Phase Winding and Flow Alignment in Freely Suspended Films of Smectic-*C* Liquid Crystals

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We present the first experimental study of shear-flow–induced behavior in a two-dimensional anisotropic fluid: circular, freely suspended films of smectic-*C* liquid crystals. Depending on the topology of the in-plane director configuration, we observe either a phase-winding state, where Φ, the phase defining the director orientation relative to the velocity field, winds up in the center of the film, or a flow-alignment state, where Φ is constant.

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A density wave in one direction and orientational order in another characterize smectic-*C* liquid crystals.[1] In general, the hydrodynamic equations for smectic *C*[2,3] are complicated, making flow studies impractical when both density wave and orientational order are involved. However, anisotropic two-dimensional flow behavior is expected for shear applied in the plane of a set of parallel layers.[2,3] This fact was used to point out[4] the possibility of flow alignment—the in-plane orientational order, **n**, is constant relative to the velocity field—and stimulated the present experiment.

Parallel layers were achieved by drawing freely suspended films across a circular hole and shear was applied by a rotating needle in the center of the film (Fig. 1). Depending on the topology of **n**, we find two regimes. The first is the phase-winding regime obtained when the film is free of defects so that the topological index, S, of the film is zero. Here, one observes concentric rings of constant orientation, Φ, with Φ a maximum, Φ_0, at the needle boundary layer, r_0, and zero at $r=R$, the film radius. Φ_0 increases by 2π with each turn of the needle. Since the orientation is fixed at the rim of the film, shear winds **n** up. The second is flow alignment that occurs when the needle is at the center of a defect of topological index $S=+1$ where **n** has circular symmetry. These results demonstrate the overwhelming importance of topology in determining the possible states for systems showing truly two-dimensional, anisotropic behavior.

The films we used had a diameter $2R \approx 0.8$ to 2.6 mm and a thickness of 0.5–2 μm. They are drawn in an oven and observed with a polarizing microscope. In the present Letter, we focus on results obtained in the smectic-*C* phase. The results for several materials showing this phase were all qualitatively similar. The quantitative data shown here were obtained with use of TB9A [terephthalylidene-bis-(4-nonylaniline)][5,6] which exhibits the *C* phase between 157.5 and 192.7 °C as well as several other smectic phases.

We start the experiment from an initial state where Φ is a constant for $r_0 \leqslant r \leqslant R$. We insert the tip of a glass needle, $r_0 \approx 20$ μm, into the film so that it is concentric with R. Rotation of a well-centered needle exerts circular shear on the film (Fig. 1). The angular speed of the needle can be as high as 5000 rpm.

In the phase-winding regime, a $2\pi k$, k an integer, rotation of the needle results in a $2\pi k$ increase of Φ at r_0 relative to the phase at R. This is seen in polarized light as $4k$ dark concentric fringes [Figs. 2(a) and 2(b)

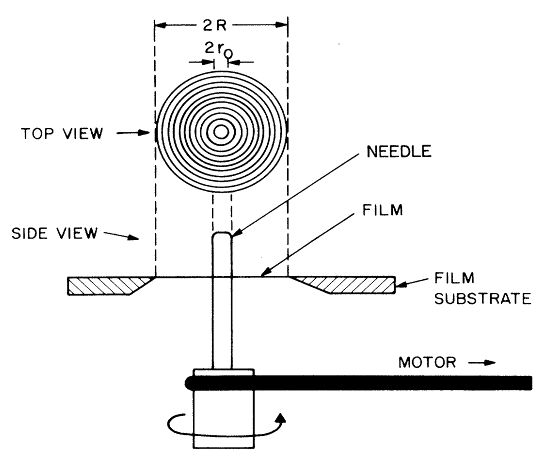

FIG. 1. The experimental setup.

