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Studies of the Temperature-Pitch Variations in Planar Cholesteric Layers and Surface Anchoring

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Analysis of the temperature dependence of the measured optical characteristics of planar oriented cholesteric (CLC) layers shows that just before the change of the number of half-pitches within the layer thickness by one, the director direction at the surface essentially differs from the alignment direction and the jump temperature is dependent on the direction of the temperature change (cooling or heating). A theoretical description of this hysteresis is presented on the basis of a continuum theory of elasticity of CLC's that takes also account of the surface anchoring. Because the observed hysteresis and deviation angle are dependent on the molecular anchoring forces at the surface the corresponding measurements fitted to the developed theory are used to obtain information about the anchoring potential. The possible ways of the, model independent, anchoring potential reconstruction and the different jump mechanisms are discussed.

Keywords: surface anchoring; hysteresis

INTRODUCTION

For completely aligned planar oriented cholesteric samples, the number of half-pitches in the sample thickness depends on the temperature and only at restricted temperatures an exact number of half-pitches will fit in the sample thickness. In previous publications^[1-3] we demonstrated that this can be seen in the transmission spectra of normal incident circular polarized light. These spectra can be described by the 4-wave model^[4,5] in which 6 parameters characterize the shape of the spectra. By introducing an angle which takes the devia-

tion of the director direction from the alignment direction at the sample surface into account the 4-wave model gives also a good description of the experimental spectra for non integer half-pitch numbers. This is demonstrated in Figure 1.

The parameters used in the calculations are:

- α Ellipticity of the incident light
- ξ Angle between polarization direction of incident light and director orientation at the entrance surface.
- Φ Deviation angle between director direction in layer and at entrance surface.
- r Ratio of refractive indices of the external media and the CLC.
- δ Dielectric anisotropy.
- N Sample thickness expressed in equilibrium number of half-pitches.
 $N=2d/p$; p is the pitch and d is the layer thickness.

The solid line is the calculated spectrum. Circles are the experimental points

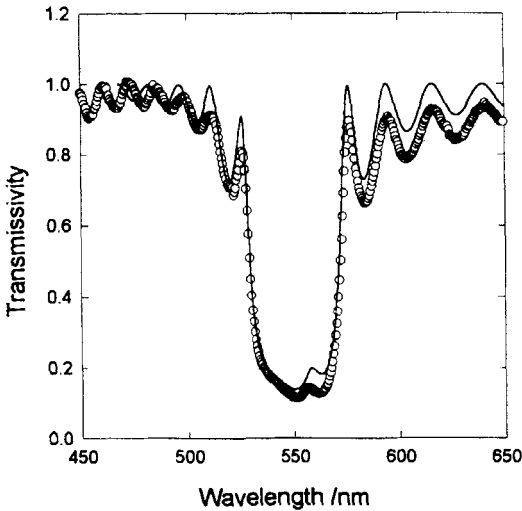


FIGURE 1. Transmission spectrum $T=40.30^{\circ}\text{C}$; $d=4.8\mu\text{m}$

The calculated spectrum is a combination of 2 spectra with weight factor w . The deviation from the director orientation at the specimen surface from the alignment direction at the jump temperature has been compared with a continuum theory of elasticity of CLC's taking account of the surface anchoring. This gives a quite good quantitative description. Subsequent measurements

showed temperature hysteresis which depends strongly on the specimen thickness and the anchoring at the surface of the layer. The parameters for the temperature hysteresis for the cholesteric pitch are determined for a mechanism where the director at the surface slips through a potential barrier.

EXPERIMENTAL

The temperature dependence of the parameters of the spectra has been determined in 2 samples of a 60% racemic mixture of CE6^[1]. A thick sample with $d=18\mu\text{m}$ and a thin sample with $d=4.8\mu\text{m}$. The temperature dependence of the pitch in the bulk CLC was assumed to be the same as in the $18\mu\text{m}$ sample. It can be described by the relation^[6,7]

$$p=p_{00}[1+a(T-T_{sc})]+b(T-T_{sc})^v \quad (1)$$

The transition temperature T_{sc} depends on the thickness of the sample. As shown earlier^[2,3], the spectra are described by a theory with non-integral number of half-turns of the cholesteric helix over the thickness of the sample.

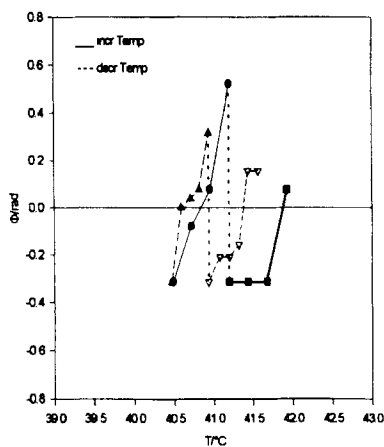


FIGURE 2. Temperature hysteresis in $4.8\mu\text{m}$ sample.

