

Theoretical simulation of the anisotropic phases of antiferromagnetic thin films

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We simulate antiferromagnetic thin films. Dipole-dipole and antiferromagnetic exchange interactions as well as uniaxial and quadrupolar anisotropies are taken into account. Various phases unfold as the corresponding parameters J , D , and C , as well as the temperature T and the number n of film layer, vary. We find (i) how the strength Δ_ν of the anisotropy arising from dipole-dipole interactions varies with the number of layers ν away from the film's surface, with J and with n ; (ii) a unified phase diagram for all n -layer films and bulk systems; (iii) a layer-dependent spin reorientation (SR) phase in which spins rotate continuously as T , D , C , and n vary; (iv) that the ratio of the SR to the ordering temperature depends (approximately) on n only through $(D + \Delta/n)/C$, and hardly on J ; (v) a phase transformation between two different magnetic orderings, in which spin orientations may or may not change, for some values of J , by varying n .

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I. INTRODUCTION

Magnetic thin films are attracting much interest. Some of it derives from applications (in electronics¹) of ferromagnetic and antiferromagnetic layered structures, where bias hysteresis arises from interactions at the interfaces of film² and in nanoparticle layers.³ Knowledge of the nature of the magnetically ordered states, as well as of the transitions between them, is important. The spin reorientation (SR) transition is most interesting. Continuous SR transitions, in which the direction of the magnetization changes continuously with temperature, were first observed in bulk ferrimagnets⁴ and in canted spin antiferromagnets (AF's).⁵ Discontinuous SR transitions were first discovered in the bulk, in AF's⁶ and in ferromagnets.⁷

Competition of various magnetic anisotropies play decisive role in SR. Simply put, minimization of the energy with respect to direction of the magnetization m (or some staggered magnetization m_s for AF's) gives the physical direction of m at very low temperatures. By proper choice of the anisotropy, the energy minimum can be controlled, and thus the direction of m . Furthermore, in a Ginzburg-Landau-like theory, the anisotropy constants can be made to vary with temperature, and thus the direction of m . This approach was first use by Horner and Varma^{8,9} for continuous SR. Mean-field as well as Monte Carlo (MC) calculations also give continuous SR in the bulk.¹⁰ Discontinuous SR, on the other hand, occurs when one local minimum in the free energy, for some spin direction, suddenly (as, for instance, the temperature varies) becomes the global minimum, at the expense of another local minimum, for another spin direction.

For films, thermally driven SR transitions, whose nature (continuous or discontinuous) was not clearly established, were first reported for ferromagnets by Pappas *et al.*¹¹ Usually,¹² but not always,¹³ SR proceeds from out of plane to in plane as the temperature increases. Variation of the number n of layers can also lead to SR transitions.¹²⁻¹⁴ Manifestly smooth SR transitions have been recently observed in ferromagnetic thin films.^{15,16} There are two main sources for the out-of-plane and in-plane anisotropies in films: (i) mis-

ing bonds at surfaces can give rise to large local magnetocrystalline anisotropies then; (ii) dipole-dipole interactions induce important anisotropies in magnetic films. In ferromagnets, dipolar fields drive spins to lie in plane, rather than out of plane, because dipolar field energies ($\sim m^2$) that obtain when m is out of plane are thus avoided.¹⁷ Dipolar fields lead to stripelike domains in thin films when magnetocrystalline anisotropies favor spins to be out of plane.¹⁷⁻¹⁹ Growth of such stripes of in plane spins, at the expense of out-of-plane domains (or the other way around), as the temperature varies, leads to continuous SR in ferromagnetic films.²⁰ No continuous SR transition is obtained if a homogeneous magnetization as well as only a lowest-order uniaxial anisotropy is assumed,^{21,22} as has sometimes been done in mean-field theory,^{23,24} MC,^{24,25} and a renormalization group calculation.²⁶

The behavior of antiferromagnetic films is qualitatively different, mainly because anisotropic effects that arise from dipolar fields in AF's are more subtle than in ferromagnets. In AF's, fields decay exponentially beyond the system's boundaries, as expected from the following simple argument. Consider an AF filling all space where $z < 0$. In the vacuum (i.e., where $z > 0$), the magnetic field $\mathbf{h}(\mathbf{r})$ follows from $\mathbf{h}(\mathbf{r}) = \nabla \phi(\mathbf{r})$, where $\phi(\mathbf{r})$ is a suitably defined field. Since $\phi(\mathbf{r})$ obeys Laplace's equation for $z > 0$, $\phi(\mathbf{r})$ can be expanded therein, in obvious notation, as $\sum_{\mathbf{k}} a_{\mathbf{k}} \cos(\mathbf{k} \cdot \mathbf{r}_{\parallel}) \exp \times (-|k|z)$. This much follows as well for ferromagnets. The difference between AF's and ferromagnets arises from the fact that whereas $a_{\mathbf{k}} \approx 0$ for $|k| < |\mathbf{G}|$ where \mathbf{G} gives the periodicity of $\phi(x)$ near the surface of an AF, for ferromagnets, $|\mathbf{G}|$ scales with the inverse ferromagnetic domain size. In addition, the previous argument suggests that anisotropic effects from dipolar fields may also decrease exponentially, away from surfaces, within AF's. Important qualitative differences between anisotropies in ferromagnets and antiferromagnets arise from this. Unfortunately, relevant experiments²⁷⁻²⁹ and MC work for one-layer antiferromagnetic films have only recently been reported.^{30,31} A discontinuous SR has been simulated in one-layer films with a weak antiferromagnetic exchange³⁰ (in which dipolar inter-

actions are dominant) as well as with a strong one.³¹ Because no high-order (beyond quadratic) site anisotropy was taken into account, *continuous* SR's did not obtain. Finally, there is a mean-field theory calculation for one-layer Heisenberg spin systems which include dipolar interactions as well as the lowest-order uniaxial anisotropy³² which also yields a thermally driven discontinuous SR.

Our aim in this paper is to study (i) how the effective surface anisotropy that arises from dipolar interactions in magnetically ordered AF films varies with film thickness and with exchange strength, (ii) how the magnetic phases depend on film thickness, as well as on exchange, the uniaxial D and quadrupolar C anisotropy constants, (iii) how spins on surface layers behave with respect to spins on inner layers, and (iv) how the continuous and discontinuous SR temperatures depend on various parameters.

