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Theoretical Studies of the Planar Lebwohl-Lasher Lattice Model

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Theoretical studies of a planar version of the Lebwohl-Lasher model are presented. The model consists of a set of interacting centres forming a simple square lattice. The pair potential is nearest neighbour and varies as the second Legendre polynomial of the relative orientation between the two particles. The system is studied with the aid of Faber's theory of nematic order, the mean field theory and the two site cluster theory. The results are compared with one of the existing simulations. We find that Faber's theory not only gives an accurate description of the model at low temperatures, but also correctly predicts the dependence of various parameters on the system size. The two site cluster theory predicts the order-disorder transition temperature with greater accuracy than the mean field theory.

Keywords: Lattice models; nematics

PACS number (s): 61.30, 61.30.Gd

1 INTRODUCTION

The Lebwohl-Lasher (LL) model, since its inception in 1972 has become a prototype for computer simulation as well as theoretical calculations [1–8] for systems which exhibit a nematic-isotropic like orientational phase transitions. One can compare its role in the field nematic-isotropic transition with that of the Ising and Heisenberg models in the field of magnetism. In its original version, the model consists of a system of rod like molecules placed at the sites of a simple cubic lattice and interacting through the pair potential

$$U_{ij} = -\varepsilon_{ij} P_2(\cos\theta_{ij}), \tag{1}$$

where θ_{ij} is the angle between the symmetry axes of the i-th and j-th molecules and ε_{ij} has a value ε if i and j are nearest neighbours and is zero otherwise. Accurate and extensive Monte Carlo (MC) simulations [4] suggest that the LL model exhibits a weakly first order phase transition at a dimensionless temperature $T_c^* = k_B T/\varepsilon = 1.1232 \pm 0.0006$, where k_B is the Boltzmann constant, and the transition value of the long-range orientational order parameter $\langle P_2 \rangle_c$ is 0.27.

The potential represented by (1) may be regarded as a simplified version of the anisotropic interaction put forward by Maier and Saupe in their celebrated mean field (MF) theory [9]. The MF theory predicts a transition temperature $T_c^* = 1.3212$ and the transition order parameter $\langle P_2 \rangle_c = 0.429$ which are higher by about 18% and 58% respectively than the values obtained by computer simulation experiments on the LL model. Remarkable improvements over the MF results were achieved by adopting cluster variation methods [5–8, 10]. The two site cluster theory (TSC), the simplest of this approach [5–7] takes into account the correlations between a pair of neighbouring molecules, a feature totally ignored in the conventional MF calculation. The TSC calculations [5, 7] predict the values of $T_c^* = 1.1598$ and $\langle P_2 \rangle_c = 0.3866$ for the LL model.

As opposed to the MF theory, Faber's theory of nematic order [11–12] (or disorder, to be more precise) takes into account the correlations between neighbouring molecules from the very beginning. In this theory the misalignment in a nematic is described by a set of thermally excited periodic distorsion modes of the director field. Faber [12] has applied his theory to the LL model and has demonstrated that except at temperatures close to the nematic-isotropic transition it provides an accurate description of the results generated for the model by computer simulation. Later Roy [13] and more recently Mukhopadhyay *et al.* [14] have applied Faber's theory to several two-, three- and one dimensional lattice models and in all cases the low temperature behaviour of those systems are accurately predicted.

More recently a number of variations of the conventional LL model have been investigated both theoretically and with the help of computer experiments. We shall use the symbol d to indicate the lattice dimensionality and n to indicate the spin dimensionality. For molecules orienting in the three dimensional space, a linear ($d = 1, n = 3$) and a planar ($d = 2, n = 3$) lattice have been discussed by Vuillermot and Romerio [15–16] and not surprisingly these authors were able to provide an analytical solution showing the absence of a true phase transition for the $d = 1$ system [15].

