Long-range order in a classical two-dimensional dipole system

1992 J. Phys.: Condens. Matter 4 75

(http://iopscience.iop.org/0953-8984/4/1/018)

J. Phys.: Condens. Matter 4 (1992) 75–82. Printed in the UK

Long-range order in a classical two-dimensional dipole system

V M Bedanov
Institute of Pure and Applied Mechanics, USSR Academy of Sciences, Novosibirsk, 630090, USSR

Received 5 November 1990, in final form 11 April 1991

Abstract. A two-dimensional system of classical dipoles on a triangular lattice with isotropic in-plane ordering is studied both in spin-wave approximation and by Monte-Carlo simulation. It is shown that the mean-square displacement $\langle\theta^2\rangle$, unlike in the two-dimensional XY model, is finite in this system in the thermodynamic limit—not divergent logarithmically. This boundedness of $\langle\theta^2\rangle$ is caused, as demonstrated, by the long-range nature of the potential.

1. Introduction

A number of physical systems, such as layered magnetics with dominant dipole–dipole interaction [1, 2], admolecules with permanent dipole moment on a solid substrate [3] and biological membranes [4], are described by models of classical dipoles. When the external field is absent, and ignoring exchange or any other interactions, the simplest Hamiltonian may be written as

$$H = (J/2) \sum_{i\neq j} \sum_{\alpha,\beta} S_i^\alpha S_j^\beta D^{\alpha\beta}(r_i - r_j) \tag{1}$$

where

$$D^{\alpha\beta}(r) = \delta^{\alpha\beta}/r^3 - 3r^\alpha r^\beta/r^5 \tag{1a}$$

is a dipole tensor, J measures the strength of the dipole–dipole interaction, and S_i is a classical spin vector of unit length located at the site r_i.

The problem of phase transitions in a system with uniaxial ordering of spins with dipolar interaction has previously been described for an Ising-like system on a cubic lattice [5], for ferromagnets of finite thickness [6] and for Langmuir–Blodgett films [7]. The method of obtaining critical exponents from the ε expansion is discussed in [8].

In this paper, we shall consider the case of isotropic in-plane ordering of classical spins with dipolar interaction when spins are confined to a two-dimensional (2D) lattice.

The interesting questions arising with this model are: the structure of the ground state, the existence of long-range order and, if it exists, phase transition.

At present the following results for 2D dipole system appear in the literature:

(i) The ground state in a system with periodic boundary conditions is angular-degenerate, ferromagnetic or antiferromagnetic depending on the rhombicity angle of

0953-8984/92/010075 + 08 $03.50

the lattice [9, 10]. The ferromagnetic state, for example, with energy $E/N = -2.758\,J/a^3$, where a is the lattice spacing, is lowest for a triangular lattice.

(ii) The ground state in a finite system with open boundaries is the macro-vortex configuration with a higher energy than in the case of a periodic system [9].

(iii) At high temperatures the system undergoes a phase transition, which is predicted in computer experiments with periodic boundary conditions for square [11] and honeycomb [2] lattices.

(iv) The spherical model for the Hamiltonian (1) gives the critical temperature tending to zero in the thermodynamic limit for triangular and square lattices, i.e. absence of long-range order [12].

(v) Dipole forces in 2D ferromagnets stabilize the ferromagnetism, according to quantum spin-wave theory [13].

(vi) A phase transition is impossible in a system of classical dipoles in both 2D and 3D cases [14].

These results reveal a contradictory situation, concerning the existence of long-range order and phase transition in a 2D dipole system. This contradiction arises because different models were considered in the studies mentioned above. In the spherical model [12], for instance, the condition $S_i^2 = 1$ is replaced by the weaker one

$$\sum_{i=1}^{N} S_i^2 = N \tag{2}$$

and this results in the components S_i^x and S_i^y of a vector $\mathbf{S}_i$ becoming unbound (they are bound only through (2)). Hence, longitudinal fluctuations and those transverse to $\boldsymbol{k}$ give independent contributions. If one of them diverges at large N, then the total integral of fluctuations diverges as well. But in the 2D dipole system with the restriction $S_i^2 = 1$ (as will be shown below), transverse and longitudinal components of fluctuations cannot be separated.

