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Isotropic phase of nematics in porous media

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We study the effect of random porous matrices on the isotropic-nematic phase transition. Sufficiently close to the cleaning temperature, both random field and thermal fluctuations are important as disordering agents. A novel random field fixed point of renormalization group equation was found that controls the transition from isotropic to the replica symmetric phase. Explicit evaluation of the exponents in $d = 6 - \varepsilon$ dimensions yields to a dimensional reduction and three-exponent scaling.

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Liquid crystalline ordering in a confined geometry has been the subject of considerable investigation during the past decade. The study of liquid crystals constrained to a random network of porous silica aerogel has been an area of current interest due to their importance in technological applications and from a fundamental point of view. One of the fundamental questions is the effect of quenched disorder on the phase transitions. Liquid crystals exhibit a variety of experimentally accessible phase transitions involving orientational and translational ordering. Most of the studies are focused on the nematic-isotropic or nematic-smectic phase transitions. For example, the first has been investigated using various experimental techniques [1]. The main results could be summarized as follows: (a) the bulk isotropic-nematic (I-N) phase transition temperature is shifted down and the character of the transition changes; (b) even for above the bulk I-N phase transition temperature, there exists a weak residual nematic ordering; and (c) Monte Carlo simulations show that in some cases the nematic order is replaced by a quasi-long-range nematic phase.

Theoretical modeling of such phenomena is difficult. The porous matrix not only geometrically confines the liquid crystal, but also induces a random orienting field that fixes the direction of the order parameter near the surface of the matrix. Some experiments with liquid crystals in random porous media [2] have stimulated a random-field (RF) model for nematic liquid crystal [3] that qualitatively explains the glasslike behavior seen in experiments for liquid crystal-aerogel systems [2, 4 – 6].

The nematic phase within the pores could be modeled as an Ising-like system with an imposed random field coupled directly to the orientational order parameter to account for the random confinement. Such a model uses a random uniaxial anisotropy on a spin system [3, 7], including a symmetric coupling between the anisotropy vector and order parameter in order to account for the "up-down" nematic symmetry. This RF term in the Hamiltonian of the nematic liquid crystal is linear coupled to the order parameter. The strength of the random field in this model should directly depend on the anchoring strength of the molecules to the surface of the gel and indirectly on the porosity.

The basic point in discussing the effect of RF on ordered nematic phases follows from the Imry-Ma argument [8,9], which suggests that this continuous symmetry system does not have nematic long-range order for dimensions less than four ($d < 4$). The possibility for the nematic phase to be replaced by a glassy state characterized by quasi-long-range order was discussed in [10], and also predicted by numerical simulations [11], and a renormalization group (RG) approach [12].

The theory [12] is the first one that extends beyond the mean field approximation for the low-temperature phase of disordered nematics. In this low-temperature phase, uniaxial nematics in random porous media can be mapped onto the RF $O(N)$ model. However, mapping becames invalid near the phase transition to the isotropic phase. In this paper, we focus on the effects of quenched disorder that are introduced by the host silica aerogel at the high-temperature phase, i.e., above the I–N phase transition temperature. An appropriate model would require a full Landau–de Gennes type Hamiltonian incorporating a random orienting field. We carry out the mean field analysis and RG treatment as well.

The order parameter for a nematic liquid crystal is a three-dimensional symmetric traceless second rank tensor Q_{ij}. The effective Landau–de Gennes free-energy

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functional appropriate to the RF nematic model near the I-N phase transition can be written as

$$F = \int d^d x \{ \frac{1}{2} r_0 \mathrm{Tr}(Q^2) + \frac{1}{2} \mathrm{Tr}(\nabla Q)^2 - \frac{1}{3} b \mathrm{Tr}(Q^3) + \\ + \frac{1}{4} c [\mathrm{Tr}(Q^2)]^2 - \mathrm{Tr}[h(x) Q(x)] \}, \quad (1)$$

where $r_0 = T - T_0$, T_0 is the second order transition temperature if $b = 0$ (bulk supercooled temperature limit), and b, c are temperature independent constants. The quenched RF $h_{ij}(x)$ is a symmetric, traceless, Gaussian random tensor with vanishing quenched average $[h_{ij}(x)]_{av} = 0$ and with variance [13]

$$[h_{ij}(q) h_{km}(-q)]_{av} = h_0^2 (\frac{1}{2}(\delta_{ik}\delta_{jm} + \delta_{im}\delta_{jk}) - \frac{1}{n}\delta_{ij}\delta_{km}), \quad (2)$$

n is the dimensionality of the tensor h_{ij}.