The plan of the paper is as follows. The model is specified in Sec. II. Section III is about antiferromagnetic ordering in the ground state and the unification that can be achieved between film and bulk phase behavior. This unification comes about because the anisotropy that arises from dipole-dipole interactions is, as surmised in this Introduction, a surface effect. In Sec. III A we define two general homogeneous spin configurations. By MC simulations, we show that all antiferromagnetically ordered phases, except for the SR phase,³³ conform to these configurations. One (the other one) general configuration holds for AF-ordered states in which exchange (dipolar) interactions dominate. We derive the anisotropy energy in each of these two configurations coming from dipolar interactions. Monte Carlo results show that the resulting effective anisotropy decays exponentially fast with distance away from films surfaces. In Sec. III B, the ground-state continuous SR transition is studied. By MC simulations, we study how surface anisotropy arising from dipole-dipole interactions drives the spin directions as a function of layer position. Section IV is about thermal effects. In Sec. IV A we report MC results for transitions between various homogeneous magnetic phases. One of them is the *discontinuous* SR transition. In addition, a transition between two ordered states, with the same spin alignment, is found as the number of film layers changes. In Sec. IV D, we study, by MC simulations, the thermally driven continuous SR transition. Defining an effective uniaxial anisotropy constant D_{eff} that takes into account the dipole-dipole induced anisotropy, we show that the ratio of the SR transition temperature to the ordering temperature depends on D_{eff}/C , but depends hardly on the exchange constant, as long as it is antiferromagnetic.

II. THE MODEL

We next specify the model system we study. Let \mathbf{S}_i be a classical three-component unit spin at lattice site i of a simple cubic (sc) lattice; let

$$\mathcal{H} = \mathcal{H}_J + \mathcal{H}_d + \mathcal{H}_A, \quad (1)$$

where $\mathcal{H}_J = -J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j$, the sum $\sum_{\langle ij \rangle}$ is over all nearest-neighbor bonds,

$$\mathcal{H}_d = \sum_{\langle ij \rangle} \sum_{\alpha\beta} T_{ij}^{\alpha\beta} S_i^\alpha S_j^\beta, \quad (2)$$

$$T_{ij}^{\alpha\beta} = \varepsilon_d \left(\frac{a}{r_{ij}} \right)^3 \left(\delta_{\alpha\beta} - 3 \frac{r_{ij}^\alpha r_{ij}^\beta}{r_{ij}^2} \right), \quad (3)$$

\mathbf{r}_{ij} is the displacement from site i to site j , a is the sc lattice parameter,

$$\mathcal{H}_A = -D \sum_i (S_i^z)^2 - C \sum_i [(S_i^x)^4 + (S_i^y)^4], \quad (4)$$

and D and C are the uniaxial and quadrupolar anisotropy constants, respectively. The nearest-neighbor dipolar energy ε_d is defined through Eqs. (2) and (3).

The boundary conditions we use are most easily grasped in one dimension. Consider first spin sites at $x_k = ka$, for $k = -\infty, \dots, 0, \dots, \infty$. For periodic boundary conditions (PBC's), $S_k^\alpha = S_{k+L}^\alpha$ for all k , and we let a spin at the k th site interact with all $L/2$ ($L/2 - 1$) spins immediately to the right (left) of the k th site. For free boundary conditions (FBC's), on the other hand, we would let a spin at the k th site interact with all spins on sites $n = 1, \dots, k-1, k+1, \dots, L$. We now return to the system of interest here, an n -layer film, by which we mean $L \times L \times n$ spins on a fully occupied sc lattice within a slab which lies flat on an xy plane. Let the z axis be perpendicular to the film layers. We use PBC's along the x and y directions and FBC's along the z direction. Thus, a spin on any given site i interacts, through dipolar fields, with all other $L \times L \times n - 1$ spins in the system which are in a box, whose top and bottom surfaces coincide with the two films surfaces but is otherwise (that is, sidewise) centered on the i th site.

Our simulations follow the standard Metropolis Monte Carlo algorithm.³⁴ In particular, after we choose an initial spin configuration, we compute the dipolar field at each site. Time evolution takes place as follows. A spin is chosen at random and temporarily pointed in a new random direction. The move is accepted if either $\Delta E \leq 0$, where ΔE is the energy change, or with probability $\exp(-\Delta E/k_B T)$, where T is the systems temperature, if $\Delta E > 0$. All dipolar fields are then updated throughout the system if the move is accepted, before another spin is chosen to repeat the process. By *in plane* and *out of plane* we will mean spins lying flat on the xy plane or along the z axis, respectively.

III. EFFECTIVE SURFACE ANISOTROPY FROM DIPOLE-DIPOLE INTERACTIONS

The spin configurations explicitly depicted (not the SR phase) in Figs. 1(a) and 1(b) were shown in Ref. 10 to be ground states for large $L \times L \times L$ systems with PBC's. Our Monte Carlo calculations show that the same spin configurations are also ground states for films, with PBC's at the film edges and FBC's on the perpendicular direction to the film. In these states, spins on the two film surfaces do not deviate at all from the direction they would point to in the bulk.³⁵ We shall refer to these states as *homogeneous*. Our Monte Carlo calculations also show (see below) that, in the SR phase, spins on surface layers tilt away from these directions. We first derive the effective surface anisotropy that arises from dipole-dipole interactions in homogeneous states.

