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Experimental investigation of the compressible continuum theory of a homeotropically aligned ferroelectric liquid crystal

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The optical tensor configuration in a homeotropically aligned ferroelectric liquid crystal (FLC), SCE13 cell, is investigated by means of optical excitation of half leaky guided modes. A thin slab of FLC is confined between a high index pyramid and a low index substrate whose indices bound those of the liquid crystal. In this geometry there exists a small angle range over which a series of sharp resonant modes may propagate in the liquid crystal. Detecting the angular dependent reflectivity for plane polarized radiation and subsequently fitting the data by iteratively modelling from multilayer Fresnel theory, a full characterization of the tilt and twist profile throughout the cell is achieved. The temperature dependence of the tilt of the principal director, which is related to the smectic cone angle, and of the optical permittivity, as well as the pitch have been obtained. The tilt director profile across the cell is interpreted using a compressible continuum theory for SmC* liquid crystals which includes the possibility of variable cone angle and layer spacing.

1. Introduction

The aim of this paper is to present the optical tensor profile characterization of a homeotropically aligned liquid crystal (LC) cell using optical excitation of half leaky guided modes [1]. A thin ferroelectric liquid crystal layer is sandwiched between a pyramid having a refractive index higher than the largest of the LC and a low refractive index substrate having index lower than the lowest of the LC. In this geometry, there exists an angular window, from the pseudo-critical angle between the high index pyramid and the effective index of the liquid crystal to the critical angle between the high index pyramid and the low index substrate, over which sharp half leaky resonant guided modes may propagate. For linearly polarized incident radiation there will be strong *s* (TM) to *p* (TE) conversion if the molecular director is twisted out of the plane of incidence. Fitting of multilayer optics theory [2, 3] to the observed angle-dependent reflectivity data gives a complete evaluation of the optical structure [4].

A particular issue which we address here concerns the variation of cone angle with smectic interlayer spacing, i.e. a change in molecular tilt with respect to the layer normal, from the surface to the centre of the sample. In a previous study [5] a simple linear tilt profile was chosen as an acceptable approximation to treat the near surface regions. A more elaborate non-linear variation in this surface regime is supplied by a theory which takes into account the layer compression as molecules move away from the layer normal. The model proposed by McKay and Leslie [6], which adopts a simple free energy for a ferroelectric liquid crystal similar to Nakagawa [7], suggests that the tilt angle profile in homeotropically aligned cells is determined by a term associated with smectic layer compression. This model is the extension of the continuum theory for smectic C liquid crystals for constant layer spacing [8] introducing chirality and variable layer thickness.

2. Experimental

The sample geometry (figure 1) includes two glass plates with indices $n_{\mathrm{High}}=1{\cdot}7305$ and $n_{\mathrm{Low}}=1{\cdot}4630$ at 632·8 nm wavelength, both coated with an approximately 610 Å thick layer of indium/tin oxide. Homeotropic alignment is induced by gently wiping the glass surfaces in one direction with a lens tissue soaked in lecithin dissolved in diethyl ether; excess of lecithin is then

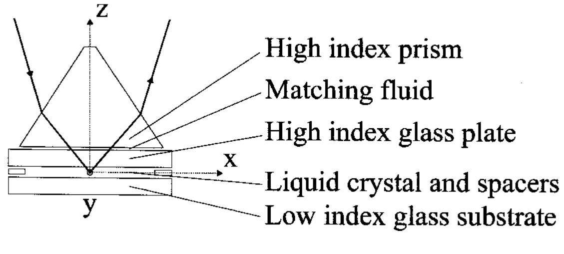

Figure 1. The sample geometry used in the experiment.

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removed by washing in solvent only. The plates are assembled in a clean room using UV hardening glue containing 3 μm diameter spacer beads, and the cell is filled by capillarity with ferroelectric liquid crystal MERK SCE13 in the isotropic phase. Once full, the cell temperature is reduced fairly quickly through the N* phase into the SmA phase; subsequently it is cooled very slowly through the SmA to SmC* phase transition (less than 0·5°C per hour) in order to obtain a large smectic monodomain. Once a temperature a few degrees below

