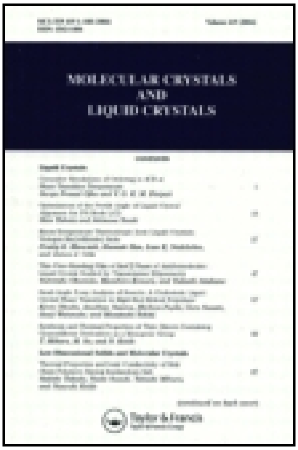

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Electroconvection in Freely Suspended Smectic C* Films

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Electrohydrodynamic convection (EC) of freely suspended smectic films is investigated by means of polarizing microscopy. The EC patterns observed in our ferroelectric S_C^* films belong to an EC type qualitatively different from the vortex convection described previously in smectic A films by Morris et al. and in smectic C films by Becker et al. It is driven by a bulk charge separation mechanism analogous to the Carr-Helfrich-instability in nematic cells. The threshold voltage is close to zero and film thickness independent. We demonstrate that the convective flow patterns can be confined by domain walls, which is not possible in the vortex mode.

Keywords: Electroconvection; Ferroelectricity; Smectic films

INTRODUCTION

Freely suspended films are useful systems to study the dynamics of smectic liquid crystals. Thin films can be used to investigate two-dimensional convective flow resulting from the interaction of an applied electric field with inhomogeneous space charge densities. Two electroconvection mechanisms have been conceived so far. One instability is driven by the interaction of the electric field with space charges at the free surfaces of the films [1, 2, 3, 4, 5, 6]. This surface charge mechanism (vortex mode, VM) has been intensively studied in smectic A films [2, 3, 4], and it has been observed also in tilted smectic phases [7, 8, 9].

The second type is a bulk charge separation effect due to a conductivity anisotropy, analogous to the Carr-Helfrich mechanism well known from nematics in sandwich cells. It has been proposed by Ried et al. [10] to be a possible mechanism also in smectic films with in-plane anisotropy, but so far it has not been unambiguously identified in experiments. We will denote it by CH here. In a previous paper [9] we have shown that the EC patterns in ferroelectric S_C^* are qualitatively different from those observed in the S_C and S_A films. The onset threshold voltage is very low. Near the threshold, convection is practically film thickness independent. This was interpreted as an evidence for the CH instability in the S_C^* material, but an unambiguous identification of the convection type is not straightforward. Although the VM and CH instabilities are driven by different effects, there is no general criterion to distinguish them from their optical appearance. In both cases, a row of flow vortices with alternating sense of rotation is characteristic. In S_A films, the CH mechanism can be excluded because the conductivity is in-plane isotropic. In S_C films studied so far, one can conclude from the experimental images [7, 8] that a VM is observed. The in-plane conductivity anisotropy necessary for CH is averaged out across the vortex.

The experimental results in S_C films cannot be generalized because in the materials investigated (a disubstituted phenylbenzoate in [7] and n-OPPy in [8, 9] and in this paper) the dielectric anisotropy $\delta\epsilon$ in the film plane has not been determined. A positive $\delta\epsilon$ would destabilize the ground state of the c-director field and prevent CH convection similar as in nematic cells. For the ferroelectric material, we have checked that $\delta\epsilon$ is positive. However, the c-director orientation in the S_C^* films is stabilized by polar interactions with the electric field [9]. Therefore, the prerequisites for CH are given independent of the sign of $\delta\epsilon$. In this study we demonstrate the instability is driven by a CH type bulk charge mechanism at onset. At higher electric fields both the surface charge mechanism and the bulk effect are involved in the convection.

EXPERIMENTAL SETUP

Our experiments are performed on smectic films freely suspended between two movable and two fixed holders (Figure 1). Films of controlled homogeneous thickness are drawn in the smectic A phase. The film width is given by the distance between the fixed holders which is about 6 mm, while the film length can be varied from 0 to 7 mm. The film holders are contained in a heating stage and the films can be stabilized at a given temperature to $\pm 0.1\,K$. Observations of the films are carried out with polarized light using a CARL ZEISS JENA NU2 reflection polarizing microscope. The film thickness is measured by means of reflection spectroscopy [11].

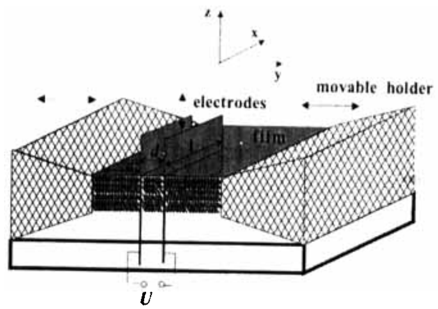

Figure 1: Schematic drawing of the film holders with micro electrodes pierced into the film (only the movable holders are sketched).

The electric fields are provided by two parallel micro electrode plates pierced through the film. Electrode distances of d of $500\,\mu m$ and $250\,\mu m$, have been used, the electrode length l is about $2.5\,mm$. The transmission images are recorded with a HAMAMATSU CCD camera and C2400 camera controller for contrast enhancement.