Ground state configurations of the longitudinal component of the field $Q(x)$ (we consider here only the uniaxial nematic) are defined by the saddle-point equation

$$-\Delta Q + r_0 Q - bQ^2 + cQ^3 = h(x). \quad (3)$$

We recall first what behavior is expected for a nematic placed in a non-random field, i.e., a homogeneous field in a uniform direction. The isotropic phase acquires some order and is transformed into a paranematic phase. The paranematic-nematic phase transition occurs at $r_{0c} = (2b^2/9c)(1 + h/2h_c)$. Here $h_c = b^3/27c^2$ is a uniform critical field that determine the nematic-paranematic critical point, $r_0^+(h_c) = b^2/3c$. For $h < h_c$ the paranematic supercooling temperature T_0 and the nematic overheating temperature T^* both have field dependence. All three temperatures T_0 , T_c, and T^* merge at the nematic-paranematic critical point r_0^+. For $h > h_c$, the order parameter Q will increase smoothly as temperature is decreased.

Apparently, the solutions of the Eq. (3) with nonhomogeneous $h(x)$ may essentially depend on a particular configuration of the quenched fields. The effect of RF is averaged over a length scale L, over which the orientation is correlated. The mean magnitude of the sum of the random fields is given by the sum of the squares of the random fields. Using the central limit theorem, the effective RF which couples to the local order parameter is approximately $h_0 L^{-d/2}$. Now, because the order parameter is changing on a length scale L, the elastic energy term is of the form $(Q/L)^2$. Combining the ideas of Landau and those of Imry and Ma, it was shown, that for low order parameters $Q < ch_0/b^2$, the correlation length L is about a molecular length scale [7]. The free energy advantage is as though there were a fixed nematic field on the molecules, and thus negatively linear in Q. There is an energy cost in changing molecular orientation from point to point, but this is negligible because it is proportional to Q^2. Thus, for the isotropic phase the effect in this mean field consideration is roughly the same whether the imposed field is random or fixed.

Let us estimate under which conditions random fields are relevant and are getting a dominant contribution for the ground state configurations. We divide the system into blocks of linear size L. As we have seen, the characteristic value of the RF in this block (averaged over realization) could be defined by $h = h_0 L^{-d/2}$. In the case when the fields can be considered as the dominant factor, the order parameter does not depend on the temperature and it happens for $h > \tau^{\beta\delta}$. Here β and δ are the order parameter critical exponents as a function of temperature and field, respectively. Now it is easy to estimate the characteristic size of the block up to which the RF can dominate: $L < h_0^{2/d} \tau^{-2\beta\delta/d}$. On the other hand, the approximation we are using is correct only on length scales much larger than the fluctuation region $\xi \propto \tau^{-\nu}$. Thus, we have another bound for L: $L > \xi$. Therefore, the temperature region where RF effects cannot be ignored is [14],

$$|\tau| < \left(ch_0^2\right)^{1/(2\beta\delta - d\nu)} = \tau_h. \quad (4)$$

Such a region of temperatures near T_c exists only if $2\beta\delta > d\nu$. This value of τ_h can be interpreted as the estimate for the temperature interval around T_c in which the order parameter configurations are essentially defined by the random fields.

In the mean field theory, using Landau critical exponents, the above nontrivial temperature interval τ_h exists only at dimensions $d < 6$ and equal $\tau_h = (ch_0^2)^{2/(6-d)}$. These simple arguments hold only in the approximation where critical fluctuations can be neglected. Thus, the temperature region τ_h where disorder induces a finite correlation length $\xi(h_0) \propto (ch_0^2)^{-1/(6-d)}$ is correct in this regime only.

