NOTIZEN

Monte-Carlo Simulation of a Two-dimensional Dipolar Lattice

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Monte-Carlo calculations were carried out on a system consisting of 256 point-dipoles, whose centres are fixed in a two-dimensional square lattice with the usual boundary condition; the Epstein-Ewald-Kornfeld algorithm was used in evaluating the electrostatic energy. No evidence of a first-order phase transition was found, and the results suggest there might be a second-order one. Additional calculations were carrierd out using the mean-field theory, which was found to overestimate the transition temperature by about a factor two.

The present note reports on Monte-Carlo simulation of a classical, pure-dipolar system in two dimensions, whose dipolar centres are fixed on a simple square lattice; all dipoles are assumed to be identical, having a dipole moment of magnitude μ; calculations showed that the ground-state for this system is antiferroelectric, with an energy $-2.549\,\Lambda$ particle^{-1} versus -2.17 for the ferroelectric configuration. The pair potential consists of the dipole-dipole interaction only

Table 1. The ground-state antiferroelectric lattice.

Particle	Coordinates	Orientation
1	$(0, 0)$	$(1, 0)$
2	$(0, \frac{1}{2})$	$(-1, 0)$
3	$(\frac{1}{2}, 0)$	$(1, 0)$
4	$(\frac{1}{2}, \frac{1}{2})$	$(-1, 0)$

$$\Phi(i, j) = (\boldsymbol{\mu}_i \cdot \nabla_i)\,(\boldsymbol{\mu}_j \cdot \nabla_j)\left(\frac{1}{r}\right) = -\mu^2\,\boldsymbol{u}_i \cdot \mathbf{T}_{ij} \cdot \boldsymbol{u}_j\,, \tag{1}$$

$$\mathbf{T}_{ij} = \frac{1}{r^3}\left(\frac{3}{r^2}\,\boldsymbol{r}\,\boldsymbol{r} - \mathbf{I}\right) i \neq j\,; \quad \mathbf{T}_{ii} = 0\,, \tag{2}$$

$$\boldsymbol{r} = \boldsymbol{r}_i - \boldsymbol{r}_j\,, \quad r = |\boldsymbol{r}|\,, \tag{3}$$

for dipole moments $\boldsymbol{\mu}_i$, $\boldsymbol{\mu}_j$ located at $\boldsymbol{r}_i$, $\boldsymbol{r}_j$; $\boldsymbol{u}_i$, $\boldsymbol{u}_j$ are unit vectors defining their orientations, $\mathbf{T}_{ij}$ is the interaction sentor and $\mathbf{I}$ is the identity matrix.

All thermodynamic properties were expressed in reduced units; let Λ be defined by

$$\Lambda = \mu^2/a^3 \tag{4}$$

where a is the nearest-neighbour distance; temperatures are given in units Λ/k and energies in units Λ particle^{-1}; note that the thermodynamic properties are exclusively functions of the reduced temperature. There has been recently a number of theoretical and computer-simulation calculations of the properties of a three-dimensional dipolar lattice [1–4] whereas no calculations seem to have been carried out for a two-dimensional one.

The Epstein-Ewald-Kornfeld [5–9] algorithm was used in order to achieve a faster convergence in the evaluation of the electrostatic energy; a similar treatment for a three dimensional dipolar lattice has been developed by Hoskins, Perram and Smith [10, 11].

Results for the energy and the specific heat (in units k particle^{-1}) are listed in Table 2; both the

Table 2. Results for energy and specific heat.

T	$-U$	C_v
0.025	2.5371 ± 0.0002	0.42 ± 0.03
0.050	2.5246 ± 0.0006	0.50 ± 0.09
0.100	2.4992 ± 0.0005	0.56 ± 0.12
0.150	2.4742 ± 0.0006	0.50 ± 0.04
0.200	2.449 ± 0.002	0.56 ± 0.07
0.250	2.4200 ± 0.0004	0.48 ± 0.03
0.320	2.384 ± 0.002	0.54 ± 0.07
0.400	2.340 ± 0.003	0.68 ± 0.06
0.500	2.281 ± 0.005	0.64 ± 0.06
0.625	2.212 ± 0.009	0.67 ± 0.11
0.700	2.15 ± 0.01	0.61 ± 0.09
0.750	2.096 ± 0.008	0.80 ± 0.05
0.800	2.04 ± 0.01	0.80 ± 0.10
0.850	1.981 ± 0.004	0.86 ± 0.07
0.900	1.93 ± 0.01	0.91 ± 0.08
0.950	1.87 ± 0.01	0.88 ± 0.09
1.000	1.828 ± 0.005	0.82 ± 0.09
1.0625	1.77 ± 0.01	0.9 ± 0.1
1.125	1.720 ± 0.005	0.82 ± 0.08
1.250	1.620 ± 0.005	0.68 ± 0.05
1.500	1.465 ± 0.005	0.60 ± 0.03
1.750	1.327 ± 0.005	0.52 ± 0.02
2.000	1.208 ± 0.006	0.38 ± 0.04
2.500	1.024 ± 0.005	0.29 ± 0.02
3.000	0.88 ± 0.01	0.27 ± 0.01
4.000	0.680 ± 0.005	0.136 ± 0.008
6.000	0.475 ± 0.004	0.069 ± 0.001
8.000	0.354 ± 0.007	0.046 ± 0.003

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statistical errors and the specific heat were calculated in the usual way.

