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The optical tensor configuration in a surface stabilized ferroelectric liquid crystal determined by using half leaky guided modes

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The optic tensor configuration in a surface stabilized ferroelectric liquid crystal cell is investigated using optical excitation of half leaky guided modes. A thin ferroelectric liquid crystal layer is confined between a high index pyramid, with an index greater than the maximum of the liquid crystal, and a glass substrate having an index less than the minimum of the liquid crystal. Using standard attenuated total reflection experimental procedures, over a small angle range a series of sharp resonant peaks are recorded in the s-polarized reflectivity using p-polarized incident light. These peaks are extraordinarily sensitive to details of the optical tensor configuration within the cell. Fitting theoretically modelled reflectivities from multilayer Fresnel theory to the data allows determination of near surface alignment, bending of the chevron, surface tilt angle and biaxiality. To give a clear physical explanation for the great sensitivity of the technique, the electromagnetic field component distributions in the cell are also presented and analysed. The results confirm that the half leaky guided mode method has enormous potential for the study of the optic tensor configuration in liquid crystal layers.

1. Introduction

The possibility that a cell containing a chiral smectic C ferroelectric liquid crystal would exhibit bistable properties was first predicted by Meyer [1] in 1975. Using surface stabilization to obtain two switched states a device of this nature was demonstrated by Clark and Lagerwall [2]. In this device the ferroelectric properties of the chiral smectic C (S_C^*) liquid crystal were used to allow switching between two states of differing optical tensor orientation by application of DC pulses. Since then a substantial body of work has been accumulated with the objective of determining the S_C^* structure within the cell to give a clear physical understanding of the switching mechanism.

One of the primary techniques for examining the director profile in such devices has been polarized optical microscopy [3]. Examination of a S_C^* filled cell, with surface alignment, using a polarizing microscope shows various coloured states which may be interpreted in terms of director configurations within the cell. For several years, the simple bookshelf geometry in which the smectic layering lay perpendicular to the cell walls (the density wave normal being in the plane of the cell) was the favoured model and results were interpreted in terms of optic tensor configuration within this structure [2]. Subsequently X-ray scattering studies [4, 5] have revealed that the smectic layering is tilted away from this simple bookshelf configuration into a chevron structure. The realization of this chevron structure meant that the optical microscopy data had to be re-interpreted [5] and further indicates the need for development of more powerful

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optical techniques to evaluate the full details of the optical tensor configuration in the cell. The optical microscopy techniques are all integral methods, in that they integrate the optical response throughout the thickness of the cell. However, the X-ray studies show that there is a variation in structure through the cell and hence a method for probing the spatial variation across the cell needs developing.

This spatial probing is achieved by the excitation of a series of guided modes in a thin liquid crystal layer, each mode having a different spatial distribution of its optical field across the cell. Thus by monitoring the optical response of a cell which supports several such modes, more detail of the optical tensor, but not the layering, is obtained [6]. Using this technique Elston and Sambles [7,8] have excited a series of guided modes, as well as a surface plasmon resonance, in a metal-clad surface stabilized ferroelectric liquid crystal waveguide. Their results indicate that the optic tensor configuration in the cell is largely equivalent to a uniform slab of material with the optic axis rotated from the surface alignment direction (defined in the S_A phase) by a few degrees. This is consistent with the X-ray data indicating a chevron structure in the smectic layering.

This technique of using metal-clad waveguides, while quite powerful, suffers from several problems. Firstly, because of the absorption of radiation by the metal layer, and its strong reflectivity for transverse electric (TE or s) polarized radiation, then the transverse magnetic (TM or p) to s conversion, the signature of twist and tilt in the cell, is very weak. This limits the usefulness of the technique for studying subtleties in the optic tensor configuration, for example small pre-tilt angles, small changes in twist, biaxiality, etc. Further, the requirement of metal-coating also inhibits the study of practical device structures, since virtually all such structures use transparent conducting electrodes (ITO) and rubbed polyimide aligning layers. With the relatively soft metallic layers, it is quite difficult to use rubbed polyimide alignment. Since the director alignment throughout the cell depends on the nature of the surface anchoring, then this limitation on the use of rubbed polyimide is quite severe.

A way of overcoming these limitations of the metal coatings would appear straightforward—dispense with them altogether. Lavers and Sambles [9] did this, exciting now a series of leaky guided modes in an ITO-coated, rubbed polyimide aligned surface stabilized ferroelectric liquid crystal layer. The problem here, very clearly illustrated by their results, is that the leaky modes are very broad and this severely limits the precision with which we may determine the optical tensor configuration within the cell.

