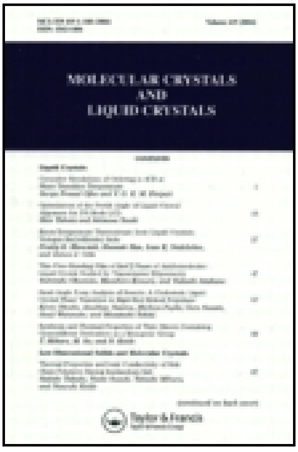

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Intrinsic Torsional Viscosity in a Narrow Tube of Nematic Liquid Crystal†

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We describe two phenomena which can occur in crystalline liquids and which are analogues of effects that occur in superconductors. The first concerns the acquisition of torsional viscosity, rather than rigidity, by long and narrow nematic liquid crystals, due to the occurrence of topologically accessible thermal director fluctuations; it is the analogue of fluctuation-induced resistance in superconductors. The second concerns the torques and forces between a pair of grooved plates immersed in a pre-nematic fluid; its superconducting analogue is the Josephson effect, *e.g.*, at an *SNS* junction. We conclude with approximate numerical estimates for the magnitudes of these phenomena.

Keywords: thermal fluctuations in nematics, intrinsic viscosity, interfacial forces in pre-nematic fluids

The purpose of this paper is to describe briefly two phenomena which occur in nematogenic systems and are analogues of phenomena occurring in superconductors. The first concerns the conversion, from rigid to viscous, of the intrinsic response of a long and narrow channel of nematic liquid crystal to externally applied torques.[1] This qualitative change is caused by topologically accessible thermal fluctuations in the orientation of the nematic director; it is the analogue of the intrinsic electrical resistance exhibited by long and narrow channels of superconducting metal due to thermal fluctuations in the amplitude of the complex superconducting order parameter.[2] The second phenomenon concerns the equilibrium torques and forces between a pair of grooved plates immersed in a *pre-nematic* fluid, *i.e.*, a fluid whose bulk equilibrium state is isotropic, but which is close to the isotropic-nematic transition.[3] The plates are coupled when there is appreciable overlap between regions of fluid in which nematic order is induced by proximity to the grooved plates.[4] This effect is the analogue of the Josephson effect,[5] as exhibited, *e.g.*, by a metallic superconducting-normal-superconducting (*SNS*) junction, due

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to the overlap in superconducting order induced in the normal region by the nearby pieces of superconductor.

We will attempt to give an account of these phenomena that emphasizes the crucial physical ingredients without dwelling on analytical technicalities. More detailed discussions can be found of intrinsic viscosity in ref. 1, and of Josephson-type inter-plate coupling (which has been investigated in collaboration with M. B. Tarlie) in a forthcoming paper.[6]

First we shall discuss the response to external torques of nematic liquid crystalline tubes. Our approach is an adaptation of the method introduced by Langer and Ambegaokar (LA) in their work on the resistance of narrow superconducting channels,[2] although there turn out to be significant differences in both the nature of the solution and the resulting physical character of the dissipative phenomenon. The central question is this: If a constant torque (about the long axis) is applied to the director at one end of a long and narrow sample of nematic liquid crystal, with the director at the other end held fixed, how does the system respond? To address this, consider a sample of nematic liquid crystal which is sufficiently long and narrow that the local preferred direction of molecular alignment, *i.e.*, the director $\mathbf{n}$, depends only on the position on the long axis (*i.e.*, the z-axis) of the sample, so that $\mathbf{n} = \mathbf{n}(z)$. Then imagine applying end-plates to the sample, perpendicular to the long axis, whose effect is locally to constrain the director to a particular direction in the plane of the plates. Now, for fixed end conditions conferred by the strongly-anchoring end-plates, and provided the Frank constrants satisfy $K_b > 2K_t$, there is a discrete family of locally stable minima of the Frank free energy.[7] This comprises the uniformly twisted configurations labelled by the integer m. For example, when the directions of alignment caused by the boundary plates at $z = \pm L/2$ are identical, these minima are given by $\mathbf{n}(z) = \cos\phi(z)\mathbf{e}_x + \sin\phi(z)\mathbf{e}_y$, as depicted in Figure 1, with $d\phi/dz = m\pi/L$, and where $\{\mathbf{e}_i\}$ is an orthonormal basis. In these configurations the director uniformly rotates in the plane of the end-plates as one moves through the sample; they have a free energy given by $F_s = ALK_t(m\pi/L)^2/2$, where L is the length of the sample and A is its cross-sectional area. We shall refer to them as *metastable states*, as they are destabilised by fluctuations (provided $|m| \geq 2$). If the end-plates are held parallel, and if director fluctuations are neglected, then the system will adopt one of these states as its equilibrium state.

