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Optical Characterisation of a Homeotropically Aligned Ferroelectric Liquid Crystal Using Half Leaky Guided Modes

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Optical excitation of a half leak guided modes has been used for characterising in detail the optical tensor profile in a homeotropically aligned ferroelectric liquid crystal (SCE8) layer.

In the S_A phase it is found, by careful fitting of multilayer Fresnel theory to angle dependent transverse magnetic to transverse electric conversion reflectivity data, that at 1°C above the S_C^* to S_A phase transition there is a finite pre-tilt of the S_A layers.

Further fitting of similarly recorded data in the S_C^* phase gives details of the cone angle and the chiral pitch of the S_C^* phase. These results not only serve to obtain material parameters for this S_C^* phase but also illustratee fully the enormous potential of the half leaky guided mode technique.

Keywords: Ferroelectric liquid crystal, homeotropic, half-leaky guided mode

INTRODUCTION

Since the discovery of ferroelectricity in S_C^* liquid crystals by Meyer *et al.*,[1] Ferroelectric S_C^* liquid crystals (FLCs) have been studied extensively primarily because of the possibility of their application in high speed optical switches and in displays. Experimental attention has focused mainly on the homogeneously aligned state particularly after bistable switching was demonstrated in such a surface stabilized FLC (SSFLC) by Clark and Lagerwall.[2] However the homeotropically aligned cell also has potential interest since it may allow the experimental determination of the FLC parameters such as pitch and perhaps some elastic constants.[3–7] The weaker homeotropic alignment, by comparison to the homogeneous, may also allow for a closer approximation of the FLC structure in a homeotropic cell to the "free" material structure.

Of a range of methods for determining the pitch of an FLC in a homeotropically aligned cell the most common one is to measure the transmittance as a function of incident wavelength. At a fixed temperature the observed change in transmittance, due to selective reflection[4,5,7] allows the determination of the FLC pitch. An alternative is to fix the wavelength and vary the temperature,[6] again selective strong reflection occurs when the Bragg condition is satisfied. It must however be appreciated that although the helical structure of the homeotropically aligned FLC may give rise to strong Bragg reflection and thereby accurate pitch determination there are several problems.

Firstly, with light incident normally on the homeotropically aligned cell surface and the helical axis of the FLC along this incidence direction, in general only the first-order Bragg reflection may generally occur.[3–6] This of course means that only short FLC's ($\leq 2\,\mu m$ pitch) may be readily measured using conventional visible and near-infra-red sources. Modifications to this simple technique using applied fields[3] or oblique incidence[7] still does not help with longer pitch material. Thus in general for longer pitch FLC's it is only the pitch of the N^* phase, often determined by the Cano wedge technique, that is given.

A second problem with the selective reflection technique is that it is the optical pitch $n_{av}P$ which is determined.[5] To obtain the real pitch P, the "average" refractive index n_{av} also has to be established.[6] This is not very satisfactory since the analytic formula used to give n_{av} is a rather crude approximation while a full treatment of the situation demands rather more elaborate experimentation.

Thirdly, even if the crude approximation for n_{av} is to be used to determine P, the cone angle of the long axis director of the FLC, the ordinary index and the extraordinary index (if the FLC is assumed uniaxial) have still to be determined by some other procedure.[3–7] Often the measurements needed to discover these parameters are performed on the FLC aligned in a different manner in another cell about which further assumptions have to be made with respect to the alignment of the director. This adds an extra level of uncertainty in parameterising the material.

Finally (and less severely) the optical pitch measurement using selective reflection is often based on the assumption that the helical axis is perpendicular to the cell walls. This may not be true, the FLC layers may have some pre-tilt.

Thus, up till now, there is no really satisfactory procedure for determining the pitch of FLC materials, even for relatively short pitch materials. For long pitch materials ($\geq 2\,\mu m$) the situation is even worse. But there is no need to resort just to the simple, and convenient, selective reflection technique to determine the helical pitch. If for example the full optical tensor profile within a cell may be determined then the helical structure will be well quantified also and thereby the helical pitch.

