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Pretransitional fluctuations of the Lebwohl-Lasher model of a nematic liquid crystal

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We carry out Monte Carlo simulations of the Lebwohl-Lasher model of a nematic liquid crystal at temperatures just above the orientational ordering transition. We calculate the correlation functions for the order parameter fluctuations which are compared to the predictions of Landau theory in the Gaussian approximation in order to infer the values of the inverse susceptibility A and the elastic constant L. We also observe the distribution function for the invariant $\text{tr}Q^2$, which has simple analytic form if the fluctuations of the order parameter have a Gaussian distribution. At those temperatures for which the distribution of order parameter fluctuations is Gaussian, A is found to follow the Landau theory behavior $A(T) = a(T - T^*)$, with $(T_c - T^*)/T_c = 3 \times 10^{-3}$, where T_c is the nematic-isotropic phase transition temperature.

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INTRODUCTION

A nematic liquid crystal is characterized by a traceless, symmetric order parameter tensor Q in its ordered phase. Macroscopically, Q can be identified with the anisotropic part of the magnetic susceptibility, for example. For an idealized system of rodlike molecules, Q can be defined microscopically as

$$Q_{\alpha\beta} = \left\langle \frac{1}{N} \sum_i \frac{3}{2} \left(u^i_\alpha u^i_\beta - \frac{1}{3}\delta_{\alpha\beta} \right) \right\rangle , \qquad (1)$$

where $\mathbf{u}^i$ is a unit vector describing the orientation of molecule i. Within Landau theory, the free energy density associated with the nematic ordering is given by

$$f = \frac{1}{2}A\text{tr}Q^2 - \frac{1}{3}B\text{tr}Q^3 + \frac{1}{4}C(\text{tr}Q^2)^2 \qquad (2)$$

plus higher order terms [1,2]. It is conventionally assumed [1,2] that only the coefficient A is temperature dependent. The cubic term cannot be ruled out, since the transformation $Q \to -Q$ corresponds to the transformation from a prolate to an oblate distribution of the molecular axes and is thus not expected to leave the free energy invariant. The presence of the cubic term in the free energy expansion leads to a first order phase transition at a temperature T_c given by $A(T_c) = \frac{1}{27}\frac{B^2}{C}$, which is higher than the lower limit of stability of the isotropic phase T^*, which corresponds to $A(T^*) = 0$.

The linear response of the order parameter to an external field in the disordered phase is governed by a susceptibility χ, which is proportional to $1/A$. Experimentally, this corresponds to field-induced birefringence, which has been seen in both static magnetic fields [3] and strong optical fields [4]. The experiments indicate that the coefficient A is linear in temperature,

$$A(T) = a(T - T^*) \qquad (3)$$

as is usually assumed in the Landau theory. T^* is typically found to be on the order of 1 K below the nematic-isotropic phase transition temperature, so that $(T_c - T^*)/T_c \sim 3 \times 10^{-3}$. The proximity of the phase transition temperature to the hypothetical second order transition temperature T^* leads to strong enhancement of the ordering susceptibility just above the phase transition. Since the susceptibility is related by linear response theory to the correlation function of the order parameter fluctuations, these fluctuations are also strongly enhanced near the phase transition, which can be seen directly by light scattering techniques [3].

This paper is concerned with the study of pretransitional fluctuation effects in the Lebwohl-Lasher model. The Lebwohl-Lasher model [5] is a statistical mechanical model for a nematic liquid crystal in which molecules, whose states are specified by the orientation of a unit vector $\hat{\mathbf{u}}$, are confined to the sites of a cubic lattice. The interactions between the molecules are governed by the Hamiltonian

$$H = -\epsilon \sum_{\langle ij \rangle} P_2(\hat{\mathbf{u}}_i \cdot \hat{\mathbf{u}}_j), \qquad (4)$$

where the sum includes all pairs i,j of nearest neighbors. Mean field theory applied to this model yields the Maier-Saupe theory [6] of the nematic-isotropic phase transition. From its inception, the model has been the subject of simulation studies in order to clarify the role of the mean field approximation in the Maier-Saupe theory. In general, comparisons of simulations with mean field theory indicate that the mean field approximation gives a too strongly first order transition [5,7–9]. The latent heat at the transition is overestimated by a factor ≈ 2.5 [7,9], and recent simulation studies [8,9] indicate that the stability limit of the isotropic phase T^* is very close to the nematic-isotropic transition temperature, with $(T_c - T^*)/T_c \leq 3 \times 10^{-3}$, whereas mean field theory predicts $(T_c - T^*)/T_c = 10^{-1}$.

