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J. V. Gandhi

Kent State University - Kent Campus

X. D. Mi

Kent State University - Kent Campus

Deng-Ke Yang

Kent State University, dyang@kent.edu

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Effect of surface alignment layers on the configurational transitions in cholesteric liquid crystals

J. V. Gandhi, X.-D. Mi, and D.-K. Yang*

Liquid Crystal Institute, Kent State University, Kent, Ohio 44242

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To understand the mechanism behind the pitch change during the transient planar to the planar state transition of a cholesteric liquid crystal (while it is relaxing from the homeotropic state to the planar state), we studied a more controlled situation of pitch change in a thermochromic liquid crystal. We studied the pitch change in the liquid crystal, as a function of temperature, under various surface alignment conditions such as homogeneous and homeotropic surface alignments. In a cell with homogeneous surface treatment, the liquid crystal showed a discontinuous pitch change, while the pitch change was rather continuous in a cell with homeotropic treatment. The numerical simulations also showed an existence of continuous pitch change in a homeotropic cell, but not in a homogeneous one. This led to a prediction that when a cholesteric material is switched from the homeotropic state to the planar state, the transition may be much faster for the homeotropically treated cell as compared to the homogeneously treated cell. We performed experiments to verify this prediction with a chiral nematic mixture. Indeed, the homeotropic to planar state transition time of a homeotropic cell was 5 msec, much faster compared to about 200 msec for corresponding transition in a homogeneous cell. [S1063-651X(98)12306-1]

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I. INTRODUCTION

Cholesteric liquid crystals (ChLC) are chiral nematic materials. ChLC molecules like to align themselves, on the average, along a common director. In addition to that, they also have a helical structure in which the director is twisted uniformly in space as a function of position along the helical axis, perpendicular to the director. The helix is characterized by its intrinsic pitch P_0 , which is the distance along the helical axis required to twist the director by an angle of 2π . When either a small amount of polymer is dispersed in the ChLC (known as polymer stabilized cholesteric textures or PSCT) or a suitable surface alignment layer is used on glass surface (known as surface stabilized cholesteric textures or SSCT), the ChLC, sandwiched between two glass plates, exhibits two stable states (textures) at zero field [1–4]. These two states have substantial optical contrast between them and hence are of great interest for display applications. One state is the reflecting planar state (or texture) in which the helix axes are perpendicular to the cell surface and reflects colored light depending on the pitch of the ChLC. The wavelength of the light reflected is given by $\lambda_0 = \tilde{n}P_0$ where \tilde{n} is the average refractive index of the liquid crystal. The other state is the scattering focal conic state in which the helix axes are oriented more or less parallel to the cell surface.

A cholesteric liquid crystal material with positive dielectric anisotropy ($\Delta\epsilon$) can be switched from the planar (P) state to the focal conic (F) state by applying a small electric field but the reverse is not true. To switch the material from the focal conic to the planar state, the material has to be first switched to the homeotropic (H) state (where all the molecules are aligned perpendicular to the cell surface) and then it relaxes to the planar state under low field or the focal conic state under high field. The configurational transition from

homeotropic to planar state (H - P) is a slow process. In order to utilize the ChLC for display applications, this H - P transition time needs to be reduced. A clear understanding of the mechanism of the transition is required to reduce the H - P transition time of the ChLC. It is known that the ChLC in the homeotropic state first undergoes a rapid homogeneous transition to a transient planar (P^*) state [5]. In this transient planar state, the pitch of the ChLC is about twice its intrinsic pitch [6]. This transient planar state then relaxes to the planar state with the intrinsic pitch P_0 . Depending on the surface anchoring, the second transition may be a nucleation transition and slow. Surface alignment layers and polymer networks have been shown to play an important role in reducing the transition time in a PSCT [7]. Our motivation for this investigation was to understand the mechanism behind the pitch change from transient planar state to planar state when different surface alignment layers including homogeneous, random tangential and homeotropic alignment layers are used and to further predict and demonstrate effects of the surface alignment layers on the H - P transition times.

