Topology and Nematic Ordering

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We consider the weakly first-order phase transition between the isotropic and ordered phases of nematics in terms of the behavior of topological line defects. Specifically, we present analytical and Monte Carlo results for a new coarse-grained theory of nematics which incorporates the inversion symmetry of nematics as a local gauge invariance. Increasing the disclination core energy makes the nematic-isotropic transition more weakly first order, and eventually splits it into two continuous transitions which involve the unbinding and condensation of defects, respectively. We find a novel isotropic phase with topological order.

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Both magnetic and nematic media have orientationally ordered phases, which are described by a vector and a "director" (headless vector), respectively. Yet while the order-disorder transition in magnets is generally continuous, the observed nematic-isotropic transition is *discontinuous*. The conventional explanation of this difference appeals to Landau theory and symmetry [1]. For nematics, the order parameter is a traceless symmetric tensor $Q_{\alpha\beta}$ which specifies the local distribution of molecular orientations. The Landau free energy then contains a third-order term proportional to $\mathrm{Tr}Q^3$, which distinguishes between molecular ordering parallel to ($\mathrm{Tr}Q^3 > 0$) or perpendicular to ($\mathrm{Tr}Q^3 < 0$) a common axis. For magnets the order parameter is a vector—the magnetization—and a third-order term is forbidden by symmetry. The presence of a cubic term in the Landau free energy implies, according to Landau theory, a generically strong first-order nematic-isotropic phase transition. We will show, contrary to this conventional wisdom, that (a) the nematic-isotropic transition is not *necessarily* first order (and can in principle share the universality class of the ferromagnetic-paramagnetic transition), and (b) a new, topologically ordered phase of nematics is possible.

The nematic-isotropic transition is observed to be only *weakly* first order [1]—the correlation length at the transition is typically an order of magnitude greater than the molecular size, and the difference between the spinodal and transition temperatures is small. In Landau theory this implies an anomalously small coefficient for the third-order term. This would imply the existence of a *biaxial* nematic phase at slightly lower temperatures, which is not seen experimentally [2].

We propose a new picture of the nematic transition which focuses on disclinations—topological line defects which are found in nematics but not in magnetic systems [3]. A disclination is a curve (Fig. 1) whose encirclement produces a 180° rotation of the local molecular axis (director). Since nematogens have inversion symmetry, the director is continuous outside the defect core. We find that by adding an explicit core energy [4] for disclinations, the first-order nematic-isotropic transition can be made arbitrarily weak. Eventually, the transition splits into a pair of *continuous* transitions with a novel topologically ordered intermediate phase, as shown in Fig. 2. From this point of view the observed weakness of the nematic-isotropic transition follows from the similarity of magnets and nematics when defects are suppressed.

Our work is motivated by other phase transitions that

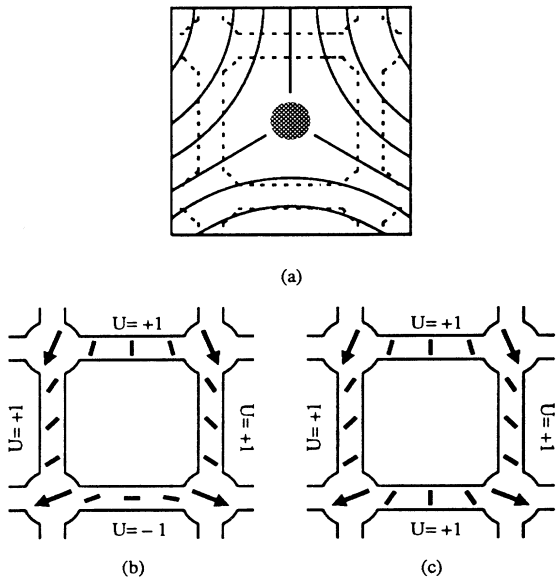

FIG. 1. (a) A cross section through a disclination. When the defect is encircled, the local molecular axis rotates through 180°. On a lattice the defect can occur inside a plaquette. The presence (b) or absence (c) of a defect depends on the product of the U_{ij} around the plaquette.

