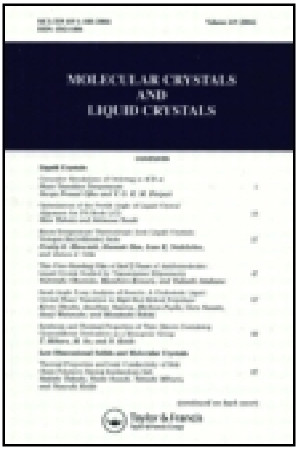

Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl19

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Version of record first published: 24 Sep 2006

Mol. Cryst. Liq. Cryst., Vol. 362, pp. 203-215
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Time-resolved X-Ray Scattering Studies of Reversible Layer Flexing in a Surface Stabilised Ferroelectric Liquid Crystal Device

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Recent time resolved x-ray scattering studies of a surface stabilised ferroelectric liquid crystal (SSFLC) device have demonstrated that the smectic layers move in two orthogonal planes during low field switching of the device. This paper extends our initial studies of the layer motion and presents a detailed examination of the influence of low field switching on the chevron layer structures in a 3 μm SSFLC device. A symmetric bipolar pulse was applied to the device at a temperature 5°C below the ferroelectric chiral smectic-C to smectic-A phase transition. X-ray scattering data, corresponding to the intensity and position of the Bragg peak, were collected with microsecond time resolution. Results are presented which show a rapid, reversible flexing of the smectic layers to much lower angles during switching with fields of magnitude ±3 $V\mu m^{-1}$ followed, in some cases, by a slow (millisecond) return to the original layer structure. The data are to some extent consistent with the layer flex model proposed by Gießelmann, followed by domain motion within the device.

INTRODUCTION

The layer and director configurations of surface stabilized ferroelectric liquid crystal (SSFLC) display devices have been investigated by many authors[1] and it has long been known that they have a significant impact on the electro-optic switching of such devices. In 1987, Reiker *et al*[2] first used x-ray diffraction techniques to observe the now well-known chevron structure which occurs in many ferroelectric devices. It is the chevron structure which is responsible for the zig-zag defects that are commonly observed in SSFLC devices, and the various complex director geometries that can be accommodated in the chevron structure have

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a significant influence on the electro-optic properties of the devices[3]. The effect of an applied electric field on the layer structure in an SSFLC device depends, amongst other things, on its magnitude. An electric field applied across the substrates of an SSFLC device couples with the spontaneous polarization of the liquid crystal to switch the device between two stable states. High electric fields have been shown to alter the out-of-plane tilt of chevron layers irreversibly in order to align the ferroelectric dipoles more closely to the field (so-called chevron to bookshelf switching[4,5,6]). X-ray diffraction has proven to be an invaluable tool for investigating such phenomena directly.

In this paper, we concern ourselves with the influence of relatively low magnitude electric fields on the layer structure in SSFLC devices. The switching in an SSFLC device experiencing low magnitude electric fields is very complex, and has been studied almost exclusively by optical means. Switching within the chevron structure involves overcoming the energy associated with the chevron interface in addition to that associated with the surfaces. At relatively high fields (though still lower than those necessary to induce the chevron to bookshelf transition), the directors above and below the chevron interface can homogeneously decouple and switching occurs without the need for domain wall motion[1]; the directors above and below the chevron interface are considered to rotate around the cone in opposite directions. At lower fields, decoupling is not possible and the switching process is mediated by domains that occur at the chevron interface, nucleated both homogenously and heterogeneously[3]. Stroboscopic experiments have shown that field-dependent stressed states form, which on field removal would return to the original unswitched director configuration[7]. The chevron interface switches as a domain wall passes.

