Journal of Statistical Physics, Vol. 88, Nos. 1/2, 1997

Effect of Local Field Fluctuations on Orientational Ordering in Random-Site Dipole Systems

B. E. Vugmeister[1] **and H. Rabitz**[1]

Received October 21, 1996

Some peculiarities of dipole ordering in systems with uniaxial or cubic anisotropy with an arbitrary degree of dilution are analyzed in terms of random local field theory. The approach takes into account the effect of thermal and spatial fluctuations of the local fields acting on each particle from its neighbors with an accuracy corresponding to that of the Bethe–Paierls pair clusters approach. We show that ferromagnetic (ferroelectric) structure for uniaxial Ising dipoles distributed on a simple cubic lattice is intrinsically unstable against the fluctuations of the local fields for any concentration of the dipoles. This result is quite different from the prediction of the mean-field theory which implies the possibility of ferromagnetic ordering as a metastable state in field-cooled experiments. The local field fluctuations do not exclude, however, antiferromagnetic ordering above a certain critical concentration. Ferromagnetic ordering is possible for other types of lattice geometries and for an amorphous-like dipole distribution above a certain critical concentration. A simple physical explanation of such behavior is given based on the specific angular dependence of the dipole–dipole interaction that results in a relatively high value of the local field second moment for simple cubic lattice.

KEY WORDS: Oreintational ordering; dipole–dipole interactions; soft dipole spheres; simple cubic lattice; local field; long-range order; fluctuations.

I. INTRODUCTION

The ordering of particles due to dipole–dipole interactions arises in many contexts. Examples include ferrofluids and electrorheological fluids,(1–3) electronic and nuclear magnetism,(4, 5) ferroelectrics and electric dipole glasses,(6–8) ferroelectric nematics,(9) etc. The dipole–dipole interaction is a

---

[1] Department of Chemistry, Princeton University, Princeton, New Jersey 08544.

long-range and sign-changeable interaction that makes the analysis of dipole ordering nontrivial. Even for dipoles occupying the regular positions in a periodic lattice the situation is not completely understood. It has been established long ago(10, 12) that the nuture of the dipole ordering depends on the geometry of the lattice. In particular, conventional mean-field theory predicts that for a simple cubic lattice the energy of a ferromagnetic (ferroelectric) structure is higher than that for an antiferromagnetic structure, contrary to body-centered or face-centered cubic lattices, possessing the ferromagnetic ground state.

Also mean-field theory predicts that the ferromagnetic transition temperature, for a simple cubic lattice would be only slightly below the antiferromagnetic transition temperature. These results, although correctly reproducing the fact that for simple cubic lattice ferromagnetic type of dipole ordering would not be realized in equilibrium, imply that ferromagnetic state could be achieved as a metastable state by cooling down the system in a strong external field.

Mean-field calculations do not properly tract the stability of the structure against thermal fluctuations. A stability analysis of the ferromagnetic phase against the spin-wave fluctuations has been performed by Cohen and Keffer.(11) The conclusion was that the ferromagnetic structure is unstable against the formation of spin waves for freely rotating dipoles located at the sites of a simple cubic lattice. A stability analysis has not been done, however, for Ising uniaxial dipoles. It is apparent that in this case a different approach should be developed, since fluctuations of the spin-wave type are forbidden for anisotropic Ising spins. A stability analysis of dipole systems adequate for uniaxial symmetry will be performed in the present paper. We will show that in anisotropic systems the situation is similar to that for freely rotating dipoles and ferromagnetic ordering is not possible for a simple cubic lattice even as a metastable state.

The situation is more complex in dilute dipole systems, as the dilution is an additional source of fluctuations. Therefore a general question concerns the possibility of long-range order due to dipole–dipole interactions in dilute systems. This question for isotropic 3-dimensional dipole systems has been originally considered by Aharony,(13) who concluded that long-range ferromagnetic order is not possible for dilute isotropic dipole systems due to their instability against transverse fluctuations of the local field. The analogous conclusion for very dilute dipole systems with uniaxial or cubic anisotropy has been obtained by one of the authors,(14) concluding that very dilute dipole systems are unstable against the longitudinal fluctuations of the local field as well.

New impetus to investigate dipole ordering in dilute systems is given by the molecular dynamic simulations of Wei and Patey(9) for soft dipolar spheres. They have shown that for systems of dipole spheres there exists a reduced critical density of particles above which ferroelectric long range order is possible. Recently the conclusion that ferroelectric dipole ordering could exist for completely random spatial particle configurations (and uniaxial orientational anisotropy) has been confirmed in simulations.(15) To the same conclusion came Zhang and Widom(16) using the continuous version of the random local field theory.(17, 18)

A key to understand is the physics permitting ferroelectric ordering of randomly distributed dipole spheres while disallowing an ordering for point dipoles distributed on the simple cubic lattice. Intuitively one would expect the opposite result to be true since randomness in dipole configurations might increase the role of fluctuations. In order to explore this matter a general formalism is needed capable of spanning both situations. We will show below that random local field (RLF) theory taking into account the effect of pair correlations between dipole orientations is sufficient to elucidate the main difference between the two situations indicated above.

