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Quenched disorder and spin-glass correlations in XY nematics

L Petridis and E M Terentjev

Cavendish Laboratory, University of Cambridge, J J Thomson Avenue, Cambridge, CB3 0HE, UK

E-mail: emt1000@cam.ac.uk

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Abstract

We present a theoretical study of the equilibrium ordering in a 3D XY nematic system with quenched random disorder. Within this model, treated with the replica trick and Gaussian variational method, the correlation length is obtained as a function of the local nematic order parameter Q and the effective disorder strength Γ . These results, $\xi \sim Q^2 e^{1/Q^2}$ and $\xi \sim (1/\Gamma) e^{-\Gamma}$, clarify what happens in the limiting cases of diminishing Q and Γ , that is near a phase transition of a pure system. In particular, it is found that quenched disorder is irrelevant as $Q \rightarrow 0$ and hence does not change the character of the continuous XY nematic–isotropic phase transition. We discuss how these results compare with experiments and simulations.

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(Some figures in this article are in colour only in the electronic version)

1. Introduction

Classical nematics possess long-range orientational order as a result of anisotropically biased pair interaction between their long rod-like molecules. Below a certain temperature such systems are in the ordered phase, where the rods are on average aligned with their long axes parallel to each other. Macroscopically this preferred direction is defined by a unit vector called the director, labelled as \mathbf{n} [1]. In this paper we investigate the effects a quenched random field—disorder coupled to the local order parameter—has on the ordering of XY nematics. This is a special sub-class of systems where the orientational ordering direction is confined to a plane (hence the name XY), while the overall system dimensionality may remain the usual 3D (or any other). This is of course fully analogous to the famous XY magnets [2, 3], but with the quadrupolar (nematic) order rather than a dipolar. Physically such systems are

increasingly common in thin planar cells of a liquid crystal, or in new liquid crystalline elastomers made into thin free standing films with the director confined in their plane.

The presence of quenched disorder is implicit in many systems, as different as random ferromagnets [4], vortex lattices in type-II superconductors [5] and randomly crosslinked nematic elastomers [6]. It arises from impurities whose distribution in the sample and the constraints they impose on the local field conformation do not change on the timescale relevant for the evolution of the main ordering.

The influence of quenched disorder is well understood from a theoretical point of view. In the limit of weak disorder¹, it was first shown by Larkin that impurities lead to the destruction of long-range order [5]. This result was then generalized by Imry and Ma who demonstrated that, in less than four dimensions, an arbitrarily weak random field confines the long-range order to hold only at length scales smaller than the ‘domain size’ ξ . The system breaks into uncorrelated regions and therefore loses overall order and alignment [5, 7]. Much work has been done on XY random anisotropy magnets, showing magnetization correlations to decay exponentially [4, 8]: $\langle \mathbf{m}(0) \cdot \mathbf{m}(r) \rangle \sim e^{-r/\xi}$. More sophisticated analysis was done on flux lattices with a two-component order parameter ($N = 2, d = 3$) whose Hamiltonian incorporates an elastic term and a random potential [9]. It shows that correlations in fact decay as a power-law $(r/\xi)^{-1}$, giving rise to quasi-long-range order (QLRO). A functional renormalization group analysis gives a similar behaviour for XY ferromagnets, but also finds that quasi-long-range order is absent in the case of a magnetization field with more than three components [10]. Our work follows a similar line to [9] and also predicts QLRO. However, another non-perturbative functional renormalization group analysis disputes the presence of QLRO in $N = 2, d = 3$, where the system is found to always stay disordered [11]. The authors find that for $N = 2$ the critical dimensionality below which QLRO exists is 3.8.

In many systems with such glassy nature of macroscopic ordering, the experimental observation of key theoretical predictions is difficult and often indirect. The loss of the long-range orientational order is directly seen in equilibrium polydomain nematic elastomers. These materials, formed by crosslinking liquid crystalline polymers in their isotropic phase, have inherent quenched disorder. There are two plausible explanations why disorder is present, related to each other. Residual heterogeneous strains established at the moment of crosslinking give rise to random stresses (a), acting locally on the nematic order [12]. Alternatively (b), the locally anisotropic crosslinks provide randomly oriented anisotropy axes whose disordering effect competes with Frank elasticity, which favours a uniform director configuration [6]. There is also a model essentially treating an ad hoc random temperature in a nematic system [13] but its approach, neglecting the fundamental couplings of nematic ordering to the underlying elastic network, is not relevant for our work. It may be possible that a careful analysis of director correlations could resolve the physical origin of quenched disorder due to the network crosslinks, since computer simulations based on model (a) predict a faster than exponential decay of director correlations [12], whereas simulations based on model (b) find the decay to be exponential [14]. Nevertheless, in both simulations the disorder correlation length has been a rapidly decreasing function of the effective disorder strength. The dependence is exponential and therefore poses the problem that the correlation length does not diverge at the limit of vanishing disorder.

In practice, the polydomain nematic elastomers have the following features. On cooling below the nematic-to-isotropic transition temperature (T_{NI}) a high order parameter phase is formed. Measurement of the order parameter is difficult, with perhaps only NMR a suitable

¹ The empirical criterion of ‘weak disorder’ is when the quenched impurities can be treated as acting on the background of an establishing mean ordering field—as opposed to the strong disorder when one cannot define the usual order parameter in the same way as in a pure system.

technique [15, 16]. There are differing reports in the literature over the years, in all cases finding the transition to be continuous—approaching a critical point in better quality samples. It is well established that deep in the nematic phase the orientation of the nematic director is very non-uniform: \mathbf{n} varies continuously across the sample, pointing roughly along one direction across a very small region of space: following a classical spin-glass pattern which is perhaps misleadingly called a ‘polydomain’ structure. Correlations between director orientations at different points in space decay rapidly and eventually vanish at distances much larger than ξ so that long-range order is eventually lost [17]. This characteristic length scale, ξ , is often called the domain size or correlation length. It is well established to be of the order of microns [18, 19] in many different systems. Therefore, light passing through such a sample is strongly scattered on birefringent regions with a randomly oriented optical axis. As a result, such a sample is completely transparent at high temperatures, but becomes opaque below T_{NI} due to the multiple scattering of the disordered nematic phase. The dependence of ξ on the magnitude of the order parameter (Q) and the effective strength of quenched disorder (Γ) is the main focus this work. The experimental work quoted above was done in thin films leading us to consider directors that are confined to the xy -plane, though being dependent on all three spatial coordinates. This choice would also adequately describe nematic membranes [20, 21].

