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Determination of coherence length of a smectic liquid crystal

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A model for the near surface director profile in a homeotropically aligned smectic liquid crystal is developed based on the idea of the two independent anchoring energies separately associated with the director and the density wave at the surface. These anchoring energies are counterbalanced by the tendency to form the smectic C phase in the bulk. The model yields simple distance-dependent cone angle profiles which are compared with experimental data obtained from the half-leaky waveguide technique to obtain the coherence length for the penetration of the smectic C phase into the smectic A phase and the ratio of the surface to bulk cone angles.

1. Introduction

Since the realization in the early 80s of the surface stabilized ferroelectric liquid crystal (S_C^*) geometry [1] with its potential for bistable switching and hence device technology, there has been much interest in the S_C^* phase of liquid crystals. Many of the studies have been concerned, quite naturally following Clark and Lagerwall's paper [1], with the homogeneous alignment. Yet this is notoriously difficult to model. Even reduced theories [2–4] which simplify the mathematics of this structure by imposing special constraints are relatively unwieldy and not readily applied. Details of director anchoring at the aligning surface, the nature of the chevron interface and the compressibility of the smectic layers are all important yet unknown. Thus even though there is a substantial body of work studying homogeneous cells using both optical probing [5–10] and X-rays [11, 12], only very sketchy information on fundamental elastic properties have been forthcoming.

Recently, we have shown [13, 14], that while the rather less studied homeotropic alignment does not resemble device structures, it nevertheless provides a good geometry for the study of some fundamental characterizations of smectic materials. In particular, because the surface anchoring constraints of a homeotropically aligned cell are very weak, then the 'almost-infinite' liquid crystal response may be obtained. Using the very powerful half-leaky guided wave technique [15, 16] to probe the configuration of the optical tensor within the liquid crystal, we have been able to confirm extended mean field theory [13], determine for the S_C^* phase the cone angle, as well as the natural helical pitch [14] and more recently one of the elastic constants, B_3 [17], by application of in-plane electric fields.

In this present study we explore the nature of the near surface director profile in a homeotropically aligned cell in order to unravel some further fundamental characteristics of the S_C^* phase.

The orientational interaction of the smectic phase with the surface results in competition between different types of anchoring forces, one external, the other essentially bulk. For example, for the S_C or S_C^* phase, homeotropic anchoring (director normal to the surface) may well be in opposition to the cone angle as a consequence of the layer normal also attempting to be normal to the surface.

We already have shown [14] from our optical studies of homeotropically aligned smectic cells that there is a surface region in which the tilt angle varies spatially as we move away from the surface. Furthermore the results have also shown that, contrary to popular expectation, the smectic layers are not necessarily parallel to the surface even in the S_A phase. This latter behaviour almost certainly depends on the temperature dependence of the density wave spacing in the S_A phase and will be material dependent.

It is important to appreciate that the tilt of the layers is not a consequence of a prepared surface designed to give such a tilt, as for example in high tilt SiO_x alignments. There is no specific orientation of the layer normal imposed by the simple lecithin coated surface used in these experiments. As far as we were able to discover, the layer tilt is very simply a consequence of layer shrinkage in the S_A phase and space filling requirements.

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Here we turn to interpreting this surface reorientation region with the help of a simple continuum theory of the S_C phase, supplemented by appropriate surface energy terms. We note that the analysis of the surface region, and in particular the variation of the smectic C cone angle near the aligning surface has been performed by several authors. In 1981 Pikin and Yoshino [18] presented analytic results for the variation of cone angle in an S_C film, establishing the dependence of the transition temperature on the thickness of the film. They used the following simple expression for the bulk free energy of the S_C phase:

$$F_C = a\theta^2 + b\theta^4 + L\left(\frac{d\theta}{dz}\right)^2$$

where θ is the cone angle and the z axis is the surface normal. The surface free energy was taken in the Rapini form:

$$F_S = W\theta^2$$

A similar bulk free energy has also been used by Kraus *et al.* [19] in the analysis of the first order S_A to S_C phase transition in very thin films. Very recently, Ul Islam and co-workers [20, 21] have analytically modelled and numerically evaluated the cone angle profiles near the surface of a homeotropic cell using the same bulk free energy density and surface boundary conditions.