The remainder of the paper is organized as follows. In Section II we derive the basic equations for the local polarization given by the microscopic version of the RLF theory. For systems with uniaxial anisotropy the RLF theory is based on the exact equations for local polarization and employs the decoupling approximation in which only pair correlations are taken into account. This approximation was originally proposed by Zernike(19) and further developed by Kaneyoshi(20) (who used the techniques of differential operators), for the Ising model with nearest neighbor interactions. The advantage of the RLF theory in the microscopic form presented below is that it allows a simple and convenient way to analyze the criteria of spontaneous appearence of the stable structures of various symmetries (i.e., ferro- or antiferromagnetic ordering or other types of ordering) depending explicitly on the interaction potential and particle concentration.

The application of the formalism for the dipole–dipole interaction is given in Section III. We show that for a simple cubic lattice the effect of fluctuations of the local field prevents the appearence of ferromagnetic structure for any dipole concentration. The RLF theory predicts, however, the well defined critical concentration of dipoles above which the antiferromagnetic type of ordering takes place. We show that the absence of the ferroelectric type of ordering for a simple cubic lattice is a consequence of the specific angular dependence of the dipole–dipole interaction $\sim(1-3\cos^2\theta)$ that causes very large fluctuations of the local fields in a simple cubic lattice compared to body-centered lattices and those with a amorphous-like dipole distribution.

II. BASIC EQUATIONS

We consider Ising spins randomly occupying sites on a 3-dimensional lattice. The Hamiltonian of the system is

$$H = -\frac{1}{2} \sum_{i,j=1}' J_{ij} l_i l_j, \qquad l_i = \pm 1 \tag{1}$$

where $\sum'$ denotes the summation over the occupied sites, J_{ij} describes the interaction between spins l_i and l_j located at sites i and j with the coordinates $\mathbf{r}_i$ and $\mathbf{r}_j$, $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$.

In the development of the RLF approximation we start from the identity⁽²¹⁾

$$\langle l_i \rangle = \left\langle \tanh\left(\frac{E_i}{T}\right)\right\rangle \tag{2}$$

where $\langle \ldots \rangle$ denotes the thermal average at temperature T, and

$$E_i = \sum_j' J_{ij} l_i \tag{3}$$

is the local field acting on every dipole due to interaction with its neighbors. Eq. (2) can be written in the form

$$\langle l_i \rangle = \int_{-\infty}^{\infty} dE \tanh\left(\frac{E}{T}\right) f_i(E) \tag{4}$$

where

$$f_i(E) = \langle \delta(E - E_i) \rangle \tag{5}$$

Note that the function $f_i(E)$ in Eq. (4) is the *exact* distribution function of the local field acting on dipole i. Mean-field theory corresponds to the replacement $f_i(E) = f(E) = \delta(E - \langle \bar{E}_i \rangle)$, where the overbar denotes the additional average over spatial configurations of the particles randomly occupying the lattice sites, $\langle \bar{E}_i \rangle$ is a conventional form of the mean-field value of the local field. Klein *et al.*⁽¹⁾ introduced the so called random mean field approach representing $f_i(E) = \delta(E - \langle E_i \rangle)$. In Klein's approach only configurational fluctuations of the local fields are taken into account compared with Eq. (5) which takes into account thermal fluctuations of the local fields as well.

Using the integral representation of the delta-function and the fact that one has $l_i^{2n} = 1$ and $l_i^{2n+1} = l_i$, we rewrite Eq. (5) as

$$f(E) = \frac{1}{2\pi} \int_{-\infty}^{\infty} d\rho\, e^{i\rho E} f_{i\rho} \tag{6}$$

in which

$$f_{i\rho} = \langle \exp[-i\rho E_i] \rangle = \left\langle \prod_k [\cos(\rho J_{ik}) - i l_k \sin(\rho J_{ik})] \right\rangle \tag{7}$$

In order to calculate the phase transition temperature it is sufficient in Eq. (7) to keep only terms linear in $\langle l_i \rangle$. We also neglect correlations between different dipoles k, k' interacting with dipole i, which is equivalent to the Zernike approximation. When this is done, the expression for $f_{i\rho}$ assumes the form

$$f_{i\rho} = \prod_k \cos(\rho J_{ij}) - i \sum_j{}' \prod_{k \neq j} \cos(\rho J_{ik}) \sin(\rho J_{ij}) \langle l_j \rangle \tag{8}$$

