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Relationship between the pitch and elastic constants of the transient planar state in cholesteric liquid crystals

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We investigate experimentally the relationship between the pitch and the elastic constants of a transient planar state in cholesteric liquid crystals. The pitch of the transient planar state is measured by the optical reflection technique. The twist and bend elastic constants of cholesteric materials are determined from the measured values of the threshold voltages for the transitions between a focal-conic state and a homeotropic state. By comparing these values, we experimentally confirm the relationship for the pitch of a transient planar state; $P_{\text{transient}} = (K_{33}/K_{22})P_0$, which has been suggested theoretically.

1. Introduction

The cholesteric phase of liquid crystals differs from the nematic phase in that the director rotates in space about a helical axis. In general, the cholesteric phase has two stable states between parallel glass plates: a planar (P) and a focal conic (FC) state. In the equilibrium P state, the helical axes of the cholesteric material are normal to the plates and the directors are in the planes parallel to the substrates. This periodic helical structure of the P state shows Bragg reflection of light in the spectral range $n_o P_0 \leq \lambda \leq n_e P_0$, where P_0 is the intrinsic pitch of the P state and n_o and n_e are the ordinary and extraordinary refractive indices of the liquid crystal, respectively. The FC state also exhibits a twisted structure, although the helical axes are not normal to the plates but bend between the two substrates or are even parallel to the plates and not aligned with each other. The cholesteric system in the FC texture appears milky due to the scattering of light by many defects.

Under the influence of an external electric or magnetic field, the P state of a cholesteric liquid crystal undergoes a transition to a homeotropic (H) state, which is the field-induced nematic state, through an FC state. This transition was first described by de Gennes [1]. As shown in figure 1(a), the helical axis of the P state rotates to be parallel to the plates and the system proceeds to an FC state, as the electric field is increased. At a still higher field, all the helical structures are unwound and the system undergoes transition to an H state. Now, upon decreasing the electric field

gradually, the liquid crystal material makes a transition to the FC state. The transmittance for each director configuration as a function of the applied field is shown in figure 1(b). As one can see in the figure, the relation of transmittance to the external field is different when the field is increasing from when it is decreasing. This hysteretic behaviour of cholesteric liquid crystals has been observed by many researchers [2–4] and efforts have been made to describe this phenomenon [5, 6]. Van Sprang and van de Venne obtained an expression for the threshold field for each transition by considering the surface interaction terms [7]. According to their

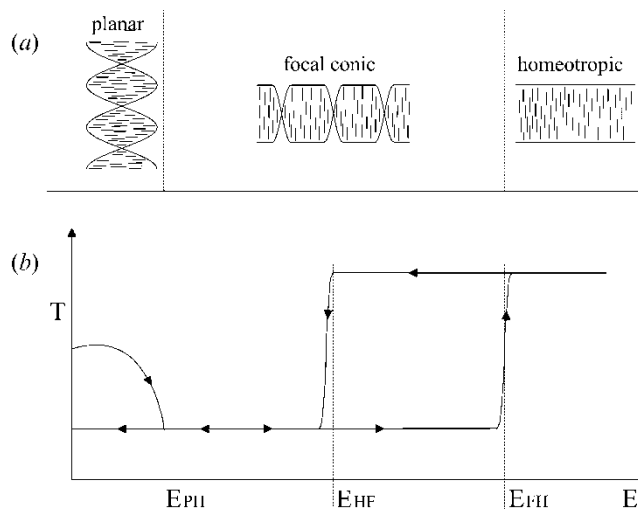


Figure 1. Change of (a) the director configuration and (b) the transmittance as a function of external electric field in cholesteric liquid crystals.