The $d = 2$, $n = 3$ system, now known as the planar Lebwohl-Lasher system, has been the subject of an elaborate Monte Carlo simulation by Chiccoli *et al.*

[17] and an earlier but more restricted, investigation by Mountain and Ruijgrok [18]. According to a rigorous study by Vuillermot and Romerio [16], a class of such systems having Gegenbauer polynomial interactions, cannot have any conventional long-range order. However after the Kosterlitz-Thouless [17] findings of the existence of topological phase transitions in a class of systems of the $O(2)$, $n = 2$ type which fail to exhibit conventional phase transitions, interest has developed on the $d = 2$, $n = 3$ class of systems. In their study on a 20×20 lattice, Mountain and Ruijgrok excluded the existence of a first order transition and concluded that the ordering takes place through a higher order mechanism. Chiccoli *et al.*, have investigated five lattice sizes starting from 5×5 to one as large as 80×80 (which is necessary for revealing the estimates of a strongly fluctuating system) using the potential of equation (1) and the periodic boundary condition and their study of the orientational pair correlation function reveals a power law decay in the ordered phase and an exponential decay above the pseudo-transition temperature. Their results are consistent with the absence of a true phase transition but indicate a low temperature phase with long short-range order.

It must be admitted that so far there is no indication of the existence of topological defect structures in the planar LL system which could lead to the existence of the algebraic decay of order in the low temperature phase. There is however scope for further investigation as has been discussed in detail by Chiccoli *et al.* [17] and we have no intention of repeating these arguments.

The problem of the nature of the phase transition in the planar LL system, having thus been taken care of and discussed in detail by Chiccoli *et al.* [17], we now turn our attention to the application of the Faber's theory of misalignment to explain their MC results. As is well known, Faber's theory is valid for small distortions of the director field and is thus unable to explain the behaviour of a system in the vicinity of the nematic-isotropic transition. Keeping this in mind, it remains to be seen whether this theory is capable of predicting the temperature dependence of various parameters like $\langle P_2 \rangle$, $\langle P_4 \rangle$ and the pair correlation function $\sigma_2(\ell)$. Since the work of Chiccoli *et al.* has revealed a strong lattice-size dependence of those parameters and the MF and TSC theories being intrinsically insensitive to the system size, it will be of interest to test how far Faber's theory is capable of explaining both the systems-size and the temperature dependence of those parameters. Apart from this, we also discuss the behaviour of the system in the light of the MF and the TSC theories and findings of all three approaches are compared.

2 THEORETICAL INVESTIGATIONS

2.1 The Model in the Light of Faber's Theory

Computer simulation for this model have been performed by Chiccoli *et al.* [17]. Following these authors, we consider a system of $N(\equiv L^2)$ molecules, confined to a square lattice in which the separation between nearest neighbours is a. The anisotropic interaction is restricted to the nearest neighbours and takes the form as in equation (1). The periodic boundary conditions are assumed to hold good. Around each molecule in the lattice neighbouring molecules are coordinated in circles of radii ℓa, where

$$\ell^2 = \ell_1^2 + \ell_2^2 \quad (\ell_{1,2} = 0, 1, 2, \ldots). \tag{2}$$

In the present calculation attention has been confined to all allowed circles depending on the lattice size and in few cases these circles are degenerate, in the sense that more than one choice of ℓ_1 and ℓ_2 can be made, e.g. 5 and 0 or 3 and 4 both of which correspond to $\ell = 5$.

Following the assumptions and procedures which Faber [11] has fully explained, we first evaluate a quantity $\alpha(\ell)$, which is defined by the relation

$$\sum_{q \neq 0} \langle (\Delta\psi)_\ell^2 \rangle = \alpha(\ell) \sum_{q \neq 0} \langle \psi^2 \rangle, \tag{3}$$

where $\langle \psi^2 \rangle$ is the mean square angle through which the molecular axes are rotated when a single mode is thermally excited, $\langle (\Delta\psi)^2 \rangle$ is the mean square value of $(\psi_j - \psi_i)$ for two molecules separated by $r_{ij} = \ell a$, and the sums are over all possible values other than zero for the wave vector $\mathbf{q}$ of the periodic distorsion modes of the director field. We exclude $q = 0$ mode since it costs no energy and affects the orientation of the director for the specimen as a whole.

