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Use of mode mixing to determine the optic tensor configuration of a thin ferroelectric liquid crystal layer

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It is possible to probe directly the optic tensor profile within a thin ferroelectric liquid crystal layer by the propagation of prism-coupled leaky guided modes. Plane parallel monochromatic radiation of single polarization is made incident through a prism into a planar ferroelectric liquid crystal layer. Monitoring the reflected signal as a function of incident angle results in a series of sharp dips at angles corresponding to the excitation of modes in the layer. For a ferroelectric layer in which there is in-plane or out-of-plane tilt, conversion of one linear polarization to the orthogonal polarization may occur, by detecting this conversion it has been possible to show the existence of thin boundary regions at the two surfaces of the thin ferroelectric liquid crystal layer.

1. Introduction

The discovery by X-ray scattering that the layers of a chiral smectic C (S_C^*) liquid crystal in a thin cell form a chevron structure [1–2] is of great importance in the understanding of ferroelectric liquid crystal devices, although it does not evaluate the director (and thereby the optic dielectric tensor) directly. High resolution X-ray diffraction studies of smectic layers in thin ($\leqslant 3\,\mu$m) S_C^* liquid crystal samples gives information about the layering and layer spacing but it is insensitive to the director configuration of the liquid crystal. It is necessary therefore to use alternative techniques to probe the optical dielectric tensor configuration in detail. Guided optic modes may be propagated along a thin layer of ferroelectric liquid crystal material [3–5] and the resonant momenta of these modes used to characterize this. Recent work has involved the use of the Kretschmann–Raether [6] geometry, using a thin metal film as the tunnel barrier, in the visible part of the spectrum. If the metal film is removed it is still possible to propagate prism-coupled leaky modes whose properties are sensitive to the ferroelectric liquid crystal index profile. In the geometry used here the metal layers are replaced with transparent conductive layers.

2. Experimental

If an air-filled cell is assembled (see figure 1) and optically probed with radiation at 632·8 nm with the plane of the electric field polarization in the *xz* plane (*p* polarized) then it is possible to excite leaky Fabry–Perot modes such that at particular angles of incidence the reflections between the front and the back interfaces of the bounding media interfere constructively. By varying the incident angle of the light at the glass/overlayer interface the component of photon momentum along the interface also changes. If at a particular incident angle this momentum matches that of a possible resonant mode within the gap the mode is excited and a dip in the reflected intensity occurs. A series of weak resonant modes may be observed extending down in angle from just below the glass/air critical angle. Both the sharpness and the envelope of these modes are sensitive to any overlayer present on the glass. By comparing such data of reflectivity versus incident angle (R versus Θ) with Fresnel's theory the overlayer parameters may be obtained. When the gap between the coated pyramids is subsequently filled with ferroelectric liquid crystal the resulting optic modes may then be observed and used to characterize the layer.

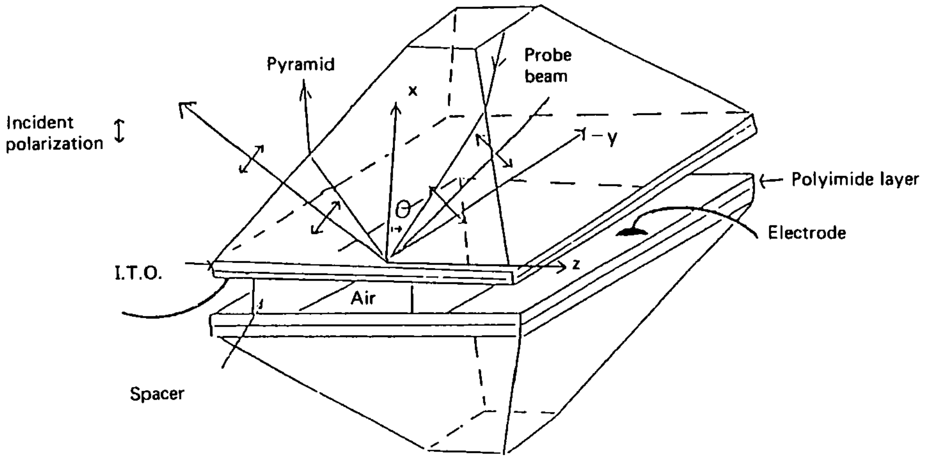

Figure 1. An air-filled cell showing the plane of incidence and electrode connections within the xyz frame of the cell.