Towards this goal, we carried out the following experimental investigation. Our aim in this experiment was to simulate pitch change in the ChLC at a rate slow enough to be studied carefully. We studied a liquid crystal system where pitch change could be induced by varying temperature. We used a thermochromic liquid crystal (TLC), which exhibited a wide pitch change over a relatively short temperature range. We varied the surface boundary conditions using various alignment layers including homogeneous, random tangential, and homeotropic surface treatments and observed the corresponding effects on the temporal changes in the pitch. From these results, we predicted the effect of surface alignment layer on the H - P transition time. To test our prediction, in a separate experiment, we measured the H - P transition time of a chiral nematic mixture in a cell with different surface treatments at room temperature. Comparison of the transition time of a cell with homeotropic treat-

*Author to whom correspondence should be addressed.

ment with the transition time of a cell with homogeneous treatment clearly showed the dramatic effect of homeotropic treatment in reducing the H - P transition time, completely agreeing with the prediction from the first experiment.

This paper is organized in the following manner. In Sec. II, we present the details of the materials and experimental arrangement. In Sec. III, results of the first experiment and related numerical simulations are presented. The results of the second experiment are reviewed in the light of the results of the first experiment. In Sec. IV, we present the conclusion.

II. EXPERIMENT

For our study of pitch change with temperature, we chose TM75A (manufactured by BDH Chemicals Ltd.) as the thermochromic liquid crystal (TLC) because it undergoes a smectic- A to cholesteric transition at a rather low temperature of 41.3 °C and a cholesteric-isotropic transition at 53.2 °C. As temperature is increased, from the infinitely long pitched smectic- A phase, the TLC changes to the cholesteric phase with the natural pitch changing from long IR wavelengths through short UV wavelengths and then to the isotropic phase. In the cholesteric phase, within a very short temperature range from about 41.6 °C to 42.3 °C, when the pitch is comparable to vacuum wavelength of light in the visible region divided by the refractive index of the liquid crystal, ChLC reflects visible light and hence can be used as a tool to measure the effects of the temperature change.

Glass cells with the following surface alignment layers were used: (1) homogeneous alignment layer, (2) random tangential alignment layer, and (3) homeotropic alignment layer. For the homeotropic alignment layered cells, in the cholesteric phase, the TLC exhibited the scattering focal conic state. To obtain a reflective planar state for ease of investigation, we added about 10% by wt. of a negative $\Delta\epsilon$ material, ZLI2806 (manufactured by E. Merck). The mixture had a negative $\Delta\epsilon$. Then application of electric field forced the LC molecules to align in the planar state, which strongly reflected light and the changes in the pitch were much more evident. We used the same mixture in the homogeneous and tangential cells, which produced planar states with a fewer number of defects upon the application of electric field. We confirmed that addition of a relatively small amount of negative $\Delta\epsilon$ material did not drastically change the relevant properties of the TLC. The cells were coated with polyimide to obtain random tangential boundary conditions and further rubbed to obtain homogeneous alignment layer. The cells with homeotropic boundary conditions were prepared by coating a layer of ODTCS (Octadecyltrichlorosilane) on SiO₂ overcoated indium-tin-oxide (ITO) glass.

Glass cells of about 5 μm thickness were filled with the TLC mixture using vacuum filling method. At room temperature, the TLC was in the smectic- A phase with a very high viscosity. After vacuuming and lowering the cells in the TLC mixture, they were heated in an oven. Upon heating, the TLC changed to the isotropic phase, which had a much lower viscosity and filled up the cells under air pressure. The cells were then cooled down, compressed, and sealed.

