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Ordering of Ising dipoles

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Abstract. The ordering properties of Ising dipoles are studied in mean field theory, and by Monte Carlo simulations. The boundary conditions are such that there is no net depolarizing field and both regular lattices and various random arrangements are considered. In the mean field approach we employ the replica method with a Gaussian approximation for the distribution of dipole–dipole interactions, while a Kirkwood approximation is used for the spatial distribution of dipoles. The low-temperature phase for a system of randomly parked dipoles and diluted face centred cubic and body centred cubic lattices is found to be ferroelectric above a critical concentration. Below this concentration the mean field theory predicts a spin glass. The simulations are only carried out for the body centred cubic lattice. The transition temperature to the ferroelectric state is determined from finite size scaling of the mean square polarization. The critical concentration for the occurrence of a spin glass phase is estimated by zero temperature Monte Carlo simulations using the simulated annealing method. The results are found to be in qualitative agreement with those of the mean field theory described above.

1. Introduction

There is a long history associated with the question of whether a system of randomly distributed dipoles can order ferroelectrically. Debye (1912) noted that it was necessary to consider the effect of molecular electric dipole moments in order to explain the temperature dependence of the dielectric constant in certain liquids containing polar molecules. He started from the Lorentz expression for the internal field inside a cavity that is cut out from a medium

$$E_{\text{in}} = E + P/3\epsilon_0 \quad (1)$$

where P is the polarization, E the macroscopic field, and ϵ_0 is the permittivity of vacuum. Debye proceeded to construct a simple mean field theory, the predictions of which include ferroelectric ordering. For reasons which we will make clear later, the case of dipoles with discrete degrees of freedom will be of particular interest to us. We will, therefore, restrict ourselves to Ising spins ($s_i = \pm 1$) rather than the free rotors considered

by Debye. For Ising spins, the self-consistent equation determining the value of $m = \langle s_i \rangle$ at temperature T is

$$m = \tanh(\beta[\mu E + \mu^2 Nm/3V\epsilon_0]) \quad (2)$$

where β is $1/k_B T$, μ is the dipole moment, V is the volume of the sample, and N is the number of dipoles. In this case, the transition to a ferroelectric state with spontaneous polarization occurs below a Curie temperature, which is given by

$$T_C = \mu^2 N/3V\epsilon_0 k_B. \quad (3)$$

Although ferroelectricity is, and was, a well known phenomenon, it has not been observed in isotropic or nematic polar fluids. This motivated Onsager (1936) to argue that only a part of the internal field (1), which he called the cavity field, E_{ca} , gives rise to a torque on a dipole inside the cavity. The remaining part, which he called the reaction field, has no orienting effect. In the case of permanent dipoles randomly distributed in vacuum, Onsager's theory gives, in place of (1)

$$E_{ca} = E + \frac{P}{2\epsilon + \epsilon_0} \quad (4)$$

where ϵ is the dielectric constant of the medium. When this constant is determined self-consistently in a generalized Clausius-Mossotti theory there is always a locally stable solution with zero spontaneous polarization and, as a consequence, many people believe that a system of randomly distributed dipoles will not exhibit a ferroelectric state. Nevertheless, Pirene (1949), and later in more detail Zernik (1965), have shown that the Onsager theory does permit a ferroelectric solution. The possibility of ordering can be seen by noting that in general the dielectric function is given by $\epsilon = \epsilon_0 + \partial P/\partial E$. In a spontaneously polarized state near saturation $\partial P/\partial E$ will be small, and we return to a situation described qualitatively by Debye theory (except the transition to the spontaneously polarized state is now first order).

It is possible that the failure to observe ferroelectricity in polar fluids and nematics is largely due to 'molecular association' resulting from the fact that the molecules are free to move. Since dipolar interactions are also important in certain crystalline and amorphous materials we move on to theories where the dipoles have frozen positions. A number of authors have studied the energetics of dipoles on various crystalline lattices. Sauer (1940), whose results were later confirmed by Luttinger and Tisza (1946), found that thin needles of face centred cubic (FCC) and body centred cubic (BCC) crystals ordered ferroelectrically while the simple cubic lattice of dipoles had an antiferroelectric ground state. Their results depended on sample shape because of depolarizing effects. This result is subject to criticism since Griffiths (1968) showed rigorously for lattice systems that in the thermodynamic limit the free energy in zero external field is independent of sample shape. The depolarization energy cost of a uniformly polarized sample will be avoided by shape-dependent macroscopic domain wall structures (Kittel 1949) with energy per dipole going to zero in the thermodynamic limit. In order to study the thermodynamics and the existence of a ferroelectric transition, one can therefore restrict oneself to boundary conditions which allow uniform polarization without a depolarization field. Examples of such boundary conditions are metallic boundaries with shorted capacitor plates (Zernik 1965, Vugmeister and Glinchuk 1985) or periodic boundary condition with Ewald summation (Kretschmer and Binder 1979).