Thus in the simple case of homeotropic anchoring, the general form of the cone angle profile near a surface is well known. However, the detail of the interaction between the S_C liquid crystal and surface is poorly known. In the above studies, the surface energy has been modelled in the form of a simple nematic, depending on the director **n** at the surface. However in the S_C phase there is an independent contribution to the surface energy with the minimum corresponding to the director lying on the smectic cone (see figure 1). As a result the surface free energy of the S_C may have two minima. These minima coincide only if the surface promotes S_A type ordering. The situation becomes even more complex if there is layer tilt at the surface, as in our experimental situation. In this case the smectic plane normal **k** does not coincide with the surface normal **e** (see figure 1) and hence the surface cone angle θ is different from the angle θ' which determines the deviation of the director from the homeotropic alignment direction. In this paper we endeavour to consider these competing effects and draw some conclusions regarding the form of the surface free energy in a real S_C cell. For this purpose we determine the S_C coherence length ξ_c by comparing with experimental data. By fitting our model at various temperatures to the data obtained we find the temperature dependence of ξ_c.

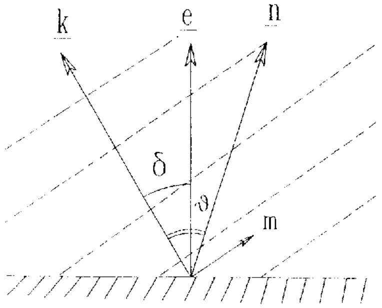

Figure 1. Model of the homeotropic smectic C phase near a surface.

As far as we are aware, the S_C coherence length has not been directly measured and there is little experimental information on this quantity. In the system under study the tendency to form the S_C phase is different at the surface from that in the bulk. As a result, the thickness of the boundary layer region is determined by the penetration length of the S_C phase into the S_A phase. This is particularly obvious for the reverse situation [22, 23] when the bulk is in the S_A phase while there still exists a surface region with non-zero surface tilt, that is it is S_C like.

2. Anchoring of the smectic C phase at a planar surface

The full description of the anchoring of the ferroelectric smectic, S_C^*, phase should account for the possible interaction between the spontaneous polarization and the surface. However the contribution of the polarization terms to the total free energy in the bulk is rather small [24] and these terms do not significantly influence the cone angle (a racemic mixture of an S_C^* material will have a measureably different cone angle only close to the S_A/S_C phase transition). In addition, for planar, structureless surfaces covered with lecithin, we do not expect any specific, strong, interaction between the surface and the spontaneous polarization. Thus as a first approximation we assume that the tilt angle profile near such a surface is negligibly influenced by chirality and therefore consider the much simpler problem of the anchoring of the achiral S_C.

The orientation of the S_C is determined by the smectic plane normal, **k** (the density wave vector) and the director **n**. We specify the orientation of the structureless planar homogeneous surface by the unit normal **e** (see figure 1).

We assume that in the nematic phase, to minimize the surface free energy, the director, **n** would like to be normal to the wall. This tendency is preserved also in the S_C phase, but it is accompanied by two other effects.

(1) The density wave normal (the layers) may be tilted with respect to the surface normal — there is good experimental evidence for this [13, 14].

(2) When the bulk liquid crystal is in the S_C phase, the director at the surface may have an independent tendency to be tilted relative to the smectic plane normal. This may well be sensitive to surface treatment.

In the general case there will be competition between the desired homeotropic alignment of the *director* at the surface and the possible tilt of the director with respect to the smectic plane normal — the density wave vector.

The total free energy per unit volume of the S_C may be written

$$\frac{F}{\sigma} = F_S + \int \mathrm{d}z \left\{ \alpha(T - T_{AC})\theta^2 + b\theta^4 + \cdots + L\left(\frac{\mathrm{d}\theta}{\mathrm{d}z}\right)^2 \right\} \tag{1}$$

where L is an appropriate elastic constant, σ is the surface area and F_S is the surface free energy, located within a few molecular lengths of the surface. This surface free energy, F_S, depends on the orientation of the smectic layers with the surface, on the orientation of the director with respect to the surface normal $\mathbf{e}$ and on the cone angle between the director $\mathbf{n}$ and the layer density wave vector $\mathbf{k}$.

