PHYSICAL REVIEW B VOLUME 51, NUMBER 1 1 JANUARY 1995-I

Mean-field theory for spin-reorientation phase transitions in magnetic thin films

Xiao Hu and Yoshiyuki Kawazoe
Institute for Materials Research, Tohoku University, Sendai 980-77, Japan
(Received 4 August 1994)

A mean-field theory for spin-reorientation phase transitions in magnetic thin films is worked out, solving analytically the relevant differential equation. The role of the exchange stiffness A in the competition between the in-plane shape anisotropy K_v and the perpendicular surface anisotropy K_s is addressed. The spin-reorientation transition from the perpendicular configuration to the in-plane one with increasing thickness is derived. Two different stable spin configurations are presented at large thickness for $K_s < \sqrt{AK_v}$ and vice versa. The phase transition with temperature is also explored. Asymptotic formulas are given for energy stocked in the film per unit area, which enable one to evaluate the surface anisotropy.

I. INTRODUCTION

Much attention has been paid to metallic thin films in recent years, since sophisticated epitaxial techniques enable us to control the thickness, the surface condition, and the interface smoothness between the film under consideration and substrate, and many new phenomena have been observed. Among them it is found experimentally in nonmagnetic-magnetic-nonmagnetic sandwich structures that, the easy-axis direction for magnetization changes around a thickness of the transition metallic film: At lower thickness, it is normal to the film plane, while at higher thickness, it lies in the film plane.[1–5] A similar phenomenon has also been discussed in magnetic bilayer systems used for magneto-optical recording.[6,7] The phase transition is also observed when the film thickness is fixed, while the temperature is varied.[8–11]

From the application point of view, the presence of perpendicular magnetization in thin film of transition metal is of great potential for high-density recording. This phenomenon in thin-film geometry is of fundamental interest as well. According to the theorem of Mermin and Wagner,[12] no long-range order can exist in two-dimensional (2D) isotropic Heisenberg system. Therefore, the magnetization in film should be triggered by anisotropy. The group of Mills[13] has shown with renormalization-group (RG) approach and Monte Carlo simulations that a phase transition to ferromagnetism occurs for arbitrary small anisotropy. In sandwich and bilayer structures, a surface anisotropy normal to the film is produced by the breaking of translation invariance at the interface, as considered first by Néel.[14] In contrast with 3D systems, the magnetization in film also produces shape anisotropy, which favors in-plane ordering. In the present paper, we will assume the presence of the long-range order in the thin film under a surface anisotropy, and proceed to discuss the spin-reorientation phase transitions with the variance of the thickness and the temperature.

Although the role played by the anisotropies has been explored by both experimental and theoretical arguments, the exchange stiffness has not been paid enough attention to. In most of the literature, the inverse thickness dependence of the effective volume anisotropy is taken as a satisfied fitting of experimental data or as an ansatz *a priori* in theory. The effect of the exchange stiffness was studied first by Rado and his co-workers, resorting to a linearization procedure to the nonlinear differential equation.[15] In the present study we explore the role of the exchange stiffness in the competition of the shape anisotropy and the surface anisotropy, both around the transition point and at large thickness limit, by solving directly a nonlinear differential equation.

The remaining part of this paper is organized as follows: In Sec. II the formalism is presented briefly and the phase transition is settled. In Sec. III we show the thickness dependence of the effective anisotropy from our approach. A summary is given in Sec. IV.

II. SPIN-REORIENTATION TRANSITIONS

Consider a magnetic layer with thickness $2a$. Within it, one has the ferromagnetic exchange stiffness A and a volume anisotropy K_v. At the surface, a perpendicular surface anisotropy K_s is supposed to exist. As a well-chosen simplification, we take the quadratic Néel-type approximation. Taking the z axis to be normal to the film and the origin at the bottom surface as shown in Fig. 1, half of the total energy per unit area is expressed by

$$\gamma = \int_0^a \left[A \left(\frac{d\varphi}{dz} \right)^2 - K_v \sin^2\varphi \right] dz + K_s \sin^2\varphi(0) , \quad (1)$$

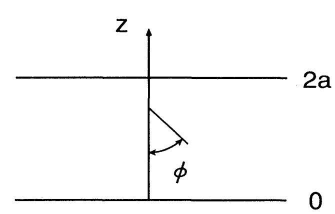

FIG. 1. Geometry and coordinates of the system.

where K_s and K_v are positive through the present study. The volume anisotropy K_v includes the shape anisotropy from the dipole interaction and possibly an intrinsic part. The stable state is considered to be uniform in the directions parallel to the surfaces.