The role of the smectic layers in the switching process is very poorly understood and generally the layers are believed to remain undisturbed during low field switching. Indeed, there are convincing arguments that an elastic response of the layers at low electric field should not be expected[8]. However, models have been proposed[9,10,11] which predict that the low electric fields used in switching may be expected to reversibly flex or reorganise the smectic layers in a chevron structure and there is evidence for such motion from optical[10,12] and x-ray experiments[13,14]. The switching model proposed by Hartmann[9] relies on allowing the layer tilt to vary during director reorientation. It assumes the director to be uniform and planar-oriented everywhere within the device at all times. Switching occurs via a reversible bending of the smectic layers to slightly higher tilt near to the chevron interface, which allows the rotation of the director around the cone without violation of the planar condition. According to the Hartmann model, layers flex to *larger* angles during switching. Gießelmann[10] also proposes a switching model incorporating bend of the smectic layers, for tilted layers with an initially splayed director profile. During field application the splay becomes confined to thin regions close to the surfaces. In the bulk regions between the director remains uniformly oriented (although not necessarily planar) but with a much *reduced* layer tilt. On field reversal director reorientation occurs via a coupled reversible layer movement which superimposes the motion of the director around the cone, the layer tilt decreases to enable the director to move around the cone. Gießelmann presents electro-optic data which fit well to the results of mathematical modelling. He also accepts that an elastic deformation of the smectic layers is unlikely, but points out that a reorganisation of the smectic layers via a shift of the defects (a layer reorganisation) is energetically feasible.

In our previous work[13,14] we reported motion of the smectic layers in a ferroelectric liquid crystal device switched with relatively low electric fields. The layer motion was observed directly through time resolved x-ray scattering studies of the device. The use of an area detector in the experiment allowed us to observe motion in two orthogonal planes. We determined that during the switching of the device the chevron angle changed and at the same time the smectic layers rotated in the plane of the device. Both motions occurred on a 10 microsecond time scale and the angular distortion of the layers in the plane of the device was approximately 1°. Neither the magnitude nor the sign of the change in chevron angle was determined. An argument was presented based on conservation of layer thickness which indicated that the Hartmann model could explain the data presented. In this paper, we report a more detailed study of the motion of the layers during switching in response to a bipolar pulse driving waveform. This driving waveform is different from that employed previously which considered only switching from one driven state to another, though the fields applied and the temperature at which the experiments were carried out are the same. Further, we report here details of the layer structure at angles up to 5° from the chevron angle in the unswitched state, providing a far more detailed picture of the chevron motion during switching than previously presented. Our results are discussed with respect to possible switching mechanisms in SSFLC devices.

EXPERIMENTAL

X-ray diffraction was carried out on the devices as described previously[13] at station 2.1 at the SRS, Daresbury Laboratory, UK[15]. A 1 mm × 1 mm beam of

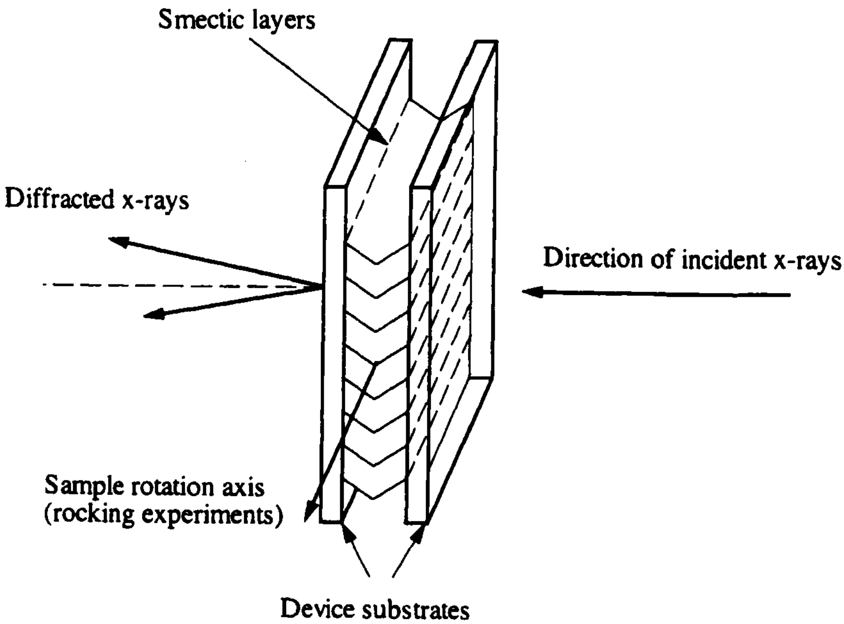

FIGURE 1 A schematic of the experimental arrangement of the experiment, showing the axes of rotation with respect to the liquid crystal device geometry