The incident light source was composed of a voltage regulated halogen lamp. A polarizing microscope was used in the experiment because it was important to focus on a very

small region of the cell comprising of only one or a few domains. The region of study in the first experiment was only about 200 μm in diameter. Two types of investigations were performed. In the qualitative investigation, a video camera and a VCR were used to record the change in domains in the TLC as a function of temperature. This provided a qualitative picture of the pitch change as a function of temperature. In a quantitative investigation, the spectrum of the light reflected by the TLC cell was measured under crossed polarization and the center wavelength of the spectrum was measured as a function of temperature. The light reflected from the TLC cell entered an ORIEL MS257 imaging spectrometer via an optical fiber. The measurement of the light intensity was done by a charge-coupled device array and data were collected using ORIEL Instaspec Software. The temperature of the LC cells was controlled using an INSTEC temperature controller. The temperature as well as the rate of change of temperature could be controlled through a software with temperature accuracy better than 0.005 °C. The temperature was changed in steps of 0.01 °C for the spectrometer measurements.

In the second experiment, to measure the H - P transition time, we used as the cholesteric liquid crystal a chiral-nematic mixture composed of 24 wt. % E44 (nematic) and 76 wt. % BLO61 (chiral) (both manufactured by E. Merck). This mixture reflected light with a pitch of 0.37 μm (or a vacuum wavelength of 590 nm). We measured the H - P transition time of this ChLC mixture filled in a cell with homeotropic alignment layer as well as in a cell with homogeneous alignment layer (PSCT). The cells were filled in a manner similar to the above, except that it was not necessary to heat the material since the material was already in the low viscosity cholesteric phase at room temperature. Black paint was applied on the back of these cells so that the scattering focal conic state would appear black. White light passed through a monochromator before falling on the cell at an incident angle of about 22.50°. The dynamic response of the cells was measured at the wavelength where its reflectance was maximum. The reflected light was detected at the specular angle by a silicon diode detector. Crossed polarizers were used to minimize the reflection from glass-air interfaces. A 1 kHz square wave pulse of 60 V amplitude and 100 msec duration was applied to the cells to switch them to the homeotropic state. Immediately after the removal of the pulse, as the material started to relax to the reflecting state, the reflectance of the cell was measured as a function of time.

III. RESULTS AND DISCUSSION

In this section, we present our results in the following manner. In part A, we present the results of the study of pitch change in TLC as a function of temperature for different alignment layers. We will present the qualitative results of the video recording along with the quantitative results of the spectrometer measurements. Effects of surface alignment layers on the pitch change mechanism will be highlighted and related numerical simulations will also be presented. In part B, we present the results of the measurement of the H - P transition time for homeotropically aligned cells and the homogeneously aligned cells. These results will be compared in the light of the results from the previous experiment on the

mechanism of pitch change for different surface alignment layers.

A. Pitch change of the TLC with temperature

a. The cell with homogeneous alignment layer: At room temperature, the TLC cell was in the smectic-A phase. Under the microscope, it appeared to have a fan-type structure. On heating the sample slowly (about 0.1 °C/min), these patterns melted into a gray region as the temperature increased towards the smectic-cholesteric transition. On further heating at a slow rate, the material changed into the cholesteric phase. In our cells, because of the polyimide surface treatment and rubbing, the material was in the planar state in the cholesteric phase. Application of electric field further enhanced the reflectivity of the cell by producing a more uniform planar state (i.e., with larger domains and less defects). Different colors started to appear in these large domains. These colors appeared in strict order from yellow-green towards blue as the temperature was increased. When the material undergoes a phase transition from the smectic-A phase to the cholesteric phase and then to the isotropic phase, the pitch of the cholesteric phase would shift from longer wavelengths (red) to shorter wavelengths (blue). The longer pitches would occupy a shorter temperature interval than the shorter pitches [8,9]. We observed the above two characteristics in our studies of a pure TLC, without any additive like the negative $\Delta\epsilon$ material. When we added the negative $\Delta\epsilon$ material to the TLC, we could not observe the long pitch region (red) of the transition, but only the mid range (yellow-green) to short pitches (blue) could be observed. It may be possible that the addition of negative $\Delta\epsilon$ material would have reduced the already short temperature range over which long pitches could exist to even smaller value that we could not observe them. But other features, described below, of the transition of the TLC with and without the negative $\Delta\epsilon$ material were almost identical, giving us reasonable confidence that our study of the mixture gave us useful information about the transitions of the pure material subjected to different alignment layers.

