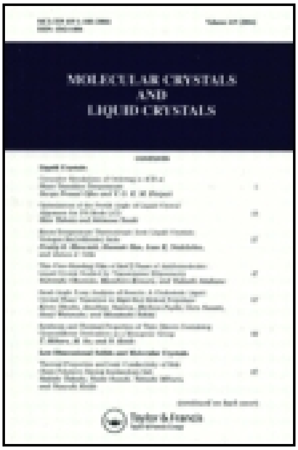

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Vertically Suspended Smectic Films with In-Plane Temperature Gradients

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We present a study of vertically suspended smectic films under the influence of temperature gradients in the film plane. It is shown that such gradients lead to the transport of smectic material, even against the action of the gravitation forces, from the hot to the cold film edge. In addition, we observe thermally driven convection in these films, and it is demonstrated that the surrounding air plays an essential role for this instability. We compare this result with convective patterns in soap films reported by Martin and Wu in *Phys. Rev. Lett.* **80** 1892 (98) and give some evidence that their interpretation has to be modified, the contact with surrounding air has to be taken into account.

Keywords: Smectic films; Rayleigh-Benard-convection; dissipative pattern formation

INTRODUCTION

It is well known that thermal gradients in a liquid layer can lead to different types of convective instabilities. Rayleigh-Benard convection (RBC) in isotropic liquids has been discovered already in the beginning of the previous century. Benard observed hexagonal vortex patterns in a thin liquid layer heated from below. Later, these patterns turned out to be driven by surface tension gradients (Benard-Marangoni instability). Rayleigh's theory from 1916 explained convection patterns in a liquid sandwiched between two solid plates with temperatures T_1 and T_2 of the upper and lower boundaries, resp. by a buoyancy driven mechanism. The latter effect is nowadays designated as Rayleigh-Benard convection. The dimensionless parameter which describes the onset of this instability is the Rayleigh number

$$R = \frac{g\beta}{\nu\kappa}\Theta h^3$$

where g is the acceleration of gravity, β is the expansion coefficient, $\rho = (1 - \beta\Theta)\rho_0$ the mass density, ν is the kinematic viscosity and κ is the thermal diffusivity. The critical Rayleigh number R_c depends upon the boundary conditions of the system and is 1708 for a liquid enclosed between two solid interfaces, but much lower if one of the surfaces is open. In nematic liquid crystals, the same mechanism is effective. In addition, the anisotropic material properties provide another instability mechanism, heat focussing, which is due to the anisotropic thermal conductivity. A lateral temperature modulation can build up as a consequence of spatial director tilt fluctuations, and the resulting flow field destabilizes the uniform director alignment. This may reduce the onset threshold of convection dramatically. In homeotropic cells the instability may occur even when the sample is heated from above.

While nematics have been extensively studied in the past (see e.g. Ahlers [1]), there is little known about thermal convection in smectics. Godfrey and van Winkle [2] have investigated surface tension driven convection in a freely suspended horizontal film, where the temperature gradient is in the film plane. They reported the onset of convection driven by the temperature dependence of the surface tension σ of the smectic material. Such an effect leads to convection vortices when the temperature gradients along the lateral film exceeds only a few degrees. The rotation sense in the rolls (in the film plane) is directed such that the flow of the smectic material at the edges with the largest temperature gradient runs from the cold to the hot plate. They interpret this observation with the assumption of a positive temperature slope $\alpha = d\sigma/dT > 0$. Another related experiment has been performed with vertical freely suspended soap films. Martin and Wu [3] have reported a study of convection in these films with a vertical temperature gradient in the film plane. They have found certain peculiarities of the observed patterns which they characterize as double diffusive convection. In this paper we investigate thermotropic smectic films in the geometry of Martin and Wu. We study the influence of thermal gradients on the onset of convective flow in the film. The experimental setup is described in the next section, a report of the experimental observations follows in the third section, and in the final section an interpretation and summary are given.