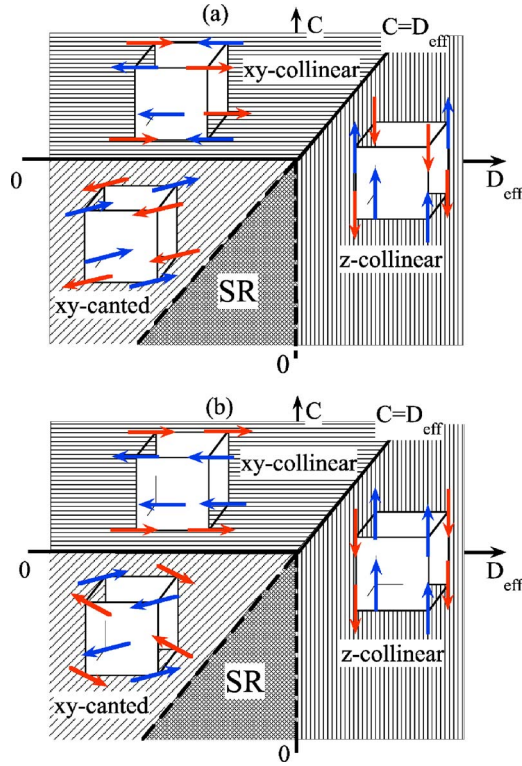


FIG. 1. (Color online) (a) Magnetic phase diagram for AF's in which exchange dominates. In the SR phase, spins tilt away from the z axis, $\phi = \pm \pi/4$, and all spins that are nearest neighbors to any one given spin point opposite to it. (b) Same as in (a) but for AF's in which exchange is very weak and dipolar interactions dominate. A spin configuration in the SR phase is depicted in Fig. 2. Solid and dashed thick lines stand for first- and second-order transitions, respectively.

A. Homogeneous states

Consider first the phases in Fig. 1(a), in which

$$\mathbf{S}_i = (\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta) \boldsymbol{\eta}_i, \quad (5)$$

where

$$\boldsymbol{\eta}_i \equiv (-1)^{x(i)+y(i)+z(i)} \quad (6)$$

and $x(i), y(i), z(i)$ is the three-dimensional position of the i th site. By a proper choice of θ and ϕ , the above equations define the three spin configurations shown in Fig. 1(a). We shall refer to these spin configurations, which minimize E_J , as AF_J configurations. Spins in these configurations are clearly collinear.

Note first that Eqs. (5) and (6) imply

$$E_J = \left(3 - \frac{1}{n}\right) J, \quad (7)$$

independently of θ and ϕ , and $E_A = -D \cos^2 \theta - C(\sin^4 \phi + \cos^4 \phi) \sin^4 \theta$. To calculate E_d , we substitute Eqs. (5) and (6) into Eqs. (2) and (3). Note first that $\sum_{ij} T_{ij}^{\alpha\beta} \boldsymbol{\eta}_i \boldsymbol{\eta}_j = 0$ if $\alpha \neq \beta$ (by x and y reflection symmetry). Similarly, $\sum_{ij} T_{ij}^{xx} \boldsymbol{\eta}_i \boldsymbol{\eta}_j = \sum_{ij} T_{ij}^{yy} \boldsymbol{\eta}_i \boldsymbol{\eta}_j$. Therefore,

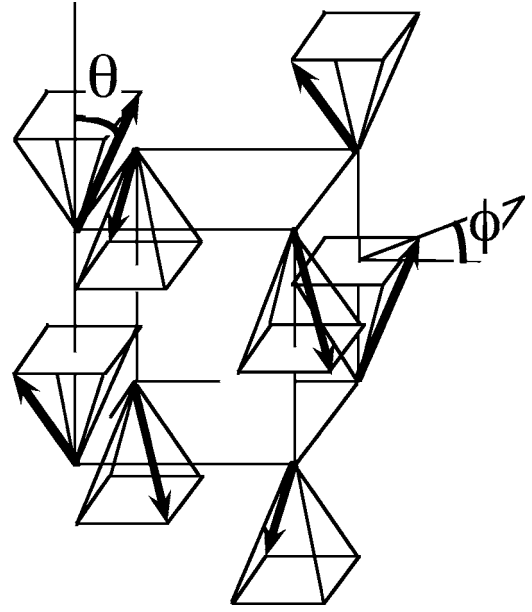


FIG. 2. Spin configuration defined by Eqs. (11) and (12). In the SR phase of Fig. 1(b), $0 < \theta < \pi/2$ and $\phi = \pm \pi/4$; for other phases of Fig. 1(b), θ and ϕ are as depicted therein.

$$E_d = (2N)^{-1} \sum_{ij} [T_{ij}^{xx} \boldsymbol{\eta}_i \boldsymbol{\eta}_j + (T_{ij}^{zz} - T_{ij}^{xx}) \boldsymbol{\eta}_i \boldsymbol{\eta}_j \cos^2 \theta] \quad (8)$$

follows, which, by numerical evaluation, gives

$$E_d = \frac{0.67 \epsilon_d}{n} - \frac{\Delta}{n} \cos^2 \theta, \quad (9)$$

where

$$\Delta = 1.984 \epsilon_d \quad (10)$$

for AF_J spin configurations.

We next calculate $E_J + E_d + E_A$ for the three spin configurations shown in Fig. 1(b), given by

$$\mathbf{S}_i = (\tau_i^x \sin \theta \cos \phi, \tau_i^y \sin \theta \sin \phi, \tau_i^z \cos \theta), \quad (11)$$

where $\boldsymbol{\tau}_i \equiv [\tau_i^x, \tau_i^y, \tau_i^z]$ is given by

$$\boldsymbol{\tau}_i = [(-1)^{y(i)+z(i)}, (-1)^{x(i)+z(i)}, (-1)^{x(i)+y(i)}]. \quad (12)$$

We shall refer to the above spin configurations, depicted in Fig. 1(b) for some values of θ and ϕ and in Fig. 2 for arbitrary θ and ϕ , as AF_d configurations. Spins in these configurations are in general noncollinear. It is worth pointing out that, in $L \times L \times L$ spin systems in cubic lattices, \mathcal{H}_d is invariant with respect to both θ and ϕ in these AF_d configurations.^{10,36}

Note first that Eqs. (11) and (12) imply

$$E_J = \left(1 - \frac{1}{n}\right) J + \frac{2J}{n} \cos^2 \theta \quad (13)$$

and $E_A = -D \cos^2 \theta - C(\sin^4 \phi + \cos^4 \phi) \sin^4 \theta$. We now calculate E_d . For systems with complete cubic symmetry—that is, with a cubic lattice structure and a cubic shape—with the same type of boundary conditions on all surfaces, we have

shown¹⁰ that E_d is invariant with respect to θ and ϕ in Eq. (11). By the arguments preceding Eq. (9), we obtain

$$E_d = (2N)^{-1} \sum_{ij} [T_{ij}^{xx} \tau_i^x \tau_j^x + (T_{ij}^{zz} \tau_i^z \tau_j^z - T_{ij}^{xx} \tau_i^x \tau_j^x) \cos^2 \theta] \quad (14)$$

for n -layer films with PBC's at the edges and FBC's on the top and bottom surfaces. Straightforward numerical calculations give

$$E_d = - \left(2.68 - \frac{0.13}{n} \right) \varepsilon_d + \frac{1.23 \varepsilon_d}{n} \cos^2 \theta \quad (15)$$

for AF_d spin configurations.

