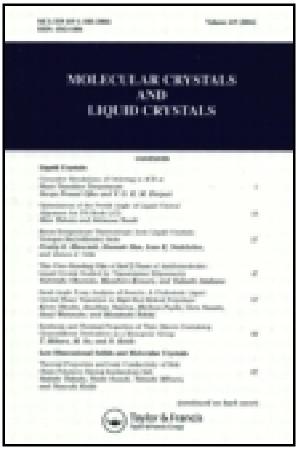

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Cholesteric Liquid Crystal Transmission Profile Asymmetry

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Cholesteric Liquid Crystal Transmission Profile Asymmetry

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The selective reflection profile of a cholesteric liquid crystal is asymmetric when (a) for light incident parallel to the cholesteric helix axis, the incident polarization vector, *E*, is at an angle other than 45° to the molecular director at the first surface or (b) for light incident oblique to the cholesteric helix axis, the incident polarization is S- or P-polarized. We confirm these effects using cyclic polysiloxanes. Further, we show that the use of a new form of cholesteric liquid crystal called flakes allows the first experimental separation of the asymmetry due to the surface and that due to the bulk.

Keywords: Cholesteric liquid crystal; CLC flakes; polysiloxanes

Cholesteric liquid crystals (CLC's) are characterized by a structure in which the average direction of molecules in a given plane, indicated by a unit vector called the director, **n**, rotates about an axis. The continuously rotating director traces out a helix. The length through which the director makes a complete 360° cycle is called the pitch, *P*, of the cholesteric. A schematic diagram of the rotation of the molecular director is shown in Figure 1. When the orientation of the helix axis is normal to the boundary surfaces, the texture produced is called Grandjean or planar [1]. This structure is uniaxial [2] where the optic axis is the helix axis. The molecules in a given plane are approximately aligned in the same direction similar to a

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[*] Corresponding author.

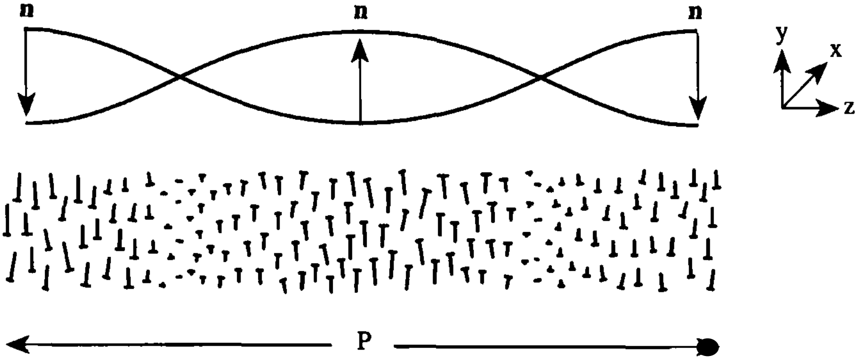

FIGURE 1 Schematic model of CLC structure.

nematic liquid crystal so the layers are sometimes referred to as quasinematic [3].

The periodicity of the Grandjean texture is responsible for the Bragg-like selective wavelength and polarization reflection unique to CLC's. Incident light that is superimposable on the helix, that is, of the proper wavelength and of the proper circular polarization, will be reflected [4]. The center of the reflection band, λ_o, is given by the De Vries equation:

$$\lambda_o = \bar{n}_n P \qquad (1)$$

where $\bar{n}_n$ = the average refractive index of the quasinematic layers. The width of the reflected wavelength band, for visible-reflecting CLC's (λ_o ranging from 350 to 800 nm) can typically range from 10 to 100 nm [5].

When the incident light is either circularly polarized such that it is superimposable on the CLC helix or randomly polarized, the reflectance peak or the transmittance valley is symmetrical about λ_o. This has been predicted [6] and demonstrated [7] in reflection and transmission.

In experiments where the incident light is *linearly* polarized [8–11], reflection and transmission scans of Grandjean-aligned CLC's show asymmetry around λ_o. In other words, one side of the reflectance peak has a higher value than the other, or conversely, one side of the transmittance valley has a lower value than the other.

