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## Liquid Crystals

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## The alignment of a liquid crystal in the smectic A phase in a high surface tilt cell

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Liquid crystal alignment is studied using propagating optical mode techniques for a cell with a high surface tilt SiO alignment. The director configuration is determined for both the nematic and smectic A phases. In the nematic phase a uniform splay across the cell is demonstrated, as predicted by continuum theory. In the smectic A phase the structure is seen to be nearly uniform in the central region of the cell with large splay in boundary layers of about  $0.5 \mu\text{m}$  thickness. The reason for this appears to be competition between the tilted surface alignment forces and internal forces within the bulk of the smectic A layer which would induce a homogeneous configuration.

### 1. Introduction

Early studies of the alignment of liquid crystals in the smectic A and smectic C phases assumed that the smectic layers formed on crossing the nematic to smectic A phase transition were perpendicular to the cell surfaces in homogeneously aligned cells. It was assumed that this structure was retained when the smectic C phase was entered and that the molecular director (and hence observed optic tensor) was constrained to take up a configuration consistent with this [1]. Later studies using X-ray scattering techniques revealed that while it is true that the layers are perpendicular to the cell surfaces in the smectic A phase, this is not so in the smectic C phase [2]. This is because the pitch of the layer density wave decreases when crossing the smectic A to smectic C phase transition due to the tilt of the molecular axis away from the layer normal. The result of this is a corresponding tilt in the smectic layers in order to retain the same packing density of layers in the cell, leading to the observation of a 'chevron' structure in the smectic layering [2, 3]. Molecular director configurations are then constrained to form within this layer structure. Formation of this structure illustrates the importance of the way that smectic layer formation dominates the configuration in smectic liquid crystal cells.

Clearly the structure which forms on entering the smectic A phase if the nematic phase is not initially homogeneously aligned is more difficult to ascertain. In a symmetric cell with parallel high surface tilt alignment, the nematic phase is expected to be in a 'splayed' state, with the tilt angle smoothly varying from  $+\delta_s$  to  $-\delta_s$  across the liquid crystal layer. (This is due to the requirement of minimization of splay energy.) If when entering the smectic A phase the molecular director structure were to stay the same, then bent smectic A layers would form. However as these would not have a

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common centre of radius, the layers would be thicker in the centre of the cell than at the edges. This would lead to a variation in the pitch of the layer density wave (measured along the direction of the molecular director) across the cell; such a variation however is not allowed, as it is a material parameter [4]. Once again X-ray scattering data have revealed what happens. It has been shown that in the smectic A phase the smectic layers are perpendicular to the cell surfaces for a high surface tilt aligned cell, as they are in the homogeneous alignment case [5, 6]. Thus it appears that in formation of the smectic A layers in the case of non-homogeneous alignment, the average molecular direction dictates the direction of the layer density wave and layer normal. Normally, the molecular axis is perpendicular to the layering and therefore the X-ray data seem to imply that a high surface tilt cell which has splayed tilt in the nematic phase will be of homogeneous director (and optic tensor) alignment in the smectic A phase. This raises the issue of what happens near the surfaces of the cell, as here it is preferable for the molecular axis to lie tilted relative to the surface plane due to the surface alignment forces. However, if in the bulk of the cell the smectic A layering has formed perpendicular to the surfaces there must be some competition between the bulk alignment and surface alignment in the regions near the surfaces.

Here we use the excitation of propagating optical modes in a layer of liquid crystal material in a high surface tilt aligned cell to examine the optic tensor and hence director configuration throughout the sample in the nematic and smectic A phases. In this way we will be able to determine the optic tensor profile across the cell from surface to surface and study the relation between this optical result and the X-ray scattering data.