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results, the transition fields are expressed as

$$E_{PF} = 2\sqrt{2} \left(\frac{F_{SF} - F_{SP}}{d\epsilon_0\Delta\epsilon} \right)^{\frac{1}{2}} \quad (1)$$

$$E_{FH} = 2\sqrt{2} \left(\frac{\pi^2 K_{22}}{P_0^2 \epsilon_0 \Delta\epsilon} + \frac{F_{SH} - F_{SF}}{d\epsilon_0\Delta\epsilon} \right)^{\frac{1}{2}} \quad (2)$$

$$E_{HF} = \left[\frac{\pi^2 4K_{22}^2 - K_{33}^2 (P_0^2/d^2)}{K_{33}\epsilon_0\Delta\epsilon} + 4 \frac{F_{SH}}{d\epsilon_0\Delta\epsilon} \right]^{\frac{1}{2}} \quad (3)$$

where E_{PF} , E_{FH} , and E_{HF} are the threshold fields for the P to FC, FC to H and H to FC transitions, respectively. K_{22} is the twist elastic constant and K_{33} is the bend elastic constant of the cholesteric material. ϵ_0 is the permittivity of free space and $\Delta\epsilon$ is the dielectric anisotropy. F_{SP} , F_{SF} , and F_{SH} are the surface free energy densities of the P, FC, and H states, respectively.

Now if the electric or magnetic field is removed suddenly, the H state undergoes a transition to the P state instead of the FC state. There have been many reports of another metastable state observed during this H to P transition in cholesteric liquid crystals [2, 8–11]. This state, called a transient planar (TP) state, has a longer pitch than the equilibrium planar pitch P_0 and comes about in the early stage of the H to P transition. Yang and Lu solved the Frank–Oseen equation approximately and derived equations for the elastic free energy of a cholesteric system during the H to TP transition as a function of the polar and azimuthal angles of the director [11]. Their theoretical model predicted that the TP state exists with an estimated pitch of approximately $(K_{33}/K_{22})P_0$ assuming a one-dimensional, conical relaxation of directors. A more recent theoretical study reports that any value of pitch

greater than $0.5(K_{33}/K_{22})P_0$ is allowed in the initial stage of the H to TP transition [12]. However, the rate of increase of the polar angle for the director in time has its maximum value when the pitch is $(K_{33}/K_{22})P_0$. Therefore, although initially there exist pitches of various values in the system, only the pitch of value $(K_{33}/K_{22})P_0$ becomes dominant as the transition proceeds. As a result, the TP state with this value of pitch exhibits the strongest reflectance, and thereby one measures the value of $P_{\text{transient}}$ to be $(K_{33}/K_{22})P_0$. However, to the best of our knowledge, this relationship between $P_{\text{transient}}$ and the two elastic constants of cholesteric liquid crystals is only a theoretical result, and has not previously been verified experimentally.

In this study, we attempt to verify this relationship experimentally for the TP state of a cholesteric system. The equilibrium pitches P_0 and $P_{\text{transient}}$ are measured by the optical reflection technique and the elastic constants are obtained from the measured values of the threshold fields for samples of different thicknesses.

2. Experimental procedures

As shown in figure 2, our experimental system consists of a light source block, an optical components block and an electronic components block. The light source block includes a high-power Xe lamp and a monochromator. Monochromatic radiation was incident on liquid crystal cells and the transmitted and reflected radiation from the cells was measured by a photomultiplier tube (Hamamatsu R-405). The electronic components block includes a digital oscilloscope (Hewlett Packard 54501A), a function generator (Tektronix FG5010) and a voltage amplifier (Krohn-Hite 7500).