A number of studies have been done to elucidate the source(s) of the asymmetry:

For linearly polarized light incident parallel to the CLC helix axis, Tur [12] has calculated that a large difference between the external medium index and the average index of the cholesteric itself affects the symmetry of the selective reflection profile within the selective band. He attributes the effect to interference between selective reflection from the cholesteric planes and dielectric reflection from the interface of the cholesteric with the external medium. He considers two variables: (i) whether $\bar{n}_n$ is larger or smaller than the refractive index, n_{med}, of the bounding medium (air or glass support, for example) and (ii) whether the extraordinary refractive index of a given layer, $n_{e,n}$, is larger or smaller than the ordinary refractive index of the layer, $n_{o,n}$. For the case, for example, of $\bar{n}_n > n_{\mathrm{med}}$ and $n_{e,n} > n_{o,n}$, the transmission profile is lower at wavelengths longer than λ_o when the incident polarization vector, **E**, is perpendicular to the director, **n**, at the first surface. For the same conditions, the transmission profile is lower at wavelengths shorter than λ_o when **E** is parallel to **n** at the first surface. The greater the index difference, the more pronounced is the effect.

For light incident at an angle to the CLC helix axis, the shapes of the reflection/transmission profiles were predicted in a number of studies using various approximation methods to solve Maxwell's equations for oblique propagation of light in the cholesteric medium.

One of the earliest approximate solutions to the Maxwell wave equation at oblique incidence for CLC's was developed in 1969 by Taupin [13] using a method of truncated infinite matrices. Taupin's results showed that there should be multiple orders of selective reflection bands at oblique incidence and that each order would have three distinct zones: a short-wavelength zone that reflects only when P-polarized light is incident, a long-wavelength zone that reflects only when S-polarized light is incident, and a middle zone reflecting no matter what the incident polarization state is. Taupin called this middle part of each triplet a "total reflection region".

Later, Teitler and Henvis [14] used a 4 × 4 matrix formulation of the electromagnetic wave equations applied to a model of finite discrete layers of homogeneous anisotropic media. Berreman and Scheffer [15, 16] extended this theory to the continuously varying anisotropy of the cholesteric structure and wrote the characteristic matrix for one thin layer as a series expansion in powers of thickness that was assumed small enough so that higher powers could be ignored. This approach led to predictions about the shape, symmetry, polarization-dependence, and multiple orders of the selective reflection profile that were consistent with Taupin's predictions. The 4 × 4 matrix method improved on all the other solution techniques by predicting not only the position of the reflection band and multiple orders and the linear polarization response, but also the shape of the selective reflection profiles. The middle zone of each order reflects both S- and P-polarized light. S-polarized incident light is strongly reflected at the long-wavelength edge of the selective band, and P-polarized light is strongly reflected at the short-wavelength edge of the selective band. The 4 × 4 approach indicates that this behavior manifests itself as a marked asymmetry in the selective reflection profiles for S- *versus* P-polarized obliquely incident light.

The S – P effect has been demonstrated using low-molecular-weight CLC's (LMCLC's) by Berreman and Scheffer [15] and more recently by Takezoe *et al.* [10]. To our knowledge, the asymmetry due to the relative orientations of **E** and **n** at the first surface has never been experimentally demonstrated. For any given sample, both causes of asymmetry occur simultaneously and have never been experimentally isolated. In this paper, we confirm the two types of asymmetry behaviors for higher-weight oligomeric CLC's. Using a new form of CLC called flakes, we will show how the two sources of asymmetry can be examined separately.

SAMPLE PREPARATION

The CLC used in this study is a cyclic polysiloxane [17, 18] we will refer to as CLC670. Samples to be tested were prepared as continuous films and as discrete flakes.

Continuous films were prepared by single-side release layer from a sandwich cell in the following way: Glass disk substrates made of a conveniently available float glass ($n_{D,20} = 1.533$, 1-in. diameter, 1 mm thick) were cleaned. One substrate was placed on a spin-coater [19] and flooded by syringe with a 0.5%-by-weight solution of polyvinyl alcohol (PVA, 87 – 89% hydrolyzed) [20]. The substrate was spun at 2000 rpm for 60 sec. The application of PVA solution was repeated once more, and the substrate was left to dry for 24 h in a Class 1000 cleanroom.

