### Size effects in frustrated dipolar spin systems

A. V. Klopper\* and R. L. Stamps

School of Physics M013, University of Western Australia, 35 Stirling Hwy, Crawley, Western Australia 6009, Australia (Received 16 November 2004; revised manuscript received 22 September 2006; published 1 December 2006)

The effect of classical dipole interactions on the paramagnetic to glass phase transition is studied in threeand quasi-two-dimensional site-disordered Ising spin systems. A field theory is developed using a Gaussian variational approach to estimate transition temperatures and study phase stability. Long-range dipolar interactions are found to permit a transition to the glass state. An investigation of the role of relative length scales within the model highlights restrictions on the existence of the glass phase in the two-dimensional case, and this effect is shown to be sensitively dependent on Ising spin geometry.

DOI: 10.1103/PhysRevB.74.214402

PACS number(s): 64.60.My, 75.10.Nr, 75.30.Kz, 75.70.Ak

# I. INTRODUCTION

It is well known that dipolar interactions can have pronounced effects on long-range ferromagnetic order in spin systems.<sup>1</sup> There is some evidence that these interactions can also play a significant role in the establishment of glassy order. Indeed, results from simulations of Ruderman-Kittel-Kasuya-Yosida coupled classical vector spins suggest that a spin-glass phase only occurs when dipolar interactions are present.<sup>2</sup> It would appear as though local magnetic anisotropies arising from dipolar shape effects are important for stabilizing the glassy-ordered state.<sup>3</sup>

Understanding the effect of dipolar interactions on the spin-glass phase is valuable for experimental studies of dilute magnetic alloys. In these systems clustering of moments is possible and can lead to large effective local moments which may produce appreciable dipolar fields.<sup>4</sup> In particular, dipolar effects can be significant in insulating alloys where exchange interactions are generally short ranged. This includes systems such as  $Eu_xSr_{1-x}S$ ,<sup>5</sup>  $Eu_xSr_{1-x}Te$ ,<sup>6</sup> LiHo<sub>x</sub>Y<sub>1-x</sub>F<sub>4</sub>,<sup>7</sup> LiTb<sub>x</sub>Y<sub>1-x</sub>F<sub>4</sub>,<sup>8</sup> and certain granular alloys.<sup>9,10</sup> For sufficiently dilute systems it is possible for the average dipole interaction strength per cluster to be comparable to the direct exchange between neighboring clusters, averaged per cluster.<sup>11</sup>

It has been suggested that such clusters may be effectively modeled as superspins.<sup>12</sup> This analogy is convenient since considerable attention has been paid in recent years to the existence of a crossover to superspin-glass behavior in threedimensional randomly distributed assemblies of singledomain nanoparticles.<sup>13–15</sup> For certain densities and narrowsize distributions, the requisite frustration invoking such a transition is attributed to prevailing dipolar interactions,<sup>16,17</sup> at least in cases for which the production method ensures exclusion of the direct exchange between magnetic cores.<sup>18</sup> With these examples in mind, we develop a model for a site-disordered assembly of classical uniaxial (Ising) spins interacting via classical dipole interactions.

One feature characteristic of the dipolar interaction is a strong dependence on sample shape. The dipole interaction energy is given by the expression

$$E_{ij} = J(1 - 3\cos^2\theta_{ij})S_iS_j/r_{ij}^3,$$
 (1)

where J is the coupling constant,  $S_i$  is the spin variable at site i, and  $\theta_{ii}$  is the angle subtended by the spin axis and the

vector joining spins at sites *i* and *j*, which has magnitude  $r_{ij}$ . It follows from the form of this function that ferromagnetic ordering is preferred along the axial direction and antiferromagnetic order in directions perpendicular to the spins.<sup>19</sup> The implications of this shape dependence in the case of a random system are twofold. The first is the possibility for frustration between spins which may lead to a glass phase, to which we have already alluded. The second relates to interesting geometry-related effects in low dimensions; dipoles aligned perpendicular to the plane of a thin film produce strong demagnetising fields in contrast to dipoles aligned in the plane.<sup>20</sup> In this paper we concentrate on these two geometries in the context of our Ising model description, which is appropriate for certain dilute compounds created in a strong magnetic field.<sup>21</sup>