Thus, for both AF_J and AF_d configurations, an effective anisotropy

$$D_{eff} = D + \Delta/n \quad (16)$$

obtains, where Δ is given by Eq. (10) for AF_J , and by

$$\Delta = -1.23 \varepsilon_d - 2J \quad (17)$$

for AF_d states.

The anisotropy induced in AF's by dipole-dipole interactions, given by Eqs. (10), (16), and (17), differ from the one for ferromagnets in two respects: (i) it favors out-of-plane spins over in plane ones in AF_J states, and (ii) the corresponding energy for AF's varies as $1/n$ as n increases. The reason for it is given in the Introduction, in-plane orientation need not be favored in AF's, because no significant vacuum dipolar field energy exists for them. We next discuss the mechanism underlying the other effect, the $1/n$ behavior.

Equations (10) and (17) suggest that anisotropy effects arising from dipole-dipole interactions occur only on surface layers. We have (numerically) calculated how the dipolar field varies with the distance from a film's surface for homogeneous spin configurations. Let $\nu=0,1,2,\dots$ number the layers, starting with 0 for one of the two outermost surface layers. We find that the deviation of dipolar fields from their bulk value decreases exponentially as ν increases. More specifically,

$$\delta h_\nu \approx \delta h \exp(-\kappa \nu), \quad (18)$$

where $\kappa \approx 4.4$ ($\kappa \approx 7$) for all AF_J 's and out of plane AF_d 's (for all in-plane AF_d 's) ordered states we have tried. This is in agreement with the discussion in Sec. I, since the wave vector magnitude of the field near the surface of an AF_J or an out-of-plane AF_d (for an in-plane AF_d) on a sc lattice is $\sqrt{2}\pi/a$ ($\sqrt{5}\pi/a$). Therefore, to the accuracy of our numerical results, all anisotropy effects arising from dipole-dipole interactions occur only on the outer surfaces. Therein, it is given by $\Delta/2$ for each of the two surfaces on $n=2$ films and by Δ for $n=1$ layer films.

In order to characterize antiferromagnetically ordered states, we now define

$$m_J^\alpha = N^{-1} \sum_i S_i^\alpha \eta_i \quad (19)$$

and

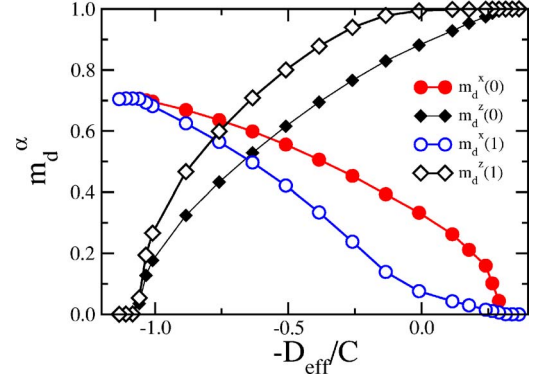


FIG. 3. (Color online) m_d^α versus $-D_{eff}/C$, for $\alpha=x,z$ on two different layers. $m_d^\alpha(\nu)$ is for the ν th layer ($\nu=0$ for a surface layer, and so on). All data follow from MC simulations of films of $16 \times 16 \times n$ spins, for $n=4$, $C=-0.8\varepsilon_d$, and $J=0$, near $T=0$.

$$m_d^\alpha = N^{-1} \sum_i S_i^\alpha \tau_i^\alpha. \quad (20)$$

In an AF_J state, $\mathbf{m}_J = (\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta)$ and $\mathbf{m}_d = 0$. On the other hand, in an AF_d state $\mathbf{m}_J = 0$ and $\mathbf{m}_d = (\sin \theta \cos \phi, \sin \theta \sin \phi, \cos \theta)$. We can define these order parameters for the whole system, by summing over all sites i , or we can define, say, order parameters for surface film layers or interior layers, by summing over surface or interior sites. We next discuss the effective anisotropy in the SR phase.

B. SR phase

In Ref. 10 the nature of the spin reorientation phase [marked as SR in Figs. 1(a) and 1(b)] in $L \times L \times L$ systems with PBC's was discussed in some detail. In the SR phase $m_J^\alpha \neq 0$ ($m_d^\alpha \neq 0$) for all three components of α if $J \leq -1.34\varepsilon_d$ ($-1.34\varepsilon_d \leq J \leq 0$); that is, spins in the SR phase are tilted some angle $0 < \theta < \pi/2$ away from the z axis. For homogeneous states, given by either Eq. (5) or (12), as for $L \times L \times L$ systems in Ref. 10, minimization of the total energy, gives

$$\mathbf{S} = (\pm u, \pm u, \pm v) \quad (21)$$

for $C < D_{eff} < 0$, where $u = \sqrt{D_{eff}/2C}$ and $v = \sqrt{1-2u^2}$. Then, varying D_{eff}/C over the 0–1 range would lead to a $\pi/2$ spin rotation. Inspection of Fig. 3 shows that the homogeneity assumption is wrong for thin films, and consequently, spins do *not* quite rotate by $\pi/2$ as D_{eff}/C sweeps over the 0–1 interval. The order parameters m_d^x and m_d^z on the surface clearly differ from the order parameters on inner layers in Fig. 3. To the accuracy of our results, spins on all inner layers do follow either Eq. (5) or (11). Thus, phase diagrams for n -layer films collapse into a single diagram if D is replaced by $D + \Delta/n$, but only approximately so for the SR phase.