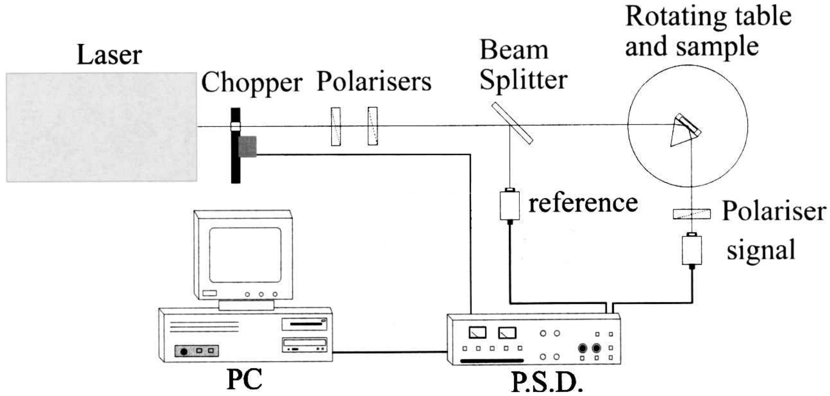

Figure 2. The experimental arrangement.

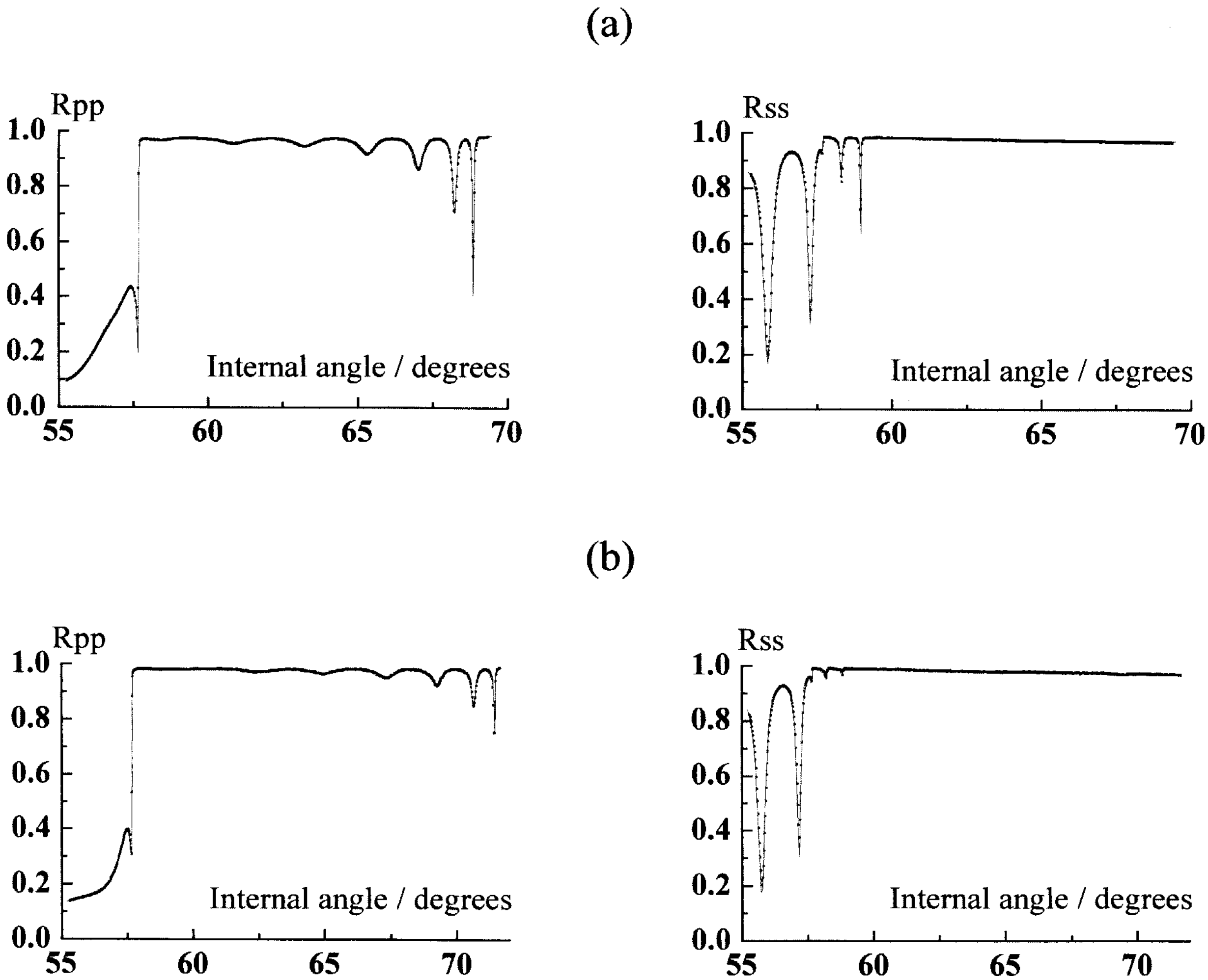

Figure 3. The recorded reflectivity scans (crosses) showing R_{pp} and R_{ss} versus the angle of incidence compared with theory (solid lines): (*a*) nematic phase, $T=90$°C; (*b*) S_A phase, $T=65$°C.