The dipolar interactions in two dimensions are approximated by expressions appropriate to a thin-film geometry and represent averages of dipolar sources taken across the thickness of a very thin film. Cutoffs are introduced in the Fourier expansion of the resultant potential, consistent with the thin-film approximation, in order to remove a longwavelength divergence. These approximations are reasonable for very thin films and allow us to avoid the appearance of unbounded fluctuations associated with instabilities that can exist for two-dimensional (2D) isotropic systems. We investigate the phase space of this system, focusing on the effects of these wave number cutoffs.

The paper is organised as follows. The theory is outlined in Sec. II, based on Refs. 22–24. In our approach a variational method using a Gaussian trial function provides a lowest-order quadratic correction to mean-field theory by averaging over fluctuations in the Ising Hamiltonian.<sup>25</sup> The replica-symmetric solution to the resulting set of selfconsistent order parameter equations is examined in Sec. III. Allowed phases and stability are discussed in terms of a general position-dependent potential. The form of the dipolar interaction potential is derived in Sec. IV by calculating the second-order correction to the leading-order term in a multipole expansion. Shape and size effects are discussed in closing.

#### **II. SITE-DISORDER MODEL**

The system comprises a finite number N of spins  $S_i$  placed randomly at positions  $\mathbf{r}_i$  throughout volume V. The number of particles remains the same for each realization of the disorder. Interactions are controlled by the pairwise potential J, which depends solely on spin separation. The Ising Hamiltonian takes the standard form

$$\mathcal{H} = -\frac{1}{2} \sum_{ij} J(\mathbf{r}_i - \mathbf{r}_j) S_i S_j.$$
(2)

This gives rise to a partition function which contains a trace over all possible configurations of the degrees of freedom,  $S_i$ . A Hubbard-Stratonovich transformation allows the partition function to be expressed as

$$Z_{N} = \sum_{(S)} \int \mathcal{D}\phi(\det\beta J)^{-1/2} \exp\left(-\frac{1}{2\beta} \int \int \phi(\mathbf{r}) J^{-1}(\mathbf{r} - \mathbf{r}') \times \phi(\mathbf{r}') d\mathbf{r} d\mathbf{r}' + \sum_{i}^{N} \phi(\mathbf{r}_{i}) S_{i}\right).$$
(3)

An average over the disorder in the partition function is obtained by integrating over position using the flat measure  $\frac{1}{V^N}\int_V \prod_i^N d\mathbf{r}_i$  (Ref. 24). The free energy follows from this expression, and the problem of averaging over a logarithm is dealt with by weighting this energy with a Poisson distribution and employing the replica trick.<sup>26</sup> One assumes that the system has been taken from a larger system of volume *V*, in which the spins have mean concentration  $\rho$  and are distributed according to the probability function

$$P(N) = e^{-\rho V} \frac{(\rho V)^N}{N!}.$$
(4)

The averaged free energy  $\overline{F}_N = -\ln \overline{Z}_N / \beta$  may then be expressed<sup>38</sup> in terms of the replicated partition function  $Z_N^n$ , which is identified as the product of *n* identical replicas of the system, each representing the same realization of disorder. The free energy is given by the weighted sum

$$-\beta F = -\beta \sum_{N} P(N)\overline{F}_{N} = \lim_{n \to 0} \sum_{N} P(N) \left(\frac{\overline{Z}_{N}^{n} - 1}{n}\right), \quad (5)$$

such that

$$e^{-n\beta F} = e^{-\rho V} \sum_{N} \frac{(\rho V)^{N}}{N!} \overline{Z}_{N}^{n}.$$
 (6)

Since the interaction potential is position dependent, one cannot obtain the free energy expression exactly and a variational approach is taken. This involves defining a variational free energy, which forms an upper bound for the free energy and can be solved using a quadratic trial Hamiltonian.

