Electroconvection and Pattern Formation in a Suspended Smectic Film

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We have observed convective flow in a thin freely suspended film of smectic-*A* liquid crystal when a sufficiently large electric field is applied in the plane of the film. The convective flow neither reorients the director nor deforms the free surfaces of the film. Thus the film behaves as a two-dimensional isotropic fluid. We have studied the onset of this flow in two film holders, one which allows injection of charges from the electrodes into the film and one which does not. We present measurements of the threshold voltage and of the wavelength of the vortex pattern as a function of the frequency of the applied field, and briefly discuss the nature of the mechanisms driving the flow.

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In order to simplify the study of pattern formation in nonequilibrium nonlinear systems, it is useful to consider patterns which are periodic in only one spatial dimension.[1] Such patterns have been studied in Rayleigh-Bénard convection in a narrow box,[1,2] Taylor vortex flow,[1,3] and electroconvection in a nematic liquid crystal.[4-6] In each of these systems there is a range of parameters for which the fluid velocity forms a one-dimensional periodic pattern. In all cases the flow becomes three dimensional at higher values of the control parameters.

We have observed a novel type of electroconvection which displays simple patterns, but for which the flow is constrained to remain two dimensional over a substantial range of the control parameter. The convection takes place in the plane of a freely suspended film of a slightly impure smectic-*A* liquid crystal. The smectic *A* is a layered phase[4,5] which can very readily form suspended films[7] which are liquidlike in the plane of the film, but act like a soft solid in the perpendicular direction. The molecular axis, or director, of the smectic-*A* phase is oriented perpendicular to the plane of the film in the absence of flow. The electroconvective flow studied here does not reorient the director, indicating that the anisotropy of the smectic phase is not essential to the mechanism driving the flow: Similar flow has been observed in isotropic liquid films.[8,9] Furthermore, our material has a large positive dielectric anisotropy, so the Williams domain instabilities[4-6] are not expected. The anisotropy of the material contributes to our results only in that it causes the flow to be confined to the plane of the smectic layers and prevents deformation of the free surfaces of the film; the film can therefore be viewed as a two-dimensional isotropic liquid. This behavior is qualitatively different from that of suspended films of nematic or isotropic liquids[8-10] in which the flow is three dimensional and the free surfaces are deformed.

In our experiment the smectic film was supported at its edges by a rectangular frame. We used two frames with different electrode configurations to investigate the effect of injected charge; these are shown in Fig. 1. In the first, which we refer to as the *contact* holder, the sides of the film were supported by electrodes made of 15-μm tungsten wire. In the second, the *noncontact* holder, the sides were supported by 125-μm nylon threads which were separated from the wire electrodes by a 125-μm air gap. In both holders the ends of the film were supported by wipers made of coverslip glass. The film was drawn across the frame by one of the wipers which was motorized; its position defined the length L of the film. The width of the film d was mechanically variable in the range 0–3 mm. In the experiments described here, d was about 2 mm and the aspect ratio L/d was > 10.

The liquid crystal used was octylcyanobiphenyl (8CB) which is in the smectic-*A* phase at room temperature. It has a smectic layer spacing of 3.16 nm,[11] and a large

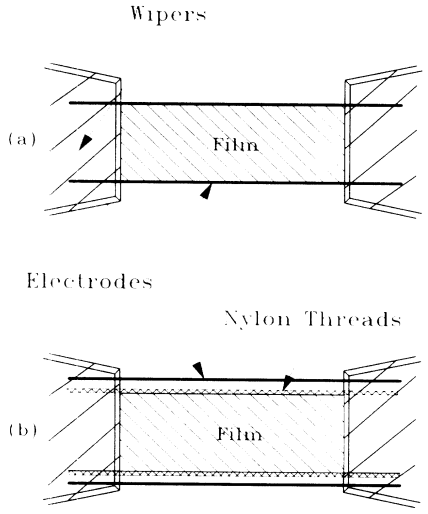

FIG. 1. The two types of film holder used: (a) the contact holder and (b) the noncontact holder.

FIG. 2. Photograph of the vortex pattern observed at dc in the contact holder. The width of the film was 2.16 ± 0.02 mm. Only a portion of the length of the film is shown.

positive dielectric anisotropy $\Delta\epsilon = \epsilon_{\parallel} - \epsilon_{\perp} \approx 8.5$, where $\epsilon_{\parallel}$ and $\epsilon_{\perp}$ are the dielectric constants parallel and perpendicular to the director, respectively. We doped the pure material as received from the manufacturer[12] with tetracyanoquinodimethane (TCNQ) at a concentration of 7.5 ± 0.2 mM/l to control the concentration and species of impurities. TCNQ has been shown to give strong, reversible unipolar injection of $TCNQ^-$ when used as a dopant in nematics.[13] The experiments were performed at room temperature on material that had been dried under vacuum.

