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Optical guided-wave study of a homeotropically aligned ferroelectric liquid crystal

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The excitation of optical modes is used to study the optical tensor configuration in a thin ferroelectric liquid crystal layer, cooled from the initially homeotropically aligned nematic phase. By monitoring the angular dependent reflectivity for plane polarized radiation coupled into the guided modes in the smectic C* layer and subsequently fitting the recorded data to predictions from multilayer optics theory, the optical tensor configuration in the layer is fully evaluated. Iteratively modelling the full tilt/twist profile in the cell, progressively converging the predicted reflectivity to experimental data, gives a complete and very well specified picture of the optical tensor throughout the cell. By studying the cell at various temperatures, the temperature dependence of the tilt of the major axis of the optic tensor (which may be related to the cone angle if the smectic layers are parallel to the cell surface) has been established. The temperature dependent optical dielectric constants have also been obtained.

1. Introduction

The excitation of optically resonant guided modes in a thin layer of liquid crystal has proved to be a very powerful tool for the study of the detailed director profile in a liquid crystal [1]. In recent years this method has attracted much attention because of the detail that may be obtained about the director alignment and consequential implications for displays and other liquid crystal devices [2, 3]. While this does not excite too much interest in the area of nematic liquid crystals which are now well-understood and for which there is a well-tested continuum theory, the same is not true for the ferroelectric smectic C* phase. At present, in the absence of a satisfactory continuum theory for this phase we have to rely on X-ray [4] and optical waveguiding techniques [5] to characterize the smectic layers and the optical tensor, respectively. In previous guided wave studies of the smectic C* phase, work has centred on the alignment formed on cooling the originally homogeneously aligned nematic phase [6, 7]. This results generally in chevroned layers and the quite complex tilt and twist profiles of the optical tensor. In this study we begin with the nematic phase homeotropically aligned and cool very slowly through the smectic A to smectic C* phase transition. A simple model would then suggest that if the smectic layers parallel to the cell surfaces are retained the optic tensor will progressively tilt away from, and twist about, the normal to the cell surface as the temperature is reduced. Detailed fitting of the angular dependent reflectivity establishes this behaviour for the material MIX 783 over the temperature range 32°C to 63°C.

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2. Experimental

The cell configuration for this study is illustrated in figure 1. A high index glass pyramid ($n = 1{\cdot}8$ at 632·8 nm) is coated with a thin (~ 39 nm) layer of silver which acts both as a mirror for light guiding and as tunnel barrier for the incident radiation. This silver layer also supports a surface plasmon which may be used to probe the near surface director profile. The other bounding medium is a thick silver layer (opaque) on a glass plate. The silver is characterized by performing an angular dependent p-polarized reflectivity experiment with $\lambda = 632{\cdot}8$ nm radiation incident from the prism side of the silver. This causes excitation of the surface plasmon, resulting in a minimum in the angular dependent reflectivity which, when fitted to theory, yields the silver parameters.

This gave a thickness of $38{\cdot}6(\pm 0{\cdot}5)$ nm with the dielectric constant

$$\epsilon = -17{\cdot}82(\pm 0{\cdot}03) + i0{\cdot}75(\pm 0{\cdot}03).$$

Having characterized the silver, both layers are coated with lecithin deposited from a dilute solution of lecithin in diethyl ether. Excess of lecithin is removed by wiping the surface in ether soaked lens cleaning tissue. The cell is then assembled in a clean environment using $\sim 3{\cdot}5\ \mu$m mylar spacers to space the silver coated glass plate away from the prism. This complete assembly is then mounted into an oven and heated to about 125°C, at which point it is capillary filled with the liquid crystal in its isotropic phase. MIX 783 has the phase sequence

$$\text{I } 119{\cdot}6^\circ\text{C}\text{–}116{\cdot}7^\circ\text{C N*}92^\circ\text{C S}_\text{A}\ 61{\cdot}2^\circ\text{C S}_\text{C}^* < -15^\circ\text{C C}.$$

Because the silver layers will significantly alter if held at elevated temperature (> 120°C) for several minutes, then the length of time the cell is kept at this temperature is minimized, with rapid cooling to well into the S_A phase being implemented. Once a temperature a few degrees above the S_A to S_C^* phase transition is reached (~ 65°C), cooling is stopped and the optical studies are begun. The sample in its oven is placed on a computer controlled rotatable table with an angular resolution of 0·01°, in a system