In [14] 2D and 3D classical dipole systems were studied, where the interaction was described by the tensor $k^\alpha k^\beta/k^2$. However, this cannot be considered as a Fourier transform of the dipole tensor (1a) in leading orders in k because the dipole tensor at small k is given by [13, 15]

$$\tilde{D}^{\alpha\beta}_{2\mathrm{D}}(k) = a_1\delta^{\alpha\beta} + b_1\, k^\alpha k^\beta/k + \mathrm{O}(k^2)$$

$$\tilde{D}^{\alpha\beta}_{3\mathrm{D}}(k) = a_2\delta^{\alpha\beta} + b_2\, k^\alpha k^\beta/k^2 + \mathrm{O}(k^2)$$

for 2D and 3D cases respectively, where the first terms cannot be neglected.

We conclude that points (iv) and (vi) are not relevant, strictly speaking, to our 2D dipole system and there is no restriction, from the literature, against the existence of long-range order and phase transition in a 2D dipole system.

Computer experiments [2, 11] were carried out for precisely the same model as described here, but for finite systems without size-effect analysis. In this paper we present spin-wave and Monte-Carlo studies of the low-temperature behaviour of a 2D system of classical dipoles on a triangular lattice. The mean-square angular displacement and correlation functions are calculated for different system sizes. We shall discuss also a system with isotropic $1/r^3$ potential on an arbitrary 2D lattice.

2. Spin-wave approximation

Since the ground state of the 2D dipole system on a triangular lattice is an angular-degenerate ferromagnetic state, we suppose the low-temperature state to be nearly ferromagnetic with small angular displacement in every lattice site. Consider the finite system with periodic boundary conditions. Let $N = 2N_xN_y$ be the number of dipoles, and $L_x = N_xa$ and $L_y = N_y(3)^{1/2}a$, where a is the lattice spacing, be the sides of the periodic cell, which is chosen as a rectangle, consistent with the triangular lattice. Let θ_0 be the angle of the vector of spontaneous magnetization, θ_i be angular displacement in the site i. Changing variables S_i^α to θ_i and expanding the Hamiltonian into a θ_i series up to square terms, we obtain

$$H = N\varepsilon_0 + (J/2)\sum_{i,j} \theta_i A(\boldsymbol{r}_i - \boldsymbol{r}_j)\theta_j \tag{3}$$

where

$$\varepsilon_0 = (J/2)\sum_{\boldsymbol{r}} D^{xx}(\boldsymbol{r}) = (J/2)\sum_{\boldsymbol{r}} D^{yy}(\boldsymbol{r}) \tag{4}$$

is the energy of the ground state, where $\boldsymbol{r}$ denotes the lattice sites.

It will be noted that the dipole tensor for a periodic system is given by

$$D^{\alpha\beta}(\boldsymbol{r}) = \sum_{\boldsymbol{p}} \left(\frac{\delta^{\alpha\beta}}{|\boldsymbol{r}+\boldsymbol{p}|^3} - \frac{3(r^\alpha + p^\alpha)(r^\beta + p^\beta)}{|\boldsymbol{r}+\boldsymbol{p}|^5} \right) \tag{5}$$

where $\boldsymbol{p}$ is a translational vector with the components nL_x and mL_y, where $n, m = 0, \pm 1, \pm 2, \ldots$. Function $A(\boldsymbol{r})$ resulting from expansion has the form

$$\begin{aligned} A(0) &= -2\varepsilon_0/J + (\sin^2\theta_0)D^{xx}(0) + (\cos^2\theta_0)D^{yy}(0) \\ A(\boldsymbol{r}) &= (\sin^2\theta_0)D^{xx}(\boldsymbol{r}) - \sin(2\theta_0)D^{xy}(\boldsymbol{r}) + (\cos^2\theta_0)D^{yy}(\boldsymbol{r}) \qquad \boldsymbol{r} \neq 0 \end{aligned} \tag{6}$$

where

$$D^{\alpha\beta}(0) = \sum_{\boldsymbol{p}\neq 0} \left(\frac{\delta^{\alpha\beta}}{p^3} - \frac{3p^\alpha p^\beta}{p^5} \right).$$