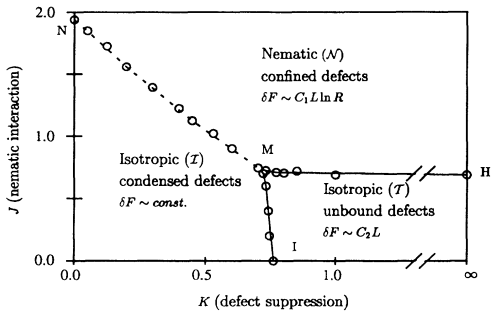

FIG. 2. Phase diagram for the lattice gauge theory Eq. (3). The phase boundaries are very straight except near their intersection. The dotted line denotes a first-order transition and the unbroken lines continuous transitions.

have been understood as a proliferation of topological defects. The best-known example is the two-dimensional XY model; point defects (vortices) unbind at the transition in accordance with the scenario of Kosterlitz and Thouless [5]. Similarly, the three-dimensional XY model can be exactly mapped onto a system of interacting vortex loops, whose unbinding accompanies the transition [6,7]. Unbinding of point defects [8] has also been proposed as a mechanism for the disordering of the three-dimensional Heisenberg model. It remains unclear how the description in terms of defects relates to the Landau-Ginzburg-Wilson theory of these last two cases [9].

Before we present an explicit calculation, let us discuss the general features of the phase diagram for our lattice gauge theory of nematics, shown in Fig. 2. The horizontal axis specifies the bare defect core energy K, while the vertical axis corresponds to the microscopic nematic interaction J. To distinguish the three phases of Fig. 2, let us compare the free energies of a nematic fluid confined to a cylinder of height L and radius R with two different boundary conditions. The first allows no disclinations to pierce the boundary, while the second constrains a single disclination to pass through the centers of the upper and lower faces of the cylinder, but allows no other defect lines to cross the surface.

In the nematically ordered phase $\mathcal{N}$, the free energy difference between the two boundary conditions varies as

$$\delta F_{\mathcal{N}} \sim C_1 L \ln R , \tag{1}$$

with C_1 given by the (long wavelength) nematic stiffness. Similarly, there will be a logarithmic potential between a pair of externally imposed defects. Thus, spontaneous defect loops are small and sparse; those with hedgehog character are bound in pairs [10].

As the interaction strength is reduced while suppressing defects (reducing J at fixed large K), the nematic stiffness eventually vanishes, just as the spin stiffness of a magnet vanishes at its critical point. Indeed, if line defects are completely forbidden in the nematic (i.e., for infinite K), then the local molecular axis can be consistently assigned a direction throughout the system, effectively converting the nematic order parameter into a vector. The phase transitions on the line MH should therefore be in the Heisenberg universality class. Despite the absence of nematic order, there remains a nonzero free energy cost per unit length of disclination in the resulting disordered phase $\mathcal{T}$, so the free energy difference between the two boundary conditions is

$$\delta F_{\mathcal{T}} \sim C_2 L , \tag{2}$$

where C_2 is a constant which depends on the bare core energy as well as the elastic energy within a nematic correlation length of the defect. In this phase there is no long-range interaction between a pair of externally imposed disclinations. Long defect loops are exponentially suppressed by an effective string tension [11].

As the core energy is diminished at fixed, weak nematic interaction (reducing K at fixed small J), an externally imposed defect line will meander through the sample to gain an entropy proportional to its length L. C_2 will therefore diminish and eventually vanish at the phase boundary IM. (We will describe this transition in terms of an Ising lattice gauge theory whose critical point lies in the universality class of the three-dimensional Ising model.) In the resulting disordered phase $\mathcal{I}$ the nematic stiffness and the defect free energy per length both vanish, so the free energy difference between the two boundary conditions remains finite as L and R tend to infinity. In this phase line defects proliferate, and there is a nonzero density of infinitely long defects.