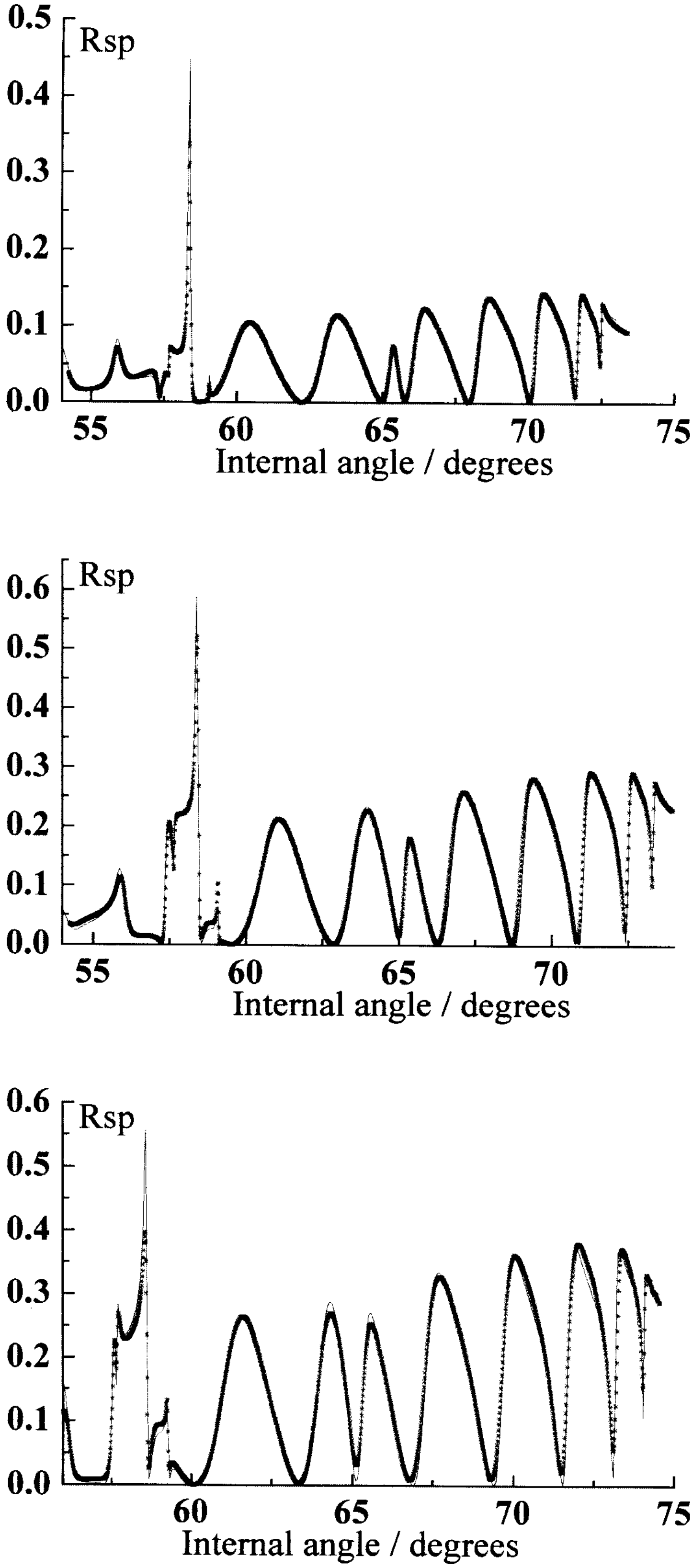

Figure 4. Fitted conversion reflectivity, R_{sp} versus the angle of incidence in the SmC* phase for three different temperatures. From the top curve: $T = 50°C$, 40°C and 30°C; the solid lines are the fitted theory to the experimental data (crosses).