It is easy to estimate the Ginzburg criterion of the applicability of this approximation. For our model (1), one can get $\tau_G \propto \max[b^{4/(6-d)}, c^{2/(4-d)}]$, and the above result is valid only for $\tau > \tau_G$. On the other hand, the Ginzburg temperature region is larger than the metastable interval of the first order I-N phase transition $\tau_G > b^2/c$. For weak RF such that $\tau_h < \tau < \tau_G$, critical exponents get renormalized by thermal fluctuations, and in the region $\tau < \tau_h$, RF fluctuations are important as well.

The following qualitative arguments may be constructed. Actually, multiple global solutions of the sad-

dle point equation (3) can appear due to the double-well local potential. This potential has two local minima for $T_0 < T < T^*$ and for the values of the field $h < h_c$. At temperatures above T^*, the disordered local minima solution is unique. The energy of the nematic solution is higher than typical energy of the disordered solutions. At further temperature lowering, the interaction of the local minima solutions is getting not small. Like in spin-glasses [15], there are a large number of the disorder dependent local energy minima. In contrast to the usual spin-glass phase, these minima probably are separated by finite energy barriers. In this state the standard nematic order parameter equals zero, $[\langle Q\rangle]_{av} = 0$. Therefore, it is possible to expect the existence of a finite temperature interval between isotropic and nematic phases where the glassy-type behavior occurs, rather than the real spin-glass phase. At the same time, the application of external magnetic field H restores the long-range orientational order, and the magnetic field threshold is determined from the condition that the nematic coherence length $\xi_H \propto H^{-1}$ is less than the disorder induced correlation length $\xi(h_0)$.

In the glass-type phase, thermodynamics is defined by numerous disorder dependent local energy minima. The most developed technique in this case is the Parisi replica symmetry breaking method [16]. Using this technique, it has been proven that for the N-component ($N >> 1$) spin systems with RF, the usual scaling replica-symmetry solution is unstable with respect to the replica symmetry breaking in the phase transition point. Moreover, it turns out that the spin-glass transition, which is believed to take place at replica symmetry breaking temperature, always precedes the low temperature phase and obeys the equation $\tau_{RSB} \propto (h_0^2)^{4/(6-d)}$ [17]. If we compare τ_{RSB} with a RF controlled temperature region τ_h, we see that $\tau_h > \tau_{RSB}$.

Now we consider the disordered I-N model, defined by Eq.(1) within the high-temperature, i.e., isotropic phase. We assume the existence of rather strong fluctuations of the order parameter in the isotropic phase near the I-N transition, for which experimental evidence exists [18]. The Landau–de Gennes Hamiltonian has cubic, quartic interaction terms, and the RF term, therefore, there are three length scales in the fluctuation theory: $\xi_c \propto c^{-1/(4-d)}$, $\xi_b \propto (b^2)^{-1/(6-d)}$ and $\xi_h \propto (ch_0^2)^{-1/(6-d)}$.

Let one remove the fast modes and rewrite the Hamiltonian in terms of the block order parameter, corresponding to the scale $L = al$. Here a is the ultraviolet cutoff, and $l > 1$. Then we make rescaling such a way that the Hamiltonian would restore its initial form with new constants $b(L)$, $c(L)$, and $h_0(L)$. Dimensional analysis provides estimations

$$b(L) = l^{(6-d)/2} b(a),\ c(L) = l^{4-d} c(a),\ h_0(L) = l h_0(a). \tag{5}$$

If one considers the combination $\Delta = ch_0^2$ as a new parameter, we immediately get