Only the second term in Eq. (8), which depends on $\langle l_j \rangle$, contributes to Eq. (4). Taking the configurational average of the Eqs. (4) and (8) we finally obtain

$$m_i = c \sum_{j=1}^{N_0} \Phi_{ij}(T)\, m_j \tag{9}$$

where $m_i = \langle l_i \rangle$. The summation in Eq. (9) is taken over all N_0 lattice sites and $c = N/N_0$ is the atomic concentration.

$$\Phi_{ij}(T) = T \int_0^{\infty} d\rho \frac{\exp[-F_1(\rho)] \sin(J_{ij}\rho)}{\sinh(\pi \rho T/2)} \tag{10}$$

$$\exp[-F_1(\rho)] = \prod_{k \neq j} (c \cos(\rho J_{ik}) + (1-c)) \tag{11}$$

Φ_{ij} can be considered as the effective interaction between dipoles i and j renormalized by the effect of fluctuations. $\Phi_{ij} \to J_{ij}$ as $J_{ij} \to 0$, however Φ_{ij} saturates to the value $\Phi_{ij} \to 1$ as $J_{ij} \to \infty$.

While obtaining Eq. (9) we used the identity

$$\int_{-\infty}^{\infty} dE \tanh\left(\frac{E}{T}\right) \sin(\rho E) = \frac{\pi T}{\sinh(\pi \rho T/2)} \tag{12}$$

and the decoupling approximation

$$\overline{\prod_{k\neq j}\cos(\rho J_{ij})\langle l_j\rangle}=\overline{\prod_{k\neq j}\cos(\rho J_{ij})\langle l_j\rangle} \tag{13}$$

Eq. (13) is the basic equation for the evaluation of the phase transition temperature (assuming a second order phase transition). Since Eq. (13) is a translationally invariant equation, its solution can be found by spatial Fourier transform, that yields

$$m(\mathbf{q})=c\Phi(T,\mathbf{q})\,m(\mathbf{q}) \tag{14}$$

where

$$m(\mathbf{q})=\sum_j m_j e^{i\mathbf{q}\cdot\mathbf{r}_j} \tag{15}$$

$$\Phi(T,\mathbf{q})=\sum_{r_{ij}}\Phi_{ij}(T)\,e^{i\mathbf{q}\cdot\mathbf{r}_{ij}} \tag{16}$$

The bifurcation points of Eq. (15) (at which nontrivial solutions $m(q_s)\neq 0$ appear) satisfy the condition

$$c\Phi(T_c,\mathbf{q}_s)=1 \tag{17}$$

If there are two or more different solutions of Eq. (17), only the temperature $(T_c)_{\max}$, the higher temperature among possible solutions of Eq. (17), would be the true second-order transition temperature. Below $(T_c)_{\max}$ one should take into account the appearence of the order parameter corresponding to the wavevector q_s (for which $T_c=(T_c)_{\max}$) that makes invalid the linear approximation, Eq. (8), below $(T_c)_{\max}$.

In order to calculate the susceptibility we use the fluctuation–dissipation relation

$$\chi_q=\frac{1}{T}S_q \tag{18}$$

where S_q is the static structure factor

$$S_q=c+c^2\overline{\langle l_i l_j\rangle} \tag{19}$$

In order to obtain the equation for S_q we use the Ornstein–Zernike approach[22] and assume that in Eq. (2) spin l_0 has a given random orientation. We also use the definition

$$\Gamma_{i0}\equiv\overline{\langle l_i l_0\rangle}=\overline{\langle\langle l_i\rangle_0\, l_0\rangle} \tag{20}$$

where $\langle l_i \rangle_0$ is the average value of l_i at given value of l_0. The equation for Γ_{i0} can also be obtained by multiplying Eq. (2) by l_0 and averaging. In the linear approximation, Eq. (8), we obtain

$$\Gamma_{ij} = c \sum_k \Phi_{ik}(T)\, \Gamma_{kj} + \Phi_{ij}(T) \tag{21}$$

Taking the Fourier transform of Eq. (21) and using Eqs. (18) and (19), we obtain finally

$$\chi_q = \frac{c}{T} \frac{1}{1 - c\Phi(T, q)} \tag{22}$$

One can give also a convenient criteria for the existence of solutions of Eq. (17) with $T_c \geqslant 0$. First, note that the function $\Phi(T, q)$ monotonically increases with temperature. Taking the limit $T \to 0$ one can introduce the parameter

$$\xi(\mathbf{q}) = \lim_{T \to 0} c\Phi(T, \mathbf{q}) = \frac{2}{\pi} c \int_0^\infty \frac{d\rho}{\rho} \exp[-F_1(\rho)] \sum_{r_{ij}} e^{i\mathbf{q}\mathbf{r}_{ij}} \sin(J_{ij}\rho) \tag{23}$$