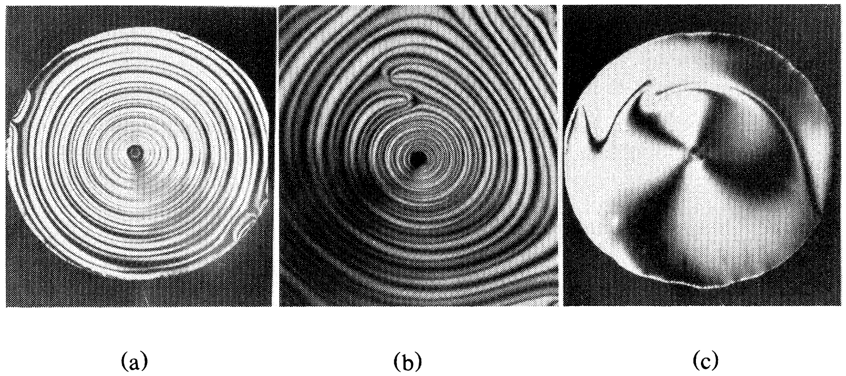

FIG. 2. Photomicrographs showing the following: (a) Phase winding in the smectic-C phase of TB9A. The needle diameter in the center of the film is $\approx 60\ \mu$m. The film diameter is ≈ 2 mm. (b) Disclination pairs forming when rings break (Ref. 7). They mediate the transition between the phase winding in (a) and the flow alignment in (c). (c) Flow alignment in the smectic-C phase of TB9A. The flow-alignment angle ($\approx 45°$) is deduced from the orientation of the four dark brushes, or extinction bands, radiating from the needle. This contrast is the signature of an $S=1$ defect and occurs when $\mathbf{n}$ is parallel (or perpendicular) to the four directions of a crossed polarizer and analyzer. Furthermore, because of the circular symmetry of $\mathbf{n}$, the pattern is invariant with respect to sample rotations about the viewing axis.

with a schematic of the director orientation shown as an inset in Fig. 3(a)]. The number of rings is determined by the number of rotations until a saturation is reached where the phase apparently slips at the needle boundary layer. This occurs when the radial width of the rings is comparable to the film thickness. Thinner films support more rings.

As long as the needle remains in the film, the picture of concentric rings does not change even if the needle stops rotating because the boundaries of the film pin Φ. Furthermore, reversing the rotation direction leads to an unwinding of the ring structure.

If the needle is pulled out of the film, the $S=0$ topology is revealed as the needle area is replaced by uniformly oriented material. This is schematically shown as an inset in Fig. 3(b). The phase at the center of the film unwinds in a characteristic time τ proportional to $K/\gamma_1 \approx 10^{-4}$ cm^2/sec, where K is an elastic constant and γ_1 a viscosity. This is observed as concentric fringes that shrink and vanish at the center of the film.

The flow-alignment regime is seen in the polarizing microscope [Fig. 2(c)] as four dark brushes radiating from the needle. The topological index for this configuration is $S=+1$ since the orientation of $\mathbf{n}$ changes by $1\times 2\pi$ around any circuit that includes the needle. The needle forms the core of the disclination. Pulling out the needle creates a singular core at $r=0$. As we will see, flow alignment cannot be obtained in circular shear without an $S=+1$ defect centered at the needle.

To describe the phenomena observed, we note that the hydrodynamic equations for the in-plane director of smectic C[2,3] specialized to the present geometry are isomorphic to those of a two-dimensional nematic and we can equivalently use the equations of Ericksen[1,8] specialized to two dimensions. We use polar coordinates (r,ϕ,z) and the in-plane director $\mathbf{n}$ is given by $n_r=\cos\Phi$, $n_\phi=\sin\Phi$. A uniformly oriented film is thus described by $\Phi=\Phi_0-\phi$ and an isolated disclination of index $S=+1$ by $\Phi=$const. In general, for index S, $\Phi=(S-1)\phi+$const, where S is an integer.

In our geometry, only two of the elastic constants for a smectic-C liquid crystal[9,10] are needed and, assuming those two to be equal, we have for the elastic energy

$$F_{\mathrm{el}}=\frac{K}{2}\int\left[\left(1+\frac{\partial\Phi}{\partial\phi}\right)^2+\left(r\frac{\partial\Phi}{\partial r}\right)^2\right]\frac{dr}{r}\,d\phi, \qquad (1a)$$

when

$$\Phi=(S-1)\phi+f(r). \qquad (1b)$$

Minimizing Eq. (1a) with boundary conditions $\Phi=-\phi$ at $r=R$ and $\Phi=-\phi+\Phi_0$ at $r=r_0$, we obtain the static solution,