The structure of the paper is as follows. In section 2 we summarize a physical model of quenched disorder in nematic systems following [17]. Section 3 applies the Gaussian variational method [22] to this problem. We find that the replica symmetry is broken in this system and most of the calculations are performed within the hierarchical replica symmetry breaking framework. In section 4 we solve the variational equations and then obtain ξ as a function of Q and Γ . We also examine how the phase transition is changed by presenting a predicted $Q(T)$ plot. Finally, in section 6, we conclude by discussing our results and comparing them with experiments.

2. Model

2.1. Sources of quenched disorder

In nematic elastomers crosslinked in the isotropic phase the network crosslinks act as sources of quenched orientational disorder. The crosslinks always contain anisotropic groups that provide easy anisotropy axes \mathbf{k} , similar to the sources in random anisotropy magnets [4] (see figure 1). We assume that it is favourable for the local director to align along \mathbf{k} in the vicinity of the crosslink. The orientation of the anisotropy axes and the spatial distribution of the crosslinks are quenched variables. They remain unaltered with time even if external conditions, such as temperature, change.

We follow previous work [17, 23] modelling the local coupling between the nematic order and the random field. For a crosslink positioned at \mathbf{R}_i with an anisotropy axis \mathbf{k}_i a free energy $F_i = -\gamma \mathbf{k}_i \cdot \underline{\underline{Q}} \cdot \mathbf{k}_i$ is attributed to it, where γ is the coupling strength and $Q_{ij} = Q(n_i n_j - \frac{1}{2} \delta_{ij})$ is the XY nematic tensor order parameter at spatial position \mathbf{R}_i . The magnitude Q of the nematic order parameter is different from the Edwards–Anderson order parameter of spin glasses, but serves the purpose: $Q = 1$ refers to a system of locally completely ordered rod-like molecules, whereas $Q = 0$ to a fully isotropic average orientation. Employing a continuum density of impurities $\rho(\mathbf{r}) = \sum_{\mathbf{R}_i} \delta(\mathbf{r} - \mathbf{R}_i)$ and substituting the full expression of Q_{ij} we get the coarse-grained form of random field energy for the whole sample:

$$F_{r.f.} = -\gamma \sum_i \mathbf{k}_i \cdot \underline{\underline{Q}} \cdot \mathbf{k}_i = - \int d^3 \mathbf{r} \gamma Q \rho(\mathbf{r}) (\mathbf{k} \cdot \mathbf{n})^2 + \frac{1}{2} N_{\text{cr}} \gamma Q, \quad (1)$$

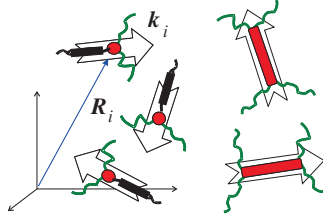


Figure 1. Schematic representation of how crosslinks provide easy anisotropy axes $\{\mathbf{k}\}$. The nematic director is forced to be aligned, in the vicinity of the crosslink, with the axes, which are represented by the arrows. Both the orientation of $\{\mathbf{k}\}$ and the positions of the crosslinks $\{\mathbf{R}_i\}$ are random. Since the crosslinks are confined by the network topology, they add quenched disorder to the nematic system.

where N_{cr} is the total number of impurities (crosslinks). The positive linear term ($N_{\text{cr}}\gamma Q$) is a byproduct of the requirement that Q_{ij} is traceless. It cancels out when random disorder is treated properly in subsection 2.2, and it is dropped for now.

When the (one-constant approximation) Frank elasticity term is included, the full continuum Hamiltonian of the nematic system reads

$$H = \int d^3\mathbf{r} \left[\frac{1}{2} K (\nabla\mathbf{n})^2 - \gamma Q \rho(\mathbf{r})(\mathbf{k} \cdot \mathbf{n})^2 \right]. \quad (2)$$

A simple dimensional argument gives $K \sim k_B T/a$, where a is the size of the mesogenic unit below which the continuum limit of elasticity no longer holds. Both the microscopic and the phenomenological Landau–DeGennes theories of nematic transition give the elastic constant K to scale as Q^2 for $Q \ll 1$. Combining these two estimates, we take that for small Q the elastic constant is approximated by $K \simeq k_B T Q^2/a$. Finally, it is noted that the local magnitude of the order parameter is taken to be constant across the sample and $\nabla Q = 0$, that is, the effect of quenched disorder lies only in the resulting equilibrium director texture. It is assumed that the sample is chemically homogeneous and boundary effects are neglected.

This Hamiltonian has been shown [6] to explain domain formation in nematic elastomers using the classic Imry–Ma argument [7]. Even a weak random field destroys long-range order in length scales greater than the correlation length $\xi_d \sim K^2/\rho_0(\gamma Q)^2$, where in our notation ρ_0 is the number of crosslinks per unit volume. It is a well-known fact that the correlation length of the order parameter fluctuations should diverge close to the phase transition. In direct contrast, when the Imry–Ma argument is applied to nematic elastomers the disorder correlation length tends to zero as the order parameter vanishes ($\xi_d \propto Q^2$) because K is a quadratic functions of Q . In section 4 we are able to clarify this rare shortcoming of the Imry–Ma argument.

2.2. Replica method

We are interested in results averaged over the random distributions of the quenched variables $\rho(\mathbf{r})$ and \mathbf{k} because these variables cannot be controlled experimentally. In other words, one looks for the macroscopic averages over a variety of realizations of $\rho(\mathbf{r})$ and \mathbf{k} . Crosslinking the sample above T_{NI} makes the easy anisotropy axes point at random directions in the XY plane, with an isotropic probability of orientation $P(\mathbf{k}) = \frac{1}{2\pi}$. The crosslinks are dispersed

randomly in the sample with the density $\rho(\mathbf{r})$ and the probability that a particular distribution ρ occurs Gaussian [24]:

$$P[\rho] \sim \exp \left[- \int d^3r \frac{(\Delta\rho)^2}{2\rho_0} \right], \quad (3)$$

where $\Delta\rho = \rho - \rho_0$ is the deviation of local density from ρ_0 , the mean density of crosslinks.

