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## Mechanism of Relaxation from Electric Field Induced Homeotropic to Planar Texture in Cholesteric Liquid Crystals

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We have investigated the relaxation process from a homeotropic structure to the planar one in a cholesteric liquid crystal. The relaxation occurs after switching off the electric field. We have developed a new experimental photography technique with a time resolution of about 20 $\mu$ s and applied it to the investigation of textural changes during the transition. After the liquid crystal goes through a defect-free transient planar structure, the development of the cholesteric helices occurs in two stages. Direct observations have shown that formation of the final equilibrium state involves a nucleation process. We used optical retroreflection spectroscopy and measurement of light scattering to observe changes of the cholesteric pitch and orientation of helices as well as the size of domains in time for various treatments of the glass substrates. We have also proposed a theoretical model that describes the experimental data satisfactorily.

**Keywords:** cholesteric liquid crystals; bistable reflective displays; liquid crystal texture; relaxation time

### INTRODUCTION.

Bistable Cholesteric Texture (BCT) displays attract a lot of interest because of their many possible applications [1-3]. A limitation in the practical use of BCT displays is the comparatively long relaxation time, especially when switching between the field-induced homeotropic (nonreflecting) and equilibrium reflecting textures [3]. Many efforts have been directed recently to understand the nature of the relaxation of BCT's and to make the relaxation time shorter. In particular, the production of a "perfect" intermediate transient planar (TP) state during the transition from the homeotropic to planar structure was observed. This state is characterized by a "perfect" cholesteric structure with a pitch  $P_p > P_o$  ( $P_o$  is the equilibrium pitch) and with helical axes perpendicular to the substrates [4-6].

The characteristic times of the TP relaxation are shown to be determined by parameters of the liquid crystal and not to be related to surface alignment [7]. Our previous investigations have shown that the equilibrium pitch is achieved in the cholesteric structure in about 10ms and the following relaxation to the equilibrium

planar state is due to macroscopic structure changes only. The reflection spectra can not explain what structural changes determine the long relaxation time. In this paper we examine the relaxation process by direct observation and analysis of the structure

## EXPERIMENT.

To study the dynamics of the BCT, we designed a new technique which allows one to obtain microphotographs of LC textures with a time resolution of about 20  $\mu$ s. It consists of a Nikon polarizing microscope with a photocamera, an illuminator and electronic components. The regular Nikon microscope illuminator was replaced with Oriel series 6427 Xe flashlamp (5J maximum output and a full width half max of 9  $\mu$ s flash duration). The electronic components include: a digital DS345 Synthesized Function Generator from Stanford Research System, a 120V bipolar amplifier (A) and a Delay Time Control Unit (DTCU). The voltage applied to the sample is an AC signal at 1kHz and 45-50V<sub>pp</sub> modulated by a 0.5Hz square wave (100% amplitude modulation). The negative slope of the modulation signal is used as the trigger source for the DTCU, which flashes the lamp at a preset time after the trigger signal is obtained. The energy of the flash is sufficient so that ordinary 35mm Kodak Max 800 DIN film at 100x-400x microscope magnification can be used.

Two different initial cholesteric structures were used for investigations. Both were prepared in a 5  $\mu$ m sandwich type cell filled with a mixture of Merck liquid crystal 18349 and CB15 chiral additive. The reflected wavelength is  $\lambda_{\text{max}} = 600 \pm 10$ nm. It gives an estimate  $P_0 = \lambda_{\text{max}}/n = 375$ nm, where  $n$  is average index of refraction of the liquid crystal. The only difference between those cells was in the surface treatment. A rubbed Dupont-2555 polyimide was used as an alignment layer to obtain planar structure for one cell, and octadecyltrychlorsilane from Aldrich (silane) was used to obtain strong homeotropic alignment for other.

## RESULTS AND DISCUSSION.

The equilibrium texture of the first cell is a typical planar cholesteric texture with

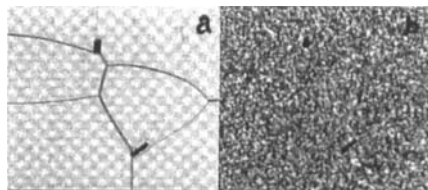


FIGURE 1. (a) equilibrium planar texture and (b) an intermediate polydomain planar texture obtained 100ms after electric field was removed. The spacer on all microphotographs is 5  $\mu$ m wide.