By applying the variational method to energy (1), we obtain the following differential equation:

$$2A\frac{d^2\varphi}{dz^2}+K_v\frac{d\sin^2\varphi}{d\varphi}\varphi=0 \tag{2}$$

with boundary conditions

$$\left.\frac{d\varphi}{dz}\right|_{z=a}=0\ ,$$

$$\left.A\frac{d\varphi}{dz}\right|_{z=0}=K_s\sin\varphi(0)\cos\varphi(0)\ . \tag{3}$$

The above differential equation can be transformed into a nonlinear equation, concerning, for example, the direction of magnetization at $z=a$:[7]

$$\frac{K_s}{\sqrt{AK_v}}=\frac{sn\,[a\sqrt{K_v/A}\,,\sin\varphi_a]dn\,[a\sqrt{K_v/A}\,,\sin\varphi_a]}{cn\,[a\sqrt{K_v/A}\,,\sin\varphi_a]} \tag{4}$$

where $\varphi_a\equiv\varphi(a)$, and $sn\,[x,k]$, etc., are the Jacobian elliptic functions. The total configuration in the film is expressed by the variable φ_a directly as

$$\varphi(z)=\sin^{-1}\left[\sin\varphi_a\frac{cn\,[(a-z)\sqrt{K_v/A}\,,\sin\varphi_a]}{dn\,[(a-z)\sqrt{K_v/A}\,,\sin\varphi_a]}\right] \quad \text{for } 0\le z\le a\ . \tag{5}$$

The configuration in the upper half of the film can be given by noting the mirror symmetry of the film concerning $z=a$. This approach is useful for the discussion of variance of easy axis with the thickness a, since the left-hand side of (4) is independent of the film thickness.[7]

A. Onset of in-plane magnetization

It is found that there is a bifurcation of the solutions to (4) concerning the thickness with the mechanism discussed in Ref. 7: Below the bifurcation point there is only the trivial solution $\varphi(z)=0$ for $0\le z\le 2a$, the nontrivial configuration appears only for thicknesses higher than the bifurcation point. The bifurcation thickness a_{c1} is derived analytically from (4) by noting $\varphi_a=0$:

$$a_{c1}=\sqrt{A/K_v}\tan^{-1}\frac{K_s}{\sqrt{AK_v}}\ . \tag{6}$$

For values of magnetic quantities like $K_v=5.0\times10^8$ mJ/m^3, $A=2.0\times10^{-9}$ mJ/m, and $K_s=1.5$ mJ/m^2, the above critical thickness is evaluated as $a_{c1}\simeq 20$ Å. The variance of the direction of magnetization is described by the exponent $\beta=\frac{1}{2}$, as

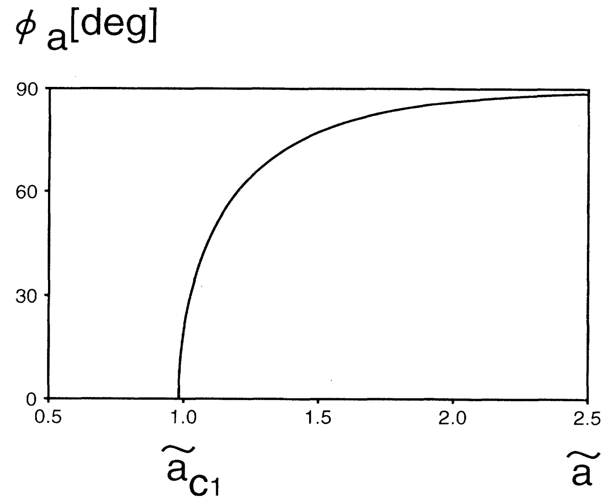

FIG. 2. Phase transition concerning the magnetization direction from normal ($\varphi=0$) to in-plane ($\varphi=\pi/2$), where $\tilde{a}\equiv a\sqrt{K_v/A}$.

$$\varphi\sim\left[\frac{a}{a_{c1}}-1\right]^\beta \quad \text{for } a\ge a_{c1}\ , \tag{7}$$

and depicted in Fig. 2 for φ_a.

