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## Liquid Crystals

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# The transition mechanism of the transient planar to planar director configuration change in cholesteric liquid crystal displays

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In cholesteric liquid crystals with a positive dielectric anisotropy, the relaxation from the electric field-aligned director configuration to the stable zero field director configuration proceeds via a metastable transient planar director configuration which has a pitch distinct from the equilibrium state. The transition from the transient planar to the equilibrium zero-field state is shown here to occur via a Helfrich-type instability which continuously leads to an in-plane helical structure. The equilibrium planar state is then seen to grow continuously from the in-plane state, leaving behind walls whose length then spontaneously shrinks.

## 1. Introduction

Cholesteric liquid crystals whose equilibrium pitch causes the reflection of visible light [1] have been suggested for use in several practical applications [2–4]. Devices used in these applications have two states, labeled planar (P) and focal conic (FC), that are stable with no applied voltage. Switching from the FC to P requires the application of a high voltage (usually 40 V or more) which forces the liquid crystal system into a homeotropic (H) state, where the director is along the cell-normal direction, parallel to the applied field. On removal of this field, a relaxation process ultimately resulting in the P state occurs. However, in such applications, the transition time from the field-aligned homeotropic director configuration to the final equilibrium planar director configuration is often considered to be a problem [5].

The relaxation from H to P is somewhat complicated, and not fully understood. The transition begins with the director relaxing in a one dimensional, conical manner to a transient planar (TP) state, which has the director basically within the cell plane [6–8]. The primary structural difference between TP and P is the number of director twists between the two substrates. Yang *et al.* derived the elastic free energy of a cholesteric material during the relaxation from H to TP from the Frank–Oseen equation as a function of the polar angle and the chirality [9], and have theoretically estimated and experimentally observed the TP pitch to be about  $P_0 K_{33}/K_{22}$ . Here,  $P_0$  is the equilibrium material pitch,

$K_{22}$  is the twist elastic constant, and  $K_{33}$  is the bend elastic constant for the liquid crystal material. This derivation assumes a conical relaxation to TP. For most available nematic host materials,  $(K_{33}/K_{22})$  is between 2 and 3, yielding a TP pitch significantly longer than the equilibrium pitch.

If we assume that TP is a perfect Grandjean state, its twist free energy density (inserting the TP pitch into the Frank–Oseen free energy density expression [10]) is

$$f_{\text{twist}} = 2\pi^2 \left( \frac{K_{22}}{P_0^2} \right) \left[ 1 - \left( \frac{K_{22}}{K_{33}} \right) \right]. \quad (1)$$

This energy density is the difference between the free energy densities of the TP and P states. Since the energy of the TP state is higher, a transformation from TP to P is observed to occur.

Our earlier work investigated several aspects of the transformation process from TP to P [11, 12]. We found that during the transition, the angular distribution of reflecting cholesteric helices changes from very narrow (nearly perfect Grandjean) to very wide (cholesteric helices oriented at a significant angle away from the cell normal direction), then returns to very narrow. Also during this process, the texture changes from a very uniform appearance in TP to being highly domain-divided in a transition state, with a slow growth of domains (on the order of seconds) leading to a perfect Grandjean planar equilibrium state.

The mechanism through which the transformation from TP to P occurs is not well understood. The relationship between the formation of domains and the appearance of reflecting helices away from the cell-normal direction

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is unclear. Whether a domain nucleation and growth process (depending on the motion of defect walls) or a bulk modulation process occurs has not been established. A diagram of the full H to TP to P transition, as it is currently understood, may be seen in figure 1 [4]. This picture shows that the transition begins with a well-understood conical relaxation to the TP state, but the mechanism of the transition from the TP to P has not been elucidated. In this study, we will explain the transition mechanism based on observations of two systems using optical microscopy.

## 2. Experimental

We have employed an experimental set-up consisting of a Nikon Optiphot 2 polarizing microscope with a Leitz 35 mm camera, a Xe lamp, and electronic components to obtain photomicrographs of textures with time resolution of about 20  $\mu\text{s}$ . The regular Nikon microscope illuminator was replaced with an Oriel series 6427 Xe flash lamp (5 J maximum output and a full width half max of 9  $\mu\text{s}$  flash duration). The voltage applied to the sample was a 40 V a.c. square wave signal at 0.5 kHz produced by an Analogic Model 2020 Polynomial Waveform Synthesizer. The voltage was applied for 1 s to ensure that the texture was fully homeotropic. The voltage was then set to zero. At specified times after the voltage was removed, a trigger signal from the waveform synthesizer activated the flash lamp system. The energy of the flash is sufficient to use ordinary 35 mm Kodak Max 800 DIN film at 200 $\times$  microscope amplification. Delay times of 1 ms to 5 s were employed.