Recently the most powerful optical technique for determining the director profile of liquid crystals in a thin cell (the half leaky guide mode-HLGM method) has been reported.[8,9] In this new technique, the chosen experimental geometry is that of a high index glass pyramid (replaced by a prism, matching fluid and high index plate if required), an aligned liquid crystal layer and a low index glass substrate. Ideally the high index glass should have an index above the largest of the liquid crystal, while the low index plate (the substrate) should have an index less than the lowest index of the liquid crystal. This gives a range of incidence angles, from the pseudo-critical angle between the high index glass and the liquid crystal and the real critical angle between the high index glass and the low index glass, over which half-leaky guided modes are excited. Furthermore for any out-of-incidence plane director alignment there is significant TM(p) to TE(s) conversion in the cell. Then in the half-leaky guided wave angle window the p to s conversion reflectivity gives a series of sharp peaks which are remarkably sensitive to the director profile within the cell. The reflectivity peaks are sharp because the optical field is fully reflected at the liquid crystal substrate boundary, while being relatively strongly reflected at the high index glass-liquid boundary. Also by measuring the p to s or s to p conversion signals which result from the director being

out of the plane of incidence the technique becomes in effect of higher order than measurement of the simple p to p or s to s reflectivity. This thereby allows the detailed characterisation of the optical tensor profile in the cell.

Thus, in principle, if we study a homeotropically aligned FLC with the HLGM method then we should be able to obtain the optical tensor cone angle from the tilt of the primary axis and from the twist of this axis through the cell we will obtain the pitch. In practice even the relatively weak homeotropic alignment forces may disturb the FLC material from its "free" state.

In this present study a thin homeotropically aligned FLC layer has been investigated using the HLGM technique. By fitting predictions from multilayer Fresnel theory to the p to s conversion reflectivities for the HLGM region full optical characterisation is established.

EXPERIMENTAL

The sample geometry used in the experiment is illustrated in Figure 1. An approximately 3.5 μm thick layer of FLC (BDH-SCE8) is homeotropically aligned in the N^* phase between a high index pyramid ($\varepsilon = 3.2400$ at 632.8 nm) and a low index substrate ($\varepsilon = 2.1415$ at 632.8 nm). The homeotropic alignment is established by the use of a thin lecithin layer. This layer is deposited on both the pyramid and substrate from diethyl-ether solution, excess after drying being carefully washed off with diethyl-ether. The pyramid and glass substrate with their lecithin coatings are then clamped together with 3.5 μm mylar spacers and the complete assembly placed in a temperature controlled oven with the temperature stabilised to ± 0.1°C. This empty cell is heated to 120°C and capillary-filled with SCE8. Once full the cell temperature is quickly reduced to a temperature low in the N^* phase (≈ 100°C). Subsequently the cell is cooled very slowly through the S_A to S_C^* phase transition (≈ 59°C) in order that large domains

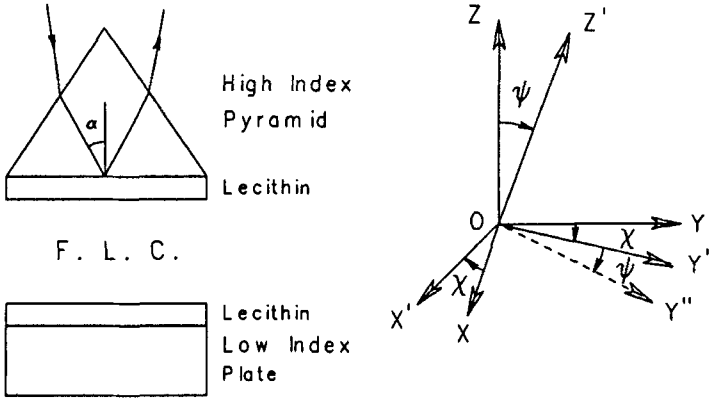


FIGURE 1 The sample geometry used in the experiment.

