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A preliminary investigation of the anomalous out-of-plane tilt alignment in an orthogonal-twist ferroelectric liquid crystal cell using the prism-coupling technique

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Recent prism-coupling results are presented which clearly demonstrate that in ferroelectric liquid crystal cells, prepared with the alignment directions of the two surfaces orthogonal, with sputtered indium tin oxide conductive layers and spun polyimide alignment layers, there is no out-of-plane tilt of the liquid crystal optic axis. Unlike most techniques which give an integrated optical response throughout a cell, the prism-coupling technique allows a separate analysis of in-plane and out-of-plane tilt. Conventional optical polarized microscopy yields good lateral resolution but poor section resolution. For a uniform sample we can obtain information on the section (across the cell) resolution. We report that an optical dielectric tensor configuration is formed in which the major optic axis lies in the plane of the surface across the cell, but that as expected, in an orthogonal-twist cell, there is no axis of preferred alignment within this surface plane.

The prism-coupling technique has been widely used to study different classes of liquid crystal [1-3]. Extensive research into the ferroelectric phase has revealed the proposed chevron in the smectic layering [4], verified by X-ray scattering experiments [5-6]. Information about the director and layer elastic constants of thin aligned ferroelectric liquid crystal cells is necessary for the development of a ferroelectric continuum mechanics model and for the next generation of electro-optic devices. It was decided to investigate the reflectivity obtained from the propagation of optical guided modes in a liquid crystal layer in a parallel aligned cell where the alignment preparation permitted the formation of the chevron layer structure and in an orthogonal-twist cell where the layer formation was prohibited.

Outline of prism-coupling technique: pyramids are used to permit optical probing of the sample in two orthogonal orientations. First the bare prism substrates are cleaned by heating them for 1 hour in boiling 1,1,1-trichloroethane and then for a further hour in isopropyl alcohol. The pyramids are then baked for 1 hour at 120°C. Indium tin oxide (ITO) was then sputtered on to the prisms and a layer of 40 nm deposited. A polymer alignment layer of approximately 50 nm was spun onto the substrates using a solution of the monomer (Nolimid 32), soft baked at 80°C for 10 min, buffed unidirectionally half a dozen times using a rubbing machine and then hard-baked at

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200°C for 1 hour. Cells were then assembled such that, in one case, the direction of the buffing of the two alignment layers was in the same direction (parallel aligned cell), and in a further cell the two alignment directions were orthogonal (orthogonal-twist configuration).

The cells were spaced with nominally 3.5 μm thick clear mylar, and placed in a temperature controlled oven (accurate to $\pm 0.1^\circ\text{C}$). These empty cells were then filled with liquid crystal in the isotropic phase by capillary action and the temperature reduced continuously into the ferroelectric S_C^* phase. SCE 8 with a cone angle of 19.5° at 20°C was used to fill both cells.

For an accurate optical determination of the important material parameters the refractive indices and the thickness of the cell—data acquisition is considerably simplified if only two orthogonal polarizations of incident radiation, transverse magnetic (TM or p-polarized) and transverse electric (TE or s-polarized) radiation, are

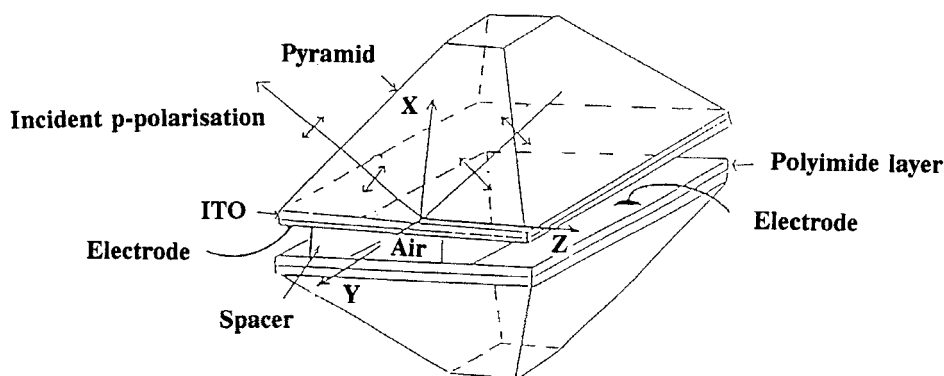


Figure 1. The assembled cell.