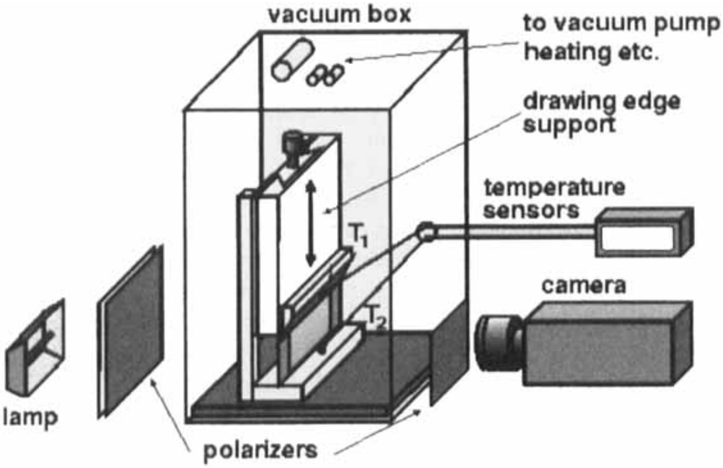


Figure 1: *Experimental setup consisting of film holder with temperature sensors, illumination and camera and the vacuum box.*

EXPERIMENT

Fig. 1 depicts the experimental geometry. The freely suspended film is drawn on a frame consisting of one movable and three fixed edges. The horizontal frame edges are mounted on copper thermoblocks which are connected to hot water thermostats. Their temperatures can be controlled separately with an accuracy of about 0.1 K. We have refrained from using electric heating to avoid possible stray fields in the film. If not stated otherwise we have used horizontal edges of 80 mm length to obtain a large aspect ratio. In a few other experiments 33 mm edges have been used. The lateral vertical frames are made from high-grade steel. The whole setup is enclosed in a transparent vacuum box. The film is illuminated with white or monochromatic light and is observed in transmission or reflexion by means of a video camera, connected to a HAMAMATSU 2400 video controller for contrast enhancement. The mesogenic material investigated was *FELIX16-100*, a commercial ferroelectric mixture from *Hoechst*, with phase sequence X -20°C SmC* 72°C SmA 85°C N 94-90° I.

In the beginning of the experiment, the horizontal holders are brought in contact and smectic LC material is spread between them. Then, the upper edge is carefully elevated by means of a micrometer set screw and the freely suspended film forms in the frame. The film is drawn with both horizontal edges at the same temperature in the SmC* phase. After the desired film height is adjusted, the upper and lower edges are heated to temperatures T_1 and T_2, respectively. By means of this procedure, we obtained films up to 30 mm height and 80mm width. The film thickness depends on the drawing speed, temperature, and viscosity of the material (usually of the order of 10-50 layers). After the film is drawn, the vacuum box is sealed and the pressure can be adjusted between atmospheric pressure and about 10 $mbar$.

The Rayleigh number gives a rough estimate at which film heights thermal convection can be expected. For typical LC parameters, $\beta=10^{-3}$, $\nu=10^{-4}m^2/s$, $\kappa \approx 8 \cdot 10^{-8}m^2/s$ ([4]) films height h and temperature difference θ we obtain $R \approx 1.2 \cdot 10^3 K^{-1}cm^{-3} \cdot h^3\,\theta$. When θ is $10K$ and $h > 5mm$, one reaches values of R above 2000. However, this simple calculation is not adequate to determine thresholds in the free standing films since it does not consider the finite film thickness (see last section). The Prandtl number ν/κ is approximately 10^3. As will be demonstrated, air convection plays an important role for the observations. For an air layer of height h, the Rayleigh number is approximately $110K^{-1}cm^{-3} \cdot h^3\,\theta$, where we have assumed $\lambda_{air}=2.6{\cdot}10^{-2}Wm^{-1}K^{-1}$, $\rho_{air}=1.3\ kgm^{-3}$, $c_p=1000Jkg^{-1}K^{-1}$ and $\nu_{air}=1.5{\cdot}10^{-5}m^2/s$.