1.54Å x-rays was incident on FLC devices approximately 3 μm thick. The devices were constructed using 150 μm thick ITO coated glass and were filled with the commercially available ferroelectric liquid crystal, SCE13 (Merck Ltd.). The surfaces of the device were treated with a rubbed polyvinylalcohol (PVA) alignment layer. The device was held in a Linkam heating stage, suspended in a cradle that allowed rotation about the axis of the x-ray beam and an axis perpendicular to it. The device was pre-rotated so that the substrates were perpendicular to the beam and the smectic layers were vertical in the substrate plane, see Figure 1. The heating stage had a temperature stability of ±0.1°C over the mesophase range of SCE13.

The device was cooled slowly to a temperature approximately 5°C below the S_C^* to S_A phase transition at which temperature the layer repeat of SCE13 generates a Bragg angle of $\theta \approx 1.3°$. All further experiments were undertaken at this temperature. A rocking curve experiment was carried out to determine the layer structure in the device prior to any switching experiments, as shown in Figure 2. The diffraction pattern was determined every 0.5° over the range −15° to +15° and the rocking curve in figure 2 deduced from the intensity of the Bragg peaks

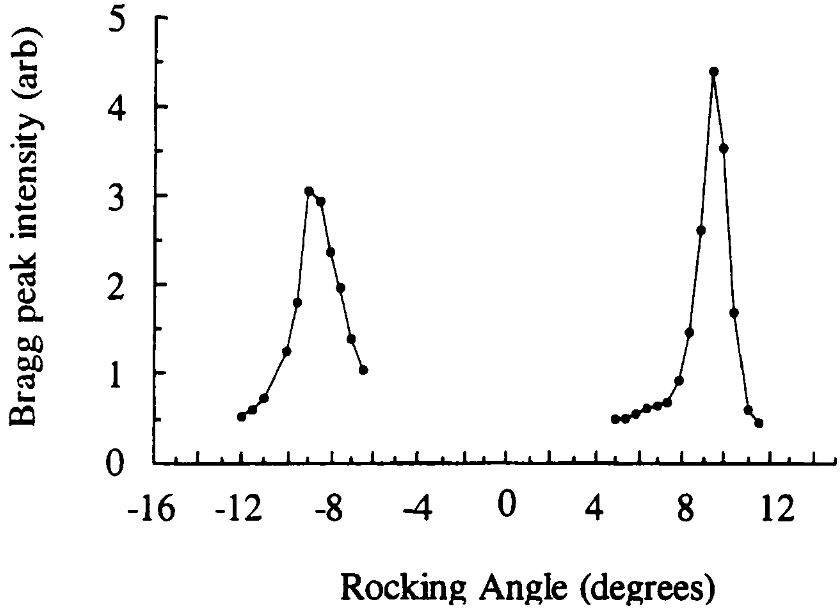

FIGURE 2 A rocking curve of the device prior to switching

at each angle. The figure indicates that there are two symmetric chevron peaks, though we did also observe some bookshelf structure (not shown). Observation of the texture of this device via polarizing microscopy indicated a largely quite well aligned area with a small, poorly aligned domain within the area of incidence of the x-ray beam which we believe was responsible for the bookshelf region. The device was rocked to specific angles for the switching experiments.

All the switching experiments were carried out using a symmetric bipolar pulse of total duration 10 ms and of amplitude $9V_{pp}$. The field across the device (approximately 3 V/μm) is far less than is needed to induce the irreversible chevron to bookshelf transition in this device. The pulse generator was triggered synchronously by the data acquisition system. Data were collected in time slots of 20 μs distributed through the switching cycle and the experiment cycled to give approximately 2 s of data per slot, as indicated schematically in figure 3. This technique clearly assumes that the switching of the device is repeatable over many cycles. A two dimensional area detector was used, providing information on both the layer structure in the substrate plane (the position of the Bragg peak) and the chevron structure (the Bragg peak intensity as a function of rocking angle).