We write

$$F_S = -W_o(\mathbf{n}\cdot\mathbf{e})^2 - U_S(\mathbf{k}\cdot\mathbf{e})^2 + a\theta_o^2 + B\theta_o^4 \tag{2}$$

where θ_o is the surface cone angle

$$\cos\theta_o = (\mathbf{n}\cdot\mathbf{k}) \tag{3}$$

The last two terms in equation (2) characterize the tendency to form an S_C phase at the surface. This is an energy due to the director being forced to deviate from the bulk S_C cone angle at the surface. This energy must be distinguished from the 'nematic' anchoring energy $W_o(\mathbf{n}\cdot\mathbf{e})^2$ which describes the energy cost of deviation from the surface normal, $\mathbf{e}$. We also note that the coefficients in the expression for the surface free energy (3) in powers of the cone angle can be different from the corresponding coefficients in the bulk.

Thus W_o is the 'nematic' anchoring strength and U_S is the smectic anchoring strength which characterizes the energy of the smectic layer being tilted with respect to the planar surface. This energy is related to the energy of the defects needed to allow the layer tilt.

We note in the general case while the constant a in the equation (2) is of order $\alpha(T - T_{AC})$, it does not necessarily vanish at the S_A to S_C phase transition. When $a < 0$ then a surface S_C tilt can exist in the S_A phase. By contrast, if $a > 0$ the S_C phase is suppressed at the surface.

The equilibrium director profile $\mathbf{n}(z)$, the orientation of the surface director $\mathbf{n}_o$ and the equilibrium orientation of the smectic plane normal $\mathbf{k}$ correspond to the minimum of the free energy using equations (1) and (2).

We now minimize this free energy with respect to the director $\mathbf{n}$, keeping the smectic layers fixed. The orientation of the director $\mathbf{n}$ is determined by the cone angle, θ, and the azimuthal angle ψ:

$$\mathbf{n} = \mathbf{k}\cos\theta + \mathbf{c}\sin\theta \tag{4}$$

where the unit vector $\mathbf{c}$ is given as

$$\mathbf{c} = \mathbf{m}\cos\psi + \mathbf{l}\sin\psi$$

where $\mathbf{m}$ and $\mathbf{l}$ are two unit vectors orthogonal to $\mathbf{k}$. For simplicity we set $\mathbf{l}$ to lie orthogonal to the surface normal $\mathbf{e}$; $\mathbf{l}\cdot\mathbf{e} = 0$.

Minimisation of the free energy with respect to the director $\mathbf{n}$ is equivalent to the minimisation with respect to θ and ψ. We note that only the surface free energy F_S (given by equation (2)) depends on ψ. Thus we can minimise F_S with respect to ψ and the tilt free energy with respect to θ.

In equation (2) only the first term, $-W_o(\mathbf{n}\cdot\mathbf{e})^2$ depends on ψ; thus we minimize this anchoring energy $-W_o(\mathbf{n}\cdot\mathbf{e})^2$ with respect to ψ at constant θ_o. Then

$$\begin{aligned} -W_o(\mathbf{n}\cdot\mathbf{e})^2 &= -W_o\{(\mathbf{k}\cos\theta + \mathbf{c}\sin\theta)\cdot\mathbf{e}\}^2 \\ &= -W_o\{(\mathbf{k}\cos\theta + \mathbf{m}\cos\psi\sin\theta \\ &\quad + \mathbf{l}\sin\psi\sin\theta)\cdot\mathbf{e}\}^2 \\ &= -W_o\{\cos\delta\cos\theta + \sin\delta\cos\psi\sin\theta\}^2 \end{aligned} \tag{5}$$

where we have taken into account that $(\mathbf{k}\cdot\mathbf{e}) = \cos\delta$, $(\mathbf{m}\cdot\mathbf{e}) = \sin\delta$ and $(\mathbf{l}\cdot\mathbf{e}) = 0$.

From the definition of the angles θ and δ it follows that $0 < \theta < \pi/2$ and $0 < \delta < \pi/2$.

It is obvious that the minimum of the anchoring energy (5) corresponds to $\cos\psi = 1$. Now the surface free energy, equation (2), can be written as

$$F_S = -W_o\cos^2(\theta_o - \delta) - U_S\cos^2\delta + a\theta_o^2 + B\theta_o^4 \tag{6}$$

With this and equation (1) we have to minimize the total free energy with respect to $\theta(z)$. The appropriate $\theta(z)$ profile is determined through

$$2L\frac{\mathrm{d}^2\theta}{\mathrm{d}z^2} + 2\alpha(T - T_{AC})\theta + 4b\theta^3 = 0 \tag{7}$$

for which the boundary condition is $\theta(z) \to \theta_b$ as $z \to \infty$, θ_b being the bulk cone angle.