The weighted free energy can be recast in terms of an average in the trial ensemble,

$$e^{-n\beta F} = e^{-\rho V} \langle e^{-(\mathcal{H}[\phi] - \mathcal{H}_t[\phi])} \rangle_t e^{-F_t}, \tag{7}$$

where  $\mathcal{H}$  is the Hamiltonian associated with this free energy and is given by identifying the right-hand side of Eq. (6) with  $e^{-\rho V} \int \mathcal{D} \phi e^{-\mathcal{H}}$ .  $\mathcal{H}_t$  is the trial Hamiltonian which describes a system with free energy given by  $e^{-F_t} = \int \mathcal{D} \phi e^{-\mathcal{H}_t}$ . Note that the ensemble average in Eq. (7) is defined selfconsistently according to this expression. In this case, the trial Hamiltonian is chosen as

$$\mathcal{H}_{t} = \frac{1}{2} \int \sum_{ab} \left[ \phi_{a}(\mathbf{r}) - \bar{\phi}_{a} \right] G_{ab}^{-1}(\mathbf{r} - \mathbf{r}') \left[ \phi_{b}(\mathbf{r}') - \bar{\phi}_{b} \right] d\mathbf{r} d\mathbf{r}'.$$
(8)

The matrix  $G_{ab}(\mathbf{r}-\mathbf{r}') \equiv \langle \phi_a(\mathbf{r}) \phi_b(\mathbf{r}') \rangle_c$  is the connected correlation function for which the trial free energy is the generating functional.<sup>27</sup>

The expression for the free energy in Eq. (7) can be further simplified by invoking the minimum principle<sup>28</sup> which states

$$\langle e^{-(x_1 - x_2)} \rangle \ge e^{-\langle x_1 - x_2 \rangle}. \tag{9}$$

Using this inequality, the bounded free energy is written as<sup>24</sup>

$$n\beta F_{var} = \rho V + \langle \mathcal{H} - \mathcal{H}_t \rangle_t + F_t \ge n\beta F.$$
(10)

The averaged term here in Eq. (10) represents the difference between the Hamiltonian corresponding to the averaged, replicated Ising system and the trial Hamiltonian, averaged in the trial ensemble. It should be noted that in the resulting expression for the variational free energy, the trace over  $\ln \beta J$  comprises spatial summation only, whereas the trace over  $\ln G$  sums both spatial variables and replicas. This free energy is

$$n\beta F_{var} = \rho V + \frac{1}{2\beta} \int \int \sum_{a}^{n} \left[ \bar{\phi}_{a} J^{-1} (\mathbf{r} - \mathbf{r}') \bar{\phi}_{a} + G_{aa} (\mathbf{r} - \mathbf{r}') J^{-1} (\mathbf{r} - \mathbf{r}') \right] d\mathbf{r} d\mathbf{r}' + \frac{n}{2} \mathrm{Tr} (\ln \beta J - 1) - \rho \int_{V} \Omega d\mathbf{r} - \frac{1}{2} \mathrm{Tr} (\ln G).$$
(11)

This expression is analogous to the first equality in Eq. (3.3) of Ref. 24. The third term has been corrected,<sup>29</sup> and the fourth term involves an integral over a reduced partition function for the system in the Gaussian approximation of the form

$$\Omega = \sum_{(S)} \exp\left(\sum_{a} \overline{\phi}_{a} S_{a} + \frac{1}{2} \sum_{ab} S_{a} G_{ab}(\mathbf{r}, \mathbf{r}) S_{b}\right).$$
(12)

The variational equations are obtained by minimizing the free energy with respect to the mean field and the matrix appearing in the trial Hamiltonian. Application of the variational derivatives  $\frac{\delta F_{var}}{\delta \overline{\phi}_a} = \frac{\delta F_{var}}{\delta G_{ab}} = 0$  results in expressions for averages over  $S_a$  and  $S_a S_b$ ,

$$\frac{\phi_a}{\beta} J^{-1}(\mathbf{r} - \mathbf{r}') \,\delta(\mathbf{r} - \mathbf{r}') = \rho V \langle S_a \rangle_{\Omega}, \tag{13a}$$