The cells used in this study employed ITO-coated soda lime glass coated with Dupont 2555 polyimide. The polyimide was rubbed in an antiparallel fashion. Cell spacings of 15 and 47  $\mu\text{m}$  were used. Cells were filled with a mixture of Merck ZLI 4792 liquid crystal with 3% CB15 chiral additive. The intrinsic pitch of this mixture was measured to be 5.4  $\mu\text{m}$  using the Grandjean–Cano method. Since ZLI 4792 has a value of 2.815 for the ratio of  $K_{33}/K_{22}$ , we expect the transient planar pitch to be approximately 15  $\mu\text{m}$  in an unrestricted system. With the surfaces fixed in our systems, the transient planar state in the 15  $\mu\text{m}$  cell should consist of 1 full twist, while the equilibrium planar state will have

about 3 full twists. Similarly, for the 47  $\mu\text{m}$  cell, we can expect the transient planar state to have 3 twists, and the equilibrium state to have  $8\frac{1}{2}$  twists. The cells were observed between crossed polarizers.

## 3. Experimental results

Observation of photomicrographs of the 15  $\mu\text{m}$  cell shown in figure 2 reveals that in the early stages of the relaxation process, a domain-free texture is present. While the voltage is applied, a black texture is observed. This is consistent with the cell being in the H state, where the director is along the cell-normal direction, and thus no retardation is observed. We will define time  $t = 0$  as the time at which the voltage is removed. By  $t = 1$  ms, a coloured texture has become visible. The colour of the cell remains uniform across the entire viewed area. Various colours are observed until about 15 ms into the process, at which time the cell has achieved a uniform green texture. This texture does not change for the next 200 ms. These observations agree with the expected one dimensional relaxation to a Grandjean transient planar state. The appearance of colours corresponds to an increase in observed birefringence as the director changes from being aligned normal to the cell (in the H state) to within the cell (TP state).

After the 15  $\mu\text{m}$  cell has been in the transient planar state for about 200 ms, a deformation distorts the uniformity of the texture. This deformation, which initially appears as a series of violet stripes running perpendicular to the rub direction, increases in scope and magnitude between times  $t = 225$  to 275 ms. Note that the stripes appear to be grouped in pairs. As time progresses, the modulation increases in magnitude, and the colour changes to yellow, then lavender. The lavender regions then grow, with wall boundaries separating the region. This lavender colour is seen to correspond to the retardation of the equilibrium P texture.

In the 47  $\mu\text{m}$  cell, a similar process is observed, although here the distortion is more intricate, and appears to have a three dimensional rather than a two dimensional structure (see figure 3). Note that in this system, the material remains in the TP state for only about 35 ms before the transformation begins. The observed modulation is seen to be non-directional,

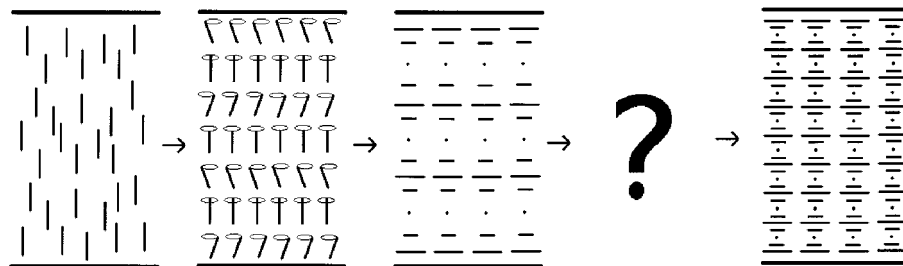


Figure 1. The homeotropic to planar transition as currently understood.