In the present work, we carry out simulation studies to observe directly the correlation functions for order parameter fluctuations in the isotropic phase of the

Lebwohl-Lasher model. Comparison of the simulation results with expressions obtained from Landau theory in the Gaussian approximation allow us to extract the value of the coefficient A in the free energy expansion, as well as the value of the elastic constant L, which governs the coupling of order parameter fluctuations at nearby points. Due to the finite size of the simulated system, both the ordered and disordered phases contribute to the statistical averages near the phase transition, whereas in the thermodynamic limit only the phase of lowest absolute free energy contributes. To check whether this finite size effect is contributing to the measured susceptibility, we examine the distribution function for the invariant trQ^2. This distribution is predicted to have a simple form in the thermodynamic limit, where the fluctuations of the spatially averaged order parameter have a Gaussian distribution. Very near the phase transition, significant departures from a Gaussian distribution for the fluctuations are found, but at sufficiently high temperatures a Gaussian distribution is observed. For those temperatures for which the fluctuations obey a Gaussian distribution, the coefficient A is well described by the Landau theory behavior, Eq. (3).

An essential element of our analysis here is the procedure we have developed to account for finite size effects near the transition thereby obtain improved estimates of the correlation length and susceptibility in the thermodynamic limit.

NEMATIC FLUCTUATIONS IN THE ISOTROPIC PHASE

Spatial correlations of the fluctuations of the order parameter can be described, in the long wavelength limit, in terms of a continuum Landau-Ginsburg theory. By comparing the Landau-Ginsburg result, obtained in the Gaussian approximation in which only invariants of second order in Q are retained, with the results obtained from the simulations, we can infer the values of the coefficient A and the elastic constant L. This approach for obtaining the elastic constant is closely analogous to the method used by Cleaver and Allen [10] to calculate the Frank elastic constant K in the nematic phase. Fluctuations above the nematic-isotropic transition are discussed in Refs. [11] and [12] and references therein. Here we give a brief discussion in order to introduce some ideas and notation used in analyzing the simulations.

If the free energy density is expanded about the isotropic state $Q = 0$, retaining only quadratic terms and considering the possibility of spatial variations in the order parameter, we have

$$f = \frac{1}{2}AQ_{\alpha\beta}Q_{\beta\alpha} + \frac{1}{2}LQ_{\alpha\beta,\gamma}Q_{\beta\alpha,\gamma}, \tag{5}$$

where we use the notation

$$X_{,\gamma} \equiv \frac{\partial X}{\partial x_\gamma}. \tag{6}$$

It is understood that (5) applies to long wavelength variations of Q, for which the underlying lattice should be irrelevant, so that we can treat Q as if it varies continuously in space.

For general nematics, there is another quadratic invariant in the gradients of Q of the form

$$\frac{1}{2}L_2Q_{\alpha\beta,\alpha}Q_{\gamma\beta,\gamma}, \tag{7}$$

but symmetry considerations imply that $L_2 = 0$ for the Lebwohl-Lasher model. In general, rotational symmetry requires that the free energy be invariant under the transformation

$$Q(\mathbf{x}) \to RQ(R^{-1}\mathbf{x})R^{-1}, \tag{8}$$

where R is a rotation matrix. This property holds for both (5) and (7). However, the invariance of the Lebwohl-Lasher Hamiltonian under rotations of the molecules implies that the free energy is invariant under rotations of the tensor indices alone,

$$Q(\mathbf{x}) \to RQ(\mathbf{x})R^{-1}. \tag{9}$$

Since the term (7) does not have this property, its coefficient L_2 must be zero. A more general Hamiltonian containing dependence on, for example, $\hat{\mathbf{u}}_i \cdot \mathbf{r}_{ij}$ would have $L_2 \neq 0$.

For the discussion of fluctuations, it is convenient to expand the order parameter as

$$Q_{\alpha\beta}(\mathbf{x}) = \frac{1}{\sqrt{V}}\sum_{\mathbf{k}}\sum_{i=1}^{5} q_{\mathbf{k}}^i\, E_{\alpha\beta}^i\, e^{i\mathbf{k}\cdot\mathbf{x}}, \tag{10}$$

where the E's are a set of five basis tensors satisfying

$$\begin{aligned} E_{\alpha\beta}^i &= E_{\beta\alpha}^i, \\ E_{\alpha\alpha}^i &= 0, \\ E_{\alpha\beta}^iE_{\beta\alpha}^j &= \delta_{ij}. \end{aligned} \tag{11}$$

An explicit representation having these properties has been given in Ref. [13]. In terms of the expansion coefficients $q_{\mathbf{k}}^i$, the total free energy $F = \sum_{\mathbf{x}} f$ becomes a sum of squares

$$F = \frac{1}{2}\sum_{\mathbf{k}}\sum_{i=1}^{5}(A + Lk^2)|q_{\mathbf{k}}^i|^2. \tag{12}$$

Then the probability distribution function for the $q_{\mathbf{k}}^i$ is a product of independent Gaussian functions and it is straightforward to evaluate the correlation function [14]

$$\begin{aligned} c^i(\mathbf{k}) &= \langle |q_{\mathbf{k}}^i|^2 \rangle \\ &= T/(A + Lk^2). \end{aligned} \tag{13}$$

The functions $c^i(\mathbf{k})$ are related to the real-space correlations of the order parameter tensor by a Fourier transform

$$\langle Q_{\alpha\beta}(\mathbf{x})Q_{\gamma\delta}(\mathbf{x}')\rangle = \frac{1}{\sqrt{V}}\sum_{\mathbf{k}}\sum_{i=1}^{5} c^i(\mathbf{k})\, E_{\alpha\beta}^i\, E_{\gamma\delta}^i\, e^{i\mathbf{k}\cdot(\mathbf{x}-\mathbf{x}')}. \tag{14}$$

By linear response theory, $c^i(\mathbf{k})/T$ is also the susceptibility which gives the response of the order parameter to an external field F, which couples to the system with an interaction energy $\sum_{\mathbf{x}} F^i E^i_{\alpha\beta} Q_{\alpha\beta}(\mathbf{x}) e^{i\mathbf{k}\cdot\mathbf{x}}$.