If the liquid crystal material filling the cell is in the isotropic phase or the nematic or smectic A uniaxial phase with the optic axis either parallel or perpendicular to the direction of propagation, then modes will be of a simple form. For p polarized (TM) incident light, that is with the electric field parallel to the plane of propagation, the modes excited will be TM modes. Alternatively if the incident light is s polarized (TE), with the electric field perpendicular to the plane of propagation excited modes will be TE modes. With no tilting of the optic axis within the liquid crystal media out of the incident plane of the electric field vector there will be no sp mode mixing. However, if the liquid crystal optic axis is tilted out of the incident plane then this creates off-diagonal terms in the optic tensor in the xyz frame defined by the plane of incidence and the plane of the cell. These off-diagonal elements couple between p and s polarization fields and consequently modes may be excited of mixed polarization, TEM mixed modes, and the reflectivity data is significantly more complicated.

Thus, for example with incident s polarized light, the liquid crystal layer converts s to p polarized light within the cell and the light exits the system as partially p polarized light. For the systems studied here the conversion factor is of the order of 3–4 per cent for the plane of incidence parallel to or perpendicular to the original nematic alignment axis. Thus the s to p converted signal is more sensitive to in-plane tilting and out-of-plane tilting of the director configuration than are the single polarization modes. (The in-plane tilt is the tilt of the director away from the z axis in the yz plane (see figure 1) and the out-of-plane tilt is the tilt of the director away from the z axis in the xz plane.) If the sample is heated from just below the $S_C^* \rightarrow S_A$ phase transition into the S_A phase the sp mixed mode signal is abruptly extinguished.

Observation of TEM modes is hence an excellent tool for examination of the S_C^* phase.

It is helpful to consider the electric field profile of the modes across the cell to understand probing the refractive index profile. At an angle of incidence just below the critical angle for coupling between the glass and the material in the cell the first optic mode will be excited. This is termed the $m = 1$ mode and has one electric field maximum in the middle of the cell. As we decrease the incident angle the $m = 2, 3, \ldots, n$, etc., modes will be excited with correspondingly two, three, ..., n, etc., electric field maxima across the cell. If the material possesses a non-uniform refractive index profile then the different electric field profiles for different modes allows for detailed study of this index profile. For example, a region of low refractive index in the middle of the sample would cause the angles at which the odd order modes are excited to be displaced relative to the even modes simply because odd modes have field maxima in the middle of the cell while even order modes have a minimum.

3. Sample fabrication

Fabrication of high quality aligned thin film cells is vital to the experimentation. In the present geometry conventional glass plates are replaced by pyramids made from high refractive index glass (E06 soveril glass $n \approx 1{\cdot}80$ at 632·8 nm), this allows angle dependent reflectivity data to be taken in two orthogonal orientations. 40 nm of indium tin oxide (ITO) was deposited on the square face of the two pyramids by a sputter deposition method with a deposition rate of $\approx 10\,\mathrm{nm\,min^{-1}}$. This is characterized by comparing the angular dependent reflectivity (from inside the prism) with Fresnel's theory. Next polyimide (Nolimid 32) was spun on the substrate using a standard spinning technique, cured and then buffed unidirectionally in a conventional manner. The pyramids were then assembled with the rubbing direction parallel at the two surfaces and with the two faces displaced relatively by 4 mm to expose electrode regions, the cell was spaced with nominally 3·5 μm mylar and characterized once more using the reflectivity technique.