It is apparent from Eq. (17) that the criterium of the existence of a solution $T_c \geqslant 0$ for a given wavevector q_s is

$$\xi(\mathbf{q}_s) \geqslant 1 \tag{24}$$

In order to illustrate the quality of the formalism above, we shall calculate the transition temperature for an Ising model with ferromagnetic type interactions between nearest neighbor spins. For nearest neighbor interactions the equation for T_c follows from Eqs. (10) and (11) with the form

$$\frac{1}{T_c} = cM \int_0^\infty \frac{d\rho \sin J\rho}{\sinh(\pi\rho T_c/2)} (c \cos J\rho + 1 - c)^{M-1} \tag{25}$$

For a 3-dimensional lattice we find from Eq. (25) that $T_c(c = 1) \approx 5J$ compared with the result of a high temperature series $T_c \approx 4.5J$.(22) The critical concentration c_{cr} below which the ferromagnetic phase transition does not exist is $c_{cr} \approx 0.29$ compared with the Monte Carlo result(23) $c_{cr} \approx 0.31$. These results show that the RLF theory is a significant improvement over the conventional mean field theory (that predicts $T_c = 6$ cJ) and can be considered as a first step towards accounting for the effect of fluctuations. The accurracy of the RLF approximation is comparable to the accuracy of the Bethe–Paierls pair clusters formalism. Equation (25) also reproduces the well known fact that there is no long-range order for one-dimensional systems. For one-dimensional systems one should use the value $M=2$ in Eq. (25) for the number of nearest neighbors that results in the absence of positive solutions for T_c.

III. EFFECT OF LOCAL FIELD FLUCTUATIONS ON DIPOLE ORDERING IN A SIMPLE CUBIC LATTICE

In this section we will apply the formalism developed above to the analysis of the dipole ordering in a simple cubic lattice. For two-orientable dipoles the interaction Hamiltonian can be written in the form of Eq. (1) with

$$J_{ij}=J(\mathbf{r}_{ij})=-\frac{\mu^2}{r_{ij}^3}(1-3\cos^2\theta_{ij}) \tag{26}$$

where μ is the magnitude of the magnetic or electric dipole moment oriented along the z direction and θ_{ij} is the angle between $\mathbf{r}_{ij}$ and z. The variable l_i in Eq. (1) describes the two possible directions of the dipole moment.

As well known, for dipole–dipole interaction the integral $\int d\mathbf{r}\, J(\mathbf{r})$ depends on the boundary conditions. We will consider the boundary conditions corresponding to a thin slab with the axis z being parallel to the plane of the slab. In this case

$$\int d\mathbf{r}\, J(\mathbf{r})=\frac{4\pi}{3}\mu^2 \tag{27}$$

Such a geometry corresponds to the cancellation of the depolarizing field and to the maximum possible value of the mean-field experienced by each dipole from its neighbors. The effect of the boundary should be taken into account in the evaluation of $\Phi(T,\mathbf{q})$ at $\mathbf{q}=0$. In order to take into account the effect of boundary conditions one should replace $c\sum_{r_{ij}}$ in Eq. (16) for sufficiently large $\mathbf{r}_{ij}$ by $n\int d\mathbf{r}$, where $n=N/V$ is the dipole number density and V is the volume of the crystal.

According to Eq. (24), in order to establish the possibility of ferromagnetic or antiferromagnetic ordering one needs to calculate the parameter $\xi(q_x, q_y, q_z)$ for the corresponding wavevector. In Fig. 1 we present the values of $\xi(0,0,0)$ and $\xi(\pi/d, \pi/d, 0)$ as a function of concentration (d is the lattice constant). The calculated value of $\xi(0,0,0)$ is equal to 0.98 at $c=1$ meaning that ferromagnetic structure is unstable with respect to thermal fluctuations. The values of $\xi(0,0,0)$ decrease further with a decrease