In order to look further into this effect, we have also performed MC simulations of n -layer films with anisotropy constants D and C on all sites, except for all surface sites, where a variable quantity ΔD is added to D . We calculate $|m_d^\alpha(in)|$

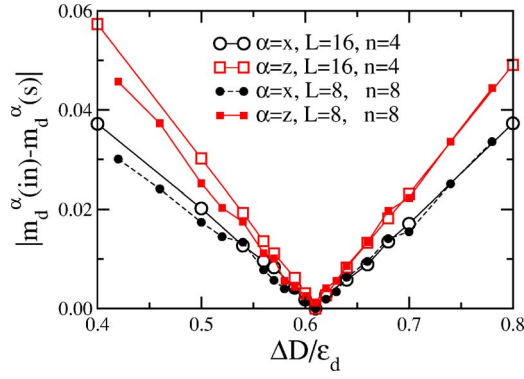


FIG. 4. (Color online) $|m_d^\alpha(in) - m_d^\alpha(s)|$ versus ΔD for slabs of $L \times L \times n$ dipoles for $J=0$, $D=-\epsilon_d$, and $C=-2D$. We use FBC's in the z direction and PBC's in the xy plane. The value of D on the two outermost layers of the slab differs by ΔD from the value it takes on the inner layers.

for inner layers and $|m_d^\alpha(s)|$ for the two surface layers. A plot of $|m_d^\alpha(in) - m_d^\alpha(s)|$, for $\alpha=x, y, z$, vs $\Delta D/\epsilon_d$ is shown in Fig. 4 for $T \sim 0$, $D=-\epsilon_d$, $C=-2D$, and $J=0$. $|m_d^\alpha(in) - m_d^\alpha(s)|$ vanishes for all α at $\Delta D=0.615\epsilon_d$, as was to be expected from Eqs. (17) and (18). Further MC simulations we have performed for films of various thicknesses yield analogous results.

IV. PHASE TRANSITIONS

Up to this point we have assumed which of the two, AF_J or AF_d , states the system is in, but we are now able to specify which of these two obtain given the value of J . Much of this section, the portions having to do with phase transitions as the number of film layers change at very small temperatures follow from the following considerations. Comparison of Eqs. (7), (9), and (10), with Eqs. (16) and (17) shows that (i) AF_J (AF_d) order ensues in out-of-plane spin configurations when $J \lesssim -1.34\epsilon_d$ ($-1.34\epsilon_d \lesssim J < 0$); AF_J [AF_d] order ensues for in-plane spin configurations when $J \lesssim -(1.34+0.27/n)\epsilon_d$ [$-(1.34+0.27/n)\epsilon_d \lesssim J < 0$]. From these conditions on $|J|$ one can decide whether $|J|$ is sufficiently small for a system to qualify as a dipolar antiferromagnet.³⁷ As $n \rightarrow \infty$, the results obtained in Ref. 10 for bulk systems follow. Note also that a transformation between AF_J - and AF_d -ordered states for in-plane configurations can occur as the number n of layers changes if $-1.61\epsilon_d \lesssim J \lesssim -1.34\epsilon_d$.

A. $C=0$ transition

The phase transition at $C=0$ for $D_{eff} < 0$ is illustrated in Fig. 5. We know of no previous experimental or MC work on this transition in films. It appears to be of first order, as predicted by Landau's theory, because no symmetry group in any of these phases is a subgroup of another one. The transition moves slightly off the $C=0$ line as T departs from 0, as shown for a one layer film in Fig. 6, giving rise to a reentrant transition. The transition at $C=0$, however, remains unmoved at $T=0$ as the number of film layers varies. This is in agree-

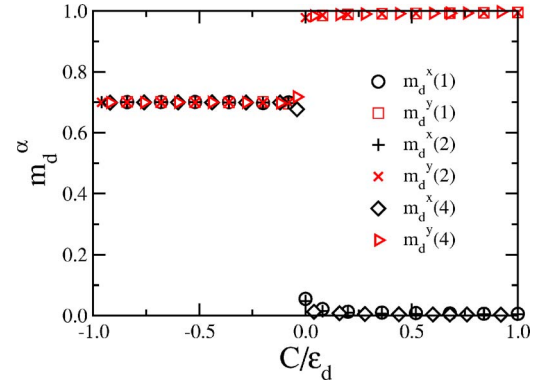


FIG. 5. (Color online) Order parameter m_d^α , for $\alpha=x, y$ vs C for slabs of $16 \times 16 \times n$ dipoles on sc lattices at $T=0.05\epsilon_d/k_B$, where k_B is Boltzmann's constant, for $D=-\epsilon_d$ and $J=0$. A transition between the xy -collinear and xy -canted phases is clearly exhibited at $C=0$ for different values of n . We use FBC's in the z direction and PBC's in the xy plane. C was lowered from $C=\epsilon_d$ in $\Delta C=-0.02\epsilon_d/k_B$ steps. Each data point follows from averages over 10^5 MC sweeps.

ment with the statement that dipolar interactions shift the value of D but not of C .

B. Discontinuous SR transitions

Consider first the phase transition between in-plane and out-of-plane spin configurations at low temperature for $C > 0$. Assume temporarily the same AF_J or AF_d order in both in-plane and out-of-plane phases, and recall that the ground-state energy variation with spin tilt angle θ is given by \mathcal{H}_A if D is replaced by D_{eff} in Eq. (4). Then,

$$\mathcal{H}_A = C \left[\left(\frac{D_{eff}}{C} - \sin^2 \theta \right) \sin^2 \theta \right] - D_{eff} \quad (22)$$

follows from Eq. (4), replacing D by D_{eff} and assuming $S_x^2 = 1$ or $S_y^2 = 1$. Clearly, (i) $D_{eff}/C < 1$ ($D_{eff}/C > 1$) implies an