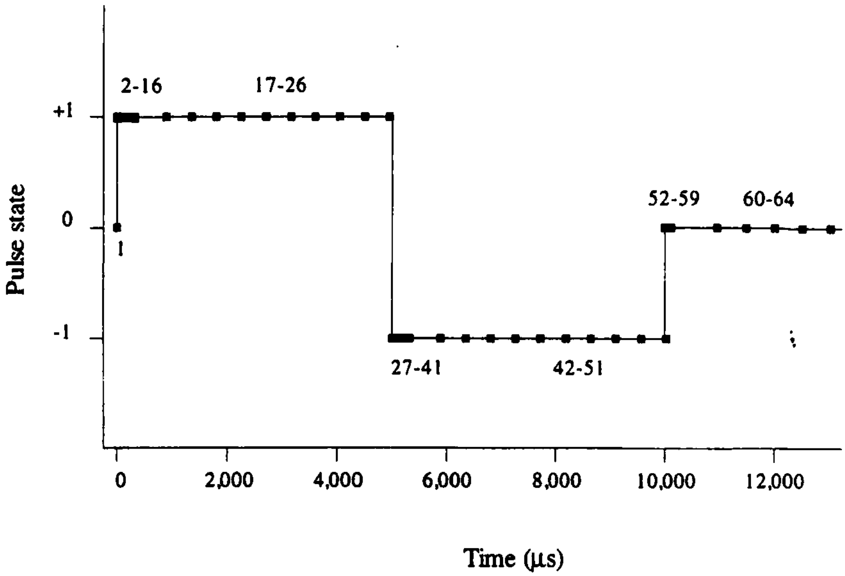

FIGURE 3 A schematic of the timing of the experiment. The points indicate the times at which the 64 frames of 20 μs duration were located. The experiment was repeated and data added to each of the frames until the cumulative data collection time for each data point was approximately 2 seconds. This was the minimum time that gave an acceptable signal to noise ratio on the Bragg peaks

RESULTS

Figure 4 shows the integrated intensity of the Bragg peak as a function of time for the device rotated to the chevron angle of –9° during the application of a symmetric positive-negative bipolar pulse. In figure 4 the intensity of the peak corresponding to layers at the chevron angle (-9°) changes slightly on first application of the positive pulse and then remains approximately constant during the positive region of the pulse. On field reversal there is a large and rapid decrease in intensity, which reaches a minimum after approximately 400 μs. This is followed by a slow rise (over approximately 4 ms), back to the initial intensity. An almost mirror image is seen for the second spot in the Friedel pair, corresponding to layers with tilt of around –6.5° (the two peaks in the Friedel pair differ in orientation angle by twice the Bragg angle). The number of layers in the diffraction condition at –6.5° increases as those contributing to the diffraction at –9° decreases, indicating that the layers flex reversibly to *lower* angles as the field is reversed. Again there is only a slight change in the intensity of the Bragg peaks on removal of the field, the final peak intensities at both –9° and –6.5° being approximately the same as they were prior to the application of the pulse.