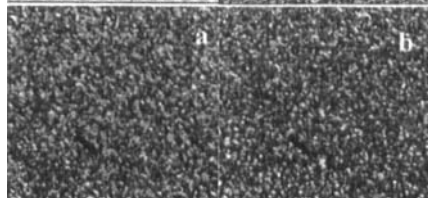
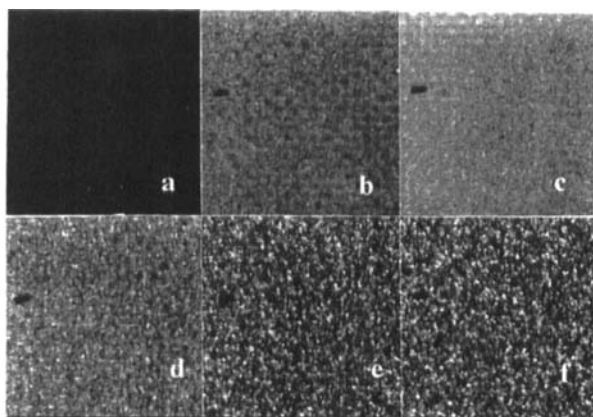
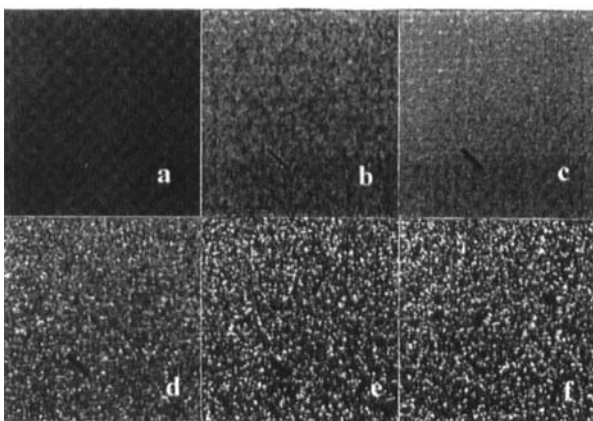


FIGURE 2. (a) equilibrium polydomain texture in the cell with homeotropic surface treatment and (b) an intermediate polydomain texture obtained 100ms after the electric field was removed.

characteristic oily streak defects (fig.1a). Short pulse application of an electric field caused the appearance of an intermediate polydomain structure (Fig.1b). Typically, this is a distorted planar texture consisting of small domains. The average size of the domain is about  $4\mu\text{m}$ . The helical axes of the domains are randomly tipped off from the substrate normal by an angle of about  $15^\circ$ [7]. This intermediate polydomain structure is not stable, however, and transforms slowly to equilibrium. The equilibrium texture in the second cell is a polydomain structure with large angular distribution of helices (Fig.2a). Short pulse application of an electric field result in an intermediate polydomain structure similar to that occurring in the planar cell (Fig.2b). The next relaxation process of the state depicted in Fig.2b only involves a small increase in the domain sizes.



**FIGURE 3.**  
Intermediate textures obtained:  
a)-500 $\mu\text{s}$ ;  
b)-750 $\mu\text{s}$ ;  
c)-1.25ms;  
d)-2ms;  
e)-5ms;  
f)-10ms after electric field was removed (planar cell).



**FIGURE 4.**  
Intermediate textures obtained:  
a)-500 $\mu\text{s}$ ;  
b)-750 $\mu\text{s}$ ;  
c)-1.25ms;  
d)-2ms;  
e)-5ms;  
f)-10ms after electric field was removed (homeotropic cell).

Fig.3 shows textural changes during the transition in the planar cell. Fig.4 shows textural changes during the transition in the cell with homeotropic surface treatment. The character of the texture changes in both cells during the first 10 ms after the electric field is turned off it seem to be surprisingly similar. During the first 500 $\mu$ s, the homeotropic structure is still present in the cells (Fig.3a,4a). After this time, a marble-like texture starts to form, remaining for 1.25ms (Fig.3b,4b). Our investigations of reflection spectra has shown the existence of a reflective state with  $\lambda_{\max} \sim 450$ nm in time interval  $\sim 500$ -1000 $\mu$ s [7] for both cells. At 1.25 ms the "perfect" transient planar texture occurs in the cells (Fig.3c,4c) with  $\lambda_{\max} = 975$ nm. After 1.25ms, small domains start to form (Fig.3d,4d). The density of domains reaches its maximum at approximately 4ms and then decreases gradually (Fig.3e-f,4e-f).

We can divide the relaxation process into four stages. First, a delay time, when the homeotropic structure is still present in the cells. Second, a fast relaxation period, when the transient planar structure is formed. The liquid crystal goes from the homeotropic through an intermediate conical structure to the quasiequilibrium transient planar state by changing the polar angle of director orientation (the angle

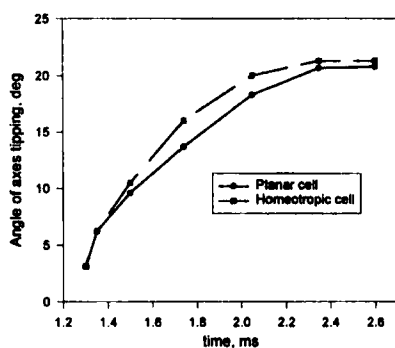


FIGURE 6. Plot of average axes tipping during nucleation process for cells with different alignment layer.

between the director and the normal to the cell surface) from 0 to  $\pi/2$  (about 1.25ms). Fig.3c and 4c shows that for both surface alignments this is a defect-free relaxation, occurring very fast. After the transient planar pitch is reached and the quasiequilibrium transient planar structure is formed, a subsequent twisting process is impossible without structure symmetry breaking. Third stage, when the equilibrium pitch is reached.