A third, and the most powerful, variant on this guided wave technique has now appeared [10]. In this new technique, the chosen geometry is that of a high index glass pyramid (which may be replaced by a pyramid, matching fluid and glass plate if required), an aligned liquid crystal layer and a low index glass plate. Now there is a range of incident angles over which half leaky guided modes are excited within the liquid crystal layer. Furthermore the strong p to s conversion in this angle range, not inhibited by metal layers, is very sensitive to details of the optical tensor profile within the cell. This geometry, in which ideally the high index glass should have an index greater than the highest index of the liquid crystal, and the low index glass should have an index lower than the lowest index of the liquid crystal, allows ITO coatings and rubbed polyimide layers without degrading the optical sensitivity.

Analytic treatment, numerical modelling and experimental data all confirm the existence of an incident angle range, a window of in-plane momentum values, over which half leaky modes exist, from the pseudo-critical angle between the high index

pyramid and the effective index of the liquid crystal and the real critical angle between the high index pyramid and the low index substrate. In this momentum window, because the optical field is fully reflected at the liquid crystal substrate boundary, being thus evanescent in the substrate, while propagating in the liquid crystal layer, there are quite sharp half leaky guided modes in the cell. Also the early results [10] show that if the liquid crystal optical tensor axes are tilted and twisted from the plane of incidence, then strong p to s conversion may occur over this momentum window, conversion which relies entirely on the lowering of symmetry brought about by the liquid crystal optical tensor. Thus, using crossed polarizers between input and output beams, and studying in detail the optical response over this momentum window, a very powerful technique is established. In effect this is a second order technique—the more usual p to p and s to s reflectivities giving less detailed information on the tilt/twist profile of the liquid crystal optic tensor.

In the present study a surface stabilized ferroelectric liquid crystal (SCE3) has been investigated using the half leaky guided mode (HLGM) technique at two wavelengths, 632·8 nm and 514·5 nm. A p to s conversion as large as 60 per cent has been recorded in the momentum window discussed above. Fitting angle dependent reflectivities to predictions from Fresnel multilayer optics theory yields, in extraordinary detail, the optic tensor configuration within the cell. A small tilt angle ($\sim 1°$–$2°$) is noted at room temperature, and together with a biaxiality ($\varepsilon_{xx} - \varepsilon_{yy}$) of $\sim 0{\cdot}003$–$0{\cdot}004$, a near surface region of $\sim 0{\cdot}3\ \mu$m thickness and a slightly bent chevron layer structure are all detailed. The use of the two wavelengths also shows a small amount of optical axis dispersion. To underline the reasons for the sensitivity of this technique, an analysis of the electromagnetic field distribution within the cell for various modes is presented. This shows clearly why certain resonances are sensitive to some parameters, while different ones are sensitive to others.

2. Experimental

The sample geometry used in the experimental study is illustrated in figure 1. An approximately 4 μm thick layer of liquid crystal is aligned between a high index glass pyramid and a low index glass substrate. As in previous studies [7, 8], the surface layers chosen to give homogeneous alignment in the nematic phase are thin (~ 20 nm) layers of 60° evaporated SiO. The coated pyramid and glass substrate are clamped together with 4 μm mylar spacers and the apparatus is placed in a temperature controlled oven

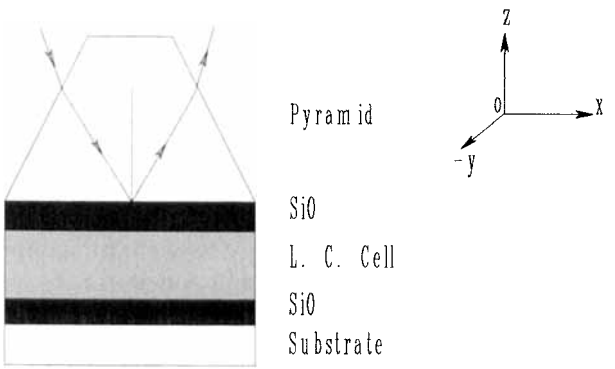


Figure 1. The sample geometry used in the experiment.

in which the sample temperature may be stabilized to $\pm 0{\cdot}02°C$. The empty cell is heated to 140°C and filled with SCE3 liquid crystal using capillary action. Once the cell is filled the temperature is quickly reduced to a point low in the N* phase ($\sim 110°C$). Subsequently the cell is cooled very slowly through the S_A to S_C^* phase transition ($\sim 72{\cdot}4°C$) in order that large domains form in the S_C^* phase. This need for slow cooling is helped by the thermal inertia of the glass pyramid and substrate. Typically the rate of change across the phase transition is 0·5°C per hour. Once a temperature a few degrees below this transition is reached ($\sim 70°C$) cooling is accelerated, the final temperature being set to 24·5°C.