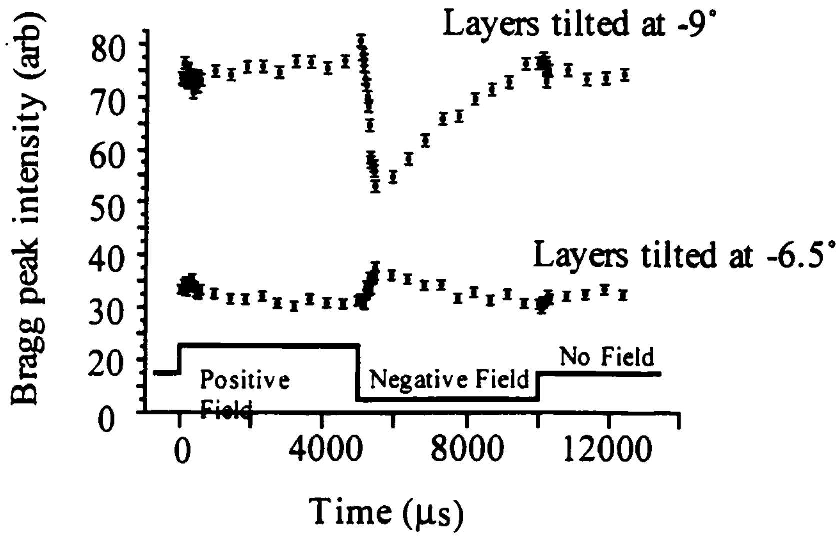

FIGURE 4 Intensity of Bragg peak as a function of time during the $9V_{p\text{-}p}$ bipolar switching pulse. The sample is rocked to −9° and the two sets of information correspond to each of the Friedel pair recorded on the area detector

The experiment was repeated for a range of rocking angles around the negative chevron angle. For angles higher than the field-off chevron angle (i.e. larger than −9.5° in the negative rocking direction), there is no significant change in the already small Bragg peak intensity during the switching cycle. At −10°, there was a larger increase in the Bragg peak intensity on first application of the switching field, not observed at any other rocking angles. The reduction in intensity on field reversal shown for the Bragg peak at the −9° rocking position is observed for angles between −9.5° and −7.5°. The behaviour shown for the −6.5° position in figure 4, where the layer distribution increases on field reversal is seen at angles as low as −5°. The decrease in the intensity of the Bragg peaks on first application of the pulse becomes more marked at low rocking angles.

Care must be taken in making quantitative comparisons between the intensities of the Bragg peaks at different rocking angles during the switching experiment. Each data set took some hours to record and the beam intensity at Daresbury is subject to long term changes, particularly between beam dumps which occurred approximately every 12 hours at the time this experiment was carried out. The data in this experiment are not corrected for fluctuations in the intensity of the incident beam. Thus, although intensity data corresponding to different peaks in the Friedel pair *may* be quantitatively compared (any fluctuation in experimental

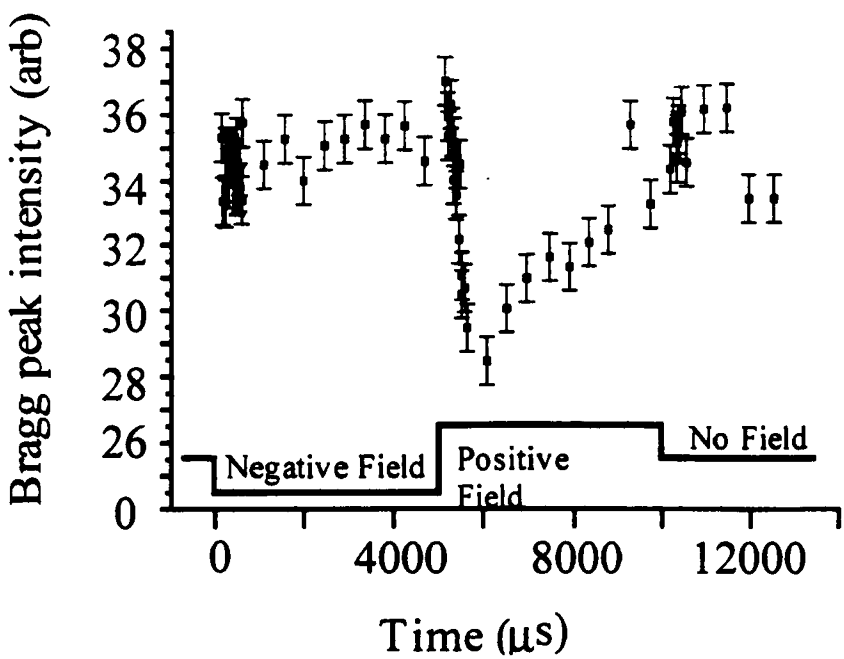

FIGURE 5 The intensity of Bragg peak as a function of time, at a rocking angle of −9°, for a bipolar negative-positive pulse of amplitude $9V_{p\text{-}p}$

