

## Spontaneous transition from chevron to striped texture of a planar smectic- $C^*$ liquid crystal

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(Received 28 October 1991)

We observed a spontaneous transition from a vertical chevron texture to a striped texture (striped “bookshelf” texture) in a surface-stabilized film of ferroelectric liquid crystal mixture (ZLI 4237-000, from E. Merck). The transition takes place gradually, at temperatures more than  $20^\circ\text{C}$  below the smectic- $A$ –smectic- $C^*$  transition. The layers in the stripe texture are perpendicular to the plate, but in successive stripes, the orientation of the layer normal alternates by  $\pm 23^\circ$ . The polarization is perpendicular to the film and alternates between up and down, while the director orientation is in plane and alternates by  $\pm 5^\circ$ . The width of the stripes was typically  $50\ \mu\text{m}$  for samples of about  $5\ \mu\text{m}$  thickness. The area of the kink interfaces between the stripes is accordingly an order of magnitude smaller than in chevron textures, which energetically favors the stripe texture. The formation of this texture is slow because it requires sliding of the layers along the surface. The observed transition can be of importance for display technology because it may enable one to obtain a uniform bookshelf texture.

PACS number(s): 61.30.Eb, 64.70.Md, 61.30.Gd

### I. INTRODUCTION

The preferred texture for electro-optic devices using surface-stabilized ferroelectric smectic films is the homogeneous striped or “bookshelf” texture. Accordingly, since the first paper on an electro-optic device using surface-stabilized liquid crystals was presented [1], efforts have been made to obtain this texture. It can be readily obtained in smectic- $A$  (Sm- $A$ ) phases but, on cooling to smectic  $C$  (Sm- $C$ ), the texture changes because the layer distance decreases. The decrease causes an undulation instability. The most common resulting texture is a (vertical) chevron texture with a kink of the layers at the mid-plane of the film [2,3]. The chevron texture and the presence of domains with opposite kink orientation are disadvantages. The homogeneity of displays is affected and the contrast is reduced because the layers are tilted against the surface normal. A number of attempts were made to overcome these difficulties. With proper surface treatments, fairly homogeneous chevron textures that have bistability can be made [4], but the contrasts are still not optimal. Good progress in obtaining the bookshelf texture was made by the use of mixtures containing naphthalene derivatives [5]. These mixtures give a very small layer tilt angle at room temperatures ( $\alpha = 4^\circ$ , quasi-bookshelf texture). Recently, it was observed that the application of strong electric fields of low frequencies may induce a transition from the vertical chevron texture to a striped bookshelf texture [6–8] (horizontal chevron texture). In these samples the layers are perpendicular to the surface, zigzagging in the horizontal plane. The contrast and the homogeneity of displays with this texture were significantly better than in samples with the normal chevron texture.

We observed with a different material the spontaneous transformation from a vertical chevron texture to a striped bookshelf texture. The transformation was observed on a surface stabilized ferroelectric-liquid-

crystalline film without application of an electric field. In the following, we describe the optical and electromechanical properties of this texture and analyze its microscopic structure.

### II. EXPERIMENT

The studied material is a ferroelectric liquid crystal mixture from E. Merck (ZLI-4237-000). It has a smectic- $C^*$  (Sm- $C^*$ ) phase between  $-20^\circ\text{C}$  and  $63^\circ\text{C}$  and a long helical pitch ( $> 40\ \mu\text{m}$ ) over the whole Sm- $C^*$  range. The director tilt angle and the spontaneous polarization of the material increase with decreasing temperature from zero at the Sm- $A$ –Sm- $C^*$  transition to a tilt angle of the director of about  $26^\circ$  and a spontaneous polarization of about  $7\ \text{nC}/\text{cm}^2$  at room temperature. Samples of  $5\ \mu\text{m}$  thickness were prepared by using polystyrene spheres as spacers. The sample cells consisted of two indium tin oxide-coated glass plates. The conducting inner surfaces were also coated with polyimide and rubbed parallel ( $Z$  direction). The samples were filled with the material at an elevated temperature ( $> 79^\circ\text{C}$ ) in the isotropic phase. The samples were then slowly cooled through the cholesteric range, while periodically shearing the cover plates perpendicular to the rubbing direction at a frequency of 3 Hz and with an amplitude of about 0.1 mm. After reaching the Sm- $A$  phase (below  $72^\circ\text{C}$ ), the shearing was halted. In this way fairly homogeneous textures with few focal conic domains were obtained. The textures in the measurement cell and their changes in temperature and under various electric fields were observed with a polarizing microscope mounted to the heating stage. The electromechanical responses were detected by three small accelerometers (2.3 g each, from Bruel & Kjaer) fixed to the cover plate so that they measured the acceleration in three orthogonal directions. The cell was placed horizontally in a heating stage with the bottom plate fixed to the oven and the top plate free to move. The accuracy of temperature regulation was