The boundary condition at $z = 0$ is determined by minimization of the total surface free energy including the gradient term which appears after the minimization of the volume integral in equation (1). As a result we obtain

$$W_o \sin 2(\theta_o - \delta) + 2a\theta_o + 4B\theta_o^3 = 2L \left.\frac{d\theta_o}{dz}\right|_{z=0} \qquad (8)$$

It is now reasonable to go on to consider separately the solutions of equations (7) and (8) for both the S_A and the S_C phases.

3. Surface anchoring in the smectic A phase

In the bulk S_A phase, the cone angle, θ_b, is zero and we neglect the cubic term in equation (7), because the likely induced tilt is very small. Then equation (7) has the explicit solution

$$\theta(z) = \theta_o \exp\left(\frac{-z}{\xi_c}\right) \qquad (9)$$

where ξ_c is the S_C coherence length (i.e. the length of penetration of the S_C phase into the S_A phase)

$$\xi_c = \left(\frac{L}{\alpha(T - T_{AC})}\right)^{1/2} \qquad (10)$$

The surface cone angle, θ_o, is determined by the following equation, which comes from (8) and (9)

$$W_o \sin 2(\theta_o - \delta) + 2a\theta_o + \frac{2L}{\xi_c}\theta_o + 4B\theta_o^4 = 0 \qquad (11)$$

Assuming $\theta_o \ll 1$ and also $\delta \ll 1$, we can rewrite equation (11) as

$$\left(W_o + a + \frac{L}{\xi_c}\right)\theta_o \simeq \delta W_0 \qquad (12)$$

Thus equation (12) establishes a linear relationship between the surface cone angle θ_o and the layer tilt angle, δ, which is considered to be set by the layer shrinkage in the S_A phase. It is apparent that if $\delta = 0$, then correctly $\theta_o = 0$ and the cell will be a simple homeotropically aligned S_A phase with the layers parallel to the walls. On the other hand if δ is finite, then θ_o will also be finite, differing from δ as a consequence of the extra terms a/W_0 and $L/W_0\xi$.

4. Surface anchoring in the smectic C phase

In the S_C phase, the bulk cone angle, θ_b, is no longer zero and the cubic term in equation (7) cannot necessarily be neglected. However there is still an exact solution to the equation, the cone angle profile being given by

$$\theta_z = \theta_b \tanh\left(\frac{z}{\xi_c} + C\right) \qquad (13)$$

where ξ_c is as before, but with $T - T_{AC}$ reversed, that is $\xi_c = [L/\alpha(T_{AC} - T)]^{1/2}$, and C is a constant which is related to the surface cone angle θ_o through

$$\theta_o = \theta_b \tanh(C) \qquad (14)$$

The surface cone angle, θ_o, is now given by

$$W_o \sin 2(\theta_o - \delta) + 2a\theta_o + 4B\theta_o^3 = 2L\left(\frac{\theta_b}{\xi_c}\right)\left(1 - \frac{\theta_o^2}{\theta_b^2}\right) \qquad (15)$$

Then once more making the very reasonable assumptions that θ_o and δ are small, we arrive at the equation

$$\left(\frac{\theta_o}{\theta_b}\right)^2 + \gamma\left(\frac{\theta_o}{\theta_b}\right) = 1 + \kappa\delta \qquad (16)$$

with $\gamma = \xi_c/\xi_1$ and $\kappa = \xi_c/\xi_2$, where ξ_1 and ξ_2 are two additional lengths which characterize the surface properties of the system:

$$\xi_1 = \frac{L}{W_o + a} \qquad (17)$$

$$\xi_2 = \frac{L}{W_o}. \qquad (18)$$

We note that the relationship between the surface cone angle θ_o and the bulk cone angle θ_b strongly depends on the parameter γ. In the case of small γ, that is $\gamma \ll 1$, then $\theta_o/\theta_b \simeq 1 + \kappa\delta/2 - \gamma/2$, so the surface cone angle θ_o is greater than θ_b. By contrast, at larger γ,

$$\frac{\theta_o}{\theta_b} \simeq \frac{1 + \kappa\delta}{\gamma} \qquad (19)$$

which means $\theta_o < \theta_b$.