Averaging the logarithm of the partition function Z to obtain the free energy is not algebraically possible, so the replica trick [25] is employed to overcome this difficulty. The expression for the free energy arising from disorder then reads

$$\begin{aligned} F_d &= -k_B T \langle \log Z \rangle_{\rho, \mathbf{k}} = -k_B T \left. \frac{\partial}{\partial m} \langle Z^m \rangle \right|_{m=0} \\ &= -k_B T \left. \frac{\partial}{\partial m} \right|_{m=0} \prod_a^m \int D\mathbf{n}_a \exp[-\beta H_{\text{rep}}], \end{aligned} \quad (4)$$

where we now have m identical ‘replicas’ of the system, labelled by the index a . Equation (4) provides the definition of the replica Hamiltonian, which no longer depends on \mathbf{k} and ρ . After averaging over disorder this Hamiltonian is found to be

$$H_{\text{rep}} \equiv \sum_{a,b=1}^m \int d^3\mathbf{r} \left\{ \frac{1}{2} K (\nabla \mathbf{n}_a)^2 \delta_{ab} - \Gamma (\mathbf{n}_a \cdot \mathbf{n}_b)^2 \right\}, \quad (5)$$

where the subscripts a and b are the replica indices and m is the number of replicas that is set to zero at the end of the calculation. The parameter Γ , arising from completing the Gaussian square with the random-field term in equation (2), reflects the strength of the disorder and has a quadratic dependence on the order parameter magnitude:

$$\Gamma = \frac{\gamma^2 \rho_0 Q^2}{8k_B T}. \quad (6)$$

It is noted that all replicas are assumed to have equal disorder strength and equal magnitude of the order parameter; that is, $\gamma_a = \gamma_b$ and $Q_a = Q_b$ for all a and b .

The simplest way to ensure that the director is a unit vector confined in a plane is to parameterize it by a single angle θ : $\mathbf{n} = \{\cos \theta(r), \sin \theta(r)\}$. In this notation the replica Hamiltonian, now a functional of θ , reads

$$H_{\text{rep}}[\theta(r)] = \sum_{a,b}^m \int d^3\mathbf{r} \left[\frac{K}{2} (\nabla \theta_a)^2 \delta_{ab} - \frac{\Gamma}{2} \cos 2(\theta_a - \theta_b) \right], \quad (7)$$

where the irrelevant constant term $-\Gamma V/2$ arising from the conversion of $\cos^2 \theta$ to $\cos 2\theta$ is dropped.

3. Gaussian variational method

The cosine in the random field term of equation (7) poses an obstacle to the development of the model. The correlation function and the free energy can be obtained from equation (4) as long as the replica Hamiltonian is quadratic in θ . This is clearly not the case in our model and to overcome this obstacle we employ the Gaussian variational method (GVM), first used in the context of random manifolds by Mezard and Parisi [22].

3.1. GVM in nematic systems

A model Hamiltonian H_0 is assumed to describe the system, meaning that all its physical properties, such as correlations and free energy, are fully defined by $H_0[\theta]$ instead of the original $H_{\text{rep}}[\theta]$. It is essential that H_0 is quadratic in the fields $\theta(\mathbf{q})$, in Fourier space:

$$\beta H_0 = \frac{1}{2} \sum_{a,b}^m \int \frac{d^3q}{(2\pi)^3} \theta_a(q) G_{ab}^{-1}(q) \theta_b(q). \quad (8)$$

Here and throughout the paper we use the shorthand notation $\beta = 1/k_B T$ that makes all energy functions non-dimensional. The (unknown) propagator can be written as

$$G_{ab}^{-1} = \beta K q^2 \delta_{ab} - \sigma_{ab}, \quad (9)$$

where $K q^2 \delta_{ab}$ is chosen to match the elastic contribution in H_{rep} , and σ_{ab} is a set of yet undetermined parameters approximating the random field effects. The true free energy of the system can be cast as an expansion in $\langle H_{\text{rep}} - H_0 \rangle$, assumed small:

$$F \approx F_0 + \langle H_{\text{rep}} - H_0 \rangle_{H_0}, \quad (10)$$

where $F_0 = -k_B T (\det G)^{1/2}$ and the angular brackets indicate averaging with the Gaussian $e^{-\beta H_0}$ as measure. The ultimate aim is to minimize $F[G]$ with respect to variations in G_{ab} and obtain the best upper bound for F . Using equation (8), the average of equation (10) leaves the variational energy density:

$$\beta F_{\text{var}} = \frac{1}{2} \int_q \left[-\text{Tr} \log \mathbf{G} + \sum_a^m \beta K q^2 G_{aa} \right] - \frac{1}{2} \sum_{a,b}^m \beta \Gamma \exp \left[-2 \int_q (G_{aa} - 2G_{ab} + G_{bb}) \right],$$

where the notation \int_q is a shorthand for the full $\int d^3q/(2\pi)^3$ and the irrelevant constant $\langle H_0 \rangle = -k_B T/2$ has been dropped. Before proceeding with the minimization it is convenient to define a new function that appears regularly in the random field term:

$$B_{ab} = \int_q (G_{aa} - 2G_{ab} + G_{bb}). \quad (11)$$

It is related to the correlations between replicas since $\langle [\theta_a(r) - \theta_b(r)]^2 \rangle = B_{ab}$.

