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Studies of the alignment properties of antiferroelectric liquid crystals by X-ray diffraction

L. A. PARRY-JONES*, S. M. BELDON, D. RODRIGUEZ-MARTIN†,
R. M. RICHARDSON† and S. J. ELSTON

Department of Engineering Science, University of Oxford, Parks Road,
Oxford OX1 3PJ, UK
†H. H. Wills Physics Laboratory, University of Bristol, Tyndall Avenue,
Bristol BS8 1TL, UK

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It is observed optically that in a parallel rubbed antiferroelectric liquid crystal device, the texture consists of domains with two distinct optic axes, which make equal and opposite angles with the rubbing direction. It is proposed that this is caused by a large electroclinic effect at the surfaces during layer formation in the SmA* phase. This hypothesis is verified by finding the layer structure in single, parallel and skew rubbed devices by using X-ray diffraction.

1. Introduction

Antiferroelectric liquid crystals (AFLCs) [1–3] are among the most promising materials for display applications. Being chiral tilted smectic liquid crystals, each layer has a spontaneous polarization to which an electric field applied parallel to the layers can couple. So, like ferroelectric liquid crystals, they are capable of fast, in-plane switching. In addition, however, their tristable switching properties offer the possibility of symmetric driving schemes and increased ease of grey scale generation. The main difficulties in using AFLCs for displays are the so-called 'pretransitional effect' and the difficulty in obtaining good quality alignment, both of which limit the achievable contrast. Having previously considered the pretransitional effect elsewhere [4], this paper will concentrate on the issue of alignment.

When an AFLC is used to fill a cell whose inside surfaces have been coated with a polymer layer and rubbed in the same direction (parallel rubbing), the texture as viewed using a polarizing microscope is as shown in figure 1 (*a*). The cell is subdivided into domains with two distinct optic axis directions, which are rotated by equal and opposite angles, $\pm\alpha$, from the rubbing direction. However, when the AFLC is used to fill a cell which has had only one of its polymer coated surfaces rubbed, there is only one optic axis orientation, figure 1 (*b*), which is rotated by a similar angle α from the rubbing direction. By making a cell in which both surfaces are rubbed, but the directions are skewed by an angle 2α, it is also possible to obtain a single optic axis orientation, figure 1 (*c*). The details of this method of 'skew' rubbing have been reported elsewhere [5, 6].

We propose that in the single rubbed device the rotation of the optic axis away from the rubbing direction is caused by the surface electroclinic effect [5, 7]. After filling the cell in the isotropic phase, as the temperature is reduced into the SmA* phase, the molecules at the surface prefer to lie along the rubbing direction. In the absence of the surface electroclinic effect this would result in the formation of smectic layers perpendicular to the rubbing direction, figure 2 (*a*). However, we suggest that the interaction between the polar alignment layer and the large spontaneous polarization of the liquid crystal (characteristic of materials exhibiting a SmC$^*_A$ phase) induces the SmC* phase at the surface: the surface electroclinic effect. The fact that the molecules prefer to lie along the rubbing direction, and must tilt with respect to the layer normal, means that the layers form with their normal at an angle to the rubbing direction, as shown in figure 2 (*b*).

If, however, both surfaces are rubbed parallel to each other, then the equal and opposite polarizations at either surface will cause layer formation with conflicting layer normals, figures 2 (*b*) and 2 (*c*). Instead of forming one layer arrangement in the top half of the cell, and the other arrangement in the bottom half, in practice the layer formation in the bulk will be seeded randomly from either one surface or the other. The resulting layer structure is therefore a pattern of domains of two types, one with a layer structure as in figure 2 (*b*) and the other as in 2 (*c*). The texture of the cell will therefore appear

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*Author for correspondence;
e-mail: lesley.parry-jones@eng.ox.ac.uk

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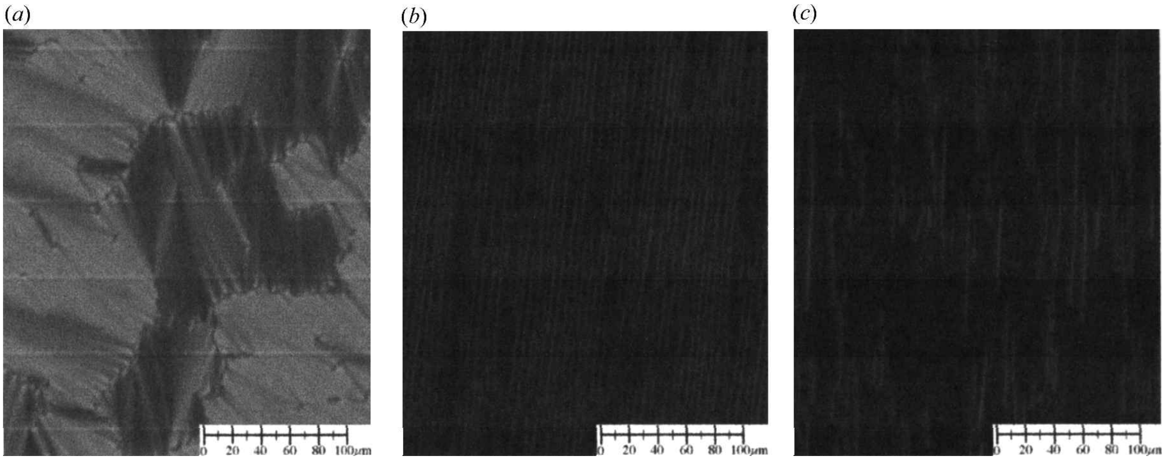