Introducing the Green function

$$G(\boldsymbol{r}) = \sum_{\boldsymbol{k}} \frac{e^{i\boldsymbol{kr}}}{A(\boldsymbol{k})} \tag{7}$$

where $A(\boldsymbol{k})$ is the Fourier transform of $A(\boldsymbol{r})$, allowing θ_i to take values from $-\infty$ to $+\infty$, and performing the necessary integration, one derives the following expressions for the mean-square angular displacement and correlation function, respectively,

$$\langle\theta^2\rangle = (k_B T/NJ)G(0) \tag{8}$$

$$c(\boldsymbol{r}) \equiv \langle S(0)S(\boldsymbol{r})\rangle = \exp\{(k_B T/NJ)[G(\boldsymbol{r}) - G(0)]\}. \tag{9}$$

For the Fourier transform of $A(\boldsymbol{r})$, taking account of expression (6), we obtain

$$A(\boldsymbol{k}) = -2\varepsilon_0/J + (\sin^2\theta_0)D^{xx}(\boldsymbol{k}) + (\cos^2\theta_0)D^{yy}(\boldsymbol{k}) - \sin(2\theta_0)D^{xy}(\boldsymbol{k}) \tag{10}$$

where $D^{\alpha\beta}(\boldsymbol{k})$ is the Fourier transform of the dipole tensor. It is clear from (10) that only excitations transverse to the magnetization vector give contributions to $\langle\theta^2\rangle$.

Tensor $D^{\alpha\beta}(\boldsymbol{k})$ has been calculated in different ways [12, 13, 15, 16]. It can be expressed in terms of Ewald sums

$$D^{\alpha\beta}(\boldsymbol{k}) = -\frac{2\sqrt{\pi}}{v}\sum_{\boldsymbol{q}}\frac{(k^\alpha+q^\alpha)(k^\beta+q^\beta)}{|\boldsymbol{k}+\boldsymbol{q}|}\Gamma(\tfrac{1}{2}, |\boldsymbol{k}+\boldsymbol{q}|^2/4M) - \frac{2}{\sqrt{\pi}}\sum_{r\neq 0}\frac{\cos(\boldsymbol{kr})}{r^3}$$
$$\times\left(\delta^{\alpha\beta}\Gamma(\tfrac{3}{2}, rM^2) - \frac{2r^\alpha r^\beta}{r^2}\Gamma(\tfrac{5}{2}, r^2M)\right) + \frac{4}{3\sqrt{\pi}}M^{3/2}\delta^{\alpha\beta} \tag{11}$$

where v is the elementary cell area ($v = (\sqrt{3}/2)a^2$ for a triangular lattice), $\Gamma(b, z)$ is an incomplete gamma function, M is an arbitrary parameter, the value of which is chosen to ensure that both series converge rapidly, and $\boldsymbol{q}$ is a vector of the reciprocal lattice. Expanding (11) into a Taylor series near $\boldsymbol{k} = 0$ we obtain the small-$\boldsymbol{k}$ behaviour of the dipole tensor

$$D^{\alpha\beta}(\boldsymbol{k}) \simeq \delta^{\alpha\beta}(2\varepsilon_0/J + D_1k^2) + D_2\,k^\alpha k^\beta/k - D_3k^\alpha k^\beta \tag{12}$$

where $D_1 = 0.2633\,a^{-1}$, $D_2 = 2\pi/v = 7.255\,a^{-2}$, $D_3 = 1.5800\,a^{-1}$ and $\varepsilon_0 = -2.7585\,Ja^{-3}$.