$$\left(\frac{\delta_{ab}}{\beta}J^{-1}(\mathbf{r}-\mathbf{r}') - G^{-1}_{ab}(\mathbf{r}-\mathbf{r}')\right)\delta(\mathbf{r}-\mathbf{r}') = \rho V \langle S_a S_b \rangle_{\Omega},$$
(13b)

where

$$\langle S_a \rangle_{\Omega} = \sum_{(S)} S_a \exp\left(\sum_a \bar{\phi}_a S_a + \frac{1}{2} \sum_{ab} G_{ab}(\mathbf{r}, \mathbf{r}) S_a S_b\right),$$
(14a)

$$\langle S_a S_b \rangle_{\Omega} = \sum_{(S)} S_a S_b \exp\left(\sum_a \bar{\phi}_a S_a + \frac{1}{2} \sum_{ab} G_{ab}(\mathbf{r}, \mathbf{r}) S_a S_b\right)$$
(14b)

are the expectation value of the replica-dependent spin,  $\langle S_a \rangle \Omega$ , and the correlation function of spins between replicas,  $\langle S_a S_b \rangle \Omega$ . These terms are related to the physical operators of the system. The spin-density operator is defined as

$$m_a(\mathbf{r}) = \sum_i \,\delta(\mathbf{r} - \mathbf{r}_i)S_i^a \tag{15}$$

and appears in the replicated, unaveraged action as a source for the field,  $\phi_a$  (Ref. 24). The Edwards-Anderson spin-glass operator is analogously written as<sup>26</sup>

$$q_{ab}(\mathbf{r}) = \sum_{i} \,\delta(\mathbf{r} - \mathbf{r}_{i})S_{i}^{a}S_{i}^{b}.$$
(16)

The expectation value of this operator indicates the orderdisorder transition.

In order to examine the expectation values of these operators within the bounds of the Gaussian approximation, it is convenient to introduce sources for the magnetization and spin glass operators. A linear response approach circumvents the problem of being unable to express the relationship between  $q_{ab}(\mathbf{r})$  and  $\phi_a$  directly. The sources are  $h_a(\mathbf{r})$  and  $j_{ab}(\mathbf{r})$ for the magnetization and spin-glass operators, respectively. The partition function becomes a generating functional for the correlation function and the free energy a generating functional for the connected correlation functions. The only part of the free energy affected by these sources is the reduced partition function

$$\Omega(\mathbf{r}) = \sum_{(S)} \exp\left(\sum_{a} \left[\bar{\phi}_{a}(\mathbf{r}) + \beta h_{a}(\mathbf{r})\right]S_{a} + \frac{1}{2}\sum_{ab} \left[G_{ab}(\mathbf{r},\mathbf{r}) + \beta j_{ab}(\mathbf{r})\right]S_{a}S_{b}\right).$$
(17)

Following Dean and Lancaster,<sup>24</sup> the spin density operator takes the form

$$m_a(\mathbf{r}) = -\frac{1}{\beta} \frac{\delta(n\beta F_{var})}{\delta h_a(\mathbf{r})} = \rho V \langle S_a \rangle_{\Omega(\mathbf{r})}, \qquad (18)$$

where the function  $\langle S_a \rangle_{\Omega(\mathbf{r})}$  is analogous to that defined in Eq. (14a). The first variational equation (13a) thus gives  $m_a(\mathbf{r}) = \overline{\phi}_a / \beta J(\mathbf{0})$ , which is in fact position independent. The expectation value of the spin-glass operator is similarly determined as

$$q_{ab}(\mathbf{r}) = -\frac{2}{\beta} \frac{\delta(n\beta F_{var})}{\delta j_{ab}(\mathbf{r})} = \rho V \langle S_a S_b \rangle_{\Omega(\mathbf{r})}, \tag{19}$$

where the form of the function  $\langle S_a S_b \rangle_{\Omega(\mathbf{r})}$  assumes that of its source-independent counterpart in Eq. (14b).