We now consider what happens to these states if one boundary plate is held fixed and a constant external torque is applied to the other. In the absence of thermal fluctuations the torqued end-plate rotates (changing the boundary conditions) and the system remains in an instantaneous local minimum of the Frank free energy, being continuously transported through a sequence of metastable states (on the spiral of Figure 2). The twist and internal torque continue to increase until the internal torque matches the applied torque; at this time macroscopic rotation ceases. Regarded as a *black box*, the nematic medium *rigidly* couples the pair of end-plates.

What happens to the externally torqued system when thermal fluctuations in the director orientation are permitted (even without the introduction of fluctuations in the *degree* of molecular alignment) so that the system no longer necessarily

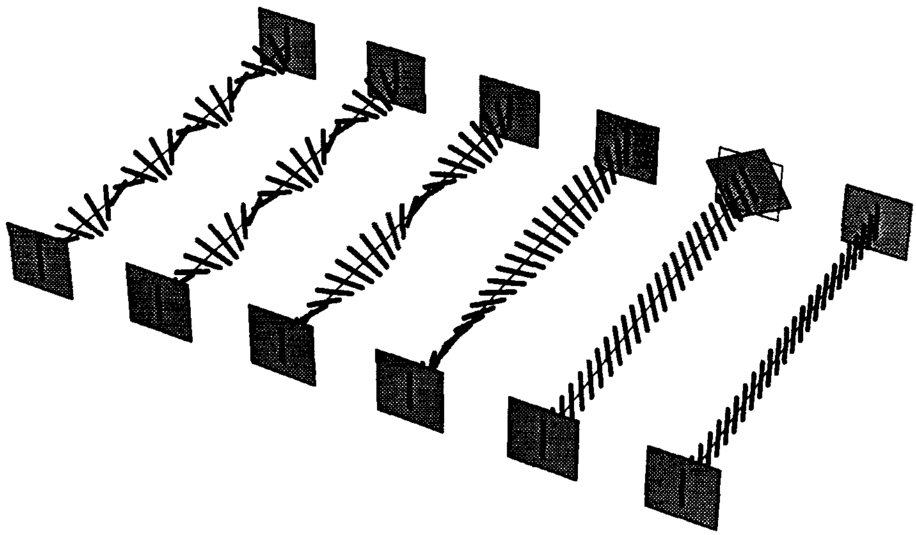

FIGURE 1 The variation of the direction for a variety of twisted configurations of nematic liquid crystal. From right to left, the total twist across the sample in radians is: 0; $\pi/6$; π; 2π; 3π; 4π. Apart from the slightly twisted case ($\pi/6$), these configurations comprise part of a family of metastable states, as they are locally stable (for $K_b > 2K_t$) and have common end-plate orientations.

adopts a configuration which locally minimises the Frank free energy? It can respond by adopting a dissipative non-equilibrium steady state, in which topologically accessible fluctuations intermittently relieve (and less often enhance) the twist, after which the torqued end-plate rotates so as to restore the internal torque to a value which balances the applied torque. This process is depicted in Figure 2. Regarded as a *black box*, the medium *viscously* couples the pair of end-plates, so that a relative torque produces, on average, a constant rate of relative rotation. Our goal is to characterize this viscosity. Whilst under some circumstances the incorporation of thermal fluctuations produces only minor quantitative changes to the properties of liquid crystals, the response to torque provides an example in which fluctuations can induce a qualitative change from rigid to viscous.