## 2. Experimental

Optical mode techniques for the study of liquid crystal alignment have been described elsewhere [7, 8] and will only be outlined briefly here. The experimental configuration consists of a cell formed between high index glass pyramids ( $n = 1.8$ ), which allow coupling into propagating optical modes in the liquid crystal layer. Thin silver films of about 45 nm thickness are coated on to the surface of the pyramids forming the cell. On to these silver films are then evaporated high surface tilt aligning layers of SiO of around 20 nm thickness: these are evaporated at an incident angle of  $85^\circ$  which gives a liquid crystal alignment with a surface pre-tilt of about  $30^\circ$  [9]. The cell is assembled with parallel alignment (meaning that the surface tilts are equal and opposite, to lead to a splayed configuration) and  $3.5 \mu\text{m}$  thick spacers, and the gap is then capillary filled with the Merck Ltd liquid crystal material SCE12 in the isotropic phase. Cooling into the nematic and smectic A phases is done slowly ( $< 1^\circ$  per hour) in order to form good alignment at all times.

With the cell constructed in this way it is possible via light incident in one of the pyramids to excite both guided modes in the liquid crystal layer and surface plasmon polaritons (SPPs) at the silver liquid crystal interface [7, 10]. The guided modes are sensitive chiefly to the liquid crystal alignment in the bulk of the cell, and the SPP, being a surface bound mode, is sensitive to alignment near the surfaces. In a cell which is homogeneously aligned, these modes will be of pure polarization: the guided modes can be either s-polarized (TE) or p-polarized (TM), and the SPP is a TM mode only. Modes are observed by examining the reflectivity from the cell as a function of angle of incidence for light entering through one of the pyramids. Data of this nature are taken with the cell oriented such that the homogeneous liquid crystal director is parallel to the plane of light propagation, and also with the director perpendicular to the plane of propagation. It is then possible by comparison with theoretically predicted reflectivity

curves to determine the relative permittivities of the liquid crystal optic tensor. Prior to the work presented here this was done with a homogeneously aligned SCE12 cell in order to determine the optical permittivity tensor of the material at various temperatures in the nematic and smectic A phases.

The optical situation in the case studied here is a little more complex due to the non-homogeneity of the alignment. Tilt in the surface alignment will lead to off-diagonal terms for the optical tensor in the plane of light propagation if the cell is orientated such that the average director is perpendicular to the incident plane. These off-diagonal terms will couple s-polarized and p-polarized light and the excited optical modes will no longer be plane polarized, but will be mixed polarization s-p (TE-TM) modes [11]. This mixing allows detailed study of the liquid crystal alignment if the cell is oriented in this way [12]. In the nematic phase with uniform splay expected across the cell, we anticipate s-p mixed modes, and fitting reflectivity theory to data taken in this state should confirm this. However, in the smectic A phase, the situation will depend on the molecular director (and hence optic tensor) configuration which forms. If as the X-ray scattering data suggest the structure is forced into a relatively homogeneous alignment, then little mixing of modes will be observed and a similar result to that for the simple homogeneous alignment will be obtained.

### 3. Results

Reflectivity data taken for incident p-polarized light at 632.8 nm with the SCE12 in the nematic phase at 85°C and the cell oriented such that the average director is perpendicular to the plane of propagation are shown in figure 1. The series of sharp dips