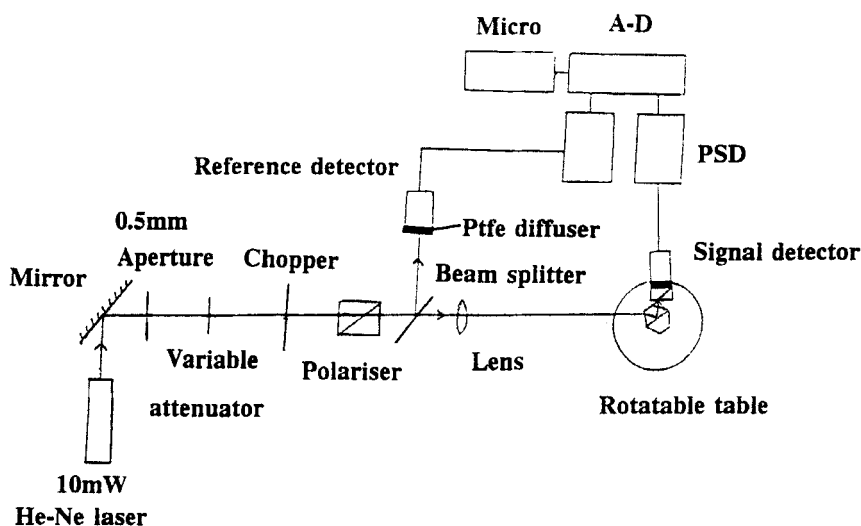


Figure 2. Experimental angular spectrometer system.

used. However, data become much more complicated if the layer to be examined is not uniaxial ($\epsilon_{xx} \neq \epsilon_{yy}$). Two possible cell orientations may arbitrarily be defined, where the major optic axis lies in the plane of incidence along the z axis in the xz plane (horizontal orientation), or perpendicular to the xz plane along the y axis (vertical orientation), see figure 1. The assembled cell, see figure 1, is placed on a rotating spectrometer table and light from a helium–neon laser at 632.8 nm is frequency chopped for phase sensitive detection of both a reference beam (to account for fluctuations in laser beam intensity) and also to monitor a signal from the sample to be studied (see figure 2).

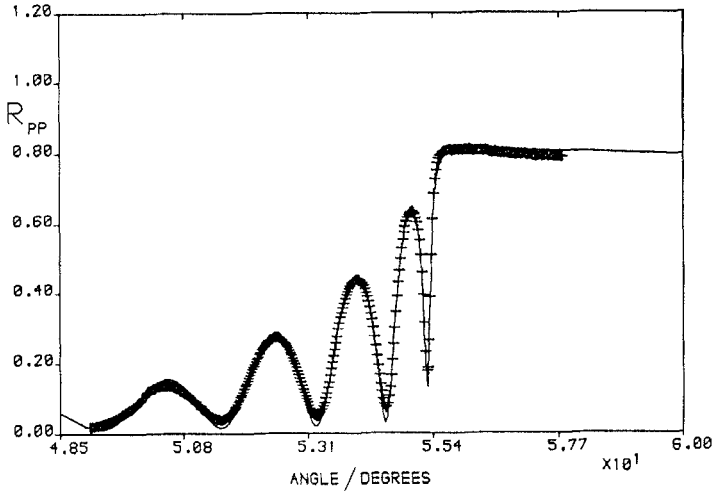
Reflectivity data may be taken in the S_A phase at 78°C at an incident wavelength of 632.8 nm from the parallel aligned SCE 8 cell, with TE (s-polarized) light incoming and with TE polarized light exiting the cell. The experimental data appear as a series of minima corresponding to the solution of the leaky Fabry–Perot optical guided modes below the critical angle θ_C , for total internal reflection at the prism/liquid crystal interface (see figure 3(a)). The optical guided modes of the reflectivity curve exhibit no structural features and agree well with theory. The data are represented by crosses and the theory by a continuous solid line. TM polarized reflectivity spectra may also be recorded in a similar manner to the TE data. If the temperature of the parallel aligned system is now lowered well into the S_C^* phase ($\sim 39^\circ\text{C}$), probing with a wavelength of 632.8 nm, TM-polarized spectra may be obtained (figure 3(b)).