A special case of (13) of interest is $\langle \mathrm{tr}\bar{Q}^2\rangle$ where $\bar{Q}$ is the average order parameter $\bar{Q} = \frac{1}{V}\sum_{\mathbf{x}} Q(x)$,

$$\begin{aligned}\langle \mathrm{tr}\bar{Q}^2\rangle &= \frac{1}{V}\sum_i c^i(0)\\ &= \frac{5T}{VA}.\end{aligned} \tag{15}$$

In addition to the average value of $\mathrm{tr}\bar{Q}^2$, it is also of interest to consider its distribution function. [The case $\mathbf{k} = 0$ is exceptional in that q^i_0 is real and there is no double counting of $\mathbf{k}$ and $-\mathbf{k}$. These two factors cancel in the calculation of $c(k)$.] Introducing the shorthand

$$\begin{aligned}X &= \mathrm{tr}\bar{Q}^2\\ &= \frac{1}{V}\sum_i (q^i_0)^2,\end{aligned} \tag{16}$$

the distribution function for X is

$$\begin{aligned}P(X) &= \frac{1}{Z_0}\int d^5q\,\delta\left(X - \frac{1}{V}\sum_i q_i^2\right)\\ &\quad\times\exp\left(-\frac{1}{2}A\sum_i q_i^2/T\right)\\ &= \frac{4}{3\sqrt{\pi}}\left(\frac{VA}{2T}\right)^{5/2} X^{3/2}\exp(-VAX/2T),\end{aligned} \tag{17}$$

where Z_0 is a normalization constant.

MONTE CARLO SIMULATIONS

In our simulations, we have used a vectorized algorithm in which the lattice is divided into two sublattices which form the pattern of a chess board, which we refer to as black and white sublattices. Since all the neighbors of a molecule on the black sublattice are on the white sublattice, and vice versa, a Metropolis update of one black molecule has no effect on the other black molecules, and a whole sublattice can be updated as a vector. This idea has been widely used in simulations of lattice models with neighbor interactions. Some of these applications have been reviewed recently by Landau [15]. The validity of the method follows from the fact that it is a special case of the algorithms using sequential updates analyzed by Hastings [16].

At every temperature, the simulations were initialized in a perfectly ordered state, then equilibrated for 20 000 sweeps. Production runs of 500 000 sweeps followed for the calculation of averages. We have checked that the configurations were well equilibrated by performing a separate run at $T = 1.13$ starting from the final configuration of the previous run and comparing the distribution function for the order parameter. The correlation functions $c^i(\mathbf{k})$ were calculated for $\mathbf{k} = (k_x, 0, 0)$ by summing the order parameter tensor $Q_{\alpha\beta}$ over slices x =const, decomposing $Q_{\alpha\beta}(x)$ onto the basis of the E^i's, and performing a one-dimensional Fourier transform to obtain the coefficients $q^i_{\mathbf{k}}$. The values of $\langle |q^i_{\mathbf{k}}|^2\rangle$ were then obtained by averaging over the run. According to Eq. (13), $c^i(k)$ is actually independent of the index, i. Since the $q^i_{\mathbf{k}}$ are statistically independent in the Gaussian approximation, we use the variations of the individual c^i with i as a measure of the statistical error in the c^i. In Fig. 1 we show $c(\mathbf{k})$, which is the average over the index i of the calculated $c^i(\mathbf{k})$. The error bars shown are simply $1/\sqrt{5-1}$ times the rms deviations of the individual $c^i(\mathbf{k})$ from the average. Note that all results in this section are reported with ϵ taken as the unit of energy, the lattice spacing as the unit of length and ϵ/k_B as the unit of temperature.