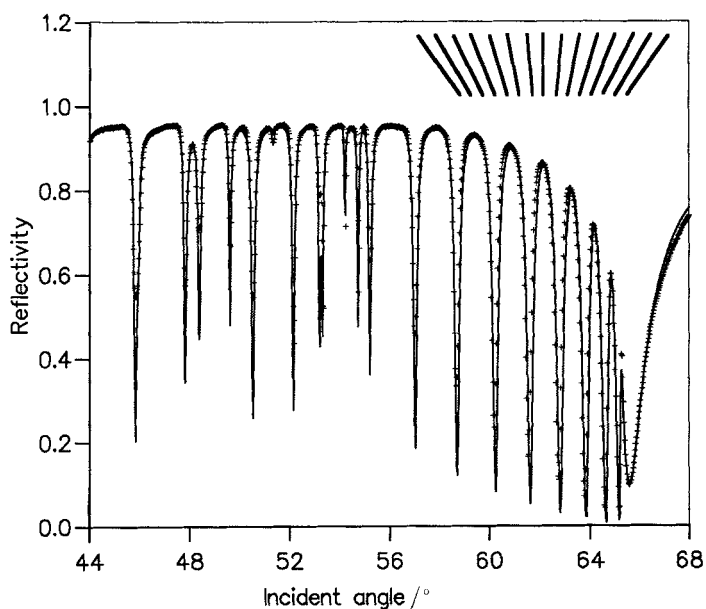


Figure 1. Reflectivity data (crosses) for incident p-polarized light with a high surface tilt cell with the average alignment direction perpendicular to the plane of light propagation. The cell is filled with SCE12 in the nematic phase at 85°C. The continuous line represents a theoretical fit to the data, using a model with uniform splay across the cell, as illustrated in the inset.

below  $56^\circ$  are due to the excitation of mixed polarization TM-like and TE-like modes: both mode types are excited because there are off-diagonal terms in the optic tensor in the plane of light propagation (the uniaxial axis, the director being not everywhere parallel to the cell surfaces). The sharp dips above  $56^\circ$  are due to excitation of TE-like modes, supported in the high index region in the centre of the cell. As the nematic director is splayed across the cell, the TE-like modes sense a larger index in the centre of the cell, with lower indices towards the surfaces, and this enhances the guiding effect. These TE-like modes are excited with TM incident light because of the tilt of the director out of the surface plane. At about  $66^\circ$  a broader dip is seen; this is due to the excitation of the SPP at the silver/SiO/liquid crystal interface.

A theoretical comparison with the data is also shown for a model with uniform splay in the nematic director, as illustrated by the inset in figure 1. This theoretical reflectivity curve is calculated by splitting the non-uniform liquid crystal layer into many thin homogeneous slices and then using multi-layer-optic matrix techniques to determine the reflectivity [13, 14]. It can be seen that the comparison between theory and experiment is very good, confirming that, as expected, the optic tensor (director) configuration is uniformly splayed across the cell. Only one parameter exists in addition to the cell thickness when fitting this uniform splay model for the nematic phase; this is the surface pre-tilt angle, assumed to be equal and opposite at the two surfaces, as the aligning layers were prepared simultaneously. In this case this is shown to be  $29 \pm 2^\circ$ .

When the cell is cooled into the smectic A phase, there is a considerable change in the form of the reflectivity data. Data taken with p-polarized light for the cell oriented as before, but now in the smectic A phase at  $75.9^\circ\text{C}$ , are shown in figure 2. There are a number of things which can be noted about these data:

- (i) The director configuration is (as expected) no longer uniformly splayed across the cell, as can be seen by comparison of these data with those shown in figure 1. Small changes in refractive indices of the material take place due to the decrease in temperature, but these would not lead to the large change seen in the data.
- (ii) The director configuration is not simply homogeneous, as initially indicated by the X-ray scattering data [5]. A reflectivity curve calculated for a uniform homogeneous alignment is shown together with the data in figure 2. This shows no TE–TM mode mixing (apparent from the absence of sharp dips in the SPP resonance), because in this structure there are no off-diagonal terms in the optic tensor in the plane of incidence. This contrasts with the strong degree of mode mixing shown by the data which indicates that there is still significant tilt of the director somewhere within the cell.
- (iii) The TE-like mixed modes now extend to higher angles. In the nematic phase, the cut-off for mode excitation was about  $65^\circ$ ; now it extends to about  $66^\circ$ . This indicates that more of the cell has the director oriented perpendicular to the plane of propagation, leading to a larger effective index for TE-like modes.
- (iv) The SPP has moved to a lower angle, now being excited at about  $63^\circ$ , instead of about  $66^\circ$ . As the dominant electric field component in the SPP is perpendicular to the silver surface, this indicates less tilt of the director in the surface regions leading to a lower effective index in this direction.