Once $\alpha(\ell)$ is known we may evaluate the three roots p', p'', p''' of equation (14) in paper IV of the series by Faber [12], and the associated amplitudes Q', Q'' and Q''', and proceed to evaluate the long range order parameter $\langle P_2 \rangle$ as a function of the reduced temperature from the equation,

$$T^* = k_B T/\varepsilon = -(4/3)\left(\frac{N}{N-1}\right) \ln \langle P_2 \rangle \Sigma, \tag{4}$$

where

$$\Sigma = Q' p' \langle P_2 \rangle^{(2/3)p'} + Q'' p'' \langle P_2 \rangle^{(2/3)p''} + Q''' p''' \langle P_2 \rangle^{(2/3)p'''}. \tag{5}$$

The long range order parameters are defined in the usual way as

$$\langle P_M \rangle = \langle P_M(\cos\theta_i) \rangle, \tag{6}$$

where θ_i is the angle between the i-th molecular axis and the direction in which it would lie if all the modes other than the $q = 0$ mode could be switched off.

The corresponding short range order parameter for two molecules separated by ℓa, defined as

$$\sigma_2(\ell) = \langle P_2(\cos\theta_{ij})_\ell \rangle, \tag{7}$$

are then obtainable from

$$\sigma_2(\ell) = (Q' \langle P_2 \rangle^{(2/3)p'} + Q'' \langle P_2 \rangle^{(2/3)p''} + Q''' \langle P_2 \rangle^{(2/3)p'''})_\ell, \tag{8}$$

and the reduced energy per molecule from

$$U^* = \langle U \rangle / N\varepsilon = -2\sigma_2(1). \tag{9}$$

To find $\alpha(\ell)$ we use the following equation which is a straight forward extension of one of Faber's results:

$$\alpha(\ell) = 2 \frac{\sum_{q \neq 0} (C_\ell / C_1)}{\sum_{q \neq 0} (1/C_1)}, \tag{10}$$

where

$$C_\ell = 1 - (z_\ell)^{-1} \sum_j \cos(\mathbf{q}.\mathbf{r}_{ij})_\ell. \tag{11}$$

z_ℓ is the coordination number for the ℓ-th shell and the summation on the right-hand side of (9) is over all neighbours of the i-th molecule which are separated from it by ℓa.

The values of q permitted by the boundary condition satisfy

$$\mathbf{q} = (2\pi/La)(\hat{n}_1 + \hat{n}_2), \tag{12}$$

with $-L < 2n_1 \leq L$, $-L < 2n_2 \leq L$.

Table I lists computed results for five values of L. A simple approximation, proposed by Faber [12], that $\langle P_4 \rangle = \langle P_2 \rangle^{(10/3)}$ is used to obtain the fourth rank order parameter.

2.2 The Model in the Light of MF and TSC Theory

The mean field approximation can be developed, leading to the one particle potential

$$\tilde{V}(\theta) = -zSP_2(\cos\theta), \tag{13}$$

where z is the coordination number of the first coordination shell and S is some variational parameter. The difference in the free energy per particle between the ordered and the isotropic phases is given by

$$\beta F_0^* = (z/2)\beta S^2 - \log Z_1 + \log 2, \tag{14}$$

where,

$$Z_1 = \int_0^{\pi} d\theta \sin\theta \exp\left[(z/T^*) SP_2(\cos\theta)\right], \tag{15}$$

is the single particle pseudo-partition function. Here $F_0^* = F_0/\varepsilon$ and $\beta = 1/T^*$.

The minimization of the free energy with respect to the variational para meter S gives $S = \langle P_M \rangle = \langle P_M \rangle_{z_1}$ i.e.,

$$\langle P_M \rangle = \frac{\int_0^1 d(\cos\theta)\, P_M(\cos\theta)\, P(\cos\theta)}{\int_0^1 d(\cos\theta)\, P(\cos\theta)}, \tag{16}$$

where θ is the orientation of the molecular axis with respect to the director and $P(\cos\theta)$ is the singlet orientational distribution, given by,

$$P(\cos\theta) = \frac{1}{Z_1} \exp\left[(z/T^*) SP_2(\cos\theta)\right]. \tag{17}$$

TABLE I Variation of $\alpha(\ell)$ with the system size

L	5	10	20	60	80
$\alpha(\ell)$	1.5789	1.1929	0.9491	0.7137	0.6699

Given this distribution, $\langle P_2 \rangle$ and $\langle P_4 \rangle$ can be obtained as a functions of T^* by applying equation (13) for both $M = 2$ and $M = 4$.