$$\Delta(L) = l^{6-d}\Delta(a). \tag{6}$$

Iteration until $\Delta(L_0) = 1$ yields $L_0 = \xi_h$, i.e., the length scale beyond which the RF fluctuations are significant. The same arguments are true for the order parameter fluctuations coming from cubic term in (1). The quartic term is an irrelevant variable in the RG sense. Hence, the two length scales are important for IN phase transition near $d = 6$. Thus we interpret this result physically by noting that sufficiently close to T_c, the dominant disordering agent is not the RF only, but the thermal fluctuations caused by cubic interaction also. Using the RG method for disordered systems, recursion relations are established for the parameters of the effective replica Hamiltonian. Then replica symmetry is assumed and the RG equations become simple functions of replica number. In that respect, the use of replica is a trick of diagram counting. One can generally establish identical RG equations directly by considering disorder correlation functions, a method which is usually called a replica symmetry perturbation theory. After standard RG transformations, the one-loop equations in differential form are the following

$$\frac{dr}{d\ln L} = (2-\eta)r - \frac{7}{6}b^2(1-2r) + 7\Delta(1-2r), \tag{7}$$

$$\frac{d\ln b^2}{d\ln L} = \varepsilon - 3\eta - b^2 - 24\Delta, \tag{8}$$

$$\frac{d\ln \Delta}{d\ln L} = \varepsilon - 3\eta + \frac{22}{3}b^2 - 26\Delta. \tag{9}$$

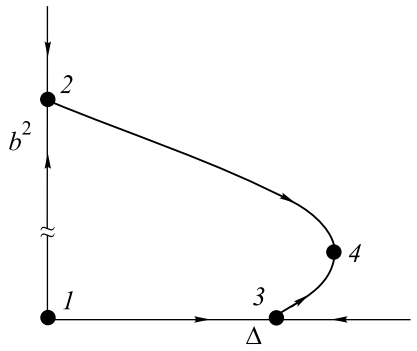
Here we put $n = 3$ for a nematic liquid crystal, and $\varepsilon = 6 - d$.

The exponent η determines the behavior of the two-point correlation function $G(q)$, which is defined by means of the relation

$$G(q) = [\langle Q(q)\, Q(-q)\rangle]_{av} - [\langle Q(q)\rangle\langle Q(-q)\rangle]_{av}. \tag{10}$$

At the critical point, $G(q)$ diverges as $q^{\eta-2}$, and to the lowest order in the perturbation expansion,

$$\eta = 7b^2/18. \tag{11}$$



Phase diagram of the RG equations. Points 1, 2, 3, and 4 stand for the fixed points μ_0, μ_b, μ_Δ, and μ^*, respectively.

We find that the fixed points $\mu(b^2, \Delta)$ of the RG equations are given by $\mu_0(0,0)$, $\mu_b(6\varepsilon/13, 0)$, $\mu_\Delta(0, \varepsilon/26)$, and $\mu^*(6\varepsilon/613, 25\varepsilon/613)$. The RG flow diagram in the (b^2, Δ) plane is illustrated in figure.

In addition to the trivial Gaussian fixed point μ_0, these equations possess three nontrivial fixed points. The fixed point μ_b describes the critical behavior of the pure nematic and the coefficient r at this point is greater than zero. Thus fixed-point Hamiltonian has a minima at $Q = 0$ and at $Q \approx b/c$. The first-order transition occurs if the order parameter falls into the later deep minimum. It is likely that the μ_b fixed point corresponds to the critical fluctuations about the metastable minimum at $Q = 0$ [19]. The fixed point μ_Δ is exactly RF Heisenberg fixed point for the five component $O(N)$ model and it describes the RF behavior at the isolated Landau point on the phase diagram, where $b = 0$ [20]. All the above fixed points are unstable.

The only stable fixed point is μ^*, that controls the behavior of the relevant parameters of the Hamiltonian below six dimensions, and corresponds to the replica symmetric phase with an infinite correlation length.