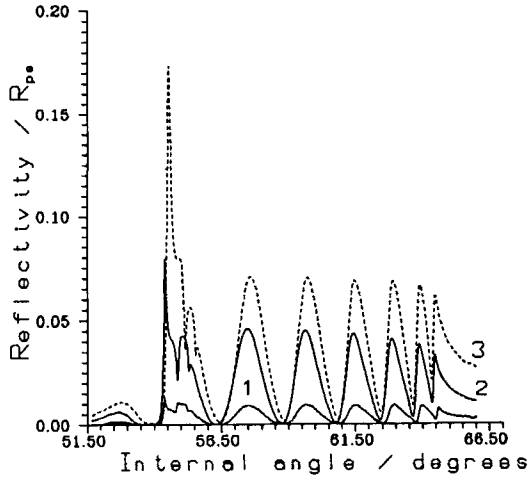


FIGURE 2 The experimental data for three different temperatures. Curve 1: 60.3°C, Curve 2: 55.1°C and Curve 3: 34.4°C.

(bigger than the sampling beam area of 2 mm^2) form in the S_C^* phase. This requirement of slow cooling is aided by the thermal inertia of the glass pyramid and substrate. Typically the rate of cooling across the phase transition is less than 1°C per hour. Once a temperature a few degrees below the S_A to S_C^* transition is reached ($\approx$ 55°C) cooling is accelerated to a few degrees per hour.

In order to obtain the data in the required form of reflectivity from the pyramid-liquid crystal boundary as a function of angle of incidence the complete cell plus oven assembly is set on a computer controlled rotation stage. A parallel monochromatic light beam (He–Ne, 632.8 nm) mechanically chopped at 1.7 kHz to allow the use of phase sensitive detection, is incident on one face of the pyramid such that it arrives at the liquid crystal layer at the desired angle of incidence. The incident beam is plane polarised, either *s* or *p*, in the experimental results presented here primarily *p* is utilised and a second polariser is placed in front of the detector to give either *p* to *p* or *p* to *s* reflectivities. In this article we only present details of the *p* to *s* (the conversion) signals. To allow for any variation in laser source intensity a small ($\approx$ 4%) reflection is taken from the input beam to act as reference.

The data presented begins with the material just in the S_A phase at 60.3°C. It might be anticipated that with perfect homeotropic alignment the director in the S_A phase would be normal to the cell walls with the density wave normal similarly aligned. Then there can be no *p* to *s* conversion signal. As shown in Figure 2 (curve 1) this is not the case. There is no question that there is a finite *p* to *s* conversion, of order 1% over much of the HLGM angle window. This shows that the director in the S_A phase is somewhat surprisingly not normal to the walls but tilted off by some finite angle.

On cooling the sample further into the S_C^* phase the reflectivity features apparent in the S_A phase strengthen with the conversion strength rising as the sample is cooled. Two other sets of data are here presented, one at 55.1°C and the other well into the S_C^* phase at 34.4°C.

RESULTS AND DISCUSSION

Firstly we need to define our coordinate frame to allow a clear description of the director tilt and twist profiles. We choose on Eulerian definition in which the original axis is the normal to the cell wall and the original x axis is in the plane of incidence of the radiation. Then twist is the angle of rotation of the xy axes about the z axis, rotating to $x'y'$ and tilt is the angle of rotation of the zy' axes about the new x axis. These angles are illustrated in Figure 1. Of course strictly it is known that the S_C^* phase is biaxial but evidence suggests that this is to such a limited extent[9] that for the homeotropic alignment used here it is negligible. In this case the third Eulerian angle is irrelevant and the major axis of the optical tensor also coincides with the long axis director.