How can we be sure that the required fluctuations are topologically accessible? For simplicity, consider the situation where, in the absence of fluctuations, the applied torque happens to confer the same orientation on the ends of the sample. Then all the possible non-singular configurations $\mathbf{n}(z)$ fall into one of two classes: those which can be smoothly deformed into the untwisted (*i.e.* spatially constant) configuration, and those which cannot.[8] The implication of this result of homotopy theory is that by varying the orientation alone, and not destroying the local degree of molecular ordering (*i.e.* without the free energy cost of creating an isotropic droplet) it is possible for spontaneous thermal fluctuations to relieve (or enhance) the degree of twist m (so long as the parity of m is conserved). So, once we allow fluctuations, we are no longer guaranteed that, under large distortions of the nematic, the system has the rigidity that Goldstone's theorem implies for infinitesimal distortions. Alternatively, if we fix the end-plate orientations and measure the torque across a twisted sample, instead of remaining constant for all time, as

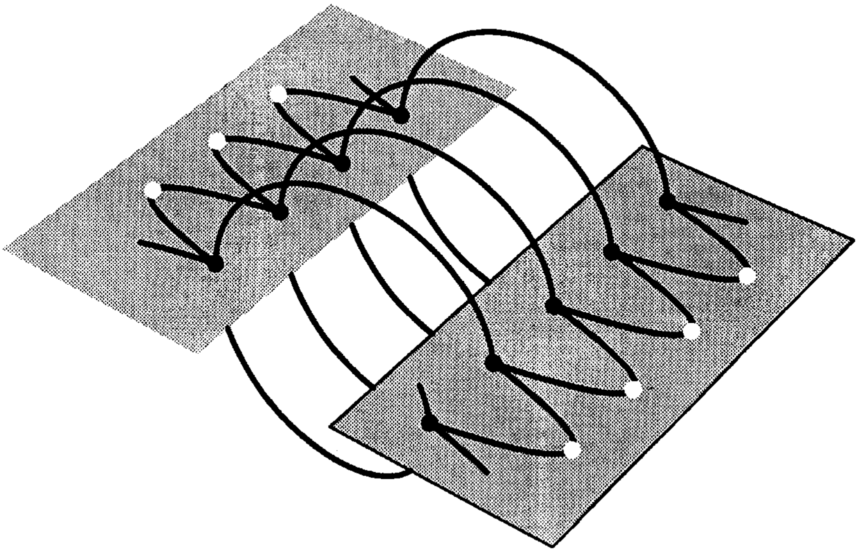

FIGURE 2 A schematic representation of a portion of the space of configurations; each point represents a configuration $\mathbf{n}(z)$. The bordered plane contains those configurations with end conditions $\mathbf{n}(\pm L/2) = \mathbf{e}_x$ which can be smoothly deformed into the untwisted configuration; the unbordered plane contains those with end conditions $\mathbf{n}(-L/2) = -\mathbf{n}(L/2) = \mathbf{e}_x$ which are not smoothly deformable into the untwisted configuration. The black dots represent uniformly twisted metastble states; those of Figure 1 with total twist 0, 2π and 4π radians (and more generally $2m\pi$) would appear on the bordered plane, whilst those with total twist π and 3π radians (and more generally $(2m + 1)\pi$) would appear on the unbordered plane. The black spiral represents the evolution of the system in the absence of fluctuations as an externally applied torque quasi-statically rotates one of the end-plates, introducing twist and isothermally transporting the system between metastable states of alternating parity. The white dots represent non-uniform, unstable saddle-point configurations through which the system passes when making topologically allowed fluctuations between pairs of metastable states. The black in-plane arcs represent paths between pairs of metastable states; the system relieves (or enhances) twist by making fluctuations which move it along similar paths. The viscous non-equilibrium steady state at constant external torque includes processes where the system fluctuates, within a plane, out of a metastable state, and subsequently returns to it along the spiral.

it would in the absence of fluctuations, the torque would diminish in time at a rate which we shall determine.