The simplest model consistent with all these observations is that the director in the smectic A phase is largely homogeneous, but that near the surfaces, highly splayed

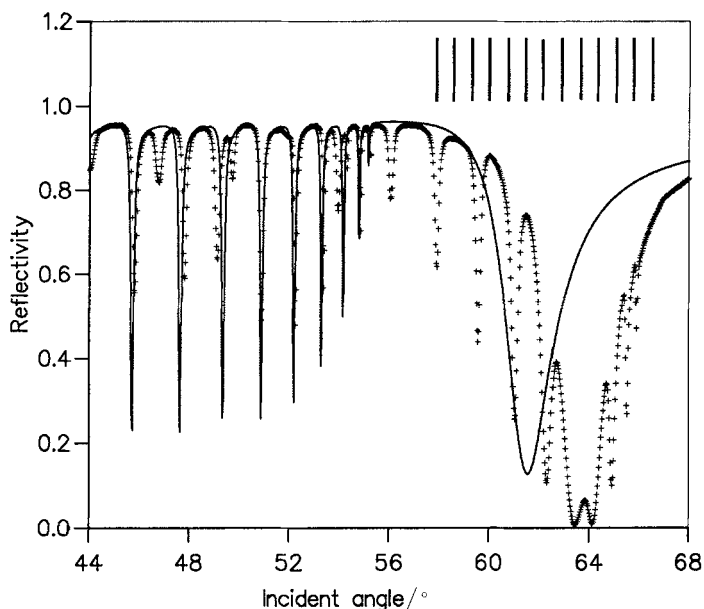


Figure 2. Reflectivity data (crosses) for the same configuration as in figure 1, but with the sample in the smectic A phase at 75.9°C. The continuous line is the theoretical reflectivity for a uniform slab of material (see inset), as would be expected if the smectic A layers were everywhere perpendicular to the cell surfaces.

regions occur: we need to consider what form these regions take. It was commented earlier that we might expect competition between the forces which would cause the smectic layering to lie in a uniform structure and those which would induce the director at the surfaces to lie at the surface tilt angle. In order to incorporate this in a model of the director profile, we will introduce an empirical elastic energy expression which assumes that the energy cost of non-uniform layering is to be balanced by the energy cost of director splay. We could then write a simple integral for the elastic energy as follows:

$$E \propto \int_{z=0}^{z=1} \delta^2 + K \left( \frac{d\delta}{dz} \right)^2 dz$$

where  $\delta$  is the director tilt angle,  $z=0$  is in the centre of the cell and  $z=1$  is at the cell surface. As the layering is assumed to form preferably perpendicular to the cell surfaces (as seen in the X-ray work), the  $\delta^2$  represents a cost in layer compression energy due to tilting the layering away from this: small tilts lead to small deformations in the layer density wave, and as the tilt increases so does the deformation and cost in energy. The  $d\delta/dz$  term represents the energy cost of director splay, and the constant  $K$  represents the balance between these two terms. The resulting expression inside the integral is a simplified form of that presented by Ahmadi [15]. Now applying variational calculus to this expression, and setting the correct boundary conditions, we obtain

$$\delta(z) = \delta_s \frac{\sinh(z/\sqrt{K})}{\sinh(1/\sqrt{K})}$$

Consideration of this form for the molecular director configuration, with a small value for the constant  $K$ , shows that it satisfies the points (i) to (iv) listed above. Using this