We have studied the chiral mixture FELIX 016-100 (Hoechst) and a mixture of achiral *5-n-Octyl-2-(4-n-alkyloxyphenyl)-pyrimidines* (n-OPPy) [9]. The samples show the following phase sequences:

FELIX 016-100:	$< -20°C$	S_C^*	$72°C$	S_A	$85°C$	Ch	$94°C$	I
n-OPPy:	$< -10°C$	S_C	$50.5°C$	S_A	$53°C$	N	$68°C$	I

EXPERIMENTAL OBSERVATIONS

When a sufficiently large DC voltage is applied to the electrodes, a periodic flow pattern consisting of pairs of alternating vortices sets in. With increasing voltage, the vortex rotation velocity increases. Due to the optical anisotropy of the tilted smectic phases, convective flow can be identified from the motion of optical textures (Figure 2). In addition, the flow field can be visualized in films of inhomogeneous thickness when islands of excessive smectic layers advected with the flow. Of course, homogeneous films are preferable for quantitative measurements. In S_A films convective flow has been determined from measurements of the electric current [5] or by means of small probe particles suspended on the film [1, 2]. We used this probe particle method to measure small vortex velocities just above the onset of convection. The rotation frequency of the vortices was determined from trajectories of particles.

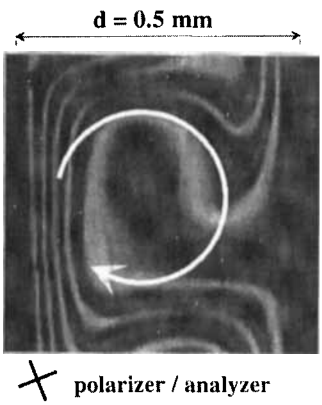

Figure 2: Instant image of vortex flow electroconvection in a smectic C film at a DC voltage $U = 60\,V$ ($U_C = 10.75\,V$). The film thickness s is $2493\,nm$. The white arrow visualizes the vortex flow field. The c-director orientation changes by 45° between adjacent bright and dark areas.

Smectic C

In the S_C films investigated, the threshold field for the formation of convective vortices is of the order of several kV/m. It increases linearly with film thickness. The measured vortex frequencies are consistent with the assumption of a forward bifurcation, with the appropriate control parameter $\epsilon = \left(\frac{U}{U_C}\right)^2 - 1$ and threshold voltage U_C where the amplitude (vortex frequency f_v) of the pattern above the bifurcation increases proportional to $\sqrt{\epsilon}$ [2]. A fit of $f_v(U) \propto \sqrt{\epsilon}$ gives the critical voltage U_C where the convection sets in. The threshold voltages obtained [9] are in qualitative agreement with the results for S_A films presented in [1, 2, 6] when the different conductivities σ are taken into consideration.

Smectic C^*

The essential new feature in our S_C^* films is the interaction of the spontaneous polarization $\mathbf{P_S}$ with the electric field $\mathbf{E}$. When DC Voltage is applied, $\mathbf{P_S}$ tends to align parallel to $\mathbf{E}$. In thin films the critical threshold field for polar alignment is practically zero.

At DC excitation, the pure electroconvection effects can be studied. When the excitation field is AC, a superposition of ferroelectric switching (c-director reorientation) and convective flow occurs. This switching behaviour will be analyzed in detail elsewhere. In the following description of the electroconvection effects only DC excitation fields are considered.

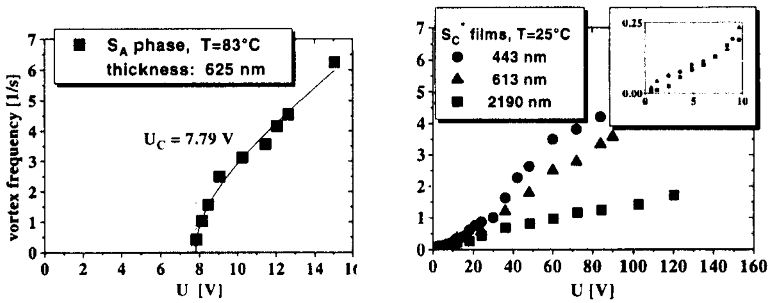

Figure 3: Measured vortex frequencies for smectic A films (left) and smectic C* films (right) of FELIX-016/100. The small box box in the upper right corner of the S_C^* plot shows the results for the low voltage range between zero and $10\,V$.

The c-director initially reorients parallel to the electrodes and remains nearly fixed there. The torque of the convective flow field on the c-director is much smaller than that excerted by **E** on $\mathbf{P_S}$. However, one finds a small deflection of the c-director with the sense of the vortex rotation. The vortex pattern can be visualized with crossed polarizers when a small angle between polarizer and electrode is adjusted. As seen in Fig. 4, vortices rotating in opposite directions are recognized from brightnes changes in the texture. The reflexion of the film is minimum when the c-director is along one of the polarizers, therefore a small deflection of the c-director from the ground state towards the polarizer axis darkens the image, an opposite deflection brightens it. Thus, we can extract the sense of rotation directly from the textures without probe particles.