OBSERVATIONS

After the film is drawn, its thickness is in general not homogeneous, but the thickness inhomogeneities gradually disappear. However, when a temperature gradient of a few K is established between the horizontal edges, additional smectic layers start to climb from the hot edge to the cold edge, gradually thickening the film (Figs. 2,3).

Initially, the film is about 80 nm thick, but in the bottom part, regions of continuously increasing film thickness appear. This is de-

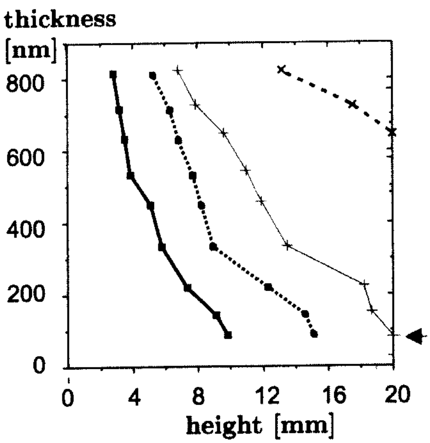


Figure 2: *Vertical thickness profiles determined from interference colors in reflexion. [5] FELIX16 film with $T_2 = 70^0C$, T_1 room temperature. The film area is 33mm×20mm. Profiles measured at t=315s (solid thick) 555s (dotted), 885s (solid thin) and 1365s (dashed line) after heating was started.*

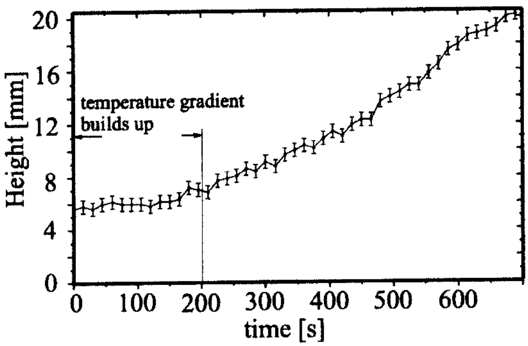
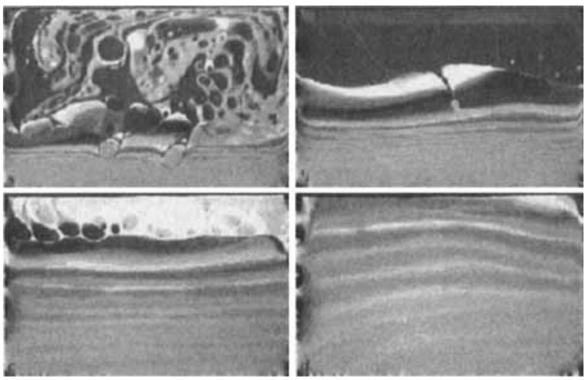


Figure 3: *Propagation of the first thickness step. When the temperature gradient has built up, the propagation velocity is approximately constant $30\mu m/s$. Bottom: corresponding reflexion images*



t=315s 555s 815s 1365s

See Color Plate XXIII at the back of this issue.

picted in Fig. 2. The thickness profiles have been determined from crystallographic interference colors [5]. One finds a continuous progression of the thickness front. The film thickness after 20 minutes was $1.17 \pm 0.1 \mu m$.