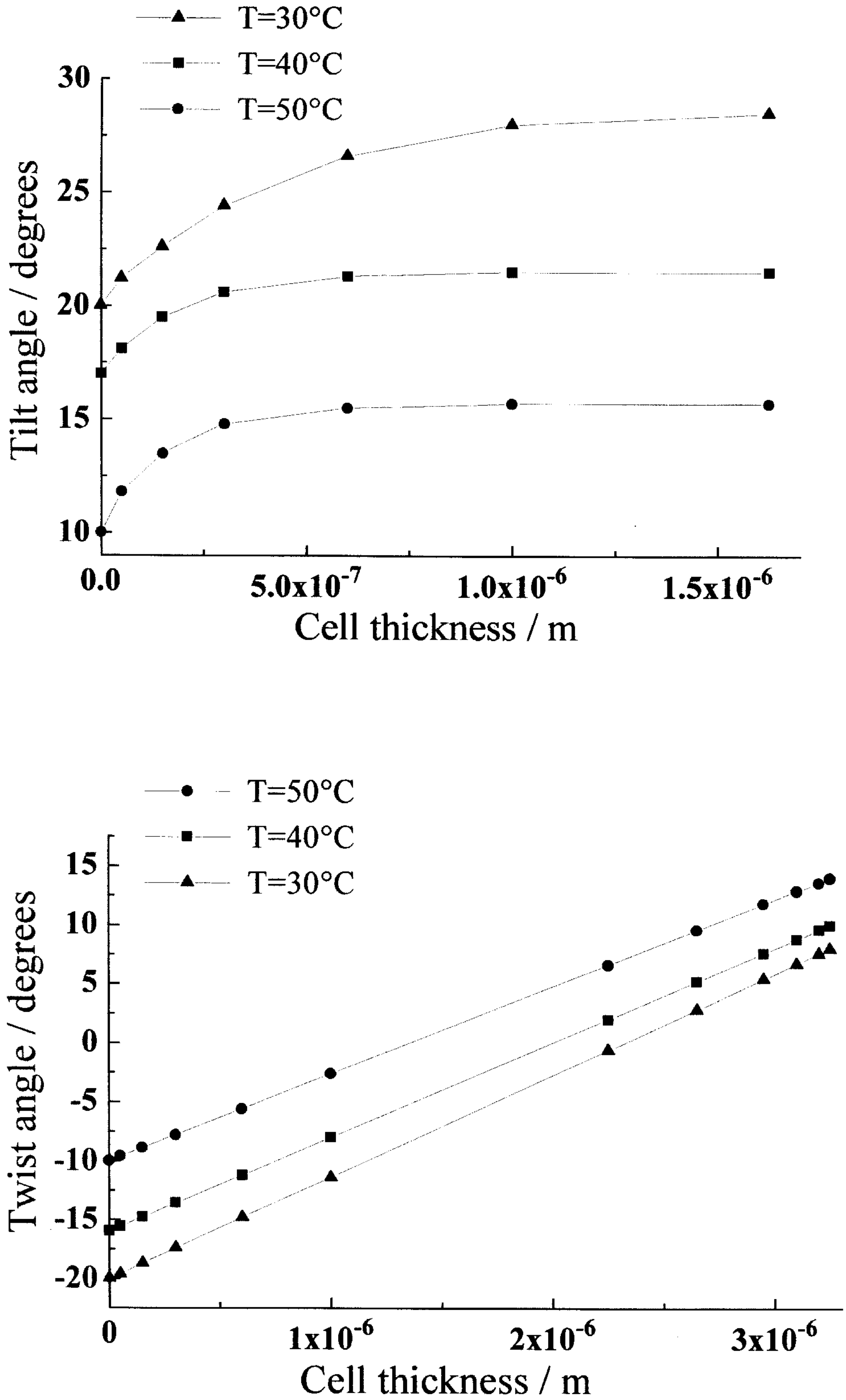

Figure 5. The fitted tilt angle profile from the surface to the middle of the cell (the distribution in the second half is symmetric) and the fitted twist angle profile from one cell surface to the other in the SmC* phase for three different temperatures.

this transition is reached, cooling is then speeded up to about 5°C per hour.