## **III. ORDER PARAMETERS**

Order parameters are derived by examining the replicasymmetric case in momentum space. The field is taken to be replica independent  $\overline{\phi}_a \rightarrow \overline{\phi}$  and the matrix  $\widetilde{G}_{ab}(\mathbf{k})$  is parametrized to be  $\widetilde{f}(\mathbf{k}) + \widetilde{g}(\mathbf{k})$  on the diagonal and  $\widetilde{g}(\mathbf{k})$  elsewhere. In Fourier coefficients, the free energy takes the form

$$\beta F_{var} = -\frac{\rho V}{2} f(\mathbf{0}) - \rho V \int_{-\infty}^{\infty} \frac{\mathrm{d}x}{\sqrt{2\pi}} e^{-x^2/2} \ln\{2 \cosh[\bar{\phi} + \sqrt{g(0)}x]\} \\ + \frac{\bar{\phi}^2}{2\beta} J^{-1}(\mathbf{0}) + \frac{V}{2\beta} \int \frac{\mathrm{d}^d \mathbf{k}}{(2\pi)^d} [\tilde{f}(\mathbf{k}) + \tilde{g}(\mathbf{k})] \tilde{J}^{-1}(\mathbf{k}) \\ + \frac{V}{2} \int \frac{\mathrm{d}^d \mathbf{k}}{(2\pi)^d} [\ln \beta \tilde{J}(\mathbf{k}) - 1] \\ - \frac{V}{2} \int \frac{\mathrm{d}^d \mathbf{k}}{(2\pi)^d} \left( \ln \tilde{f}(\mathbf{k}) - \frac{\tilde{g}(\mathbf{k})}{\tilde{f}(\mathbf{k})} \right).$$
(20)

The variational equations for the replica-symmetric case give the magnetization expectation value as  $\langle m_a(\mathbf{r}) \rangle \rightarrow \langle m \rangle$  with

$$\langle m \rangle = \frac{\overline{\phi}}{\beta J(\mathbf{0})} = \rho V \int_{-\infty}^{\infty} \frac{\mathrm{d}x}{\sqrt{2\pi}} e^{-x^2/2} \tanh[\overline{\phi} + x\sqrt{g(\mathbf{0})}].$$
(21)

The spin-glass order parameter is expressed as  $\langle q_{ab}(\mathbf{r}) \rangle \rightarrow \langle q \rangle$  with

$$\langle q \rangle = \rho V \langle S_a S_b \rangle_{\Omega}^{a \neq b} = \rho V \int_{-\infty}^{\infty} \frac{\mathrm{d}x}{\sqrt{2\pi}} e^{-x^2/2} \tanh^2[\overline{\phi} + x\sqrt{g(0)}].$$
(22)

The form of the function  $g(\mathbf{0})$  is also given by the variational equations of this system. The variational derivatives with respect to the components of  $\tilde{G}_{ab}$  give rise to the relations

$$\widetilde{f}(\mathbf{k}) = \frac{\beta J(\mathbf{k})}{1 - (\rho V - \langle q \rangle)\beta \widetilde{J}(\mathbf{k})},$$
(23a)

$$\tilde{g}(\mathbf{k}) = \langle q \rangle \tilde{f}(\mathbf{k})^2.$$
 (23b)

In the long-range limit of small  $J(\mathbf{k})$ , the function  $g(\mathbf{0})$  can be approximated as

$$g(\mathbf{0}) = \langle q \rangle \int \frac{\mathrm{d}^d \mathbf{k}}{(2\pi)^d} \beta^2 \widetilde{J}(\mathbf{k})^2.$$
(24)

This leading-order term forms the position-dependent equivalent of the mean-field term in the Sherrington-Kirkpatrick solution.<sup>30</sup> The order parameters can be expressed self-consistently as

$$\langle m \rangle = \rho V \int_{-\infty}^{\infty} \frac{\mathrm{d}x}{\sqrt{2\pi}} e^{-x^2/2} \tanh[\beta J(\mathbf{0}) \langle m \rangle + x \sqrt{g(\mathbf{0})}],$$
(25a)



FIG. 1. Scaling function for the finite system with  $x_{\ell} = ak_{\ell}/\pi$ .