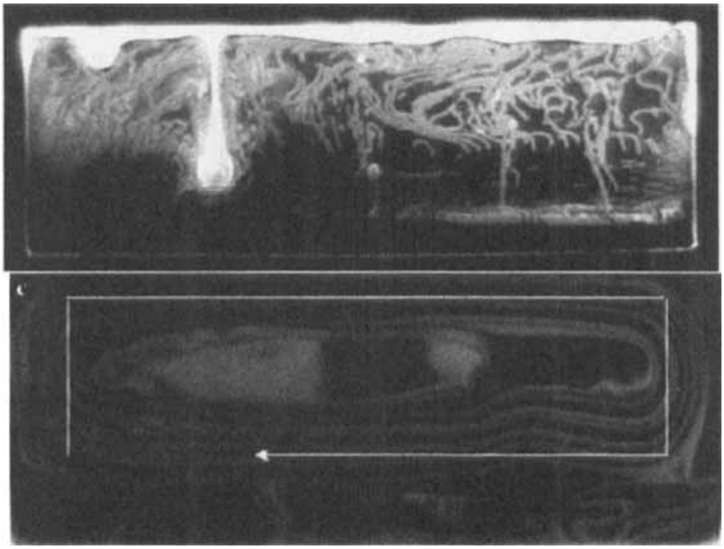


Figure 4: *Top: Instability of the 'bulge' of excess material at the upper edge. FELIX16 film at* $T_1 = 50.0^oC, T_2 = 82.0^oC$*, film height h=11 mm, (transmission image with crossed polarizers). Bottom: large single convection roll,* $T_1 = 30.0^oC, T_2 = 50.0^oC$*, p= 800 mbar.*

The excess material collects in a bulge at the upper film edge, and undergoes a sort of Rayleigh-Taylor instability. The interface undulates and excess material drops down the film again (Fig. 4 top). This periodic process causes strong flow in the film plane and disturbs all other convection processes. Therefore, it was necessary to remove excess material carefully by means of a capillary. The resulting films are fairly uniform (≈10% thickness variation). Since the absolute interference order can be counted while the material climbs up, one can determine the local thickness from the reflective color within 5% accuracy. In the quite uniformly thick films, a convective motion sets in when the temperature gradient exceeds ≈ 10 K. Often, the formation of one single convection roll is found which moves along the frame, parallel (in opposite directions resp.) to the thermostated edges (Fig. 4 bottom). In homogeneously thick films, where one cannot observe the flow of islands on the film, it is detected walls in the c-director structure created by the flow field (observed with crossed polarizers in transmission). When the aspect ratio is kept very low (33 mm by 20 mm frame), we often observe two counterrotating rolls. If the horizontal thickness step in the film has not yet reached the upper edge (Fig. 3), convection remains confined in the homogeneous region.

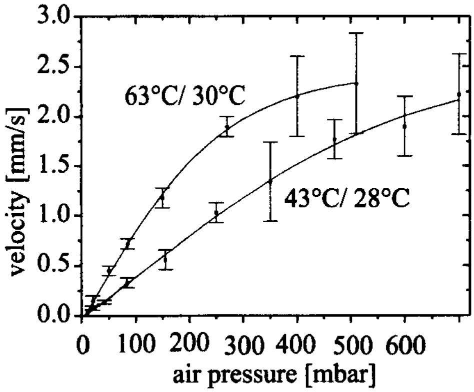


Figure 5: *Flow velocity in the FELIX16 film in dependence of the surrounding air pressure. The observed convection in the film can be fully suppressed by reducing the air pressure (solid lines guide the eye).*

The measurement of the flow velocity of convection rolls as a function of the air pressure in the vacuum chamber makes clear that the flow is driven by air convection between the upper and lower thermoblocks. Figure 5 shows flow velocities for two thermal gradients as a function of pressure p.

When the air flow is conducted by an array of glass sheets placed about 1 mm next to the film, the convection rolls in the film adopt the arrangement of the compartments. The transmission image of a film in such a geometry is shown in Fig. 6. Glass sheets (100μm thickness) divide the front in compartments of approximately $18 \times 9\,mm$. The temperatures are $T_1 = 48°C$, $T_2 = 58°C$. Since the sheets themselves are not in contact with the film, but the compartment structure is reflected in the convection pattern of the film, we assume that it is mediated by air flow near the film.