Consider now the S_A phase. It has already been mentioned that there is significant p to s conversion. In which case the uniaxial axis cannot be everywhere normal to the cell wall. Then we need to fit the data of Figure 2 (curve 1) to Fresnel model predictions based on a reasoned profile of the S_A director. The Fresnel multilayer modelling uses a scattering matrix method[10] with the liquid crystal layer divided into 90 sub-layers. A guess is first made of the director profile including the cell thickness, $\varepsilon_\parallel$, $\varepsilon_\perp$ and the tilt and twist angles through the cell. This produces a model prediction of the angle dependent reflectivity which is compared with the data. Adjustments are then iteratively made to all the parameters until a minimum least squares fit to the data is obtained.

In Figure 3 is shown in detail the fit of model theory to the data. The fit, the continuous line, is superb with two minor discrepancies, one in the vicinity of 52.2°, which is partly associated with scattering in the fully leaky regime, the other at the sharp minimum at 54.7°. The shallowing here of the experimental data is partly, but not fully, attributable to the angle spread in the incident laser beam and suggests further detailed refinement in our model, such as a near surface phase change, which we have

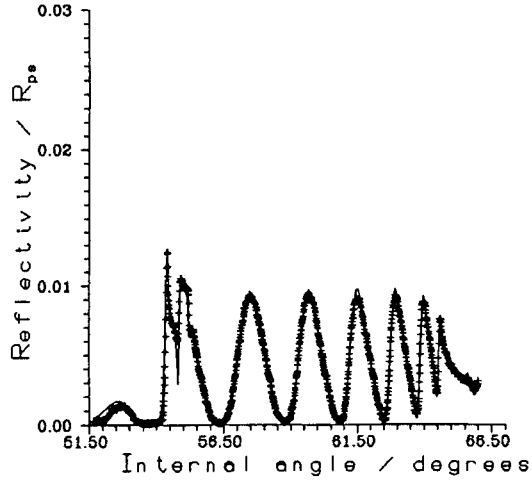


FIGURE 3 The experimental data (crosses) and theoretically fitted results (full line) for a temperature of 60.3°C. The fitted parameters of the geometry are: wavelength, $\lambda = 632.8$ nm; pyramid, $\varepsilon = 3.2400$; liquid crystal, $\varepsilon_\parallel = 2.6512 + i0.0002$, $\varepsilon_\perp = 2.1920 + i0.0002$, thickness 3.33 μm, tilt angle 3.9° and twist angle 15°; substrate, $\varepsilon = 2.1415$.

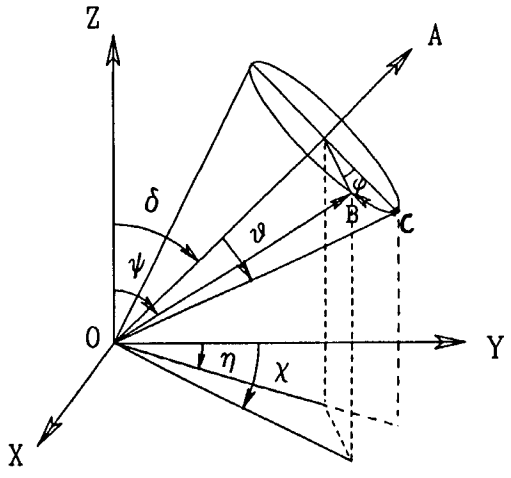


FIGURE 4 The relation between the layer normal and the director of the FLC.

yet to incorporate. The model used to produce the excellent fit has the following parameters: the cell thickness is $d = 3.33\ \mu m$, the two optical permittivities for the liquid crystal at this temperature are $\varepsilon_{\parallel} = 2.6512 + i0.0002$ and $\varepsilon_{\perp} = 2.1920 + i0.0002$ with a tilt angle in the majority of the cell of 3.9° and a twist angle of 15°. The lecithin anchoring is quite weak and because of this there appears to be only a very thin surface region of 0.15 μm thickness at the pyramid interface where there is a variation in tilt. A linear variation from 1.7° to 3.9° over this 0.15 μm region is satisfactory to fully fit the data. This change of tilt implies layer bending in the S_A phase while the 3.9° tilt raises questions over assumptions about the uniaxial axis being normal to the cell walls in the S_A phase of FLC material. Fortunately, the 3.9° tilt does allow for layer shrinkage when going from S_A to S_C^* phase, since material and extra layers can flow in from the cell edge. Already then the HLGM technique has provided a rather surprising result, the S_A layers, and therefore presumably the S_C^* layers are not necessarily parallel to the cell walls. Before however going on to look in detail at the S_C^* phase results we need to consider a little more the geometry of this phase.