better than  $0.03^\circ\text{C}$ . The sample was electrically driven by the output of the internal oscillator of a lock-in amplifier (EG&G Princeton Applied Research, Model 5209) and the signals of the accelerometer due to mechanical vibrations of the cover plate were filtered and measured separately by the same lock-in amplifier. At a given temperature the responses in the three directions were measured as a function of frequency and voltage. The consistency of the data was checked by repeating the measurements in the first direction before the next temperature step. The field and frequency sweeps and the data collection were made automatically under control of a computer.

### III. EXPERIMENTAL RESULTS

#### A. Microscopic studies

For detailed microscopic studies, the cell was removed after the electromechanical measurements and after realignment, examined with a Leitz polarizing microscope using a Mettler heating stage. We observed with typical samples on cooling from Sm-*A* to Sm-*C*\* (at a rate of about  $2^\circ\text{C}/\text{min}$ ) below  $55^\circ\text{C}$  the formation of zigzag domains that are characteristic for vertical chevron textures [2,3]. The domain sizes were on the order of  $1\text{ mm}^2$ . Below  $40^\circ\text{C}$  stripes formed gradually parallel to the rubbing direction. In repeating this cycle, it was found that the stripes fade on heating above  $50^\circ\text{C}$ , but they reappeared on cooling below  $40^\circ\text{C}$ .

At room temperature the sample also relaxed into a quite regular striped texture which, after a couple of days, covered the whole sample area. At this stage zigzag domains could no longer be observed. The width of the stripes was typically  $50\text{ }\mu\text{m}$  but it varied over the sample area and increased slightly in time; after a week it reached equilibrium with stripes as wide as  $100\text{ }\mu\text{m}$  in some areas. Reheating to the isotropic phase and repeating the alignment procedure and the cooling cycle gave again zigzag domains that persisted down to room temperature but within a few days the stripes formed again and the zigzag domains disappeared (Fig. 1). The formation of the stripes can also be induced by a small periodic shear parallel to the rubbing direction or by application of a small ac voltage.

The experiments show that at low temperature the striped texture is the more stable and gradually replaces the chevron texture which appears to be stable at elevated temperatures, but the zigzag domains do not reform on heating from the striped texture and they can be recovered only when the sample is heated above the Sm-*C*\*-Sm-*A* phase transition.

The optical contrast between the stripes with crossed polarizers is largest when the polarization makes an angle of  $\pm 5^\circ$  or  $\pm 85^\circ$  to the rubbing direction. Under this orientation, dark and bright stripes alternate. The dark and bright stripes interchanged when the sample was rotated by  $10^\circ$  in the proper direction. Applying dc voltages we found that every second stripe remained almost unchanged, while the others changed more strongly and became similar to the first ones. Reversing the polarity of

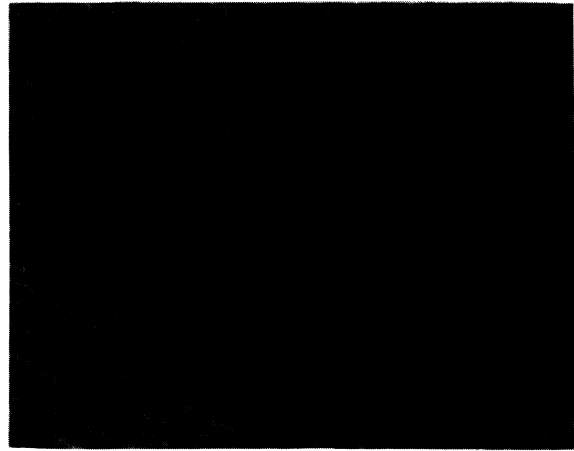


FIG. 1. Spontaneously formed striped texture, surface-stabilized Sm-*C*\* film (ZLI 4237-000, E. Merck);  $5\text{ }\mu\text{m}$  thick,  $T = 28^\circ\text{C}$ , stripe width  $\sim 50\text{ }\mu\text{m}$ , crossed polarizers. The stripes are parallel to the rubbing direction and perpendicular to the shear, polarization angle  $5^\circ$  to stripes.

the field interchanged the responses of the stripes. This observation shows that the polarization of the stripes alternates between parallel and antiparallel to the surface normal.