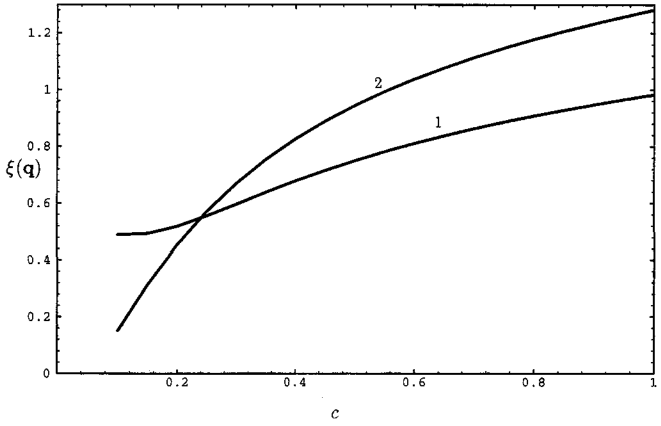

Fig. 1. Concentration dependence of the parameter $\xi(0, 0, 0)$ (1) and $\xi(\pi/d, \pi/d, 0)$ (2).

of the dipole concentration. At the same time the parameter $\xi(\pi/d, \pi/d, 0)$ corresponding to antiferromagnetic structure is approximately equal to 1.3 at $c = 1$, indicating that antiferromagmetic phase transition does exist.

Since the value $\xi(0, 0, 0) = 0.98$ is very close to the critical value $\xi(0, 0, 0) = 1$, one could argue that the conclusion of the intrinsic instability of the ferromagnetic structure could be an artifact of the method used. We emphasize, however, that RLF approximation underestimates the effect of correlations (e.g., shown above from the comparison with the nearest neighbor interactions model). This implies an overestimation of T_c and ξ.

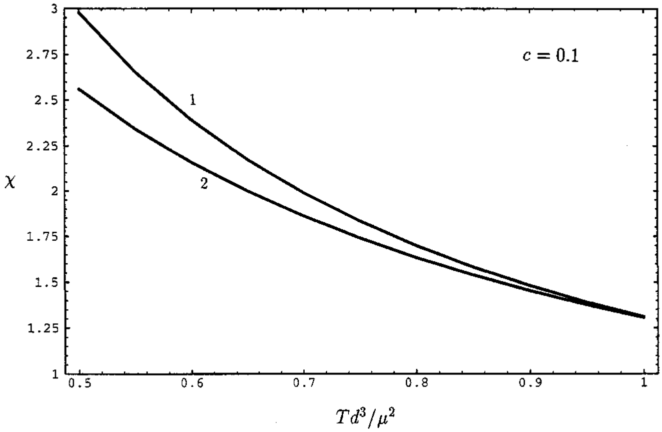

Fig. 2. Temperature dependence of the ferromagnetic (1) and antiferromagnetic (2) susceptibility for $c = 0.1$.

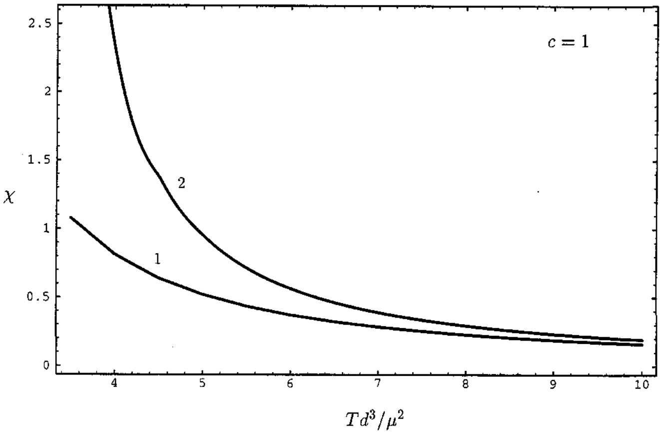

Fig. 3. Temperature dependence of the ferromagnetic (1) and antiferromagnetic (2) susceptibility for $c = 1$.

In the Appendix this conclusion is confirmed by a more exact evaluation of the correlation effects.

In Fig. 1 a remarkable feature of the parameter $\xi(0, 0, 0)$ is that it saturates to the relatively high value $\xi(0, 0, 0) \approx 0.44$ as $c \to 0$ compared with the parameter $\xi(\pi/d, \pi/d, 0)$ which rapidly decreases with concentration. Such different behavior of $\xi(0, 0, 0)$ and $\xi(\pi/d, \pi/d, 0)$ implies that at low concentrations the values of the homogeneous ferromagnetic susceptibility would be higher than those of the antiferromagnetic susceptibility. We illustrate this conclusion in Fig. 2, where it is shown that at $c = 0.1$ the values of the ferromagnetic susceptibility calculated from Eq. (22) exceed the value of the antiferromagnetic susceptibility, although the opposite situation takes place at $c = 1$ (Fig. 3) due to the existence of the antiferromagnetic phase transition at $T_c = 5.35J$.