The pitch of the cholesteric-phase is forced to obey the surface boundary conditions. In the case of a homogeneous alignment layer, this would mean that an integral number of half pitches have to fit in the cell. In that case, the pitch change with temperature is expected to be discontinuous. Our observation of the video tape supported this expectation. The pitch in a single domain remained constant over a small range of temperature. Further change in temperature then induced a pitch jump. The variation of the pitch or color always originated from disclinations, separating two domains with different pitches, e.g., in a cell with thickness d , adjacent domains with pitches $P_1 = 2d/m$ (m being an integer) and $P_2 = 2d/(m+1)$ will have a disclination between them. As temperature increased, this line moved from domain with pitch P_2 to domain with pitch P_1 [10]. These disclinations seemed to originate from fixed points on the surface during different temperature runs and provided nucleation centers for the pitch change. The range of the temperature over which the pitch was constant also increased as the pitch decreased (towards blue). This is to be expected because twist speed may increase as the pitch decreases, thereby requiring

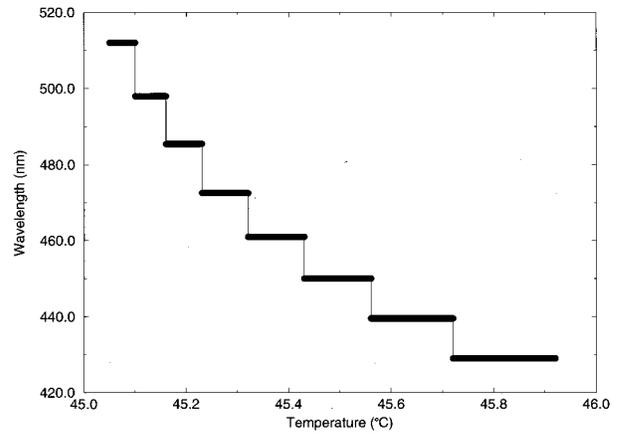


FIG. 1. Center wavelength of the reflection spectrum of the homogeneous cell as a function of temperature.

higher thermal energy to induce a pitch change.

The spectrum of the reflected light from the TLC was characterized by its center wavelength corresponding to the pitch of the ChLC. Figure 1 shows the center wavelength as a function of temperature. (The pitch of the TLC that can reflect light of this wavelength would be equal to this vacuum wavelength divided by the average refractive index of the TLC.) Due to the design restriction of our temperature controller setup, only the relative value and not the absolute value of the temperature should be noted. As Fig. 1 shows, the change of pitch of the TLC was discontinuous with temperature. The pitch (or the wavelength of light reflected) stayed constant over a certain range of temperature. Then as the temperature increased, the pitch jumped to a lower value. The measured wavelength of the reflected light agreed well with the values calculated using the simple model of allowing only an integral number of half pitches in a 5 μm thick cell. Figure 2 shows the plot of inverse wavelength versus the number of half pitches in the cell. The straight line fit supports the above simple model.

The rapid change in the pitch or color of the ChLC with temperature is described as ‘pretransition effect’: it occurs as the helix of the ChLC unwinds when temperature is de-

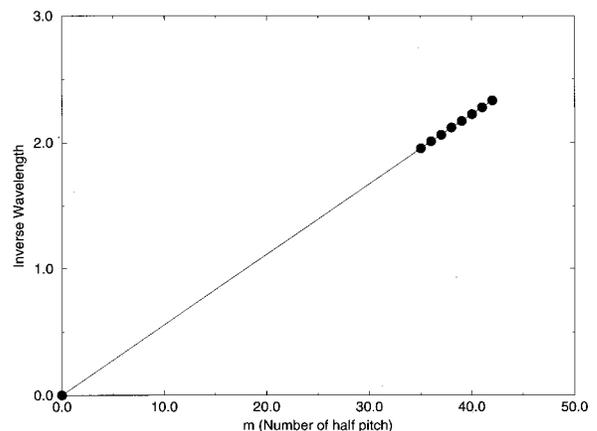


FIG. 2. Inverse wavelength vs number of half pitches for the homogeneous cell. The fit assumes that the only integer number of half pitches are allowed in a homogeneous cell.