The liquid crystals used were a mixture of Merck ZLI

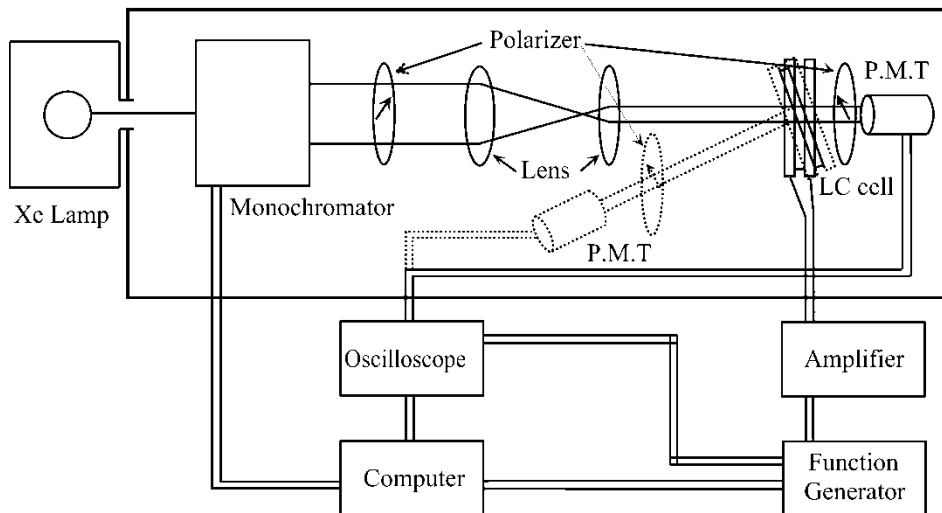


Figure 2. Experimental arrangement.

2293 nematic liquid crystal and ZLI 811 cholesteric dopants in the weight ratio 2.2:1. The intrinsic pitch P_0 of this mixture was measured to be (300 ± 10) nm using the Grandjean–Cano method. The cells used in this study employed ITO-coated soda lime glass, and the surfaces were treated with a rubbed planar SE 5291 polyimide (Nissan Chemical Ind., Ltd). The thicknesses of cells measured interferometrically were 4.3, 5.2, 6.0, 6.8, 7.5, 8.1 and 9.0 μm .

Set-ups for measuring the pitch for the TP state and the twist and bend elastic constants, K_{22} and K_{33} , were basically the same except for the location of the photomultiplier tube. For the elastic constants the transmittance of cholesteric liquid crystals was measured with the LC cells located at right angles to the incident monochromatic light; for the pitch, the reflectance was measured with two crossed polarizers in order to prevent the light reflected at the glass from entering the photomultiplier tube. The voltage applied to the sample was a 1 kHz square wave, whose amplitude could be varied between 0 and 200 V_{p-p} with a step of 0.5 V. The threshold fields, E_{FH} and E_{HF} , were determined from the transmittance vs. voltage curves. Measurement of the output transmittance was made 30 s after the voltage application to ensure the system was in an equilibrium state.

3. Results and discussion

3.1. Measurement of the pitch of a TP state

Figure 3 shows the measured reflection spectrum for the planar textured sample. The interference fringes in the reflection spectrum indicate that the cholesteric material is in a nearly perfect planar state and a monodomain is formed in the cell. The strong reflection

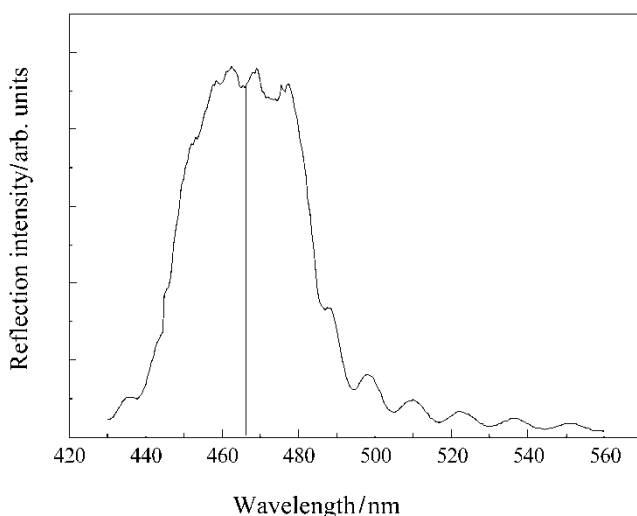


Figure 3. Measured reflection spectrum for the planar state. The centre of the reflection peak is 466 nm.

region ranges from wavelength 440 to 490 nm with the peak centre at about 466 nm, and from the relationship between the centre wavelength of the reflection peak ($\lambda_{C,P}$) and the pitch of the equilibrium P state, $\lambda_{C,P} = \bar{n}P_0$, the average refractive index \bar{n} is calculated to be 1.55.