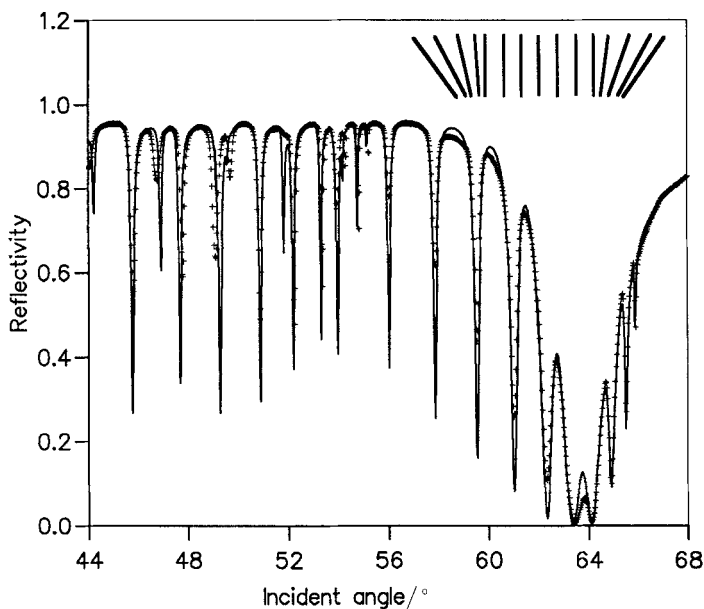


Figure 3. A theoretical fit (continuous line) to the same data (crosses) illustrated in figure 2. The model has a non-uniform director splay across the cell, concentrated in boundary regions, with approximately uniform alignment in the bulk of the cell. The profile is illustrated in the inset.

form we can once again calculate reflectivity curves for comparison with the data, using the surface tilt,  $\delta_s$ , and constant  $K$  as fitting parameters. Doing this allows us to obtain the comparison between data and theory illustrated in figure 3. This is obtained with  $\delta_s = 24 \pm 2^\circ$  and  $K = 0.025$ . Using this simplistic model, most of the excited optical modes are at the correct angles, with one or two subtle differences between theory and experiment.

#### 4. Conclusions

The small differences between the data and the theory in figure 3 are due to the simple form used for the director profile, and are not likely to indicate a fundamental flaw in its general form. Thus we see that the profile, as illustrated by the inset in figure 3, is nearly uniform in the central region of the cell, but with a large splay in transition regions near the surfaces. These regions are determined to be about  $0.5 \mu\text{m}$  in thickness. It should be noted that a fit of near equal quality can be obtained with a uniform central region and simple smoothly varying tilted regions for the boundary layers. However, the continuum nature of the liquid crystal material indicates that this is less likely to be a correct description of the director profile. In applying this simple model to the data in the smectic A phase, there has been a decrease in the surface tilt angle from  $\sim 29^\circ$  (in the nematic phase) to  $\sim 24^\circ$ . This is due to competition between the bulk alignment (which tends to be homogeneous) and the surface pre-tilt of the SiO alignment. As the pre-tilt forces are not very strong, the extra stress in the smectic A phase leads to the observed decrease in surface tilt angle. It should also be noted that normally the cost of change in the layer density wave is very large [2] and this would lead to a very small value for the constant  $K$  in the expression used here. The result of  $K = 0.025$  is rather larger than expected and is still to be explained.

As the above description appears to be very nearly the correct optical tensor profile for the high surface tilt cell in the smectic A phase, a question of consistency with the X-ray scattering data is raised. Near surface transition regions of around 0.5 μm thickness should have been large enough to lead to an effect on the X-ray scattering data (the variation in layer tilt broadening of the diffraction peak), and yet the X-ray work indicated a uniform homogeneous layering [5, 6]. A possible explanation is that the layering is broken near the surface and ceases to typify a true smectic A phase. Such a structure could then lead to a smooth splay in the molecular director as recorded here (and may also be the reason for the larger than expected value of *K*), but would only contribute to the background scattering signal in X-ray work. A surface induced decrease in ordering of this nature has previously been used to explain a large difference (~18°) between the surface alignment direction and the smectic A orientation which can occur in a homogeneously aligned cell [16]. However the balance between molecular chirality and degree of smectic ordering then led to a transition region thickness of ~250 Å much less than seen here for high surface tilt alignment. Surface induced changes in ordering of liquid crystals are common, often leading to the pre-transitional appearance of higher order phases [17]; thus the tilt observed near the surface in this work may indicate the existence of a pre-transitional smectic C phase. But we would then expect the transition region to be of the same nature as that seen in the surface electroclinic effect, and previous studies have revealed the thickness of this to be ~50 Å [18]. Thus, these effects do not provide a full explanation and the issues remain to be clarified.

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