In order to obtain the data in the required form of reflectivity as a function of angle of incidence, the complete cell plus oven assembly is positioned on a computer controlled rotatable table, which controls both the angular position of the detector and that of the sample. A parallel monochromatic light beam from a laser source, 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 incident angle. The incident beam is pure p-polarized, and a second polarizer is placed in front of the detector allowing a choice of detection of p or s polarization. In this article, only results for the orthogonal configuration, s-polarized output, are presented. Other data were recorded for p-polarized output, but these are not as sensitive to details of the director profile. In order to allow for any drift in source intensity, a small (~ 4 per cent) beam is taken by partial reflection before the sample to act as a reference.

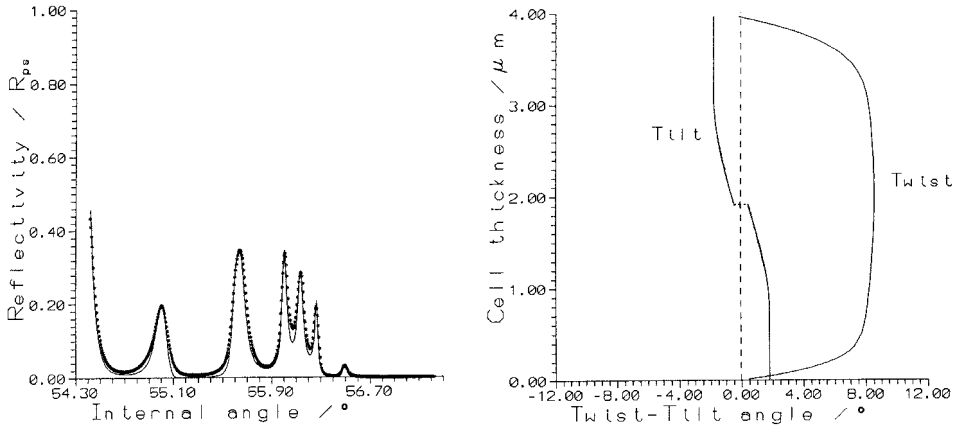
It is very important, in order to measure accurately any twist of the director relative to the initial alignment, that the initial alignment of the cell defines accurately the homogeneous direction as zero twist. In order to check this, the p to s conversion signal is monitored in the S_A phase. If there is a twist of greater than $\sim 0{\cdot}5°$ out of the alignment direction, this would show itself as a signal greater than that recorded. Thus we conclude that the pyramid and glass substrate alignment directions are the same to better than 0·5° and both directions are to the same precision in the plane of incidence of the radiation.

Finally in the experimental set up, it was considered useful to have an alternative check of our model for the tensor profile by fitting data at two wavelengths. For the accuracy required, two monochromatic parallel sources were needed. A He–Ne laser provides a beam at $\lambda_1 = 632{\cdot}8$ nm corresponding, at room temperature, to $\varepsilon_1 = 3{\cdot}2400$ for the pyramid and $\varepsilon_3 = 2{\cdot}1418$ for the substrate, while an argon-ion laser was used to give $\lambda_2 = 514{\cdot}5$ nm, corresponding to $\varepsilon_1 = 3{\cdot}3133$ and $\varepsilon_3 = 2{\cdot}1590$, again at room temperature.

3. Results and discussion

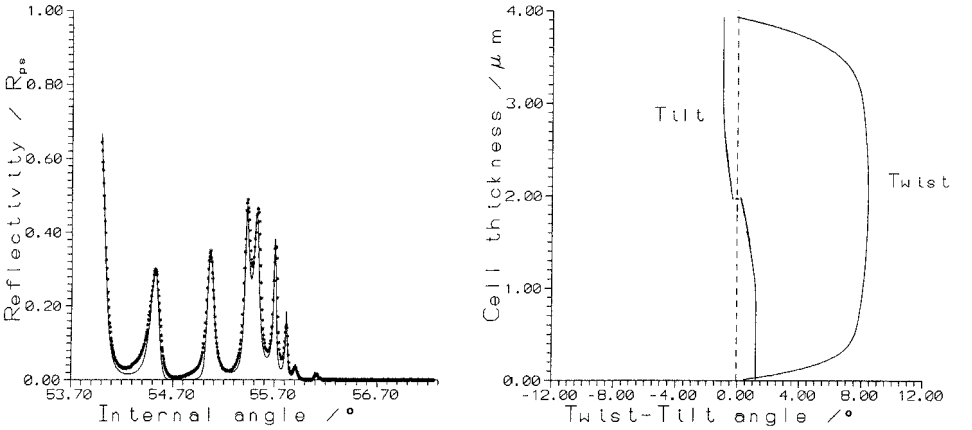
Experimental results (crosses) compared with theoretical predictions (full line) are shown in figures 2 (*a*) and 3 (*a*) for the wavelengths 632·8 nm and 514·5 nm, respectively. As mentioned above this is the p to s conversion data, R_{ps}, for the cell configured with the homogeneous alignment director in the plane of incidence.