B. Large thickness limit

The behavior of the system with large thickness can also be investigated by (4). For $K_s>\sqrt{AK_v}$, one can set $a\to\infty$ and $\varphi_a=\pi/2$ in (4). In this case, one has from (4) and (5)

$$\varphi(0)=\sin^{-1}\frac{\sqrt{AK_v}}{K_s} \quad \text{as } a\to\infty\ . \tag{8}$$

Figure 3 shows the thickness dependence of φ_a and that

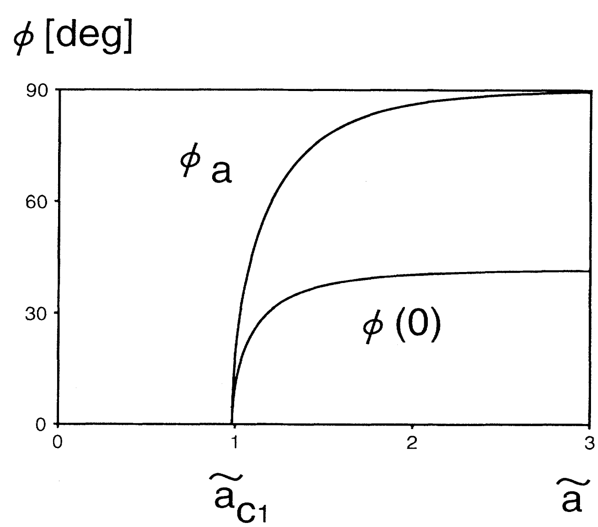

FIG. 3. Thickness dependence of the magnetization direction for $K_s>\sqrt{AK_v}$.

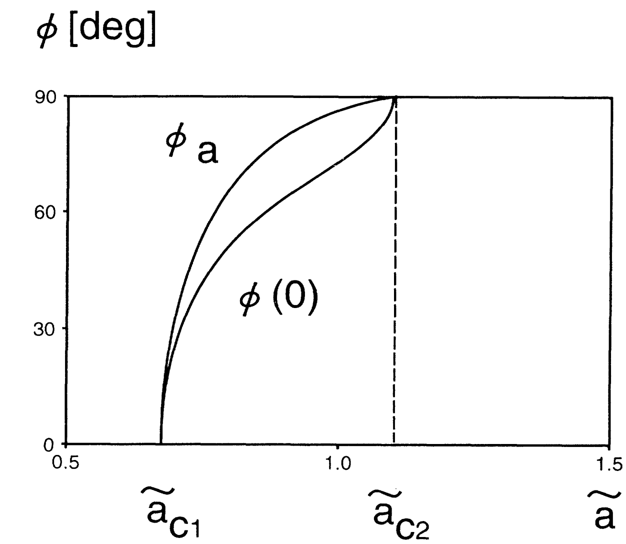

FIG. 4. Same as Fig. 3 except for $K_s < \sqrt{AK_v}$.

of $\varphi(0)$. The magnetization in the inner part of the film saturates to in-plane direction gradually with the thickness, while the direction at the surface insists at about 40° in Fig. 3.

On the other hand, for $K_s < \sqrt{AK_v}$, a different thickness dependence of configuration, as shown in Fig. 4, is obtained. Namely, there exists a second critical thickness a_{c2}, above which the magnetization is totally aligned within the film. The spin-reorientation phase transition is continuous.[4] The thickness a_{c2} is again analytically derived from (4) by noting $\varphi_a = \pi/2$, as

$$a_{c2} = \sqrt{A/K_v}\tanh^{-1}(K_s/\sqrt{AK_v}) \ . \tag{9}$$

It is then clear from (6) and (9) that in the present approach, one always has $a_{c1} < a_{c2}$ for $0 < K_s/\sqrt{AK_v} < 1$.

For values of magnetic constants given in the preceding subsection, except for $K_s = 0.7\ \mathrm{mJ/m^2}$, which makes $K_s < \sqrt{AK_v}$, one has $a_{c1} \simeq 12$ Å and $a_{c2} \simeq 17$ Å. The resultant phase diagram is shown in Fig. 5 for the case of $K_s < \sqrt{AK_v}$.

C. Phase transition with temperature

We can also discuss the spin-reorientation phase transition with temperature in the film with fixed thickness, by considering (1) as a mean-field expression for the free energy. For simplicity, the effect of thermal fluctuation is included only in the exchange stiffness as $A = A_0(1 - T/T_c)$.