Demanding F_{var} to be stationary with respect to variations on G_{ab} provides a relation that determines σ_{ab} .

$$\frac{\delta \beta F_{\text{var}}}{\delta G_{cd}} = 0 \quad \Rightarrow \quad G_{ab}^{-1} = \beta K q^2 \delta_{ab} - 4\beta \Gamma \left(e^{-2B_{ab}} - \delta_{ab} \sum_c e^{-2B_{ac}} \right). \quad (12)$$

Comparing this expression with F_{var} , it is easily seen that the stationary equation for the matrix of parameters σ takes the form

$$\sigma_{ab} = 4\beta \Gamma \left[e^{-2B_{ab}} - \delta_{ab} \sum_c e^{-2B_{ac}} \right]. \quad (13)$$

The above equation has a reassuring property—that σ is zero when there is no disorder in the system ($\Gamma = 0$). After all, σ_{ab} was introduced to approximate the random field imposed by the crosslinks.

It is convenient to decompose the equation into two separate conditions [22]—one for the off-diagonal and the separate for the purely diagonal part of σ_{ab} . The off-diagonal part is easily obtained by setting the Kronecker delta to zero in equation (13):

$$\sigma_{a \neq b} = 4\beta \Gamma V e^{-2B_{ab}}. \quad (14)$$

This is a transcendental equation because B_{ab} is itself a function of G_{ab} and therefore of σ_{ab} as well. To be able to solve it a certain form of σ_{ab} must be assumed so that G_{ab}^{-1} can be inverted to obtain B_{ab} . The diagonal part is fixed by noting that summation over the free index a in equation (13) is zero because $B_{ab} = B_{ba}$. Hence after finding the off-diagonal elements the condition

$$\sum_{a=1}^m \sigma_{ab} = 0 \tag{15}$$

provides a way to determine the diagonal matrix element σ_{aa} .

3.2. Replica symmetry

In a first attempt to determine the variational parameter σ_{ab} from the stationary equation, let us assume all the non-diagonal elements to be equal to $\sigma_{a \neq b} = \sigma$, a constant which will be determined by equation (14). Using equation (15) we then find the diagonal part to be $\sigma_{aa} = (1 - m)\sigma$. This scheme based on the constant- σ assumption is called the replica symmetry (RS) limit. Under this assumption, the model Hamiltonian acquires the simple form

$$G_{ab}^{-1}(q) = (\bar{K}q^2 + m\sigma)\delta_{ab} - \sigma \mathbb{1}_{ab}, \tag{16}$$

where $\bar{K} = \beta K$ and $\mathbb{1}_{ab}$ is the matrix with all its elements equal to one. The matrix can be trivially inverted:

$$G_{ab} = \frac{1}{\bar{K}q^2} \delta_{ab} + \frac{\sigma}{\bar{K}^2 q^4} \mathbb{1}_{ab}. \tag{17}$$

The stationary equation (14) is easily solved because B_{ab} does not depend on σ . This is a unique feature of RS in this problem. We shall see later that less symmetric forms of σ_{ab} generate a transcendental equation more difficult to deal with. The replica symmetric solution is

$$\sigma = 4\beta\Gamma \exp\left(-4 \int_q \frac{1}{\bar{K}q^2}\right) = 4\beta\Gamma \exp\left(-\frac{2q_{\max}}{\pi^2 \beta \bar{K}}\right), \tag{18}$$

where $q_{\max} = 2\pi/a$ is the ultraviolet cut-off in momentum space. If we now use the earlier estimate for the Frank elastic constant, $K \sim k_B T Q^2/a$, the solution becomes

$$\sigma = 4\beta\Gamma V \exp\left(-\frac{4}{\pi Q^2}\right) \propto Q^2 \exp[-4/\pi Q^2], \tag{19}$$

where the quadratic order parameter dependence of Γ has been also substituted. This function has a singular dependence on the order parameter, dropping to zero as $Q \rightarrow 0$. As a result, the disorder is irrelevant close to the nematic–isotropic transition.

A necessary condition for the RS solution to be applicable is that the disorder energy must be stable for infinitesimal variations from that solution. The stability analysis was first introduced in the context of spin glasses [26] and later applied to the GVM [22]. First, the variational parameter σ_{ab} is allowed to vary $\sigma_{ab} = \sigma_{ab}^{RS} + \epsilon_{ab}$, with the replica symmetric ansatz labelled as σ^{RS} and ϵ_{ab} the infinitesimal deviation from it. $F_{\text{var}}(\sigma)$ is then expanded to second order in ϵ_{ab} , naturally, without the linear term, represented by the zero in equation (12):

$$\tilde{F}_{\text{var}} = F_{\text{var}}(\epsilon = 0) + \frac{1}{2} \int_q \sum_{a,b,c,d} H_{ab,dc}(q) \epsilon_{ab} \epsilon_{cd}.$$

The term second-order in ϵ_{ab} involves the four-dimensional tensor of coefficients $H_{ab,dc}$, which is called Hessian and plays a vital role in determining the stability of this solution: for RS to

be stable, this term must be positive definite for arbitrary ϵ . This is ensured as long as all the eigenvalues of the Hessian are non-negative. In our XY system there are only two non-trivial eigenvalues. One of them, the so-called replicon eigenvalue, diverges to negative infinite [9, 22]:

$$\lambda = 1 - \sigma \int \frac{d^3q}{(2\pi)^2} \frac{1}{(\bar{K}q^2)^2}.$$

Therefore the replica symmetric solution does not give an established free energy minimum and is not appropriate.

3.3. Hierarchical RSB

The stability analysis of the RS solution as well as much previous work on disorder systems invites us to look for the hierarchical replica symmetry breaking solutions [27]. This is known to be the real equilibrium state (i.e. has both Hessian eigenvalues positive definite) of spin glasses [28] and random manifolds [29]. It is reasonable to expect a similar behaviour in our model and therefore the hierarchical method was chosen to explore replica symmetry breaking.