We have observed convection in films from twenty to several hundred layers thick. The film is viewed with a low-power microscope in reflected white light. Films of this thickness show bright interference colors and the eye can pick out the change in color accompanying a change of one layer in thickness.[7] White light was therefore used to check the thickness uniformity of the film. The thickness itself was determined by measuring the power transmitted and reflected by the film at six wavelengths of a 10-mW Ar^+ laser beam polarized perpendicular to the plane of incidence. A fit to the reflectivity[14] as a function of the wavelength gives the thickness to ± 1 layer for films of less than about 100 layers, and to ± 2 layers for thicker films.

We allowed a small amount of dust to fall on the film and illuminated it using an expanded beam from the Ar^+ laser. The light scattered by the dust is visible as bright specks which were used to visualize the flow. Our results are not affected by the composition or number of the dust particles within reasonable limits.

In all cases described below, observation of the films between crossed polarizers shows no reorientation of the director due to the flow. Furthermore, observation of the film in reflected white light as described above shows no change in the number of smectic layers in the film and no distortion of the film surface at the onset of convection. This is in contrast to what occurs in freely suspended films of nematic or isotropic materials,[8-10] where the flow is accompanied by large changes in thickness and/or reorientation of the director.

When a dc voltage is applied to the contact holder, there is a well-defined critical voltage $V_c(0)$ above which the film convects in a stationary pattern of vortices of roughly square shape as shown in Fig. 2. Below $V_c(0)$ there is generally only a slow disorganized stirring of the film, with some vortexlike flow at the ends of the film holder.

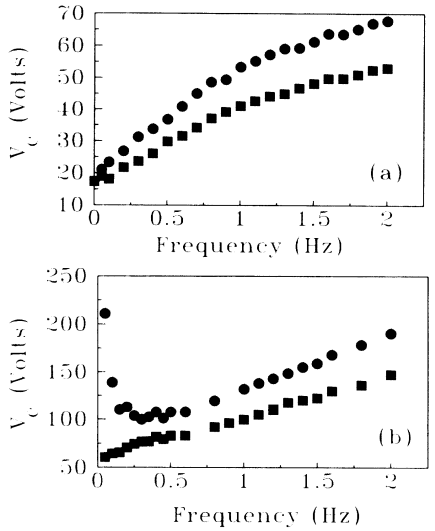

FIG. 3. The frequency dependence of the critical amplitude of an applied voltage for sinusoidal (●) and square (■) waves for (a) the contact holder, for a film 61 ± 1 layers thick and width $d = 2.10 \pm 0.02$ mm, and for (b) the noncontact holder, for a film 86 ± 1 layers thick and width $d = 1.89 \pm 0.03$ mm.

When a low-frequency sinusoidal or square-wave voltage is applied to the contact holder, we observe flow when $|V(t)| > V_c(f)$. The sense of rotation of the vortices reverses every half cycle with the polarity of the field. Figure 3(a) shows a plot of the critical amplitude $V_c(f)$ for the contact holder. V_c increases monotonically with frequency even at higher frequencies where the wavelength of the pattern changes (see below). The ratio $V_c^{\sin}/V_c^{sq}$ is approximately $4/\pi$ (the ratio of the first Fourier components) at all frequencies except very close to dc, where the ratio drops to 1. We observe no hysteresis in the critical voltage within the uncertainty in visually determining the onset of flow.

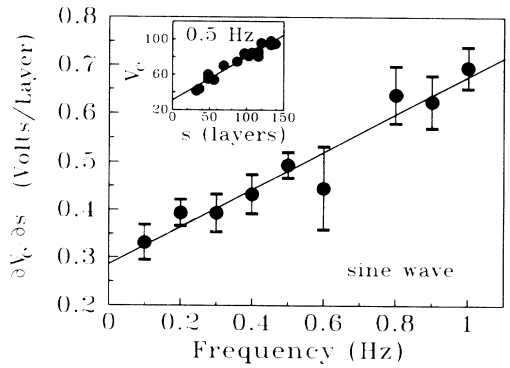

FIG. 4. The rate of change of V_c with film thickness, as a function of frequency for a sinusoidal applied voltage. Inset: The linearity of $V_c(s)$ at 0.5 Hz.

FIG. 5. The high-frequency pattern in the contact holder.

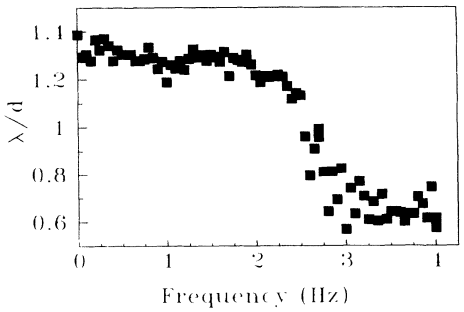

FIG. 6 The wavelength of the vortex pattern as a function of frequency for a film 128 ± 2 layers thick in the contact holder.