The topological considerations, however, only tell us that transitions may *in principle* occur between certain pairs of metastable states; they tell us nothing about the free energy barriers that must be overcome in order to make such transitions. Our remaining task is to characterize the rate at which such transitions between states with differing degrees of twist occur. To do this, we follow LA in borrowing techniques familiar from classical mechanics. The central point is that whilst metastable states are characterized as stationary points of the free energy which are *local minima*, the states at the top of the free energy barriers *also* make the free energy stationary, but are not minima. Instead of being stable with respect to small variations in all directions in configuration space, they are saddle-points, stable in all directions except one, and we refer to them as saddle-point states.[9] That unstable direction is the direction of steepest descent along which the system crosses the barrier. Armed with this characterization we can estimate the barrier heights and, using the Arrhenius formula, the rate of transitions between metastable states.

Classical mechanics enters through the following observation. If the director **n** only depends on z then the total free energy reduces to a one-dimensional integral over z, reminiscent of an action integral, with z interpreted as the time variable and the free energy density interpreted as a lagrangean. As discussed in detail in ref. 1, the total free energy of the liquid crystal problem is then equivalent to the action of a particle moving on a disc. Our goal is to determine the form of the metastable and saddle-point states and, in particular, the height of the barriers between metastable states (*i.e.* the difference free energy between the metastable states and the saddle-point states). This corresponds to solving Newton's equations for two classes of classical trajectory—those which minimize the action, and those which make it stationary but with one unstable direction—and determining the action of these solutions. The classical mechanics problem is unusual but solvable, involving a point particle which moves on a disc[10] of radius unity in the absence of a potential but with a moment of inertia and mass which depend on the radial degree of freedom and the ratios of Frank constants. (This *skater* rotates more slowly as she pulls in her arms. Analogously, during the fluctuation event the twist is pushed away from the tilting region.) The lagrangean admits two useful conserved quantities, the analogues of energy E and angular momentum J. Not only do they allow us to reduce the classical mechanics problem to quadratures but also, due to the absence of a potential energy term,[11] energy conservation is equivalent to lagrangean conservation. Thus, any configuration of the director which makes stationary the Frank free energy has a free energy density which is spatially uniform, even when the configuration is not. This provides a great simplification when we come to compute the total free energy of any saddle-point configuration: although their variation with z is determined implicitly in terms of elliptic functions, we can compute their free energies exactly, without having to (i) solve for the configurations explicitly, (ii) insert into the Frank free energy density, and (iii) integrate over the sample length. We suspect that this simplification may be useful for other systems, such as superfluid ^{3}He, which may be described by the gradient energy of a non-linearly constrained field.

Using the mechanics analogy it is not difficult to establish an expression for the barrier heights. We consider three states: the metastable state with twist m and free energy F_s^+; the metastable state with 2π radians less of twist and free energy F_s^-; and the saddle-point state with free energy F_u through which the system passes as it fluctuates between them. It can then be shown that the barrier heights are given by $F_u - F_s^{\pm} = K_t A(m\pi/L)(\delta + \pi \mp \pi)$, where the *phase shift* δ is determined entirely by ratios of Frank constants through Equation (4) of Reference 1. Notice that the barrier heights *increase* with the degree of twist m.