The coated substrate was buffed using a device developed in-house at the Laboratory for Laser Energetics (LLE) [21]. It consisted of a spinning (2000 rpm) cylinder, 25 cm long and 10 cm in diameter and covered with a polyaramide fiber sheet [22]. The substrate was held to the platform by vacuum and then passed beneath the rotating cylinder. The height of the roller was adjusted until passing the substrate under the stationary roller just caused light behind the roller to be extinguished. The direction of buffing was defined to be the direction of the roller motion (same as the movement of the platform for the first pass). The substrate was passed forward and backward four times under the spinning roller.

The CLC670 was mixed with glass-fiber spacers [23] until removal of an amount comparable to what would be used in the cell showed about 20 spacers under a microscope. The spacers ranged from 20 to 100 μm in length and were 13 ± 0.3 μm in diameter. The CLC670 was degassed for 24 h under vacuum [24] (approximately < 1 Torr) at 130°C to remove any trapped air or solvent remaining from the manufacturing process. The bottle with CLC670 and substrate with PVA coating were then heated on a hotplate to 130°C. The CLC was dabbed onto the warmed substrate using a microspatula. A second clean substrate, without PVA coating and unwarmed, was then placed onto the CLC. The whole cell remained at the elevated temperature while the top substrate was gently manually sheared until the CLC thickness had reached that of the fiber spacers. The temperature of the hotplate was then slowly ramped down (1°C/h) to 45°C, 2°C below the glass transition temperature, T_g, of CLC670 and remained at that temperature for 24 h. This annealing process allowed the CLC molecules to align into the Grandjean texture and eliminated any flow lines and disclinations caused by the manipulation of the cell. Finally, the temperature was ramped down (1°C/h) to room temperature.

The cell was submerged in water. After a week, the PVA dissolved and the previously coated substrate slid off the CLC surface. There was no obvious swelling, but samples mounted for optical testing were equilibrated in air for at least 24 h to allow evaporation to occur before testing began. Since the PVA had been buffed before the cell was made, it acted as an alignment layer as well as the release layer.

Discrete flakes were prepared by a procedure referred to as "knife-coating" followed by thermal fracturing. A 100 mm-diam. 0.5 mm-thick silicon wafer [25] was heated to 130°C on a hotplate. CLC670 was melted onto the substrate surface. A microscope slide, held on edge to act as the "knife," was used to smear the polysiloxane across the surface. This knife-coating process caused the CLC to be aligned in the Grandjean texture as observed by reflective color. The substrate was then quickly removed from the hotplate, allowing the CLC to be quenched to room temperature, "freezing in" the selective reflection structure.

The "open-faced" film was placed in a Petri dish and liquid nitrogen poured over it. The CLC fractured and lifted slightly and temporarily off the silicon surface. Methanol was used to wash the fractured CLC off the substrate. These new fractured CLC polymer pieces are called "flakes."

To separate flakes by size, a stack of sieves [26] was used. They were stainless steel screens of mesh sizes 80, 170, 325 and 635. Flakes in methanol slurry were dripped into the sieve stack, using methanol to wash the flakes through. The flakes were separated into five groups: > 180 μm, 90 – 180 μm, 45 – 90 μm, 20 – 45 μm and < 20 μm.

To examine the optical properties of CLC flakes, they were prepared as samples in a process we refer to as "water bead orientation." One mL of deionized water was dropped by graduated pipet onto each substrate (the same float glass described earlier) to cover it to the edge. Flakes in methanol slurry were dripped onto the water bead by medicine dropper. The flakes oriented themselves with their largest surface parallel to the meniscus. When the surface was visually covered with flakes, the underlying fluid was removed by medicine dropper, and the sample was allowed to dry. At regular intervals, the sample was weighed until three identical readings were obtained, indicating that the samples had completely dried (about 48 h total). For this study, CLC670 flake results from only the 45 to 90 μm size-group are reported. However, all flake size-groups were tested and gave similar results regarding asymmetry effects. These results as well as additional optical and physical characteristics of flakes are reported elsewhere [27].