For a film of $a = 15$ Å with $T_c = 270\,°\mathrm{C}$, $A_0 = 2.0 \times 10^{-9}$ mJ/m and with other quantities the same as those given in Sec. II A, the variance of the spin direction at $z = a$ is shown in Fig. 6. At the ground state, the perpendicular phase is stable. As the temperature increases to values higher than a critical temperature T_R, the free energy favors the in-plane phase. The onset of in-plane magnetization in the present approximation is described by $\beta_T = \frac{1}{2}$ as

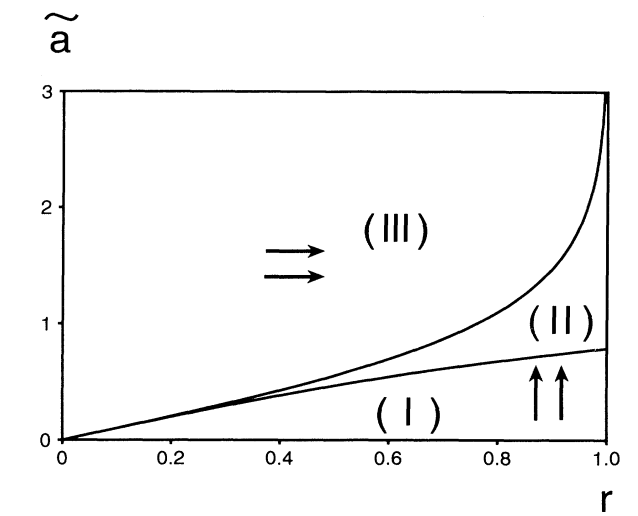

FIG. 5. Phase diagram for $K_s < \sqrt{AK_v}$, where $r \equiv K_s/\sqrt{AK_v}$. The phase boundaries are given by $\tilde{a} = \tanh^{-1} r$ (the upper curve) and $\tilde{a} = \tan^{-1} r$ (the lower curve): (I) perpendicular phase, (II) intermediate phase, and (III) in-plane phase.

$$\varphi_a \sim \left[\frac{T}{T_R} - 1 \right]^{\beta_T} \quad \text{for } T \geq T_R \ , \tag{10}$$

as depicted in Fig. 6. The spin-reorientation transition temperature is determined by

$$\left[A_0 K_v \left[1 - \frac{T_R}{T_c}) \right] \right]^{1/2} \times \tan \left[a \left[\frac{K_v}{A_0(1 - T_R/T_c)} \right]^{1/2} \right] = K_s \ . \tag{11}$$

It is about $T_R \simeq 160\,°\mathrm{C}$ for the system described above, which is quite below the Curie temperature.

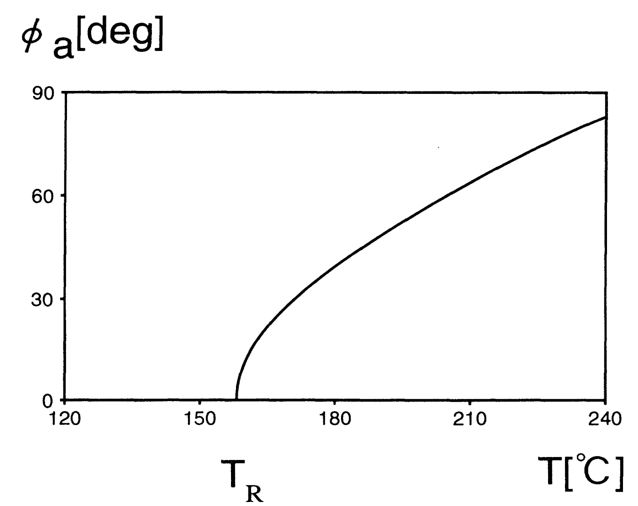

FIG. 6. Spin-reorientation phase transition with temperature.

III. THICKNESS DEPENDENCE OF VOLUME ANISOTROPY

Let us investigate the energy stocked in the film per unit area. Integrating (1), one obtains the following energy expression as a function of φ_a for $a > a_{c1}$,

$$\gamma = -aK_v \sin^2\varphi_a + K_s \left[\frac{cn[a\sqrt{K_v/A}, \sin\varphi_a]}{dn[a\sqrt{K_v/A}, \sin\varphi_a]} \right]^2 \sin^2\varphi_a$$
$$+2\left\{ E\left[\frac{\pi}{2}, \sin\varphi_a\right] - E\left[\sin^{-1}\left[\frac{cn[a\sqrt{K_v/A}, \sin\varphi_a]}{dn[a\sqrt{K_v/A}, \sin\varphi_a]}\right], \sin\varphi_a\right]\right\}$$
$$-2\cos^2\varphi_a \left\{ F\left[\frac{\pi}{2}, \sin\varphi_a\right] - F\left[\sin^{-1}\left[\frac{cn[a\sqrt{K_v/A}, \sin\varphi_a]}{dn[a\sqrt{K_v/A}, \sin\varphi_a]}\right], \sin\varphi_a\right]\right\} . \tag{12}$$