To test these ideas experimentally, one needs to vary independently the core energy of disclinations (i.e., K) and the local nematic interaction strength (i.e., J). This might be achieved by adding a "defectophilic" substance which exacts an entropic cost for defects by accumulating in their cores. Conversely, long "defectophobic" molecules which align with the nematogens but entangle unpleasantly within the cores would contribute to the free energy of the cores (while also altering the elastic constants). Since the two disordered phases will be difficult to distinguish, a crucial experimental signature would be singularities (in, for example, the dielectric constant or specific heat) at the continuous transitions into and out of the new isotropic phase $\mathcal{T}$. Less dramatically, one could also look for a weakening (strengthening) of the first-order transition as defects are suppressed (favored).

Lattice gauge theory.—The phase diagram of Fig. 2 was obtained by combining analytical and Monte Carlo studies of a lattice gauge theory for nematics which incorporates in a natural way (a) the local inversion symmetry of the nematic molecules, and (b) a variable suppression of disclinations. Consider coarse graining the orientational degrees of freedom of a nematic fluid. We imagine a cubic lattice immersed in the fluid, with a lattice spacing comparable to the nematic correlation length. A well-defined unit *vector* $\mathbf{S}_i$ is determined by the alignment of molecular axes near site i. Since the indivi-

dual molecules of the fluid are effectively head-to-tail symmetric, the choice of *orientation* for the vector $\mathbf{S}_i$ is arbitrary. The Hamiltonian of the system must therefore be invariant under the local symmetry operation $\mathbf{S}_i \rightarrow -\mathbf{S}_i$ for any particular site i.

Now consider the interaction along a link (ij). There are two ways that the local molecular axis can vary between the two lattice sites—it can interpolate directly [Figs. 3(a) and 3(b)] or by a large (nearly 180°) rotation [Figs. 3(c) and 3(d)]. To account for this local degree of freedom we introduce the link variables U_{ij}, which take on values ± 1. Each U_{ij} is associated with the rotation of the molecules which lie in a tube connecting small spheres surrounding sites i and j, as illustrated in Fig. 3. If the (smoothly varying) molecular axes between i and j can be consistently assigned *orientations* continuous with $\mathbf{S}_i$ and $\mathbf{S}_j$, then U_{ij} is $+1$ [Figs. 3(a) and 3(c)]; otherwise, it is -1 [Figs. 3(b) and 3(d)].

By introducing the link variables U_{ij}, we have expressed the Z_2 invariance of a nematic fluid as a gauge invariance [12], since for any site i we may invert the spin ($\mathbf{S}_i \rightarrow -\mathbf{S}_i$) if we also negate all of the link variables involving site i ($U_{ij} \rightarrow -U_{ij}$). The simplest Hamiltonian with this local gauge symmetry is

$$-\beta\mathcal{H} = J\sum_{\langle i,j\rangle} U_{ij}\mathbf{S}_i\cdot\mathbf{S}_j + K\sum_{\{ijkl\}} U_{ij}U_{jk}U_{kl}U_{li}\,, \tag{3}$$

where the second sum is over all elementary plaquettes $ijkl$. The first term is a nematic interaction which favors minimal variation of the director along link (ij) [see Figs. 3(a)–3(d)]. The second term is a defect core energy, since if the product of the link variables U_{ij} around a plaquette is negative, then the local molecular axis rotates by 180° as the plaquette is encircled [compare Figs. 1(b) and 1(c)]. The partition function is the trace of $e^{-\beta\mathcal{H}}$ over all configurations of both $\{\mathbf{S}_i\}$ and $\{U_{ij}\}$.

Analytic results.—We now analytically determine the behavior of the lattice gauge theory (3) near the $K=\infty$, $K=0$, and $J=0$ boundaries of Fig. 2, which then has the simplest topology consistent with these limits.