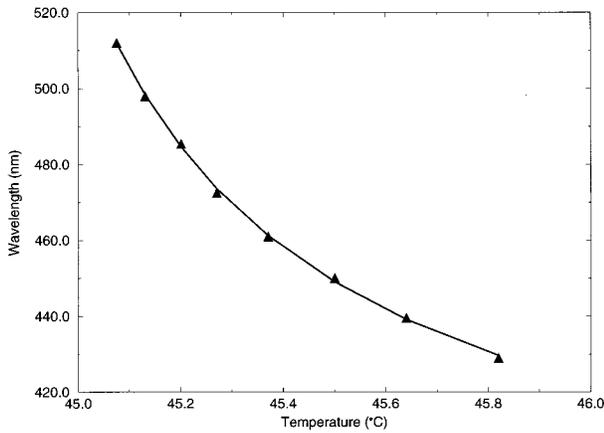


FIG. 3. Center wavelength of the reflection spectrum vs the center of the temperature range over which the wavelength remains constant. The fit is obtained using Eq. (1). See text for more details.

creasing just before the cholesteric-smectic phase transition. Theoretical and empirical models have been presented to describe the pitch change with temperature [11–13,8]. We fit our data with the following theoretical model. It describes the pitch change as [12]

$$\lambda = \gamma \left(1 + \frac{\beta}{T - T_0} \right)^2, \quad (1)$$

where γ, β are constants and T_0 is a temperature that is usually lower than the transition temperature.

The measured wavelength of the ChLC was fit against the center of the temperature range over which it stays constant. The result of the fit is shown in Fig. 3. There is a good agreement in the dependence of the pitch on temperature with the theoretical model used to describe the pretransition effect [12]. The best fit parameter values are $\gamma = 372 \pm 4$ nm, $\beta = 0.096 \pm 0.01$ °C, and $T_0 = 44.52 \pm 0.04$ °C. The values of γ and β will be compared with the best fit values of the homeotropic cell below.

b. The cell with random tangential alignment layer: For the random tangential cell, the behavior of the TLC was similar to that of the homogeneous cell. The material was in the planar state when it was in the cholesteric phase. But the domains were smaller than the domains formed in the rubbed cell. Accordingly, the number of defects also increased. Upon heating of the cell, changes of colors or pitches and growth of domains were quite similar to the one for the homogeneous alignment layered cell.

c. The cell with a homeotropic alignment layer: In the smectic-A phase, this cell showed a fan-type focal conic structure. In the cholesteric phase, because of the intrinsic twist and the surface anchoring energy, the material was not in either the planar or homeotropic state but was in the focal-conic state. At the surface, the molecules were expected to be aligned perpendicular to the surface. We induced a planar state by applying electric field to the cell. A trick to obtain a more defect-free planar state was to cool from the isotropic phase to the cholesteric phase rather than to heat from the smectic-A phase to the cholesteric phase, then apply high electric field to obtain a planar state. Although this planar state was not as reflective as the one in the homogeneous

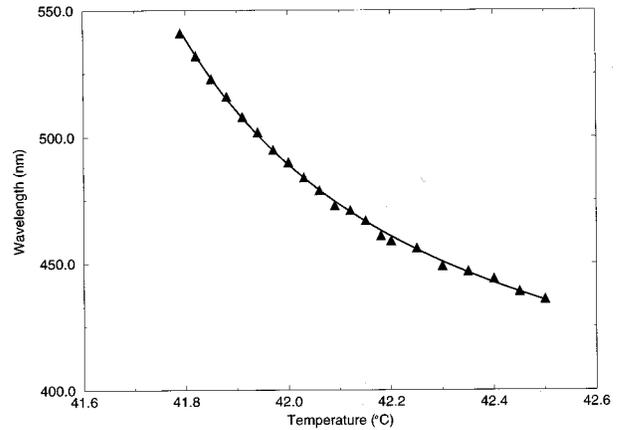


FIG. 4. Peak wavelength of the reflection spectrum as a function of temperature of the homeotropic cell. The fit is obtained using Eq. (1). See text for more details.