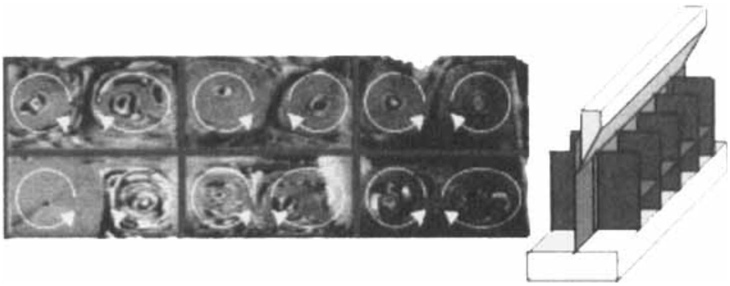


Figure 6: *Vortex flow in a FELIX16 film when the air convection is conducted by a glass plate array (shown schematically at the right). The position of the plates is marked by horizontal and vertical lines. The flow field in the film is visible by advected SmC* domain walls. A arrows indicate the reorientation sense of the vortices.*

DISCUSSION and SUMMARY

The migration of LC material from the hot to the cold plate (Figs. 2,3) is not only characteristic for vertical films, it has also been observed before by Godfrey and van Winkle [2] in horizontal films when the temperature gradient exceeded about 5 K/cm. The physical reason is obviously the same as in our experiment. The mechanism driving this effect must overcome the gravitational forces and we can estimate the necessary energy to lift the excess layers to be much larger than $0.1 J/mol$. This is a small fraction (about 1/5000) of the intermolecular cohesion in typical liquids. Thermal expansion of the LC material, about $10^{-3}/K$, leads to an increase of intermolecular distances ($\approx 3 \cdot 10^{-4}/K$) with temperature and consequently lowers cohesive intermolecular forces. This explains the observed migration of smectic material against the temperature gradient. Since the surface energy is practically uninfluenced by the thickening of the film, only weak gravitational forces counteract the process. The process stops only after the reservoir at the hot film edge is exhausted.

Surface tension driven convection observed previously in horizontal films [2] could not be detected in the vertical film geometry. However, we observe a similar convection mode when the films are brought in horizontal orientation. In FELIX16, the temperature slope $d\sigma/dT$ is negative [6], and the direction of thermocapillary flow is that of ordinary liquids.

The convective flow patterns observed in our films are obviously coupled to the air convection between the hot and cold edges of the setup. The onset of convective rolls and the flow velocity both depend upon ambient gas pressure. The observation of a single elongated roll (Fig. 4 bottom) for films with large aspect ratio is not unusual. Such single rolls occur in RBC e. g. when the thermal conductivity of the plates is small compared to that of the fluid. We have not controlled the upper boundary conditions for air convection in our experiment, air flow is practically driven by a lateral heating, and the convection cell is not bounded on top. The critical Rayleigh number for air in such a geometry is probably around 600 and is easily reached when the gap between the heated edges is larger than 1 cm and the temperature difference exceeds $5K$. But still, the Rayleigh number calculated for the liquid crystal at given film heights and temperature gradients is larger than for the gas. Therfore, it seems to be difficult to develop a simple model of the convection mechanism. Since the convection patterns can be completely suppressed by evacuating the chamber, it follows that the observed structures are not related to an 'intrinsic' RBC mechanism that can be treated without considering thermal contact with ambient air. There is evidence (Fig. 6) that the convection of air is directly involved in the formation of convection vortices in the film.

In any case, presence of air is necessary to provide the ground state of a linear temperature gradient in the film plane: in the non-convecting state, the vertical thermal gradient in the surrounding air is rather linear, and the film is in thermal equilibrium with the air everywhere at the film surface. In absence of air, or at low pressures, heat loss by irradiation at the film surfaces is dominating over heat diffusion in the film plane (see [2]) and all temperature gradients in the film plane are restricted to small zones near the film edges.

The geometry considered in this study is in many respects comparable to that chosen in [3] and it seems that some conclusions can be adopted. There is strong evidence that the same mechanism is responsible for the patterns observed in the experiment of Martin and Wu, where soap films were exposed to a temperature gradient in air under atmospheric pressure. Their model for the description of the observed patterns seems therefore inappropriate and should be revised.

We note in addition that the influence of surrounding air on the threshold of electrically driven convection in thin smectic films has been studied by Langer [7]. A slight decrease of the onset voltage with ambient pressure has also been found in that system.

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