A brief examination of the data shows a series of sharp and quite strong p to s conversion peaks in the internal angle window range which coincides with the theoretical analysis of the HLGM technique [10]. The optimum fits obtained were iteratively produced by very careful and progressive adjustment of all the optical tensor parameters characterizing the cell. This procedure forces the introduction of biaxility and gave the tensor profile across the cell illustrated in figures 2 (*b*) and 3 (*b*) for the two



(*a*) (*b*)

Figure 2. (*a*) The experimental data (crosses) and theoretically fitted results (solid line) for a wavelength of $\lambda = 632{\cdot}8$ nm. (*b*) Twist and tilt angle profiles in the cell. The fitted parameters of the geometry for (*a*) are: pyramid, $\varepsilon = 3{\cdot}2400$; SiO, $\varepsilon = 2{\cdot}4500 + i0{\cdot}0010$, thickness 20 nm; liquid crystal, $\varepsilon_{xx} = 2{\cdot}2430 + i0{\cdot}0003$, $\varepsilon_{yy} = 2{\cdot}2465 + i0{\cdot}0007$; $\varepsilon_{zz} = 2{\cdot}8959 + i0{\cdot}0006$, thickness 3·96 μm. Twist and tilt angle profiles as shown in (*b*): rotation angle 90°; substrate, $\varepsilon = 2{\cdot}1418$.



(*a*) (*b*)

Figure 3. (*a*) The experimental data (crosses) and theoretically fitted results (solid line) for a wavelength of $\lambda = 514{\cdot}5$ nm. (*b*) Twist and tilt angle profiles in the cell. The fitted parameters of the geometry for (*a*) are: pyramid, $\varepsilon = 3{\cdot}3133$; SiO, $\varepsilon = 2{\cdot}6500 + i0{\cdot}0010$, thickness 20 nm; liquid crystal, $\varepsilon_{xx} = 2{\cdot}2716 + i0{\cdot}0003$, $\varepsilon_{yy} = 2{\cdot}2754 + i0{\cdot}0006$; $\varepsilon_{zz} = 2{\cdot}9210 + i0{\cdot}0004$, thickness 3·96 μm. Twist and tilt angle profiles as shown in (*b*): rotation angle, 90°, substrate, $\varepsilon = 2{\cdot}1590$.

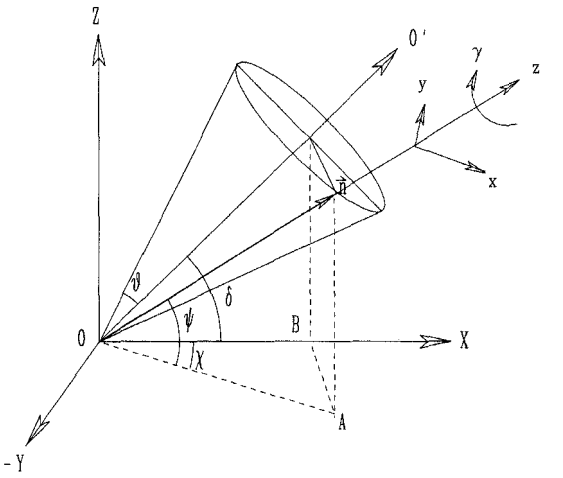


Figure 4. The geometry showing the relation of the cone angle, θ, the director twist angle, χ, tilt angle, ψ, and the layer tilt angle, δ. The rotation angle, γ, for the biaxial permittivity is also shown in the figure.

wavelengths. It is clearly a substantial advance from the earlier models of a uniaxial uniform slab twisted away from the surface alignment direction [7, 8]. The relation between the laboratory XYZ and the optic tensor coordinate system xyz with the definition of the twist angle (χ), tilt angle (ψ) and rotation angle (γ) of the tensor is illustrated in figure 4.

From figures 2 (*b*) and 3 (*b*) it is clear that there are thin boundary layers in the cell where the optic tensor twists gradually (exponentially) from the surface alignment direction out to the bulk twist angle. There is slight asymmetry of this surface region between the two boundary surfaces, although in both cases 90 per cent of the change occurs at $\leqslant 0{\cdot}4$ um. These results are basically in agreement with earlier studies [11] using the technique of evanescent field polarization conversion to probe the boundary layer. There, with a model of linear twist from the surface alignment out to the bulk twist a surface layer of $\sim 0{\cdot}2\,\mu$m was determined. Because the guided modes of the metal-clad liquid crystal waveguide are rather insensitive to these boundary layers and so were not probed by that technique [7, 8] the appearance of such a region in the present fitting is not surprising. There is also a further difference of the present study from the earlier work in the tensor profile throughout the bulk of the cell. We find that, whilst in the earlier work a uniform twisted slab provided a satisfactory fit to the data, this is here no longer true, and the twist angle, much more realistically, varies continuously throughout the cell. This together with the somewhat thicker boundary layer suggests that in the absence of the metal layers there is stronger surface anchoring.

