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Computer Simulations of Nematic Ordering with Random Disorder

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A Monte Carlo simulation of a nematic Lebwohl-Lasher liquid crystal model with a certain percentage of randomly oriented frozen spins is presented. The site-site distance dependence of the nematic correlation functions obtained are generally well fitted by stretched exponentials, which evolve into simple exponential at low temperature. The dependence of the correlation length on the disorder density is presented and compared to corresponding experimental results.

Keywords: Phase Transitions; Monte Carlo; Random Disorder

INTRODUCTION

The physics of liquid crystals incorporated in a silica porous matrix and of nematic liquid crystals containing dispersed colloidal nanoparticles has attracted considerable interest both from the experimental and the theoretical standpoints [1]. In particular the Isotropic (I) - Nematic (N) transition in these materials has been extensively studied experimentally, mainly by means of optical, calorimetric and magnetic resonance techniques [1]. Although the results offer a generally coherent description of the system indicating the

loss of nematic long range order, it remains unclear if, and under what circumstances, the disordered nematic structure is really short ranged or rather if it presents new disorder induced behavior, such as a "nematic Bragg glass" phase [2], characterized by quasi long range order. So far, theoretical modeling and computer simulation studies [3-8] of the spacial range and stability of the nematic order in the presence of quenched disorder, have tackled the problem in two ways:

i) by assuming that all relevant observed properties result from confinement, thus treating the disordered nematic as a collection of independent liquid crystal (LC) domains having suitable boundary conditions;

ii) by describing the system as a unconfined LC that contains a certain concentration of uncorrelated disordering sites. This approach allows to examine possible phenomena related to the high interconnectivity of the LC domains in the real systems.

All the proposed spin models present, in agreement with the experimental observations, a local orientational phase transition characterized by a rounded heat capacity peak increasingly shifted to lower temperatures as the silica density increases [1]. Here we wish to discuss a modified Lebwohl-Lasher model that we have previously proposed [9] where, during the simulation a certain number of spins is kept frozen in random orientations [8]. These frozen spins mimic the silica inclusions and are randomly dispersed inside the sample. A comparison with experimental observations is also proposed.

THE SIMULATION MODEL

Our simulation model, called Sprinkled Silica Spin (SSS) model [8], is based on a Lebwohl - Lasher (LL) lattice spin system [4,5] with the particles, assumed to be three dimensional "headless spins", interacting through the second rank pair potential:

$$U = -\sum_{i,j\in\mathcal{N}} \epsilon_{ij}\left[P_2(\mathbf{s}_i\cdot\mathbf{s}_j)\right] + J\sum_{k\in\mathcal{N},l\in\mathcal{S}} \epsilon_{kl}\left[P_2(\mathbf{s}_k\cdot\mathbf{s}_l)\right] \qquad (1)$$

where ϵ_{ij} equals $\epsilon > 0$ for nearest neighbors particles i and j and is zero otherwise, P_2 is the second rank Legendre polynomial. The

spin $\mathbf{s}_i$ represents a cluster of molecules whose short range order is maintained through the temperature range examined [6]. The second part of the hamiltonian of our model has been introduced to take into account the random aligning impurities and the parameter J denotes the strength of coupling between the nematic ($\mathcal{N}$) and these silica perturbers ($\mathcal{S}$). Although this parameter could be varied, e.g. to improve agreement with experiment, the effects of random disorder we wish to simulate should be universal and we have opted to keep J fixed to a value that represents the minimum departure from the presence of a nematic spin. Thus in the present simulation we have considered only the case $J = 1$. The concentration of disorder is then $p = \mathcal{S}/(\mathcal{N} + \mathcal{S})$ and the SSS model reduces to the LL system when the concentration p of the frozen spins goes to zero, correctly reproducing the bulk behavior of a nematic liquid crystals. In the present work the perturbers have an orientation $\mathbf{s}_k, k \in \mathcal{S}$ selected uniformly from a random distribution on a 3D sphere.

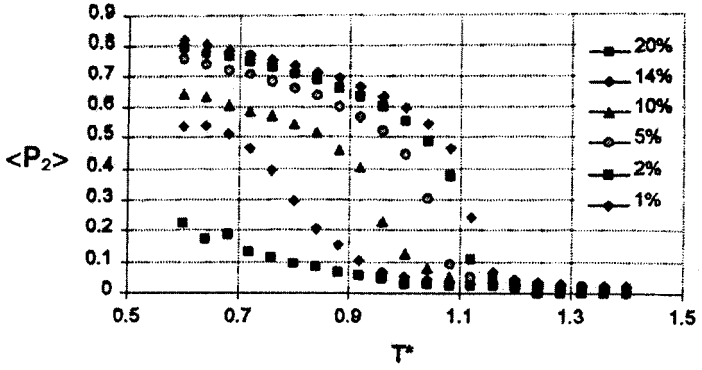

FIGURE 1 The second rank order parameter $\langle P_2 \rangle$ dependence on reduced temperature $T^* = kT/\epsilon$ as obtained from Monte Carlo simulations of a $20 \times 20 \times 20$ SSS system with various percentages of sprinkled disordered spins.

