Quasi-Long-Range Order in Nematics Confined in Random Porous Media

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(Received 8 December 1999)

We study the effect of random porous matrices on the ordering in nematic liquid crystals. The randomness destroys orientational long-range order and drives the liquid crystal into a glass state. We predict two glass phases, one of which possesses quasi-long-range order. In this state the correlation length is infinite and the correlation function of the order parameter obeys a power dependence on the distance. The small-angle light-scattering amplitude diverges but slower than in the bulk nematic. In the uniaxially strained porous matrices two new phases emerge. One type of strain induces an anisotropic quasi-long-range-ordered state while the other stabilizes nematic long-range order.

PACS numbers: 64.70.Md, 61.30.Gd, 82.70.–y

Quenched disorder is inevitably present even in the most pure solids. This explains a lot of phenomena, e.g., the residual resistance of metals. On the other hand, liquids are usually homogeneous and introducing quenched disorder in them requires special efforts. One of the approaches consists in pouring a liquid into a randomly interconnected network of pores. Such liquid-porous-matrix systems emerge in many natural and technological processes giving rise to a lasting scientific activity. The recent surge of interest in the field is due to a new micropore material: silica aerogel. Its density can be varied in a wide range up to more than 99% void volume fraction. This allows the investigation of both strongly and weakly confined fluids. The most interesting situation emerges in systems with many degrees of freedom, e.g., He-3 [1] and liquid crystals [2–7]. In these substances the porous matrix not only geometrically confines the liquid but also induces a random orienting field that fixes the direction of the order parameter near the surface of the matrix. The random-field (RF) disorder is known to cause spin-glass effects [8] and such phenomena were indeed observed experimentally in liquid-crystal-aerogel systems. In particular, a slow glassy dynamics was reported in Refs. [2,3]. Another effect of the disorder is the suppression of the isotropic-nematic phase transition.

In many experiments [2,4,5] the sharp isotropic-nematic transition inherent to the bulk liquid crystal was substituted by continuous ordering. The genuine phase transition was observed [6] only in highly porous aerogels. However, even these systems do not have nematic long-range order (LRO) as follows from the Imry-Ma argument [9,10] and the results of deuteron NMR measurements [7]. Hence, a question arises about the nature of the new phase emerging below the transition. This issue was addressed in several recent numerical simulations [11–14]. Most of them used simplified models with a discrete symmetry [11–13]. Although the structure of the phase diagram [11–13] agrees with the experiments the nematic LRO emerging in these models is an artifact of the discrete symmetry which allows LRO in three-dimensional RF systems [9]. An attempt to investigate numerically a more realistic continuous model [14] suggested an interesting scenario of quasi-long-range order (QLRO) that is the infinite correlation length and power dependences of the correlation functions on the distance. However, the results of Ref. [14] should be taken with care since the Imry-Ma domain length [15] is comparable with the sample size used for this simulation.

In the absence of reliable numerical results analytical approaches are especially desirable. However, the existing theory [13] does not extend beyond the mean field approximation. As in the other RF problems it underestimates fluctuations and incorrectly predicts nematic LRO. The present paper for the first time studies the low-temperature phase of disordered nematics beyond the mean field theory. We apply the functional renormalization group (RG) in $4 - \epsilon$ dimensions. RG flows have different character depending on the type of ordering. LRO corresponds to the fixed point in which the disorder and temperature are zero. If there is only short-range order the RG flow enters a strong coupling regime. A nontrivial fixed point describes QLRO. It was known for a long time that QLRO is possible in two-dimensional pure systems with the Abelian symmetry [16]. Recently the same type of ordering was discovered in three-dimensional RF systems [17,18]. In these systems QLRO is not prohibited by the non-Abelian symmetry [19]. Thus, QLRO is more common in disordered systems than clean ones. We demonstrate that QLRO emerges in our problem whereas LRO is absent. An experimental signature of this ordering is the divergence of the small-angle light-scattering cross section (9).

The free energy density $F = F_{\mathrm{d}} + F_{\mathrm{pm}}$ of the nematic in the porous matrix includes the Frank distortion energy [20] $F_{\mathrm{d}} = [K_1(\mathrm{div}\mathbf{n})^2 + K_2(\mathbf{n}\,\mathrm{curl}\mathbf{n})^2 + K_3(\mathbf{n} \times \mathrm{curl}\mathbf{n})^2]/2$, where $\mathbf{n}$ is the director, and the interaction F_{pm} with the surface of the random matrix. The interaction tends to align the director parallel to the surface [21]. We model the interaction as $F_{\mathrm{pm}} = (\mathbf{hn})^2$, where $\mathbf{h}$ is a random vector representing the normal to the surface. This is the simplest choice compatible with the equivalence of the opposite orientations of the director. Because of the

 0031-9007/00/84(21)/4886(4)$15.00

universality the model captures all large-scale physics and allows quantitative prediction of the critical exponents in the QLRO state. The average amplitude of the random vector $\mathbf{h}$ is a measure of the disorder strength. It is a phenomenological parameter which depends on the pore size, anchoring energy, and fractal structure of the porous matrix. The microscopic expression for $\mathbf{h}$ is discussed in Ref. [22].