Experimentally, the interest in dipolar systems has concentrated on situations where the dipoles are arranged in a diluted lattice, as impurities. When the dipoles are randomly

distributed in a weakly polarizable medium (such as off-centre Li^+ ions in K^+OH^-) they appear to exhibit only a spin glass phase but no ferroelectricity (Fiory 1971). In contrast, similar impurities in a strongly polarizable medium (such as K^+TaO_3^-) appear to exhibit a ferroelectric phase (Vugmeister and Glinchuk 1965, Kleemann *et al* 1987) although this conclusion is controversial (Höchli and Maglione 1989). It is also expected theoretically that the presence of polarizable ions in the lattice may play an important role in determining the nature of the low-temperature phase (Mahan 1967). Tagaki (1952), for example, showed in a mean field calculation for dipoles on a simple cubic lattice and polarizable ions in the body centred positions that the low-temperature phase could be ferro- or antiferroelectric depending on the magnitude of the polarizability. For reasons of simplicity we will not address the question of polarizability here.

In order to explain the occurrence of a spin glass phase arising from dipole-dipole interactions one must consider fluctuations in the spatial distribution of the dipoles. These were neglected in the theories mentioned so far. Klein *et al* (1976) made an attempt to correct for spatial fluctuations in the case of pure dipolar interactions. The boundary conditions employed by Klein *et al* preclude uniform ferroelectric ordering. The reason is that they employ spherical geometry, i.e. they sum up all interactions within a radius R and let $R \rightarrow \infty$. A uniformly polarized state would then have to overcome the depolarizing field of a sphere and a ferroelectric state would have a complicated domain structure (Arnott 1968). Similar considerations apply to the Monte Carlo simulations on pure dipoles performed by Medina *et al* (1984). Calculations have also been carried out (Klein *et al* 1979, and Kirkpatrick and Varma 1978) with interactions which scale with distance as r^{-3} , but are not of the pure dipolar form. The question of whether a purely dipolar random system can order ferroelectrically thus remains open. We wish to address this question.

We will, in section 2, construct a mean field theory using available techniques from spin glass theory (Kirkpatrick and Sherrington 1978). As shall be seen, the theory predicts ordering, for sufficiently high concentrations of dipolar impurities. This ordering may be either ferro- or antiferroelectric depending on structure. Below the critical concentration the theory predicts a low-temperature spin glass phase. The critical concentration is estimated by zero temperature Monte Carlo simulations described in section 3. The predicted ferroelectric ordering at sufficiently high concentrations is confined by finite temperature Monte Carlo simulations discussed in section 4. A summary is given in section 5.

2. Model and mean field theory

A system of N dipoles that can be oriented in the $\pm z$ directions is considered. Assuming that the sample shape is such that the depolarization field is uniform the Hamiltonian can be written

$$H = - \sum_{i < j} J_d(r_{ij}) s_i s_j - \sum_i J_0 s_i = - \sum_{i < j} J(r_i, r_j) s_i s_j \quad (5)$$

where the dipole-dipole interaction is

$$J_d(r_{ij}) = \frac{\mu^2}{4\pi\epsilon_0 r_{ij}^3} (3 \cos^2(\theta_{ij}) - 1) \quad (6)$$

where θ_{ij} is the angle between the vector r_{ij} connecting two dipole sites and the z axis. If

there is a net polarization, the first sum in the middle expression in (5) will depend on sample shape, because of the depolarization field. With metallic boundary conditions J_0 is such that it cancels this field, while $J_0 = 0$ with periodic boundary conditions (Ewald summation). In general, for a homogeneous system, J_0 will be proportional to the total polarization allowing us to define an effective dipole-dipole interaction $J(r_i, r_j)$.

Depending on θ_{ij} , the dipole pair interaction can be either positive or negative. In the case of random spatial distribution of the spins this suggests the application of spin glass theory. We note that the restriction to Ising spins greatly simplifies the calculations, in that the part of the interaction which depends on the spatial separation enters as a scalar factor multiplying the spins. It is this feature which allows us to carry out the replica trick to be discussed later.

In the case of off-centre Li^+ impurities, however, either of the six $\langle 100 \rangle$ orientations or the eight $\langle 111 \rangle$ orientations are possible. The coupling $J(r_{ij})$ then becomes a tensor with spatially dependent components. Since we are dealing with an idealized situation we will not attempt to incorporate such effects.

We complete the definition of the model by specifying the spatial distribution of the dipoles. First, we consider the 'amorphous' case in which we assume that the continuous pair distribution function $g(r)$ is given. We approximate the N -particle distribution function by using the Kirkwood superposition approximation

$$g_N(r_1, r_2, \dots, r_N) = \prod_{\text{permutations}} g(r_i - r_j). \quad (7)$$

The probability distribution for strength of the Ising coupling constant J_{ij} for a randomly selected pair of dipoles is then given by

$$P(J) = \frac{1}{V} \int d^3r g(r) \delta(J - J(r)) \quad (8)$$

where the normalization

$$\int d^3r g(r) = V \quad (9)$$

has been used.