SIMULATIONS AND RESULTS

We have performed a number of MC simulations for various concentrations p of random spins on a $20 \times 20 \times 20$ lattice.

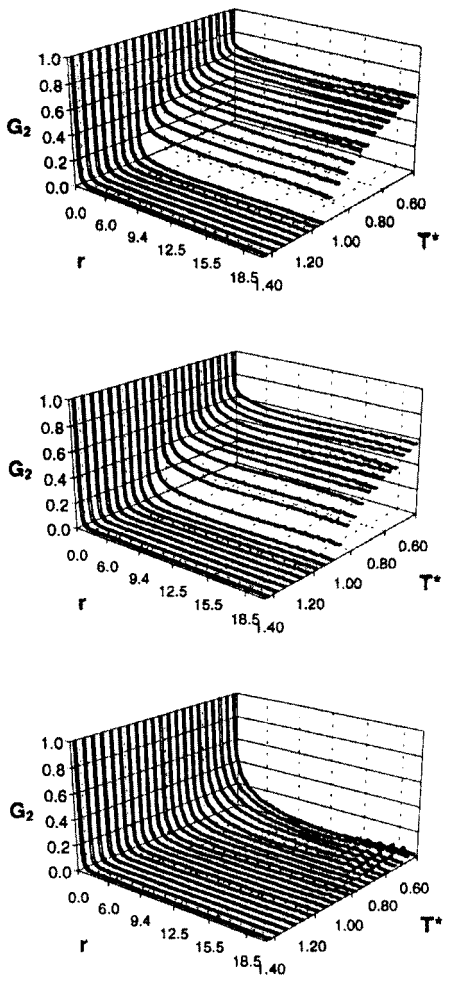

FIGURE 2 The dependence on distance and temperature T^* of the nematic orientational correlation function G_2^{NN} as obtained from MC simulations of a $20 \times 20 \times 20$ SSS system with various percentages of sprinked disorder: $p = 0.02$ (top), $p = 0.05$ (middle) and $p = 0.20$ (bottom).

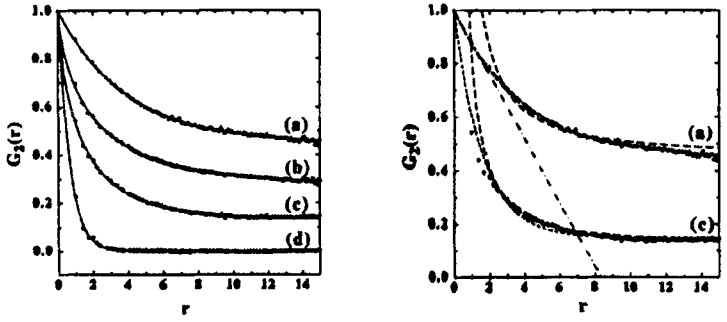

FIGURE 3 The orientational correlation function $G_2^{NN}(r)$ as obtained from Monte Carlo simulation on a $20 \times 20 \times 20$ SSS model with $p = 0.14$. *Left plate:* Simulation results (symbols) at different temperatures, i.e. $T^* = 0.2$ (a); $T^* = 0.6$ (b); $T^* = 0.8$ (c); $T^* = 1.2$ (d) and their fits with a Stretched Exponential function (lines). *Right plate:* Fit for cases (a) and (c), i.e. with exponential function (Short dashed lines); power law function (Long dashed lines) The dot dashed line denotes the limiting initial slope of curve (a).

In particular we have considered $p = 0.$, 0.02, 0.05, 0.1, 0.14, 0.2, 0.3, 0.4, 0.5 over a wide range of temperatures.

The updating of the lattice is done according to a standard Metropolis Monte Carlo procedure [10], reorienting one spin at a time. We have calculated energy, heat capacity, order parameters and correlation functions. The heat capacity, not reported for reasons of space, reproduces qualitatively the experimental results [8]. However, as in the case of other simulation models, the disorder induced temperature shifts are ten times larger than those observed in real experiments. This behavior is also confirmed by looking at the second rank order parameter shown in Fig. 1. At low concentration of disorder we find that the order is reduced but that it still propagates through the whole lattice. This is also confirmed by studying the nematic orientational correlation function given by:

$$G_2^{\mathcal{NN}}(r) = \langle P_2(\mathbf{s}_i \cdot \mathbf{s}_j) \rangle_r \qquad i,j \in \mathcal{N} \tag{2}$$