Data are also obtained, again at room temperature for the S_C^* phase, from an orthogonal-twist cell of the FLC SCE 8. TE or s-polarized modes (figure 4(a)) and TM or p-polarized modes (figure 4(b)) of the system are examined with the incident alignment interface of the cell in the horizontal orientation.

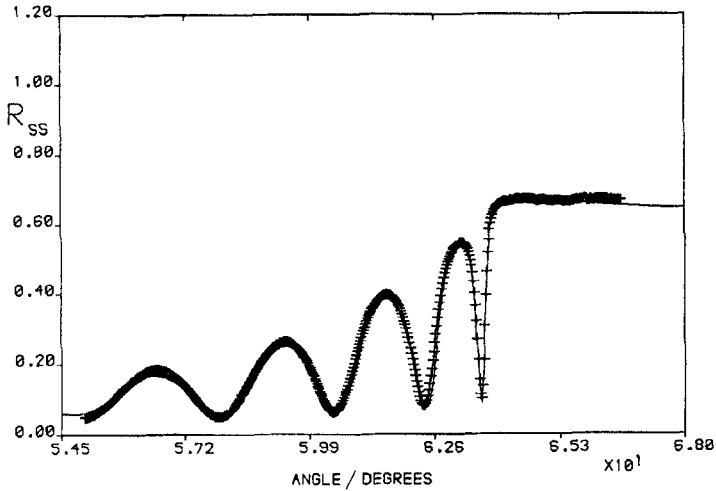
Theoretical analysis: in the isotropic phase only one reflectivity spectrum is necessary to allow the determination of ϵ_r , ϵ_i and d , where ϵ_r is the real part of the dielectric permittivity, ϵ_i the imaginary part and d the thickness of the cell. To model the experimental data of a tilted smectic correctly requires both TE and TM polarized data in both orthogonal orientations. Experimental reflectivity data are compared to theoretically predicted reflectivity spectra using a multi-layer formalism of Fresnel's equations, and the values of ϵ_r and ϵ_i which best compare to the experimental data may be related to n and k by the expression $\epsilon = \epsilon_r + i\epsilon_i = (n + ik)^2$.

The modelling method used in this paper calculates theoretical reflectivities as a function of angle of incidence for a multi-layered biaxial medium. The multi-layered stack is broken into a series of uniform biaxial slabs of dielectric material, where each layer thickness is significantly less than the wavelength of the incident light. The liquid crystal layer is consequently modelled by dividing it into as many as 91 layers of equal thickness; this allows good approximation to a continuum model of the ferroelectric liquid crystal optical director configuration. A scattering matrix formalism is used to overcome instabilities in the more conventional transfer matrix approach followed initially by Berreman [7] and then by Azzam and Bashara [8]. The scattering matrix modelling method developed by Ko and Sambles [9] couples the incoming fields at any given interface to the outgoing fields at that interface by a matrix which is implicitly stable for all incident angles at all wavelengths.

The theoretical model used to achieve a good fit to the experimental data in the S_A phase (figure 3(a)) was equivalent to a uniaxial slab of dielectric with the major optic axis along the z axis. Similarly, the theoretical model used successfully to fit the chevron cell data in the S_C^* phase shown in figure 3(b) was equivalent to a uniaxial slab of dielectric with the optic axis now uniformly twisted out from the z axis in the yz plane by a constant in-plane tilt χ , over the majority of the cell, with no out-of-plane tilt (tilt



(a)



(b)

Figure 3. (a) Comparison of experimental data (crosses) with a theoretically predicted reflectivity curve (continuous line) for incident TE polarized light taken from a $3.5\text{ }\mu\text{m}$ parallel aligned SCE 8 FLC cell in the S_A phase at 78°C . (b) Comparison of experimental data (crosses) with a theoretically predicted reflectivity curve (continuous line) for incident TM polarized light taken from a $3.5\text{ }\mu\text{m}$ parallel aligned SCE 8 FLC cell in the S_C^* phase at 39°C .