In this model the matrix σ_{ab} is assumed to have a nested block-diagonal form with off- and on-diagonal parts map to $\sigma(v)$ and $\tilde{\sigma}$ respectively, with a continuous index variable $v \in [0, 1]$. As a result, G_{ab}^{-1} and its inverse also have this hierarchical form and the quantity of interest when solving the stationary equation (14) is now written as $B_{ab} = 2 \int_q [\tilde{g} - g(v)]$, where

$$\tilde{g} - g(v) = \frac{1}{\bar{K}q^2 - \tilde{\sigma} + \sigma(1)} + \int_v^1 du \sigma'(u) \left(\frac{1}{\bar{K}q^2 - \tilde{\sigma} + \langle \sigma \rangle + [\sigma](v)} \right)^2, \quad (20)$$

where the shorthand notations $\sigma' = d\sigma/dv$ and $[\sigma](v) \equiv v\sigma(v) - \int_0^v du \sigma(u)$ are used after Parisi [27]. Finally, the condition on the diagonal part of σ in equation (15) becomes

$$\tilde{\sigma} = \int_0^1 dv \sigma(v) = \langle \sigma \rangle. \quad (21)$$

It should always be kept in mind that the variable v determines which diagonal block an element σ_{ab} belongs to. The smaller it is, the closer the element is positioned to the diagonal. More interestingly, we shall see later that the long-range behaviour of the system is specifically associated with these small values of v .

The original stationary equation is written in terms of hierarchical matrices as

$$\sigma(v) = 4\beta\Gamma V \exp \left\{ -4 \int_q [\tilde{g}(q) - g(v)] \right\}. \quad (22)$$

Substituting equations (20) in (22),

$$\sigma(v) = 4\beta\Gamma V \exp \left\{ -4 \int_q \left[\frac{1}{\bar{K}q^2 - \tilde{\sigma} + \sigma(1)} + \int_v^1 \frac{\sigma'(u) du}{[\bar{K}q^2 - \tilde{\sigma} + \langle \sigma \rangle + [\sigma](u)]^2} \right] \right\}. \quad (23)$$

The parameter v appears only on the lower limit of the u -integral in equation (23). As a result, the stationary equation is greatly simplified by differentiating it with respect to v :

$$\sigma'(v) = 4\sigma(v) \int_q \frac{\sigma'(v)}{[\bar{K}q^2 + [\sigma](v)]^2}, \quad (24)$$

where it is noted that equation (22) shows that $\sigma(v)$ has no q -dependence. This stationary equation for $\sigma(v)$ has two solutions. The first one is obvious: $\sigma'(v) = 0$. Had this been the

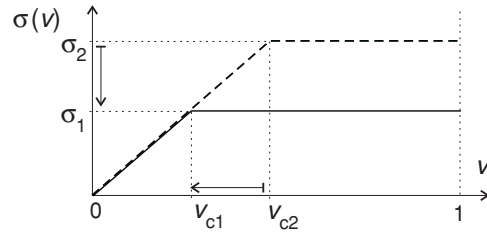


Figure 2. The function $\sigma(v)$, plotted for two different disorder strengths (dashed line for higher Γ). The weakening disorder decreases both the crossover point v_c and the plateau value $\sigma(v_c)$, but leaves the slope of the function unchanged.

real relevant solution, the unique RS would have been the only possibility (the parameter σ does not depend on v). However, a second solution also exists for $\sigma'(v) \neq 0$, and it is given by

$$1 = 4\sigma(v) \int \frac{dq}{(2\pi)^2} \frac{q^2}{[\bar{K}q^2 + [\sigma](v)]^2} \Rightarrow \sigma(v) = 2\pi \bar{K}^{3/2} [\sigma]^{1/2}. \quad (25)$$

Differentiating equation (25) w.r.t. v and observing that $[\sigma]'(v) = v\sigma'(v)$ gives the second solution for σ :

$$[\sigma](v) = \pi^2 \bar{K}^3 v^2 \Rightarrow \sigma(v) = 2\pi^2 \bar{K}^3 v. \quad (26)$$

The linear dependence on v is an interesting feature. It implies that the effect of disorder—a measure of which is the value of σ —is smaller for small v (that is, further away from the diagonal in the discrete form of σ_{ab}). As we will see in the next section, the large-distance director correlations are controlled by the small- v solution. Therefore RSB predicts that disorder has a milder effect in large-distance correlations, in marked contrast to the RS solution where σ has a constant value.

The full RSB form of $\sigma(v)$ incorporates both the linear and the constant ($\sigma' = 0$) solutions. In fact there is a crossover between the two at a value v_c , where $\sigma(v)$ changes from one regime to the other,

$$\sigma(v) = \begin{cases} 2\pi \bar{K}^3 v & \text{for } v \in [0, v_c] \\ 2\pi \bar{K}^3 v_c = \text{const} & \text{for } v \in [v_c, 1], \end{cases} \quad (27)$$

as sketched in figure 2. This form of branched function appears in spin glasses [28] as well as other disorder systems where the GVM has been used, such as random manifolds [22] and flux lattices [9].

The reason that both solutions are accepted is illustrated in figure 2. As we shall see shortly, in equation (28), v_c reduces to zero in the absence of disorder ($\Gamma = 0$). This in turn means that $\sigma(v)$ is identically zero for the whole range of $v \in [0, 1]$. The no-disorder limit is an essential part of an acceptable solution for $\sigma(v)$ and would be absent if only the linear branch of equation (27) was accepted.

The only undetermined parameter is the crossover boundary v_c , to which we turn our attention to. Setting v equal to v_c in equations (22) and (27) and observing that they are equal, we find

$$\sigma(v_c) = 2\pi \bar{K}^3 v_c = \frac{4\Gamma}{k_B T} \exp \left[-\frac{2q_{\max}}{\pi^2 \beta K} + \frac{2v_c}{\pi} \tan^{-1} \left(\frac{q_{\max}}{\pi \beta K v_c} \right) \right].$$

The term linear in the ultraviolet momentum cut-off (short-distance, $q_{\max} = 2\pi/a$) is identical to equation (18) encountered in the replica symmetry limit. Since $\beta K \sim Q^2/a$, this term does not diverge. The same applies for the argument of the arctangent, which is also approximated by $2/(Q^2 v_c)$, after which the equation determining v_c becomes

$$v_c \simeq \frac{2(k_B T)^2 \Gamma}{\pi K^3} \exp \left[-\frac{4}{\pi Q^2} + \frac{2v_c}{\pi} \tan^{-1} \left(\frac{2}{v_c Q^2} \right) \right]. \quad (28)$$

The approximately equal sign refers to the use of $K \sim Q^2 k_B T/a$.