Figure 1 shows the simulation results for $c(\mathbf{k})$, together with fits to the Landau theory form, Eq. (13), for temperatures ranging from 1.13 to 1.18. The nematic-isotropic transition temperature has been estimated for the Lebwohl-Lasher model to be [8,9] $T_c = 1.1232$. Clearly evident in the figure is the enhancement of the susceptibility on approaching the phase transition from above. The fits to the Landau theory appear to be reasonable, although the simulation data are systematically above the curves for larger wave vectors. This is more apparent when the reciprocal of $c(\mathbf{k})$ is plotted. Figure 2 shows $T/c(\mathbf{k})$ at $T = 1.16$ as a representative case. The open circles are the simulation data for a 30 × 30 × 30 simulation box, corresponding to what is shown in Fig. 1. In the Landau theory, this function is simply a quadratic $A + Lk^2$. Landau theory is expected to describe the long wavelength limit, but at shorter wavelengths the underlying lattice structure, along with the details of short-range correlations, become significant. In the present case, the lattice structure requires that $\partial c(\mathbf{k})/\partial k_x = 0$ at $\mathbf{k} = (\pi, 0, 0)$, which is reflected in Fig. 2. In light of the periodicity of $c(\mathbf{k})$ as a function of k_x, we have fitted

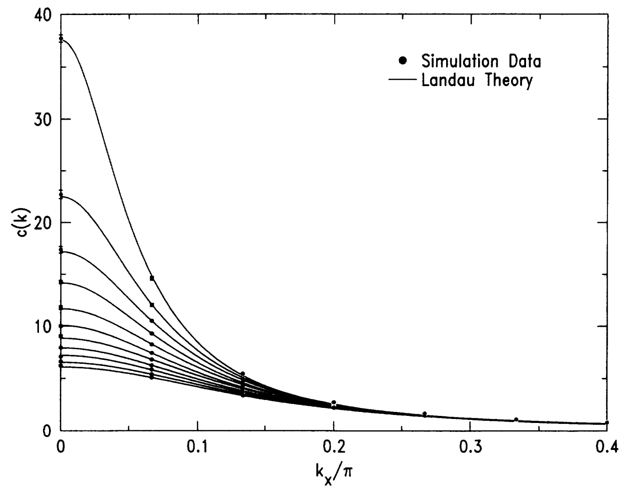

FIG. 1. The correlation function $c(k)$. The temperatures shown are, from top to bottom, T = 1.13, 1.135, 1.14, 1.145, 1.15, 1.155, 1.16, 1.165, 1.17, 1.175, and 1.18. Solid dots with error bars are the simulation data. Solid lines are fits to the Landau theory form, Eq. (13).

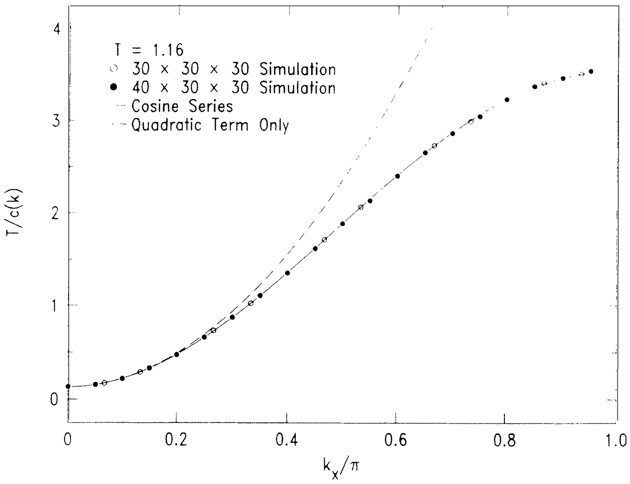

FIG. 2. The reciprocal of the correlation function $T/c(k)$ at $T = 1.16$. Open circles are the simulation data for a periodic box of dimensions $30 \times 30 \times 30$ in lattice units. Solid line is a cosine series fit. The dashed line shows only the quadratic term from the cosine fit. The solid dots are from a simulation with the x dimension of the box extended to 40 lattice units.

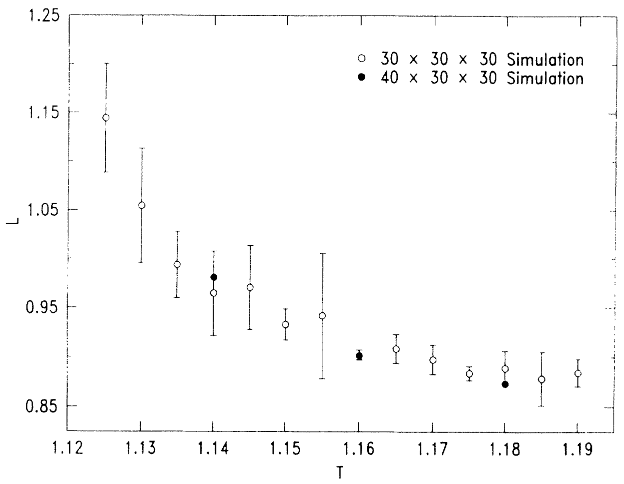

FIG. 3. Temperature dependence of the elastic coefficient L. Open circles are for a box of dimensions $30 \times 30 \times 30$ in lattice units. Solids dots are from a simulation with the x dimension of the box extended to 40 lattice units. The unit of temperature is ϵ/k_B.

$T/c(\mathbf{k})$ to a cosine series of the form

$$T/c(k_x) = \sum_n a_n \cos(n k_x) \,. \tag{18}$$

The coefficient L, as used in the plots in Fig. 1, was then obtained by differentiating the cosine series.