REFRACTIVE INDEX MEASUREMENTS

The ordinary and extraordinary cholesteric refractive indices of CLC670, $n_{o,ch}$, and $n_{e,ch}$, were measured on an Abbe refractometer [28] at four wavelengths – 546.1, 589.6, 632.8 and 1047 nm, all at $T = 20°\text{C}$ – and are listed in Table I. These values were used to calculate the ordinary and extraordinary refractive indices, $n_{o,n}$ and $n_{e,n}$ of the quasinematic layers by the method outlined in the Appendix. Over the spectral range to be examined (400–900 nm), the $\bar{n}_n$ of CLC670 is ~ 1.6.

OBTAINING TRANSMISSION PROFILES

To examine the asymmetry of the selective transmission profile, CLC film and flake samples were scanned in the Perkin-Elmer Lambda-9 spectrophotometer using the setup of Figure 2. Linearly polarized incident light was

TABLE I Refractive index dispersion at $T = 20°\text{C}$ for CLC670

$\lambda\,(nm)$	$n_{e,ch} = n_{o,n}$	$n_{o,ch} = \bar{n}_n$	$n_{e,n}$	$\bar{n}_{ch}$
546.1	1.5315	1.6212	1.7062	1.5919
589.6	1.5277	1.6151	1.6980	1.5865
632.8	1.5250	1.6106	1.6919	1.5826
1047.0	1.5143	1.5921	1.6663	1.5666

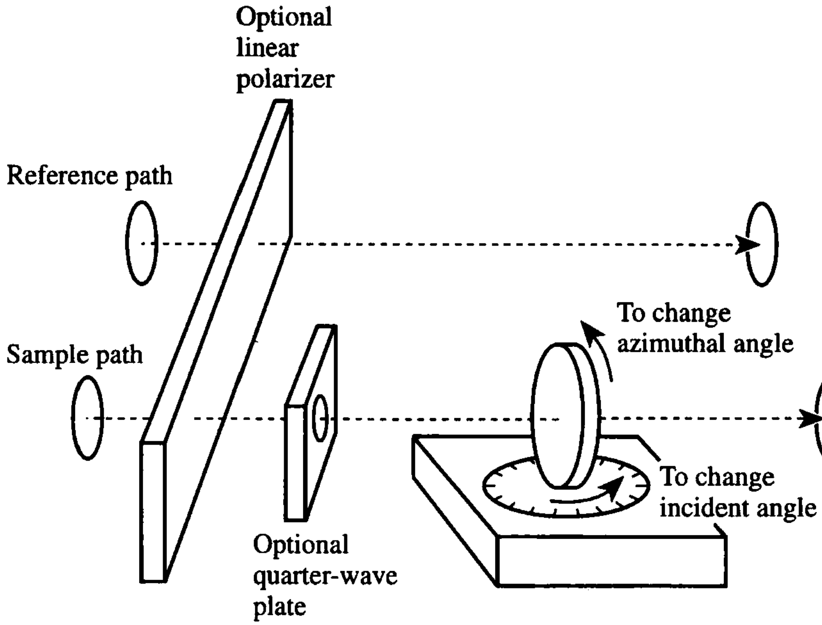

FIGURE 2 Sample and optical arrangement for Lambda-9 spectrophotometer transmission scans.

produced using HN32 Polaroid film, positioned just before the sample, placed with the transmission axis either horizontal or vertical with respect to the spectrophotometer sample base plate to produce S- and P-polarized light respectively when the sample was rotated to change incident angle.