In Figure 4 O-XYZ is the laboratory coordinate system and $\underline{OA}$ is the layer normal (density wavevector direction) with its tilt angle δ and twist angle η. The cone angle of the FLC is θ while $\underline{OB}$ is the long axis director having a tilt angle ψ and twist angle χ. Finally ϕ defines the azimuthal angle of the projection of the long axis director on the layer surface. Some geometry then shows that

$$\psi = \cos^{-1}(\cos\theta\cos\delta - \sin\theta\sin\delta\cos\phi) \tag{1}$$

and

$$\chi = \eta \pm \cos^{-1}\left(\frac{\cos\theta - \cos\delta\cos\psi}{\sin\delta\sin\psi}\right) \tag{2}$$

where when ϕ is from C to B (see Figure 4) the plus sign is taken and when ϕ is the inverse then the minus sign is taken.

Now using the above formula fitting to the S_C^* data is started with $\delta = 3.9°$ and $\eta = 15°$ as beginning conditions. Then again parameters are iteratively varied until fits are obtained, first to the data near to the S_C^* to S_A phase transition, the data at 55.1°C and then for the data taken much lower in temperature at 34.4°C. The fitted results are shown in detail in Figures 5a and 6a respectively. In Figure 5b, 5c, and 6b and 6c we show the corresponding final director twist and tilt profiles used to produce the fits to the data.

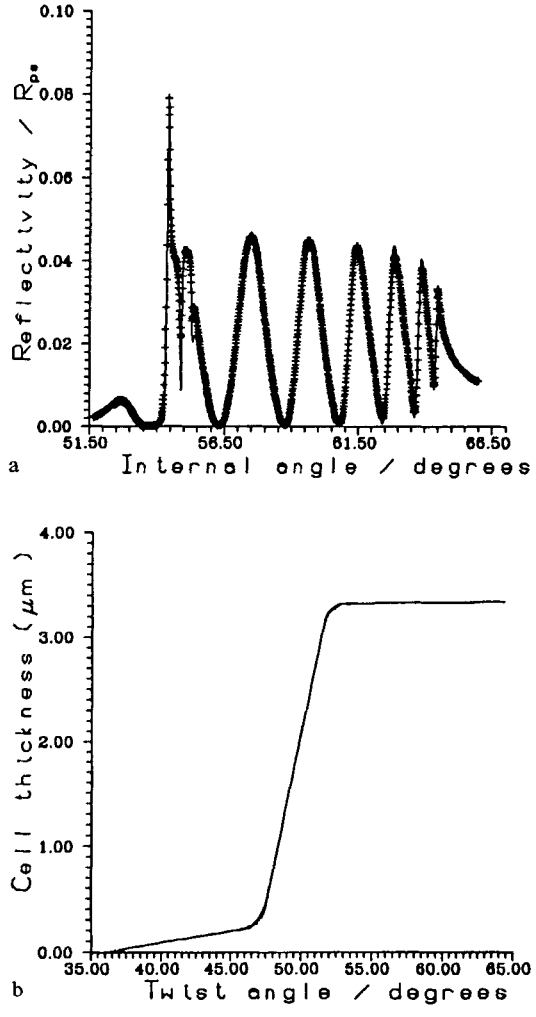


FIGURE 5 (a) The experimental data (crosses) and theoretically fitted results (full line) for a temperature of 55.1°C. (b) The twist angle profile in the cell. (c) The tilt angle profile in the cell. The fitted parameters of the geometry for (a) are: wavelength, $\lambda = 632.8$ nm; pyramid, $\varepsilon = 3.2400$; liquid crystal, $\varepsilon_\parallel = 2.6580 + i0.0006$, $\varepsilon_\perp = 2.1960 + i0.0003$, thickness 3.33 μm, twist and tilt angle profiles as shown in (b) and (c) respectively; substrate, $\varepsilon = 2.1415$.