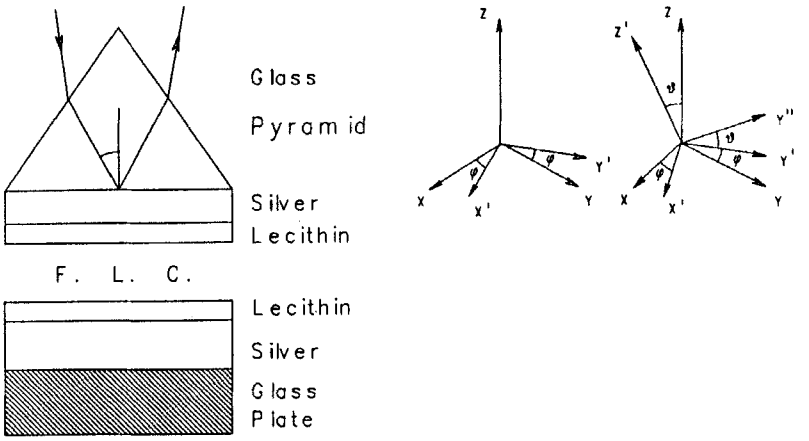


Figure 1. The sample cell construction used in the experimental work. The illuminating radiation is incident through a high index glass prism, and the cell is formed with a glass back plate, with a gap of $\sim 3{\cdot}51\ \mu$m. The coupling prism and back plate are coated with silver films and lecithin aligning layers to give homeotropic alignment. Insert: Definition of the twist and tilt angles in eulerian coordinates.

capable of monitoring angular dependent reflectivity to a precision of better than 0·1 per cent in reflectivity. Subsequently to allow formation of very large areas of well-aligned S_C^* phase, the sample is cooled at $\sim 0{\cdot}2°C\,h^{-1}$ through the S_A to S_C^* phase transition. The cooling process is then speeded up somewhat once the sample is a few degrees below the transition temperature.

3. Results and discussion

In the nematic and smectic A phases, the homeotropic alignment is very simple, with the director everywhere normal to the cell surface within an uncertainty of $\pm 1°$, if the parallel layering is retained. On cooling into the smectic C* phase, the average direction of the molecular long axis (in the smectic phase, the director) will try to spiral about the surface normal. Then, in order to discuss this alignment, it is necessary to define twist and tilt angles of the optical tensor associated with this phase. We choose an eulerian definition in which the original z axis is the normal to the surface plane and the original x axis is in the plane of incidence of the radiation (x, z is the plane of incidence). Then twist is the angle of rotation of the xy axis about the z axis rotating to $x'y'$ and tilt is the angle of rotation of the zy' axis about the new x' axes. These two angles are illustrated in figure 1.

If we suppose the material is uniaxial, then the third eulerian angle is irrelevant. Further the major axis of the optical tensor also coincides with the long axis director. Furthermore, the tilt angle, provided the density wave normal remains coincident with the z axis, which certainly at lower temperatures is rather unlikely, is always the cone angle. In actuality the S_C^* phase has to be biaxial, but evidence suggests that this is to such a limited degree that the uniaxial model used here is adequate.

Consider first data obtained from the homeotropically aligned S_A phase. At 63°C MIX 783 material is still in the S_A phase, giving data such as shown in figure 2, for p-in p-out reflectivity, R_{pp}. There is insignificant p to s conversion, indicating that the director is everywhere perpendicular (to $\pm 1°$) to the cell wall. By fitting the predictions