EXPERIMENTAL RESULTS AND DISCUSSION

(a) Normal Incidence and Unfractured CLC Film

For incident light linearly polarized either parallel or perpendicularly to the director at the first surface with an index mismatch, the selective band is expected to show the asymmetry predicted by Tur for LMCLC's [12]. Our study confirms, for the first time, the same behavior for higher-weight oligomeric CLC films. Figure 3 shows transmission scans of a PVA-released CLC670 cholesteric film for three azimuthal positions of the buff direction – 0° or parallel, 45° and 90° or perpendicular – with respect to the incident polarization vector, **E**. Because it is released on one side, there is an index mismatch to the surrounding air of ~0.6. In Figure 3(a), the incident **E** is

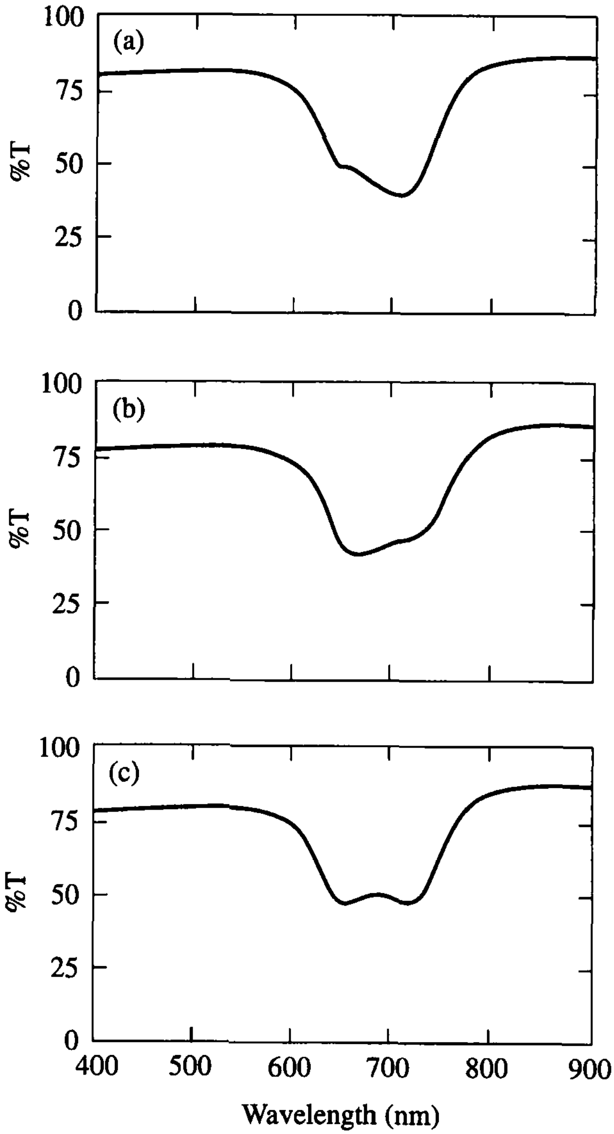

FIGURE 3 Asymmetry of selective transmittance profile for CLC670 film. Linearly polarized light at normal incidence to the air interface of this CLC film shows asymmetry when **E** is (a) perpendicular or (b) parallel to **n** at the first surface but no asymmetry when **E** is (c) at 45° to **n** at the first surface. In each case, the buff direction is the same as the direction of **n**.

perpendicular to the buffing direction. The minimum transmission/maximum reflection is at the long-wavelength edge of the selective band, consistent with Tur's calculation of **E** perpendicular to **n**. Similarly, in

Figure 3(b), in which **E** is parallel to the buff, the transmission profile is consistent with Tur's calculation of **E** parallel to **n**. We can conclude, then, that the CLC polysiloxane aligns parallel to the buff direction of a PVA alignment layer. Figure 3(c) confirms this, exhibiting no asymmetry when **E** is at 45° to the buff direction and therefore the molecular director at the first surface.