Shown in figure 4 is a three-dimensional graph of reflection intensity vs. wavelength and time constructed from the measured reflectance for light of a given wavelength as a function of time during the transition from the H to P state. The incident angle of monochromatic light on the LC cell was 3.0° . The wavelength was scanned from 430 to 640 nm in intervals of 0.5 nm. Time was also scanned to 16 ms in intervals of 0.04 ms. As can be seen in figure 4, the reflection spectra at the early stages of the H to P transition consist of interference fringes. The envelope of interference fringes, which we call a broad reflection band, corresponds to the reflection spectrum of the TP state. This band with its centre at 592 nm at 0.6 ms in the figure lasts for a few ms before the system evolves to an equilibrium P state. The band centred at about 510 nm at $t = 3.0$ ms represents the planar reflection and it slowly evolves with time to the wavelength of the equilibrium P state, 466 nm. The interference fringes observed in the reflection spectra suggest that a Fabry–Perot interferometric structure is formed in the cell during the transition [13]. Under planar boundary conditions, the transition from the H to TP state begins from the surfaces and grows into the bulk [14]. Thus, during the process of transition, a spatially cholesteric–nematic–cholesteric structure will be formed in the cell. Because the cholesteric structure reflects light of the same twist sense of the circularly polarized light as the helix, the cholesteric phase near the two surfaces acts to give reflecting mirrors and the nematic phase in the bulk region acts as a cavity resonator. This Fabry–Perot interferometric structure formed in the cell is believed to result in interference fringes in the measured reflection spectra.

The time dependence of the centre of the reflection band (λ_c) and the amplitude of the reflection peak are shown in figure 5 for the first few ms of the H to P transition. λ_c starts at about 520 nm at $t = 0$ the instant the applied voltage is removed, and evolves towards longer wavelengths. However, the reflection amplitude in this time region is very small, meaning that the helical structure is barely formed in the cell. As the system proceeds to the TP state, the reflection amplitude increases and λ_c reaches its maximum value of 592 nm at $t = 0.6$ ms, which is the reflection wavelength $\lambda_{C,TP}$ for the TP state. Then λ_c slowly decreases with the reflection amplitude increasing, as the cholesteric material relaxes into the planar texture.