Figure 1. Texture of an AFLC cell as viewed using a polarizing microscope under different alignment conditions. (*a*) The alignment layers at both surfaces are rubbed parallel to each other: this gives rise to a pattern of domains with two principle optic axis orientations. (*b*) Only one surface is rubbed and (*c*) both surfaces are rubbed, with the directions skewed with respect to each other by an optimized angle: in both cases (*b*) and (*c*), there is only one optic axis orientation.

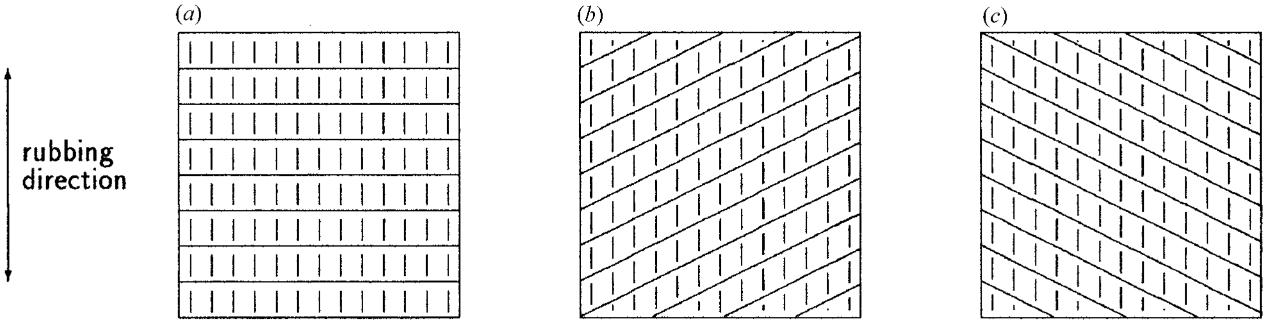

Figure 2. In a single rubbed cell (*a*) in the absence of a polar interaction with the surfaces, the smectic layers form perpendicular to the rubbing direction in the SmA phase. However, when there is such an interaction, the SmC* phase is induced at the surfaces and the layers form with their normal at an angle to the rubbing direction. The surface polarizations are equal and opposite at either surface, so the layers form one way at the top surface (*b*), and the other way at the bottom surface (*c*).

as a mixture of regions with two different optic axis alignments. If, however, the direction of rubbing at the two surfaces is skewed by the correct angle—twice the angle between the layer normal and the rubbing direction in figure 2(*b*)—then there will be uniform layer formation throughout the cell.

Clearly, the hypothesis that the surface electroclinic effect is responsible for the poor alignment observed in parallel rubbed devices is consistent with optical investigations. However, since the core of this argument is in the formation of the smectic layers, the ultimate test is to investigate the layer structure by X-ray diffraction (XRD).

2. Experimental

XRD experiments were carried out at Station 2.1 of the Synchrotron Radiation Source (SRS) at the Daresbury Laboratories, UK. Devices suitable for X-ray scattering studies were fabricated using cover slips of thickness 0.1 mm (to minimize attenuation of the X-ray beam) with single, parallel and skew rubbed polymer alignment layers. The devices (filled with AFLC mixture CS4001) were typically 2 μm thick. The optimum skew angle for the alignment material X201, and the AFLC material CS4001 was found to be 18°. The X-ray beam was of wavelength 0.154 nm, and the experiment was carried out with the cell in the Bragg geometry (figure 3), with the axis of rotation perpendicular to the rubbing/average rubbing direction (for single and parallel/skew rubbed cells, respectively). The X-ray beam was linearly polarized in the plane of incidence, and was about $3 \times 1\,\mathrm{mm}^2$ in size.