#### B. Electromechanical responses

As mentioned, we studied the mechanical responses to alternating electrical fields in three different directions by measuring the acceleration of the cover plate in three orthogonal directions (*X* normal to the plate, i.e., parallel to the field, *Y* in plane of the plate normal to the rubbing direction, *Z* in plane parallel to the rubbing direction). The response depends on temperature and on the texture. The linear effect was negligible in the *X* and *Z* directions except for weak resonancelike response around  $1.1\text{ kHz}$ . In contrast, the *Y* response was measurable also at low frequencies but it depended strongly on the texture. Figure 2 shows the *Y* displacement for a sample at  $28^\circ\text{C}$  with a vertical chevron texture and for the same sample with a striped texture. The difference is striking. The chevron texture shows a much stronger response that increases with the frequency in the range from  $0.2$  to  $1\text{ kHz}$  and forms a relatively sharp peak centered at  $1.1\text{ kHz}$ . The response drops sharply above  $1.2\text{ kHz}$ . The striped texture has a much weaker response; the displacement is fairly constant in the low-frequency range and it has a small peak at  $1.2\text{ kHz}$ . Again it drops sharply above this frequency and has another weak maximum at about  $2.3\text{ kHz}$ .

At  $60^\circ\text{C}$  (see Fig. 3), that is, close to the transition to Sm-*A*, the displacement is stronger and does not depend significantly on the history of the sample. A fresh sample obtained on cooling gives nearly the same response as a sample obtained on heating from the striped texture. The frequency dependence shows a broad peak at  $0.8\text{ kHz}$ . There is a sharp decrease between  $1.0$  and  $1.2\text{ kHz}$ , then the displacement fades out gradually in the range to  $2\text{ kHz}$ .

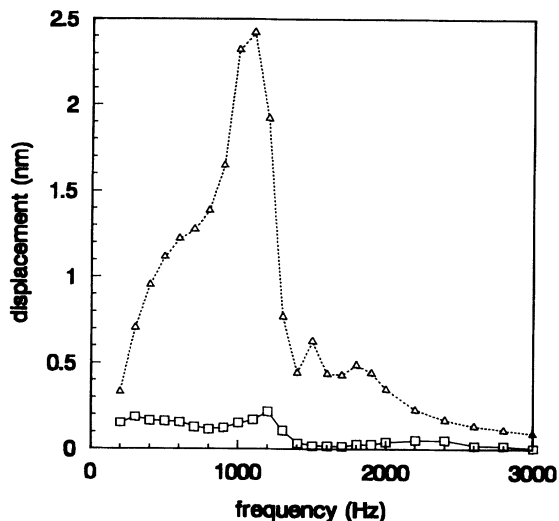


FIG. 2. Frequency dependence of displacement in  $Y$  direction,  $T=28^\circ\text{C}$ ,  $U=4.2$  V. Triangles, fresh sample, chevron texture obtained on cooling; squares, relaxed sample, striped texture.

Figure 4 shows the field dependence of the displacements at  $23^\circ\text{C}$  for a sample with a chevron texture and for the same sample with the striped texture. For the chevron texture, the displacement goes, as expected, linear with the field at low fields at least up to about 5 V. With the striped texture the results are quite different. There is no response up to about 15 V, then there follows a sharp rise of the displacements which seems to flatten somewhat at about 32 V. In decreasing the voltage, the displacements follow first a line that is proportional to  $U$ , as expected for a linear response, but below 22 V they decrease much faster. Below 14 V the mechanical response is zero.