The non-integer number of half turns is given by $N \pm \Delta N$ and $\Delta N=2\Phi/\pi$.

Figure 2. demonstrates the presence of temperature hysteresis for the pitch jump in the specimen with $d=4.8\mu\text{m}$. In this example the jump is from $N=30$ to $N=32$ and vice versa and $N=31$ is not present. The deviation angle Φ is plotted in function of the temperature. The lines are as a guide to the eye.

TEMPERATURE HYSTERESIS OF THE PITCH JUMP IN THE CONTINUUM THEORY OF ELASTICITY

The condition^[2] for a jump in the cholesteric pitch resulting in a restructuring of the director configuration from a cholesteric helix with N half-turns to a helix with $N+1$ half-turns over the layer thickness, corresponds to the temperature at which the free energies F for these configurations are equal. However, in order for the transition between the director configurations to occur, the system must overcome an energy barrier separating the initial and final states. This is a reason for possible hysteresis observation in the temperatures of the pitch jump during heating and cooling of the layer. It is obvious that the hysteresis depends on the surface anchoring. The mechanism whereby the director configuration in the layer is actually restructured is very important. One possibility is a continuous rotation of the director at the surfaces of the layer, resulting in deflections away from the alignment direction. Another possibility is sharp fluctuation-induced variations in the director structure in the volume of the cholesteric layer. The second possibility cannot be described by the standard continuum theory of elasticity of cholesterics, where only spatial derivatives up to the second order of the director field appear. Therefore we shall consider only the first case in which the potential barrier between configurations with N and $N+1$ half-turns of the director in the layer is overcome in a continuous manner during the transformation. To find the temperature behaviour of the deviation angle Φ we write the free energy of the layer as

$$F(\Phi) = 2W_s(\Phi) + \frac{K_{22}}{2} \left[\frac{2\pi}{p_{N+\Delta N}} - \frac{2\pi}{p(T)} \right]^2 d \quad (2)$$

K_{22} is the elastic torsional modulus, $W_s(\Phi)$ is the surface anchoring potential, $p(T)$ is the equilibrium value of the pitch in a bulk CLC, $p_{N+\Delta N}$ is the value of the pitch in the cholesteric layer. We assume that the alignment direction and the surface anchoring force are identical for both surfaces of the layer and therefore $\Delta N=2\Phi/\pi$. By minimizing $F(\Phi)$ in Eq.(2) we obtain the following expression for $\Phi(p(T))$

$$\frac{\partial W_s(\Phi)}{\partial \Phi} + K_{22} \left[\frac{2\pi}{p_{N+\Delta N}} - \frac{2\pi}{p(T)} \right] = 0 \quad (3)$$

For a fixed T there exist several solutions of Eq.(3) which correspond to different values of N in the layer. For finite anchoring forces the number of possible solutions is limited by the condition $\Phi < \Phi_c$. The critical angle Φ_c is determined by the form of the anchoring potential $W_s(\Phi)$. Only one of the solutions is stable and we denote the value by N_0 . The temperature $p(T)$ of the transition between configurations differing by one half-turn is determined by the temperature for which $\Phi = \Phi_c$. The computed $\Phi(T)$ for a Rapini model surface potential^[8] $W_s = -W \cos^2(\Phi/2)$, is displayed for strong anchoring in Fig. 3. $S = 4\pi K_{22}/Wp$. The curves above the abscissa refer to increasing temperatures and below the abscissa to decreasing temperatures.

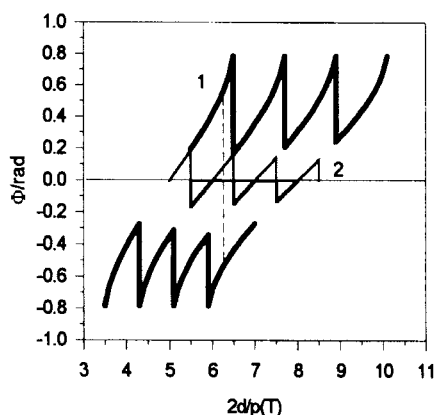


FIGURE 3. Computed deviation angle Φ in function of T for $S = 5$ and $N = 5, 6, 7, 8$; 1—hysteresis present, 2—hysteresis absent.

The potential barrier between the director configurations with N and $N+1$ half-turns is determined by ^[8]

$$B(N_0, T) = F(N_0, \Phi_c) - F(N_0, \Phi(T), T) \quad (4)$$

The computed temperature dependence for the height of the potential barrier for the same conditions as in Fig. 3, is displayed in Fig. 4.