4. Results and discussion

4.1. Director correlations

This section discusses how to obtain the domain size as a function of order parameter and disorder strength. To define the correlation length we examine how the director correlations decay. As a result of the use of a single angle to parameterize \mathbf{n} , director correlations are given by [8, 17]

$$C(r) = \langle \mathbf{n}(r) \cdot \mathbf{n}(0) \rangle = \langle \cos[\theta(r) - \theta(0)] \rangle \propto e^{-B(r)/2},$$

where $B(r) = 2 \int_q G_{aa} [1 - \cos(qr)]$.

In RSB G_{aa} is replaced by (see [22] for detail)

$$\begin{aligned} \tilde{g} &= \frac{1}{\bar{K} q^2} \left(1 + \int_0^1 \frac{dv}{v^2} \frac{[\sigma](v)}{\bar{K} q^2 + [\sigma](v)} \right) \\ &= \frac{1}{\bar{K} q^2} \left[1 + \frac{\pi \bar{K}}{q} \tan^{-1} \left(\frac{\pi \bar{K} v_c}{q} \right) + \left(\frac{1}{v_c} - 1 \right) \frac{[\sigma](v_c)}{\bar{K} q^2 + [\sigma](v_c)} \right]. \end{aligned} \quad (29)$$

The $1/\bar{K} q^2$ term which dominates at large q originates from thermal fluctuations. The remaining terms, which dominate at small q (large distances), show the effect of disorder.

In order to determine the correlations, $B(r)$ must be calculated using the random field contribution of equation (29). The q -integration is rather involved and yields

$$\begin{aligned} B(r) &= \frac{1}{2} \left\{ \text{Euler}\Gamma + SI \left(\frac{r}{\xi} \right) - CI \left(\frac{r}{\xi} \right) + \log \left(\frac{r}{\xi} \right) \right. \\ &\quad \left. - 2(1 - v_c) \sinh \left(\frac{r}{2\xi} \right) \left[\sinh \left(\frac{r}{2\xi} \right) - \cosh \left(\frac{r}{2\xi} \right) \right] \right\}, \end{aligned} \quad (30)$$

where $\xi = \pi K v_c$.

Euler $\Gamma \approx 0.58$ refers to the Euler constant, while ‘SI’ and ‘CI’ are shorthand notations for the SinhIntegral and CoshIntegral special functions, respectively. Their behaviour for small and large arguments is given by

$$\begin{aligned} SI(x) &= x + \mathcal{O}(x^3) \\ CI(x) &= \text{Euler}\Gamma + \log x + \mathcal{O}(x^2); \quad \text{and} \quad CI(\infty) = SI(\infty). \end{aligned}$$

Director correlations decay differently depending how r compares with the length scale ξ , which is identified as the correlation length or domain size:

$$\langle \mathbf{n}(0) \cdot \mathbf{n}(r) \rangle \sim \begin{cases} \exp(-r/\xi) & \text{for } r \ll \xi \\ (r/\xi)^{-1} & \text{for } r \gg \xi. \end{cases} \quad (31)$$

The power-law dependence was first derived by Giamarchi and LeDoussal [9] and is reproduced in our work. It shows that alignment order persists over greater length scales than one might first guess from the Larkin argument and the work on random anisotropy magnets [4, 5]. One also associates the exponential decay at short distances of equation (31), which is also found in the RS case, with the constant part of $\sigma(v)$ and the algebraic decay with the linear part. In this context the small- v solution can be taken to correspond to large distance correlations.

An exact analytic solution of v_c in equation (28) is clearly impossible; however, its dependence on the order parameter Q and disorder strength Γ can be deduced. It is then straightforward to find $\xi = \pi K v_c$ as a function of these two parameters, which is done in the next two sections.

4.2. Domain size as a function of order parameter

In this section we present the first theoretical model that predicts successfully the behaviour of the domain size close to the phase transition. As discussed in the introduction, the Imry–Ma argument gives $\xi \sim K^2/\Gamma \propto Q^2$. This expression cannot hold close to the nematic-to-isotropic transition since we expect that ξ should diverge as $Q \rightarrow 0$. Experiments measuring $\xi(Q)$ directly do not exist for nematic elastomers: it is practically impossible to measure simultaneously the order parameter and domain size of such systems. However, light scattering experiments measured $\xi(T)$ the domain size as a function of temperature [18]. Separately the order parameter of a *monodomain* elastomer was also measured as a function of temperature [30]. Assuming that the polydomain and monodomain samples have the same form of $Q(T)$, the data are combined with $\xi(T)$ to make a parametric plot of $\xi(T)$ versus $Q(T)$ in figure 3. As expected the domain size increases at small Q .

To determine $\xi(Q)$ for small order parameter we must first find the functional dependence of v_c on Q . From their definition both v_c and Q are smaller than one. Therefore the argument of the arctangent in equation (28) is large and $\tan^{-1} \rightarrow \pi/2$. Hence

$$v_c \approx \frac{2\Gamma}{\pi\beta^2 K^3} \exp\left(-\frac{4}{\pi Q^2} + v_c\right). \quad (32)$$

When $Q \ll 1$ the factor of $\exp(-4/\pi Q^2)$ ensures that v_c is also much smaller than one. Therefore $v_c \ll Q^{-2}$ and the term v_c in the exponent is negligible. Taking into account that both Γ and K are proportional to Q^2 for small Q , one finds

$$\xi = \pi K v_c \propto Q^2 e^{\frac{4}{\pi Q^2}}. \quad (33)$$

As we shall see later, for real elastomers $v_c \approx 10^{-4}$ even for $Q = 1$, so taking the arctangent equal to $\pi/2$ is a very good approximation.

The solid line in figure 3 shows a fit to the model expression (33). The agreement between theory and experiments is good for small Q , but diminishes for $Q \geq 0.3$. This is attributed to the fact that K is proportional to Q^2 only for $Q \ll 1$; therefore the pre-exponential factor of Q^2 arising from K^2/Γ is strictly valid only for small Q . A similar indirect study of $\xi(Q)$ was fitted with a power law $\xi \propto Q^{-2}$ [19]. The Imry–Ma correlation length was considered, forcing the authors to suggest that $\Gamma \propto Q^6$, which is hard to justify. It is not reasonable to try to differentiate the two fits, since the data do not come from direct measurements. However, our present model is theoretically consistent since it assumes $\Gamma \propto Q^2$ in accordance with the idea of crosslinks providing a random field that couples to the local order parameter, as seen in equation (1).