Another feature of the present results is the presence of a surface tilt angle. Cognard [12] indicates that for a 60° obliquely evaporated SiO alignment layer there is almost no tilt at the surface. This is for a nematic director where the optic axis and director would coincide. In the present case, since there is biaxiality, the mechanical director and the major optic axis do not have to coincide. What we are saying here then is that the major tensor axis is tilted with respect to the surface by about 1 to 2°. This tilt extends well into the cell only being lost near the centre. Evidence for this tilt is very strong as we see by examining the right hand resonance peak in figure 3 (*a*). This particular peak is

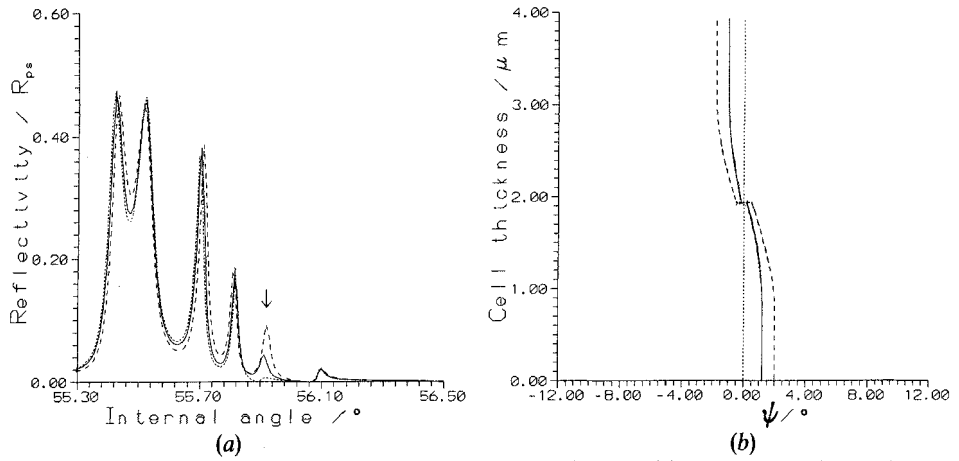


Figure 5. (*a*) The theoretical modelling curves for a wavelength of $\lambda = 514{\cdot}5$ nm. The modelling parameters are the same as shown in figure 3 (*a*) except for the tilt angle profiles which are now shown in (*b*). (*b*) The tilt angle profiles for the theoretical modelling in (*a*)