3. Results and discussion

The results of the experiments on the single, parallel and skew rubbed cells are shown in figures 4(*a*), (*b*) and (*c*), respectively. The axes of the plots are the angles γ

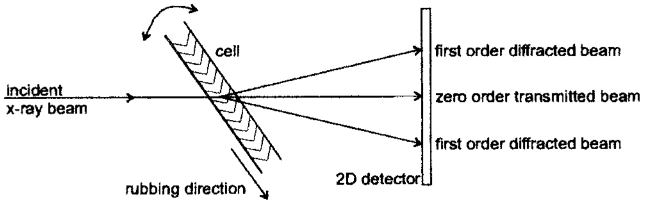

Figure 3. Bragg geometry for the X-ray scattering experiments: the cell is rotated about an axis that is normal to both the incident X-ray beam and the (average) rubbing direction (into the page). For each angle, the X-ray scattering pattern is recorded by the 2D detector.

and δ which represent twist and tilt of the layer normal, respectively, as illustrated in figure 4(*d*). The method of presenting layer normal distributions is reported elsewhere [8].

The result for the single rubbed cell shows clearly two peaks at a twist angle $\gamma \simeq 80°$, with equal and opposite values of the tilt angle $\delta = \pm 16°$. This indicates the presence of a tilted chevron structure, with the component of the layer normal that is in the plane of the glass plates at an angle of about 10° to the rubbing direction, in agreement with our hypothesis about the surface electroclinic effect. Note that one of the peaks is much more localized in terms of twist angle than the other. This is produced by the lack of a defined alignment direction on one of the substrates. The chevron arm which is induced by this substrate can slip in order to accommodate defects, whereas the direction of the other chevron's arm is fixed by the direction of the molecular director parallel to the rubbing direction. Although the layers neighbouring the unrubbed surface can slip, its distribution in twist angles is centred at $\gamma \simeq 80°$ because

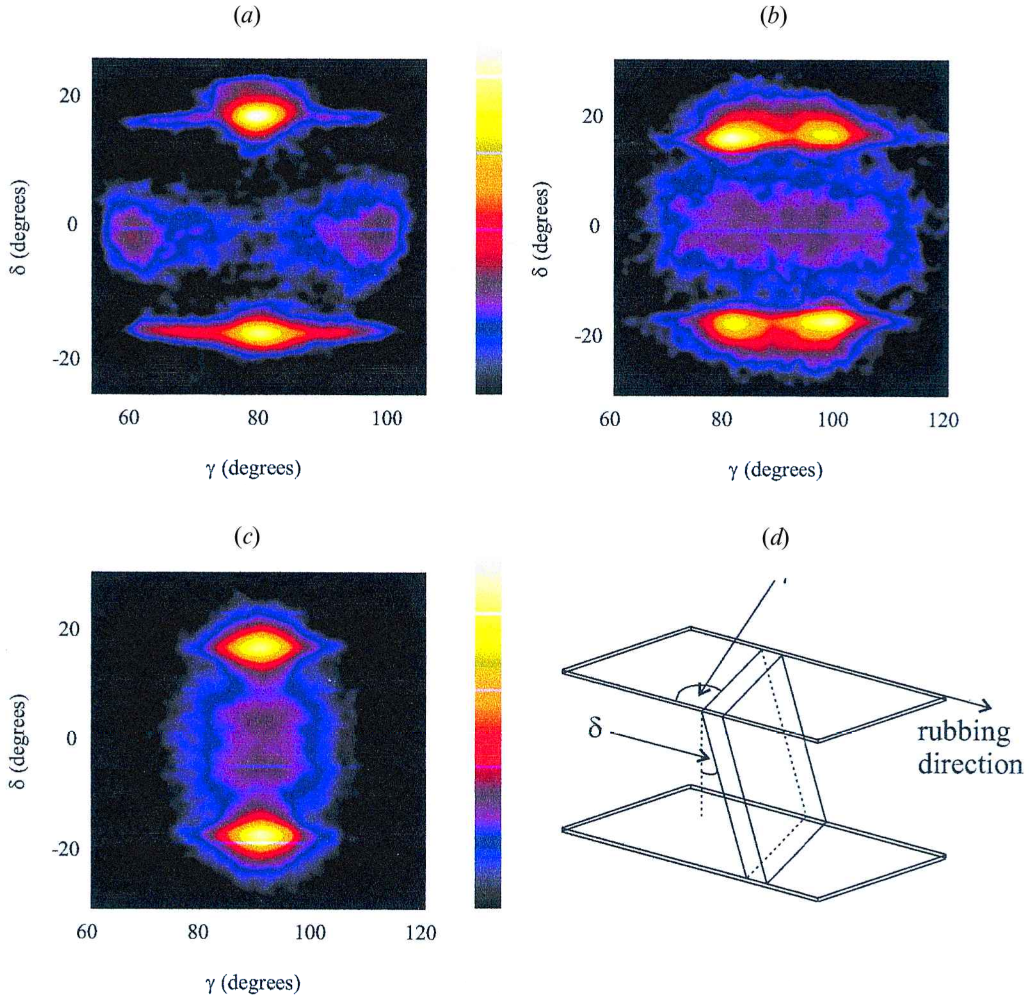

Figure 4. Comparison of the experimental results of an X-ray scattering experiment to determine the layer structure of (*a*) single, (*b*) parallel and (*c*) skew rubbed devices. The axes of the plots are δ and γ, the tilt and twist, respectively, of the smectic layer normal, as shown in (*d*).