It is clear from (12) that the small-k behaviours of eigenvalues corresponding to directions longitudinal and transverse to $\boldsymbol{k}$ are respectively

$$D_l(k) = 2\varepsilon_0/J + D_2k + (D_1 - D_3)k^2$$
$$D_t(k) = 2\varepsilon_0/J + D_1k^2.$$

Note also that these eigenvalues always appear in (10) as a linear combination and hence, unlike the spherical model [12], it is impossible to decompose longitudinal and transverse spin fluctuations into separate parts in sum (7).

Comparing (12) and (10) one can see that the function in sum (7) has a singularity at $k \to 0$. Substituting (12) into (10) and changing the variables to polar coordinates k and φ we obtain for small k

$$A(k, \varphi) \simeq D_1k^2 + (D_2k - D_3k^2)\sin^2(\theta_0 - \varphi). \tag{13}$$

This expression shows that $A(k, \varphi) \propto k^2$ only when $\varphi = \theta_0$, and $A(k, \varphi)$ depends linearly on k for other directions. For comparison, there is a stronger singularity $A(k, \varphi) \propto k^2$ for the 2D *XY* model [17, 18], which leads after integrating in $\boldsymbol{k}$-space to logarithmic divergence of displacements with system size

$$\langle\theta^2\rangle \propto T\ln(N). \tag{14}$$

However, the 2D dipole system exhibits another behaviour. Indeed, replacing the sum in (7) by an integral (this replacement is justified for sufficiently large systems) over the circular Brillouin zone of equal area we find

$$\langle\theta^2\rangle \simeq \frac{k_BT}{v_BJ}\int_{k_0}^{K} k\,dk\,4\int_0^{\pi/2}\frac{d\varphi}{D_1k^2 + (D_2k - D_3k^2)\sin^2\varphi}$$
$$= \frac{2\pi k_BT}{v_BJ[D_1(D_3 - D_1)]^{1/2}}\arcsin\left(1 - \frac{D_3 - D_1}{D_2}k\right)\Bigg|_{k=k_0}^{K} \tag{15}$$

where $v_B = 45.58\,a^{-2}$ is the area of the Brillouin zone, $K = 3.809\,a^{-1}$ and $k_0 = \min(2\pi/$

$L_x, 2\pi/L_y)$. If we consider periodic cells of nearly square shape, then $L_x \simeq L_y \simeq 0.9306\, N^{1/2}\, a$ and $k_0 \simeq 6.752\, N^{-1/2}\, a^{-1}$. The mean-square displacement is then given by

$$\langle \theta^2 \rangle \simeq 0.2341\, T^*[0.3928 + \arcsin(1 - 2.451\, N^{-1/2})] \qquad (16)$$

which gives a finite value of $\langle \theta^2 \rangle$ in the large-N limit: $\langle \theta^2 \rangle_\infty = 0.4597\, T^*$, where $T^* = k_B T a^3/J$ is the reduced temperature. Owing to this feature, the 2D dipole system essentially differs from other well known 2D systems such as the 2D XY model and 2D crystals with isotropic pair potentials for which relations like (14) are valid [17, 18].

We must not overlook the fact that certain assumptions have been made. It is clear that the assumptions concerning integration in k-space are not critical. This effect vanishes with $N \rightarrow \infty$. Our first assumption, on the existence of definite direction and small angular displacements, is more essential and may give rise to some doubts. Therefore, a Monte-Carlo computer experiment was carried out to verify the spin-wave results.

3. Monte Carlo simulation

The same system described above (i.e. 2D dipoles on a triangular lattice with periodic boundary conditions) was simulated by a Monte Carlo computer experiment. The periodic cell was chosen as nearly square in shape. The dipole tensor (5) for periodic systems of different sizes was calculated by the Ewald method before Monte Carlo (MC) runs. The usual Metropolis algorithm [19] was employed, with automatic choice of displacement amplitude ensuring an acceptance probability of about 0.5.