This is the first time, to our knowledge, that CLC polysiloxanes have been experimentally shown to have alignment parallel to the buff direction. This behavior cannot be assumed. Seo *et al.* [29] observed nematic liquid crystals aligned perpendicular to the direction of buffing. Ishihara *et al.* [30] found that nematic liquid crystal molecules may align parallel, perpendicular, or even at 45° to the buffing direction depending on the choice of alignment layer polymer and attributed the behavior to the specific chemical interaction of the liquid crystals and the polymer chains within the alignment layer. Since nematic liquid crystals do not show the twisting behavior of cholesterics, it is possible to infer surface orientation of nematic molecules from optical tests that probe the whole sample [31, 32]. These tests generally evaluate the intensity of transmitted light as a function of incident angle and azimuthal angle relative to incident polarization but not as a function of wavelength. Our results, for the first time, use the polarization *wavelength* asymmetry behavior of the CLC film to determine the nature of molecular orientation of the first surface encountered by incident light. By optical tests on the bulk sample, we are able to draw conclusions about the molecular dipole orientation of the first layer.

As Tur [12] predicted for LMCLC's, asymmetry in the transmission/reflection profile of CLC's results when there is (i) an index mismatch (ii) at the first surface which (iii) linearly polarized light encounters (iv) with an azimuthal angle other than 45° between the polarization vector and the molecular director. Freestanding and released-side cholesteric polysiloxanes meet these four requirements and therefore show the same asymmetries as LMCLC's.

(b) Normal Incidence and CLC Flakes

Samples of CLC670 flakes from the "45 to 90 μm" sieve group, oriented by the water bead method, were used to examine the polarization selectivity of CLC flakes at normal incidence.

As described earlier, asymmetry in the selective profile occurs for non-index-matched samples when the incident **E** is either parallel or perpendicular to the molecular director at the first surface. For CLC flake samples, the director at the first surface has been randomized by the water bead preparation technique. We would expect, therefore, that no asymmetry of the selective profile should be seen for CLC flake samples at any azimuthal angle for normal incidence of linearly polarized light, regardless of the index-matching of the sample. Two transmittance profiles were obtained using horizontal linearly polarized light at two azimuthal angles 45° apart (shown in Fig. 4). Since there is no predefined preferential direction, the first angle is arbitrary. Should any asymmetry exist, it should be exposed at least at one of the two azimuthal angles. However, no asymmetry of the selective profile is evident, as expected, confirming *experimentally for the first time* that one of the necessary conditions for such asymmetry is the non-randomness of the molecular director.

(c) Oblique Incidence and Unfractured CLC Film

Because the unfractured CLC670 film exhibits asymmetry due to the molecular director orientation at its surface, we first oriented the film to give a symmetric profile at normal incidence; *i.e.*, the incident **E** was at 45°

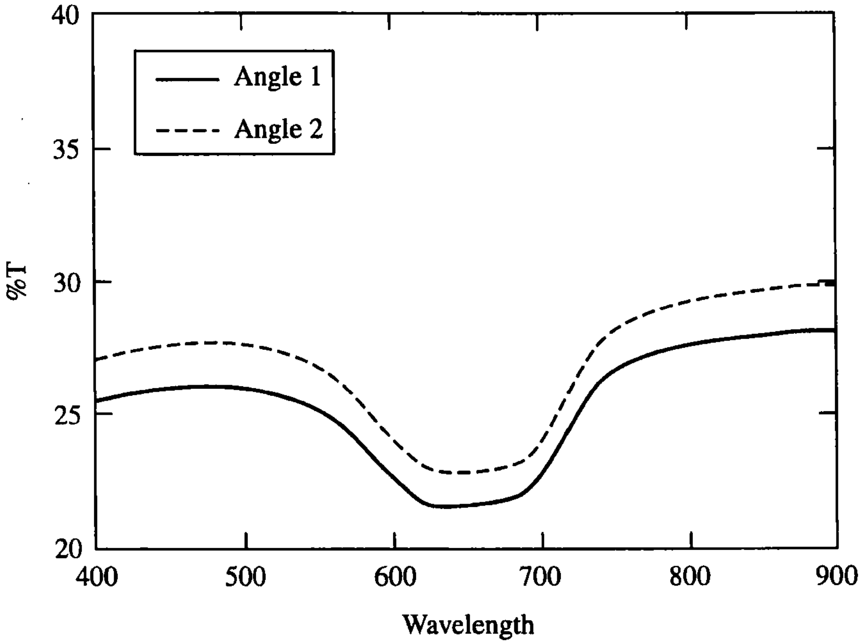

FIGURE 4 CLC flakes at two azimuthal angles. CLC670 flakes of size 45 to 90 μm are prepared by water bead orientation and scanned in linearly polarized normally incident light. For two azimuthal angles at 45° to each other, there is no evidence of the asymmetry associated with surface molecular director orientation.