Averaging over spin configurations can be carried out in closed form using the replica method if distribution (8) is approximated by a shifted Gaussian

$$P_G(J) = \frac{\exp(-N(J - (1/N)J_1)^2/2J_2^2)}{(2\pi J_2^2/N)^{1/2}} \quad (10)$$

where, since the depolarization field is subtracted,

$$J_1 = \frac{N}{V} \int d^3r J(r) g(r) \quad (11)$$

will be independent of sample shape and size (as illustrated in an example later), and

$$J_2^2 = N \int_{-\infty}^{\infty} dJ J^2 P(J) - \frac{1}{N} J_1^2. \quad (12)$$

In what follows we will neglect terms of the order $1/N$, such as, e.g., the last term on the right-hand side of (12).

Once J_1 and J_2 are known, the phase diagram can be calculated using the replica method as described, e.g., in Kirkpatrick and Sherrington (1978). We will not repeat the steps of the calculation here. There are two order parameters in the theory. One of them is the polarization $m = \langle\langle s_i \rangle\rangle$ where first a thermodynamic average is performed for given spatial arrangements of spins followed by averaging over the arrangements. The second parameter is the Edwards–Anderson (Edwards and Anderson 1975) order parameter $q = \langle\langle s_i^2 \rangle\rangle$. These two order parameters are determined from the following integral equations:

$$m = \frac{1}{(2\pi)^{1/2}} \int_{-\infty}^{\infty} dz e^{-z^2/2} t \quad (13)$$

$$q = \frac{1}{(2\pi)^{1/2}} \int_{-\infty}^{\infty} dz e^{-z^2/2} t^2 \quad (14)$$

where

$$t = \tanh[\beta(J_1 m + J_2 q^{1/2} z)]. \quad (15)$$

The phase diagram is determined by the ratio J_1/J_2 . When $J_1/J_2 > 1$ there is a low-temperature ferroelectric phase with transition temperature J_1/k_B . Note that in the limit $J_2 \rightarrow 0$ and with J_1 the Lorentz field, the self-consistent equation for m is of the same form as in the simple Debye theory of the introduction. Even if $J_2 \neq 0$ but $J_2 < J_1$ the ferroelectric transition temperature, which depends only on J_1 , will then be the same as in Debye theory, but the order parameter will in general be different due to the fluctuation term J_2 . It should also be noted that the physical properties inside the spin glass phase predicted by the replica symmetric solution given by the above theory are in part incorrect (see, e.g., Mezard *et al* 1987). The transition temperature and the behaviour in the ferroelectric phase above a de Almeida–Thouless (1978) line of instability are, however, believed to be exact within the Gaussian model for the coupling constant distribution and these are the features with which we are mainly concerned in the present paper.

In order to be more concrete, we consider as an example a system in which the dipoles are located in the centres of hard spheres of diameter d . We assume simple form for $g(r)$

$$g(r) = \begin{cases} 0 & \text{for } r < d \\ 1 & \text{for } r > d. \end{cases} \quad (16)$$

As shown in the appendix we then have

$$J_1 = \frac{2f\mu^2}{\pi\epsilon_0 d^3} \quad (17)$$

and by evaluating the integral in (12) analytically

$$J_2 = \frac{\mu^2 (\frac{32}{3} f)^{1/2}}{4\pi\epsilon_0 d^3} \quad (18)$$

where f is the filling fraction $f = \pi N d^3/6V$. Equation (16) is not a bad approximation for the system of sequentially random parked hard spheres below its jamming limit ($f_j \approx 1/3$, Gotoh *et al* (1978)). We find that $J_1/J_2 > 1$ for $f > f^* = 1/10$. The present theory thus predicts a ferroelectric phase for dipoles on a system of randomly packed hard spheres at high concentrations.

We next consider the case where the dipoles occupy discrete lattice sites with concentration c . The coupling constant probability distribution in this case is

$$P(J) = \frac{1}{N_s} \sum_i \delta(J - J(r_i)) \quad (19)$$

where N_s is the number of sites in the lattice (the number of dipoles is $N = N_s c$). Proceeding as before we find for lattices of cubic symmetry (see the appendix)

$$J_1 = c N_s \mu^2 / 3V \epsilon_0 \quad (20)$$

and

$$J_2 = \mu^2 c^{1/2} \alpha / 4\pi \epsilon_0 a^3 \quad (21)$$

where $a^3 = V/N_s$ and

$$\alpha = \left[\sum_i \frac{a^6}{r_i^6} \left(\frac{3z_i^2}{r_i^2} - 1 \right)^2 \right]^{1/2}. \quad (22)$$

The values are $\alpha = 3.65, 2.08, 1.93$ for the sc (simple cubic), FCC and BCC lattices, respectively giving $c^* = 0.76, 0.25, 0.21$, for the critical concentration above which ferroelectric ordering occurs in the three cases.