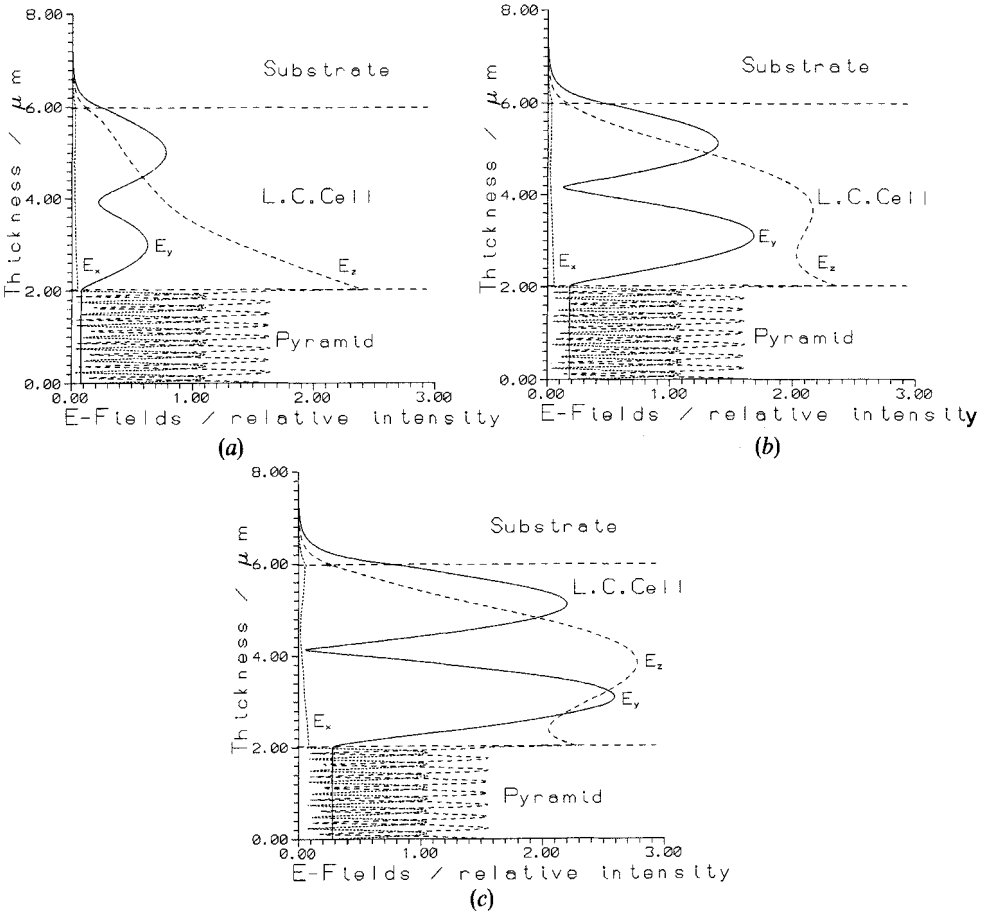


Figure 6. The field distributions in the cell for a p-polarized incident beam of the mode shown in figure 5 (*a*) by an arrow. (*a*) For a short dashed line in figure 5 (*a*), (*b*) for the solid line in figure 5 (*a*), (*c*) for the long dashed line in figure 5 (*a*).

very sensitive to the tilt as shown by figure 5 (*a*). In this figure three model curves are compared, using the same parameters as for figure 3 (*a*) except that for the short dashed line the tilt is everywhere 0° and for the long dashed line it is described in figure 5 (*b*) with a maximum of some 2°. Very clearly it is only the model of the solid line which gives the right hand peak, indicated by an arrow in figure 5 (*a*).

To illustrate more fully the reason for the sensitivity of this mode to tilt, we have examined the optical E field distributions, for the models considered, at the angle of excitation of this mode. It is clear from figure 6 that p to s conversion in the mode is strongly dependent on the tilt. As the tilt is increased so more p-polarized component (mainly E_z) is converted to s-polarized (E_y) within the cell and thus more s-polarized component is reradiated into the pyramid.

With the earlier metal-clad waveguide [7, 8], this p to s conversion signal is so weakened by the metal layer that this information is almost totally lost. Once more this shows the relative power of the new technique.

A test of the optic tensor profile model is of course to establish that it is largely independent of wavelength. Were the system uniaxial, it would have to be entirely independent of wavelength. However, it is slightly biaxial and this allows for spatial dispersion of the tensor axes [13]. Comparing the profiles of figures 2 (*b*) (632·8 nm) and 3 (*b*) (514·5 nm), we see only a minor difference associated with the tilt of the principle axis of the tensor. For a S_C^* liquid crystal, one of the axes has to be fixed, in the direction of the permanent dipole moment; this is not allowed to disperse with wavelength, but the other two may. Since the spatial dispersion is primarily tilt, then this suggests that the cone angle, in so far as it is a useful concept, also disperses with wavelength, which is as expected. The magnitude of spatial dispersion of $\sim 1°$ here recorded is well within acceptable bounds. Of course, as well as spatial dispersion, we must record biaxility, otherwise the local symmetry will be uniaxial which forbids spatial dispersion. As previously mentioned this is indeed needed to fit the data. At 632·8 nm, $\varepsilon_{xx}-\varepsilon_{yy}$ $=0{\cdot}0035+i0{\cdot}0004$, while at 514·5 nm, $\varepsilon_{xx}-\varepsilon_{yy}=0{\cdot}0038+i0{\cdot}0003$. The need for the introduction of biaxility and its magnitude is fully supported by detailed examination of the modelled reflectivity response. For example, the four peaks on the right end of figure 2 (*a*) are very sensitive to the biaxility of the material. In figures 7 (*a*) and (*b*) the