In the one-constant approximation $K_1 = K_2 = K_3$ the energy $F = F_{\rm d} + F_{\rm pm}$ reduces to the Hamiltonian of the random-anisotropy (RA) Heisenberg model [19]. Since that model possesses QLRO the same ordering is expected for the randomly confined nematic. However, in all nematics $K_1, K_3 > K_2$ and this could change the critical exponents of the correlation functions in the QLRO state in comparison with the random Heisenberg model. Below we demonstrate that this is not the case, i.e., the nematic in the porous matrix belongs to the universality class of the RA Heisenberg model. To get a simple idea why it occurs we first consider a two-dimensional nematic film with the director $\mathbf{n} = (n_x, n_y, n_z) = (\cos\phi, \sin\phi, 0)$ confined in the plane xy of the film in the absence of the disorder. The Frank energy is $F_{\rm d} = (K_1 + K_3)(\nabla\phi)^2/2 + (K_3 - K_1)\{\cos2\phi[(\partial_x\phi)^2 - (\partial_y\phi)^2]/2 + \sin2\phi\,\partial_x\phi\,\partial_y\phi\}$. The low-temperature phase of this system possesses QLRO, only the term $(K_1 + K_3)(\nabla\phi)^2/2$ being relevant at the large scales since $\langle\sin2\phi\rangle = \langle\cos2\phi\rangle = 0$.

The systematic consideration is based on the RG equations in $4 - \epsilon$ dimensions. Our method follows the line of Ref. [19] and is briefly described below. It is convenient to eliminate the disorder with the replica trick [23]. We search for a zero-temperature fixed point. We ascribe the scaling dimension 0 to the director $\mathbf{n}$. Then the scaling dimension of the temperature $\Delta_T = -2 + O(\epsilon)$. Any term of the effective replica Hamiltonian containing m different replica indices is proportional [24] to $1/T^{m-1}$. This allows us to show that all operators with three or more replica indices are irrelevant [24]. Hence, all relevant operators of the appropriate symmetry are included in the following replica Hamiltonian:

$$H_R = \int d^3r \left[\frac{1}{2} \sum_a (\lambda_1 \partial_\alpha n^a_\beta \partial_\alpha n^a_\beta + \lambda_2 \partial_\alpha n^a_\alpha \partial_\beta n^a_\beta + \lambda_3 n^a_\alpha \partial_\alpha n^a_\beta n^a_\gamma \partial_\gamma n^a_\beta) - \sum_{ab} \frac{R(\mathbf{n}^a\mathbf{n}^b)}{T} \right], \tag{1}$$

where a, b are replica indices, $\alpha, \beta = x, y, z$ label the spatial coordinates, $\lambda_1 = K_2$, $\lambda_2 = K_1 - K_2$, $\lambda_3 = K_3 - K_2$, T is the temperature, the function $R(z)$ describes the disorder, and the summation over the repeated indices α and β is assumed. Because of the symmetry $\mathbf{n}^a \leftrightarrow -\mathbf{n}^a$ the function $R(z)$ is even. Below we measure the temperature in units of K_2, and hence set $\lambda_1 = 1$. To define the energy in $4 - \epsilon$ dimensions we add to the Hamiltonian (1) the term $\lambda_0 \sum_{\alpha\beta} \partial_\alpha n^a_\beta \partial_\alpha n^a_\beta/2$, where α labels the coordinates in the $(1 - \epsilon)$-dimensional subspace, $\beta = x, y, z$. The stability conditions [20] $K_1, K_3 > 0$ lead to the inequality

$$\lambda_2, \lambda_3 > -1\,. \tag{2}$$

We represent each replica $\mathbf{n}^a(\mathbf{x})$ of the director as a combination of small-scale fields $\phi^a_i(\mathbf{x}), i = 1, 2$ and a large-scale field $\mathbf{n}'^a(\mathbf{x})$ of the unit length:

$$\mathbf{n}^a(\mathbf{x}) = \mathbf{n}'^a(\mathbf{x}) \sqrt{1 - \sum_i [\phi^a_i(\mathbf{x})]^2} + \sum_i \phi^a_i(\mathbf{x}) \mathbf{e}^a_i(\mathbf{x})\,, \tag{3}$$

where the unit vectors $\mathbf{e}^a_i(\mathbf{x})$ are perpendicular to each other and the vector $\mathbf{n}'^a(\mathbf{x})$. The fields ϕ_i change at small scales $a < r < L$, where a is the molecule size, $L \gg a$. The field $\mathbf{n}'$ changes at the scales $r > L$. The RG procedure consists in integrating out the small-scale fields ϕ_i and the rescaling such that the effective Hamiltonian of the field $\mathbf{n}'$ would have the structure (1) with new constants. The rescaling is defined in such a way that $\lambda_1 = 1$ remains unchanged. The RG equations in the first order in $\epsilon = 4 - D$ read

$$\begin{aligned} \frac{dT}{d\ln L} &= -(D - 2)T + (1 - \lambda_3) C_\phi T\,; \\ \frac{d\lambda_2}{d\ln L} &= -\lambda_2(1 + \lambda_3) C_\phi\,; \\ \frac{d\lambda_3}{d\ln L} &= -(3\lambda_3 + \lambda_3^2 - \lambda_2) C_\phi\,, \end{aligned} \tag{4}$$

where the constant

$$C_\phi = \frac{dR(z = 1)/dz}{8\pi^2\sqrt{\lambda_0(1 + \lambda_3)}} \left[1 + \frac{1}{1 + \lambda_2} \right] \tag{5}$$

describes the fluctuations of the small-scale fields

$$\langle\phi_1^2\rangle = \langle\phi_2^2\rangle = C_\phi \ln(L/a)\,. \tag{6}$$

We omit the RG equations for λ_0 and $R(z)$ since their structure is irrelevant below. Equations (4) have the only fixed point compatible with the stability conditions (2). In this fixed point $T = \lambda_2 = \lambda_3 = 0$ and Eq. (1) reduces to the Hamiltonian of the RA Heisenberg model which thus describes the large-distance physics of the randomly confined nematic. Since that model possesses QLRO in its low-temperature phase for weak disorder, QLRO is also possible in confined nematics. For strong disorder or high temperature the ordering disappears. Thus, there are three phases: the high-temperature isotropic phase and two low-temperature glass phases with and without QLRO. In both glass phases the local orientation of the director is fixed by the random potential. As discussed below the disorder driven transition between the glass phases is related with topological defects.

The large-scale correlations of the director lead to strong small-angle light scattering. We determine its intensity

in the limit of the weak optical anisotropy, i.e., assuming that in the dielectric tensor $\epsilon_{\alpha\beta} = \epsilon_\perp \delta_{\alpha\beta} + \epsilon_a n_\alpha n_\beta$ the anisotropic term $\epsilon_a \ll \epsilon_\perp$. In this case the scattering cross section can be found with the Born approximation. The scattering cross section with the change of the wave vector by $\mathbf{q}$ is given by the expression [20]

$$\sigma(\mathbf{q}) = |\omega^2/(4\pi c^2) i_\alpha \epsilon_{\alpha\beta}(\mathbf{q}) f_b|^2, \tag{7}$$

where ω is the light frequency, $\mathbf{i}$ and $\mathbf{f}$ are the unit vectors specifying the initial and final polarizations, and $\epsilon_{\alpha\beta}(\mathbf{q})$ is the Fourier transform of the dielectric tensor. Hence, $\sigma(\mathbf{q}) \sim \langle Q_{\alpha\beta}(\mathbf{q}) Q_{\alpha\beta}(-\mathbf{q})\rangle$, where $Q_{\alpha\beta} = n_\alpha n_\beta - \delta_{\alpha\beta}/3$ is the order parameter and the angular brackets denote the disorder and thermal averages. In contrast to the bulk nematic the scattering is caused not by the thermal fluctuations but by the frozen configuration of the director. The cross section $\sigma(\mathbf{q})$ is proportional to the Fourier transform of the correlation function $G(\mathbf{r}) = \langle Q_{\alpha\beta}(\mathbf{0}) Q_{\alpha\beta}(\mathbf{r})\rangle$. In the QLRO state this correlator obeys a power dependence on the distance $G(r) \sim r^{-\eta}$. To calculate the exponent η we decompose $Q_{\alpha\beta}$ into small-scale and large-scale parts with Eq. (3) and average over the small-scale fluctuations with Eq. (6):