The cell was then enclosed in a temperature controlled oven and heated above the isotropic phase transition temperature before filling with the liquid crystal BDH (British Drug House) mixture MIX 783. This has the following sequence of phases and transition temperatures

$$\mathrm{I}\quad 116{\cdot}7\text{–}119{\cdot}6^\circ\mathrm{C}\quad \mathrm{N}\quad 92{\cdot}0^\circ\mathrm{C}\quad \mathrm{S_A}\quad 61{\cdot}2^\circ\mathrm{C}\quad \mathrm{S_C^*}.$$

Temperature stabilization takes about 1 hour. Filling was then accomplished by capillary action from the bottom of the cell to prevent air bubbles forming. The system was slowly cooled through the nematic and smectic A phases to the S_C^* phase at room temperature. The temperature was controlled to $\pm 0{\cdot}1^\circ$C but absolute values are quoted here to only $\pm 1^\circ$C. The so-called uniform director state [7], and not the twisted state, is realised by using the combinations of MIX 783 and Nolimid 32 with the thin cell geometry as the use of polyimide and a ferroelectric liquid crystal mixture may result in the twisted state which is undesirable.

4. Results

The first data presented here is for the liquid crystal in the S_A phase at 84·5°C, using p polarized light whose plane of incidence is perpendicular to the surface alignment direction (see figure 2, the crosses). The sharp resonant dips are due to the excitation of *p* polarized guided modes in the liquid crystal layer. No *sp* mixing is observed since the S_A phase is a uniaxial slab of dielectric material, which is aligned with the optic axis parallel to the alignment axis. Monitoring the reflected signal with incident *p* polarization and exiting *p* polarization as a function of incident angle gives R_{pp} reflectivity data. With incident *s* polarized light and outgoing *s* polarization we record R_{ss}. By comparing the experimentally measured reflectivity data with the theoretical reflectivity curves predicted from Fresnel's equations it is possible to obtain the real and imaginary components of the optical dielectric constants and the thickness of the layer.

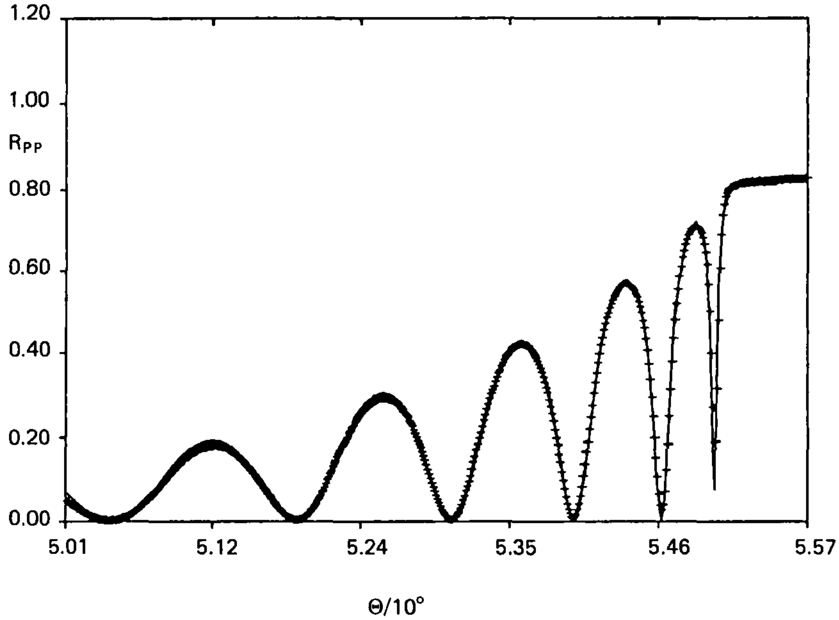

Figure 2. Data is presented (crosses) in the S_A phase at 84·5°C with incident *p* polarized light with the surface alignment direction perpendicular to the plane of incidence. Theory is given by the continuous solid line.

Data taken at a temperature just below the S_A to S_C^* phase transition with the optic axis in the plane of incidence, probing with incident *p* polarized light and monitoring the *s* polarized light exiting from the system, shows weak *sp* mode mixing in the ferroelectric liquid crystal layer with a sharp mode structure (see figure 3, continuous line). This shows that the axes of the optic tensor of the liquid crystal move out of the plane of incidence in some region of the layer, causing *sp* mixing. Decreasing the absolute temperature of the ferroelectric liquid crystal in the S_C^* phase increases the strength of this *sp* mixing, which saturates at room temperature.