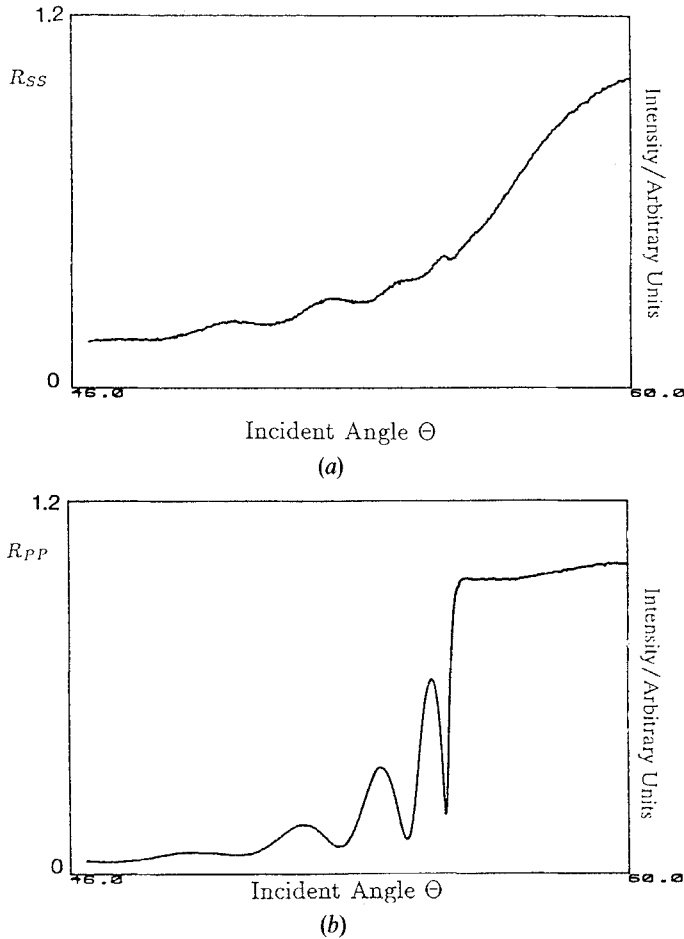


Figure 4. (a) Experimental reflectivity curve for incident TE polarized light taken from a $3.5 \mu\text{m}$ orthogonal-twist SCE 8 FLC cell in the S_C^* phase at 20°C . (b) Experimental reflectivity curve for incident TM polarized light taken from a $3.5 \mu\text{m}$ orthogonal-twist SCE 8 FLC cell in the S_C^* phase at 20°C .

away from the z axis in the xz plane) occurring in the bulk. It has been shown that the in-plane tilt (yz plane tilt) and out-of-plane tilt of the surface polyimide/liquid crystal 'boundary layer' region is non-uniform [3]; in fact it is necessary for some non-uniformity to exist as perfect extinction is not observed between crossed polarizers [10]. Also at no time have any experiments shown any significant biaxiality in the optical dielectric tensor. This model is optically equivalent to the chevron layer model proposed by Pelzl [4] and verified by Clark and Rieker [4]. The chevron model was confirmed optically using the prism-coupling technique soon after the X-ray scattering measurements were performed [2].

The good quality of the parallel aligned SCE 8 FLC cell is verified by the presence of sharp dips in the reflectivity spectra at specific angles of incidence, with a smoothly decaying envelope below critical angle. Leaky guided modes are propagated in the liquid crystal at these angles of minimum reflectivity.

For a parallel aligned cell of SCE 8, the chevron model requires a value of in-plane tilt from fitting the data with $\chi = 8.5^\circ$ in the middle of the cell. In the chevron case the required in-plane tilt of the layering is given by de Vos [11] as

$$\chi = \tan^{-1} \left[\frac{\sin \theta \cos \phi}{\cos \delta \cos \theta + \sin \delta \sin \theta \sin \phi} \right], \quad (1)$$

where θ is the cone angle of SCE 8 in our case, ϕ is the azimuthal angle around the smectic cone on which the permanent dipole sits and δ is the layer tilt.

Out-of-plane tilt, τ , is given by

$$\tau = \tan^{-1} [\cos \delta \sin \theta \sin \phi - \sin \delta \cos \theta]. \quad (2)$$

However, at the chevron interface for continuity of the director configuration $\tau = 0$

$$\cos \chi = (\cos \theta / \cos \delta). \quad (3)$$

Clearly these equations reduce to the more simplified form in the case of bookshelf model layering with $\delta = 0$.