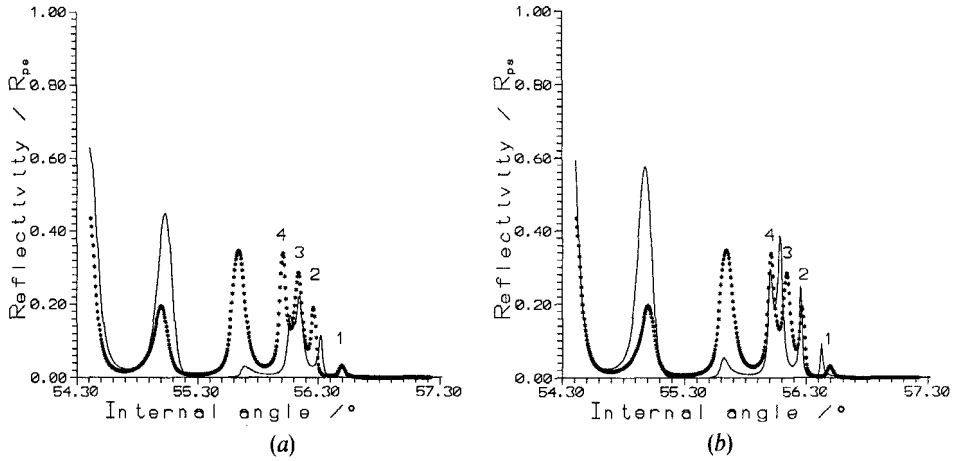


Figure 7. The experimental data (crosses) and theoretical modelling curves (solid line) for a wavelength of $\lambda = 632{\cdot}8$ nm. The theoretical modelling parameters are the same as shown in figure 2 (*a*), except for $\varepsilon_{xx}=2{\cdot}2465+i0{\cdot}0007$ in (*a*) and $\varepsilon_{yy}=2{\cdot}2430+i0{\cdot}0003$ in (*b*).

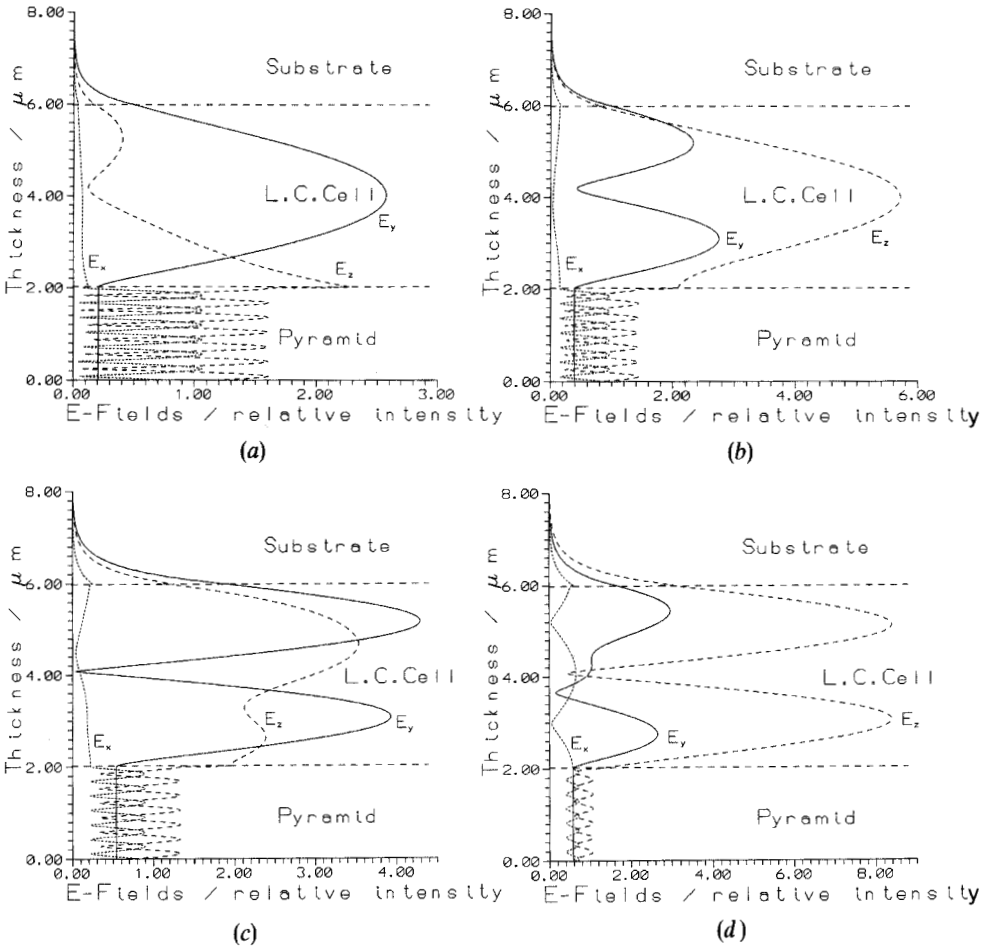
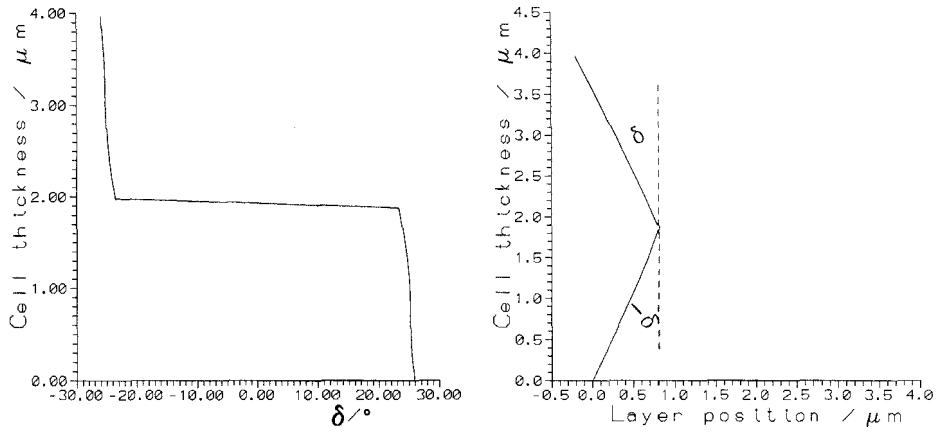