The low-temperature phase for a perfect crystalline sc lattice is antiferroelectric (Sauer 1940, Luttinger and Tisza 1946) with opposite polarization on sublattices $\langle hkl \rangle$ with $h + k$ even or odd. We can, in the spirit of Takagi (1952), recast this situation to the present formalism by reversing the sign of the spins on one of the sublattices and we find by evaluating the dipole sum numerically so that

$$J_1 = 0.426 c N_s \mu^2 / V \epsilon_0. \quad (23)$$

Since J_1 in this case is larger than the value in the ferroelectric phase, the ferroelectric transition is pre-empted. The present model thus predicts that the simple cubic lattice will have an antiferroelectric ordering at low temperatures above an impurity concentration of $c^* = 0.45$. Below this concentration the theory predicts a low-temperature spin glass phase.

3. Zero temperature Monte Carlo simulations

To estimate the minimum dipole concentration for ferroelectric ordering we search for and analyse ground state configurations of the BCC system. We use a combination of the steepest descent and simulated annealing methods for finding approximate ground states (Kirkpatrick and Sherrington 1978). The basic Monte Carlo cell is cubic and periodic boundary conditions are imposed. The effective dipole-dipole interaction is computed by Ewald summation. The relevant formulae can be found in Kretschmer and Binder (1979) and will not be repeated here. We mention that for a perfect BCC lattice we reproduce the exact value $P/3\epsilon_0$ for the local field with uniform polarization to within a factor of 10^{-7} .

For each sample a list of local field $\sum_i J_{ij} S_i$ is compiled for each initial configuration, and the spin with the largest positive local energy is flipped. The list is updated continuously until no positive local energy can be found. This steepest descent procedure is a zero temperature quench and leads to a local minimum in the energy. Further

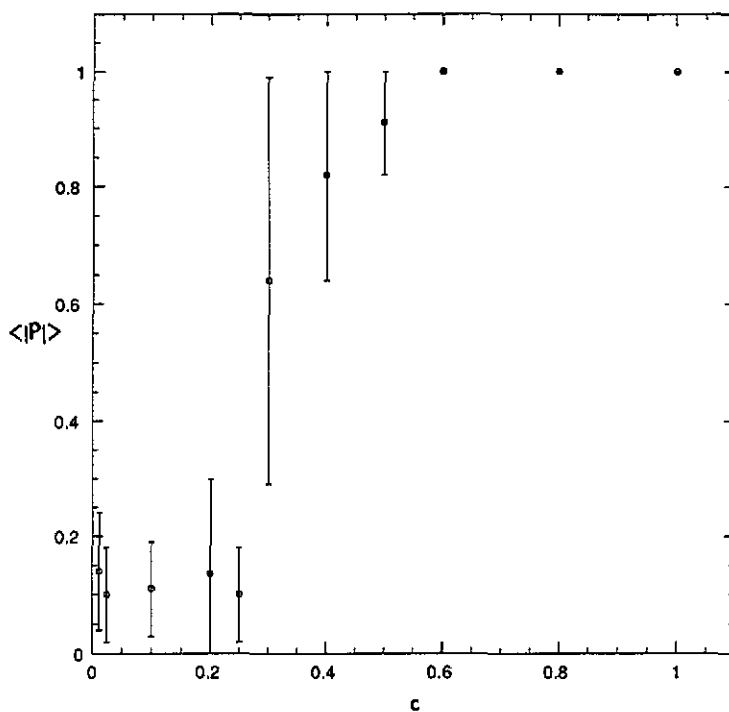


Figure 1. Ground state polarizations for the diluted BCC lattice at various concentrations. Error bars are sample to sample fluctuations.

improvement can be achieved by warming the system, using the Metropolis algorithm, to a temperature T which we took to be four times the mean field transition temperature. The system is then cooled down gradually to zero, reheated to $T/2$ and cooled down again. This procedure is repeated six times. The lowest energy configuration found during the process is saved. The energy obtained after the annealing is typically 3–10% lower than after the original steepest descent. For each sample we start from 50 different random initial spin configurations and also from an ordered state. The lowest energy is chosen to be the approximate ground state energy for the sample and we generated 50 different samples for each concentration. The number of spins in each sample was close to 450. The resulting ground state polarizations are plotted in figure 1, the error bars are the sample to sample standard deviation. We estimate the critical concentration for ferroelectric ordering to be 0.3 ± 0.1 which is somewhat larger than the mean field result 0.21 found in the previous section.