conditions applies equally to both data sets), quantitative comparisons between different data sets cannot be made. Having made this comment, it is nonetheless illustrative to compare the data at the two angles shown in figure 4 quantitatively at the point of field reversal. While the −9° data set suffers a rapid decrease in intensity of approximately 25% on field reversal, the −6.5° data sets increases by the same proportion. Thus it seems that the loss in diffraction at higher angles on field reversal is compensated by an increase in diffraction at lower angles.

The experiment was repeated for a pulse of the opposite polarity, as shown in figure 5. The behaviour is very similar to that in Figure 4, which indicates that the rapid layer reorientation to lower angles is associated with field reversal, rather than with the absolute sign of the field.

Markedly different results are shown in Figure 6 for the device rotated to the other, positive, chevron angle of +9.5°. As the field is applied the intensity immediately and rapidly rises, then maintains a roughly constant value for the reminder of this region of the waveform. On field reversal a rapid decrease in intensity, similar to that in Figures 4 and 5 is seen, followed by a further roughly

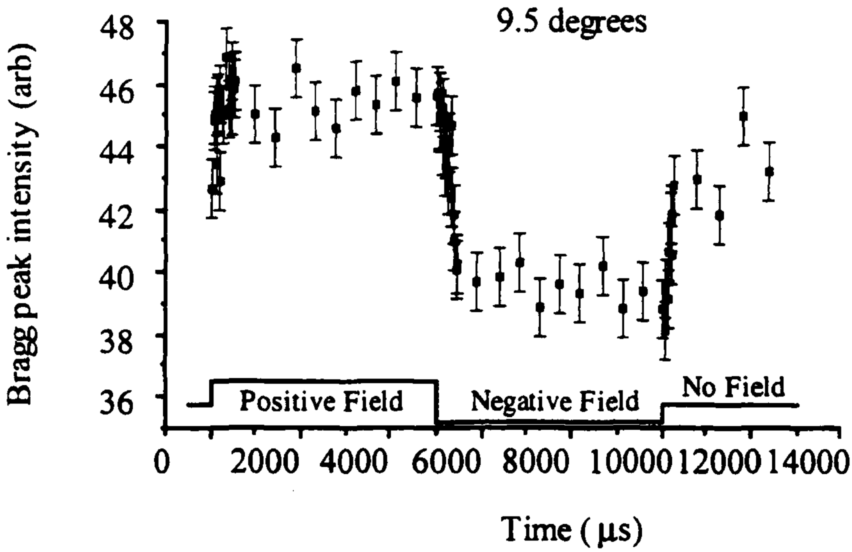

FIGURE 6 The Bragg peak intensity as a function of time for the positive chevron arm at +9.5°, in response to a positive-negative bipolar pulse. These data are taken under the same conditions as those in figure 4, but for the opposite chevron arm

constant region. When the field is removed the intensity quickly returns to approximately the initial value. The experiment was again repeated for a range of angles around the chevron angle to elucidate the response to the negative-positive pulse. Unfortunately, insufficient data were obtained at low angles to draw a direct comparison with the layer motion mechanism clearly shown by the data of figure 4. However, the data gathered at +9°, the lowest angle considered, give some indication of layers moving through that angle as the field is reversed, figure 7.