First, the ground state was tested. The arbitrary disordered initial configuration was rapidly cooled to $T = 0$. In most cases, the system came to be in a ferromagnetic state of the same energy $E_0/N = -2.7585\, J/a^3$ but with different directions of the magnetization vector. In some cases it came to be in metastable states of higher energy than E_0, but after heating and subsequent cooling it again reached the lowest-energy ferromagnetic state.

At low but finite temperatures the state was nearly ferromagnetic. The vector of spontaneous magnetization fluctuated both in amplitude about a mean value and in direction from configuration to configuration. These angular fluctuations were unrestricted. A noticeable rotation of the magnetization vector for small systems was observed, while its amplitude was nearly constant. The vector made, for example, an average rotation of approximately 180° during 4000 steps for $N = 30$.

When calculating $\langle \theta^2 \rangle$ the direction of the magnetization vector was determined for every configuration, and then the deviations from this angle were calculated along the system. About 1000 MC steps were taken for equilibration and up to 20 000 configurations were taken for averaging.

4. Results and discussion

The results of the calculations are presented in figure 1 for $\langle \theta^2 \rangle$ at reduced temperature $T^* = 0.1$. The exact result of spin-wave theory was evaluated as follows. First, the dipole tensor $D^{\alpha\beta}(\boldsymbol{k})$ was calculated for $N = 8064$, and $\langle \theta^2 \rangle$ was found as a sum over all the points in the Brillouin zone. Including only certain points from the Brillouin zone, the results for different N may be obtained. Thus, the exact spin-wave results are presented

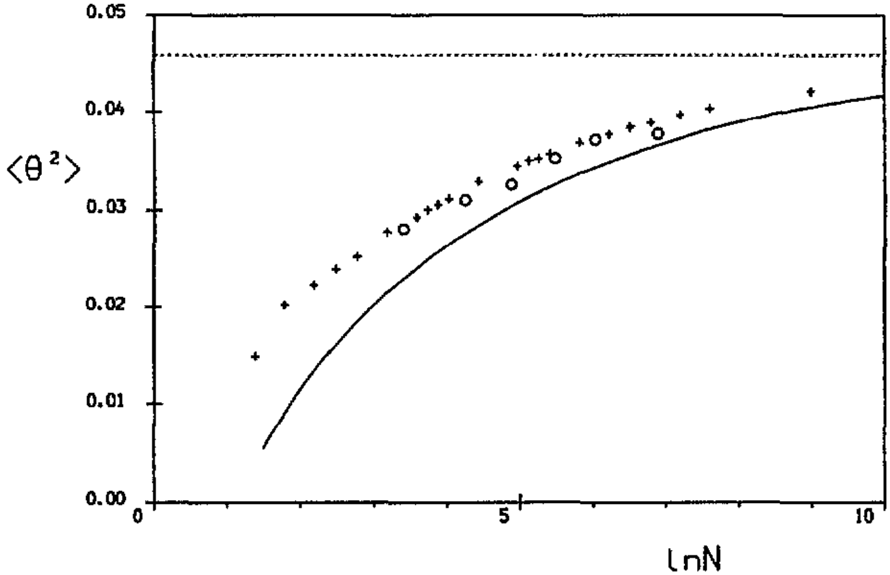

Figure 1. The mean-square angular displacement versus system size: circles are Monte Carlo values, crosses are exact spin-wave results, full curve corresponds to formula (16) and the broken line corresponds to large-N limit. The reduced temperature is $T^* = 0.1$.