Microscopic observations show that the increase of the

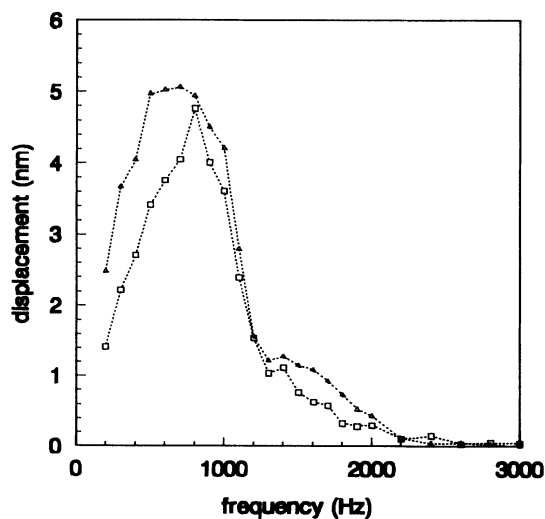


FIG. 3. Frequency dependence of displacement in  $Y$  direction.  $T=60^\circ\text{C}$ ,  $U=4.2$  V. Triangles, fresh sample, obtained on cooling from  $\text{Sm-A}$ ; squares, sample obtained on heating from striped texture.

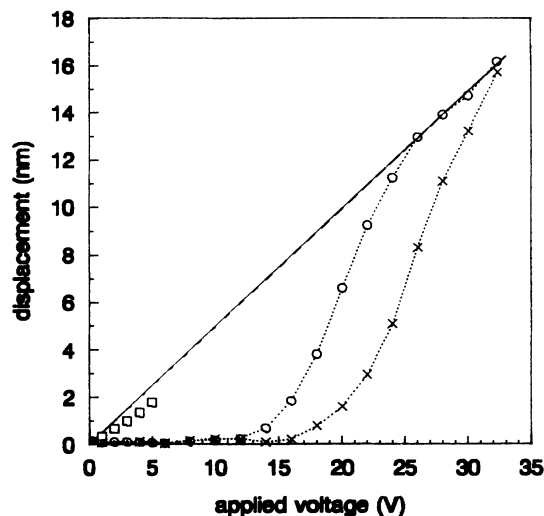


FIG. 4. Voltage dependence of displacement in  $Y$  direction.  $T=23^\circ\text{C}$ ,  $f=700$  Hz. Squares, chevron texture;  $\times$  and  $\circ$ , striped texture increasing and decreasing voltage, respectively.

response is correlated to a realignment of the  $c$ -director field.

The striped texture appears uniformly dark between crossed polarizers when the polarization is parallel to the rubbing direction. The application of the field does not cause significant changes below the voltage where the mechanical response begins to change. Parallel with the change in mechanical response the texture brightens without a change in stripe pattern. For frequencies much above 1 kHz the electric field did not cause much change in texture up to 35 V. It is important to note that above 1 kHz the mechanical responses are also small and do not show an unusual field dependence in the studied voltage interval.

The voltage dependence of the displacements is very different at  $59^\circ\text{C}$  which is close to the  $\text{Sm-A}$  phase (Fig. 5); at low fields the response is linear but it seems to satu-

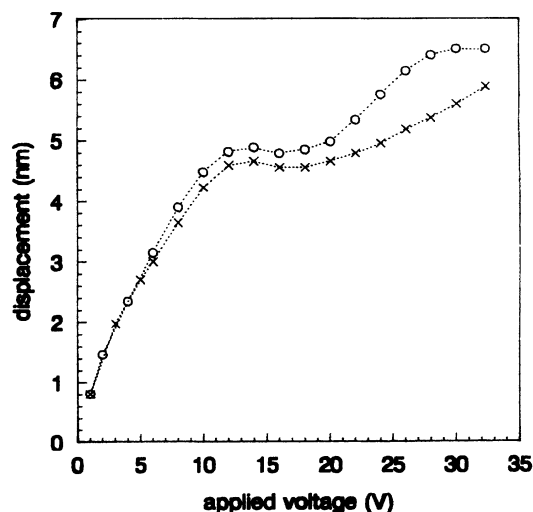


FIG. 5. Voltage dependence of displacement in  $Y$  direction.  $T=59^\circ\text{C}$ ,  $f=700$  Hz;  $\times$ , increasing;  $\circ$ , decreasing voltage.

rate near 15 V. A further increase begins above 20 V. It seems to be connected again with a reorientation of the director field since we observed a significant hysteresis in this range.