The important parameter γ is a ratio of the two lengths ξ_c and ξ_1. An order of magnitude for ξ_c is known from our earlier work [14]. That study showed the surface region to have a decay length of order 0·1 μm which for the moment we take as ξ_c, which we note is much larger than the molecular scale of order 3×10^{-4} μm. Our problem in estimating γ is that ξ_1 is unknown and so we may only make some rather crude estimates.

We take the elastic constant, L, to be of order

$$L \sim U_o l^2 \qquad (20)$$

where U_o is a typical intermolecular interaction energy. The constants a and W_o in equations (17) and (18) have the meaning of some interaction energy located at the boundary, within a thin boundary layer. Thus

$$W_o, a \sim U_o\xi \qquad (21)$$

where ξ is the thickness of the surface free energy boundary layer. Normally the surface free energy is defined within one or two molecular lengths; thus ξ_1

and ξ_2 are estimated as

$$\xi_1 \sim \xi_2 \sim l \qquad (22)$$

Hence we expect

$$\gamma \sim \frac{\xi_c}{l} \gg 1 \qquad (23)$$

We note that in practice equations (21) and (22) over estimate the value of the parameters W_o and a. For example, the estimate $W_o \sim U_o l$ corresponds to the strong anchoring case which may occur on an ideal structureless surface. In our system, the surface is covered by lecithin which gives rise to a weaker anchoring. This means that the lengths ξ_1, ξ_2 may be larger than l. Nevertheless it is reasonable to assume that ξ_1 is still smaller than ξ_c. Thus, in general we conclude that $\gamma > 1$ and θ_o/θ_b is given by equation (19), that is $\theta_o < \theta_b$. Of course, if in the S_A phase δ is finite, then as we cool into the S_C phase we go through a condition where $\theta_o = \theta_b$ before further cooling takes us into the region $\theta_o < \theta_b$.

We now go on to fit our homeotropic data with the above model.

5. Fitting data to obtain ξ_c

Three cone angle profiles at $T = 55{\cdot}1$, 50·0 and 44·8°C, determined by fitting data [14] for the homeotropically aligned (lecithin) S_C^* material SCE8, are shown in figure 2. A typical set of polarization conversion reflectivity data fitted by the simple modelling theory is shown in figure 3. The theory curve (full line), which gives profiles as in figure 2, fits the data extremely well. By using equation (13), we fit these profiles to give θ_b, ξ_c and C (or θ_o) at the three temperatures. The determined parameters are given in the table.

6. Discussion and conclusions

The data presented in the table indicate that the surface tilt angle θ_o is smaller than the bulk tilt angle θ_b. This result supports the relation (19) and we conclude that the parameter γ is greater than unity, i.e. the 'surface length', ξ_1 is smaller than the S_C coherence length ξ_c. We also conclude that the simple lecithin coated surface promotes S_A ordering and not S_C. This can be explained naturally by appreciating that one needs additional free energy to tilt the alkyl tails of the lecithin molecules at the surface. (The tilt will decrease the packing entropy of the system of tails which are on the average perpendicular to the surface.) By contrast, experimental data indicate [22, 23] that the free surface promotes S_C ordering when the bulk is in the S_A phase. In the free standing S_A films studied [22, 23], the S_C phase starts at the surface and penetrates into the bulk of the S_A phase.

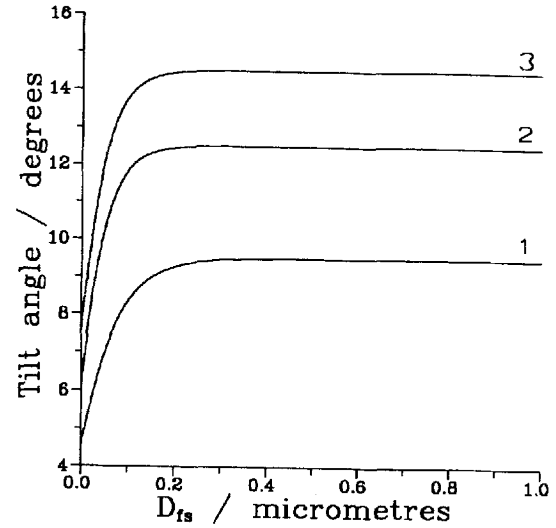

Figure 2. Cone angle against the distance, D_{fs}, from the surface of a homeotropically aligned (lecithin) S_C^* material (SCE8) at temperatures of 55·1°C (curve 1), 50·0°C (curve 2) and 48·8°C (curve 3), respectively. All three curves are determined by fitting the experimental data [14] using multilayer optics modelling theory, with the liquid crystal layer broken into a set of thin layers, each with its own director tilt. The resulting high quality fit gives the cone tilt angle profiles shown.