The use of an area detector allowed us in principle to examine both layer compression/dilation effects and any restructuring of the layers in the surface plane during the switching cycle. The diffraction peaks did not move during of any of the experiments carried out and it can be concluded that the layer spacing remained approximately constant during switching. The resolution of the experiment was such that layer spacing changes due to electroclinic switching could not be observed. The diffraction peaks themselves were quite broad, as might be expected from a sample with less than perfect alignment. There was no discernible change in their intensity distribution during the switching cycle, though the non-ideal alignment meant that it was extremely difficult to observe any such

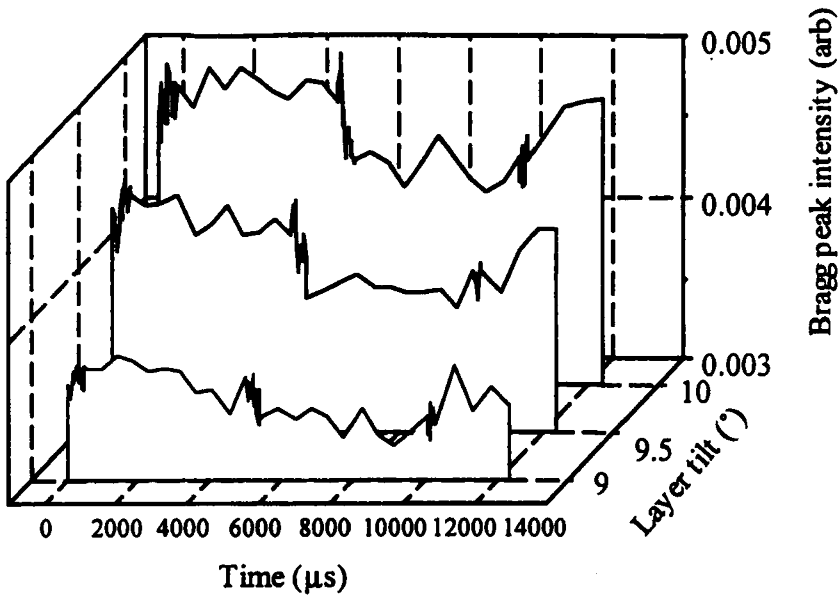

FIGURE 7 The Bragg peak intensity at +10, +9.5 and +9° chevron angles for the positive-negative bipolar pulse

changes. Thus, no conclusions are drawn regarding layer motion in the plane of the device, in contrast to our previous experiments[13,14].

DISCUSSION

In the data presented here an increase (or decrease) in intensity can be taken to indicate that the proportion of layers tilted at an angle has increased (or decreased) through deformation of the initial layer structure. In deforming, the layers may either move co-operatively, and rotate to different tilt angles whilst maintaining a periodic structure, or disruptively where the overall structure is destroyed and the layers move toward random orientation. The dominant mechanism in these results appears to be co-operative layer rotation, as any decrease in intensity at one angle is accompanied by a simultaneous increase in intensity at a different angle of layer tilt (figure 4).

There are two switching points that must be considered for each of the data sets presented; the point at which the switching pulse is first applied (or removed), and that at which the field is reversed. Consider first the change in the intensity of the Bragg peak when the field is first applied. All of the data sets show that at lower angles the Bragg peak intensity decreases, while at higher angles it increases. The effect is more marked for the positive chevron arm; the intensity changes in the negative arm at this switching point are very small. These data can be attributed to two possible phenomena, the electroclinic effect, or switching in device containing a half-splayed director. At the temperature at which the experiment was carried out (~5° away from the S_C^* to S_A transition), electroclinic switching could occur and this would tend to slightly increase the tilt angle of both arms of the chevron on application of the field, as observed. The magnitude of the effect would be comparable in both chevron arms. If the director profile was half-splayed with the splay contained in the positive chevron arm, a slight bending of the layers might result. The applied electric field should unwind the splay and cause the layers to straighten, decreasing the angular width of the chevron peak and increasing its intensity. It is likely that both effects occur in our device, the electroclinic effect being responsible for the a very small intensity increase in both arms on application of the field, and the straightening of the layers in the positive chevron arm causing the much greater intensity change apparent in those data.