cell, the change in the pitch could be clearly observed. The domains were small and not clearly defined by domain boundaries or defect lines as in the case of the homogeneous cell. Many different shades of one particular color could be seen, indicating that the pitch might not be the same even in closely spaced domains. Since the pitch was extremely sensitive to the temperature and it was not possible to achieve a very uniform temperature field, some variations in the color of the domains could be attributed to the slightly non-uniform temperature field.

On heating at a slow rate, we found a strikingly different behavior as compared to the homogeneous cell. The pitch change was not discontinuous as in the case of the homogeneous cell. The change of pitch was observed to be a smooth one. Also the change of pitch did not necessarily arise from defect lines, but was rather a more subtle change. We chose a very small region of about $200 \mu\text{m}$ in diameter and measured the spectrum of light reflected from the TLC. The spectrum differed dramatically from that of the homogeneous cell. It was more of a bell-shaped curve. Figure 4 shows the peak or the center wavelength of the reflection spectrum as a function of temperature for the homeotropic cell. Again, due to design differences in the two cells, only the relative values and not the absolute values of the temperature should be compared with corresponding values of the homogeneous cell. The peak wavelength of the spectrum decreased continuously as temperature was increased. This clearly indicates that the pitch of the cholesteric phase in the homeotropic cell did not jump with temperature as it did in the homogeneous cell. Such a continuous change can be expected for a homeotropic cell, because the boundary conditions here are completely different. As the temperature is varied, the helical structure was “injected” from the surface in such a way that the molecules in the bulk exist in their intrinsic pitch which would change in a continuous manner with the temperature.

Our numerical simulations also showed the same aspects. We started from the Oseen-Frank free energy. Using the idea of the discretization method for the tensor representation of the free energy [14], we were able to study the dynamic process from homeotropic texture to transient planar texture, and then to the planar texture. The detailed studies will be

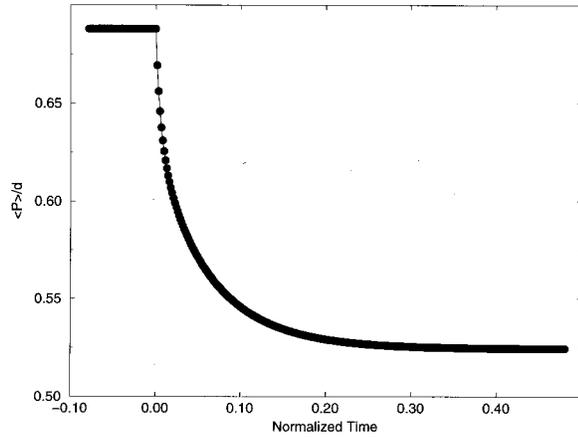


FIG. 5. The normalized average pitch vs normalized time τ (in units of $\gamma_{\text{visc}}d^2/K_{22}$) in a homeotropic cell. Relaxation is triggered by a sudden intrinsic pitch change from 1/3 to 2/5, starting from the equilibrium state with the intrinsic pitch of 1/3.