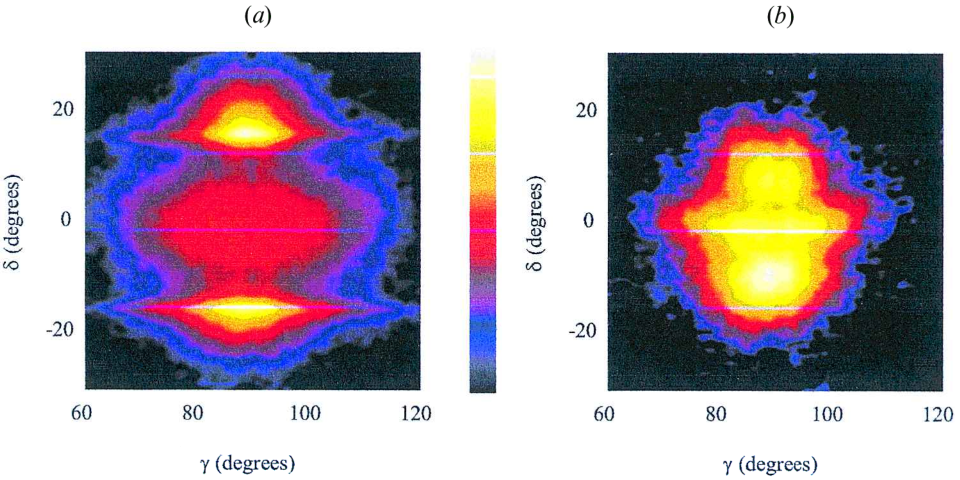

Figure 5. Comparison of the layer structure in a skew rubbed cell, (*a*) before and (*b*) after field treatment.

of the layer continuity at the chevron interface. Thus, we can say that the chevron interface acts as a surface by itself with some boundary conditions (director continuity).

Figures 4(*b*) and (*c*) show that the results for the parallel and skew rubbed devices support this conclusion. The results for the parallel rubbed cell show two predominant twist directions, as well as a chevron structure. The two directions are at $\gamma = 82°$ and $\gamma = 97°$, i.e. approximately equally spaced about the rubbing direction, in agreement with optical observations. The skew rubbed cell shows a chevron structure with the layer normal along the average rubbing direction. We therefore have full agreement between the experimental results and our hypothesis for the layer structure in these three types of device, based on the effects of the surface electroclinic effect.

It has been observed previously by optical methods [5, 6] that in the skew rubbed cells, the density of needle defects is increased by the application of an electric field. This was also investigated by XRD, and the results are shown in figure 5. The device has a non-twisted chevron structure, with a tilt angle of about 16° before field treatment, as shown in figure 5(*a*). After the application of a 0.1 Hz, ±4 Hz triangular voltage wave for ten minutes, the layer structure is greatly changed, as shown in figure 5(*b*). There is still some chevron structure (the peaks at $\gamma = 0$, $\delta \neq 0$), but at a much smaller tilt angle of about 9°. This 'bookshelfing' of the smectic layers under applied bias fields is well known in both ferroelectric [9] and antiferroelectric [10] liquid crystals, and is understood to occur due to the non-zero torque that exists between the spontaneous polarization and the electric field in the ferroelectric state of a tilted layer. In addition, figure 5(*b*) shows that other parts of the cell have a twisted bookshelf structure (the peaks at $\delta = 0$, $\gamma \neq 0$), which we interpret as originating from the needle defects observed in the cell. Needle defects occur in order to maintain layer packing density during layer reorientation towards a bookshelf structure. They grow under the application of an applied field because they have zero angle between the spontaneous polarization and the applied field in the ferroelectric state.

4. Conclusions

We conclude that the results of XRD are in excellent agreement with our hypothesis, based on optical observations, that the poor quality alignment achieved in parallel rubbed AFLC cells is due to the formation of two distinct layer directions, caused by the surface electroclinic effect. This can be improved by using only one rubbed substrate or an optimized skew angle between the two rubbing directions. We have also shown how the layer structure changes upon application of an electric field: in some parts of the cell, the chevron tilt angle reduces, whereas in others, needle defects appear and grow. The much reduced tilt and twist angles after only a short duration of applied field [10] support the assumption, often made in modelling AFLCs that the layers are in a 'quasibookshelf' configuration.

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