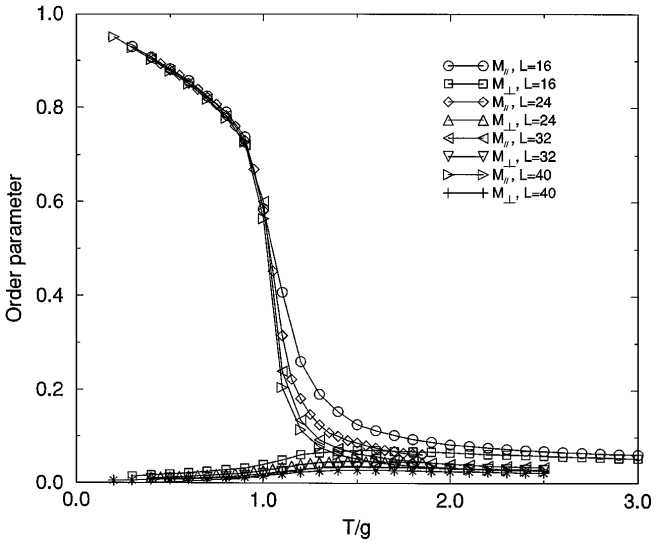

FIG. 2. Variation of the order parameters for $K = 0.50$ for various size systems.

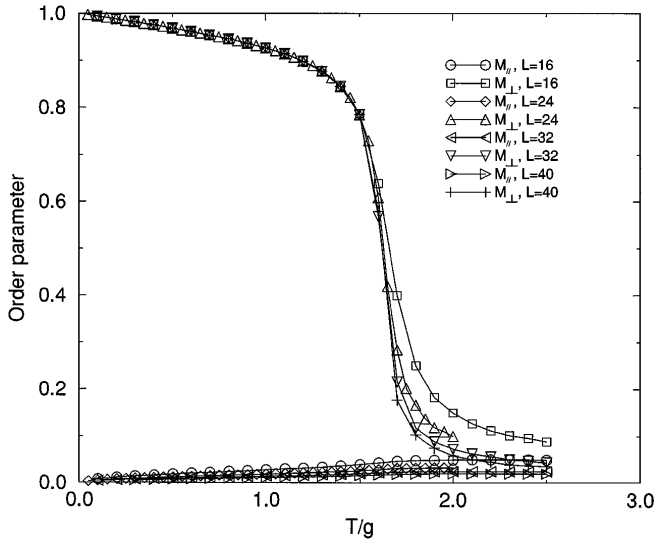

FIG. 3. Variation of the order parameters for $K = 3.00$ for various size systems.

plotted both order parameters as a function of temperature for $K/g = 0.5$ and $K/g = 3.00$, respectively. In both situations the order parameters are effectively zero at high temperature. As the temperature decreases one order parameter remains zero while the other acquires a nonzero value at a well defined transition temperature. As the system size increases we can see that the transition sharpens in a manner consistent with finite size effects at a continuous transition. This is consistent with results in the limiting cases of $K/g = \pm\infty$, where a continuous transition was reported [14,16].

The phase boundary separating region I and region II is shown in Fig. 1 as a dashed line. Along this line we have the reorientation transition between the two ordered states. The nature of this transition is distinct from that between the paramagnetic phase and the ordered phases. Figure 4 shows both order parameters, defined above, as functions of the temperature for constant $K/g = 1.50$ for a system of size $N = 40 \times 40$. At high temperature the system is paramagnetic and both order parameters are zero. As the temperature is lowered the system orders first in the out-of-plane phase, and then at a lower temperature the system switches into the in-plane phase. In Fig. 5 we have expanded the region of the reorientation transition in Fig. 4 and included results for other system sizes. We can see that the reorientation transition lacks the finite size effects that one generally associates with a continuous transition. As well the Monte Carlo simulations show that there is considerable hysteresis associated with both the order parameter and the average energy at the reorientation transition. The temperature dependence of the specific heat also shows strong indications that the reorientation transition is a first-order transition.