For infinite K, the product $U_{ij}U_{jk}U_{kl}U_{li}$ must be unity for every plaquette. There is then a gauge in which every U_{ij} is itself unity. Evidently, the *partition function* for this case is identical to that for the Heisenberg model. At $K=\infty$, the model thus has a continuous Heisenberg ordering transition at the usual critical value of J. This transition remains in the Heisenberg universality class for large but finite K, as shown in Fig. 2. For such K, one can easily sum over the U_{ij} by expanding the Boltzmann factor of the first term in (3) as

$$1 + J\sum U_{ij}\mathbf{S}_i\cdot\mathbf{S}_j + \frac{J^2}{2}\sum (U_{ij}\mathbf{S}_i\cdot\mathbf{S}_j)(U_{kl}\mathbf{S}_k\cdot\mathbf{S}_l) + \cdots\,. \tag{4}$$

The only gauge-invariant terms (hence the only ones surviving the trace over the U's) are those in which the links form closed graphs on the lattice. The coefficient of each term is the expectation value of a product of link variables in the pure gauge theory (i.e., a "Wilson loop" if the graph is a simple closed curve). In the pure gauge theory, the leading behavior of Wilson loops is known [12] to be exponential in the length of the loop. We can use this to trace over the U_{ij} to obtain an effective spin Hamiltonian which is a nearest-neighbor Heisenberg model with renormalized coupling $\tilde{J} = Je^{-\alpha}$, where $\alpha = e^{-4K}$. This merely shifts the value of $J_c(K)$, without affecting the character of the transition; there are also irrelevant, diminishing multispin corrections.

For $K=0$ the U_{ij} on different links are decoupled, and tracing over them yields the effective spin Hamiltonian $-\beta\mathcal{H}_{\text{eff}} = \sum \ln[\cosh(J\mathbf{S}_i\cdot\mathbf{S}_j)]$ [13]. The leading term is the Maier-Saupe interaction, which produces a strong first-order nematic-isotropic transition [14]. Now consider the effect of a small K. In this case, the gauge theory is in its confinement phase, and Wilson loops decay exponentially with their area. In the expansion of the partition function we need keep only graphs which are traced over in their entirety an even number of times (so the Wilson loop factor is 1), as well as those containing only single plaquette bulges. These same diagrams are generated by

$$-\beta\mathcal{H}_{\text{eff}} = \frac{J^2}{2}\sum (\mathbf{S}_i\cdot\mathbf{S}_j)^2 + J'\sum_{\{ijkl\}} (\mathbf{S}_i\cdot\mathbf{S}_j)(\mathbf{S}_j\cdot\mathbf{S}_k)(\mathbf{S}_k\cdot\mathbf{S}_l)(\mathbf{S}_l\cdot\mathbf{S}_i)\,, \tag{5}$$

with $J' \approx J^4 \tanh K$. The second term in (5) suppresses defects. The first-order transition remains for small K.

For $J=0$ the model is a pure Ising lattice gauge theory, which is dual to the ordinary Ising model in three dimensions [12]. Use of this duality shows that the free energy per length of an externally imposed defect (as discussed earlier) should vanish as $(K-K_c)^{-\nu}$ in the limit $L,R \rightarrow \infty$. At small values of J, the gauge coupling is renormalized to $K_{\text{eff}} \approx K + J^4/27$, but again the universality class is not altered.

Monte Carlo results.—Monte Carlo simulations [14] of the lattice gauge theory (3) yield the phase diagram shown in Fig. 2. These calculations used a standard Metropolis algorithm on cubic lattices of up to 16^3 sites [15]. Define the (manifestly gauge-invariant) tensor order parameter $Q_i^{\alpha\beta} \equiv S_i^\alpha S_i^\beta - 1/3\delta^{\alpha\beta}$ for each lattice site i (α and β are vector indices), with magnitude

$$q^2 = \frac{1}{N(N-1)}\,\frac{3}{2}\sum_{i\neq j} Q_i^{\alpha\beta}Q_j^{\beta\alpha}\,. \tag{6}$$

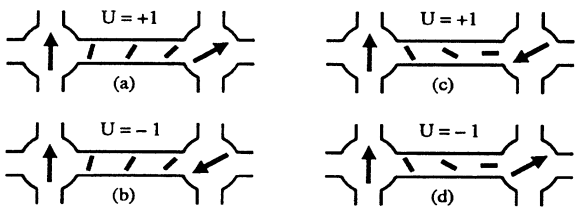

FIG. 3. The link variables U_{ij} provide information on the rotation of the local molecular axes between sites i and j.