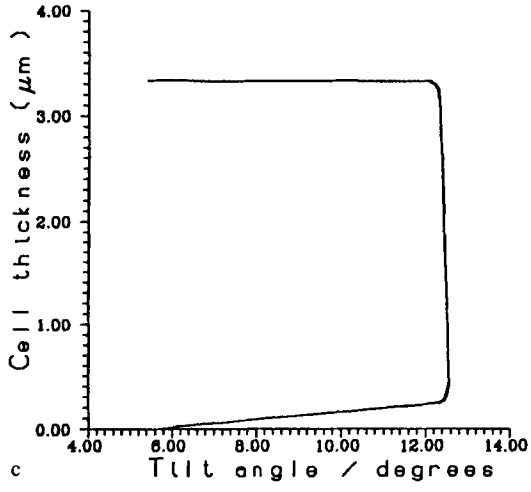


FIGURE 5 (*Continued*)

It should be appreciated that these profiles are vital to produce the fits to the data. The lack of summetry at lower temperature is essential, with tilt angles being determined to better then 0.5° and twist angles to better than 5°.

These results all indicate that there is a significant tilt of the layers at all temperatures. For example at 55.1°C where the director tilt angle is almost constant at 12.5° throughout much of the cell (see Figure 5c) we expect from the manufacturers (BDH Ltd) information a cone angle of only 9.3°. A temperature error in excess of 6° would be

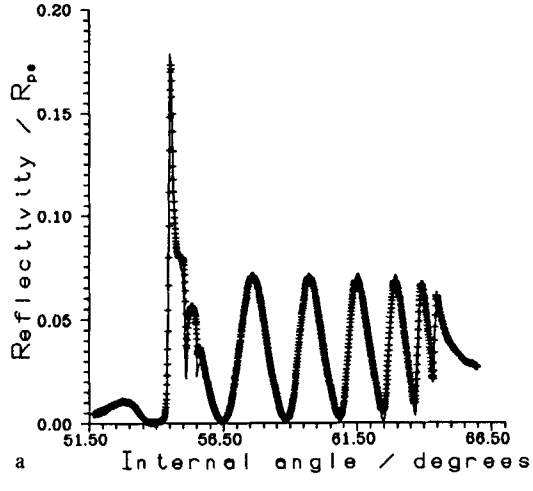
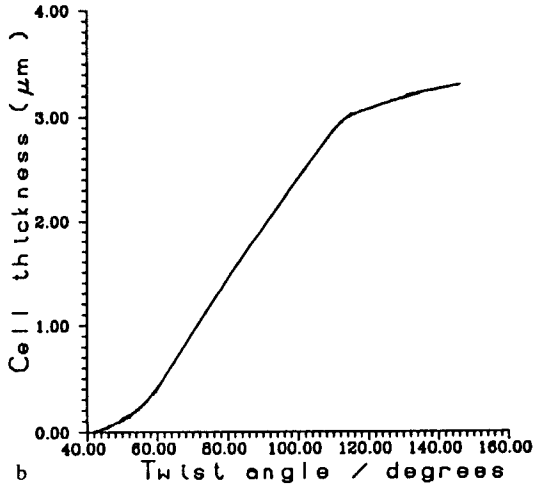
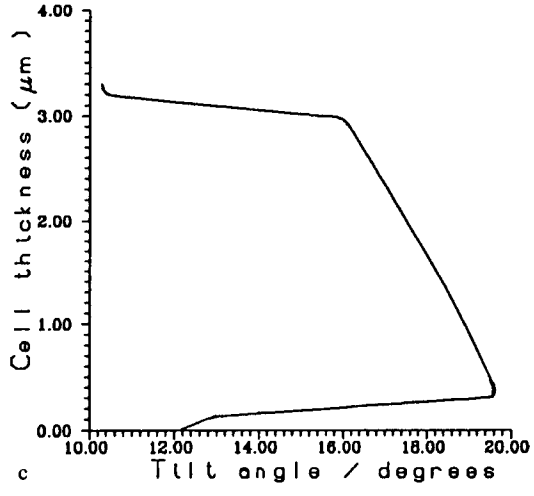