Optical continuity between the prism and the high index glass surface is achieved by a thin layer of matching fluid having the same refractive index as the neighbouring materials. Reflectivity versus angle data from

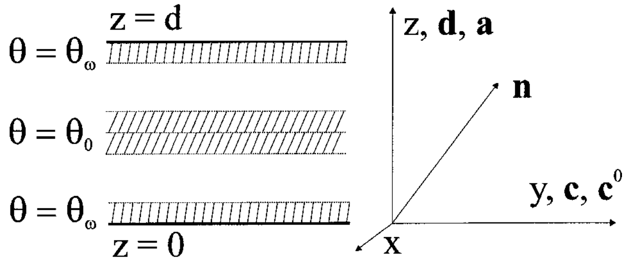

Figure 6. Sketch of the cell in the SmC* phase and the reference frame used; θ_χ and θ_0 are the surface and the bulk tilt angle, respectively.

the cell structure are taken either for *s* or *p* polarization input using either an *s* or *p* polarizer on the output; the sample in its oven is placed on a computer-controlled rotating table. Part of the input beam provides a reference to compensate for any laser intensity fluctuations (figure 2).

3. Results and discussion

The data presented begin with the material in the nematic phase, $T=90°C$ (figure 3 (*a*)). From the mode positions the liquid crystal optical permittivities are easily found; all other parameters are then fitted by iterative modelling. In this phase the fitted values are:

$$\varepsilon_{\perp LC}=(2{\cdot}2082,\ 5{\cdot}5\times10^{-5});$$

$$\varepsilon_{\parallel LC}=(2{\cdot}6110,\ 9{\cdot}0\times10^{-5});\ \text{LC thickness}=3{\cdot}283\ \mu\text{m};$$

$$\varepsilon_{ITO}=(4{\cdot}10,\ 0{\cdot}9\times10^{-2}),\ \text{ITO thickness}=610\ \text{Å}.$$

The model requires a permittivity gradient at the surface of the soft substrate glass which acquires a $\sim 1{\cdot}5\ \mu m$ strain layer during polishing. No *s* to *p* conversion is observed either in the nematic or in the SmA phase due to the very simple homeotropic alignment, with the molecular director everywhere normal to the cell surface. For $T=65°C$, in the SmA phase (figure 3 (*b*)), the permittivity values are: $\varepsilon_{\perp LC}=(2{\cdot}2037,\ 1{\cdot}0\times10^{-6})$; $\varepsilon_{\parallel LC}=(2{\cdot}6988,\ 3{\cdot}0\times10^{-6})$.

On cooling, a small *s* to *p* conversion signal becomes evident at $T=58°C$ signalling the SmA to SmC* phase transition. In the SmC* phase, fits of theory to *s* to *p* conversion reflectivity data are obtained for various temperatures (figure 4) using the tilt and twist director profiles across the cell as indicated in figure 5. In order to obtain the correct fitting, the model requires that a region near the lecithin coating has a lower tilt angle than the bulk layer. These results are satisfactorily explained by the compressible continuum theory for SmC* LC [6]. In this theory the liquid crystal is represented as having a unit director **n**, and in order to describe such a phase one must specify the spatial variation of the vectors characterizing the layer normal and the projection of the director into the smectic planes. Let **d** be the layer thickness vector and **c** the projection onto the smectic layer; then $\mathbf{a}=2\pi/|\mathbf{d}|$ is the density wave vector magnitude parallel to **d**. In the reference frame in which the *z* axis is normal to the surface plates (figure 6):

$$\mathbf{d}=\cos\theta(0{,}0{,}1);\ \mathbf{c}=\sin\theta(1{,}0{,}0);\ \mathbf{a}=[2\pi/\cos\theta](0{,}0{,}1);$$

$$\mathbf{n}=(\sin\theta{,}0{,}\cos\theta);\ \mathbf{c}^0=\sin\theta_0(1{,}0{,}0).$$

The tilt angle $\theta=\theta(z)$ does not vary within a layer, θ_0 is the cone angle expected in the bulk when no external influences take effect and $\mathbf{c}^0$ is the expected **c**-director when no boundary effects are present.

As in the nematic theory, the local energy density is assumed to take the form $W=W(\mathbf{a},\mathbf{c},\nabla\mathbf{a},\nabla\mathbf{c})$, being a quadratic function of the gradients. In this frame a simple model for the bulk free energy similar to Nakagawa is adopted:

$$W=\tfrac{1}{2}\,[K(\mathbf{c}\ \mathbf{c}-\mathbf{c}^0\ \mathbf{c}^0)^2 + A(\nabla\ \mathbf{a})^2 + C(\nabla\mathbf{c})(\nabla\mathbf{c}) - 2B(\nabla\mathbf{a})(\nabla\mathbf{c})], \qquad (1)$$

where K, A, C (>0) and B are elastic constants, $\nabla\ \mathbf{a}=a_{i,i}$ and $\nabla\mathbf{a}=a_{ij}$.