We have also investigated the local field distribution $P(E)$ in the 'ground state'. Kirkpatrick and Varma (1978) give an argument that $P(E = 0) = 0$ and they suggested that for random $1/R^3$ interactions $P(E)$ be proportional to E^α , with $\alpha = 0.5$, for small E . This behaviour is compatible with their simulations and would account for the observed $T^{3/2}$ behaviour of the specific heat of OH^- in KCl (Fiori 1971). In general the temperature dependence of the specific heat is given by $C \sim T^{1+\alpha}$ at low temperatures. We plot in figure 2 our results for the local field distribution for $c = 0.1$. In figure 3 we show a log-log plot of the field distribution for small E . Our best fits are $\alpha = 0.37$ for $c = 0.1$, $\alpha = 0.36$ for $c = 0.2$ and $\alpha = 0.46$ for $c = 0.3$. In the latter case we may be

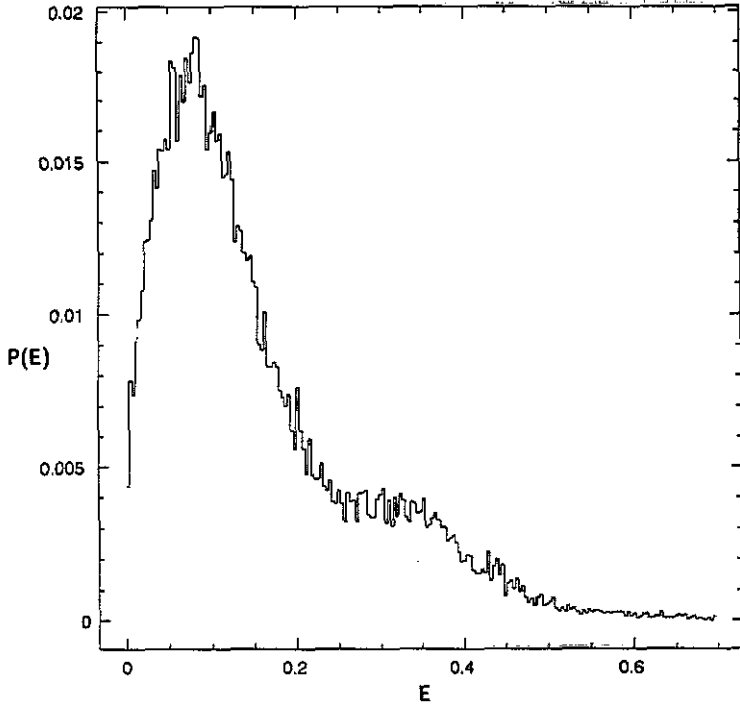


Figure 2. Ground state local field distribution at $c = 0.1$.

above the critical concentration for ferroelectric ordering. These values are significantly different from the value $\alpha = 1.0$ that have been obtained in the infinite range Sherrington-Kirkpatrick (SK) model (Palmer and Pond 1979).

4. Finite temperature Monte Carlo simulation

We have performed simulations at non-zero temperatures only for concentrations above the critical one for a ferroelectric phase. The calculations were performed starting from a random spin configuration. Each sample evolved at successively decreasing temperatures and the quantity monitored most closely was $\langle P^2 \rangle$. The initial relaxation time for this quantity was about 25 Monte Carlo steps per particle for the 800 particle systems and we used 10–20 000 steps per particle after thermalization. Periodic boundary conditions with Ewald summation was employed as described earlier. In the case of the diluted systems the thermodynamic quantities were averaged over 6–10 samples depending on sample size. The long range nature of the interactions was found, as observed by Kreschmer and Binder, to lead to important finite size effects. We therefore carried out simulations at each concentration for at least four different sample sizes and used finite size scaling theory for $\langle P^2 \rangle$ to locate the transition temperature (see, e.g., Binder 1984). In this approach one assumes a scaling form

$$\langle P^2 \rangle = N^x f(\varepsilon N^y) \quad (24)$$

for the mean square polarization, where $\varepsilon = (T - T_c)/T_c$ and the exponents x and y are related to the critical exponents, β (order parameter) and γ (susceptibility) through

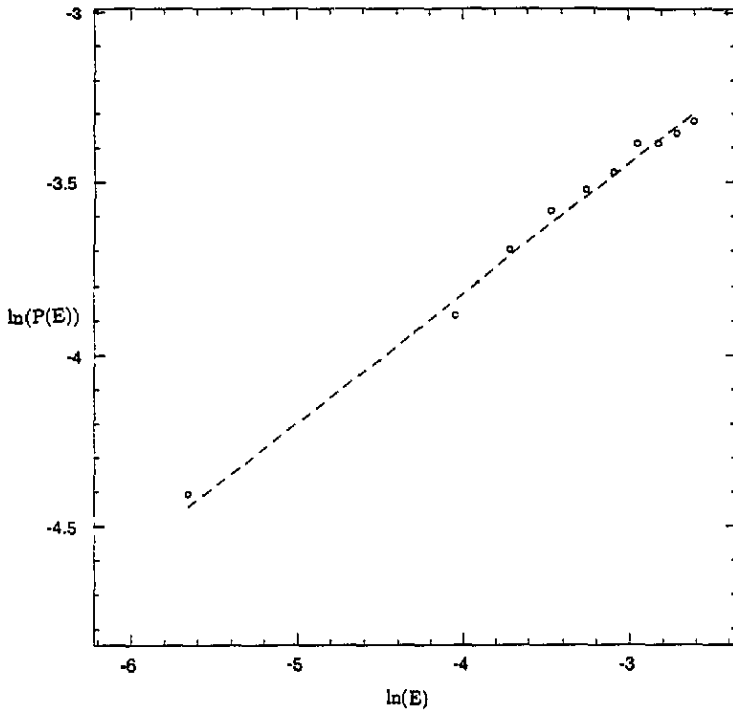


Figure 3. Log-log plot of ground state local field distribution for small E at $c = 0.1$. The broken curve is a linear fit with $\alpha = 0.37$.