5. Discussion

A modelling method which calculates theoretical reflectivity as a function of angle of incidence for a multi-layered biaxial medium was used and the theoretical reflectivity curves thus generated are compared to the corrected data. Corrections were made for the relevant polarization reflections at the air/glass interfaces and for the deviation of the probe beam on entering the pyramid, yielding the true reflectivity as a function of internal angle at the glass/liquid crystal interface. The modelling method considers the multi-layered system as a stack of uniform biaxial slabs of dielectric material, with each layer thickness $d \ll \lambda$ where λ is the wavelength of the incident light. In the present case the liquid crystal layer was modelled as 91 layers of equal thickness. An optical transfer matrix for each interface and layer was then calculated from which the reflections as a function of angle of incidence were predicited. Berreman and Scheffer [8], and Azzam and Bashara [9] developed this method; however exponentially growing fields together with *sp* mixing may cause numerical instabilities in the calculation of R_{pp}, R_{ss} etc. This problem was overcome by using the same multilayer system but in a scattering matrix formalism which couples the input and output radiation fields [10]. With this technique theoretical reflectivities were obtained for such a many layered system allowing a very good approximation to a continuously changing liquid crystal director configuration.

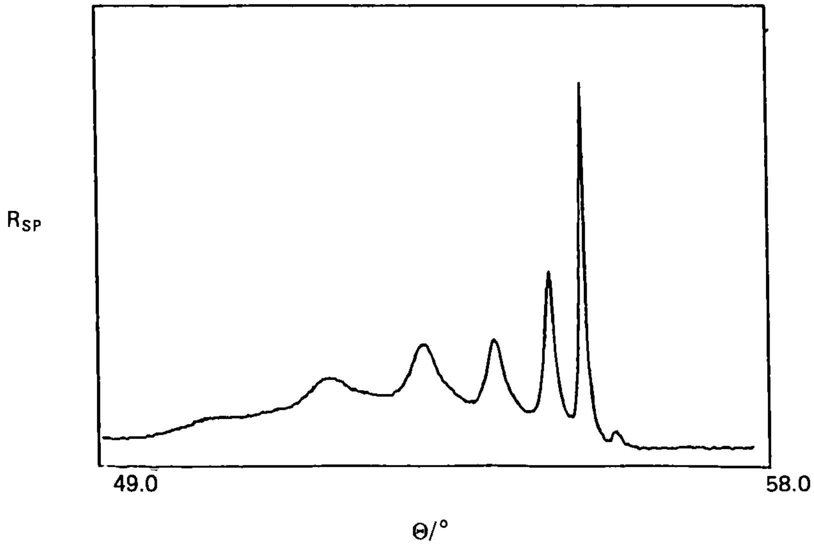

Figure 3. Weak *sp* mixing from a thin ferroelectric liquid crystal layer just below the ferroelectric phase transition, intensity in arbitrary units.

The N and S_A phases were modelled as uniaxial slabs. In the nematic phase the director aligns parallel to the rubbing direction. Additional layer ordering occurs in the S_A phase for which the data may still be fitted with a simple uniaxial slab model with the optic axis along the alignment direction (see figure 2, theory is given by the continuous line). Experimentally no *sp* mixing was observed in the S_A phase. If the director were to tilt significantly in the *xy* plane or if there were significant surface tilt then the mode structure would be altered and *sp* mixing should occur, inconsistent with the experimental data.