If we now balance the rate ω at which the end plate rotates when a torque τ is applied against the net rate at which twist is lost through fluctuations, we obtain the result $\omega = (8\pi L/t_r\ell)\exp(-\beta\tau(\pi + \delta))\sinh(\beta\tau\pi)$, where ℓ is the length of a statistically independent segment and $\beta = 1/k_B T$. Note that this result is not applicable when $\beta\tau\delta \approx 1$, but only for larger values of $\beta\tau\delta$. One can define an effective intrinsic torsional viscosity $\gamma(\omega)$ through $\tau(\omega) \equiv \omega\gamma(\omega)$. It shows rotation-thinning, decreasing strongly with ω, because transitions from states with higher twist must surmount higher barriers, and thus occur less frequently; conversely, low external torque produces low twist, for which dissipative fluctuations happen more often. Since the lightly twisted metastable states are protected only by rather low barriers, it may be difficult to produce well-twisted states by twisting, starting from the untwisted state. An alternative scheme might be to start with a cholesteric whose pitch is temperature-dependent and diverges at a certain temperature T_P. The twisted nematic might then be produced by applying end-plates to the cholesteric, and then adjusting the temperature to T_P so that the system is prepared in a well twisted metastable state, despite its equilibrium state being nematic. If the initial pitch leads the cholesteric to be blue then, with fixed end-plates, the colour would redden, logarithmically, as fluctuations lengthen the effective pitch.

Although the nature of the fluctuations in the nematic is quite different from those which give resistance to superconducting strips, being in orientation rather than amplitude, the two dissipative phenomena have a formal analogy: $\tau \sim I$, $\omega \sim V$ and $\gamma \sim 1/R$, where I, V and R are the superconducting current, voltage and resistance. However, in the superconductor, the barriers *diminish* with current, producing a highest metastable current carrying state (*i.e.* an upper critical current).

For illustrative purposes, we make an extremely crude estimate of the torque and rotation rate for a hypothetical situation. It should be emphasized that the exponential dependence of the rotation rate on the torque makes estimates extremely sensitive to the choice of parameters. We consider a sample of length $L = 1$ cm and area $A = 100\ (\mu\text{m})^2$, at a temperature $T = 400$ K, and twisted until $m = 6$. For the elastic constants we take $K_s = 1.6$ pN, $K_t = 1$ pN and $K_b = 3.2$ pN, which gives $\delta \approx 0.6$. The time-scale[12] is taken to be $t_r = 100$ ns and the statistical length is taken to be $\ell \approx 2\pi/J = L/3 \approx 0.33$ cm. This gives a torque $\tau \approx 0.19$ aJ and a rotation rate $\omega \approx 0.48\ \text{s}^{-1}$. This concludes our discussion of fluctuation-induced torsional viscosity in a narrow nematic channel.

We now turn briefly to the issue of the torques and forces between a pair of grooved plates which cause alignment in an otherwise *isotropic* fluid of nematogens.[4,6] When much farther apart than the nematic correlation length ξ the plates are essentially independent. However, they increase the free energy of the fluid because they cause nematic alignment (*i.e. condensation*) within a boundary layer coating the plates which decays exponentially into the bulk on a scale set by ξ, whilst the homogeneous equilibrium state is isotropic. If both plates cause the molecules to prefer the same alignment direction then, as they are brought closer, the boundary layers overlap and their free energy cost is diminished; hence the plates attract each other. However, if the plate alignment directions are orthogonal then this gain in condensation free energy is offset by a cost in *gradient* free energy from the distortion of the induced nematic ordering as it interpolates between the plates; this can cause a net repulsion between the plates.

This effect can be simply modelled by the Landau-de Gennes free energy[12] with gradient terms, truncated at quadratic order,

$$F = \frac{1}{2}\int_V d^3r\{a(T - T_c^*) \sum_{a,b=1}^{3} (Q_{ab})^2 + \sum_{a,b,c=1}^{3} (L_1(\nabla_a Q_{bc})^2 + L_2 \nabla_a Q_{ac} \nabla_b Q_{bc})\}, \quad (1)$$

where Q_{ab} is the symmetric, traceless nematic order parameter, $T - T_c^*$ measures the departure from what would be the critical point (if a first order transition did not preempt it), and a, L_1 and L_2 are parameters. Suppose that the plates lie in the planes $z = \pm D/2$ and have area A. We minimize F subject to: (i) the conditions at the end-plates $z = \pm D/2$ that the order is uniaxial, with magnitude q and orientation $\pm\theta/2$; and (ii) the assumption that Q_{ab} depends only on z, which gives an attractive force per unit area between the plates