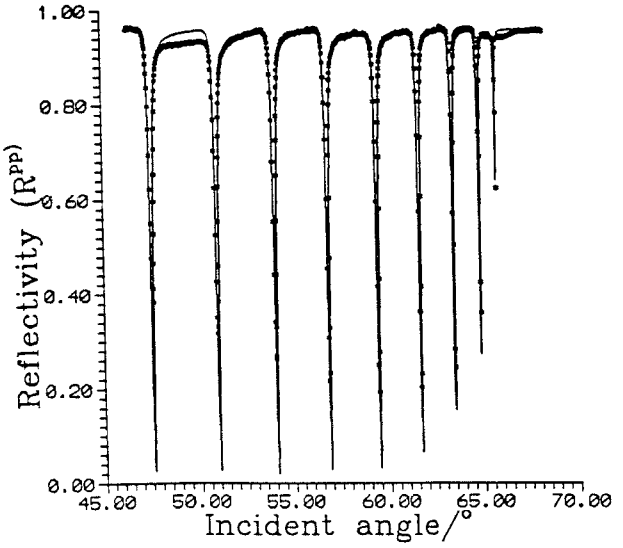


Figure 2. Reflectivity for p-polarized light. The sample temperature is 63°C, in the S_A phase of MIX783. Theoretical fit is the continuous line and the crosses are data.

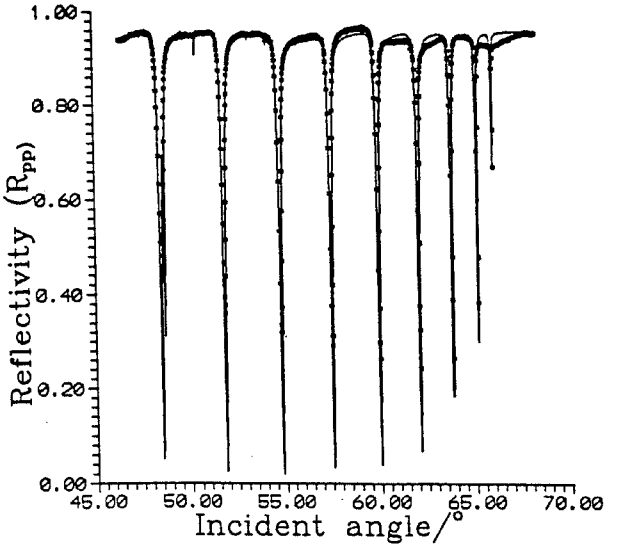


Figure 3. Reflectivity data for p-polarized light. The sample temperature is 53·8°C in the S_C^* phase of MIX 783. Theoretical fit is the continuous line and the crosses are data.

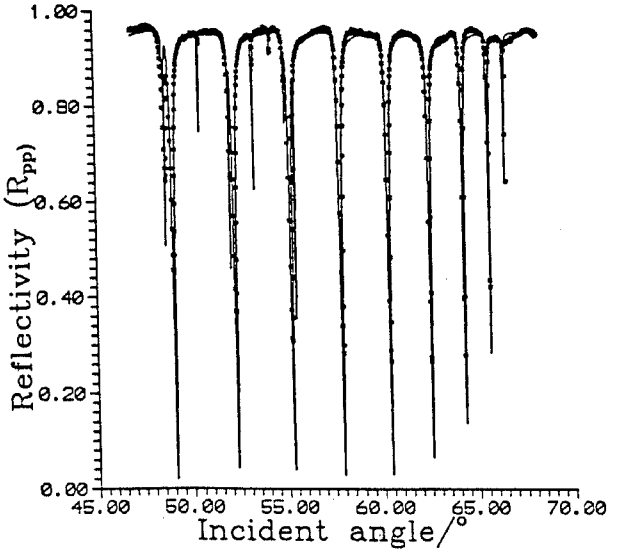
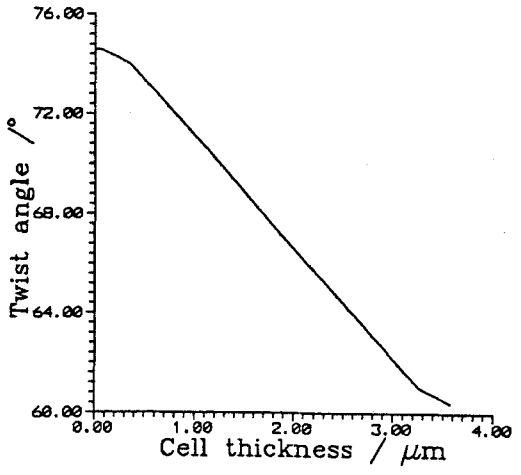


Figure 4. Reflectivity data for p-polarized light. The sample temperature is 45·9°C in the S_C^* phase of MIX783. Theoretical fit is the continuous line and the crosses are data.