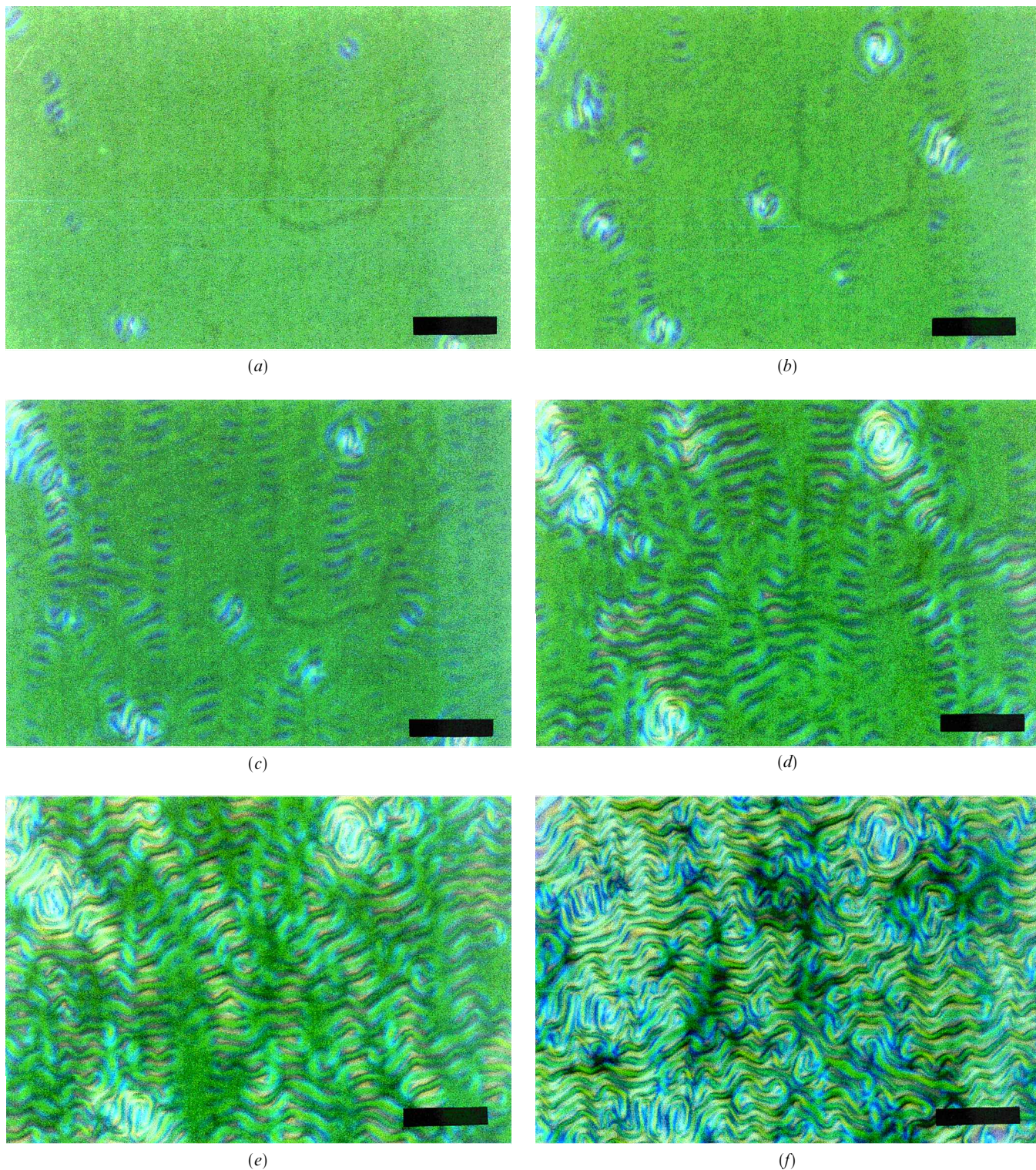
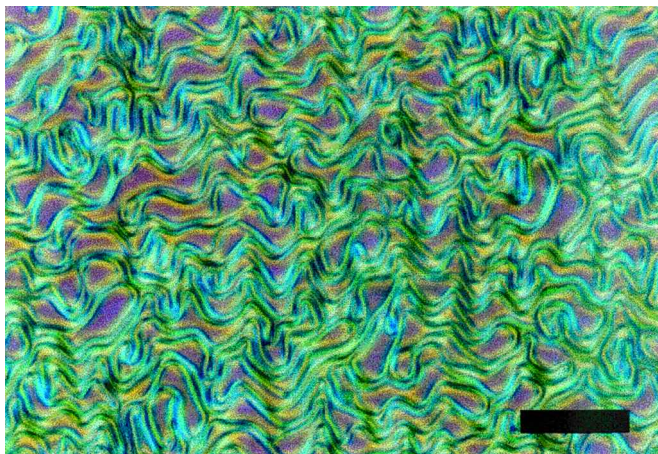