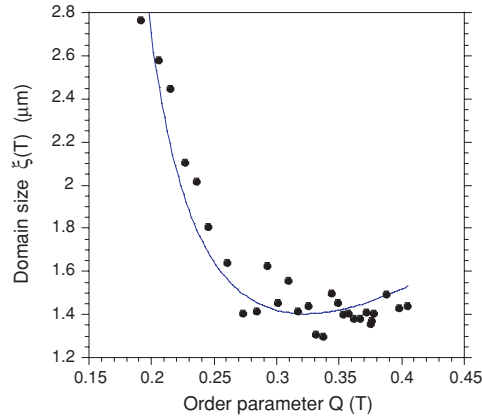


Figure 3. Combined results of two different data sets— $\xi(T)$ [18] and $Q(T)$ [30]—to form a parametric graph of $\xi(Q)$. The fit (solid line) comes from equation (33) and is good for small order parameter where the theoretical prediction holds.

4.3. Domain size as a function of disorder strength

Lattice Monte Carlo simulations have been performed to study the equilibrium ordering in a two-dimensional nematic system with quenched random disorder [14]. The long-range correlation of the director orientation was found to decay as a simple exponential. However, there is a flaw with the simple exponential decay, since it does not predict the ‘domain size’ to diverge as $\Gamma \rightarrow 0$, as one must expect. The correlation length ξ itself also decays exponentially with increasing strength of the disordering field. The weak disorder region was never probed because the amount of computation needed to produce reliable results increased sharply as Γ was reduced.

In order to obtain ξ as a function of Γ we must first find $v_c(\Gamma)$. Going back to equation (28), let us rename the dimensionless prefactor as

$$D = \frac{2(k_B T)^2 \Gamma}{\pi K^3}. \quad (34)$$

This is a measure of the relative disorder strength. To get an order of magnitude estimate for D in nematic elastomers, we consider, following the detailed experimental comparison in [6], $\gamma \sim 0.4 \times k_B T$, $\rho_0 \sim 2.5 \times 10^{26} \text{ m}^{-3}$ and $K \sim 10^{-12} \text{ Jm}^{-1}$, which gives D of the order of 10^{-4} . This means that for constant order parameter $Q = 1$, v_c is small and the arctangent in (28) is equal to $\pi/2$ with good precision.

To obtain a theoretical prediction of $v_c(D)$ let us use a self-consistent method in equation (28). At $Q = 1$ we can write

$$v_c = D \exp(-4/\pi + v_c) \approx 0.3D e^{v_c}. \quad (35)$$

In the first approximation the right-hand side dependence on v_c is dropped, leaving $v_c \approx 0.3D$. This value of v_c is placed back in the exponent of (35) to give the estimate

$$v_c \simeq 0.3D e^{0.3D}. \quad (36)$$

A numerical solution of equation (28) can easily be found without the assumption $v_c \ll 1$ and the subsequent approximation of the arctangent term. As seen in figure 4, the agreement of the estimate above with the numerical solution is very good for small values of D , which real

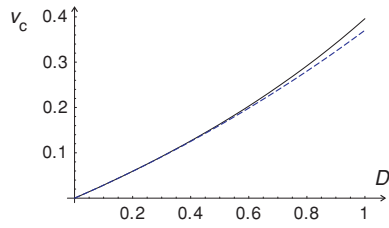


Figure 4. Numerical evaluation of $v_c(D)$ from equation (28) for fixed order parameter is plotted as a solid line. The dotted line shows the analytic approximation of equation (36). The agreement between the two curves is so good that one can hardly distinguish between them in the limit $D \rightarrow 0$.

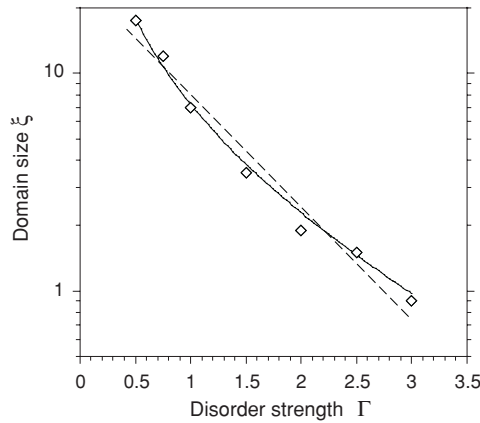


Figure 5. The log-linear dependence of the domain size on disorder strength. The data points are taken from the simulation work [14]. The dashed line is the fit by a simple exponential, provided by the authors of the original simulation work. The solid line shows the fit with our model expression (37): $(9.8/\Gamma) e^{-0.45\Gamma}$ —a clearly better fit.

systems are expected to have. An important feature of this plot is that it starts from the origin. When there is no disorder in the system ($D = 0$), $\sigma(v)$ is zero for all v because $v_c = 0$.

The domain size has a more complex functional dependence on disorder strength than a simple exponential. It is inversely proportional to v_c ; therefore, since $D \propto \Gamma$,

$$\xi(\Gamma) \propto \frac{K^2}{\Gamma} e^{-\alpha\Gamma}, \tag{37}$$

where $\alpha \simeq 0.2(\beta^2 K^3)^{-1}$. The computationally obtained $\xi(\Gamma)$ data points of [14] were fitted with the function of equation (37). The resulting fit (figure 5) is an improvement of the simple exponential function and offers, for the first time, an analytical explanation for the exponential decay found by Yu *et al* [14]. Importantly the $(1/\Gamma) e^{-\alpha\Gamma}$ function also predicts the divergence of the domain size at vanishing disorder—an important improvement of the previous work.

Nematic elastomers can potentially provide a physical system where the spin-glass theory can be extensively tested experimentally. The parameter $\Gamma \sim \gamma^2 \rho_0$ can be, in principle, controlled by changing the density or the molecular nature of network crosslinks, an important factor in determining many physical properties of elastomers.