$$\Phi=-\phi+\Phi_0\frac{\ln(r/R)}{\ln(r_0/R)}, \qquad (2)$$

predicting a logarithmic dependence of Φ on r when $S=0$. In Fig. 3(a), we plot Φ vs r measured at constant ϕ by direct observation in the microscope. The agreement with Eq. (2) is clearly excellent. In the phase-winding regime, Φ_0 is a function of time. For every 2π rotation of the needle, Φ_0 increases by 2π and the elastic energy stored in the film is, from Eqs. (1a) and (1b),

$$F_{\mathrm{el}}=\pi K\frac{\Phi_0^2}{\ln(R/r_0)}.$$

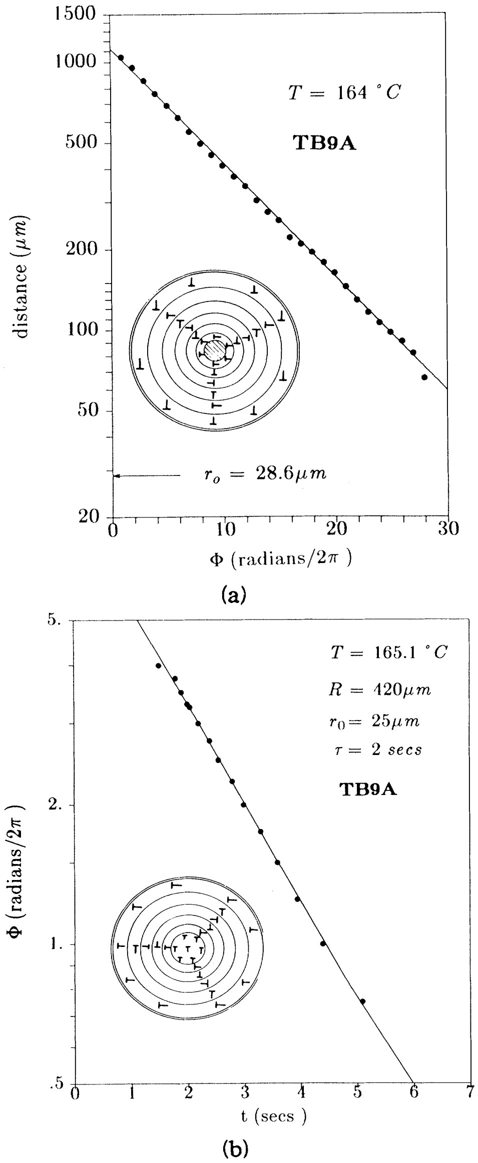

FIG. 3. (a) $\ln(r)$ as a function of phase, Φ, in units of 2π rad. The ring structure is sketched as an inset. (b) $\ln\Phi$ as a function of time, t, during phase unwinding. The inset is the ring structure after the needle has been removed from the film and is replaced by uniformly oriented smectic C. The unit for Φ is 2π rad.

To describe the relaxation of the rings when the needle is pulled out of the film, we again minimize Eq. (1a) and assume a simple exponential decay for the time dependence of the director orientation. We obtain

$$\frac{K}{r^2}\frac{\partial^2\Phi}{\partial\phi^2}+\frac{K}{r}\frac{\partial}{\partial r}r\frac{\partial\Phi}{\partial r}=\gamma_1\frac{\partial\Phi}{\partial t}. \tag{3}$$

A time-dependent solution of Eq. (3) that has our static result [Eq. (2)] as an initial condition at small r is

$$\Phi(r,\phi,t)=-\phi+\Phi_0\frac{K_0(r/R)}{K_0(r_0/R)}\exp\left(\frac{-t}{\tau}\right), \tag{4}$$

where K_0 is the modified Bessel function of the second kind[11] and $\tau=(\gamma_1/K)R^2$.

To check Eq. (4), we measured the time dependence of Φ at $r=0$ with a video camera and a photodiode. Figure 3(b) shows Φ as a function of time for a typical run. The time constant τ is the inverse of the slope in Fig. 3(b). We find again quantitative agreement between predictions and observations. We also verified the R^2 dependence of τ by repeating the experiment for various film diameters. A plot of $\ln\tau$ vs $\ln R$ gives a slope of 1.9 ± 0.1. A plot of τ vs R^2 gives $\gamma_1/K=7.7\times10^3$ sec/cm^2, that is, about a hundred times smaller than in nematics ($\gamma_1/K\approx5\times10^5$ sec/cm^2) and about the same as smectic-C films only two or three layers thick[12] ($\gamma_1/K\approx10^4$ sec/cm^2).