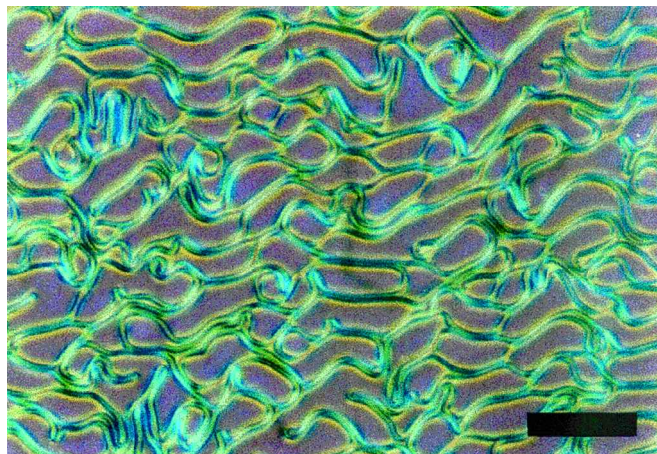
Figure 2. Photomicrograph of  $15\ \mu\text{m}$  cell, the black bar is  $80\ \mu\text{m}$  long: after (a) 200 ms, (b) 225 ms, (c) 250 ms, (d) 275 ms, (e) 300 ms, (f) 400 ms, (g) 500 ms, (h) 700 ms (i) 1000 ms. (Continued on next page.)

although a local periodicity may be observed at various locations throughout the sample. In this sample, the equilibrium state is seen to have a green colour. In a

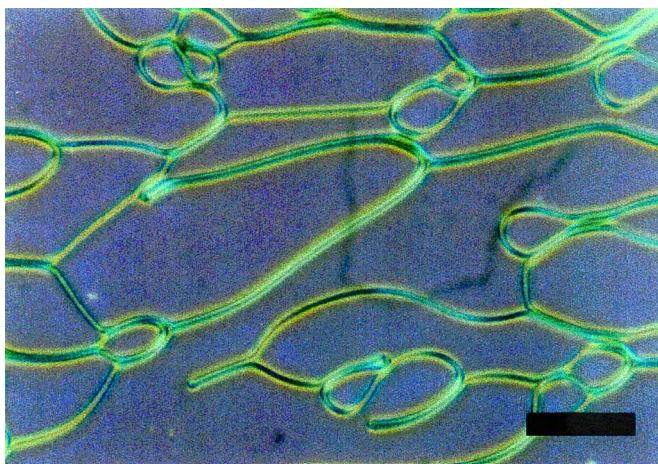
similar manner to that observed in the  $15\ \mu\text{m}$  cell, the transformation process ends with the slow removal of defect lines.



(g)



(h)



(i)

Figure 2. (Continued).

#### 4. Discussion

Let us consider what the data suggest about the nature of the TP to P transition. In the  $15\ \mu\text{m}$  cell, the transition begins with the TP director being modulated into a texture with a striped appearance. Striped configurations having similar appearance have previously been observed in cholesteric liquid crystals and explained by Helfrich [13], Hurault [14], and Chigrinov [15]. These director configurations involved sinusoidal modulation of the director structure. In these early studies, stripes occurred only in the presence of an applied field. We propose that the stripes that we have observed are configurationally similar to the field-induced stripes, but have the high energy of the TP as a driving force (rather than the desire for the director to align with an applied field). A cross-sectional view of the proposed transitional director configuration of the stripes is shown in figure 4. Consider, however, that a perfect Grandjean TP state is a metastable configuration in which no torque encourages

the director to deform away from the cell plane. Therefore, thermal fluctuations, surface irregularities, spacers, or other factors influencing the director may be required for the striped deformation to begin. Note that a helical twisted structure may be observed in the horizontal dimension of the deformation. This could conceivably lower the total free energy, making this an energetically favourable director configuration. The periodicity of a stripe deformation is given by Chigrinov as

$$\lambda = [PD(3K_{33}/2K_{22})^{1/2}]^{1/2} \quad (2)$$

where  $P$  is the equilibrium pitch and  $D$  is the thickness of the sample [13]. Note that this periodicity is independent of applied field. For our sample, since we are looking at a deformation of the transient planar rather than the planar state, we may use the transient planar pitch as our value for  $P$ . Considering that  $P = D = 15\ \mu\text{m}$  in the TP state, and  $K_{33}/K_{22} = 2.815$ , we may expect  $\lambda$  to be about  $21\ \mu\text{m}$ . This agrees very well with our observed periodicity of about  $25\ \mu\text{m}$  in figure 2.