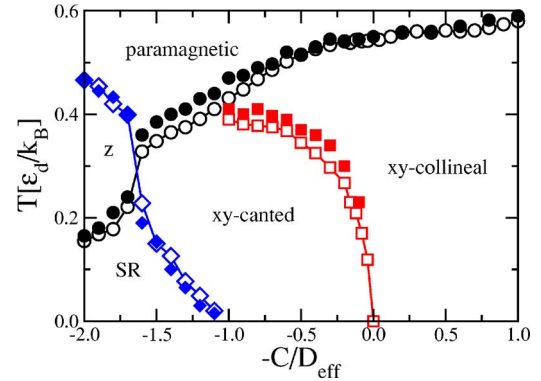


FIG. 6. (Color online) Transition temperatures vs $-C/D_{eff}$ from MC simulations of $L \times L \times 1$ spin systems, for $L=16$ and $L=32$, varying C , with $D_{eff}=-0.5\epsilon_d$ and $J=0$ fixed. Open and solid symbols stand for $L=16$ and $L=32$, respectively. xy and z stand for in-plane and out-of-plane spin configurations. In the simulations, the temperature was lowered in steps of $0.02\epsilon_d/k_B$ and $0.01\epsilon_d/k_B$ for $L=32$ and $L=16$, respectively. At each temperature, 10^6 and 2×10^5 MC sweeps were taken, for $L=16$ and $L=32$, respectively. In order to check for nonequilibrium effects, the system was heated for some values of C/D_{eff} . No such effects were found.

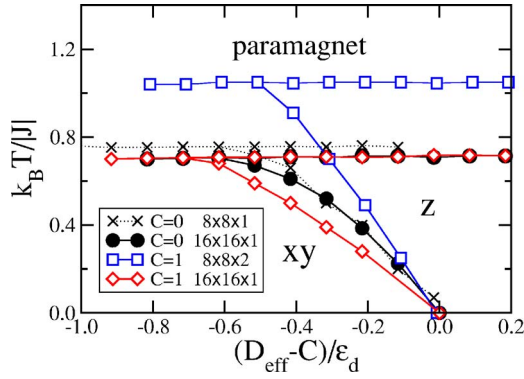


FIG. 7. (Color online) Transition temperatures vs D_{eff}/C for $n=1$ and $n=2$ layer films in which $J=-10$, $C=0$, and $C=1$. In the graph, z and xy stand for z -collinear and xy -collinear phases, respectively. All data points come from MC simulations of $L \times L \times n$ spins for the values of L and n shown in the graph. The phase transition boundaries at the top follow from the location of specific heat peaks obtained while lowering the temperature in $\Delta T=0.1$ steps. In order to make sure equilibrium is realized, the lower phase transition boundary, for SR, is obtained from counting, over MC runs of several times 10^8 sweeps, the frequency of occurrences of the two phases, which must be the same for both phases at the boundary line.

in-plane (out-of-plane) phase in the ground state and (ii) the transition is discontinuous since Eq. (22) gives an energy barrier between $\theta=0$ and $\theta=\pi/2$ when $D_{\text{eff}}=C$. This is as depicted in Figs. 1(a) and 1(b). It has been observed in experimental and numerical work on films.^{12,14} The assumption we made—that the same AF_J or AF_d order prevails in both in-plane and out-of-plane phases—holds for most values of J , as follows from Eqs. (4), (7), (9), (13), (15), and (17). More specifically, the transition occurs between two AF_d states if $-1, 34\epsilon_d \lesssim J \lesssim 0$. The transition is between two AF_J states if $J \lesssim -(1.34 + 0.27/n)\epsilon_d$. Monte Carlo results that include this transition are shown in Fig. 7 for films of $n=1$ and $n=2$.

The boundary line between out-of-plane and in-plane phases tilts away from $D_{\text{eff}}=C$ as the temperature increases, as shown in Fig. 7. Thus, the possibility of thermally driven SR transitions arises, as T varies if $-0.5\epsilon_d \lesssim D_{\text{eff}} - C < 0$. This is qualitatively as in the mean-field prediction³⁸ for $J = -10^3\epsilon_d$, $C=0$, and $n=1$ in Ref. 32.

In the small $-(1.34 + 0.27/n)\epsilon_d \lesssim J \lesssim -1.34\epsilon_d$ range the situation is a more interesting. As specified at the beginning of Sec. IV, the phase transition is then between an out-of-plane AF_J -ordered state and an in-plane AF_d -ordered state. Equation (22) does not apply, then, because the assumption underlying it—that the same spin ordering AF_J or AF_d prevails on both sides of the phase boundary—breaks down. Assuming homogeneity and making use of Eqs. (4), (7), (9), (13), and (15), the condition for discontinuous SR transitions becomes

$$D = C + 2J + (2.68 - 1.44/n)\epsilon_d, \quad (23)$$

for $C > 0$. Note that quantity D_{eff} is not well defined in this narrow J range. We can, however, use the two different val-

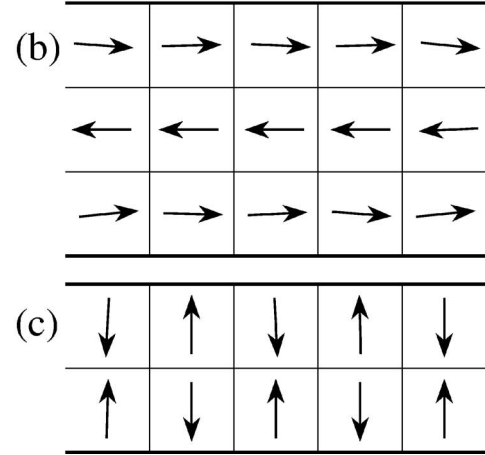
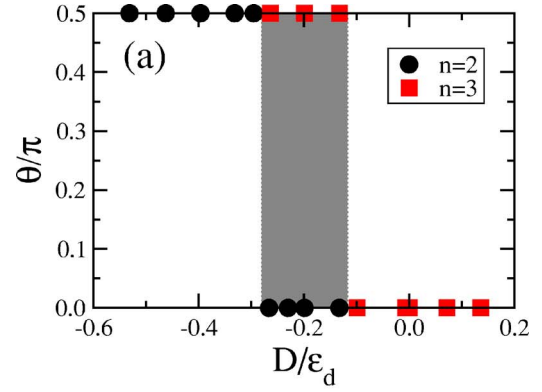