$$\langle q \rangle = \rho V \int_{-\infty}^{\infty} \frac{\mathrm{d}x}{\sqrt{2\pi}} e^{-x^2/2} \tanh^2 [\beta J(\mathbf{0}) \langle m \rangle + x \sqrt{g(\mathbf{0})}].$$
(25b)

The behavior of these order parameters can be determined analytically for some limiting cases, but in general, a numerical solution is necessary. When  $\langle m \rangle = 0$ , Eq. (25a) has a solution for all  $\tilde{J}(\mathbf{k})$  and  $\langle q \rangle$ . Conversely, Eq. (25b) has a solution where  $\langle q \rangle = 0$ , and there exists a nontrivial solution only when

$$\boldsymbol{\beta} > \left( \rho V \int \frac{\mathrm{d}^d \mathbf{k}}{(2\pi)^d} \widetilde{J}(\mathbf{k})^2 \right)^{-1/2}.$$
 (26)

Phase diagrams may be constructed by using this result coupled with the nontrivial solution to Eq. (25a), which is be obtained by linearizing both order parameter equations for small  $\langle m \rangle$  and  $\langle q \rangle$  and specifying the form of the interaction. The phases are paramagnetic with  $\langle m \rangle = \langle q \rangle = 0$ , ferromagnetic with  $\langle m \rangle \neq 0, \langle q \rangle \neq 0$ , and spin glass with  $\langle m \rangle = 0, \langle q \rangle \neq 0$ .

Stability criteria can also be determined by minimizing the first variational derivative of the bounded free energy with respect to the matrix in the reduced partition function  $\frac{\delta^2 F_{var}}{\delta \tilde{G}_{ab}(\mathbf{k}) \delta \tilde{G}_{cd}(\mathbf{k}')}$  (Ref. 31). The important result relates the fact that the converse of the condition in Eq. (26) forms the stability criterion in phase space wherever  $\langle m \rangle = 0$ . We interpret



FIG. 2. Spin-glass order parameter for the finite system with  $x_{\ell} = ak_{\ell}/\pi$ .

this as indicating that the paramagnetic phase is stable in this model, regardless of the form of the interaction. It is bounded below by a critical temperature defining the transition to an unstable spin-glass phase.

#### **IV. DIPOLAR INTERACTION**

The dipole interaction potential is determined by considering an assembly of dipoles with energy,

$$E = \frac{1}{2} \int d^d \mathbf{r} \boldsymbol{\mu}_0 S(\mathbf{r}) \hat{\mathbf{I}} \cdot \boldsymbol{\nabla} \boldsymbol{\phi}(\mathbf{r}), \qquad (27)$$

where  $\mu_0$  is the permeability of free space,  $\hat{\mathbf{I}}$  is a unit vector in the direction of the Ising-spin axis, and  $\phi(\mathbf{r})$  is the dipole field potential at  $\mathbf{r}$ , satisfying the magnetostatic form of Maxwell's equations,

$$-\nabla^2 \phi(\mathbf{r}) + \nabla \cdot S(\mathbf{r})\hat{\mathbf{I}} = 0.$$
<sup>(28)</sup>

This is solved using a Green's function  $g_{\phi}(\mathbf{r}, \mathbf{r}')$ , subject to the condition

$$\nabla^2 g_{\phi}(\mathbf{r}, \mathbf{r}') = \delta(\mathbf{r} - \mathbf{r}'), \qquad (29)$$

such that

$$\phi(\mathbf{r}) = \int d^d \mathbf{r}' g_{\phi}(\mathbf{r}, \mathbf{r}') \nabla' \cdot S(\mathbf{r}') \hat{\mathbf{I}}.$$
 (30)

The total dipole energy is therefore

$$E = \frac{\mu_0}{2} \int d^d \mathbf{r} S(\mathbf{r}) \hat{\mathbf{I}} \cdot \nabla \int d^d \mathbf{r}' g_{\phi}(\mathbf{r}, \mathbf{r}') \nabla' S(\mathbf{r}') \cdot \hat{\mathbf{I}}.$$
 (31)