We consider next the data at the field reversal point. The range of layer tilts around the negative chevron angle collectively give an almost complete picture of the layer dynamics during switching in this half of the device. The data suggest that the layers tilted at angles around the chevron angle rotate quickly and co-operatively to lower tilt angles on field reversal. At a point ~0.4 ms later the layers then begin to rotate slowly back to their original position, over ~4 ms. The director reorientation which occurs on field reversal in this device was measured optically to be completed in under 0.3 ms, a time scale that corresponds well with the rapid time scale layer motion reported here. Both the direction of the layer reorientation and its large magnitude fit well with the Gießelmann model[10], though the dynamics don't correlate completely. Their simulations indicate a layer "flexing" occurs as the director switches so that the chevron angle reduces to zero, then returns to its equilibrium value. Our data also imply that the layers in the negative chevron arm tilt to lower angles on reversal of the applied field on the same time scale as director switching. Similarly, the layers in the positive chevron arm reorient rapidly, probably to lower angles (unfortunately we could not prove this conclusively due to beam time restrictions on our experiment at the synchcrotron). In both cases this initial layer motion occurs on a similar time scale to the director motion. However, in the negative chevron arm the layers slowly return to their original position (in a few hundred microseconds), while there is no such change in the positive chevron arm.

As already discussed, SSFLC switching generally occurs via the homogenous reorientation of the bulk director to stressed-states, followed by the nucleation and growth of domains, which mediate switching at the chevron interface. These two stages of switching have been shown to have similar response times[16] to the decrease, and subsequent increase of the Bragg peak intensity seen here for the −9.5° data after field reversal in figure 4. A combined domain switching/layer flexing model is therefore proposed, in which the formation of stressed states causes a rotation of the layers to lower tilt angles in the cell bulk, while they remain anchored at the substrates. As a domain wall subsequently passes the stress is relieved and the layers return to their original configuration. At low voltages the domain walls move slowly through the device and several milliseconds elapse before complete coalescence of domains.

This domain switching model does not explain the *lack* of millisecond relaxation observed in the positive chevron arm. Unfortunately, without further data, it is impossible to offer an explanation of this effect. It is possible that the positive chevron arm remains at low angles and that the layers are distorted or rotated in the plane of the device, an effect that was clearly observed in our previous experiment on a better aligned device[13,14]. A shift of the chevron interface, enhancing the negative chevron arm, can be excluded since there is no overall increase in its diffraction intensity. Thus, although the initial decrease in Bragg peak intensity is in accordance with the Gießelmann model, the expected subsequent increase does not occur.

CONCLUSIONS

Small angle x-ray diffraction has been employed to study the layer motion in a ferroelectric liquid crystal device on application of low electric fields. The rocking curve determined prior to the switching experiments indicates that the device initially had an approximately symmetrical chevron layer structure (the peak heights in figure 2 are very similar). We believe that the director profile formed on cooling the sample, prior to application of the field is probably a half-splayed state, as a separate optical experiment confirmed that complete extinction could not be achieved between crossed polarizers.

The x-ray data presented leads us to believe that the splay is contained in the positive angle arm of the chevron. On first application of the field, the major influence is in the positive chevron arm for which the Bragg peak increases in intensity. There is little change in the intensity of Bragg peaks at negative angles.

The rapid flexing of the layers on field reversal appears to occur for both chevron arms on the same time scale as the measured optical response of the device.

Further, the negative arm of the chevron shifts to lower angles by at least 4°, following which the layers slowly return to the original angle. These data are in agreement with a model in which the rapid director switching is accommodated by layer motion to lower angles as proposed by Gießelmann, though the stressed states that form on application of the field relax via domain movement.

The data for the positive chevron arm are more difficult to interpret as they are less complete. However, there is evidence that the reduction in Bragg peak intensity reversal of the field is again due to layer motion to smaller angles. Further experiments are needed to determine the full nature of the layer motion in the positive chevron arm.

We conclude that, irrespective of the model used to describe the layer flexing, even relatively low electric fields cause the layers in ferroelectric devices to move rapidly and reversibly to lower angles by a significant amount. The fact that such motion has been observed directly, using time resolved x-ray scattering techniques, can leave no doubt as to its existence. Layer motion is an intrinsic part of the switching process in ferroelectric liquid crystal devices.

Acknowledgements

Thanks are due to Dr Nick Bowring for the development of analysis software, the EPSRC and GEC for funding and the staff of Daresbury Laboratory station 2.1 for their assistance.

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