The mean field treatment can be refined by using various cluster variational techniques [5–8, 10]. The mean field theory may be regarded as a cluster expansion theory for which the number of particles in the cluster, say N_1, is unity. In this case $\langle P_2 \rangle$ and S are the same, but the two are not identical for larger N_1. Clusters are surrounded by cages of say, N_2 nearest neighbours, and the mean field represents the effects of interactions between members of the clusters and members of the cage. When N_2 is bigger than N_1, as is the case for two particle clusters, these effects are obviously of dominating importance, but in the limit of very large clusters N_2 is much less than N_1, and in that limit $\langle P_2 \rangle$ clearly tends to zero in a way that S does not.

For the model potential given by equation (1), the approximate free energy per particle in the TSC theory is given by,

$$\beta F_2^* = -(z/2)\log Z_{12} + (z-1)\log Z_1, \tag{18}$$

where Z_{12} is the two-particle pseudo-partition function,

$$Z_{12} = \int d\omega_1\, d\omega_2 \exp\left[\beta s(z-1)\{P_2(\cos\theta_1) + P_2(\cos\theta_2)\} + \beta P_2(\cos\theta_{12})\right] \tag{19}$$

and $d\omega_i = \sin\theta_i d\theta_i d\phi_i$. The condition of minimum free energy with respect to S gives the consistency requirement

$$\langle P_2 \rangle_{z_1} = \frac{1}{2}\langle [P_2(\cos\theta_1) + P_2(\cos\theta_2)] \rangle_{z_{12}}. \tag{20}$$

After solving for S, various thermodynamic observables are obtained by differentiating the free energy and gives,

$$U^* = -(z/2)\sigma_2, \tag{21}$$

where σ_2 is a short-range order parameter

$$\sigma_2 = \langle P_2(\cos\theta_{12}) \rangle_{z_{12}}. \tag{22}$$

The TSC heat capacity can be obtained by differentiating the energy with respect to the temperature for which the expressions are given in Ref. [18].

The integrals appearing in the consistency equations were calculated using the 32-point Gaussian formula and the consistency points were located by direct minimization of the free energy in terms of the variational parameter.

3 RESULTS AND DISCUSSIONS

Comparison with MC results shows that the MF theory qualitatively explains the thermodynamic behaviour of the system. For the smallest system it slightly overestimates the simulation results, but the agreements are rather poor if one goes to the larger systems. The transition temperature predicted by the theory is at $T^* = 0.881$ which is about 30% greater than the simulation result for the 5×5 system. On the other hand the TSC method offers a better approximation to the temperature variation of $\langle P_2 \rangle$ and $\langle P_4 \rangle$ than the MF results. The TSC theory slightly underestimates the values predicted by the simulation for the smallest system, whereas the prediction is rather good for the 10×10 system. In all other cases it overestimates the simulation results. The TSC theory remarkably improves the transition temperature over the MF theory to $T^* = 0.696$, which is only about 2% greater than the MC transition temperature for the smallest system studied so far. The transition values of the different parameters are summarized in Table II.

In Figure 1 we have compared the predictions of the different models for the temperature dependence of energy, $\langle P_2 \rangle$ and $\langle P_4 \rangle$ with the MC results for the 20×20 lattice. As far as only U^* is concerned, both the TSC model and Faber's theory, satisfactorily explain the MC results almost upto the transition temperature. This is however not so for $\langle P_2 \rangle$ and $\langle P_4 \rangle$. In these cases both the MF and the TSC fail to agree with the simulation data whereas Faber's theory very closely agrees with the MC results except of course at temperatures very close to the transition. It may be pointed out that even $\langle P_4 \rangle$, calculated with the simplifying approximation $\langle P_4 \rangle = \langle P_2 \rangle^{10/3}$ very closely matches the MC results.