azimuthally to the buff direction (which is also the direction of **n**). Then, the CLC670 film was rotated so that the beam was incident at 30° from the surface normal. Figure 5(a) compares the transmission profiles of a CLC670 film in incident S- and P-polarization. In P-polarized light, there is greater reflectance (and, implicitly, lower transmittance) at the shorter-wavelength edge of the selective band. Conversely, in S-polarized light, there is greater reflectance (lower transmittance) at the longer-wavelength edge of the band. The film shows the same asymmetries in S- and P-polarized light as found by Taupin, Berreman and Scheffer, and Takezoe *et al.*, as previously discussed.

(d) Oblique Incidence and CLC Flakes

Since the CLC flake sample showed no asymmetry at normal incidence, no special positioning of the sample was necessary before oblique incidence was

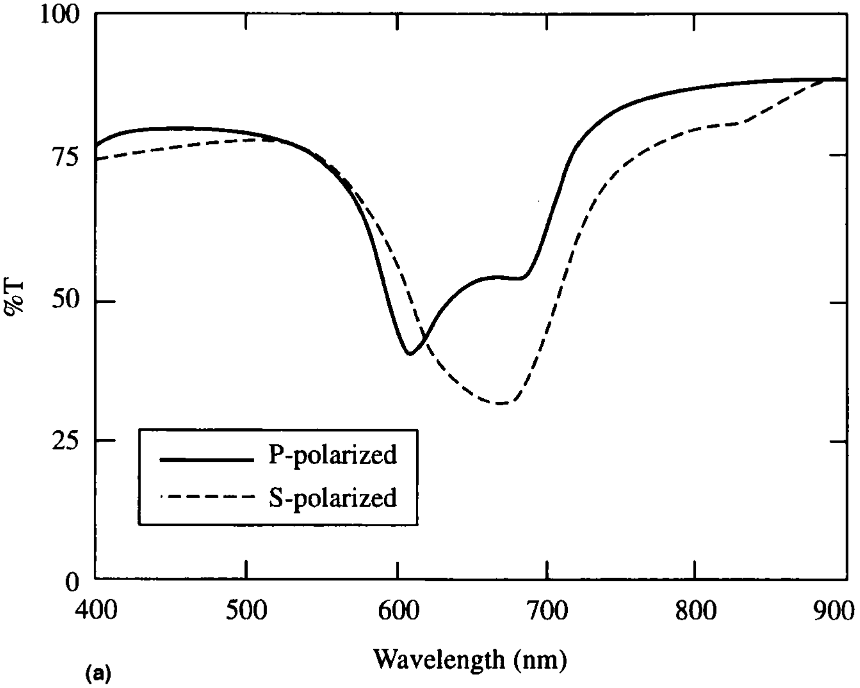

FIGURE 5 Transmittance of CLC film and flakes in linearly polarized, obliquely incident light. Linearly polarized light is incident at 30° to the surface normal of a CLC670 film and flake samples. The HN32 polarizer used in the background correction protocol was horizontally and vertically oriented respectively to give a P-, then S-polarized incident beam. The same asymmetry is exhibited by (a) unfractured film and (b) flakes. The slanted lines indicate the direction of each asymmetry.