The concentration dependence of the transition temperature is presented in Fig. 4 where also shown the mean field values of the antiferromagnetic and ferromagnetic transition temperatures. One can see that the effect of fluctuations destroys the antiferromagnetic phase transition below the critical concentration $c_{cr} \approx 0.56$.

We also estimated the effect of local field fluctuations in a body-centered cubic lattice. The positions of the atoms that correspond to a body-centered cubic structure can be represented by the radius-vector

$$\mathbf{r} = \left(m + \frac{a}{2}, n + \frac{a}{2}, k + \frac{a}{2}\right) \tag{28}$$

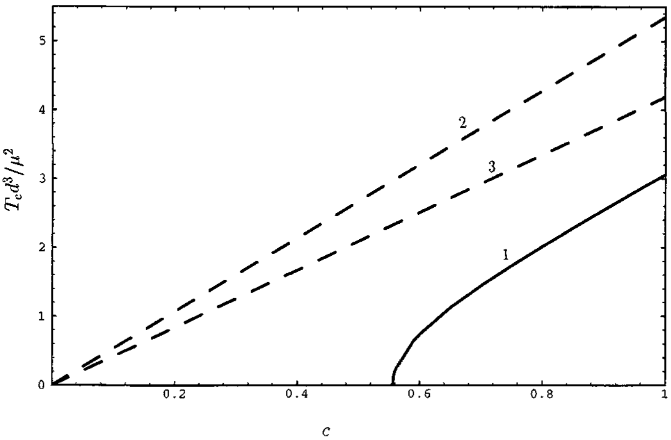

Fig. 4. Concentration dependence of the phase transition temperatures for a simple cubic lattice: (1) Antiferromagnetic transition temperature given by RLF theory. (2) Antiferromagnetic transition temperature given by mean field theory, $T_c = 5.35$ c. (3) Ferromagnetic transition temperature given by mean field theory, $T_c = 4\pi c/3$.

where $m, n, k = 0, \pm 1, \pm 2, \ldots, a = 0, \pm 1$. Note that there are two atoms per unit cell for a body-centered lattice, leading to twice the mean value of the local field, compared with that for a simple cubic lattice. At the same time the effect of local field fluctuations that shows up in the function $F_1(\rho)$ does not change significantly. For example, the local field second moment M_2 is equal to $M_2^{bc} \approx 14.9c\mu^2/d^3$ for a body-centered cubic lattice compared with $M_2^{sc} \approx 13.4c\mu^2/d^3$ for a simple cubic lattice. This results in the suppression of the effect of fluctuations in a body-centered cubic lattice and in the possibility of ferromagnetic ordering above the critical concentration $c_{cr} \approx 0.3$. At the same time the effect of local field fluctuations destroys antiferromagnetic ordering for a body-center cubic lattice. The calculated values of $\xi(\pi/d, \pi/d, 0)$ and $\xi(\pi/d, \pi/d, \pi/d)$ do not exceed 0.5.

IV. COMPARISON WITH A SPATIALLY DISORDERED DIPOLE DISTRIBUTION

The analysis of dipole ordering for spatially disordered dipole systems in terms of RLF theory has been performed recently by Zhang and Widom,(16) who considered randomly distributed dipolar hard spheres and concluded the existence of ferromagnetic ordering. We shall calculate the parameter $\xi(0, 0, 0)$ for randomly distributed dipolar spheres that establishes the possibility of ferromagnetic ordering at $T = 0$ and determines the critical volume fraction of the dipolar spheres above which such an ordering exists.

Using the continuous distribution of dipoles in Eq. (8) we obtain the parameter $\xi(0,0,0)$ in the form

$$\xi(0,0,0)=\frac{2}{\pi}\int_0^\infty d\rho\, e^{-F_1(\rho)}E_0(\rho) \tag{29}$$

where

$$F_1(\rho)=n\int d\mathbf{r}(1-\cos(J(\mathbf{r})\,\rho) \tag{30}$$

$$E_0(\rho)=n\frac{1}{\rho}\int d\mathbf{r}\,\sin(J(\mathbf{r})\,\rho) \tag{31}$$

Note that both $F_1(\rho)$ and $E_0(\rho)$ depend on the diameter of the sphere d_0 that plays the same role as the lattice constant for point dipoles distributed on the lattice sites; thus d_0 is the limiting factor for the distance separating neighboring dipoles.

At very low volume fraction one can consider the limit $d\to 0$ in Eq. (30) leading to $F_1(\rho)\sim|\rho|$. Such a limit in the analyses of dipole ordering has been considered in ref. 8 that corresponds to the Lorentzian shape of the local field distribution function. It has been shown that ferromagnetic ordering does not occur at low dipole concentration. On the other hand, at high volume fraction, the opposite limit in Eq. (30) can be considered by the expansion of $\cos(J(r)\,\rho)$ that corresponds to the Gaussian limit of the local field distribution. We obtain

$$F_1(\rho)=\tfrac{1}{2}M_2\rho^2, \qquad M_2=n\int d\mathbf{r}\,J(\mathbf{r})^2 \tag{32}$$

where M_2 is the second moment of the local field.