TABLE II Results for the transition temperature, order parameters and entropy change during transition obtained by MC simulations, the MF and the TSC treatments

Method	T_c^*	$\langle P_2 \rangle_c$	$\langle P_4 \rangle_c$	$(\sigma_2)_c$	$\Delta S/Nk_B$
MC(5×5)	0.680	0.641	0.290	0.534	–
MF	0.881	0.429	0.119	–	0.418
TSC	0.696	0.358	0.081	0.394	0.211

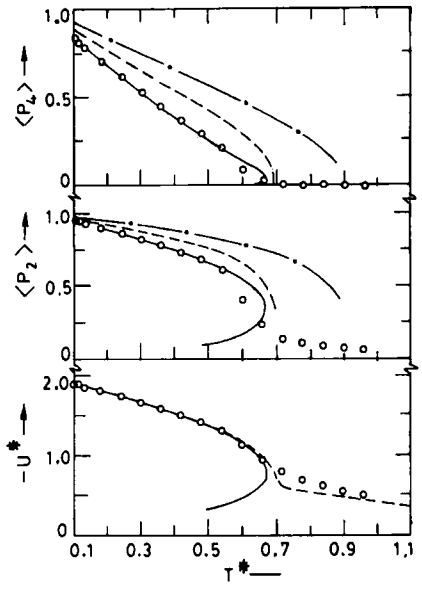

FIGURE 1 Results for the temperature dependence of energy and long-range order parameters for 20 × 20 system: a) (Circles): MC estimates; b) (Continuous line): Results of Faber's theory; c) (Dashed line): TSC results; d) (Dashed-dot line) MF results.

We now turn to the system size dependence of the various parameters as predicted by the three models. The results are depicted in Figures 2 to 4. The MF and the TSC models are evidently insensitive to the system size whereas Faber's model is not. We have chosen three temperatures, $T^* = 0.22$, 0.34 and 0.5 for the presentation of the results. The reduced energy U^* which is determined by the nearest neighbour correlation $\sigma_2(1)$ is not well explained by Faber's theory while the TSC results match closely with the MC data (Fig. 2). The mismatch is greater for large system size. However for $\langle P_2 \rangle$ and $\langle P_4 \rangle$ both the MF and TSC calculations completely fail to explain the computer data with any degree of precision.

As is evident from Figures 3 and 4, Faber's theory well explains the system size dependence of $\langle P_2 \rangle$ and $\langle P_4 \rangle$. The fact that the performance of the MF and the TSC approximations worsens as measured against increasing system size in the simulation may possibly be attributed to the strong fluctuations prevailing in the system. In a smaller system the box size cuts off long wavelength fluctuations which, given the large correlation length, are essential for the actual behaviour. Figure 5 shows the variation of $\sigma_2(\ell)$ with ℓ for the 80×80 lattice at $T^* = 0.54$ as is obtained from the MC results and Faber's theory and the agreement is excellent.

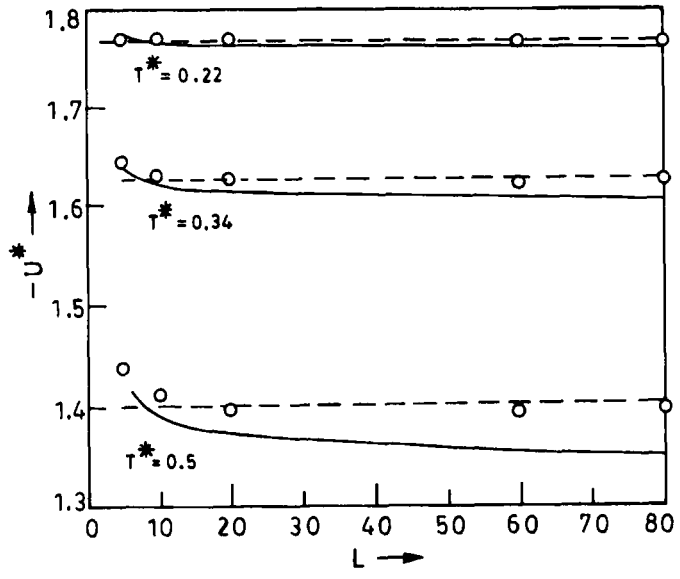

FIGURE 2 Results for the size dependence of energy: a) (Circles): MC estimates; b) (Continuous line): Results of Faber's theory; c) (Dashed line): TSC results.