The dilation term $K(\mathbf{c}\ \mathbf{c}-\mathbf{c}^0\ \mathbf{c}^0)^2$ characterizes the energy change as θ deviates from θ_0. For a strong anchoring condition ($\theta=\theta_\chi$ at $z=0,d$), the cone angle is no longer at the constant equilibrium bulk angle: $\theta_\chi\neq\theta_0$, and consequently the layers compress as the director tilts away from normal to the SmA phase. The differential equation which describes this model is as follows:

$$\left(\frac{\sin\theta}{\cos^2\theta}+F^2\frac{\cos^4\theta}{\sin^2\theta}\right)\theta''+\left(\frac{1+\sin^2\theta}{\cos^3\theta}-F^2\cos^3\theta\right)$$
$$(\theta')^2-D^2\cos^3\theta(\sin^2\theta-\sin^2\theta_0)=0, \qquad (2)$$

where:

$$'=\frac{\mathrm{d}}{\mathrm{d}z};\ \theta=\theta_\chi\ \text{at}\ z=0{,}1;\ D^2=\frac{Kd^2}{2\pi^2A};\ F^2=\frac{C}{4\pi^2A}.$$

In general it is not possible to solve this system analytically; it has to be undertaken by numerical integration. However an analytical solution is possible when $F^2=0$; in this case the previous equation reduces to:

$$(\theta')^2=\frac{D^2\cos^4\theta}{2\sin^2\theta}(2\cos^2\theta_0-\cos^2\theta-\cos^2\theta_m)$$
$$(\cos^2\theta_m-\cos^2\theta); \qquad (3)$$

where $\theta_m=\theta(\tfrac{1}{2})$ is the tilt angle in the centre of the cell.

The expression for θ' assumes $\theta'=0$ in the centre; this is a natural assumption to make requiring on both boundary plates $\theta=\theta_\chi$, thus θ must be symmetric around the middle of the layer. The solution, following from the