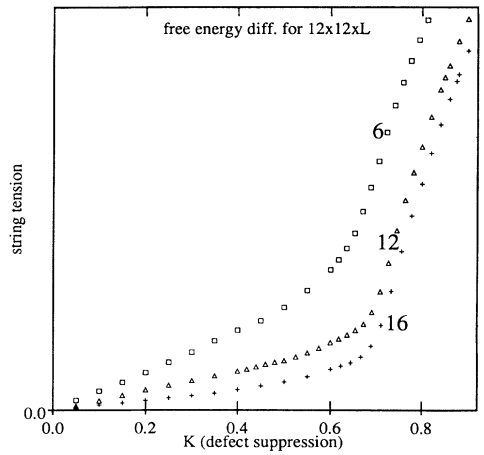

FIG. 4. The free energy difference $\delta F/L$ between a pair of $12\times12\times L$ rectangular prisms with and without externally imposed defects, for $J=0.5$ and $L=6$, 12, and 16. Below $K\approx0.7$, there is no line tension in the thermodynamic limit.

For randomly oriented spins, q^2 vanishes; for spins aligned with a common axis, q^2 is unity. Using q^2 we can find transitions into the N phase.

We have applied finite-size scaling [16] to the transitions shown in Fig. 2. To show that *gauge-invariant* quantities exhibit Heisenberg-class critical behavior, we identify $\langle\mathbf{S}\rangle$ with the magnetization m of a Heisenberg model, so $Q^{\alpha\beta}\approx m_\alpha m_\beta - m^2\delta_{\alpha\beta}/3$. Then

$$g(L,t)\equiv\frac{\langle(\mathrm{tr}Q^2)^2\rangle_L}{\langle\mathrm{tr}Q^2\rangle_L^2}\approx\frac{\langle m^8\rangle_L}{\langle m^2\rangle_L^4}=g(Lt^\nu)\,, \tag{7}$$

where $Q^{\alpha\beta}=N^{-1}\sum Q_i^{\alpha\beta}$. The last equality in (7) is a standard finite-size scaling result. The critical coupling and the exponent ν of the equivalent Heisenberg model can be determined by using this equation.

To bring β to light we use another standard scaling relation, $m=L^{\beta/\nu}f(Lt^\nu)$. Assuming J is not too much below J_c, we have $q\sim Q^2\sim m^4$, so that

$$\langle q^2\rangle_L\approx L^{4\beta/\nu}f(Lt^\nu)\,. \tag{8}$$

We have verified (7) and (8) for K as small as 0.78.

In a finite system, the delta function in the specific heat found at a first-order transition becomes a rounded peak whose height is linear in the volume of the system. We have observed this behavior for values of K as large as 0.70, and have also confirmed a double-peaked order parameter distribution [15].

The line IM between the two isotropic phases $\mathcal{I}$ and $\mathcal{T}$ can be found by studying the effective string tension $\delta F/L$, as discussed in the introduction (see Fig. 4). This is consistent with an analysis of the specific heat [15].

In conclusions, the first-order character of the ordering transition in nematic systems has been linked with the behavior of topological line defects. Surprisingly, we find *two* different disordered phases which are distinguished by the free energy cost of inserting an additional line defect through a finite sample. In the context of a lattice gauge theory for nematics which incorporates a variable suppression of defects, we find that the first-order nematic-to-isotropic phase transition can be split into two continuous transitions in the three-dimensional Ising and Heisenberg universality classes, respectively. These new transitions and phases in nematic liquid crystals should be accessible experimentally.

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Note added.—P. Poulain, P. Richetti, and D. Roux have observed what appears to be critical opalescence at a transition between two isotropic phases in nematic/polyball mixtures.

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