Let us now determine the critical exponents associated with the fixed point μ^*. The correlation length exponent ν follows directly from Eq.(7)

$$\nu^{-1} = 2 + \frac{35}{18} b^2 - 14\Delta. \tag{12}$$

Using the RG iterations and the perturbation expansion for two-point correlation function one can obtain the susceptibility exponent

$$\gamma = 1 - \frac{7}{6} b^2 + 7\Delta. \tag{13}$$

The specific heat exponent α can be calculated from a singular part of the free energy

$$F_s \propto \int_0^{\ell^*} \ln(1+\widetilde{r}\,(\ell)) e^{-d\ell} d\ell,$$

where $\widetilde{r}(\ell)$ is the coefficient of the Q^2 term in Hamiltonian averaged over the distribution of the RF, and $\ell^* = \ln \xi$. Evaluating the above integral to leading order we find $F_s \propto \tau^{2-\alpha}$, where

$$\alpha = \frac{6-d}{2} + \frac{7}{4} b^2 - 14\Delta. \tag{14}$$

Equations (11)–(14) yield the usual "thermodynamic" scaling law $\gamma = \nu(2-\eta)$, and modified hyperscaling law $2 - \alpha = \nu(d - \theta)$ with the "violation of hyperscaling" exponent $\theta = 2 - \eta$. This result is valid at first order in ε.

In the presence of the RF, the quantity $[\langle Q(0)\rangle\langle Q(x)\rangle]_{av}$ is non-zero even in phases where $[\langle Q(x)\rangle]_{av}$ vanishes. There are therefore two distinct correlation functions to consider. The first is the analog of the usual connected correlation function G (10), and the second is the disconnected function and is specific for random systems. It measures the fluctuations in the local quenched order parameter:

$$C_s(q) = [\langle Q(q)\rangle\langle Q(-q)\rangle]_{av} - [\langle Q(q)\rangle]_{av}[\langle Q(-q)\rangle]_{av}, \tag{15}$$

and diverges at small q as $C_s \propto q^{\tilde{\eta}-4}$.

Near T_c, we can write $C_s(q) = G^2(q)D(q)$ [21], where $D(q)$ is related to a dressed spectral function $h_0^2(q)$ of the RF fluctuations. If $h_0^2(q) \propto q^{-\lambda_\Delta}$ for $q\xi >> 1$, then one obtains $C_s(q) \propto q^{2\eta-4-\lambda_\Delta}$ and $\tilde{\eta} = 2\eta - \lambda_\Delta$. Note that the choice $\lambda_\Delta = 0$ yields $\tilde{\eta} = 2\eta$, a value which is on the limit of the exact inequality $\tilde{\eta} \leq 2\eta$, due to [22]. Another relation was suggested by consideration of the RF contribution to the free energy in a correlation volume which scale as ξ^θ. In contrast, for the pure system the characteristic scale of variation of the effective free energy is simply set by the thermal fluctuations, i.e., $\propto T$. On the other hand, if the local order parameter was uncorrelated with the RF this would scale as $\xi^{d/2-\beta/\nu}$. Here the factor $\xi^{d/2}$ is coming from the scaling of the total RF. The relation for β is easy to find from scaling the disconnected correlation function in a real space $2\beta = (d-4+\tilde{\eta})\nu$. Since the correlations could be included by additional factor $\xi^{\lambda_\Delta/2}$, corresponding to $h_0^2(q) \propto \xi^{\lambda_\Delta}$ for $q\xi << 1$, we expect that $\theta = 2-\eta+\lambda_\Delta$. The case $\lambda_\Delta = 0$ yields $\theta = 2 - \eta$, which is on the limit of another inequality $\theta \geq 2 - \eta$ [23]. Thus for the violation of hyperscaling exponent we recover the result $\theta = 2 + \eta - \tilde{\eta}$ [14, 17, 23, 24].

In one-loop calculations, the critical exponent η is determined by (11), and $\tilde{\eta} = 2\eta$. The Δ dependent terms

appear in $C_s(q)$ only in the two-loop diagrammatic expantion for $D(q)$

$$D(q) = h_0^2(1 + \frac{7}{3}\Delta(b^2 - 6\Delta)\ln q), \qquad (16)$$

and now the particular value $\lambda_\Delta = (7/3)\Delta(6\Delta - b^2)$ is non-zero to the second order in ε. All diagrams in (16) must be disconnected before averaging over the RF distribution.