As demonstrated earlier [5], the simplest good fit thus far to experimental data in the form of R_{pp} and R_{ss} for the S_C^* phase is the chevron model with a 0·20 μm boundary layer at each surface. The simple chevron model which is optically equivalent to a uniformly in-plane tilted slab gives, without a boundary layer, theoretical reflectivities which did not match the experimental data. In the previous study we introduced a boundary layer in which the liquid crystal optic axis increases linearly in the in-plane tilt from zero at the surface to the maximum in-plane tilt through the bulk of the

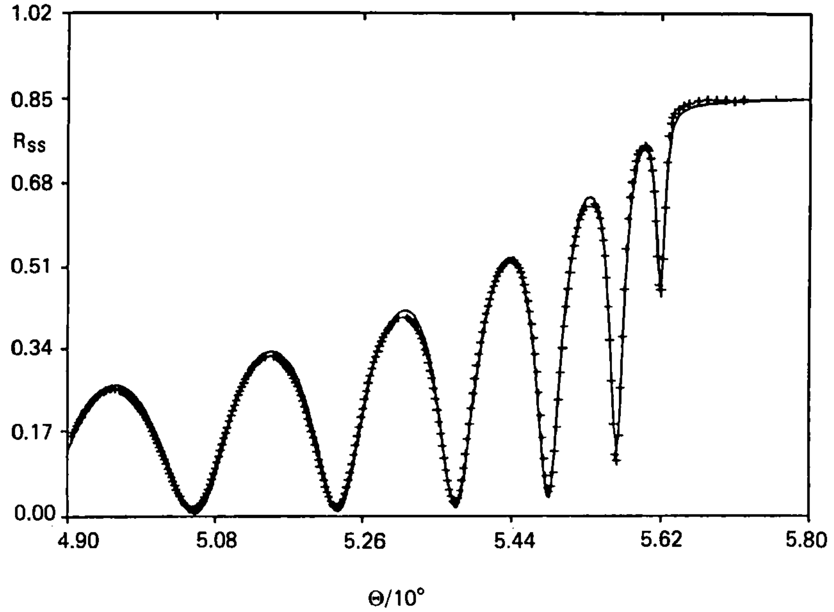

Figure 4. Data fitted with a chevron model with boundary layer regions with in-plane tilt.

sample. This model is consistent with the surface anchoring constraining the director to be parallel to the alignment direction at the bounding interfaces. The high quality of the fit of theoretical reflectivity against incident angle for the room temperature data for R_{ss} modes, where the alignment axis is parallel to the direction of propagation, shows this model matches the data well (see figure 4, data crosses).

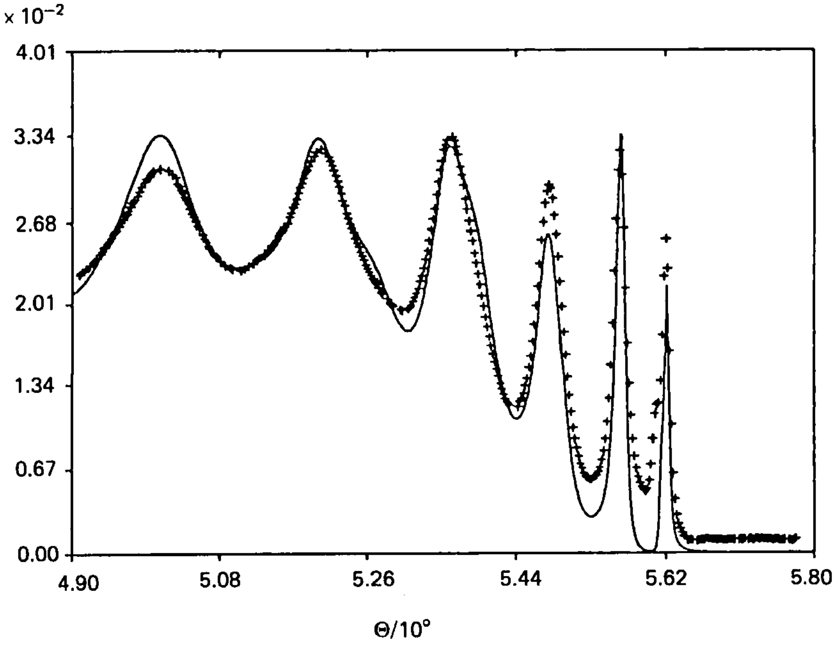

Figure 5. *sp* mixing from a thin ferroelectric liquid crystal layer at room temperature modelled with an initial 3° pretilt at both surfaces.