The low critical voltages for ferroelectric smectic C^* cannot be explained with the VM model even if we consider conductivity differences [9]. In order to compare the two convection types in one and the same material, we have studied an individual FELIX-016/100 sample in the S_A and S_C^* phases, respectively. The results are shown in Fig. 3. Even in films with a thickness of several micrometers, the threshold voltage is close to zero in the low temperature S_C^* phase while in the high temperature S_A phase of the same films U_C lies in the range typical for VM. For films with a thickness much smaller than the pitch (which is about 10 μm in FELIX-016/100), the onset of convection in the S_C^* phase is thickness independent, while it has the characteristic linear thickness dependence in S_A.

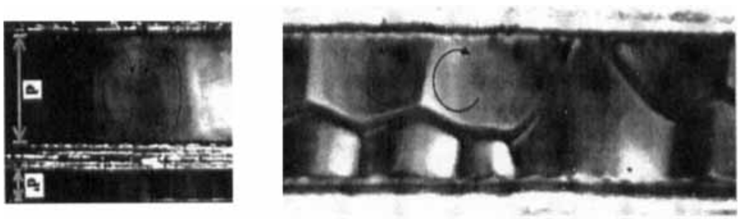

Figure 4: DC Electroconvection in S_C^* films, brightness differences of vortices rotating in opposite directions (polarizers as in Fig. 2) Left image: Large scale flow pattern in a monodomain film with electrode distance d, smaller patterns at the bottom between electrode and film holder (distance d'). Right image: Small-scale vortices in individual domains of a polydomain film between domain wall and electrodes.

Another key observation is presented in Fig. 4. We show a film which is separated into domains by inversion walls. One wall (2π) in the left part of the image extends rather parallel to the electrodes, then it approaches the upper electrode, another wall starts at that electrode in the right part. The film thickness is homogeneous, only the c-director orientation changes in the walls. The electroconvection patterns observed in this film do not extend across the electrode gap as in monodomain films. Instead, we find separate small scale vortices (dark and bright fields in the texture) in the individual domains between the walls and the electrodes. The vortex flow field deforms the domain wall, but the vortex size is determined by the distance between the wall and the adjacent electrode. With increasing voltage and flow velocity, the deformation increases and the domain walls become unstable. At high voltages, only large-scale flow patterns are observed. Obviously, the walls form some kind of "soft boundary" for the convective flow. This observation indicates that the convection mechanism depends on bulk charges. The surface charge VM model implies an inhomogenous charge distribution near the electrodes [2, 3, 4], it cannot explain the small scale vortices observed. Inversion walls should not act as a barrier for the convective flow, the VM is quite insensitive to the c-director field. Within the CH model proposed by Ried et. al. [10] one can understand that the convective vortices are trapped in domains of homogeneous orientation. The instability is driven by a lateral bulk charge separation in the film, it requires a uniform ground state of the c-director where the conductivity perpendicular to the electric field is larger than along **E**. This condition is fulfilled in each domain, the charge modulations in the individual domains are mutually independent. Interactions of vortices in different domains are mediated by the deformation of the domain wall.

On the other hand, the pure Carr-Helfrich mechanism described in [10] is not sufficient to understand convection in smectic C^* films in general, as it neglects any effect of surface charges. Morris et al. [2] have pointed out that in thin films the Debye screening length is of the order of the film thickness. This means that in our films no clear distinction between surface and bulk charges can actually be made. In fact, at higher voltages, the flow velocity becomes film thickness dependent (Fig. 3), suggesting that also contributions from the surface charge instability take effect. Therefore, we assume that the convective flow is driven by a combination of both the surface charge VM and the Carr-Helfrich effect.

CONCLUSIONS

In S_C films we observe a vortex flow similar to the S_A case. The c-director field is twisted and advected with the hydrodynamic flow field. The threshold voltage U_C depends linearly on the film thickness, indicating a surface charge mechanism as described by [1, 4]. In S_C^* films, we observe a different electroconvection mechanism. U_C is below our experimental resolution. No significant thickness dependence is found near onset for film thicknesses smaller than the pitch. A direct comparison of convection in the smectic A and C* phases of the same smectic material reveals a dramatic drop of the threshold voltage in the smectic C* phase. Unambiguous evidence against VM near onset in the S_C^* phase comes from the observation of small-scale vortices in multidomain films. Since the domain walls do not change any film properties except the rotation of the c-director, the VM effect should not be influenced by domain walls. Instead, the vortex size scales with the size of the domains, which can only be understood within a CH like convection model as proposed by Ried et al. [10]. From these results we conclude that we observe a bulk charge driven CH type instability in the smectic C^* films at low driving fields. With increasing voltage, the convection becomes thickness dependent, suggesting that the convective flow is driven by a combination of both the surface charge VM and the Carr-Helfrich effect. Therfeore, a generalized theory is necessary, which should include both the CH and VM contributions. When the film thickness is comparable to the pitch, the theoretical treatment becomes even more complicated, as the threshold field for unwinding the helix has to be considered as well.

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