presented somewhere else [15,16]. We chose homeotropic texture with small random fluctuation as initial conditions, and used fixed boundary conditions at surfaces and translational invariant boundary for the other two coordinates. For the homogeneous surface, the system would stay at the transient planar texture forever, which implied that there was no smooth way to relax to the perfect planar texture from the transient planar texture. (In our simulations defects are not considered.) However, for the homeotropic surface, the simulation did show a continuous change from the transient planar to the intrinsic planar texture. Figure 5 shows that the normalized average pitch $\langle P \rangle/d$ changes continuously with the reduced time τ . Here τ was normalized by $\gamma_{\text{visc}}d^2/K_{22}$, $\langle P \rangle \equiv 2\pi / \langle \partial\Phi / \partial z \rangle$ (the average was taken over across the cell), ϕ was the azimuthal angle, and z was the coordinate perpendicular to the cell surface. The elastic constants were chosen such that $K_{11} = K_{33} = 2K_{22}$. We chose as the initial condition the equilibrium state of the cholesteric liquid crystal with intrinsic pitch of $d/p_0 = 2.5$, and then we changed the intrinsic pitch artificially to $d/p_0 = 3$. This simulation showed a continuous relaxation to a new equilibrium state that corresponds to $d/p_0 = 3$. This process was used to simulate the smooth pitch change when the intrinsic pitch decreased due to increase in temperature.

Fitting the data with the theoretical equation [Eq. (1)] shows a similarly good fit with the following best fit parameters: $\gamma = 357 \pm 4$ nm, $\beta = 0.134 \pm 0.1$ °C, and $T_0 = 41.21 \pm 0.04$ °C. Figure 4 shows the data and the fit using the theoretical equation. The agreement of parameters γ and β with corresponding homogeneous cell parameters is also quite good.

Thus surface alignment layers have a dramatic effect on the way the pitch of a TLC changes with temperature. The homogeneous cell undergoes a discontinuous and largely nucleation driven pitch change while the homeotropic cell undergoes a more continuous pitch change whose origin may be at the surface. It can be expected that the inherently slow nucleation driven pitch change in a homogeneous cell may result in a very slow H - P transition. On the other hand, the homeotropic alignment layer allows a continuous pitch change. The pitch can be added from the surface and such a

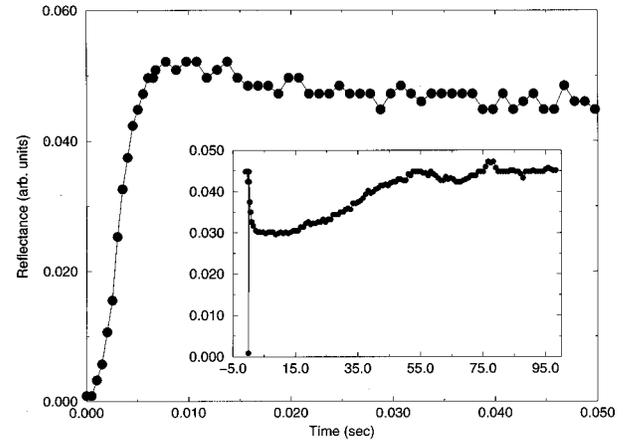


FIG. 6. Reflectance vs time of a homeotropic cell after the voltage is removed. The inset shows the long time behavior of the reflectance.

process can be very fast depending on the energy difference between the final and the initial states. The following results of the H - P transition times of the two types of cells agree with our above prediction.

B. Homeotropic-planar transition time

Our experiments with the thermochromic materials, as described above, clearly indicate dramatically different mechanisms of pitch change for homeotropic and homogeneous surface alignment layers. Such a pitch change is also involved when a ChLC relaxes from a homeotropic state to a planar state (H - P). It does so by first relaxing rapidly to a transient planar state, which then further relaxes to a planar state through a pitch change. The different mechanisms of the pitch change in the two different alignment layers (as pointed out in part A of this section) may give rise to different transition times for the homeotropic to planar state transition. We expect that the continuous change of pitch in a homeotropic alignment layer may result in a faster completion of the configurational change than the discontinuous, nucleation driven pitch change process of the homogeneous alignment layer.