FIGURE 6 (a) The experimental data (crosses) and theoretically fitted results (full line) for a temperature of 34.4°C. (b) The twist angle profile in the cell. (c) The tilt angle profile in the cell. The fitted parameters of the geometry for (a) are: wavelength, $\lambda = 632.8$ nm; pyramid, $\varepsilon = 3.2400$; liquid crystal, $\varepsilon_{\parallel} = 2.6860 + i0.0008$, $\varepsilon_{\perp} = 2.2080 + i0.0006$, thickness 3.33 μm, twist and tilt angle profiles as shown in (b) and (c) respectively; substrate, $\varepsilon = 2.1415$.



b



c

FIGURE 6 (*Continued*)

required to shift this to a cone angle of 12.5°, but our maximum uncertainty in temperature is only ±0.1°C. This then directly implies a layer tilt of order several degrees. Likewise data at 34.4°C (see Figure 6c) correspond to a director tilt angle varying from 16° to 19.5°. This is not possible for the cone angle within the cell and the obvious interpretation, once again is a layer tilt of order several degrees (also see Figure 4 and Equations (1) and (2)). Thus the results at 60.3°C which show a 3.9° tilt of the director, and we believe layers in the S_A phase are fully substantiated by the other, lower temperature, data. If we assume that the 3.9° layer tilt remains constant throughout the S_C^* phase then the fits to the data at 55.1°C and 34.4°C give a cone angle of 9.5° and 17.0° respectively. These two numbers accord very well with the information supplied by the manufactures of 9.3° and 17.2° respectively. The optical permittivities of

the FLC are also obtained by the fitting being determined as $\varepsilon_{\parallel} = 2.6580 + i0.0006$ and $\varepsilon_{\perp} = 2.1960 + i0.0003$ for 55.1° and $\varepsilon_{\parallel} = 2.6860 + i0.0008$ and $\varepsilon_{\perp} = 2.2080 + i0.0006$ for 34.4°C. From these results note that both $\varepsilon_{\parallel}$ and $\varepsilon_{\perp}$ increase with decrease in temperature as does the birefringence at this wavelength.

From the director profiles it is clear that a substantial proportion of the cell, excluding regions of order 0.3 μm in thickness near the cell surfaces are effectively bulk. Then by taking this bulk region and determining the integrated twist it is possible to establish the pitch of the FLC at these temperatures. At 55.1°C the results give a pitch of (184 ± 10) μm, while at 34.4°C this has shrunk to (17.3 ± 1.0) μm. Clearly these numbers are still to a certain extent being influenced by the lecithin anchoring and further experiments need to be undertaken on cells of different thickness to establish the influence of the boundaries on constraining the pitch of the material.

CONCLUSIONS

The half leaky guided mode method has been utilised to fully characterise the optical tensor in a homeotropically aligned FLC. In the S_A phase results show conclusively that the density wave normal is *not* everywhere perpendicular to the cell walls but instead is tilted by about 4°. On cooling into the S_C^* phase the chiral nature of this phase manifests itself resulting in much stronger p to s conversion. From careful fits to the data in the S_C^* phase not only have $\varepsilon_{\parallel}$ and $\varepsilon_{\perp}$ been determined but also the cone angle and the pitch of the helix of the FLC phase. There are also very clearly thin surface regions in which the S_C^* material is influenced strongly by the presence of the licithin aligning layer. Further experiments need to be undertaken to try to unravel the structure of the near surface region in more detail. For the time being however it is clear that the HLGM technique is already beginning to uncover behaviour which had been unexpected while it is also able to be used to quantify in substantial detail the materials.

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