#### IV. DISCUSSION

The characteristic structure of the striped texture can be deduced on the basis of the described observations. The fact that the electrical polarization in each stripe is perpendicular to the plates implies that the  $C_2$  axis of the untwisted smectic phase and the layers are also vertical. The director and layer normal are in the plane of the film. The alternating polarization indicates that the layer orientation changes at the stripe boundaries. We observed that the angle  $\alpha_d$  between director and rubbing axis changes from  $+5$  to  $-5^\circ$  at the stripe boundaries simultaneously with the polarization. We can use this observation for an estimate of the angle  $\alpha_k$  between the layer normal and the rubbing direction. The angle between layer normal and director for the given material is  $\alpha_c = 26^\circ$  (data from E. Merck). Using the relation

$$\alpha_k = \alpha_c + \alpha_d \quad (1)$$

we find  $\alpha_k = 21^\circ$  or  $31^\circ$ , depending on the assumption that we make for the sign of  $\alpha_d$ . We also determined the angle between the layer normals experimentally. We observed the sample under the polarizing microscope and applied a 0.5-kHz voltage ( $> 10$  V) to the film. The field aligns the  $c$  director parallel or antiparallel to it without changing the layer structure. In this configuration the stripes appear dark when the polarization is parallel to the layer normal. We found that one set of stripes was dark when the polarization angle was  $23^\circ \pm 2^\circ$  against the rubbing direction; turning it to  $-23^\circ \pm 2^\circ$ , the dark and bright stripes interchanged. This gives  $\alpha_k = (23 \pm 2)^\circ$  which is in agreement with the above estimate for  $\alpha_k$  with  $\alpha_d$  assumed to be negative.

The linear electromechanical response at frequencies below 1 kHz is in general dominated by the Goldstone mode. The electric field couples with the permanent polarization and exerts a torque that causes a rotation of the  $c$  director which in turn causes a mechanical oscillation of the cover plate. The torque is proportional sine of the angle between field and polarization [ $\sin(\psi)$ ]. In chevron textures the polarization is more or less normal to the field and a strong linear response is expected. However, as discussed earlier [9], the mechanical effects of different domains may cancel so that the net response often can be relatively low. The polarization of the undisturbed stripe texture is parallel or antiparallel to the field so the torque is zero and, as observed (Fig. 4), there is no linear response. When the field increases above a critical value the polarization direction changes and a linear response is observed. At sufficiently high fields we assume a polarization that is on the average perpendicular to the field and in this range the displacement is proportional to the applied voltage. The observed hysteresis between up and down sweep in voltage must be caused by a hysteresis in the reorientation of the director field.

We understand qualitatively the voltage dependence of

the response in stripe textures. The voltage dependence observed in texture close to the Sm- $A$  phase is more complicated. Presumably, we have here a chevron texture. There is a linear response that is proportional to the field strength for low voltages but at higher voltages there is a range in which the response is independent of the voltage. There is also a difference in up and down sweeps which indicates that realignments of the director field are induced. Probably the complicated voltage dependence is due to these realignments in more or less irregular chevron texture which may include some structural features of the stripe texture. The chevron texture itself can be distorted to some extent by the field which also may complicate the response curve.

The frequency dependence of the displacements (Figs. 2 and 3) is also difficult to explain on the basis of Goldstone modes, i.e., on the assumption that the mechanical effects are due to the oscillations of the  $c$ -director field. According to this assumption, the force acting on the cover plate (therefore its acceleration) for small oscillations should be proportional to torque by the electric field on the material. For an estimate here, we neglect the coupling with flow and the curvature elasticity and assume the average polarization perpendicular to the field. In this approximation we obtain from the torque balance that

$$\frac{d\psi}{dt} = \frac{P_0 E}{\gamma_1 \sin^2 \theta} \quad (2)$$

That gives for the stress [9]

$$\sigma_{xy} = \frac{1}{2} \frac{d\psi}{dt} \sin^2 \theta (\gamma_2 + \gamma_1) = \frac{1}{2} E P_0 \frac{\gamma_2 + \gamma_1}{\gamma_1} \quad (3)$$

This means the measured acceleration is independent of frequency.

