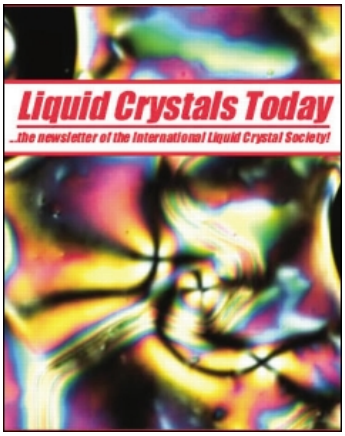

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Electroconvection in a Suspended Smectic Film

Stephen Morris[a]
[a] Department of Physics, University of California, Santa Barbara, CA, USA

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Electroconvection in a Suspended Smectic Film

from Stephen Morris, Department of Physics, University of California, Santa Barbara, CA, USA

Liquid crystals occupy an interesting and unique niche in the field of nonlinear, nonequilibrium fluid mechanics. When a fluid layer is driven away from equilibrium by an externally imposed stress, it will typically undergo a series of symmetry-breaking transitions, or "bifurcations", to flow states with more and more complex spatial and temporal patterns [1].

Eventually, the flow may become chaotic or turbulent. When the fluid is a liquid crystal, anisotropy plays a crucial and interesting role in determining the symmetry of the patterns and the nature of the turbulence. The most familiar pattern-forming instability in liquid crystals is electroconvection of the Carr-Helfrich type which occurs in certain nematics, for example MBBA [2]. It has the experimental advantages of small sample cells and the convenient control parameters of voltage and frequency.

These practical advantages are offset by the theoretical disadvantage of the complexity of nematic fluid mechanics. In this article we describe a new type of pattern-forming instability which is driven by a different mechanism and occurs in a freely suspended smectic A film [3-6]. Here, in contrast to the nematic case, the layered structure of the smectic and the geometry of the film conspire to make the fluid mechanics extremely simple, while preserving the experimental advantages of electroconvection. In addition the flow phenomena can be quite beautiful to watch.

When a smectic A liquid is drawn across an open support, it forms a robust suspended film, rather like a soap film, with the director perpendicular to the film. Unlike a soap film, however, the thickness of the smectic film cannot change continuously: the film is constrained to be an integer number of layers thick. The experimental set-up for electroconvection in a rectangular geometry is shown in ***Fig. 1***. Films of 8CB were made by drawing two glass wipers wetted with liquid crystal across an open frame made of very fine wires. The finished film was about 2mm wide and 25 mm long and was supported on the two short sides by the wipers, and on the two long sides by the wires. The thickness of the film is controlled by the wiper speed, and by the amount of material used to wet the frame. With a generous amount of material and quick drawing, one can produce films of nonuniform thickness in the submicron range. Each smectic layer is 3.16 nm thick in 8CB, so that these films are between a few dozen and a few hundred smectic layers thick. Such films appear very brightly coloured under a low power microscope using reflected white light; the colours are produced by interference between reflections from their top and bottom surfaces. The eye can easily observe the discontinuous changes of colour corresponding to changes in thickness, even steps of a single smectic layer.

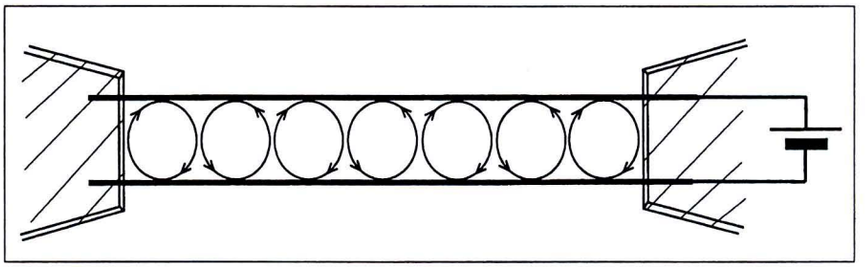

Fig. 1 The smectic film holder (rectangular geometry). The ends are formed by wipers made of coverslip glass, while the sides are 15 micron wires, about 2mm apart. An applied dc voltage drives steady convection in the plane of the film.

It was in such a nonuniform film that we first serendipitously observed electroconvection, with the thickness variations providing a crude sort of flow visualisation. The film breaks into convection when a sufficiently large dc voltage is applied to the wires. ***Fig. 2*** shows a snapshot of the spectacular swirling of the film colours just after the onset of convection. As the voltage is increased convection becomes more violent, and the film thickens and becomes more uniform as additional material is drawn onto the film from the edges.