of multilayer optics theory to this, we obtain unambiguously $\epsilon_\perp$, $\epsilon_\parallel$ and the cell thickness d. At 63°C we find for MIX 783, $\epsilon_\perp = 2{\cdot}195$, $\epsilon_\parallel = 2{\cdot}705$, and $d = 3{\cdot}57\,\mu$m. On cooling further into the S_C^* phase, a progressive twist and tilt of the director occur giving rise to some p to s mixing. This manifests itself in the appearance of weak resonances in the R_{pp} reflectivity. Examination of figure 3, taken at 53·8°C shows extra weak resonances at 48·6°, 50·5° and 53·8°. These are s type resonances which are now mixed in with the p type resonances in the R_{pp} reflectivity due to the tilt and twist of the



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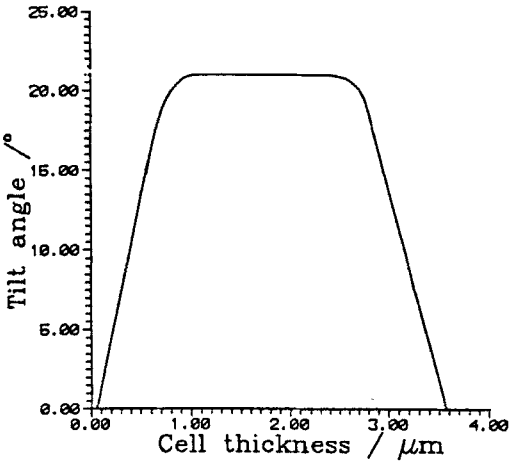

(*b*)

Figure 5. Twist (*a*) and tilt (*b*) profiles of the S_C^* phase at 45·9°C.

director. Careful fitting of the reflectivity data now yields $\epsilon_\perp = 2{\cdot}199$, $\epsilon_\parallel = 2{\cdot}715$, total twist ($\phi$) = 1·97° and centre cell tilt (θ) = 6°. In this modelling we keep the surface alignment homeotropic, allowing the director to twist and tilt throughout the cell in a manner which gives consistency with the reflectivity data with the majority of the cell having a fixed cone angle, i.e. fixed tilt, with a linear increase in twist. This is best illustrated at a lower temperature, for example 45·9°C, where much stronger p to s conversion occurs and the mixed modes are therefore stronger in R_{pp}, as shown in figure 4. At this temperature, the cone angle is now 21°, that is the majority of the cell has a director tilt of 21°, with boundary regions some 0·5 μm thick, with the twist angle varying nearly linearly across the cell. The twist/tilt profiles which best fit these data are shown in figure 5. To obtain fits to the data, a careful iteration procedure was adopted, beginning from the simplest uniformly tilted, linearly twisting slab, and ending, with constrained boundaries, with models such as those shown in figure 5. The uncertainty in cone angle here is then about $\pm 0{\cdot}1°$, with the error in the integrated twist being of the same order.

Repeating this fitting procedure for all temperatures in the S_C^* phase gives a variation of integrated twist, a measure of the pitch of the FLC (pitch = cell thickness $\times$ 360/twist). This shows, by plotting the inverse of twist, see figure 6, that the minimum pitch is of the order 78 μm, with this going to ∞ at the phase transition. Also we obtain the variation of tilt, the cone angle for this uniaxial model, with temperature as shown figure 7. The cone angle tends to zero at the phase transition. Further, in the process of obtaining tilt and twist angles, we also find values for $\epsilon_\perp$ and $\epsilon_\parallel$, all the relevant numbers being given in the table.

It has to be appreciated that this relatively simple picture of the director behaviour in the cell is only an approximation. The technique employed, although rather sensitive to director twist, is not over sensitive to tilt and at the higher temperature, a 1° tilt is not readily distinguished. Thus although we have determined rather specific tilt and twist profiles, it must be borne in mind that these have uncertainties of the order $\pm 1°$ in all angles. It is also important to appreciate that the optical technique used here does not give direct information with respect to the layering of the smectic phase.