Retaining only the quadratic term of the fit from Eq. (18) we see from Fig. 2 that only the first three wave vectors lie on the quadratic part of the curve. The spacing of the wave vectors is set by the box dimensions by $\Delta k_i = 2\pi/l_i$. As a check that our result for L is not substantially effected by discretization of k due to the finite simulation box size, we have performed simulations at $T = 1.14$, 1.16, and 1.18 using a box which is extended in the x direction to 40 lattice units. The results at $T = 1.16$ are shown in Fig. 2 as the solid dots. The results from the larger simulation box seem to interpolate well between the points from the 30^3 box. This is typical of the results at the other temperatures, and the values of L obtained from the larger simulation boxes agree with the results from the smaller ones to within the statistical error. This is shown in Fig. 3, which gives the temperature dependence of L as obtained from $c(\mathbf{k})$. It is usually assumed that L is weakly dependent on the temperature. Figure 3 seems to indicate a fairly rapid increase of L on approaching the phase transition. However, as discussed below, for $T \leq 1.14$, the order parameter fluctuations in our simulations do not follow a Gaussian distribution, and the results obtained at these lower temperatures include finite size effects and a resulting deviation from the thermodynamic limit.

An important effect of the finite size of the simulation box is that near the phase transition, statistical averages will contain contributions from both the ordered and disordered phase, whereas in the thermodynamic limit only the phase corresponding to the absolute minimum of the free energy will contribute. In our case, averages like $c(0) \propto \langle \mathrm{tr} Q^2 \rangle$ will contain contributions from the ordered phase as we approach T_c from above and the value of $\chi \propto 1/A$ will not correspond to what will be obtained in the thermodynamic limit. As a check on this possibility, we have compared the distribution function for $\mathrm{tr}Q^2$ with the theoretical form, Eq. (17). Since the distribution for order parameter fluctuations is expected to be Gaussian in the thermodynamic limit, Eq. (17) should also be valid in this limit. Figures 4 and 5 show the distribution functions for $\mathrm{tr}Q^2$ obtained from our simulations for the temperatures $T = 1.16$, an intermediate temperature, and $T = 1.13$, a low temperature close to the phase transition. In both cases the upper curves show $\ln[P(X)/X^{3/2}]$ where $X = \mathrm{tr}Q^2$. According to Eq. (17), this is expected to yield a straight line. The scales of the x axes of the two plots are different, reflecting the different values of $\langle \mathrm{tr}Q^2 \rangle$ at the two temperatures, but in both cases the variation on vertical axis of the log plots is approximately 8, corresponding to a variation of the exponential factor of 3×10^3. Errors in the distribution functions have been estimated using $\delta n_i \approx \sqrt{n_i}$, where n_i is the number of samples in the ith bin of the histogram. The statistical independence of the samples is confirmed by the fact that an independent simulation at $T = 1.13$ gives the same distribution function to within the estimated errors.

Figure 4 shows that the theoretical form (15) gives an excellent fit to the simulation data at $T = 1.16$ over the whole range for which there are significant statistics. However, at $T = 1.13$, Fig. 5 shows significant departures from the theoretical distribution function at larger values of $\mathrm{tr}Q^2$. In the upper curve, we see that a linear form describes the data well for $\mathrm{tr}Q^2 < 0.01$ and the straight line shown is a fit to the data for $\mathrm{tr}Q^2 < 0.006$. However, the weight given to larger values of $\mathrm{tr}Q^2$ is greater than is expected from the theoretical distribution function. This is consistent with the idea that the ordered phase is contributing to $\langle \mathrm{tr}Q^2 \rangle$. In light of the fact that there is a substantial linear regime in the plot of $\ln[P(X)/X^{3/2}]$, which holds near $\mathrm{tr}Q^2 = 0$, we have considered another measure for the coefficient A by equating the slope with $-VA/2T$. This measure of A may be interpreted as being related to the susceptibility for small fluctuations about the disordered phase. In the cases where the distribution function, Eq. (17), is obeyed, this measure of A will be equivalent to that obtained from Eq. (15). Any difference between the two measures of A is indicative of a breakdown of the Gaussian distribution for the fluctuations of the order parameter, related to the small system size.

The interpretation that departure of the distribution function for $\mathrm{tr}\bar{Q}^2$ from the form given in Eq. (17) is due to finite size effects is supported by a simulation of a larger system. Figure 6 shows a plot of $\ln[P(X)/X^{3/2}]$ from the simulation of the 30 × 30 × 30 system, along with the result for a 40 × 40 × 40 system at $T = 1.13$. Together with the simulation results are fits to straight lines, as predicted by Eq. (17). Again, the fits are made to the data in the linear regime at smaller values of X. The x axes of the plots have been scaled by the volume, so that the two different systems can be plotted on the same scale. It can be seen that the larger system conforms significantly better to the theoretical form for the distribution function over the same range of variation of the exponential factor. This is also confirmed by the numerical results for the coefficient A. For the 30 × 30 × 30 system, we obtain $A = 0.0371 \pm 0.0005$ by fitting the distribution function $P(X)$ at small X, whereas we obtain $A = 0.030 \pm 0.001$ using Eq. (15) for $\langle \mathrm{tr}Q^2 \rangle$. For the 40 × 40 × 40 system we obtain $A = 0.0361 \pm 0.0003$ from fitting $P(X)$ and $A = 0.0331 \pm 0.0007$ from $\langle \mathrm{tr}Q^2 \rangle$. The two values of A are closer for the larger system, indicating that the theoretical distribution function, Eq. (17), is more closely obeyed for the larger system, although for 40^3 molecules, there is still a significant finite size effect at $T = 1.13$