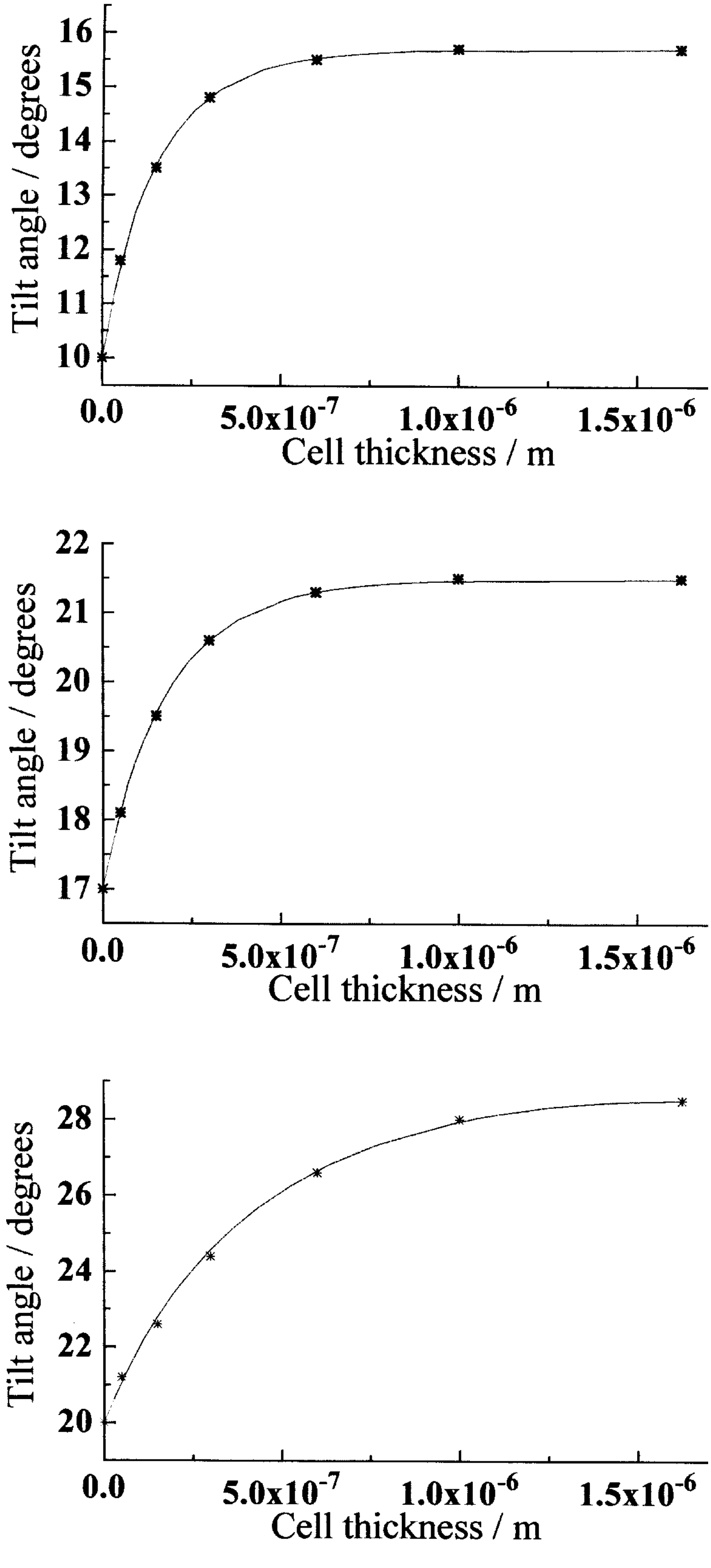

Figure 7. Comparison of the fitted tilt angle profile (crosses) to the compressible continuum theory (solid line). From the top: $T=50°C$, $D=14$; $T=40°C$, $D=14$ and $T=30°C$, $D=6$.

preceding simple assumption, gives θ in the form:

$$\int_{\theta^n}^{\theta^o} \frac{\sin\theta}{m(\theta)\cos^2\theta}\,d\theta = \frac{D}{2^{3/2}}, \qquad (4)$$

where

$$m(\theta) = [(\cos^2\theta - \cos^2\theta_m)(\cos^2\theta + \cos^2\theta_m - 2\cos^2\theta_0)]^{\frac{1}{2}}.$$

The constant F^2 is proportional to the elastic constant for the **c**-director deformation energy. In the SmC sample the tilt angle is expected to be θ_0 in a large portion of the sample. Since K is the elastic constant associated with compression of the layers as well as the deviation of θ from θ_0, the influence of the compression dilation energy in W is much greater than that of the **c**-director contribution, i.e. $Kd^2 \gg C$. As such, the assumption that $F^2=0$ allowing one to calculate an analytical solution is a physically realistic one. Furthermore the numerical results verify that the solutions for $F^2=0$ and $F^2 \neq 0$ are qualitatively similar.

In the SmC* phase a numerical solution of the tilt angle profile from equation (4) is obtained: the integration limits θ_0 and θ_χ determined from data fitting are fixed, then the D value is varied to achieve the tilt angle distributions that match the fitted ones (figure 7). Examination of the results obtained from continuum compressible theory reveals that the elastic constant ratio D (see table) seems to be constant at high temperatures, while at low temperature (30°C) its value diminishes, see figure 5. This change may be interpreted as being due to a variation of the elastic constant A which is connected to the splay deformation, while K should be almost constant depending upon the bend deformation. Unfortunately, a definite assessment of elastic constants of SmC* LC cannot be made, because they are unknown. Therefore no further aspects can be examined in depth. Additionally, the fit to the twist angle profile through the cell allows the pitch of the ferroelectric liquid crystal (pitch=cell thickness × 360/twist) to be determined as listed in the table.

Table.

Temperature/°C	Pitch/μm	D
50	48	14
40	45	14
30	42	6

4. Conclusions

The half leaky guided mode technique provides detailed information on the optical permittivity tensor and the director configuration in a thin homeotropically aligned ferroelectric liquid crystal cell. By careful fitting of multilayer theory to data in both the nematic and the SmA phase, we find that the density wave normal is everywhere perpendicular to the cell walls. In the SmC phase, the numerical solution of the tilt angle profile obtained by the continuum theory of compressible layers is in good agreement with the fitted solution. This confirms the compressible continuum theory and also provides information on the helical pitch and the elastic constant ratio, D, as a function of temperature.

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