This reorientation transition is novel as it is a reorientation from an in-plane phase at low temperature to an out-of-plane phase as the temperature is raised. The role of the two phases is reversed from that seen in the studies of ferromagnetic systems. To illustrate Fig. 6 shows schematically the two phase diagrams. For the model studied here, shown in Fig. 6(a), the sequence of phases as temperature is raised is in plane → out of plane → paramagnetic. For the exchange dominated ferromagnetic systems, the sequence is out of plane → in plane → paramagnetic, as indicated in Fig. 6(b). Related to this is the slope of the coexistence line, $\partial K_c/\partial T$. The slope is negative for the model discussed here, but positive in the case of a dominate ferromagnetic exchange in-

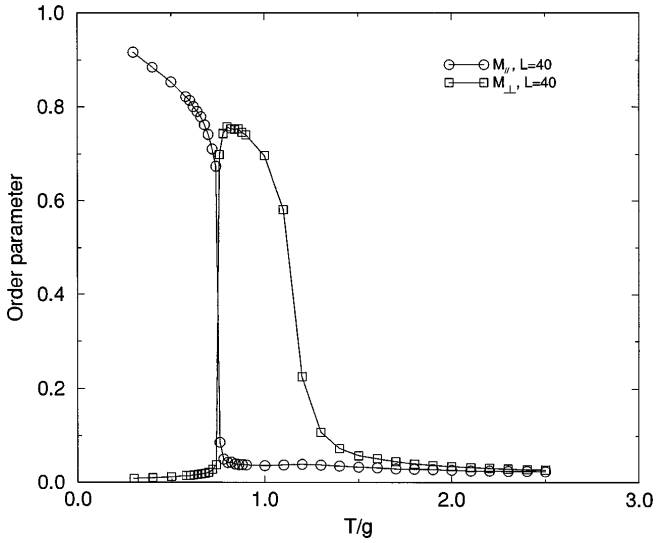

FIG. 4. Variation of the order parameters for $K = 1.50$ for a system of size $N = 40 \times 40$.

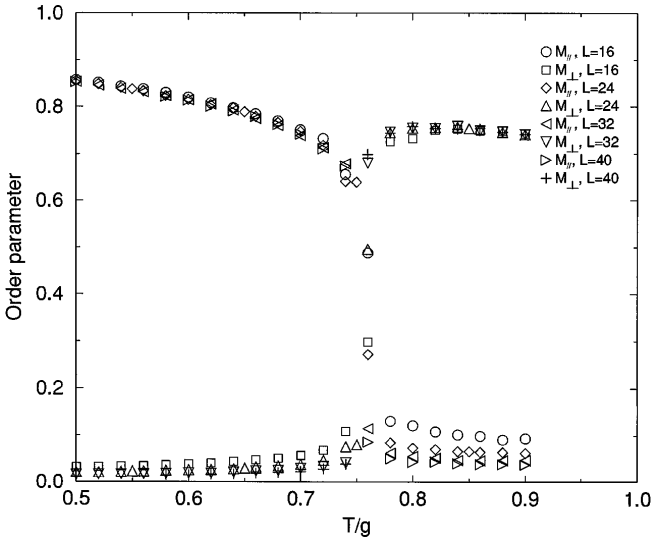

FIG. 5. Variation of the order parameters for $K = 1.50$ for various size systems, near the reorientation transition. All data shown were obtained by starting at high temperature and slowly cooling the system.

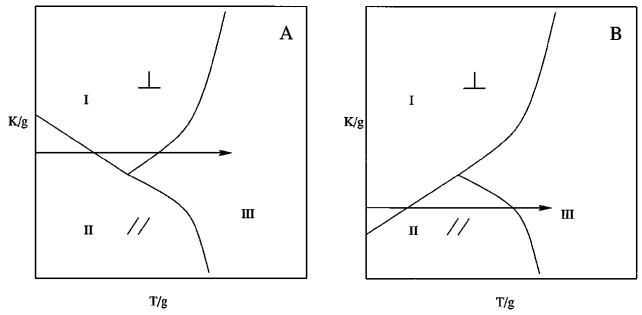

FIG. 6. Schematic phase diagram for no exchange interaction (a) and for an exchange dominated ferromagnet (b).

teraction. The question which remains is: What is the topology of the first-order surface, when one includes the relative strength of the exchange interaction, J/g, in the phase diagram? This is not a trivial problem as the lowest energy state for both the in-plane and out-of-plane orientations changes as a function of this ratio [16,23]. We hope to address this question in future work.

The lines of continuous transitions and the first-order line appear to meet at a single point. Such a point is by definition a tricritical point. Within the errors associated with the finite size of the systems and the statistical errors present, it can be determined that the tricritical point is at $T \equiv T_c \approx 0.9 \pm 0.1$ and $K \equiv K_c \approx 1.2 \pm 0.1$. This point is indicated on the phase diagram by a large triangle.