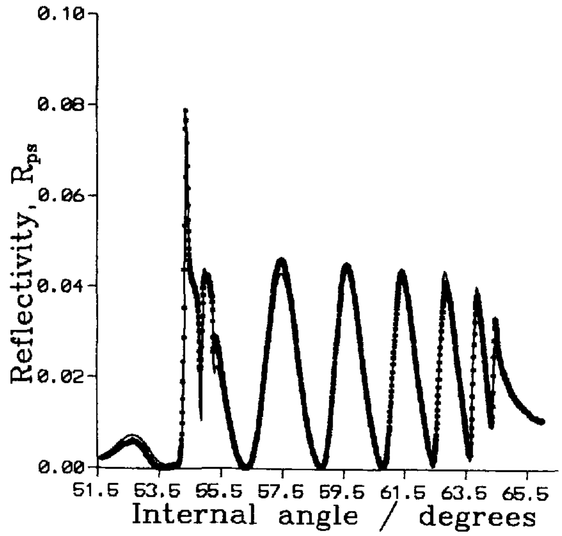

Figure 3. The experimental data (crosses) and theoretically fitted results (full line) for polarization conversion reflectivity at a temperature of 55·1°C in a homeotropically aligned SCE8 cell. The fitted parameters of the geometry are: wavelength $\lambda = 632{\cdot}8$ nm, pyramid and upper glass, $\varepsilon = 3{\cdot}2400$, liquid crystal $\varepsilon_{\parallel} = 2{\cdot}6580 + i0{\cdot}0006$, $\varepsilon_{\perp} = 2{\cdot}1960 + i0{\cdot}0003$, thickness 3·33 μm, substrate, $\varepsilon = 2{\cdot}1415$ [14].

Table. Parameters obtained using equation (13) and fitting the profiles in Fig. 2.

$T/°C$	$\theta_b/°$	$\xi_c/\mu m$	$\theta_o/°$	θ_o/θ_b
55·1	9·5 ± 0·1	0·12 ± 0·01	4·5 ± 0·1	0·47 ± 0·02
50·0	12·5 ± 0·1	0·08 ± 0·01	6·1 ± 0·1	0·49 ± 0·02
44·8	14·5 ± 0·1	0·07 ± 0·01	7·5 ± 0·1	0·51 ± 0·02

We also note that the ratio of the surface to bulk tilt angle is about 1/2 and is approximately temperature independent within the investigated range. This rather large ratio for θ_o/θ_b can be explained through equations (16) and (19), assuming that the 'surface lengths' ξ_1 and ξ_2 are both of the order of the coherence length ξ_c, but smaller. This of course makes ξ_1 and ξ_2 much larger than our estimate of a molecular length.

In this case the parameters γ_1 and κ_1 are of order 1 or a little more. Given then that δ is typically less than $\approx 0{\cdot}03(2^\circ)$, we find $\gamma \approx \theta_b/\theta_o \approx \xi_c/\xi_1$. Thus we arrive at the conclusion that the ratio of the surface to bulk director tilt angles, obtained by fitting the optical data, is a direct estimate of the ratio of the surface to bulk coherence lengths.

The large value of the surface coherence length indicates that the elastic forces which act on the nematic-like director at the surface are quite weak, i.e. the constants W_o and a are small. This corresponds to the case of weak nematic anchoring.

In conclusion, we have used the very powerful half-leaky guided wave technique to determine the director profile in a homeotropically aligned S_C^* cell. The experimental data have been fitted using a simple continuum theory of the S_C phase which takes into account the balance between the anchoring strengths of the director and of the smectic layering at the surface and the energy associated with the tilt of the director with respect to the density wavevector. In this way we have determined the coherence length for the penetration of the S_C phase into the S_A phase, the spatial variation of the cone angle near the surface and the ratio of the bulk to surface cone angles. The simple lecithin coated surface has been seen to promote S_A ordering with a characteristic coherence length only half that of the bulk coherence length.

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