At a given frequency, $V_c(f)$ increases linearly with film thickness s for both wave forms, as also observed in nematic and isotropic films.[8-10] The quantity $\partial V_c^{\sin}/\partial s$ is shown in Fig. 4; it appears to increase linearly with frequency.

At zero frequency the vortex pattern has a wavelength λ which is proportional to the width of the film d: We find $\lambda/d = 1.30 \pm 0.05$. This is similar to the value measured in Ref. 9 for an isotropic film. This wavelength persists as the voltage is increased beyond $V_c(0)$. At higher frequencies the flow at the onset of convection exhibits a crossover to a new pattern like that shown in Fig. 5. As a function of frequency, the vortices at onset become narrower and then are replaced with pairs of smaller vortices which are localized along each electrode. Figure 6 shows λ/d at the onset of convection as a function of frequency. The new pattern emerges continuously out of the old beginning above a certain frequency. The pattern shown in Fig. 5 is seen only near onset; for voltages greater than $V_c(f)$, λ/d increases continuously until the pattern is similar to that seen at low frequency.

In order to investigate the effect of charge injection by electrochemical reactions at the electrodes, we compared the above behavior to that of a film in the noncontact holder. Figure 3(b) shows the critical amplitude of a voltage applied to such a film. In the noncontact holder the vortex motion occurs near the *zero crossings* of the applied voltage, not at the peaks. In this configuration, $V_c^{\sin}(f)$ passes through a minimum and tends to infinity as the frequency approaches zero. Under a square-wave driving voltage, one observes a transient burst of convection at the zero crossings of the voltage and $V_c^{sq}(f)$ decreases to a nonzero value as $f \rightarrow 0$. The behavior of $V_c(f)$ at low frequency is due to the capacitance of the air gap between the electrodes and the film.[4]

At higher frequencies, the pattern in the noncontact holder is similar to that in the contact case.

We now briefly consider the mechanisms which drive the flow. First, the fact that the director is not reoriented by the flow, and similar convective flows are observed in isotropic films,[8,10] suggest that the basic mechanism of the instability does not depend on the anisotropy of the smectic phase. Second, the V_c data suggest that there are two distinct processes occurring, one involving the injection of charges from the electrodes into the film, and one occurring in the absence of injection. The fact that sustained convection occurs at dc in the contact holder implies the presence of injection in that case.[13,15] In the absence of injection there can be no instability at dc; any preexisting charges in the film will assume their equilibrium screening configuration and no flow will occur. Conversely, the convection observed in the noncontact holder cannot involve injection. Here, the charge distribution is thrown out of equilibrium when the applied field is rapidly changed, leading to a redistribution of the charges which is assisted by flow. In the contact holder at nonzero frequency both processes are presumably present. Since the electrochemical reactions that provide the injected charges are slow,[5] injection is expected to be important only at low frequencies. Existing theories of electroconvection[5,9] seem unable to describe all of the behavior described here.

In conclusion, we have studied electroconvection in a freely suspended film of smectic-A liquid crystal and described the thickness and frequency dependence of the critical voltage. Two types of flow pattern are observed, depending on the frequency. A comparison of the behavior between contact and noncontact holders suggests that both injection and noninjection processes may contribute to the convective instability. The smectic layering forces the system to be highly two dimensional and provides particularly simple boundary conditions on the flow. This makes smectic films a potentially very attractive system for the study of one-dimensional patterns.

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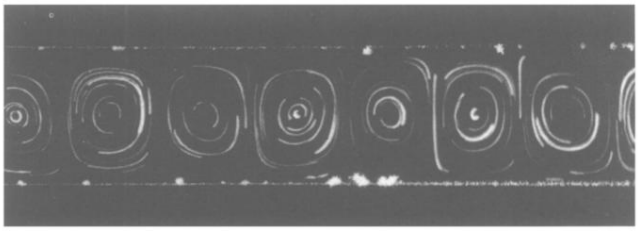
[15]We caution the reader that the usual practice of associating a simple conductivity with a liquid crystal is not justified in a contact cell under dc and very-low-frequency conditions. In this case the system is better thought of as an electrochemical cell. See Ref. 4, p. 191, and Ref. 5, Chap. 5.



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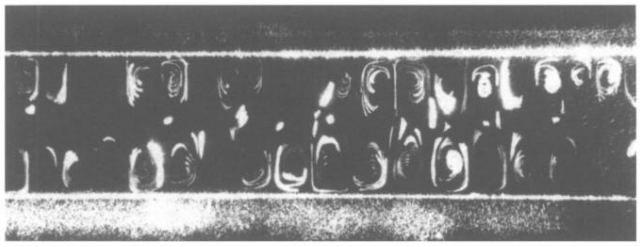

FIG. 5. The high-frequency pattern in the contact holder.