Figure 8. The field distributions in the cell for a p-polarized incident beam. (*a*) The mode 1 shown in figure 7. (*b*) The mode 2 shown in figure 7. (*c*) The mode 3 shown in figure 7. (*d*) The mode 4 shown in figure 7.

solid lines are model reflectivities produced from the same tensor profile as for figure 2(*a*), except that in figure 7(*a*), ε_{xx} is made equal to ε_{yy}, $\varepsilon_{yy}=2{\cdot}2465+i0{\cdot}0007$, and in figures 7(*b*) ε_{yy} is made equal to ε_{xx}, $\varepsilon_{xx}=2{\cdot}2430+i0{\cdot}0003$. It is obvious from these figures that of the four labelled resonances, peaks 2 and 4 clearly move to higher momentum when ε_{xx} is increased, while peaks 1 and 3 are little affected in position. Equally when ε_{yy} is decreased, peaks 2 and 4 are little affected, while peaks 1 and 3 move to lower momentum. In essence this tells us that peaks 1 and 3 are sensitive in position to ε_{yy} while peaks 2 and 4 are sensitive to ε_{xx}. This thereby allows a good determination of the biaxiality.

Once more to try to give a fuller physical picture of the modes, we examine the optical field distribution for these resonances. Using the parameters of the model giving the excellent fit to the data of figure 2(*a*), the calculated field profiles are as shown in figure 8 for the four resonances. Clearly peaks 1 and 3 are s-like with strong E_y peaks in the cell, while modes 2 and 4 are much more p-like, with very strong E_z in the cell. Hence



(*a*) (*b*)

Figure 9. (*a*) The layer tilt angle, δ, in the cell for a wavelength of $\lambda = 632{\cdot}8$ nm. (*b*) The chevron layer structure in the cell for the same director profile as (*a*).

they respectively give the strong response to ε_{yy} and ε_{xx}. Thus we have been able to determine a biaxility of 0·0035 which is on the limit of the sensitivity of the metal-clad waveguide technique [7, 8].

Finally it is instructive to try to obtain the layer tilt profile for this cell. From figure 4 the relation between the twist angle χ, tilt angle ψ, cone angle θ and layer tilt angle δ can be written

$$\sin\delta\sin\psi + \cos\delta\cos\psi\cos\chi = \cos\theta. \tag{1}$$

The problem then to find δ is to know the cone angle, which as mentioned disperses with wavelength. As yet we have been unable independently to determine this angle, but for SCE3 the quoted cone angle at room temperature is 24° [8]. Then if we use the chevron model (from X-ray scattering) we end up with a layer tilt angle δ at 632·8 nm as given in figure 9, this shows a slightly bent chevron with a maximum layer tilt of 25·8° and a minimum of 23·4°. The result of $\delta \approx \theta$ is in agreement with the results obtained in [14] in which an X-ray scattering technique has been used.

4. Conclusions

The first detailed half leaky guided mode investigation of the static optic tensor configuration in a surface stabilized ferroelectric liquid crystal layer is presented. For this investigation the ferroelectric liquid crystal layer is trapped between a high index pyramid and a low index glass plate—a configuration which allows the excitation of sharp p to s converted half leaky guided modes. From a systematic and very careful iteration of the tensor profile in the cell, progressively enabling theoretical model reflectivities to fit the experimental data, a full description of the tensor distribution is obtained. In the absence of a continuum theory for the S_C^* phase, this is the only procedure presently available, and it is with eager anticipation that we await a continuum model which may be used to evaluate elastic constants, etc. With no continuum theory, it is difficult to fully justify our model profile. To help in this, we have used two independent wavelengths, the resulting difference in the tensor profile being well within the possible range of spatial dispersion of the tensor axes. Furthermore,

examination of the optical electric field distributions within the cell gives acceptable physical reasons for the sensitivity of specific modes to certain parameters. The best fits finally obtained yield a biaxiality of order 0·003 to 0·004, a tilt of the major optic tensor axis of order 1 to 2°, surface regions of thickness 0·4 μm and a slightly bent chevron layer structure for SCE3. Presently, this work is continuing in order to study in detail voltage induced effects and rubbed polyimide alignment, as well as other interesting liquid crystal structures.

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