in figure 1 for the sequence $N = 8064, 2016, 1344, \ldots$, where every N corresponds to a nearly square cell. Note that, owing to anisotropy of the potential, $\langle\theta^2\rangle$ depends on both system shape and angle θ_0. Therefore, besides the choice of square shape we had to average $\langle\theta^2\rangle$ over θ_0. This averaging made it possible to compare Monte Carlo and spin-wave results, because of automatic averaging over θ_0 in the former case owing to fluctuations of θ_0. As one can see from figure 1 the results of spin-wave theory and Monte-Carlo simulation are in reasonable agreement. Correlation functions are presented in figure 2 for different temperatures and $N = 418$. At reduced temperatures below $T^* = 0.6$, Monte Carlo and spin-wave results again coincide. The agreement of the Monte Carlo simulation with spin-wave theory predictions is evidence of the validity of our starting assumptions and, hence, of our conclusions on the existence of long-range order in the 2D dipole system.

To clarify the role of long-range interactions of the dipole potential and its anisotropy, consider the system with long-range isotropic potential $1/r^3$ with Hamiltonian given by

$$\tilde{H} = -(J/2)\sum_{i,j}(S_i S_j)D(r_i - r_j) \tag{17}$$

where for a periodic system $D(r)$ is given by

$$D(r) = \sum_p \frac{1}{|r+p|^3}. \tag{18}$$

When $J > 0$ the ground state of this system is a ferromagnetic state, and we may apply a similar spin-wave consideration as has been applied for the 2D dipole system.

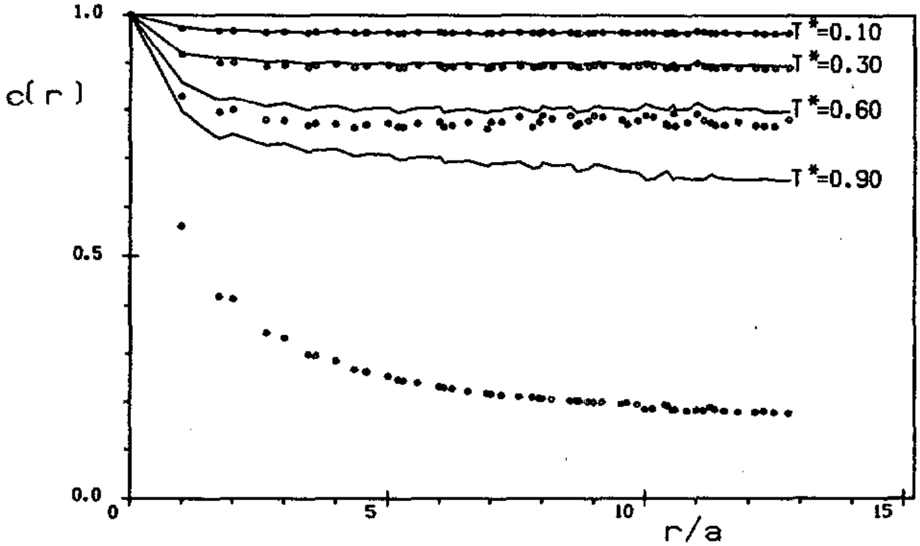

Figure 2. The pair correlation function: circles are Monte Carlo values, full curves are spin-wave predictions. The size of the system is $N = 418$.

Expanding (17) in powers of θ_i, we shall obtain the Hamiltonian in the form (3) with the ground-state energy given by

$$\tilde{\varepsilon}_0 = -(J/2) \sum_{r \neq 0} \frac{1}{r^3} \tag{19}$$

and the function $\tilde{A}(\boldsymbol{r})$ given by

$$\tilde{A}(0) = \sum_{r \neq 0} D(\boldsymbol{r}) \qquad \tilde{A}(\boldsymbol{r}) = -D(\boldsymbol{r}) \qquad \boldsymbol{r} \neq 0. \tag{20}$$

The Fourier transform of $A(\boldsymbol{r})$ now becomes

$$\tilde{A}(\boldsymbol{k}) = -2\tilde{\varepsilon}_0/J - \sum_{r \neq 0} \frac{\cos(\boldsymbol{kr})}{r^3}. \tag{21}$$