However in the present study we have now found that *sp* mixing proves to be more sensitive to the optical director configuration than the single polarization data. Not surprisingly we find once more that a uniformly twisted uniaxial slab (pure chevron model) does not fit the *sp* mixing data precisely. Again a boundary layer of 0·20 μm has been introduced to fit the data, this boundary layer being consistent with the single polarized mode data and previous modelling. However it was also necessary to include a tilt profile that started with a surface pretilt of 3° rapidly falling to zero within the boundary layer to a planar structure in the middle of the cell. A mirror plane in the middle of the sample perpendicular to the incident plane reflects the tilt profile to give 3° pretilt at the opposing surface. The presence of this small surface pretilt of 3° at the surfaces is not detectable in conventional R_{ss} and R_{pp} measurements. This model was the best fit to the room temperature R_{ps} data (see figure 5). Without the presence of a surface pretilt of 3° at the two surfaces the comparison of the theoretically predicted reflectivity against angle with the experimental R_{ps} data shows substantial discrepancies (see figure 6). A diagram of a satisfactory in-plane tilt/out-of-plane tilt profile is presented for the 3° surface pretilt (see figure 7). With this extended model for the ferroelectric liquid crystal layer at room temperature established, further experiments were performed at several different temperatures to give the results shown in the table. The in-plane tilt profile (TILT profile, the table) of the liquid crystal has a sharp cut off at the Curie temperature of the material and has a temperature dependence of the form,

$$\Theta = \alpha(T_C - T)^\beta,$$

where T_C is the transition temperature, $\alpha = 1\cdot47^\circ$ and $\beta = 0\cdot46 \pm 0\cdot01$. It is not possible to determine the temperature dependence of the surface tilt above the

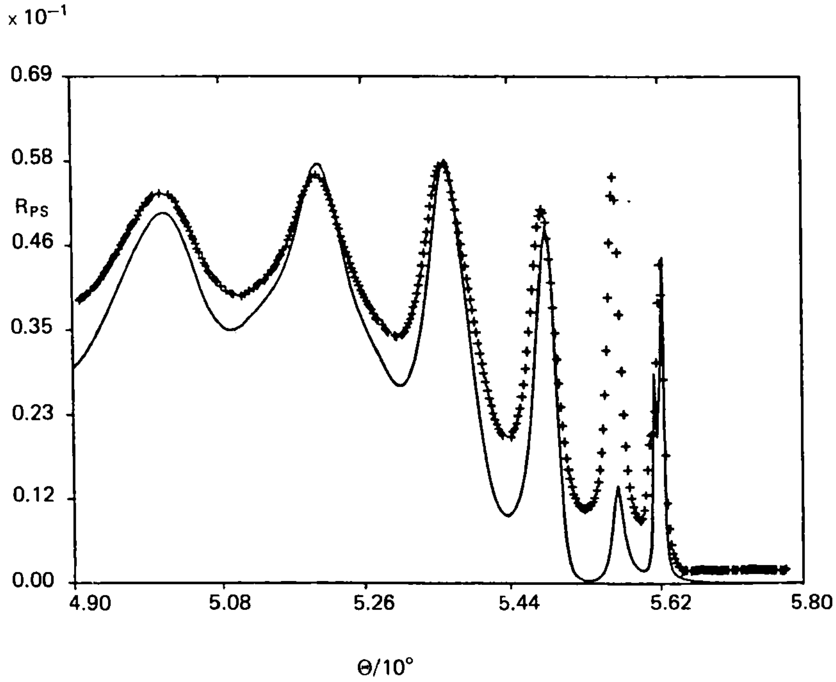

Figure 6. *sp* mixing from a thin ferroelectric liquid crystal layer at room temperature modelled without a pretilt at either surface.