FIG. 8. (Color online) Everything shown is for $J=-1.38\epsilon_d$, $C=0.5\epsilon_d$, and $k_B T=0.01\epsilon_d$. This is the lowest temperature we obtained with MC simulations in which T was lowered in $\Delta k_B T=0.1\epsilon_d$ steps, down to $k_B T=0.01\epsilon_d$. Some 10^5 MC sweeps were taken at each temperature step. (a) θ vs D for $n=2$ (●) and $n=3$ (■). The shaded rectangle covers values of D/ϵ_d where the out-of-plane AF_J state, shown in (c), transforms into the AF_d in-plane phase, shown in (b) if a layer is added to the film. (b) A $\theta=\pi/2$, AF_d , spin configuration, on lattice sites of a vertical cut of an $n=3$ layer film, obtained at $k_B T=0.1\epsilon_d$ for $C=0.5$ and $D=-0.2\epsilon_d$. (c) Same as in (b) but a $\theta=0$, AF_J , spin configuration that obtains for an $n=2$ layer film, for the same values of C , D , and T , as in (b).

ues D_{eff} has on both sides of the phase transition for comparison of the energies of AF_J - and AF_d -ordered systems. It can be checked straightforwardly that, again, there is an energy barrier between the $\theta=0$ and $\theta=\pi/2$ phases. This transition, which has not, as far as we know, thus far been observed, is illustrated in Fig. 8 with MC results for three- and two-layer films. The dark rectangle shown in Fig. 8, showing the range of values of D/ϵ_d where $n=2$ ($n=3$) films order in out-of-plane AF_J (in-plane AF_d) states, follows from our MC simulations. It is slightly displaced to the left, by $D/\epsilon_d \approx 0.12$, from the prediction that follows from Eq. (23). Irreversibility, which keeps spins from reorienting, from in plane to out of plane, at low T , as T decreases, is responsible for this effect.

C. Transitions between AF_J - and AF_d -ordered states

Phase transformations that do not involve SR can also occur between AF_J - and AF_d -ordered states, as the number n

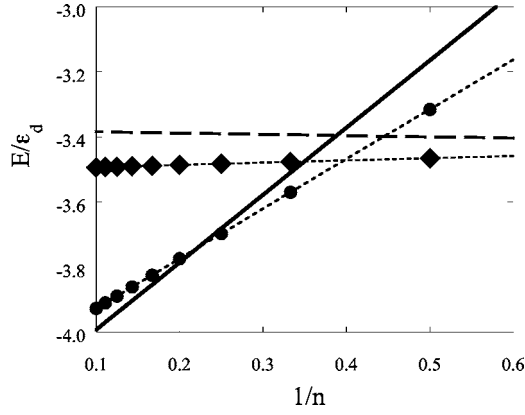


FIG. 9. Ground-state energies vs $1/n$ for AF_J and AF_d states, for out-of-plane and in-plane spin alignment, of n -layer slabs, with $J = -1.38\varepsilon_d$, $D = -0.7\varepsilon_d$, and $C = 0$. \bullet and solid line stand for in-plane (spins either along the lattice axes or diagonally to them) AF_J - and AF_d -ordered states, respectively; dashed line and \blacklozenge stand for out-of-plane AF_J - and AF_d -ordered states, respectively. Dotted lines are guides to the eye.

of layers changes, if $-(1.34 + 0.27/n)\varepsilon_d \leq J \leq -1.34\varepsilon_d$. This is illustrated in Fig. 9 for $J = -1.38\varepsilon_d$, $D = -0.7\varepsilon_d$, and $C = 0$, where a transition from an AF_J - to an AF_d -ordered state, both in plane, is shown to take place as n decreases from $n=6$ to $n=5$. (This is followed by a spin rotation, from in plane to out of plane, as n decreases from $n=3$ to $n=2$.) We are not aware of any experimental observation of this kind of phase transformation.

D. Continuous SR transitions

A thermally driven SR transition is illustrated in Fig. 10 for an $n=1$ layer film. It is rather similar to thermally driven transitions in $L \times L \times L$ systems with PBC's.¹⁰ However, as is pointed out in Sec. III B (and illustrated in Fig. 3) for $T \sim 0$, the SR phase in films with $n > 1$ is special. Spin configurations in the SR phase are not homogeneous. Whereas spins on inner layers follow Eq. (5) or (11), spins on surface layers

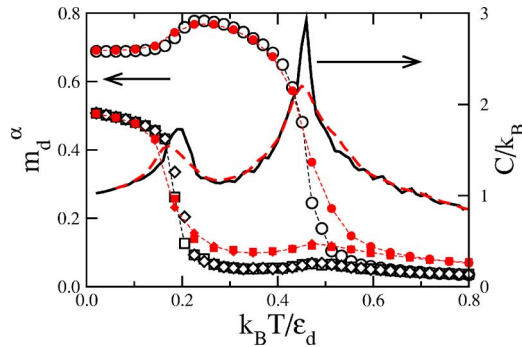


FIG. 10. (Color online) m_d^z (\circ and \bullet), m_d^y (\square and \blacksquare), m_d^x (\diamond and \blacklozenge), and C/k_B (solid and dashed lines) vs T . Open (solid) symbols and solid (dashed) lines are for MC results for systems of $32 \times 32 \times 1$ (of $16 \times 16 \times 1$) spins on sc lattices for $J=0$, $C=-\varepsilon_d$, and $D=-0.7C$. At each value of T , 4×10^5 MC sweeps were made. Lines are guides to the eye.

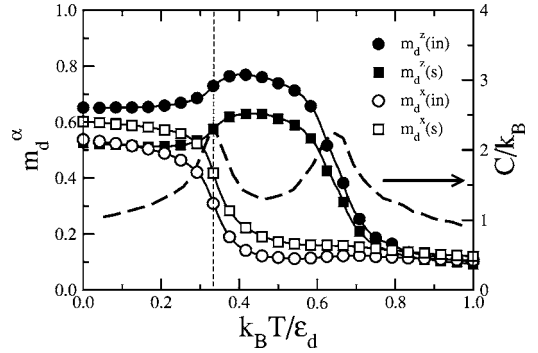


FIG. 11. Surface and inner layer order parameters $|m_d^\alpha(s)|$ and $|m_d^\alpha(in)|$, respectively, for $\alpha = x, y$, and z , and specific heat C vs T . All data points come from MC simulations of of $8 \times 8 \times 4$ spins with $J=0$. Symbols (dashed lines) are for order parameters (specific heat) of systems with spatially homogeneous anisotropy constants $D = -\varepsilon_d$ and $C = -2\varepsilon_d$ and FBC's on the top and bottom surfaces; solid lines are for films with PBC's on all of its boundaries with a uniaxial anisotropy constant $D = -\varepsilon_d$ on inner surfaces, $D = -\varepsilon_d + \Delta/2$ on its top and bottom surfaces, and $C = -2\varepsilon_d$ everywhere.

do not if D and C are homogeneous throughout the system.