In Fig. 7, we show our results for the coefficient A as a function of temperature. The open circles are obtained from $\langle \mathrm{tr}Q^2 \rangle$ using Eq. (15). The solid triangles show the result from fitting $\ln[P(X)/X^{3/2}]$ for small values of $X = \mathrm{tr}Q^2$. For temperatures ≤ 1.14, there are significant differences between the two results, indicating that

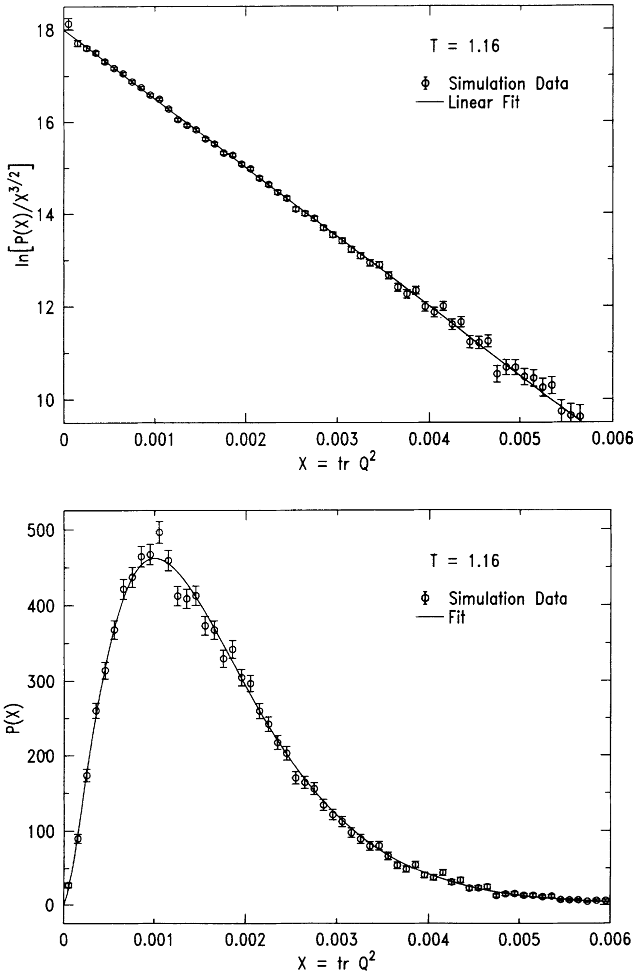

FIG. 4. The probability distribution function for $\mathrm{tr}Q^2$ at $T = 1.16$. Solid lines are a fit to the theoretical form Eq. (17).

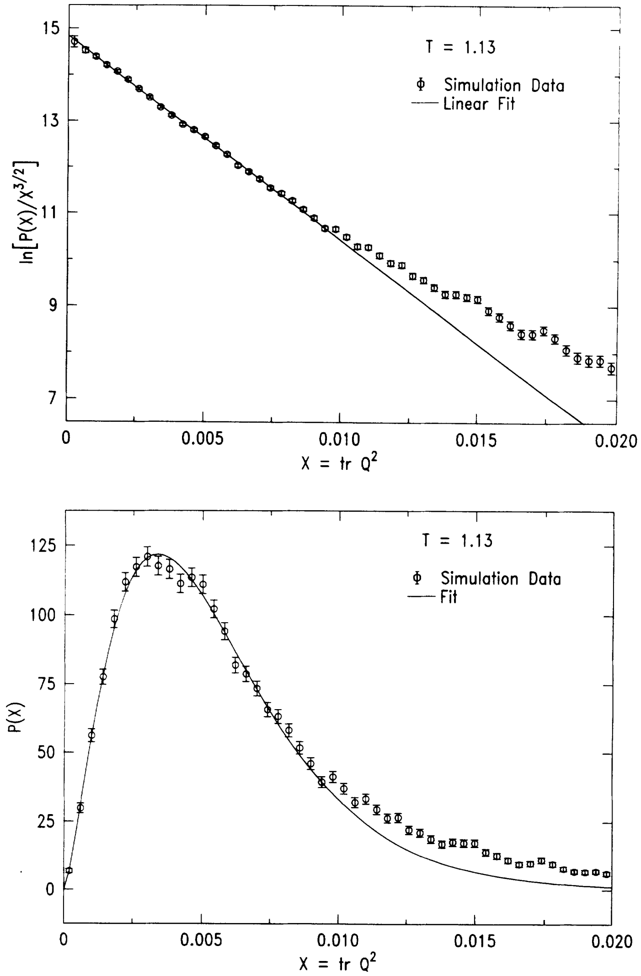

FIG. 5. The probability distribution function for $\mathrm{tr}Q^2$ at $T = 1.13$. Solid lines are a fit to the theoretical form Eq. (17).