$$\begin{aligned}\langle Q_{\alpha\beta}(\mathbf{0}) Q_{\alpha\beta}(\mathbf{r})\rangle_\phi &= \left\{ n'_\alpha(\mathbf{0}) n'_\beta(\mathbf{0}) \left(1 - \sum_i \langle \phi_i^2\rangle\right) + \sum_{ij} e^i_\alpha(\mathbf{0}) e^j_\beta(\mathbf{0}) \langle \phi_i \phi_j\rangle - \delta_{\alpha\beta}/3 \right\} \\ &\times \left\{ n'_\alpha(\mathbf{r}) n'_\beta(\mathbf{r}) \left(1 - \sum_i \langle \phi_i^2\rangle\right) + \sum_{ij} e^i_\alpha(\mathbf{r}) e^j_\beta(\mathbf{r}) \langle \phi_i \phi_j\rangle - \delta_{\alpha\beta}/3 \right\} \\ &= Q'_{\alpha\beta}(\mathbf{0}) Q'_{\alpha\beta}(\mathbf{r}) [1 - 6 C_\phi \ln L/a], \end{aligned} \tag{8}$$

where $Q'_{\alpha\beta} = n'_\alpha n'_\beta - \delta_{\alpha\beta}/3$, $\langle \cdots \rangle$ denotes the average over the fluctuations of ϕ, and the relation $\langle \phi_i \phi_j\rangle \sim \delta_{ij}$ which is valid in the RA Heisenberg fixed point is used. The constant $C_\phi = 0.309\epsilon$ [Eq. (6)] is the same as in the fixed point of the RA Heisenberg model [19]. The exponent η can be found with the iterative use of Eq. (8) at each RG step until the scale $L = r$ is reached. At the scale r the values of the renormalized director field $\mathbf{n}'$ are the same at the points $\mathbf{0}$ and $\mathbf{r}$. Hence, $Q'_{\alpha\beta}(\mathbf{0}) Q'_{\alpha\beta}(\mathbf{r}) \sim 1$ and $r^{-\eta} \sim (1 - 6 C_\phi \ln L/a)^K$, where $K = \ln(r/a)/\ln(L/a)$ is the number of the RG steps. Thus, $\eta = 6 C_\phi$. The small-angle scattering cross section is given by the expression

$$\sigma(\mathbf{q}) \sim q^{-D+\eta} = q^{-4+2.9\epsilon}. \tag{9}$$

The uniaxial stress modifies the large-distance behavior. The compression along the z axis can be described by adding to the Hamiltonian the term $F_S = A n_z^2$, where $A > 0$, since the deformation tends to make the pore surfaces parallel to the xy plane and hence favors the planar configuration of the director. The uniaxial stretch is described by $F_S = A n_z^2$ with a negative A. In both cases A is proportional to the deformation. The effect of the electric field is analogous to the effect of the stress but the sign of the electric energy [20] $F_e = -\epsilon_a (\mathbf{nE})^2/8\pi$ is fixed for a given substance. The RG flow is unstable with respect to the perturbation F_S and new regimes emerge at the scale $R = R_c$ at which the renormalized $A(R) \sim 1$. The critical length R_c can be found analogously to the correlation length of the RA Heisenberg model in the uniform magnetic field [19]. At small A the result is $R_c \sim |A|^{-1/(2-2C_\phi)} = |A|^{-0.5-0.15\epsilon}$. The stretched system is long range ordered at the scales $R > R_c$. The nematic order parameter can be calculated analogously to the magnetization of the RA Heisenberg model in the uniform magnetic field and is given by the formula $Q = \langle n_\alpha n_\beta - \delta_{\alpha\beta}/3\rangle \sim R_c^{-3C_\phi} \sim |A|^{0.46\epsilon}$. LRO can also be achieved by applying an arbitrarily weak external magnetic field to the confined nematic since the magnetic contribution to the energy [20] $F_m = -\chi_a (\mathbf{nH})^2/2$ has the same structure as the energy related with the uniaxial stretch. A more interesting situation emerges under the compression. The director averaged over a scale $R > R_c$ is confined in the xy plane. The system is thus described by the RA XY model. It possesses QLRO but the critical exponents are different from the exponents of the Heisenberg model. Thus, at the scale R_c the crossover from one QLRO state to another occurs. Using the RA XY fixed point found in Ref. [19] and repeating the derivation of Eq. (9) one finds the Born light-scattering cross section for $q < 1/R_c$: $\sigma(\mathbf{q}) \sim q^{-4+\epsilon(1+\pi^2/9)}$. In the RA XY regime the cross section (7) is anisotropic: the small-angle scattering is suppressed, if the incident or scattered light is polarized along the compression direction.