In summary, the phase diagram for a two-dimensional magnetic monolayer with a dipolar interaction and a magnetic surface anisotropy has been determined as a function of the ratio K/g and temperature. The phase diagram consists of three phases: a paramagnetic phase, an ordered phase with the moments in the plane of the film, and an ordered phase with the moments perpendicular to the film. The three phases coexist at a tricritical point. This is the point where the line of first-order transitions meets the two lines of second-order transitions. Comparisons of this phase diagram to that predicted for a system with a dominant ferromagnetic exchange interaction shows some similarities, such as the existence of a first-order reorientation transition, but there are interesting differences. In particular, the sign of the slope of the coexistence line between the two ordered phases is different; thus, the roles played by the lowest energy parallel phase and the lowest energy perpendicular phase in the reorientation transition are reversed. It will be interesting to see how the phase diagram changes as a function of J/g, when proper consideration of the striped phases is included in the analysis. This problem will be the subject of future work.

This work was supported by the Natural Sciences and Engineering Council of Canada (NSERC). The authors would like to thank the Pittsburgh Supercomputing Center and the Digital Equipment Corporation for the use of their Alpha 8400 5/300. One of the authors (A. B. M. I.) thanks the High Performance Computing Centre (HPCC) and Fujitsu Canada Inc. for the use of the Fujitsu VPX240 as part of the HPCC scholarship program.

*To whom correspondence should be addressed. Electronic address: allanb@smaug.physics.mun.ca

[1] R. Allenspach, M. Stampanoni, and A. Bischof, Phys. Rev. Lett. **65**, 3344–3347 (1990).
[2] R. Allenspach and A. Bischof, Phys. Rev. Lett. **69**, 3385–3388 (1992).
[3] D. P. Pappas, K.-P. Kämper, and H. Hopster, Phys. Rev. Lett. **64**, 3179–3182 (1990).
[4] A. Moschel and K. D. Usadel, Phys. Rev. B **49**, 12 868–12 871 (1994).
[5] D. Pescia and V. L. Pokrovsky, Phys. Rev. Lett. **65**, 2599–2601 (1990); **70**, 1185–1185 (1993); Paolo Politi, Angelo Rettori, and Maria Gloria Pini, Phys. Rev. Lett. **70**, 1183–1183 (1993); A. P. Levanyuk and N. Garcia, Phys. Rev. Lett. **70**, 1184–1184 (1993).
[6] S. T. Chui, Phys. Rev. B **50**, 12 559–12 567 (1994).
[7] Z. Q. Qiu, J. Pearson, and S. D. Bader, Phys. Rev. Lett. **70**, 1006–1009 (1993).
[8] A. Moschel and K. D. Usadel, Phys. Rev. B **51**, 16 111–16 114 (1995).
[9] A. Moschel and K. D. Usadel, J. Magn. Magn. Mater. **140–144**, 649–650 (1995).
[10] K. Ried, Y. Millev, M. Fähnle, and H. Kronmüller, Phys. Rev. B **51**, 15 229–15 249 (1995).
[11] A. Hucht, A. Moschel, and K. D. Usadel, J. Magn. Magn. Mater. **148**, 32, 33 (1995).
[12] A. Hucht and K. D. Usadel (to be published).
[13] Sona Prakash and Christopher L. Henley, Phys. Rev. B **42**, 6574–6589 (1990).
[14] K. De'Bell, A. B. MacIsaac, I. N. Booth, and J. P. Whitehead (to be published).
[15] T. Garel and S. Doniach, Phys. Rev. B **26**, 325–329 (1982).
[16] A. B. MacIsaac, J. P. Whitehead, M. C. Robinson, and K. De'Bell, Phys. Rev. B **51**, 16 033–16 045 (1995).
[17] I. N. Booth, K. De'Bell, A. B. MacIsaac, and J. P. Whitehead, Phys. Rev. Lett. **75**, 950–953 (1995).
[18] A. B. MacIsaac, J. P. Whitehead, K. De'Bell, and K. Sowmya Narayanan, Phys. Rev. B **46**, 6387–6394 (1992), and references therein.
[19] J. P. Whitehead and K. De'Bell, J. Phys. Condens. Matter **6**, L731–L734 (1994), and references therein.
[20] N. D. Mermin and H. Wagner, Phys. Rev. Lett. **17**, 1133–1136 (1966).
[21] Myron Bander and D. L. Mills, Phys. Rev. B **38**, 12 015–12 018 (1988).
[22] R. P. Erickson and D. L. Mills, Phys. Rev. B **43**, 11 527–11 530 (1991).
[23] A. B. MacIsaac, J. P. Whitehead, M. C. Robinson, and K. De'Bell, Physica (Amsterdam) **194B–196B**, 223–224 (1994).