Formally we can divide the diagrams contributing to $G(q)$ and write $\eta = \eta_1(b^2) + \eta_2(\Delta^2, b^2\Delta)$. In the one-loop approximation, $\eta_2 = 0$ and η_1 is given by (11). A straightforward evaluation of the RF depended diagrams leads to expression $\eta_2 = \lambda_\Delta$. This means that for the hyperscaling violation exponent we get $\theta = 2 - \eta_1$. In contrast to the disconnected correlation function, all diagrams here are connected before configuration averaging, and not all of them are tree-like diagrams, as it is the case for the $O(N)$ model.

More generally, in the vicinity of the fixed point μ^* the random correlation function is proportional to c^{-1} for small c. Therefore, in the critical region one expects that the random correlation function will scale as $C_s(q, \xi, c) = c^{-1}e^{(2-\eta-\lambda_c)\ell}C_s(e^\ell q, e^{-\ell}\xi)$, where λ_c is the scaling exponent of the irrelevant parameter c. For $q = 0$ one has the behavior $C_s(0, \tau) \propto \tau^{-\tilde{\gamma}}$ with $\tilde{\gamma} = \nu(2 - \eta - \lambda_c)$. Using the relation $\tilde{\gamma} = \nu(4 - \tilde{\eta})$ that follows from the scaling at small q and $\tau = 0$ we can write $\lambda_c = \tilde{\eta} - \eta - 2$. We see that $\lambda_c = -\theta$. This result is quite obvious. Really, on the other hand, the perturbation expansion for free energy is a double power series in b^2, c, and h_0^2. The first terms in this series behave like $b^2h_0^2$ and ch_0^4, or for large h_0^2 they both are proportional to h_0^2 as well. Thus, for the free energy density we have $F(h_0^2, \tau) = \tau^{\nu d}f(h_0^2\tau^{-\varphi})$, where φ is the crossover exponent. If we conclude that f is a linear function of its argument for small τ, as it follows from the perturbation expansion, one can get $F(h_0^2, \tau) \propto \tau^{\nu(d-\varphi/\nu)}$, hence $\theta = \varphi/\nu$. The crossover exponent is related to the scaling of the RF near the fixed point μ^*: h_0^2 increases as $\exp(\ell\varphi/\nu)$. Writing the recursion relation for h_0^2 up to two-loop order, as we have done, Eq.(16), we again find $\varphi/\nu = 2 - \eta + \lambda_\Delta = 2 - \eta_1$.

All our results for critical exponents suggest that $\tilde{\eta} \neq 2\eta$, in agreement with the three-exponent scaling picture [23, 24]. For example, the exponent scaling gives for the ratio $C_s(0)/G^2(0) \propto \xi^{\lambda_\Delta}$, which would diverge unless $2\eta = \tilde{\eta}$ is valid. However, this divergence is too weak to be detected, and thus this ratio may be concerned as a constant, and the concept of no self-averaging in RF systems is expected [25].

We have considered the effects of a RF (field conjugate to the order parameter) on an I-N phase transition using the $\varepsilon = 6 - d$ expansion method. We have found the novel RF fixed point that proceeds from the existence of two relevant variables in the RG approach. The first involves the effects of the RF, while the latter involves those of thermal disorder. These two agents of disorder give comperable contributions to the problem. In the pure nematic, when $h_0 = 0$, the zero cubic term means that the system is located at an isolated Landau point at the phase diagram [20]. This point is unstable with respect to b. When non-zero h_0 is switched on Δ is renormalized toward a fixed point μ^*, and all critical exponents are changed. As we believe, this fixed point governs the critical behavior at the transition from isotropic to the replica symmetric phase, that precedes the replica symmetry breaking phase. The location of this nontrivial random fixed point on a phase diagram is quite close to the fixed point μ_Δ with zero cubic term (we may call this point a random isolated Landau point). This indicates that the critical behavior of the isotropic nematic in RF is like the behavior of the RF Heisenberg model for the five component order parameter. The independent calculation of the critical exponents shows that the dimensional reduction in the hyperscaling relations for the RF isotropic nematic contain the shifted value $d - 2 - \eta + \tilde{\eta}$ instead of d. The so-called "three exponent scaling" appears in the second order in ε. The model for studing the replica symmetry breaking transition from the replica symmetric phase is clearly necessary to perform further investigations.

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