$$P \equiv \frac{1}{A}\frac{\partial F}{\partial D} = q^2 \frac{L_1}{3\xi_-^2} \{e^{-D/\xi_+} + 3e^{-D/\xi_-} \cos 2\theta\}, \quad (2)$$

and a torque per unit area

$$\frac{\tau}{A} \equiv -\frac{1}{A}\frac{\partial F}{\partial \theta} = -2q^2 \frac{L_1}{\xi_-} e^{-D/\xi_-} \sin 2\theta, \quad (3)$$

where the correlation lengths $\xi_\pm$ are defined through $\xi_-^2 \equiv L_1/a(T - T_c^*)$ and $\xi_+^2 \equiv (L_1 + 2L_2/3)/a(T - T_c^*)$. For simplicity, we have presented only the terms which dominate at large plate separation D. Notice the following features: (i) the force and torque decay exponentially with D; (ii) the torque is an oscillatory function of the relative plate orientation θ, with period π, reflecting the *head-tail* symmetry of the nematic state and the absence of metastability (*i.e.* uniqueness of the state for fixed boundary conditions); (iii) the force is also an oscillatory function of θ, which is attractive when $\theta = 0$ but can become repulsive around $\theta = \pi/2$, provided that $\xi_-^{-1} - \xi_+^{-1} < D^{-1} \log 3$; (iv) the decay lengths $\xi_\pm$ grow as the temperature T is reduced, although their divergence at T_c^* is preempted by the isotropic-nematic transition; and (v) the induced nematic state typically has a biaxial component.

What is the analogy with Josephson-type phenomena? In the static Josephson effect[5] a constant, dissipationless current I flows (in the absence of a potential difference) when a phase difference in the superconducting order parameter φ is maintained across, *e.g.*, an *SNS* junction, with I being a periodic function of φ. When thermal fluctuations are included,[13] phase slippage can occur, producing a noise voltage with a non-zero mean value. In the dynamic Josephson effect[5] an oscillatory current flows when a potential difference V causes φ to change linearly with time. In the pre-nematic, a constant reversible torque τ occurs when a mismatch angle θ is maintained in the alignment of the end-plates. In the gaussian approximation, fluctuations will renormalize this torque, in the spirit of the Casimir effect, maintaining reversibility. However, larger phase-slip-type fluctuations can cause noisy dissipative rotation of the plates with a preferred sense. Furthermore, if the plates are rotated uniformly in time at a rate ω then the torque and inter-plate force oscillate as a function of time. The analogy is then given by $\tau \sim I$, $\theta \sim \varphi$ and $\omega \sim V$. Furthermore, $M \sim C$ and $\gamma \sim 1/R$, where M and γ are the moment of inertia and viscosity of the fluid, and C and R are the capacitance and resistance of the junction, and the effect of a magnetic field on the junction can be simulated by molecular chirality.

To conclude we give a numerical estimate of the forces and torques produced in a hypothetical experiment. Suppose that $2L_1 \approx L_2 \approx 3 \times 10^{-6}$ dynes, $a \approx 42 \times 10^3$ $\text{Jm}^{-3}\text{K}^{-1}$, $(T - T_c^*) \approx 1\text{K}$ and $q \approx 0.3$. Then $\xi_- \approx 189$ Å and $\xi_+ \approx 289$ Å and, for a plate separation $D \approx 500$ Å, the attractive force per unit area is given by $P \approx 0.223\ (1 + 1.2\cos 2\theta)\text{kNm}^{-2}$, and the torque per unit area is given by $\tau/A \approx -10.1 \sin 2\theta\ \mu\text{Nm}^{-1}$.

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9. In fact we shall simply assume that the barrier-top states we find do indeed have the appropriate character. One scheme for checking this would be to extend the Jacobi test for extrema of functionals; see *e.g.*, Charles Fox, *An Introduction to the Calculus of Variations* (Oxford University Press, 1950).

10. That there are two degrees of freedom results from subjecting the three components of the director to the single non-linear constraint that it has unit magnitude.
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