Flow patterns in nonuniform films are amusing to watch, but irreproducible. Fortunately, it is possible to make films of perfectly uniform thickness. To make such a film, one must carefully restrict the amount of excess material wetting the frame, and draw the film slowly. When this is done, the film does not change its thickness while convecting, even past the onset of unsteady, turbulent flow. The thickness uniformity can be checked to within one smectic layer by monitoring the film colour using measurments of its reflectivity at various wavelengths [5]. The onset voltage and spatial wavelength of the vortex pattern above onset become sharply defined and reproducible in uniform films. We have studied convection in films as thin as 22 smectic layers.

Within the plane of the film the smectic A flows like an ordinary isotropic liquid, without director reorientation. Thus, without thickness inhomogeneities, it is not possible to visualise the vortices using the optical properties of the film itself. In order to study the flow we introduced a fine dust into the air around the film, some of which settles in the film. The dust is illuminated with an expanded laser beam: flow velocities can be quantitatively extracted from streak photos taken through the microscope [6]. Flows perpendicular to the film plane can only occur by the extremely slow "permeation" mechanism, which is negligible at the temperature of the experiment. In this way, the strongly anisotropic fluid mechanical properties of the smectic A force the film to behave as a simple two-dimensional liquid.

The following general features of the instability have been identified in uniform films under dc conditions: The film flows steadily in a vortex pattern just above a sharp critical voltage, which is the primary bifurcation for this system. A detailed study of the Fourier modes present in the flow pattern as a function of voltage, and measurements of the total current through the film show that the onset of convection is a non-hysteretic transition [6]. The flow velocity grows continuously from zero at the critical voltage, analogous to a second order phase transition. This is known as a "forward" bifurcation. The critical voltage increases linearly with the film thickness, but does not appear to scale in a simple way with the width of the film. The flow remains steady up to about five times the critical voltage, where it appears to become turbulent at a second bifurcation. This unsteady flow regime has not been systematically studied as yet. The width of the vortices in the flow pattern is independent of thickness and voltage in the steady flow regime and scales linearly with the width of the film.

No theory of this instability exists at present; we can only offer a qualitative explanation here. A discussion of the relevant electrohydrodynamical equations is presented in [6]. Since no director reorientation is involved in the smectic A film, the Carr-Helfrich mechanism [2], which depends on a director deformation to produce "charge focussing", cannot account for this instability. Furthermore, somewhat similar convection has been observed in much thicker nematic and isotropic films [7,8], although there it is accompanied by large changes in film thickness and other complications.

As in any type of electroconvection, the body force due to an electric field acting on regions of space charge is responsible for destabilising the rest state of the viscous fluid. In the liquid crystal, one has ionic impurities dissolved in a weak electrolyte. An examination of the basic equations [6] reveals two possible mechanisms for producing a spatial separation of the ions and hence regions of space charge. The first is due to the interaction of the electric field with the free surfaces of the film. Since the experiments all involve dc or very low frequencies, one must also consider direct charge injection by electrochemical reactions on the electrodes as a possible source of space charge. Such effects are notoriously difficult to control in experiments with less stable materials, such as MBBA.

Our pure 8CB was heavily doped with a charge transfer complex material in order to control the species of the carriers

Fig. 2 Convection just above onset in a rectangular film. The colours map the nonuniform film thickness which is advected by the flow. One can make out the steps in colour due to the smectic layering.

and to ensure that electrode effects did not lead to sample degradation. The electrical properties of the doped films did not change during the course of the experiment. Any additional charge due to reversible injection would be superimposed on that due to the surface diffusion layers. The experiments with ac fields and capacitively coupled electrodes suggest that, at least in those cases, injection is not necessary for the convection to occur; the charged diffusion layers alone are sufficient to drive the film.

The charged regions one expects from the geometry of the field around the film have the right sign to produce a charge "inversion" in the sense that charge builds up on the half of the film near the electrode with the same polarity in such a way that the resulting body forces tend to push the film into convection. The rest state of the film will become unstable to convection when these forces are sufficiently large that a velocity perturbation grows before it is damped by viscosity.

Many questions remain unanswered and many more experiments suggest themselves. Convection in a smectic C or C* film would be very interesting. Here one expects a coupling of the flow to the director. The nature of the regime of unsteady flow remains unexplored. ***Fig. 3*** shows an interesting variation in the electrode geometry in which the driving field is radial, a situation not often encountered in laboratory convection experiments. We plan to address these isues in future experiments.

Fig. 3 Convection in a nonuniformly thick film in an annular geometry, with a radial driving force

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