The order parameters are usually defined [12–14] as

$$Y_1 = \frac{1}{N} \left\langle \sum_{i=1}^{N} \boldsymbol{e}_i \cdot \boldsymbol{u}_i \right\rangle ;$$
$$Y_2 = \frac{1}{N} \left\langle \sum_{i=1}^{N} [2(\boldsymbol{e}_i \cdot \boldsymbol{u}_i)^2 - 1] \right\rangle , \qquad (5)$$

where $\boldsymbol{e}_i$ is the unit vector defining the orientation of the i-th particle in the ground-state lattice. It could be argued that, at least at a sufficiently high temperature, there might exist preferred orientations which do not coincide with the ground-state ones, and the order parameter Y_2 ought to be replaced by

$$Z_2 = \text{largest eigenvalue of the matrix}$$
$$\begin{pmatrix} 2\langle u_x^2 \rangle - 1 & 2\langle u_x u_y \rangle \\ 2\langle u_x u_y \rangle & 2\langle u_y^2 \rangle - 1 \end{pmatrix} . \qquad (6)$$

We calculated these matrix elements and also $\langle u_x \rangle$ and $\langle u_y \rangle$; at all temperatures we have

$$\langle u_x \rangle = \langle u_y \rangle = \langle u_x u_y \rangle = 0 ,$$

thus Z_2 coincides with Y_2, and the system does not turn ferroelectric; at $T \geqq 0.90$ we found

$$\langle u_x^2 \rangle = \langle u_y^2 \rangle = 0.5 .$$

The results for the specific heat suggest that the behaviour of the system is essentially harmonic at $T \leqq 0.32$, and anharmonicity sets in above this temperature.

The present results do not show any evidence of a discontinuity in the energy (first order phase transition); there might be a second-order phase transition between $T = 1.125$ and $T = 1.250$ but the statistics of the C_v values does not allow any conclusive result. Some additional calculations were carried out using the mean-field theory (a popular tool in dealing with order-disorder transitions), in order to test its validity.

According to the usual mean-field approximation [15, 16] Eq. (1) is averaged first over all possible orientations and then over coordinates or particle j, and the resulting pseudopotential turns out to be

$$\psi = \langle \Phi(i,j) \rangle_j = -4.34\, X \cos\Theta , \quad X = \langle \cos\Theta \rangle , \qquad (7)$$

where ψ is expressed in units Λ and X is the order parameter, which satisfies the consistency equation

$$X = \frac{1}{Z} \int_0^{2\pi} \cos\Theta \exp\left(-\frac{\psi}{T}\right) \mathrm{d}\Theta$$
$$Z = \int_0^{2\pi} \exp\left(-\frac{\psi}{T}\right) \mathrm{d}\Theta , \qquad (8)$$

where Z is the one-particle pseudopartition function. This equation can be rewritten as [17]

$$X = I_1(\zeta)/I_0(\zeta) \quad \zeta = (4.34/T)\,X , \qquad (9)$$

where I_0 and I_1 are modified Bessel functions of the first kind; this equation can be solved numerically, yielding X as function of T. The values obtained in this way (see Table 3) were found to be larger than the Monte-Carlo ones.

Table 3. Order parameters.

T	Monte-Carlo results		Mean field theory
	Y_1	Y_2	X
0.025	0.980 ± 0.002	0.920 ± 0.008	0.997
0.050	0.980 ± 0.002	0.923 ± 0.007	0.994
0.100	0.956 ± 0.008	0.84 ± 0.01	0.988
0.150	0.954 ± 0.002	0.830 ± 0.006	0.982
0.200	0.914 ± 0.007	0.69 ± 0.02	0.976
0.250	0.918 ± 0.005	0.71 ± 0.01	0.970
0.320	0.897 ± 0.004	0.643 ± 0.008	0.961
0.400	0.86 ± 0.01	0.55 ± 0.02	0.950
0.500	0.84 ± 0.01	0.48 ± 0.02	0.936
0.625	0.80 ± 0.02	0.40 ± 0.02	0.918
0.700	0.76 ± 0.02	0.34 ± 0.02	0.906
0.750	0.64 ± 0.01	0.19 ± 0.01	0.898
0.800	0.50 ± 0.05	0.07 ± 0.02	0.889
0.850	0.48 ± 0.05	0.06 ± 0.03	0.880

In the frame of this mean-field approximation the difference in the Helmholtz free energy between ordered and disordered phase for our system is given by

$$\Delta A/T = \tfrac{1}{2}\,\zeta X - \ln I_0(\zeta) ; \qquad (10)$$

when Eq. (9) is solved, X is found to be a continuous function of T, thus the previous equation can be expanded about the point $X = 0$, giving

$$\frac{\Delta A}{T} = \frac{1}{2}\zeta X - \frac{1}{4}\zeta^2 + \frac{1}{64}\zeta^4 + O(\zeta^6) \qquad (11)$$
$$= \frac{1}{2}\left(1 - \frac{4.34}{2T}\right)\frac{4.34}{T} X^2 + \frac{1}{64}\left(\frac{4.34}{T}\right)^2 X^4 + O(X^6) .$$

The transition temperature is determined by the vanishing of the coefficient of the second power of X [17, 18] and turns out to be $T = 2.17$; the vanishing of the third power of X is a necessary condition for a second-order phase transition; the coefficient of the fourth power is positive, as requested for stability. The mean-field treatment overestimates the transition temperature by a factor of about 2.

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