Inhomogeneity effects that arise from the effective surface anisotropy (induced by dipole-dipole interactions) are illustrated in Fig. 11 for a four-layer film as a function of temperature. Two kinds of data points from MC simulations are shown: (i) For films with spatially homogeneous anisotropy constants D and C and FBC's on the top and bottom surfaces and (ii) for films with PBC's on all of its boundaries³⁹ with a uniaxial anisotropy constant that is $\Delta/2$ larger on its top and bottom surfaces than on the inner layers. Note how the order parameters on a surface layer differs from the order parameters on inner layers in the former case and how direct application of an anisotropy $\Delta/2$ on surface layers of films with PBC's (no anisotropy from dipolar interactions arises then) leads to the same effect. This is as expected from the discussion in Sec. III B, concerning Fig. 4.

The continuous SR portion of the phase diagram for films is rather like the one for bulk AF's.¹⁰ We give the MC results we have obtained for $n=1$ layer dipolar films (in which $J = 0$) in Fig. 12.

Finally, we compare ordering and SR temperatures. Let T_z (T_{xy}) stand for the temperatures below which $m_j^z \neq 0$ or $m_d^z \neq 0$ ($m_j^{xy} \neq 0$ or $m_d^{xy} \neq 0$). Monte Carlo results for the T_z/T_{xy} ratio for $n=1$ films and bulk systems are shown in Fig. 13 for $J/\varepsilon_d=0$ and for $\varepsilon_d/J=0$. In the latter case, T_z/T_{xy} is, of course, independent of J , since J sets the only energy scale then. The insensitivity of T_z/T_{xy} to dimensionality and to the value of J , even when $\varepsilon_d \neq 0$, is remarkable.

V. CONCLUSIONS

We have studied a nearest-neighbor Heisenberg-spin film-like system with dipolar interactions and uniaxial plus quadrupolar anisotropies. We have found how the strength Δ of the effective uniaxial anisotropy that arises from dipolar interactions varies with the strength of the antiferromagnetic exchange interaction and with layer position. We have ar-

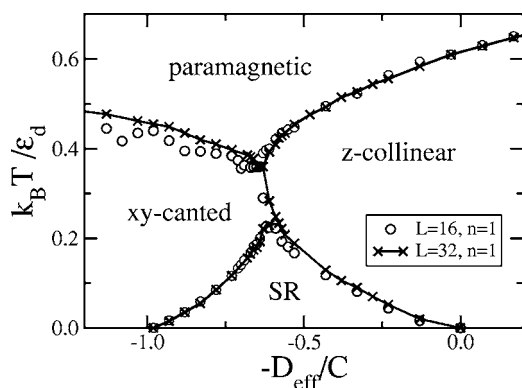


FIG. 12. (a) Phases of dipolar ($J=0$) antiferromagnetic films for $C=-\epsilon_d$. All data points come from MC simulations of (○) $16 \times 16 \times 1$ and (×) $32 \times 32 \times 1$ spins. *xy-canted* and *z-collinear* stand for the ordered phases that are depicted in Fig. 1(b); the SR phase is as pictured in Fig. 2.

gued (in Sec. I), and checked with MC simulations (in Sec. III A), that Δ decays exponentially fast with the distance from either of the two outermost film layers. We have also found (in Sec. III) that, except for the SR phase, all antiferromagnetic phases are homogeneous; that is, there are no surface states. These results, which are peculiar to AF's, imply that all but the SR phases of n -layer antiferromagnetic films fit into a single-phase diagram [see Figs. 1(a) and 1(b)] if we let $D \rightarrow D + \Delta/n$. The only exceptions are the SR phase which is associated with *continuous* SR (see Secs. III B and IV D), and phase transitions that arise from variation of film thickness, in the narrow $-1.61\epsilon_d \lesssim J \lesssim -1.34\epsilon_d$ range, in which spin orientation may (see Sec. IV B) or may not (see Sec. IV C) change. Finally, by means of MC simulations, we

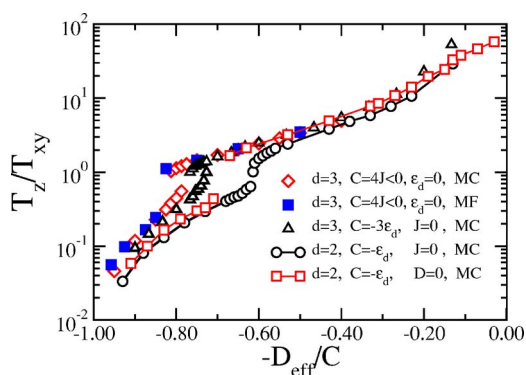


FIG. 13. (Color online) Temperature ratio T_z/T_{xy} vs $-D_{eff}/C$ for the values shown of ϵ_d , J , and C in films ($d=2$) and $L \times L \times L$ systems with PBC's ($d=3$). All data points, except the (■) mean-field (MF) results, come from MC simulations. Note that ○ and △ stand for pure dipolar systems ($J=0$), ◇ is for $\epsilon_d=0$, and □ is for a one-layer film in which both D and C are constant [D_{eff} varies, however, following Eqs. (16) and (17), because J varies]. Systems are either of $L \times L \times L$ spins (for $d=3$) or of $L \times L \times 1$ spins (for $d=2$). All symbols stand for $L=16$ except for ○, which stands for $L=32$. Lines are guides to the eye.

have found that the ratio of the continuous SR transition to the Néel ordering temperature depends very little on J , as illustrated in Fig. 13, and seems to depend on the number n of film layers only through the effective uniaxial anisotropy constant $D + \Delta/n$.

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