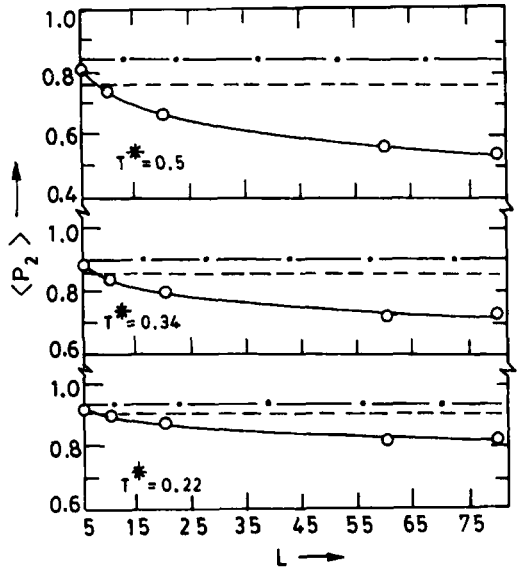

FIGURE 3 Results for the size dependence of second rank long-range order parameter: a) (Circles): MC estimates, b) (Continuous line): Results of Faber's theory; c) (Dashed line): TSC results; d) (Dashed-dot line): MF results.

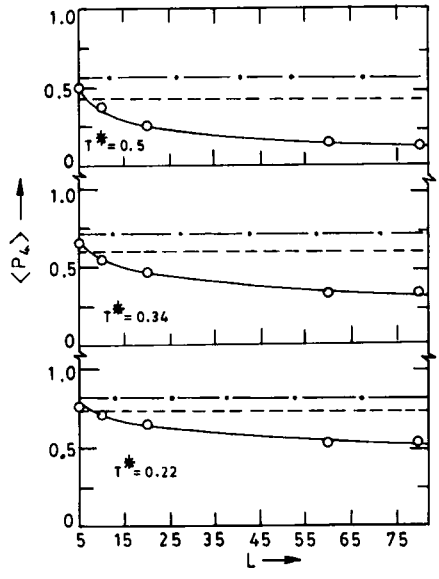

FIGURE 4 Results for the size dependence of fourth rank long-range order parameter: a) (Circles): MC estimates; b) (Continuous line): Results of Faber's theory; c) (Dashed line): TSC results; d) (Dashed-dot line): MF results.

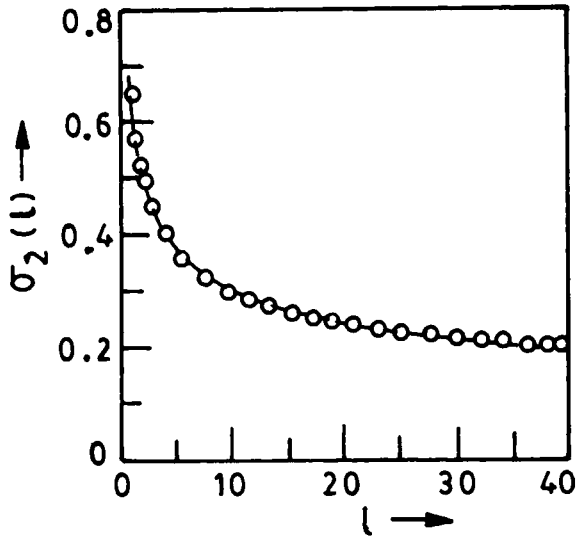

FIGURE 5 Variation of the pair correlation function $\sigma_2(\ell)$ with ℓ for 80×80 system at $T^* = 0.54$: a) (Circles): MC results; b) (Continuous line): Results of Faber's theory.

4 CONCLUSIONS

In conclusion, we may point out that although the continuum theory of nematic order as proposed by Faber fails to explain the behaviour of the present system near the order-disorder transition, it is capable of explaining the sensitivity of results on N in a satisfactory manner. The study of the size dependence of the various quantities shows that, some of the results, such as the order parameter, as is predicted by Faber's theory changes appreciably in going from 20×20 to 80×80 lattices, as is also evident from simulation results. The TSC theory clearly provides a much more realistic picture than the MF theory for the correlations of orientations between nearest neighbours, and they predict the order-disorder transition temperature with greater accuracy.

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