The Green's function for the magnetostatic potential can be expanded in a Fourier series. Using Eq. (29), the Green's function in three dimensions is

$$g_{\phi}(\mathbf{r},\mathbf{r}') = -\int \frac{\mathrm{d}^{3}\mathbf{k}}{(2\pi)^{3}} \frac{e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}}{k^{2}}.$$
 (32)

Equating the resulting energy with the Ising Hamiltonian in Eq. (2) gives the potential in momentum space as



FIG. 3. (a) Scaling function and (b) order parameter for the infinite system with  $x_u = ak_u/\pi$ . The inset in (a) shows the shaded region in greater detail (albeit on the same axes).

$$\widetilde{J}_{3}(\mathbf{k}) = -(2\pi)^{3} \mu_{0} \left(\frac{\widehat{\mathbf{I}} \cdot \mathbf{k}}{k}\right)^{2}.$$
(33)

The same problem in a quasi-two-dimensional system is solved by taking a model system that describes an infinite, homogeneous film of finite thickness a. The system is assumed to be translationally invariant along the surface, and  $g_{\phi}(\mathbf{r},\mathbf{r}')$  is expanded in a Fourier series parallel to the surface.<sup>32</sup> In the long-wavelength regime, the relevant fields are averaged over the thickness of the film under the assumption that the dipole source magnitude and direction across the film plane is uniform. This means that the spin density function is constant across the layer and vanishes outside the film as in the thin-film approximation in Ref. 33. We consider two types of potential. The first, denoted with subscript ||, refers to the geometry of spins along an axis parallel to the plane of the film. The second, denoted with subscript  $\perp$ , specifies the potential for a system with spins perpendicular to this plane. Both potentials are calculated from

$$g_{\phi}(\mathbf{r},\mathbf{r}') = -\int \frac{\mathrm{d}^{2}\mathbf{k}_{\parallel}}{(2\pi)^{2}} e^{i\mathbf{k}_{\parallel}\cdot(\mathbf{r}_{\parallel}-\mathbf{r}_{\parallel}')} \frac{e^{-k_{\parallel}|r_{\perp}-r_{\perp}'|}}{2k_{\parallel}}.$$
 (34)

Equating Eqs. (2) and (31) as before results in the following expressions:

$$\widetilde{J}_{\parallel}(\mathbf{k}_{\parallel}) = -(2\pi)^2 \frac{\mu_0(\widehat{\mathbf{I}}_{\parallel} \cdot \mathbf{k}_{\parallel})^2}{ak_{\parallel}^3} (e^{-ak_{\parallel}} + ak_{\parallel} - 1), \quad (35a)$$

$$\widetilde{J}_{\perp}(\mathbf{k}_{\parallel}) = -(2\pi)^2 \frac{\mu_0}{ak_{\parallel}} (1 - e^{-ak_{\parallel}}).$$
(35b)

#### V. RESULTS AND DISCUSSION

We calculate  $g(\mathbf{0})$  explicitly by transforming the square of these functions back into real space as described in Eq. (24). Since the dipole interaction diverges rapidly at zero distance, the minimum separation of spins must be restricted by a lattice constant, even in a randomly distributed system. Neglecting this minimum cutoff will exaggerate local fluctuations due to randomness.<sup>34</sup> We thus restrict the wave number integration with an upper cutoff  $k_u$  by assuming that the separation of spins is determined by the thickness of the monolayer, a. Then the smallest allowable separation between spins corresponds to a cutoff of  $\pi/a$ .

Similarly, we expect a lower bound  $k_{\ell}$  to apply for integration over the wave number. In finite systems, this will be imposed by the size of the sample, since the dipolar interaction is inherently long ranged. We can examine the effects of this constraint by plotting g(0) as a function of  $x_{\ell}=ak_{\ell}/\pi$  when  $k_u=\pi/a$ . After performing the required integration in Eq. (24), we find that the length and temperature dependences of all three systems (3D and 2D) is the same, such that g(0) may be expressed in terms of the following shape-dependent function:

$$g_d(\mathbf{0}; \boldsymbol{\beta}, \mu_0, a; x) = \boldsymbol{\beta}^2 \mu_0^2 a^{-d} \alpha_d(x),$$
 (36)

where *d* is the dimension. Note that both  $g_2(\mathbf{0}; \beta, \mu_0, a; x)$  and  $\alpha_2(x)$  differ depending on the geometry specified. Figure 1 shows the dependence of this scaling function on the lower wave number cutoff variable.