In the equations  $\psi$  is the angle between  $P_0$  and the field  $E$ ,  $\gamma_1$  is the rotational viscosity, and  $\gamma_2 = \eta_b - \eta_c$ , where  $\eta_c$  and  $\eta_b$  are the shear viscosities with the director in the direction of flow gradient and parallel to the flow, respectively.

In the first approximation, the amplitudes of the mechanical displacements should be proportional to  $1/\omega^2$ . Above 1 kHz there is indeed a strong decrease of the amplitude, but at low frequencies the experimental results clearly contradict this conclusion. There is no divergence of the displacements when  $\omega \rightarrow 0$ . There are two factors which may be responsible for the deviations at low frequencies. First, the oscillation amplitudes are not small at very low frequencies and, secondly, the coupling to the flow and the curvature elasticity may be important.

The coupling to flow is also needed to explain the reorientation of the director field of the striped texture in low-frequency fields. The torque due to anisotropy of the dielectric properties has no frequency dependence in the studied frequency range, but we observed a dependence of the threshold voltage on the frequency, which strongly indicates that the coupling to the Goldstone mode plays an important role in reorienting the polarization.

In addition to the Goldstone mode the induced

changes of the director tilt  $\theta$  are also of major interest. We assume that this mechanism is responsible for the “resonances” [10]. They appear particularly strong in the chevron texture (Fig. 2) but they are also present in the striped texture. The basic resonance frequency seems to be close to 1 kHz but it appears to be 10% higher in the striped texture. It is remarkable that there are indications of a higher-order resonance at about twice these frequencies. At present we cannot offer any detailed descriptions of the modes connected with the resonances.

Our present understanding of the formation and structures of the textures is the following. Due to the polyimide coating and rubbing there is a strong anchoring at the surfaces. At the surface the director is parallel to the rubbing direction and lies in the plane of surfaces (a small tilt of several degrees is possible). The shearing perpendicular to the rubbing direction helps to form a homogeneously oriented Sm-*A* film with a bookshelf texture [see Fig. 6(a)]. On cooling into the Sm-*C* phase, the director tilts away from the layer normal and the layer distance decreases. This leads to an undulation instability [11] because new layers cannot form readily. There are two limiting orientations of the undulation wave vector, perpendicular (*X* direction) or parallel (*Y* direction) to the plane of the film. The perpendicular undulation does not require a motion of the layers at the surface and has a maximum wavelength of twice the film thickness. It leads to the (vertical) chevron texture in which the smooth undulation is replaced by layer kinks [Fig. 6(b)]. The parallel undulation requires a motion of the layers along the surface, but there is no constraint for the wavelength so that it can have a smaller free energy. It will lead to the striped texture [horizontal chevron texture, see Fig. 6(c)].

If the equilibrium layer distance were strictly proportional to  $\cos(\theta)$  the kink angle  $\beta$ , would be equal to  $\theta$  at equilibrium. In that case, the director could remain parallel to the rubbing direction in the whole sample [see Fig. 7(a)]. However, the relation does not hold accurately and, therefore, the two angles deviate usually by several degrees [see Fig. 7(a')]. That accounts for the observed deviation of  $5^\circ$  of the director orientation from the rubbing direction in the striped texture and corresponding deviations in the chevron texture.

It is interesting to note that the director configuration of vertical chevron and striped texture are very similar in the sense that a side view of a chevron corresponds to a top view of a striped texture [Figs. 7(a) and 7(a')]. In the

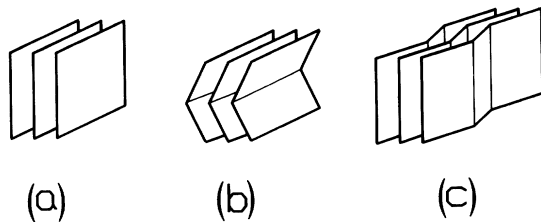


FIG. 6. Schematics of smectic layer structures. (a) Bookshelf texture (typical for Sm-*A*). (b) Vertical chevron texture (typical for planar Sm-*C*\*). (c) Striped (horizontal chevron) (also found in Sm-*C*\*).