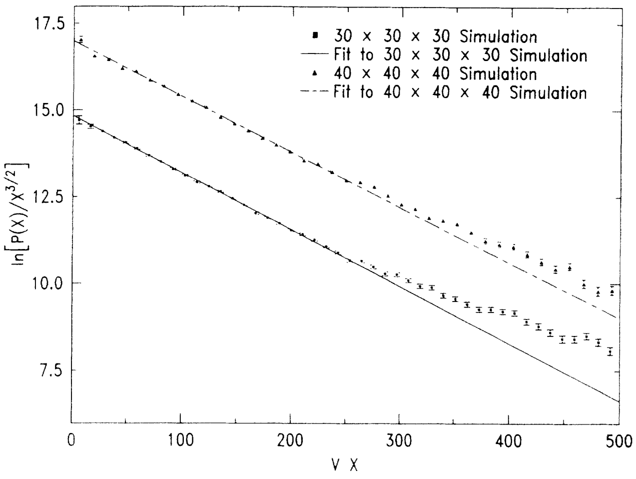

FIG. 6. The probability distribution function for trQ^2 at $T = 1.13$ for systems of 30^3 and 40^3 molecules. The X axis is scaled by the volume so that the two system sizes can be plotted on the same scale.

the order parameter fluctuations do not obey a Gaussian distribution. However at higher temperatures, the fluctuations are Gaussian, and the two results agree. The straight line is a fit to those points for which the fluctuations are Gaussian to the Landau form, Eq. (3). We obtain

$$a = 3.19 \pm 0.03,$$
$$T^* = 1.1194 \pm 0.0005, \tag{19}$$

where the uncertainties are estimates of the statistical errors. Error bars on $\langle \mathrm{tr}Q^2 \rangle$ have been obtained by standard block averaging procedures, with block lengths of 4000 Monte Carlo sweeps. Blocks of this length were found to be necessary at the lowest temperatures. The error bars in Fig. 7 are all smaller than the plotting symbols. This estimate of T^*, together with the existing estimates of the phase transition temperature [8,9] $T_c = 1.1232$, yield $(T_c - T^*)/T_c = 3.4 \times 10^{-3}$, which is comparable with the values obtained for real liquid crystals [3,4,18] and much smaller than the value obtained from mean field theory.

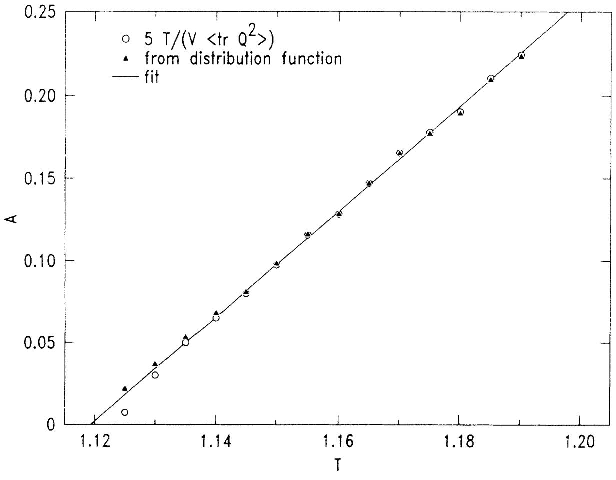

FIG. 7. The temperature dependence of the coefficient A. Open circles are obtained from $\langle \mathrm{tr}Q^2 \rangle$ using Eq. (15). Triangles are obtained by fitting the probability distribution function for trQ^2 to the form given by Eq. (17) as described in the text. The solid line is a fit to the data (open circles) for $T \geq 1.145$. The unit of temperature is ϵ/k_B.

CONCLUSIONS

We have carried out Monte Carlo simulations of the Lebwohl-Lasher model in the temperature range from $T = 1.125$ to $T = 1.19$ and observed the correlation functions for the order parameter fluctuations. These are compared with the predictions of Landau theory to obtain values of the elastic constant L and the inverse susceptibility A. We have also studied the distribution function for the fluctuating quantity tr$\bar{Q}^2$, where $\bar{Q}$ is the average over the volume of the simulation box of the order parameter tensor.

The distribution function for tr$\bar{Q}^2$ is predicted to have a simple form, Eq. (17), if the order parameter fluctuations are Gaussian. We find that this form is obeyed in our simulations for temperatures greater than ≈ 1.145. At lower temperatures, the ordered phase begins to contribute to the statistical averages, giving more weight to more ordered states. This effect is attributed to the finite size of the simulation, since in the thermodynamic limit, only the phase corresponding to the absolute minimum of the free energy contributes to statistical averages. For a system with linear dimension $l \gg \xi$, where ξ is the correlation length, the average order parameter is expected to have a Gaussian distribution as a result of the central limit theorem.