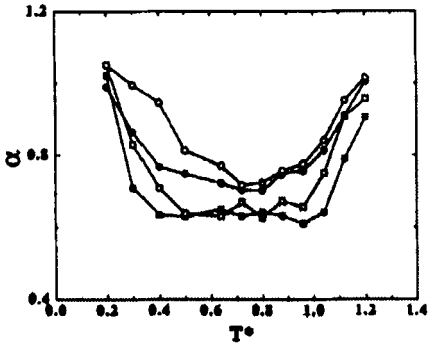

FIGURE 4 Exponent α of the Stretched Exponential function plotted vs the reduced temperature T^* as obtained by fitting the Monte Carlo results for different concentration p of quenched disorder: $p = 0.05$ (Open Squares), $p = 0.10$ (Filled Circles), $p = 0.14$ (Open Circles) and $p = 0.20$ (Filled Diamonds).

In Fig. 2 we show a set of G_2^{NN} curves as a function of distance and temperature for three different concentrations of disorder. In order to study the features of these correlations G_2^{NN}, we have systematically fitted them using power-law, exponential and stretched-exponential (SE) functions all with an added constant term to allow for any asymptotic plateau. The SE function is defined as

$$y(r) = (1 - A) + A \exp\left(-(r/\xi)^{\alpha}\right) \tag{3}$$

ξ and α being two independent fitting parameters. For every choice of p and T^* we obtain remarkably good SE fits (see Fig. 3, left plate). Power-law fits appear to be partially satisfactory only in a certain temperature range ($0.6 < T^* < 1.0$), but they are not generally acceptable, especially at low T^* (see Fig. 3, right plate). In Fig. 4 we show the T^* dependence of the SE exponent α. Both at high temperature (where the disorder is mainly provided by thermal fluctuations) and at low temperature (where the system can be described as an elastic distorted medium), the correlations are exponential ($\alpha \approx 1$). The SE shape of G_2^{NN} at intermediate temperatures is not particularly relevant *per se*, but because it continuously evolves from the low T^* exponential (short ranged) correlation, as

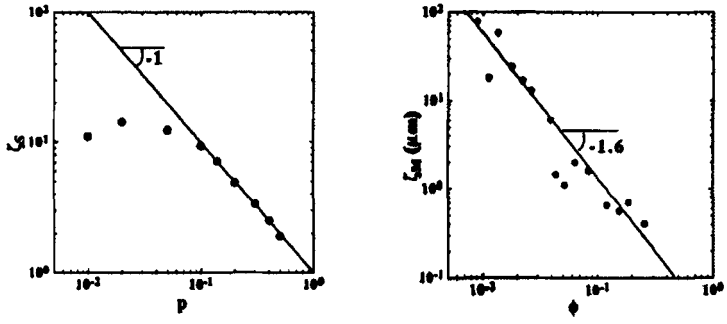

FIGURE 5 (*Left*) Correlation length ζ_S obtained from the simulation at $T^* = 0.2$, plotted as a function of the amount of quenched disorder p. The solid line fits ζ_S as obtained in the most densely disordered systems. (*Right*) Experimental nematic correlation length ζ_M measured in silica-distorted nematic systems and plotted as a function of the silica volume fraction ϕ. The line represents a power law best fit to the data.

shown in Fig. 4. This fact rules out the power law as a reasonable description of the data at intermediate temperature. On the other hand the problem of linking the SE fitting parameters to a precise microscopic origin is still to be investigated in detail. Assuming that the non-zero plateau of G_2^{NN} at large r is a finite size effect, we have extracted, from the simulated G_2^{NN} at $T^* = 0.2$ where $\alpha = 1$, the correlation length ζ_S as the inverse initial slope: $\zeta_S = \xi/A$. The p dependence of ζ_S is shown in Fig. 5 (*left plate*). We suggest that, given the size of the lattice, correlation lengths larger than $L/2 = 10$ could be heavily influenced by the finite size effects. ζ_S as obtained for $p \geq 0.1$ scale instead as $1/p$, is shown in Fig. 5 (*left plate*). In Fig.5 (*right plate*), we report the nematic correlation length ζ_M experimentally measured on samples obtained by dispersing silica nanoparticles in a thermotropic liquid crystal, plotted as a function of the silica volume fraction ϕ [11]. ζ_M has been obtained by measuring the optical turbidity of the samples as a function of T^*. The data are interpreted by using a detailed

model for the scattering of light from a distorted uniaxial medium under the assumption that G_2^{NN} has an exponential shape. The experimental and the simulated ζ can be described as power laws of the disorder density having different exponents. Although further investigation is needed to interpret this difference, we suggest that the discrepancy could be nested in the different spatial structure of the disorder, uncorrelated in the simulation while displaced on a fractal network in the experimental samples. A better comparison could be obtained once the graphs in Fig. 5 are plotted versus quantities more directly corresponding to each other, such as versus the typical length scale in both simulated and experimental systems.

Acknowledgments

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