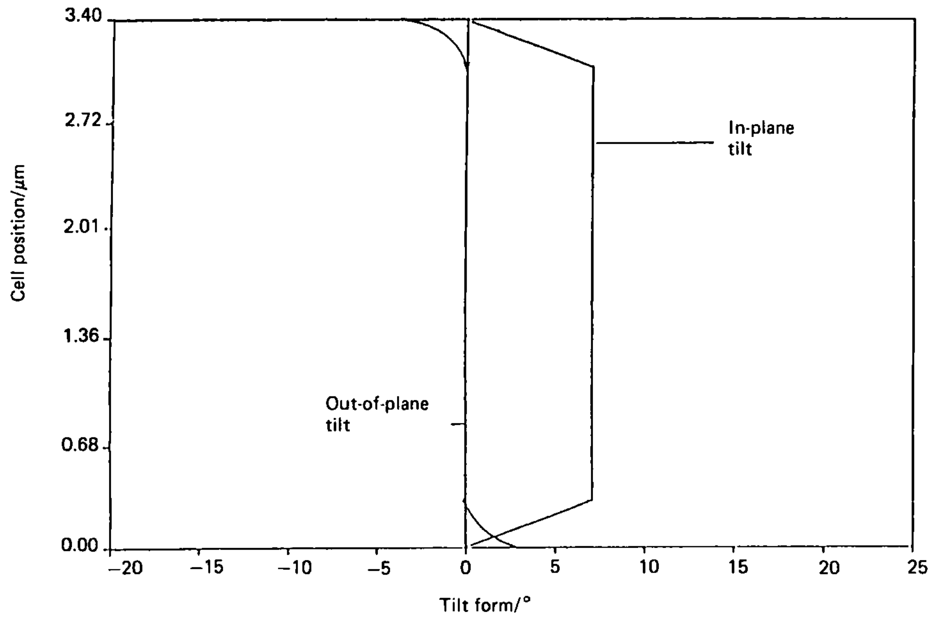

Figure 7. A diagram of the zero voltage model for in-plane/out-of-plane tilt profile in the sample.

Table of optical dielectric constants and in-plane tilt for a series of temperatures for the liquid crystal mixture MIX 783.

Temperature °C error ±1·0°C	Dielectric parameters $\varepsilon_{\parallel} = n_e^2$ ±0·001	$\varepsilon_{\perp} = n_o^2$ ±0·001	In-plane tilt angle/° error ±0·1°
101	2·259	2·189	0·0
85	2·672	2·180	0·0
61	2·710	2·199	0·0
60	2·725	2·200	1·0
59	2·726	2·201	2·0
52	2·730	2·203	4·0
49	2·744	2·205	5·4
38	2·767	2·214	6·8
19	2·804	2·241	7·5

$S_A \rightarrow S_C^*$ phase transition as we do not obtain *sp* mixing from a uniaxial slab with the optic axis perpendicular to the plane of incidence and just tilting of the optic axis in the *xy* plane. Only *sp* mixing is sufficiently sensitive to detect the presence of a surface pretilt in the system. The surface pretilt is constant through the entire S_C^* phase, the angle of which is dictated by the particular cell geometry and the surface anchoring at the liquid crystal/polyimide interface.

7. Conclusions

As the temperature of the ferroelectric liquid crystal cell is reduced the liquid crystal director reconfigures, below the $S_A \rightarrow S_C^*$ transition the change in the density wave layer thickness causes a tilt in the optic dielectric tensor and a chevron structure in the layering forms. It was found by detailed fitting that the ferroelectric liquid crystal BDH mixture MIX 783 may be modelled as a uniaxial bent chevron system, with an in-plane tilt angle less than the cone angle of the material, and with a thin surface boundary layer where it is no longer possible to maintain the same layer tilt as in the bulk of the material. Matching the theoretical reflectivity curves to the singly polarized experimental data, R_{ss} and R_{pp}, gives the magnitude of the bulk in-plane tilt and the dimensions over which it is necessary to include a surface boundary region of uniformly increasing in-plane tilt within the S_C^* temperature range.

From analysis of R_{ps} data in the S_C^* phase a thin surface boundary layer is confirmed and the surface pretilt and consequently out-of-plane tilt profile has been investigated. Optical mode mixing recorded for the S_C^* phase configured so that when heated into the S_A phase the optic axis is parallel to the alignment direction gives a quantitative measure of the surface boundary tilt of $3 \pm 0{\cdot}5°$.

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