We observe that over the time interval from 1.25 to 4 ms, as the equilibrium pitch is being established, a polydomain texture is formed for both surface alignment layers. We may assume a nucleation process (from 1.25 to 4ms) gives the possibility for the system to form helices with the equilibrium pitch  $P_0$  (at 4-5ms approximately) [7]. On the other hand, the nucleation and simultaneous twisting processes requires tilting of the helices away from the vertical position. Time resolved spectral analysis of the wavelength of maximum reflection at various angles of observation [7] makes it possible to determine the average tipping angle of the helix axes. Fig.6 represents a plot of the tipping angle against time for both cells. Finally, at 5ms the pitch of the cholesteric structure is completely formed for both alignment layers [7]. However, as a result of this process, defects are seen to form in approximately the same way for both cells (Fig.3e and 4e). At 10 ms, there are still

many defects in the system for both alignment layers. So final fourth stage is a period of slow relaxation, when the equilibrium structure is formed.

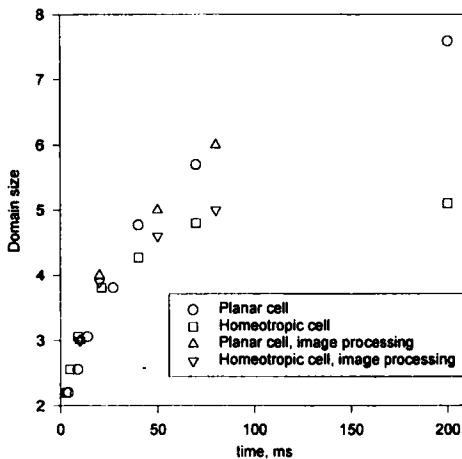
However, while the relaxation process is evidently very similar during the time scale of figures 3 and 4 for both alignment layers, the relaxation to the equilibrium texture is different. For the planar cell, there is a further slow relaxation to the final planar texture with few defects. This relaxation is obviously a slow process due to the slow removal of defects in the texture. For the silane alignment layer, it is seen that the defects that formed as the equilibrium pitch was obtained are stable.

To study changes of the domain size during the pattern relaxation we used the light scattering technique [8]. The scattering angle of the scattered light is determined by the spatial distribution of the optical non-homogeneities in scattering media. Namely, the intensity of scattering of the incident beam can be considered as the diffraction of light on the spatial harmonic of refractive index changes in the scattering media. The period  $\Lambda$  of the spatial harmonic is given by the formula

$$\Lambda = \frac{\lambda}{2 \sin(\theta/2)},$$

where  $\lambda$  is the wave length of the scattered light and  $\theta$  is the scattering angle. The domain pattern presents the complex optical structure possessing many characteristic scales. The largest characteristic scale is determined by the domain size, typically  $5\mu\text{m}$  and corresponds to the scattering angles about 7 degrees. Thus, the dependence of the light scattering intensity  $I_{sc}$  on  $\theta$ , and the change of  $\theta$  after the electric field is switched off follows the dynamics of the domain size changes. Figure 7 represent the

FIGURE 7. Domain size vs time.



dynamics of domain size during a relaxation process.

One could view the silane cell as having the faster relaxation, but our data shows that this view is incorrect. We can see from this data that a more accurate statement would be that the relaxation up to a time of about 100ms is very similar for both alignment layers, but the relaxation of the planar cell continues after that to a perfect planar structure, while the homeotropic cell does not.

Let us discuss now the shortest possible relaxation time for BCT. The typical time of transient planar formation can be estimated from the expression:  $\tau \approx \gamma P^2 / K_{22}$  [4], where  $\gamma$  is the rotational viscosity and  $K_{22}$  is the twist elastic constant of liquid crystal. For typical values of  $\gamma = 0.1 \text{ N}\cdot\text{s}/\text{m}^2$ ,  $K_{22} = 10^{-11} \text{ N}$  and  $P_0 = 3.75 \times 10^{-7} \text{ m}$  this time is  $\tau \approx 1.4 \text{ ms}$ . We can decrease this time by changing the liquid crystal material constants. For optimized mixtures we have obtained  $\tau \approx 0.8 \text{ ms}$  [9]. The further relaxation into the state with the equilibrium pitch  $P_0$  takes about 3 ms, while the minimum relaxation time can be estimated as 4 ms. This time does not include the final transformation of the polydomain texture to the equilibrium one. This means that future optimization of the BCT should make the process of domain evolution as short as possible.

### CONCLUSIONS.

In this paper we studied the relaxation process from the electric field induced nematic to cholesteric by performing direct texture change observations and analysis. We have shown here what is believed to be the first direct experimental confirmation that the transient planar structure is a defect-free structure, and that the relaxation from the transient planar to planar involves both a rapid pitch change and the nucleation of defects for both homogeneous and homeotropic alignment layers. The final long-term relaxation takes place through domain structure formation and a subsequent transformation to the equilibrium state.

### ACKNOWLEDGMENT.

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