To describe the flow regimes, we assume that the velocity profile, $v_\phi(r)=r\omega(r)$, is given simply by the large-gap approximation to the Navier-Stokes equation for circular shear,[13] i.e., $v_\phi(r)\sim1/r$. Then for fixed but arbitrary S we obtain

$$K\gamma_1^{-1}\left(\frac{1}{r}\frac{\partial}{\partial r}r\frac{\partial\Phi}{\partial r}+\frac{1}{r^2}\frac{\partial^2\Phi}{\partial\phi^2}\right)=\frac{\partial\Phi}{\partial t}+\frac{\omega_0r_0^2}{r^2}(S-\lambda^{(3)}\cos2\Phi). \tag{5}$$

Given our initial dependence [Eq. (1b)] of Φ on the spatial coordinates, (r,ϕ), Eq. (5) shows that only $S=1$ has a stationary solution. Furthermore, for constant rotation, a given ring pattern, such as shown in Fig. 2(a), is stationary only when the phase slips at the needle boundary. On the other hand, for $S=1$, a stationary flow-alignment solution, Φ_A, is possible where $1/\cos2\Phi_A=\lambda^{(3)}$ as predicted.[4] From the form of this solution, clearly, flow alignment cannot occur in materials where $\lambda^{(3)}<1$ even if $S=+1$.

A typical picture of flow alignment is given in Fig. 2(c) where the four black brushes crossing at the needle are the signature of the axial symmetry of an $S=1$ disclination. Since these brushes are nearly at 45° to the direction of the crossed polarizer and analyzer, $\Phi_A\approx45°$ so that $\lambda^{(3)}\gg1$. When the needle is in the film, it forms the core of the disclination. Pulling out the needle results in a singular core at $r=0$. The static elastic energy (in the one-constant approximation) for $S=1$ with the needle in the film is just $F_{\rm el}=\pi K\ln(R/r_0)$.

When $S=0$, the coupling of $\mathbf{n}$ to the rotational part of the shear cancels exactly with the convective term. The extensional part of the shear cannot now be balanced by the rotational part so that Φ winds as shown in Fig. 2(a). Such a scenario was first envisaged by de

Gennes[1] when flow alignment was not an option for nematics because λ was smaller than 1. Although the elastic energy increases as the ring count goes up, there are no elastic countertorques to stop the production of rings, and a stationary state is only achieved when the phase slips at the needle boundary. It is possible that the slip condition is mediated by a thin region next to the needle transforming to the nontilted smectic-*A* phase.

In conclusion, we observed two different regimes in freely suspended smectic-*C* liquid-crystal films under two-dimensional shear flow and we showed experimentally for the first time the existence of a coupling between the velocity field and the director orientation in the plane of the layers. One regime is flow alignment as predicted.[4] We measured for the first time the flow-alignment parameter of a smectic phase and found it to be large. In circular shear, a defect of strength $S=+1$ centered around the needle is required to observe flow alignment. The second regime is a phase-winding state that occurs when $S=0$. By measuring the rate of phase unwinding in this regime we determined $\gamma_1/K \approx 7.7\times 10^3$ sec/cm^2 for TB9A at $\approx 165\,°\mathrm{C}$. Our results demonstrate in an impressive manner the importance of topology on the selection of states for a two-dimensional anisotropic system in shear.

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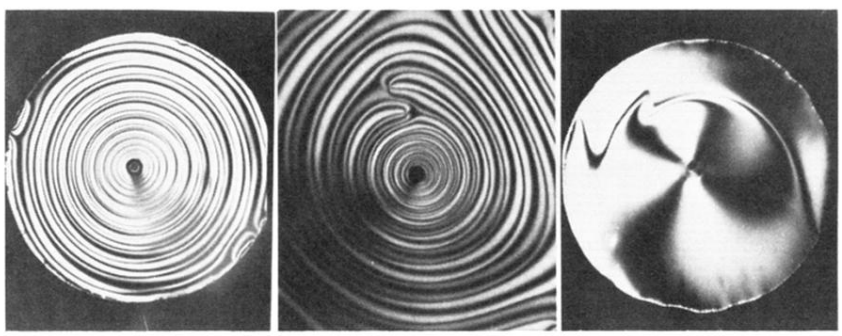

(a) (b) (c)

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