Previous prism-coupled S_C^* data indicated that as we enter the S_C^* phase from the smectic A phase it is energetically favourable for a uniformly in-plane tilted optical dielectric tensor configuration, with no out-of-plane tilt, to form in almost all experimental situations. From this we conclude that the energy involved with the formation of uniform in-plane tilted chevron smectic layers is much less than that associated with significant out-of-plane tilt of the molecular axis. However, small out-of-plane tilt of the major optic axis ($2-3^\circ$ tilt) is permitted at the surface interfacial regions in conventionally fabricated parallel aligned MIX 783 cells [12]. This non-planar in-plane/out-of-plane tilt at the interfaces has been verified by Lavers and Sambles [3] and gives rise to a boundary region, the so called boundary layer profile (BLP). The presence of a boundary layer profile has been confirmed by Elston [13] for the FLC mixtures SCE 3 and SCE 12. Anderson [14] has found that a triangular director profile (TDP) fits data well with low tilt polyimide alignment layers and that a reverse TDP gives excellent agreement for high tilt silicon monoxide alignment [15]. However, with the technique of Anderson, it is found that both the BLP and TDP give comparably good fits to the smectic layer structure.

For the orthogonal-twist cell, the poor quality (depth and sharpness) of the guided mode structure observed from an orthogonal-twist cell of SCE 8 for the TE or s-polarized modes in the horizontal orientation, (see figure 4(a)) and the complete absence of TE modes in the vertical orientation give a measure of the disorder of the optic axis in the plane of the surface throughout the cell, the lack of in-plane tilt uniformity. The laser beam was not probing the wall region between domains, as adjusting the focal spot of the beam did not reassert a good guided mode structure. There is no uniquely defined optic axis in the plane of the cell surface, and certainly no uniform orthogonal-twist of in-plane tilt between the two cell surfaces as would be supported in a simple twisted nematic cell [16]. However, the remarkable good quality guided mode structure for the TM modes (figure 4(b)) obtained at room temperature in both orientations indicate that the out-of-plane tilt is strongly constrained.

Our modelling of the chiral smectic C^* phase in parallel aligned cells as a uniaxial multi-layered optical medium with only in-plane tilt is a simple approach which models the experimental system well. Clearly in the orthogonal-twist structure, no such simple model will suffice. Quite how the optical director tensor configuration forms between the two orthogonal alignment surfaces given that only an in-planar tilt arrangement is permitted is a subject of debate.

Such an orthogonal-twist system is extremely difficult to predict theoretically, as this would really require a continuum mechanics model, and as none exists even for the much studied parallel aligned chiral smectic C* phase, modelling this experimental system to obtain any meaningful results about the elastic constants (splay, twist and bend) is not currently possible. Hopefully, if a full continuum mechanics model can be developed, it would be possible to model the data in some detail. It may be that the construction of cells with a deliberate orthogonality between the alignment surfaces, exhibiting a surprising degree of alignment in only one plane may be exploited in future ferroelectric liquid crystal devices.

In conclusion, in an orthogonal-twist cell, prepared with the two surface alignment directions orthogonal, it is shown that an in-plane tilt distribution of the major optic axis is adopted. However, within the xz plane, the molecular axis is not ordered. Thus it is possible to fabricate ferroelectric liquid crystal cells with the major optic axis to be planar across the cell, but with a 90° angle existing between the orientation of the two alignment surfaces. This planarity of the optical dielectric tensor would not otherwise have been detected by conventional optical polarized microscopy, as the out-of-plane disorder of the FLC containing system and the in-plane tilt disorder cannot be deconvolved separately, and hence disorder of in-plane tilt would have masked the out-of-plane tilt uniformity.

Conventional optical polarized microscopy through this double prism cell has shown a typical domain structure, but imaging a large area of the substrate through approximately 2 in. of glass proved extremely difficult. Further study of the S_A and nematic phases of such orthogonal-twist cells is necessary, combined with conventional polarized microscopy of a new cell design which replaces the second high refractive index prism with a thin high refractive index backplate. This new design should make viewing of the domain structure over a large area much easier. It is hoped that with increased understanding of the smectic C* phase by prism-coupling in both cells which are conventionally parallel aligned and also orthogonal-twist configured, a further generation of superior electro-optic devices may be fabricated.

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