The resultant thickness dependence of the stored energy is shown in Fig. 7. The effective anisotropy defined by $\gamma = -a\hat{K}_v$ is shown in Fig. 8. It is found that its asymptote for large thickness is given as

$$\hat{K}_v \simeq K_v - \frac{E_s}{a} . \tag{13}$$

The result explains analytically the inverse thickness dependence of the effective anisotropy, which has been adopted *a priori* in the literature[1,3,4] to explain the experimentally observed spin-reorientation transition. The quantity E_s should be considered as the effective surface anisotropy, which is observed in experiment. For the case of $K_s > \sqrt{AK_v}$, one finds

$$E_s = 2\sqrt{AK_v} - \frac{AK_v}{K_s} , \tag{14}$$

for large thickness limit. Therefore, there are two contributions to the surface energy, one from the energy of the bending structure $\sqrt{AK_v}$ and the other governed by the ratio between it and the surface anisotropy. For the case of $K_s < \sqrt{AK_v}$, (13) is established exactly for $a > a_{c2}$. No energy of the bending structure is involved, and one has $E_s = K_s$.

IV. SUMMARY

A variational study to determine the magnetic configuration in magnetic thin film is performed. The relation between the exchange stiffness, in-plane volume anisotropy, and the perpendicular surface anisotropy is addressed analytically.

It is found that when the thickness is smaller than a critical value a_{c1}, the magnetization is aligned normally to the film. In the case of $K_s > \sqrt{AK_v}$, the saturation of magnetization is gradual, and the direction of magnetiza-

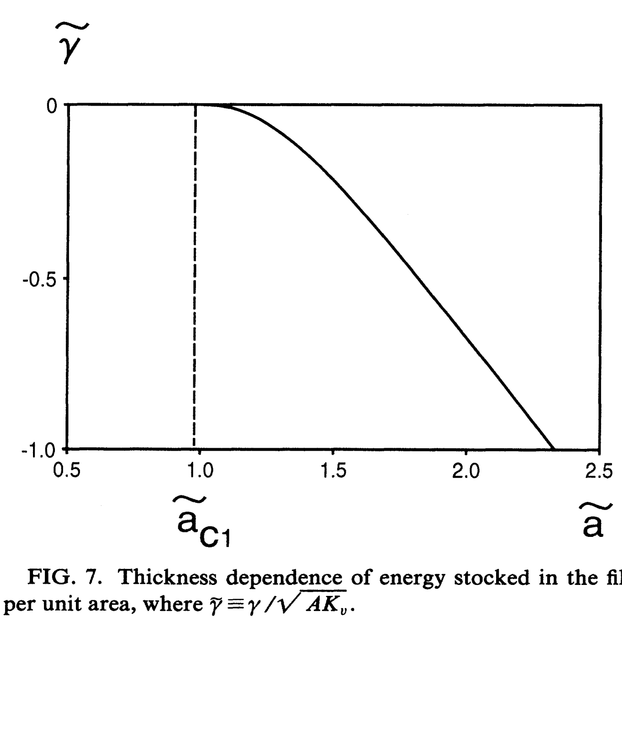

FIG. 7. Thickness dependence of energy stocked in the film per unit area, where $\tilde{\gamma} \equiv \gamma/\sqrt{AK_v}$.