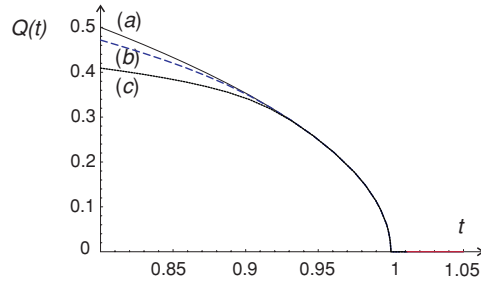


Figure 6. Plots of order parameter against reduced temperature $t = T/T^*$ that qualitatively show the effect of disorder. Plot (a) is obtained from the Landau–DeGennes theory in the absence of disorder, whereas curves (b) and (c) show the effect of increasing disorder.

4.4. Nematic–isotropic phase transition

The last question we address here is whether the presence of quenched disorder changes the character of the nematic–isotropic phase transition. In the case of XY nematics whose director is confined in a plane, the transition is second order because the cubic term in the Landau–DeGennes expansion of the free energy density is identically zero: $\text{Tr}(\underline{Q}^3) = 0$. We calculate the energy density arising from disorder as a function of order parameter using the model Hamiltonian of equation (9):

$$\beta F_0 = \frac{\partial}{\partial m} \int \mathcal{D}\theta \exp \left(-\frac{1}{2} \sum_{ab} \sum_q G_{ab}^{-1} \theta_a \theta_b \right) \Big|_{m=0} \propto \frac{\partial}{\partial m} \exp \left(-\frac{1}{2} \sum_q \text{Tr} \log G^{-1} \right) \Big|_{m=0}. \quad (38)$$

The aim is to add F_0 to the Landau–DeGennes expression to see if there is any significant change on the phase transition. In the RSB matrix algebra the trace of a logarithm is given by [22]

$$\frac{1}{m} \text{Tr} \log \mathbf{G}^{-1} = \log(\bar{K} q^2) - \frac{\sigma(0)}{\bar{K} q^2} + \int_0^1 \frac{dv}{v^2} \log \left(\frac{\bar{K} q^2}{\bar{K} q^2 + [\sigma](v)} \right). \quad (39)$$

Substituting $\sigma(v)$ and integrating over q ,

$$\begin{aligned} \frac{1}{m} \int \frac{4\pi q^2 dq}{(2\pi)^3} \text{Tr} \log \mathbf{G}^{-1} &= \bar{K}^2 \left(\frac{v_c^2}{3} - \frac{v_c}{2} \right) \left[\pi \bar{K} v_c \tan^{-1} \left(\frac{\pi \bar{K} v_c}{q_{\max}} \right) + q_{\max} \right] \\ &+ \frac{1}{2\pi} \bar{K} q_{\max}^2 \tan^{-1} \left(\frac{\pi \bar{K} v_c}{q_{\max}} \right) + \frac{q_{\max}^3}{6\pi^2} \log \left(1 + \frac{\pi^2 \bar{K}^2 v_c^2}{q_{\max}^2} \right). \end{aligned} \quad (40)$$

One way to simplify the above expression is to recall that $v_c \beta K / q_{\max} \ll 1$ and therefore the arctangent and logarithmic terms can be expanded in a Taylor series and thus obtain a Q -dependent expression. However, due to the singular dependence of v_c on the order parameter (e^{-1/Q^2}), all the terms of equation (40) vanish close to the critical point of this continuous transition. Hence the disorder energy is zero as $Q \rightarrow 0$ and the transition remains second order, with unchanged critical behaviour. This is in contrast to the case of 3D-director nematics where the addition of disorder changes the phase transition from first to second order. The overall weak effect of disorder on the average nematic order in the system is sketched in figure 6. Nematic order is weakened away from the transition, but overall there are no significant changes.

5. Conclusion

In this paper we have developed a quantitative description for the ordering of nematics with quenched disorder, following the now classical work on replica symmetry breaking in spin glasses. Our model is focused on a particular case where the director is confined in a plane (corresponding to the 3D XY model in magnets). Such nematic configuration occurs in many experimental arrangements where thin films are used, and is especially relevant for nematic elastomers. Quenched impurities (chemical crosslinks in the case of elastomers) act as sources of disorder by providing easy anisotropy axes. The competition between Frank elasticity and these random sources leads to the loss of long-range orientational order: the mesogenic units (rod-like molecules) are assumed to be locally well ordered with the magnitude of the order parameter Q uniform throughout the system, but the orientation of the director varies on a length scale ξ , called the domain size. We have extracted the dependence of ξ on Q and Γ , a measure of the strength of disorder. In addition, we have checked what effects the addition of disorder has on the nematic–isotropic phase transition in such XY nematics.

The functional forms of $\xi(Q)$ and $\xi(\Gamma)$ adequately describe the evolution of the domain size, reported by experiments and simulations. In particular, $\xi(Q) \sim (K^2/\Gamma) e^{2q_{\max} k_B T/\pi^2 K} \propto Q^2 e^{4/\pi Q^2}$ provides a consistent account of the divergence of ξ as the phase transition is approached. For $Q \geq 0.3$, where the exponential function varies much more smoothly, the Imry–Ma estimate ($\xi_d \sim K^2/\Gamma$) is recovered. When disorder is not strong, we found the domain size to diverge as $\xi(\Gamma) \sim (K^2/\Gamma) e^{-\alpha\Gamma}$. This form combines the exponential decay found in some simulations with the Imry–Ma result. It is possible that this behaviour can be verified experimentally by measuring ξ for nematic elastomers with different crosslink density.

Quenched random-anisotropy effects were found to become irrelevant as the continuous phase transition of the XY nematic is approached. The parameter σ that models the random field within the GVM is identically zero as Q approaches zero. For this reason, the addition of impurities only affects the state of local order, the $Q(T)$ plot, well below the transition temperature where $Q \geq 0.3$. In a separate paper, we consider that nematics with 3D director conformation (analogous to the Heisenberg magnet) and find that they behave in a completely different way: the presence of disorder transforms the first-order transition of pure systems to a continuous one.

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