Once the deformation is started, it spreads across the area of the cell and increases in magnitude. Two possible methods by which the deformation may progress to the equilibrium state are proposed in the sketches in figure 5. In each case, the walls separating the P domains are shown to be separated by a  $180^\circ$  twist deformation (a pi wall). Such a wall deformation could then be removed by shrinking of the defect line in the third dimension.

The deformation process that is observed in the  $47\ \mu\text{m}$  cell appears different from that of the thinner cell in that it demonstrates no directionality. That is, there is no apparent preferred direction along which the helical axis is oriented. This is not surprising, as a similar phenomenon has been observed by Chigrinov *et al.* in electric-field induced stripe deformations [15]. In that study, it was found that a striped texture was observed for low values of  $d/P$ , while a non-directional deformation

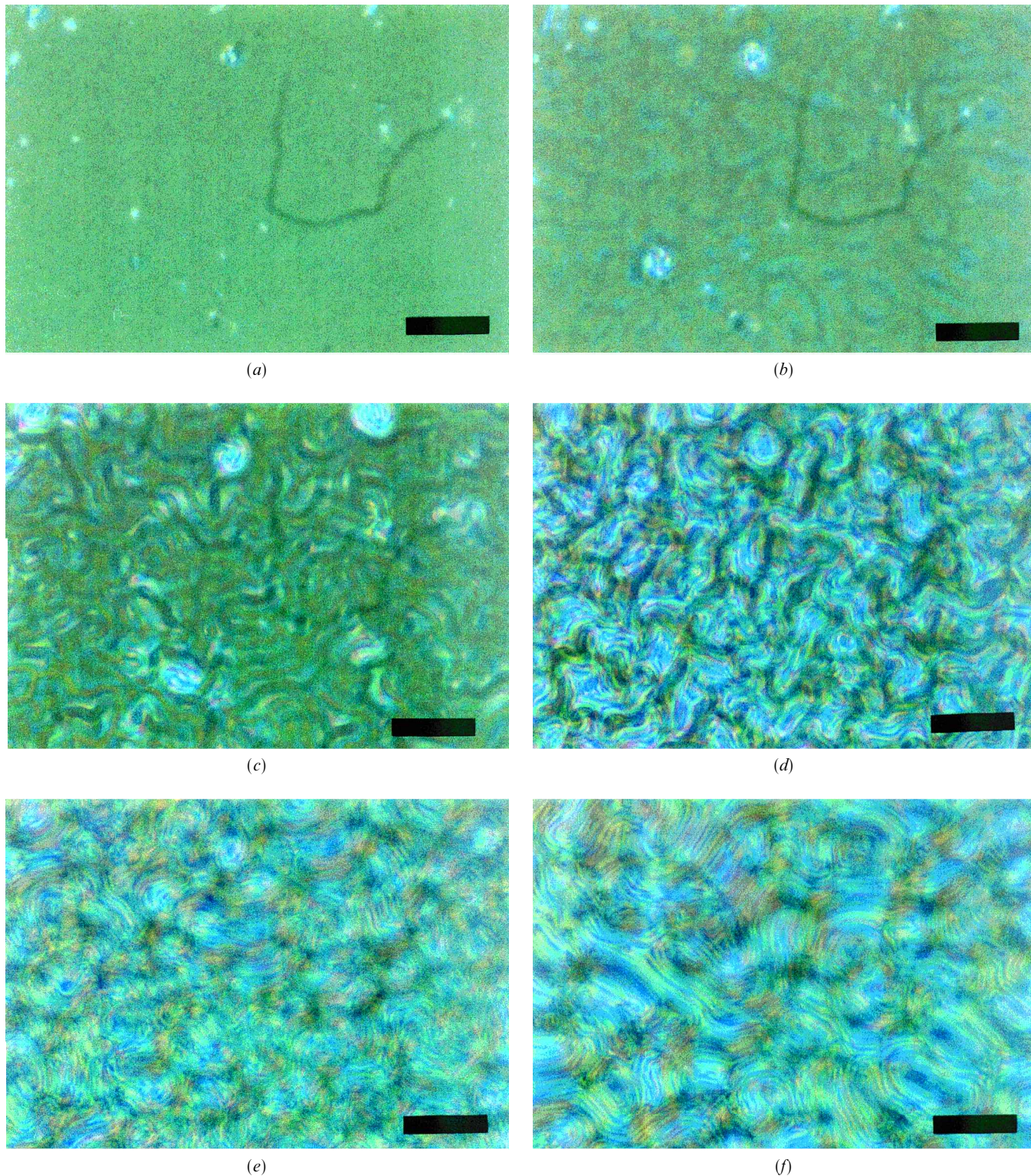


Figure 3. Photomicrograph of  $47\ \mu\text{m}$  cell, the black bar is  $80\ \mu\text{m}$  long: after (a) 25 ms, (b) 50 ms, (c) 75 ms, (d) 125 ms, (e) 450 ms, (f) 1000 ms, (g) 1200 ms, (h) 2 min. (Continued on next page.)

was seen in cases with larger  $d/P$ . The non-directional nature of the deformation that we see for larger values of  $d/P$  agrees well with the observations by Chigrinov

*et al.* of Helfrich-type distortions in cholesteric samples in ref. [15]. In that study, the authors found that a striped appearance consistently occurred for samples

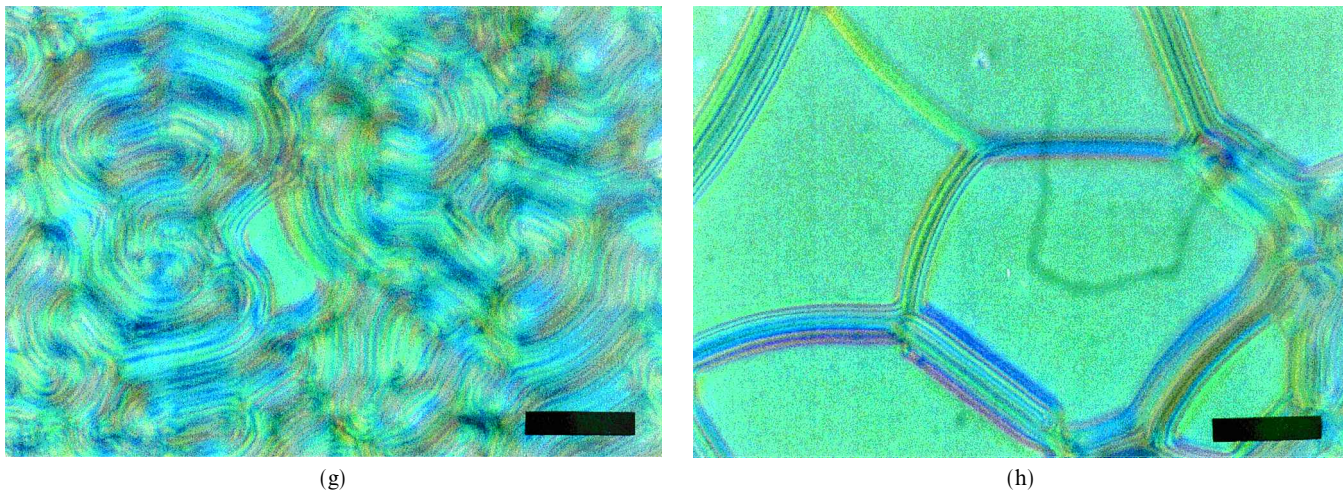
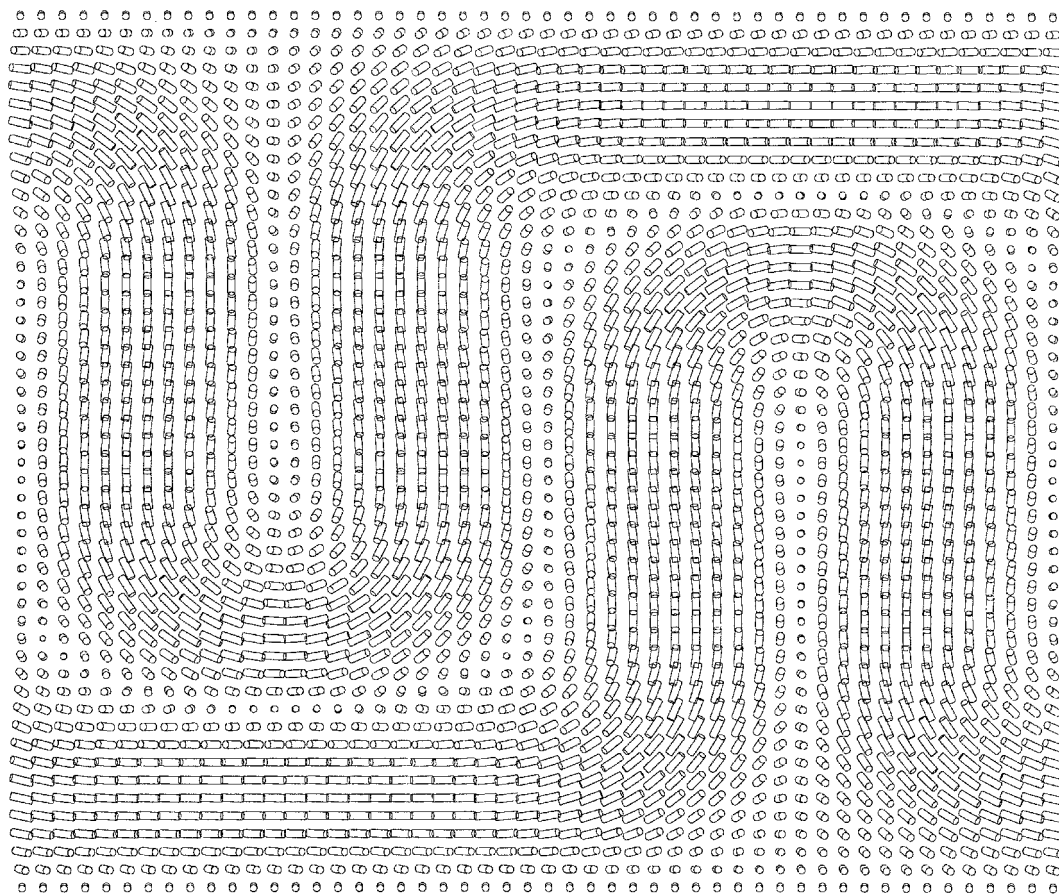