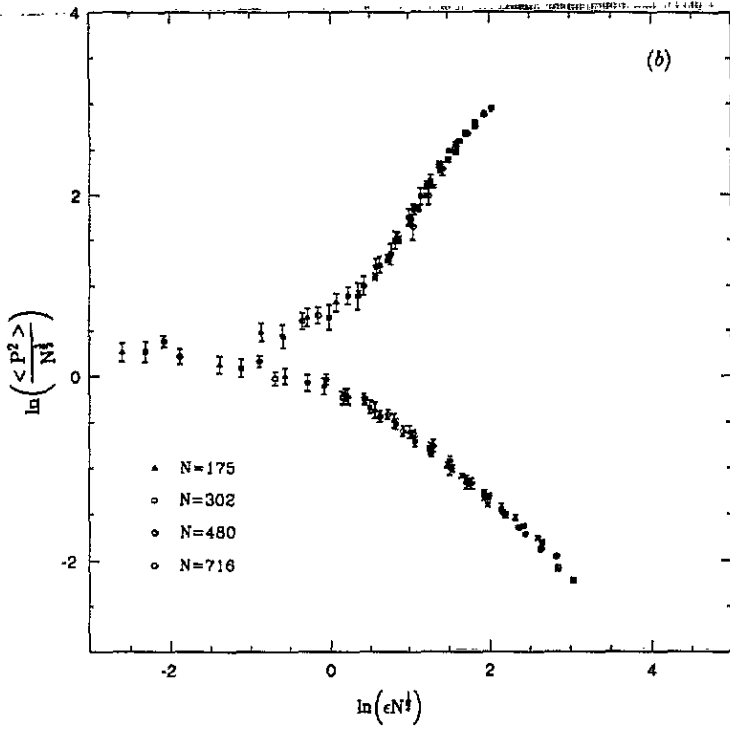
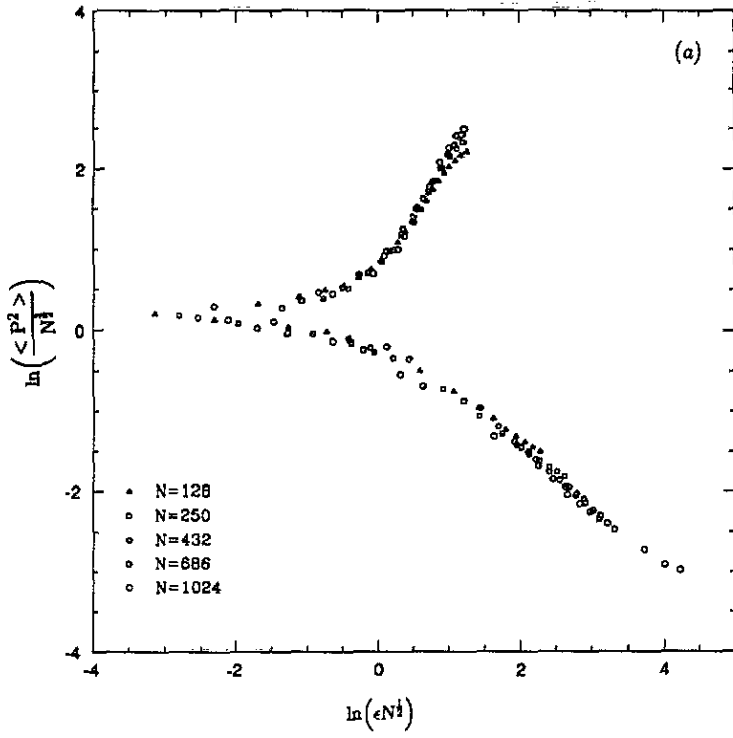
$$x - \gamma y = 1 \quad x + 2\beta y = 2.$$

In mean field theory $\gamma = 1$, $\beta = 1/2$ and $x = 3/2$, $y = 1/2$. The fit to T_c using mean field exponents is shown in figure 4(a-c). In the diluted case the points are arrived at by averaging over 6-10 samples. Without such averaging it was not possible to fit the data. All error bars represent sample to sample fluctuations and do not include thermal fluctuations which we estimate to be smaller. We were not able to significantly improve the fit by employing non-mean field values for x and y . The classical values of the exponents are in agreement with the renormalization group treatment of Larkin and Khmel'nitski (1969), and experiment (Griffin *et al* 1977). It is interesting to note that the worst fit is for $c = 1$ in which case logarithmic corrections to scaling are expected to be present.