We find that, for temperatures where the fluctuations are Gaussian, the elastic constant L is a slowly decreasing function of the temperature and that the temperature dependence of A is well described by a Landau theory behavior $A(T) = a(T - T^*)$. Our value of T^*, the stability limit of the isotropic phase $T^* = 1.1194 \pm 0.0005$, is close to the value $T^* = 1.1201 \pm 0.0006$ obtained by Fabbri and Zannoni [8]. While their method was similar in spirit to the method used here, it is understandable that their estimate of T^* is higher than ours, since their fit involved a large number of points close to the ordering transition, which tend to raise the estimated T^*. We have not used these lower temperatures in our fit when they show substantial departures from a Gaussian distribution for the order parameter fluctuations, indicating substantial size effects. Our estimate of T^* is in contrast to the result of Zhang, Mouritsen, and Zuckermann [9], from whose Fig. 3 we estimate $T^* = 1.1228$ in the limit $l \to \infty$ and even higher for the system size considered here. The result of Zhang *et al.* is based on a very different approach of using a histogram technique to obtain the dependence of the free energy on the order parameter for temperatures in the transition region. This approach is close in spirit to the Landau theory from which the concept of the stability limit derives. However, there remain difficulties of principle in defining such stability limits outside of mean field theory [17]. Our procedure of extrapolating the susceptibility from the disordered phase is equivalent to the methods used in experiments on real liquid crystals. This approach gives an operational definition to T^*, while sidestepping questions of principle.

It is expected that near T^*, modifications of the mean-field-like behavior $A \propto (T - T^*)$ will appear due to interactions between the fluctuations. Very near T^*, one expects $A \propto (T - T^*)^\gamma$, with a nontrivial exponent γ. Departures from linearity of $A(T)$ have been observed in the range of ~ 1 K above T_c in experiments on the nCB (n cyanobiphenyl) series of nematics in Ref. [19]. In Ref. [12], it was found that these deviations from linearity could be fit by considering perturbative self-energy corrections to the correlation function $c(k)$. It is likely that a similar phenomenon will occur in the Lebwohl-Lasher model. However, if we translate the experimental temperature range to our units, it corresponds to $T < 1.126$ where the simulations suffer from substantial finite size effects. Thus it is not possible to address the question of critical behavior on the basis of the present simulations. Our main goal has been to study the behavior of the Lebwohl-Lasher model in comparison with the phenomenological theories which are able to describe the fluctuations in real nematic liquid crystals well over the range from ~ 1 to ~ 20 K above the transition using a mean field exponent.

The elastic term in the free energy $\frac{1}{2}LQ_{\alpha\beta,\gamma}Q_{\beta\alpha,\gamma}$ is obtained by keeping only terms quadratic in Q. This is appropriate for treating fluctuations about the disordered phase. If the same elastic energy is assumed to be valid in the nematic phase, the Frank elastic constant K is related to L by [11]

$$K = \frac{9}{2}LS^2, \tag{20}$$

where S is the scalar order parameter, which is the largest eigenvalue of Q. Numerical values for K at different temperatures in the nematic phase have been obtained by Cleaver and Allen [10]. If we use our value of $L = 0.93$, obtained at $T = 1.15$, we obtain $K/S^2 = 4.185$, which exceeds by 13% the value obtained in Ref. [10] at $T = 1.08$ in the nematic phase. This indicates that elastic terms which are of higher order in Q than quadratic make important contributions in the nematic phase. Cleaver and Allen found that K/S^2 was a decreasing function of S, so it is consistent that our value is larger than those obtained in the ordered phase.

In comparing the Lebwohl-Lasher model with experimental data on real nematics, there are two parameters to be adjusted. These are the interaction energy ϵ and the lattice spacing d. Restoring powers of length, energy, and Boltzmann's constant, we can summarize the earlier data on the transition temperature [8,9] and our present results as

$$\begin{aligned} k_BT_c/\epsilon &= 1.1232, \\ dL/\epsilon &= 0.93, \\ d^3a/k_B &= 3.19, \\ k_B(T_c - T^*)/\epsilon &= 0.0038 . \end{aligned} \tag{21}$$

Taking the values used in Ref. [12] for the material 8CB, $T_c = 313$ K and $L = 6.62 \times 10^{-12}\,\mathrm{J/m}$ to fix ϵ and d, we obtain for the other coefficients $a = 0.277 \times 10^6\,\mathrm{J\,K^{-1}m^{-3}}$ and $T_c - T^* = 1.06\,\mathrm{K}$. These are to be compared to the actual material values $a = 0.126 \times 10^6\,\mathrm{J\,K^{-1}m^{-3}}$ and $T_c - T^* = 1.12\,\mathrm{K}$. We see that there is order of magnitude agreement. The discrepancy is on the order of the variations among different materials [18]. Thus, despite the fact that it is unrealistic at a microscopic level, the Lebwohl-Lasher model seems to give a good qualitative representation of fluctuation effects seen in real nematic liquid crystals. Simulation studies of this model will be useful in studying the effects of fluctuations on other liquid crystal phase transitions, for example, those of liquid crystals in confined geometries, where the boundary conditions induce defects in the nematic order [20].

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