Figure 3. (Continued).

Figure 4. Proposed transitional director configuration for the 15  $\mu\text{m}$  cell.

with less than about 2 twists, while a non-directional deformation was observed for samples with greater than about 3 twists. A two dimensional picture of a possible structure for the deformation of a sample with  $d/P$  of 6 is shown in figure 6.

Note that during the proposed deformation processes, the angular distribution of helical axes of the material becomes very wide. This agrees very well with previous research indicating that during the transition from TP to P, helical axes are found at very wide angles, while

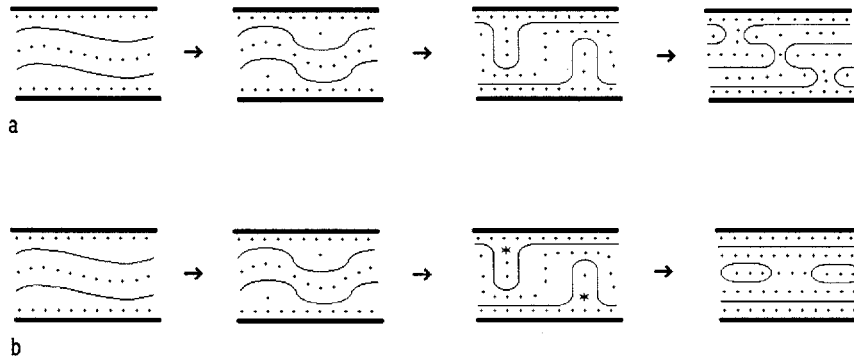


Figure 5. Two possible routes for transformation from transient planar to planar. In (a), the modulation forms an equilibrium pitch by spreading like a mushroom, whereas in (b), the action of defects (denoted by \*) allows the equilibrium pitch to form. Note that in this system, we expect a change from 1 pitch to 3 pitches. This diagram is merely intended to show the character of a possible change in pitch.