The specific heat obtained from energy fluctuations and averaged over the samples is plotted in figure 5 for $c = 1$ and $c = 0.5$ respectively and for different sample sizes. We note that in contrast to $c = 1$ there appear to be no finite size effects on the specific heat for $c = 0.5$. The concentration dependence of the transition temperature is plotted in figure 6 together with the predictions of the mean field theory of section 2.

5. Discussion and summary

We have constructed a simple mean field theory for disordered dipolar Ising spins and compared it with the results of Monte Carlo simulations. Both the theory and the



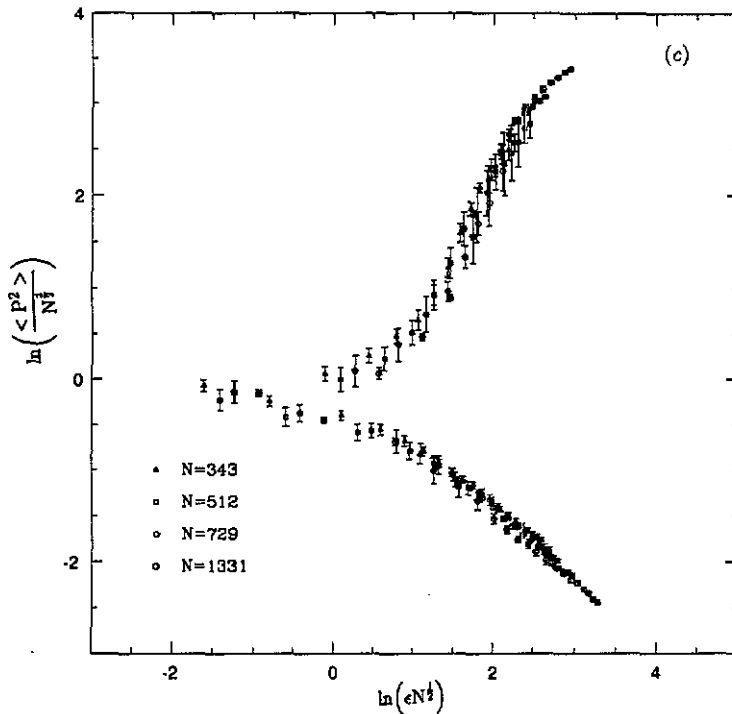


Figure 4. Scaling function (see equation (24)) at different concentrations: (a) $c = 1$, (b) $c = 0.7$, (c) $c = 0.5$. The fits are respectively $T_c = 0.79, 0.53, 0.37$ in units of $\mu^2/(4\pi\epsilon_0(a/2)^3)$.

simulations for the diluted BCC lattice show that there is a critical concentration above which a ferroelectric phase exists at low temperatures. As could be expected the transition temperature is higher ($\sim 30\%$) than found in the simulations, but the concentration dependence is in qualitative agreement with the simulation data. We have also investigated some properties of the ground state in the spin glass phase and found a local field distribution $P(E) \sim E^\alpha$ for low fields with $\alpha \approx 0.37$. The fact that this exponent is much smaller than the value $\alpha = 1$ for the SK model suggests that the low-temperature diluted dipolar system is less frozen than the spin glass with infinite range interactions. It is interesting in this context to note that Reich *et al* (1987) found that for very dilute dipolar magnets ($\text{LiHo}_x\text{Y}_{1-x}\text{F}_4$ with $x = 0.045$) the spins will not freeze at low temperatures while at higher concentrations ($x = 0.167$) they did observe the freezing of spins. It is also interesting that we have found a qualitative difference between the specific heat data for $c = 0.5$ and $c = 1$ indicating that the energy fluctuations do not diverge for the dilute system. We cannot tell if the logarithmic divergence of the specific heat disappears for all dilutions. Presently, we are also unable to state whether there is a sharp spin glass transition or just a gradual freezing of the spins at low concentration. We are currently carrying out longer and more detailed simulations in both the high- and low-concentration regimes in the hope of clarifying these questions.

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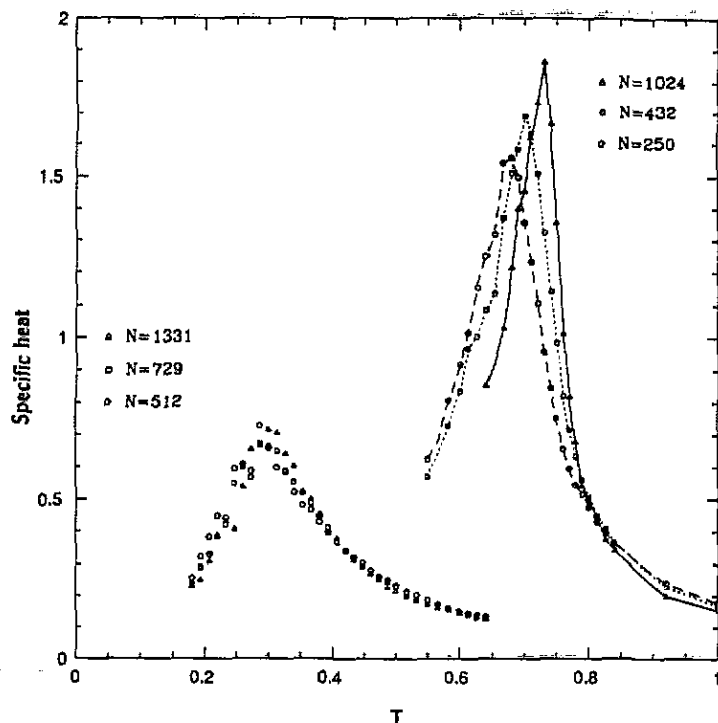