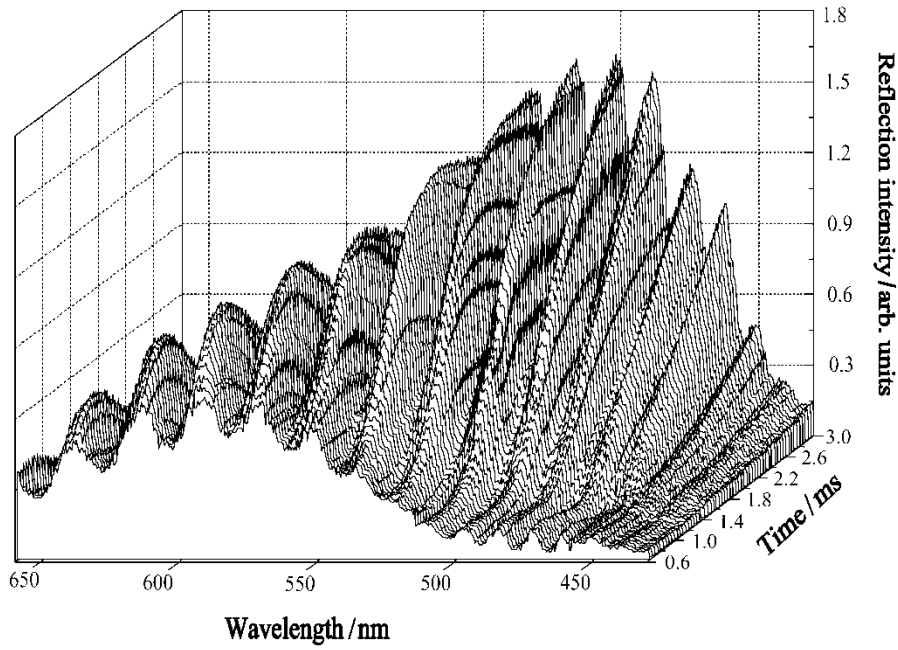


Figure 4. Reflection intensity vs. wavelength and time during the H to P transition.

The common relaxation time for the equilibrium P state is reported to be about 200 ms [15]. As one can see in figure 5, the reflection intensity of the TP state is much smaller than that of the equilibrium P state. This may suggest that the TP state is not propagated throughout the bulk region of the sample, i.e. a perfect helical structure is not formed in all regions of the cell by the time 0.6 ms.

Now the ratio of the pitches, $P_{\text{transient}}/P_{\text{planar}}$, can be obtained from the relationship, $\lambda_{C,P} = \bar{n}P_0$ and $\lambda_{C,TP} = \bar{n}P_{\text{transient}}$, with the measured values

$\lambda_{C,P} = 466 \text{ nm}$ and $\lambda_{C,TP} = 592 \text{ nm}$ as:

$$\frac{P_{\text{transient}}}{P_0} = \frac{\bar{n}P_{\text{transient}}}{\bar{n}P_0} = \frac{\lambda_{C,TP}}{\lambda_{C,P}} = 1.27. \quad (4)$$

3.2. Measurement of the twist and bend elastic constants

Equations (1)–(3) can be rewritten in terms of the transition voltage V and the cell thickness d as

$$V_{\text{PF}}^2 = \frac{8(F_{\text{SF}} - F_{\text{SP}})d}{\epsilon_0 \Delta \epsilon} \quad (5)$$

$$V_{\text{FH}}^2 = \frac{8\pi^2 K_{22}}{P_0^2 \epsilon_0 \Delta \epsilon} d^2 + \frac{8(F_{\text{SH}} - F_{\text{SF}})d}{\epsilon_0 \Delta \epsilon} \quad (6)$$

$$V_{\text{HF}}^2 = \frac{\pi^2 4K_{22}^2 - K_{33}^2 (P_0^2/d^2)}{K_{33} \epsilon_0 \Delta \epsilon} d^2 + \frac{4F_{\text{SH}}d}{\epsilon_0 \Delta \epsilon} \quad (7)$$

where V_{PF} , V_{FH} and V_{HF} are the transition voltages for the P to FC, FC to H, and H to FC transitions, respectively. The dielectric anisotropy, $\Delta \epsilon$, is obtained by measuring the capacitance of the sample. A capacitor of known capacitance was connected in series with the sample, and the voltage drop across it measured as a function of input voltage. Measured values of ϵ_{\parallel} and ϵ_{\perp} were 9.7 and 4.7, respectively, and thus $\Delta \epsilon$ was +5.0.

The transition voltages V_{FH} and V_{HF} are measured for the cells of various thicknesses as shown in figure 6. From equation (6), the dependence of V_{FH}^2 on the cell

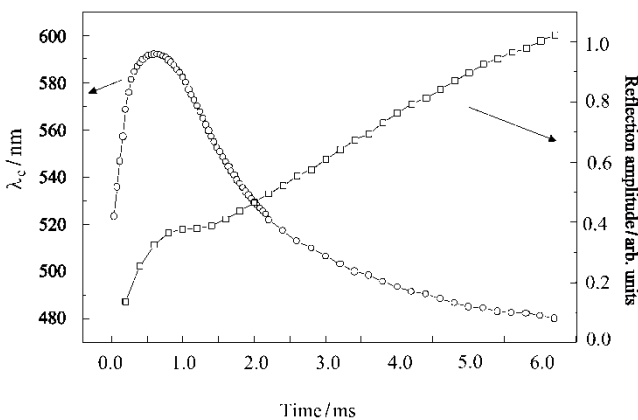


Figure 5. Time dependence of the centre λ_c and the amplitude of the reflection band during the H to P transition.