Our experimental result below shows, in agreement of our expectation from our experiment above, that the homeotropically aligned cell shows a much faster transition time as compared to the homogeneously aligned cell. Figure 6 shows the dynamic response of the homeotropic cell, which is measured as the reflectance versus time after the voltage pulse is removed. The transition time is defined as the time to reach from 10% of the maximum reflectance to 90% of the maximum reflectance. For the homeotropic cell, it is about 5 msec as can be seen from the figure. Figure 7 shows the reflectance of the homogeneous cell, which is measured as the reflectance versus time after the voltage pulse is removed. The H - P transition time for homogeneously aligned PSCT cell is about 200 msec. This transition time agrees quite well with literature results [7]. The homeotropic cell shows a much faster transition from the H to the P state as compared to the homogeneous cell. This result is very encouraging. The transition time for the homogeneous cell is longer because it may be going through an inherently slower nucleation process.

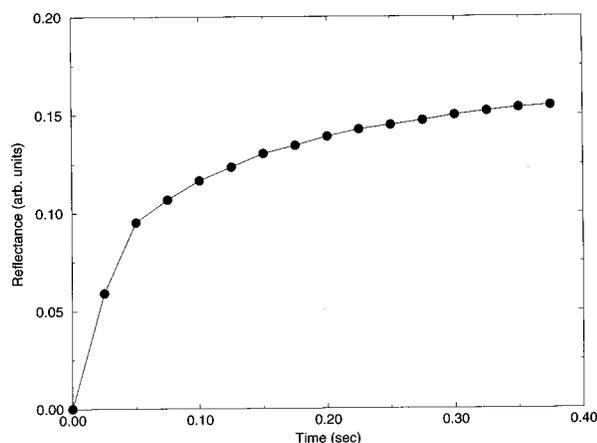


FIG. 7. Reflectance vs time of a homogeneous cell after the voltage is removed.

Pitch cannot change continuously and carrying out the large discontinuous jumps in the pitch may require crossing large energy barriers. This also requires the availability of defects in the cell. For the homeotropic cell such a process may not be required because the surface may not hinder any pitch change. In fact, surface may be helping in the process of pitch change required to bring the free energy to its minimum.

The fast H - P transition time that we have measured for the homeotropic cell also clarifies another important issue. Lu [7] reported that their polymer stabilized finger-print cholesteric texture (PSFPCT) show similarly fast H - P transition time of about 10 msec. Their cells were prepared by filling a cell with homeotropic alignment layer with a cholesteric mixture and photocurable monomer. Two parameters, the homeotropic alignment layer and the polymer network, may be responsible for the fast response in their cells. Our experiment above serves to separate the influence of the two parameters and clearly indicates the principal role played by the homeotropic alignment layer in inducing a fast homeotropic to planar transition. The effect of the polymer network does not seem to be as important as the homeotropic alignment layer. Polymer-free displays greatly simplify the manu-

facturing processes because a major step of polymerization is eliminated. This is a significant result that can have a tremendous effect during the manufacturing of liquid crystal displays.

The inset in Fig. 6 shows the long time behavior of the reflectance of the homeotropic cell. Following a rapid increase in the reflectance, there is a shallow dip. The slope of the dip is very small though. The reflectance returns to its maximum value in about 1 min. Since displays are usually refreshed faster than that, the decrease in the reflectance is not noticeable. The cell with homogeneous surface alignment layer typically show a monotonic increase in reflectance to its maximum value as can be seen from Fig. 7.

IV. CONCLUSION

In hopes of understanding the mechanism behind the pitch change that occur when a cholesteric liquid crystal undergoes transient planar to planar state transition, we have studied the role of surface alignment layers on the pitch change with temperature of thermochromic liquid crystals. From this experiment, we found that a homogeneously aligned cell experiences discontinuous pitch change and requires nucleation centers for the origination of one. On the other hand, in a homeotropically aligned cell, the pitch change is smooth and mostly independent of nucleation centers. From this, we predicted that a homeotropic alignment layer will enhance the speed of transient planar to planar state transition, in comparison with a homogeneous alignment layer. Accordingly, we measured the homeotropic to planar transition times of chiral nematic materials in homeotropically and homogeneously aligned cells. In agreement with our prediction, the homeotropic cell indeed showed a much faster transition time (5 msec) as compared to the homogeneous cell (200 msec). This fast cholesteric display is of great importance in video rate applications.

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