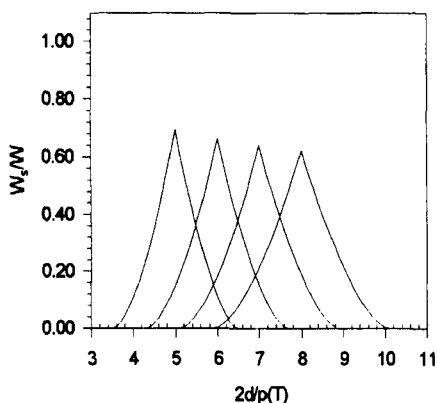


FIGURE 4. Temperature dependence of potential barrier

The transition temperature for a jump from N to $N \pm 1$ half-turns can be obtained from Eq.(3) by substituting $\Phi = \Phi_c$ ^[8]. The assumption that W_s is an even function of Φ leads to the following equation for Φ_c :

$$W_s(\Phi_c) = \frac{1}{2} W_s\left(\frac{\pi}{2}\right) = \frac{1}{2} W_{\max} \quad (5)$$

The admissible values for Φ are restricted by the condition $-\frac{\pi}{4} < \Phi < \frac{\pi}{4}$

since it follows from formula (5) that $\Phi_c = \pi/4$ for the Rapini potential.

The difference between the number $2d/p(T)$ of half-turns across the layer for the equilibrium value of the pitch and N half-turns in an unstressed configuration $\Delta N_j = N - 2d/p(T)$, found from Eq.(3) is described by

$$|\Delta N_j| = \frac{Wd}{2\pi K_{22}} + \frac{1}{2} = \frac{N}{S} + \frac{1}{2} \quad (6)$$

The jump hysteresis can be found from

$$\Delta N_h = 2d \left[\frac{1}{p(T)_{N \rightarrow N+1}} - \frac{1}{p(T)_{N+1 \rightarrow N}} \right] = \frac{Wd}{\pi K_{22}} = 2 \frac{N}{S} \quad (7)$$

As can be seen from Fig.3. that,for strong anchoring ($S=5$) after the jump through the potential barrier ($\Phi_c=\pi/4$), the jump director configuration does not correspond to the configuration with minimum free energy for the jump temperature. $|\Phi_j^s| > |\Phi_j|$ where Φ_j is the deflection angle at the jump point and Φ_j^s is the actual realized deflection angle. This also holds for weak anchoring forces and large N .

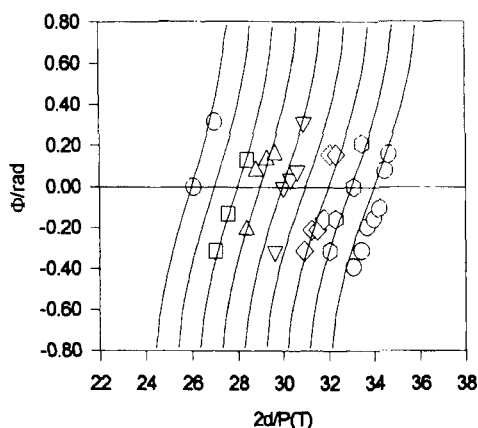


FIGURE 5. Temperature dependence of the deflection angle Φ for a $4.8\pi\text{m}$ thick sample (cooling). Calculations are for $S=25$. Different symbols for experimental points with $N=26 \rightarrow 34$.

Analysis of the hysteresis phenomena based on the mechanism whereby the director slips through a barrier for the parameters of the problem which corresponds to the experiment performed ($S \approx 20, N \approx 30$) leads, according to Eq. (6), to ΔN_j of order 3 before the jump and 2 after the jump, when $\Phi = \Phi_j^s$.

This means that in the higher mentioned mechanism, the director configuration in the layer is strongly distorted and metastable not only before but also after the pitch jump.

DISCUSSION

The fit of the theoretical curves $\Phi(T)$ with respect to the parameter S for temperatures preceding the pitch jump to the measured values of this quantity (Fig. 5) demonstrates a reasonable agreement between theory and experiment for $S=25$ during both cooling and heating of the specimen^[8]. If this value of S is used to calculate the hysteresis with Eq. (7) for example for $N=30$, then we obtain $\Delta N_h=2.4$. An estimate of the same quantity from the measured values of the temperature of the pitch jumps and the temperature changes of the equilibrium pitch in a bulk cholesteric (i.e. the 18 μm sample), see Fig. 3, gives us a much smaller value, equal to only 0.3.

If one draws a straight line parallel to the Y axis near $2d/p(T)\approx 6$ in Fig. 3 it becomes clear that the continuum theory predicts in addition to a stable configuration, several metastable configurations with different values for N .

If such metastable configurations were present in the layer for a sufficiently long time, the experimental spectra would consist of more than two spectra with different values of N . Moreover, structural changes with N changing by more than 1 should be observed. However the experimental measurements do not give a clear indication for this.

Thus, it must be concluded that in the specimen studied, the restructuring of the director configuration does not occur by means of continuous changes in the orientation of the director at the surface. This means that some kind of fluctuations of the director or instabilities of the director configuration prevent Φ from reaching the value Φ_c . It should be expected that these factors influence the appearance of hysteresis in the pitch jump especially if several metastable configurations of the director with $|\Phi_o| < |\Phi_c|$ exist in the layer.

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