One can show also that in the evaluation of the function $E_0(\rho)$ the main contribution comes from long-distance dipoles for which one can replace $\sin(J(\mathbf{r})\,\rho)$ by its argument leading to $E_0(\rho)=E_0\equiv 4\pi n/3$. Using Eq. (29) we obtain finally

$$\xi(0,0,0)=\frac{4\pi}{3}n\int_0^\infty d\rho\, e^{-M_2\rho^2/2} \tag{33}$$

The solution of the equation

$$\xi(0,0,0)=1 \tag{34}$$

gives the critical volume fraction $\phi_c r \approx 0.2$ (which is almost identical with the result obtained in ref. 16) above which ferromagnetic ordering exists at $T = 0$. The volume fraction of dipole spheres is defined by

$$\phi = \frac{\pi}{6} n d_0^3 \tag{35}$$

Note that in the case of dipoles distributed on the lattice sites Eq. (33) follows from Eq. (23) with the same degree of accuracy as it follows from Eq. (29) for a continuous dipole distribution. This means that the second moment M_2 is the main characteristic of the local field fluctuations if the dipole concentration is not very low. This fact allows one to simply analyze why ferromagnetic ordering is possible for a spatially disordered dipole distribution and is not possible for a simple cubic lattice. Let us compare the second moment M_2 given by Eq. (32) with the local field second moment M_2^{sc} for the dipoles distributed on a simple cubic lattice.

$$M_2^{sc} = c \sum_j J_{ij} \approx 13.4 c \frac{\mu^2}{d^6} \tag{36}$$

From Eq. (32) we obtain

$$M_2 = \frac{4\pi n}{3} \frac{\mu^2}{d_0^3} = 6.4 \phi \frac{\mu^2}{d_0^6} \tag{37}$$

In order to compare M^2 with M_2^{sc} we account the fact that volume fraction for a continuous distribution of hard spheres has the same meaning as the atomic concentration for point particles distributed on the lattice sites, and, as we pointed above, the diameter of the hard spheres has the same meaning as the lattice constant for point dipoles. Thus we can identify c with ϕ and d with d_0. One can see then that M_2^{sc} is twice as large as M_2 thereby explaining the different effect of fluctuations in both systems.

It is important to emphasize also that such a large difference between M_2 and M_2^{sc} comes mainly from the angular part of the dipole–dipole interaction. If we neglect the angular part in Eq. (26) and consider $J^2(r) \sim r^{-6}$, we obtain $M_2^{sc} \approx 8.5 c\mu^2/d^6$, $M_2 \approx 8\pi\mu^2/d_0^6$.

V. CONCLUSION

This paper presents a local field theory formulation that predicts the types of ordered structures in random site systems with Ising anisotropy for various forms of interactions between particles. It was shown that for dipoles distributed on a simple cubic lattice ferromagnetic structure is intrinsically unstable against the fluctuations of the local fields at any dipole concentrations. Contrary to that behavior antiferroelectic type of ordering appears above a well defined critical concentration. It is shown also that the possibility of ferromagnetic dipole ordering for a spatially disordered dipole distribution stems from the suppressed value of second moment of the local field compared with that for a simple cubic lattice, where the angular part of dipole–dipole interaction $(1-3\cos^2\theta)$ leads to the relatively large value of the local field second moment caused by nearest neighbor dipoles.

APPENDIX

The RLF approximation neglects the effect of correlations between dipoles located at sites i and j. We will show below that accounting for correlation effects leads to an additional decrease of the parameter $\xi(\mathbf{q})$ compared with Eq. (23).

According to the exact relation in Eq. (7), the function $f_{i\rho}$ can be written in the form

$$f_{i\rho}=\sum_{m}\frac{1}{m!}(-i)^m\sum_{j_1\cdots j_m}\prod_{k\neq j_1\cdots j_m}\cos(J_{ik}\rho)\prod_{m}\sin(J_{ij_m}\rho)\langle l_{j_1}\cdots l_{j_m}\rangle \tag{A1}$$