In the S_A phase, the results are fully consistent with the director being everywhere normal to the cell surface, the layers being parallel to the cell walls. On cooling into the S_C^* phase, once again the results are still fully consistent, in the bulk of the cell with this idea. However, it has to be appreciated that as the layer spacing is reduced on cooling, some small tilt of the layers or disclination mediated flow of material must occur. Indeed below 35°C, the optical modes start to distort and there is strong evidence for multidomains being present in the beam area of 1 mm^2. If the sample is cooled quickly, the onset of this disclination dominated region occurs at a much higher temperature. Thus we conclude that with slow cooling, for temperatures above 35°C, as the layers shrink, enough molecular mobility occurs to allow the structure to reorganize itself without disclinations being an observable problem.

A final point worthy of note is that as the temperature is lowered, the boundary region, in figure 5 shown to be about 0·7 μm thick, progressively shrinks towards the surface. Then it becomes very difficult to discover in detail the director profile in this region. We have chosen as an approximation to treat this region as having a simple linear variation in tilt and twist, since this gives an acceptable fit to the data. To use a

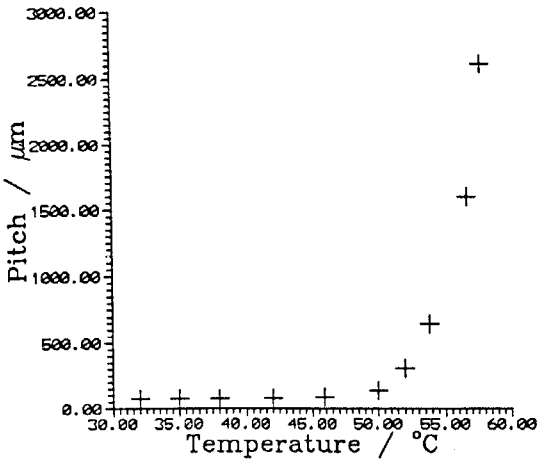

Figure 6. Pitch of the helix as a function of temperature.

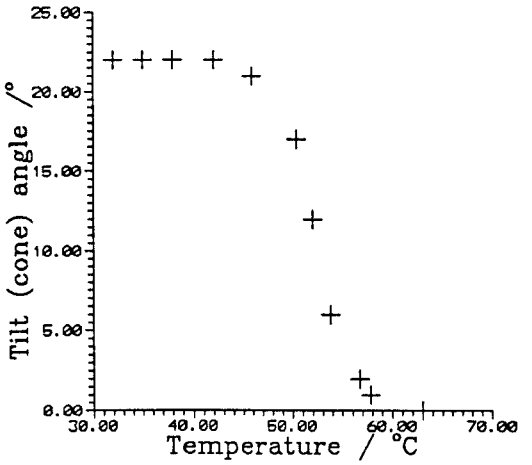

Figure 7. Tilt(cone) angle as a function of temperature.

Optical dielectric constants and twist, tilt angles at a series of temperatures for MIX783.

T/°C	$\epsilon_\perp$	$\epsilon_\parallel$	ϕ/°	θ/°
63·0	2·183	2·701	0·00	0·0
57·8	2·188	2·706	0·49	1·0
56·7	2·190	2·707	0·80	2·0
53·8	2·199	2·715	1·97	6·0
52·0	2·201	2·724	4·16	12·0
50·4	2·202	2·732	9·18	17·0
45·9	2·207	2·738	14·19	21·0
42·0	2·207	2·740	16·14	22·0
38·0	2·208	2·742	16·59	22·0
35·0	2·218	2·750	16·59	22·0
32·0	2·219	2·753	16·59	22·0

more elaborate non-linear variation in this surface regime would perhaps be more physically sensible, but we cannot readily distinguish between the near infinity of models then available; we have therefore kept to the simplest, linear, approximation. Because of this we cannot be confident that our model of the director profile in the vicinity of the surface, at low temperatures, is the correct one; it is only an acceptable model as far as the present data are concerned.

4. Conclusions

Using the simple structure of a homeotropically aligned smectic C* material, Merck BDH MIX783, probed by the optical excitation of guided modes, we have studied the twist and tilt of the optical tensor as a function of temperature. The results are consistent with the smectic layers remaining parallel to the cell surface, the tilt angle being a measure of the cone angle, which progressively increases as the temperature is decreased, with the integrated twist, a measure of the inverse pitch also increasing as the temperature is decreased. These are the first published results of this type of study of a homeotropically aligned ferroelectric liquid crystal and serve to illustrate the power of the technique.

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