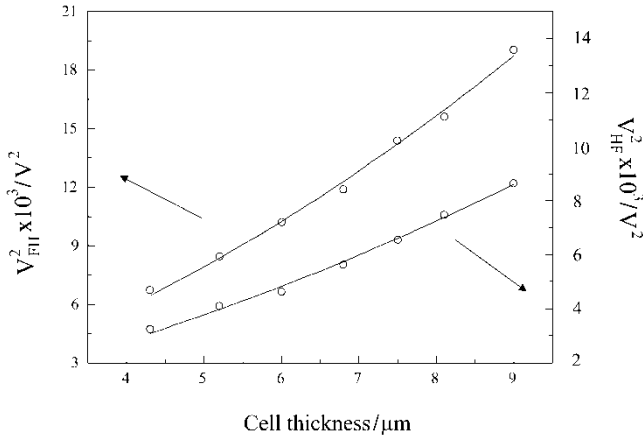


Figure 6. Transition voltages squared vs. cell thickness for the FC to H and the H to FC transitions. Circles show the measured values, solid lines represent the fitted curves.

thickness d can be expressed as:

$$V_{FH}^2 = \alpha d^2 + \beta d. \quad (8)$$

The fitted curve for V_{FH}^2 as a quadratic function of d in figure 6 gives $\alpha = 1.24 \times 10^{14} \text{V}^2 \text{m}^{-2}$, yielding $K_{22} = 6.24 \times 10^{-12} \text{N}$ for our sample. Equation (7) for V_{HF}^2 can also be written as a quadratic function of d :

$$V_{HF}^2 = \frac{\pi^2}{P_0^2} \frac{4K_{22}^2}{K_{33}\epsilon_0\Delta\epsilon} d^2 + \frac{4F_{SH}}{\epsilon_0\Delta\epsilon} d - \frac{\pi^2 K_{33}}{\epsilon_0\Delta\epsilon} \quad (9)$$

$$= \alpha' d^2 + \beta' d + \text{const.}$$

From the fitted result, α' is $4.82 \times 10^{13} \text{V}^2 \text{m}^{-2}$ and K_{33} is calculated to be $7.94 \times 10^{-12} \text{N}$. Therefore, the ratio between K_{33} and K_{22} is given by

$$\frac{K_{33}}{K_{22}} = 1.27. \quad (10)$$

Equations (4) and (10) confirm the previously obtained theoretical relationship [11, 12];

$$P_{\text{transient}} = (K_{33}/K_{22})P_0. \quad (11)$$

4. Conclusions

In this study, we experimentally investigated the relationship between the pitch of a TP state and the

elastic constants in cholesteric liquid crystals. The equilibrium planar pitch was measured to be $P_0 = (300 \pm 10) \text{nm}$ using the Grandjean–Cano method; the TP pitch, $P_{\text{transient}}$, was found to be 381 nm from the measured reflection spectrum during the H to TP transition for our cholesteric system, and these values yield the ratio $P_{\text{transient}}/P_0 = 1.27$. The twist and bend elastic constants were determined from the threshold voltages of the FC to H and H to FC transitions for cells of different thickness. For our cholesteric material K_{22} was $6.24 \times 10^{-12} \text{N}$ and K_{33} $7.94 \times 10^{-12} \text{N}$, yielding the ratio $K_{33}/K_{22} = 1.27$, which is the same value as the ratio between the transient planar pitch and the unstrained intrinsic pitch. This experimental result confirms that the pitch of a TP state of a cholesteric liquid crystal system is determined by the two elastic constants, and the equilibrium planar pitch of the material as $P_{\text{transient}} = (K_{33}/K_{22})P_0$, which agrees well with the suggestions of previous theoretical studies.

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