Function $1/\tilde{A}(\boldsymbol{k})$ has a singularity at $\boldsymbol{k} = 0$. Noting that the $1/r^3$ potential can be expressed in terms of the dipole tensor

$$-1/r^3 = D^{xx}(\boldsymbol{r}) + D^{yy}(\boldsymbol{r}) \tag{22}$$

we can write the expression

$$\tilde{A}(\boldsymbol{k}) = -4\varepsilon_0/J + D^{xx}(\boldsymbol{k}) + D^{yy}(\boldsymbol{k}). \tag{23}$$

Knowing the small-k behaviour of $D^{\alpha\beta}(\boldsymbol{k})$ we now derive

$$\tilde{A}(\boldsymbol{k}) = D_2 k + (2D_1 - D_3)k^2. \tag{24}$$

We again have a term linear in k, and hence the value of $\langle\theta^2\rangle$ is bounded. Indeed, integrating on k for $N \to \infty$ ($k_0 \to 0$) one finds $\langle\theta^2\rangle_\infty \simeq 0.105\, T^*$.

Consider now the effect of lattice type. The expression (21) may be written in the form

$$\tilde{A}(k) = \sum_{r \neq 0} \frac{1 - \cos(kr)}{r^3} \tag{25}$$

from which it is clear that, at small k, the terms with small r can be neglected. The terms with large r give the main contribution; but for large r, the lattice sum gives the same result for different lattices. This result may be evaluated by replacing the sum (25) with the integral. Performing the integrations and expanding Bessel functions in powers of k, one finds

$$A(k) \simeq (2\pi/v)[k - (a/4)k^2] \tag{26}$$

where the first term exactly coincides with that of (24). Thus, our conclusion about the boundedness of $\langle\theta^2\rangle$ for the 2D dipole system on a triangular lattice is also valid for an isotropic ferromagnetic interaction $1/r^3$ on the arbitrary 2D lattice.

References

[1] Ibrahim A K and Zimmerman G O 1987 *Phys. Rev.* B **35** 1860
[2] Zimmerman G O, Ibrahim A K and Wu F Y 1988 *Phys. Rev.* B **37** 2059
[3] Ruiz-Suarez J C, Klein M L, Moller M A, Rowntree P A, Scoles G and Xu J 1988 *Phys. Rev. Lett.* **61** 710
[4] Seelig J, Macdonald P M and Scherer P G 1987 *Biochemistry* **26** 7535
[5] Kretschmer R and Binder K 1979 *Z. Phys.* B **34** 375
[6] Garel T and Doniach S 1982 *Phys. Rev.* B **26** 325
[7] Andelman D, Brochard F and Joanny J F 1987 *J. Chem. Phys.* **86** 3673
[8] De'Bell K and Geldart D J W 1989 *Phys. Rev.* B **39** 743
[9] Belobrov P I, Voevodin V A and Ignatchenko V A 1985 *Zh. Eksp. Teor. Fiz.* **88** 889
[10] Brankov J G and Danchev D M 1987 *Physica* A **144** 128
[11] Romano S 1987 *Nuovo Cimento* D **9** 409
[12] Danchev D M 1990 *Physica* A **163** 835
[13] Maleev S V 1976 *Zh. Eksp. Teor. Fiz.* **70** 2374
[14] Sadreev A F 1986 *Phys. Lett.* A **115** 193
[15] Aharony A and Fisher M E 1973 *Phys. Rev.* B **8** 3323
[16] Rozenbaum V M, Artamonova E V and Ogenko V M 1988 *Ukr. Fiz. Zh.* **33** 625
[17] Berezinskii V L 1970 *Zh. Eksp. Teor. Fiz.* **59** 1970 907
[18] Kosterlitz J M and Thouless D J 1973 *J. Phys. C: Solid State Phys.* **6** 1181
[19] Binder K (ed) 1979 *Monte Carlo Methods in Statistical Physics* (Berlin: Springer)