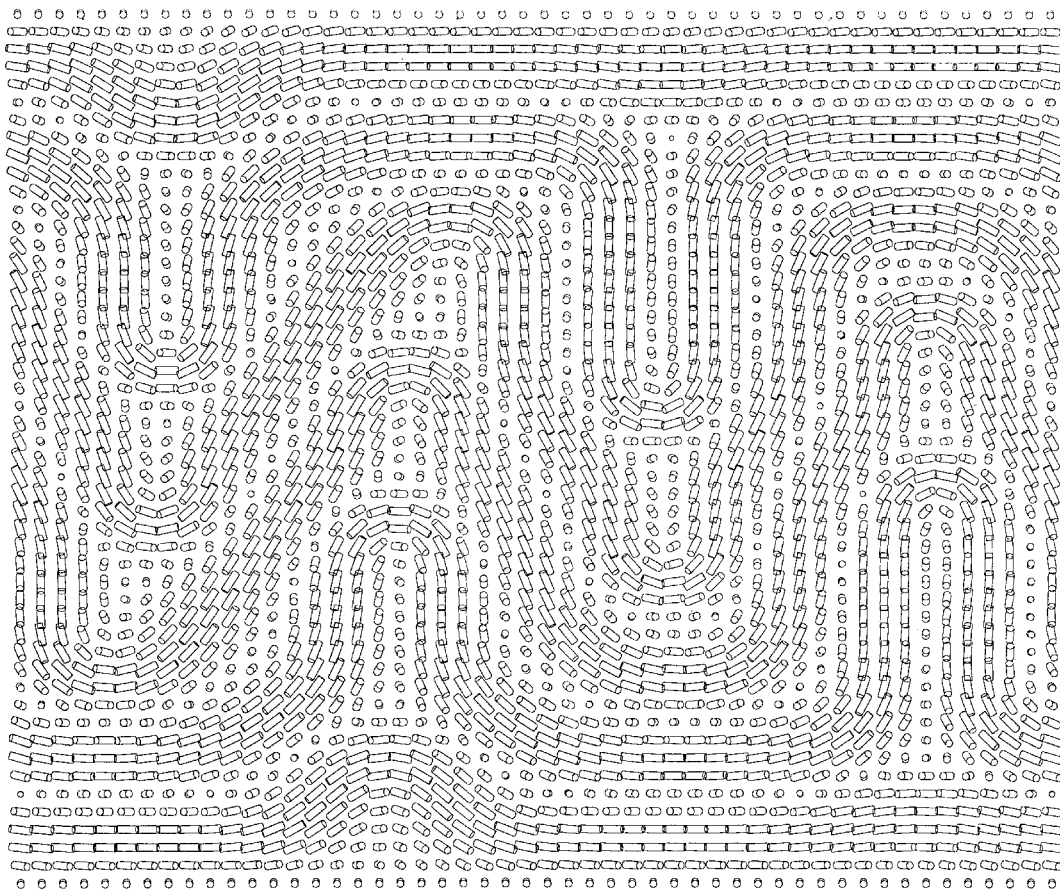


Figure 6. Proposed transitional director configuration for the 47  $\mu\text{m}$  cell.

both the TP and P states have very narrow angular distribution of reflecting helices [12].

### 5. Conclusions

Based on experimental observations, a mechanism of transformation between the transient planar and equilibrium planar states of cholesteric liquid crystals is

proposed. We have observed that the relaxation of texture from the transient planar (TP) to the equilibrium planar (P) state of cholesterics occurs through a deformation similar to the Helfrich stripe deformation observed in cholesteric devices in the presence of an electric field. In thicker samples with a greater total twist, a similar deformation is observed, only without the directional



nature observed in thinner cells. This loss of directionality concurs with observations by Chigrinov of field-induced modulations [15].

The driving force behind this modulation is thought to be the high energy of the TP state compared with the P state. Because in the TP to P transition there is no applied field supplying a torque to move the director out of the cell plane, it is likely that thermal fluctuations or surface imperfections may be necessary to initiate the deformation. The formation of a helical axis within the cell during such a deformation may lower the free energy enough to make the deformation favourable. The formation of a Helfrich-type modulation is supported by previous research [12] showing that the angular distribution of helical axes becomes very wide during the time that the material pitch is changing from its transient planar value to its equilibrium value.

The equilibrium planar state is seen to grow continuously from the in-plane state, leaving behind defect walls whose length spontaneously shrinks, resulting in a monodomain structure.

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