Our RG procedure is based on the decomposition (3) which makes sense only if the director change is slow at the microscopic scale a. This condition is broken in the core of a topological defect. There are two types of the defects: disclination loops and point defects [20]. The disclination loop is a line the rotation by 2π around which reverses the director: $\mathbf{n} \to -\mathbf{n}$. The structure of the point defect is analogous to the structure of the hedgehog in the Heisenberg model. The topological defects are irrelevant at small $\epsilon = 4 - D$ for weak disorder. This can be understood from the consideration of the contribution of the disclination loops and the pairs of point defects of size $l \gg a$ to the RG equations at the scale l. After averaging over the small-scale fluctuations the size l of the topological excitations plays the role of the ultraviolet cutoff. The renormalized temperature is small: $T(l) \ll 1$. Hence, the thermal fluctuations are irrelevant. The disorder-induced term $R(\mathbf{n}_a \mathbf{n}_b) \sim \epsilon$ in the renormalized replica Hamiltonian (1) is of order $\langle h^4\rangle$, where the random vector $\mathbf{h}$

describes the (renormalized) random anisotropy $F_{\rm pm} = (\mathbf{hn})^2$, $\langle \cdots \rangle$ denotes the average over the realizations of the disorder. Inside a defect the director change is of the order of 1 at the cutoff scale. Hence, the elastic excitation energy determined by the renormalized Hamiltonian $H(l)$ (1) can be compensated by the interaction with the disorder only in the positions where $h \sim 1$. The concentration of such positions is exponentially small $\sim \exp(-1/\epsilon)$. Thus, the defects produce the corrections of order $\exp(-1/\epsilon)$ to the RG equations and do not modify the results of the paper qualitatively. The concentration of the topological excitations of size l is not more than of order $l^{-D}\exp(-1/\epsilon)$. The above discussion is valid, if the disorder is weak. In the case of the strong disorder the topological defects are present at the microscopic scale a and QLRO is absent. Thus, the topological defects drive the system into another glass state in which the orientation of the director is determined only by the local random potential. The critical strength of disorder at which QLRO disappears can be estimated by comparison of the elastic and random contributions to the energy. Since the Frank constants have usually the same order, QLRO is stable for $a^2\langle h^2\rangle \lesssim K_{1,2,3}$. The irrelevance of the topological defects for weak disorder can also be demonstrated with the energy argument [18] modified to take into account the scale dependence of the interaction. A possible fractal structure of the large disclination loops can lead to their strong suppression [25].

Recently it was suggested that the RF model (1) describes nematic elastomers [26]. However, it is unclear if this model and hence the above results are applicable to nematic elastomers because of elasticity-mediated nonlocal interactions in these substances [27]. The nonlocal dipole interactions are present also in the amorphous magnets which could be described by the RA Heisenberg model in the absence of the dipole forces. The effect of the long-range interactions on the stability of QLRO is an open question. Such forces are absent in randomly confined nematics which provide a genuine realization of the RA Heisenberg model and can be used for an experimental test of the possibility of QLRO in the non-Abelian systems. In conclusion, we have demonstrated that weakly disordered nematics possess QLRO in their low-temperature phase while LRO is absent for arbitrarily weak disorder. The uniaxial stretch and the external magnetic field stabilize LRO, and the uniaxial compression drives the liquid crystal into another QLRO state (see the phase diagram Fig. 1).

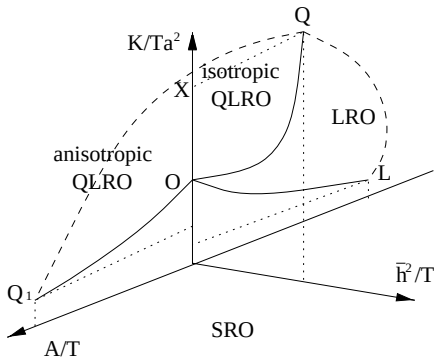

FIG. 1. The phase diagram in the one-constant approximation $K = K_1 = K_2 = K_3$. $\bar{h}^2$ is the average square of the random field, T the temperature, A the anisotropy, and a the molecule size. Isotropic QLRO exists in region OXQ that is the border between the LRO and anisotropic QLRO phases. Surfaces OLQ and OQQ_1 indicate the borders between the region of SRO and the regions with LRO and anisotropic QLRO, respectively.

The author thanks A. Kamenev, V. V. Lebedev, V. P. Mineev, and V. Steinberg for useful discussions, and R. Whitney for the critical reading of the manuscript. This work was supported by RFBR Grant No. 00-02-17763.

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