We use this relation to investigate the nontrivial solution to the order parameter function in Eq. (25b). In Fig. 2, we plot the behavior of the spin-glass order parameter in the regime for which net magnetization is zero and  $\rho V$  unity, as the lower wave number cutoff is varied. The example given takes parameter values which correspond to those appropriate for a superspin system near the critical region, with T=50 K and a=50 nm (Ref. 35). It is clear that in the threedimensional case, there is a nonzero solution to Eq. (25b) corresponding to a transition to spin-glass behavior, for all lower-wave-number cutoffs  $k_{\ell} < k_{\mu}$ . The two-dimensional geometries have the effect of rounding out this transition. In these cases, the spin-glass phase appears to deteriorate and eventually vanish for smaller lower-wave-number cutoffs, indicating that spin-glass order cannot be sustained in twodimensional systems, below a critical system size.

The nontrivial solution to Eq. (25b) with  $\langle m \rangle = 0$  exists in all three cases of an infinite system, for which the lowerwave-number cutoff is zero. It is instructive to examine the effect of upper cutoff in this particular limit. As a function of  $x_u = ak_u/\pi$ , g(0) scales in the form given in Eq. (36), with the scaling function  $\alpha_d(x)$  resembling those plotted in Fig. 3(a). The corresponding dependence of the spin-glass order parameter on the higher-wave-number cutoff is shown in Fig. 3(b). The three-dimensional system permits a nonzero solution to the spin-glass order parameter equation, except in the unphysical case of  $k_u \leq k_\ell$ . Again, the sensitive dependence of the dipole interaction on sample shape is manifest in the rounding of the transition in two dimensions. Both two-dimensional geometries exhibit a minimum upper cutoff below which spin-glass ordering cannot exist in the infinite system. The limiting cutoff in the spin in-plane case greatly exceeds that in the out-of-plane case, indicating that the spin-glass phase in the infinite system is more unstable to fluctuations when there are both ferromagnetic and anti-ferromagnetic competing interactions.

# **VI. CONCLUSION**

The effects of long-range dipolar interactions on the boundary between paramagnetic and spin-glass phases have been studied for a site-disordered Ising spin system in a thinfilm geometry. A variational field theory has been used to construct self-consistent order parameter equations for a system characterized by dipolar interactions. Wave number cutoffs provide a means of avoiding unphysical contributions to the free energy due to artifacts in the continuum approximation. In addition, these cutoffs facilitate a study of finite-size effects associated with the shape dependence of the dipolar interaction. In particular, our results show that the spin-glass phase in an infinite quasi-two-dimensional system of sitedisordered Ising spins interacting via a long-range dipolar potential can only be predicted within this model if one probes a sufficiently large wave number range. That is, neglecting larger wave numbers can have the effect of suppressing the glass transition. That this effect is more prominent in the spins in-plane case relates the sensitive shape dependence of the low-dimensional dipolar spin system.

We note that one cannot rule out the possibility that the Gaussian variational approximation is too severe to accurately describe allowed phases. However, one may consider this ansatz justified for systems in which each spin interacts with many effective neighbors, as is the case in high-dimensional systems or in those with long-range interactions.<sup>24</sup> The variational method becomes exact in the limit that the number of components of the degrees of freedom is infinite.<sup>36</sup> In the case of an Ising model, then, the method is merely an approximation with limited bounds of validity. Indeed, a stability analysis of this model shows that the spin-glass state is unstable. The stability of phases in the replica-symmetric solution in general is not clear,<sup>25</sup> but the behavior of the system *above* the paramagnetic transition temperature is exact in this theory.<sup>37</sup>

#### ACKNOWLEDGMENT

We acknowledge the Australian Research Council for support.

- \*Present address: Max Planck Institute for the Physics of Complex Systems, Nöthnitzer Straße 38, 01187 Dresden, Germany. Electronic mail: avk@pks.mpg.de
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