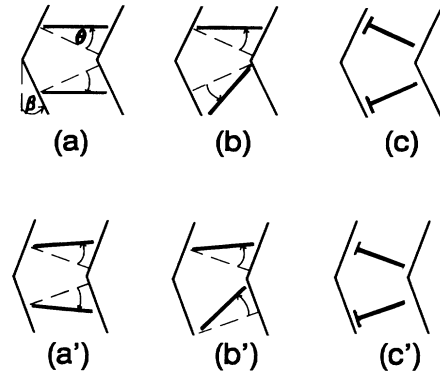


FIG. 7. Schematics of the director orientation in Sm-*C*\*.  $\theta$  angle between director (thick solid line) and layer normal (dashed lines),  $\beta$  kink angle of layers [indicated in (a) only]. (a)  $U=0$ . Relaxed states with  $\beta=\theta$ . Corresponds to a side view of a vertical chevron texture. Polarization horizontal, but opposite in lower and upper half. May also be considered as top view of a striped texture with polarization vertical. (a') Similar to (a), but with  $\beta<\theta$ . (b) Striped texture in strong electric field ( $U_{dc} > U_{th}$ ),  $\beta=\theta$ . Polarization in all stripes parallel to the electric field. (b') Similar to (b), except  $\beta<\theta$ . (c) Side view of vertical chevron texture in strong electric fields.  $\beta=\theta$ . The polarization is parallel to the layers and tends to be parallel to the field. May also be considered as top view of striped texture in strong ac fields. The polarization is perpendicular to the field and parallel to the layers. (c') Similar to (c), but  $\beta<\theta$ .

presence of a vertical electric field this correspondence does not hold [see Figs. 7(b) and 7(b')] but can be found corresponding analogy between the side view of a chevron in the strong dc field and the top view of the striped texture in the strong ac fields [see Figs. 7(c) and 7(c')].

At this time we do not understand why the chevron texture is reformed on heating from the striped texture. It may be that the surface energy favors the chevron texture. A better understanding of these problems is important for practical applications.

## V. SUMMARY

We observed on a surface-stabilized ferroelectric-liquid-crystalline film a spontaneous transition from a chevron texture to a striped texture and found that in the subsequent stripes the polarization alternates between up and down in contrast to the chevron texture where the spontaneous polarization lies in the horizontal plane. This structure has a lower free energy than that created by the application of the strong electric field [see Figs. 7(b) and 7(b')]. It also has a much lower free energy than the chevron structure, but for their creation translations of the layers along the rubbing direction take place, which takes time. According to our model, the striped bookshelf texture should be more stable than the chevron. We expect that with other materials a transition to a striped texture can also be found because, with proper surface conditions, it has a lower free energy.

There are significant differences in the electromechanical responses of the two textures. The stripe texture does not have a linear electromechanical effect at low fields;

only at higher fields does the mechanical vibration have a component of the frequency of the field. This indicates that the spontaneous polarization has rotated and is no longer parallel to the electric fields. In contrast to the director reorientations, the layer structure is unchanged by the application of the field.

#### ACKNOWLEDGMENTS

Research supported in part by DARPA/ONR Contract No. N00014-86-K-0766 and the National Science Foundation under the ALCOM Center Grant No. DMR89-20147.

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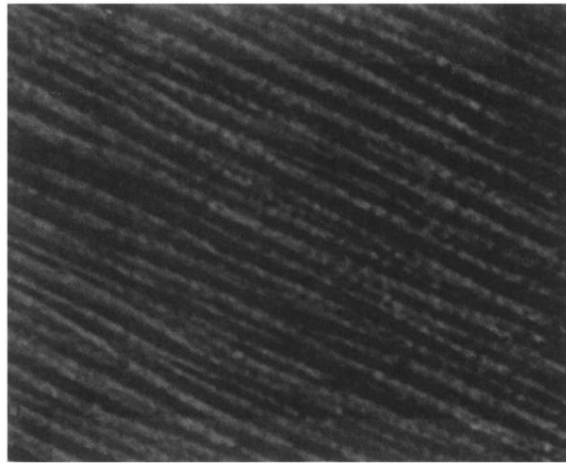


FIG. 1. Spontaneously formed striped texture, surface-stabilized Sm-C\* film (ZLI 4237-000, E. Merck);  $5\ \mu\text{m}$  thick,  $T=28\ ^\circ\text{C}$ , stripe width  $\sim 50\ \mu\text{m}$ , crossed polarizers. The stripes are parallel to the rubbing direction and perpendicular to the shear, polarization angle  $5^\circ$  to stripes.