Figure 5. Specific heat at $c = 1.0$ (right), and 0.5 (left).

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Appendix. Evaluation of J_1 for randomly parked dipoles and diluted cubic lattices

We wish to evaluate J_1 defined in (11) and show that this quantity will be shape independent if the boundary conditions are such that there is no net depolarizing field, e.g. because of metallic boundaries. Let us start by assuming that the sample shape is such that the free sample has a uniform depolarizing field, E_{dp} (as in an ellipsoid, sphere, needle or slab) and is of the form

$$E_{dp} = -\frac{\mu}{4\pi\epsilon_0} \frac{D_s}{V} \sum_i s_i \quad (A1)$$

where D_s is the shape dependent depolarization factor, e.g. $D_s = 4\pi$ for a thin slab, $D_s =$

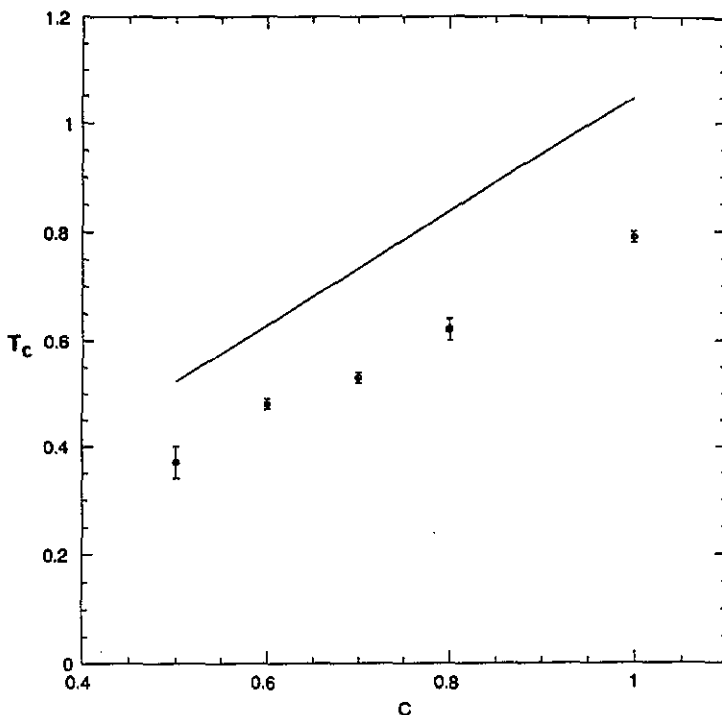


Figure 6. Critical temperature for the ferroelectric transition at various concentrations. The 'experimental points' are from Monte Carlo simulations while the full line is the mean field theory of section 3. The temperature unit is the same as in figure 4.

$4\pi/3$ for a sphere, and $D_s = 0$ for a needle. With metallic boundary conditions we have $J_0 = -E_{dp}\mu$ in the Hamiltonian (5). Therefore

$$J_0 = \frac{\mu^2}{4\pi\epsilon_0} \frac{D_s}{V} \sum_i s_i. \tag{A2}$$

For a diluted lattice from (19) we have

$$J_1 = c \sum_i J(r_i) = c \sum_i \left(J_d(r_i) + \frac{\mu^2}{4\pi\epsilon_0} \frac{D_s}{V} \right) \tag{A3}$$

where the sum i runs over all the lattice sites and c is the dipole concentration. The local field at a site when the sample is uniformly polarized (all the spins point up) is

$$\frac{1}{\mu} \sum_i J_d(r_i) = E_1 + E_2 + E_{dp} \tag{A4}$$

where $E_1 = N\mu/3V\epsilon_0$ is the Lorentz field due to polarization charges on the surface of an imaginary spherical cavity and E_2 is the field due to the dipoles inside the cavity. For a lattice of cubic symmetry $E_2 = 0$ and

$$J_1 = cN_s\mu^2/3V\epsilon_0. \tag{A5}$$

Similarly for randomly packed dipoles we find

$$J_1 = \mu^2 N/3\epsilon_0 V \tag{A6}$$

since $E_2 = 0$ when averaged over dipole spatial distributions.

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