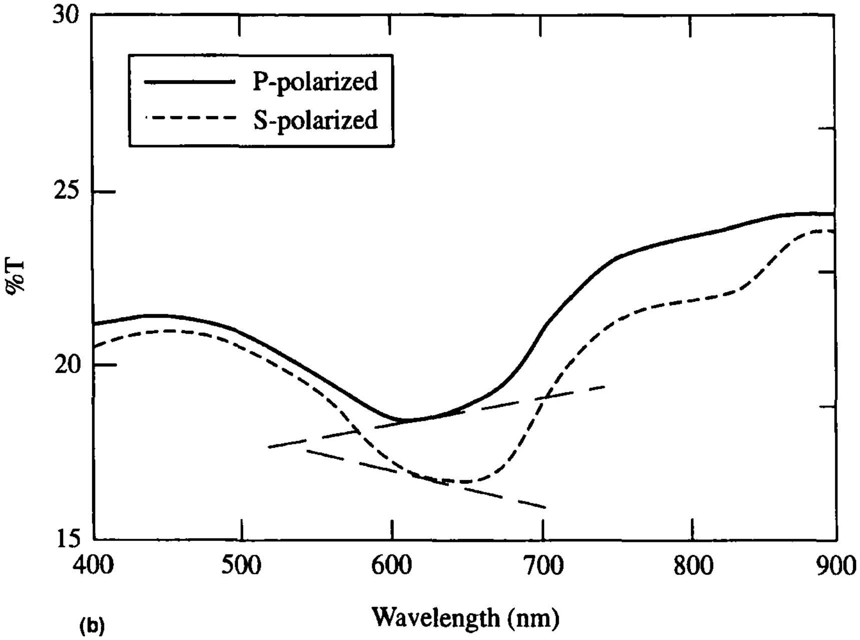

FIGURE 5 (Continued).

examined. Again, the incident angle was 30° from the surface normal. Figure 5(b) illustrates the comparison of S- versus P-polarization incident on CLC flakes. The same asymmetry appears for the CLC flake sample as for the CLC film sample. This comparison indicates that this asymmetry is a bulk phenomenon not related to the molecular orientation at the surface.

CONCLUSIONS

The asymmetry of a CLC transmission profile is due to two effects: the relative orientation of the incident polarization vector with respect to the molecular director at normal incidence and the relative orientation of the incident polarization vector with respect to the incident plane. We have confirmed the behavior for a CLC oligomer. We have used the asymmetry to draw conclusions about the molecular orientation at the surface of a CLC. Finally, we have used the new form of CLC – flakes – to experimentally separate the asymmetry phenomena.

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APPENDIX: MEASUREMENT AND CALCULATION OF THE CHOLESTERIC REFRACTIVE INDICES

Using three principal dielectric constants [33], ε_1 parallel to the molecular axis and $\varepsilon_2 = \varepsilon_3$ perpendicular to the molecular axis but parallel to the helix axis, Müller and Stegemeyer [34] defined the principal indices of refraction for a CLC in the Grandjean texture:

$$n_{e,n} = \sqrt{\frac{\varepsilon_1}{\varepsilon_o}} \tag{A.1}$$

$$n_{o,n} = n_{e,ch} = \sqrt{\frac{\varepsilon_2}{\varepsilon_o}} \tag{A.2}$$

$$n_{o,ch} = \sqrt{\frac{\varepsilon_3}{\varepsilon_o}} \tag{A.3}$$

where $n_{e,n}$ = extraordinary refractive index of the quasinematic layer
$n_{o,n}$ = ordinary refractive index of the quasinematic layer
$n_{e,ch}$ = extraordinary index of the cholesteric helix
$n_{o,ch}$ = ordinary index of the cholesteric helix
ε_o = permittivity of free space

They also defined these relationships between the cholesteric and nematic indices

$$n_{e,n} = \sqrt{2n_{o,ch}^2 - n_{e,ch}^2} \tag{A.4}$$

$$n_{o,n} = \sqrt{\frac{n_{e,n}^2 + n_{o,n}^2}{2}} \tag{A.5}$$

We define the average values as

$$\bar{n}_{ch} = \sqrt{\frac{2n_{o,ch}^2 + n_{e,ch}^2}{3}} \tag{A.6}$$

$$\bar{n}_n = \sqrt{\frac{n_{o,n}^2 + n_{e,n}^2}{2}} \tag{A.7}$$

Light rays whose **E** vector oscillates parallel to the helix axis sample the index of refraction designated as $n_{e,ch}$. Light rays whose **E** vector oscillates perpendicular to the helical axis/optic axis sample $n_{o,ch}$.