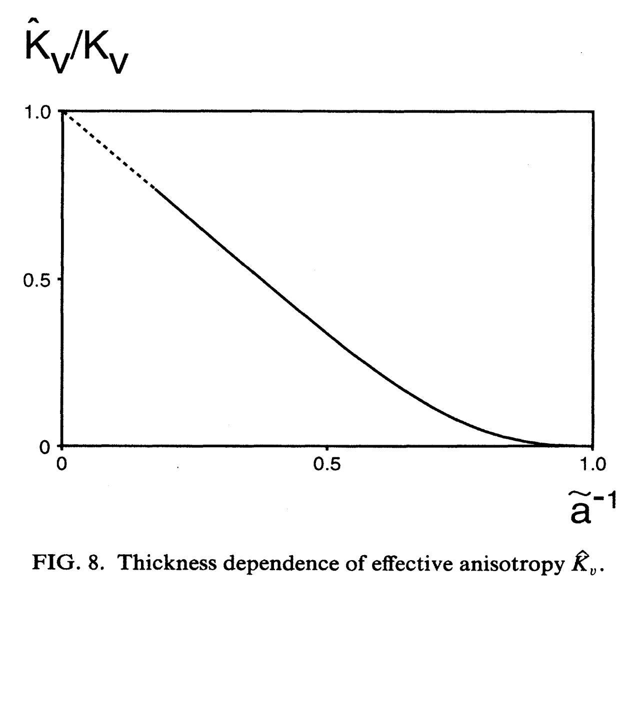

FIG. 8. Thickness dependence of effective anisotropy $\hat{K}_v$.

tion on the surface takes an intermediate direction even at the infinite thickness limit. On the other hand, in the case of $K_s < \sqrt{AK_v}$, there exists a second critical thickness a_{c2}, above which the magnetization is aligned completely in plane. These different stable configurations in the large thickness limit, coming from the relationship between the surface anisotropy and the energy of the bending structure, which is proportional to the domain-wall energy in bulk, is expected to be detected experimentally.

The surface anisotropy is treated analytically, and it becomes clear that the total effective anisotropy shows an inverse thickness dependence in a wide range of thicknesses. Therefore, our result verifies the usually adopted ansatz. Large-scale Monte Carlo simulations are now in progress in order to clarify the effect from fluctuation in the spin-reorientation phase transitions for Heisenberg models in film geometry, where surface anisotropy and dipole-dipole interactions play important roles.

[1] C. Chappert and P. Bruno, J. Appl. Phys. **64**, 5736 (1988).

[2] R. Allenspach, M. Stampanoni, and A. Bischof, Phys. Rev. Lett. **65**, 3344 (1990).

[3] V. Grolier, J. Ferre, A. Maziewski, E. Stefanowicz, and D. Renard, J. Appl. Phys. **73**, 5939 (1993).

[4] H. Fritzsche, J. Kohlhepp, H. J. Elmers, and U. Gradmann, Phys. Rev. B **49**, 15 665 (1994).

[5] Dongqi Li, M. Freitag, J. Pearson, Z. Q. Qiu, and S . D. Bader, Phys. Rev. Lett. **72**, 3112 (1994).

[6] H. Wakabayashi, H. Notarys, and T. Suzuki, J. Magn. Soc. Jpn. **15**, Suppl. S1, 87 (1991).

[7] X. Hu and Y. Kawazoe, Phys. Rev. B **49**, 3294 (1994); X. Hu, T. Yorozu, Y. Kawazoe, S.Ohnuki, and N. Ohta, IEEE Trans. Magn. **MAG-29**, 3790 (1993); Y. Kawazoe, X. Hu, and S. Honma, Mater. Res. Soc. Symp. Proc. **131**, 513 (1993); X. Hu and Y. Kawazoe, J. Appl. Phys. **75**, 6486 (1994); X. Hu and Y. Kawazoe, Acta Metall. (to be published).

[8] J. J. Krebs, B. T. Jonker, and G. A. Prinz, J. Appl. Phys. **63**, 3467 (1988).

[9] M. Stampanoni *et al.*, Phys. Rev. Lett. **59**, 2483 (1987).

[10] D. Pescia and V. L. Pokrovsky, Phys. Rev. Lett. **65**, 2599 (1990).

[11] D. P. Pappas, K. P. Kamper, and H. Hopster, Phys. Rev. Lett. **64**, 3179 (1990).

[12] N. D. Mermin and H. Wagner, Phys. Rev. Lett. **17**, 1133 (1966).

[13] M. Bander and D. L. Mills, Phys. Rev. B **38**, 12 015 (1988); R. G. Erickson and D. L. Mills, *ibid.* **43**, 11 527 (1990).

[14] L. Néel, J. Phys. Rad. **15**, 376 (1954).

[15] G. T. Rado, Phys. Rev. B **26**, 295 (1982); L. Zhang and G. T . Rado, *ibid.* **36**, 7071 (1987); R. J. Hicken, G. T. Rado, G. Xiao, and C. L. Chien, Phys. Rev. Lett. **64**, 1820 (1990); R. J. Hicken and G. T. Rado, Phys. Rev. B **46**, 11 688 (1992).