From Eqs. (4) and (6) only the imaginary part of the function $f_{i\rho}$, corresponding to odd terms over m in Eq. (A1), contributes to the values of $\xi(\mathbf{q})$. Consider the first order correction to the linear approximation, Eq. (8),

$$i\frac{1}{3!}\sum_{j_1j_2j_3}\prod_{k\neq j_1,\,j_2,\,j_3}\cos(J_{ik}\rho)\sin(J_{ij_1}\rho)\sin(J_{ij_2}\rho)\sin(J_{ij_3}\rho)\langle l_{j_1}l_{j_2}l_{j_3}\rangle \tag{A2}$$

We replace

$$\langle l_{j_1}l_{j_2}l_{j_3}\rangle\approx\langle l_{j_1}\rangle\langle l_{j_2}l_{j_3}\rangle+\langle l_{j_2}\rangle\langle l_{j_1}l_{j_3}\rangle+\langle l_{j_3}\rangle\langle l_{j_1}l_{j_2}\rangle \tag{A3}$$

In order to estimate the upper limit of the effect of correlations we can further replace

$$\langle l_{j'}l_{j''}\rangle\approx 1 \tag{A4}$$

Also when evaluating

$$\sum_{j_2j_3}\prod_{k\neq j_2,\,j_3}\cos(J_{ik}\rho)\sin(J_{ij_2}\rho)\sin(J_{ij_3}\rho) \tag{A5}$$

we consider the major contribution from long-distance dipoles that allows one to replace Eq. (A5) by

$$E_0^2 \rho^2 \prod_k \cos(J_{ik} \rho) \tag{A6}$$

where $E_0 = \lim_{\rho \to 0} E_0(\rho)$ and we have taken into account that for long-distant dipoles one can neglect the limitation $k \neq j_2, j_3$ in Eq. (A5).

With the above approximations we obtain that function $F_1(\rho)$ should be replaced by $F_1(\rho) + \frac{1}{2} E_0^2 \rho^2$. Therefore, the correlation effects lead to the increase of the width of $f(E)$ resulting in the decrease of the parameter $\xi(\mathbf{q})$.

ACKNOWLEDGMENT

The authors acknowledge support from the National Science Foundation.

REFERENCES

1. R. E. Rosensweig, *Ferrohydrodynamics* (Cambridge University Press, Cambridge, 1985)
2. W. Luo, S. R. Nagel, T. F. Rosenbaum, and R. E. Rosensweig, *Phys. Rev. Lett.* **67**:2721 (1991).
3. J. Popplewell, P. Davis, A. Bradbury, and R. Chantrell, *IEEE Trans. Magn.* **22**:1128 (1993).
4. D. H. Reich, B. Ellman, J. Yang, T. F. Rosenbaum, G. Aeppli, and D. P. Belanger, *Phys. Rev. B* **42**:4631 (1990).
5. M. Goldman, *Phys. Rep.* **32**:1 (1971).
6. M. E. Lines and A. M. Glass, *Principles and Applications of Ferroelectrics* (Clarendon Press, Oxford, 1977).
7. U. T. Hohcli, K. Knorr, and A. Loidl, *Adv. Phys.* **39**:405 (1990).
8. B. E. Vugmeister and M. D. Glinchuck, *Rev. Mod. Phys.* **62**:993 (1990).
9. D. Wei and G. N. Patey, *Phys. Rev. Lett.* **68**:2043 (1992).
10. M. Luttinger and L. Tisza, *Phys. Rev.* **70**:954 (1946); **72**:257 (1947).
11. M. H. Cohen and F. Keffer, *Phys. Rev.* **99**:1128 (1955).
12. J. Villain, *Phys. Chem. Solids* **11**:303 (1959).
13. A. Aharony, *Solid State Commun.* **28**:667 (1978).
14. B. E. Vugmeister, *Sov. Phys. Sol. State* **26**:1483 (1984); B. E. Vugmeister and V. A. Stephanovich, *Sov. Phys. JETP* **70**:1053 (1990).
15. G. Ayton, M. J. P. Gingras, and G. N. Patey, *Phys. Rev. Lett.* **75**:2360 (1995).

16. H. Zhang and M. Widom, *Phys. Rev. B* **51**:8951 (1995).
17. M. W. Klein, C. Held, E. Zuroff, *Phys. Rev. B* **13**:3576 (1976).
18. B. E. Vugmeister and V. A. Stephanovich, *Solid State Commun.* **67**:323 (1987).
19. F. Zernike, *Physica* **7**:565 (1940).
20. T. Kaneyoshi, I. Tamura, and R. Honmura, *Phys. Rev. B* **29**:2769, (1984).
21. H. B. Calen, *Phys. Lett.* **4**:161 (1963).
22. E. Stenli, *Introduction to Phase Transitions and Critical Phenomena* (Clarendon Press, Oxford, 1971).
23. V. K. Shorte and S. Kirkpatrick, *Adv. Phys.* **20**:1279 (1971).