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Mechanisms of switching in an antiferroelectric liquid crystal device revealed by time-resolved X-ray scattering

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Time-resolved small angle X-ray scattering has been used to study the electric field-induced motion of the smectic layers in an antiferroelectric liquid crystal on switching between the field-induced ferroelectric states. The intensity and position of the Bragg peak was recorded with 10 μs time resolution across a bipolar switching pulse of period 2 ms. Reversible layer reorganization and motion were observed to occur within the first 500 μs of applying the switching field. The data show that the antiferroelectric liquid crystal switches from one ferroelectric state to the other with an increase in layer spacing of around 0.1 Å, a rotation of the layer normal of 0.5° and a reduction in the angular width of the diffraction peak by 1.7°. Subsequent relaxation of the layers occurs with the characteristic of a van der Pol oscillator. A model is presented that illustrates the form of the rapid and reversible layer motion.

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1. Introduction

Ferroelectric liquid crystal devices [1] are well known to the extent that they are now finding use in commercial applications such as the viewing panels of some cameras. The rapid, tristate switching of antiferroelectric liquid crystals has also generated significant interest in their application to display devices and optical elements such as spatial light modulators, though they have yet to be used commercially [2–4]. Antiferroelectric and ferroelectric liquid crystal phases are closely related. Both are formed from chiral, rod-like molecules arranged in the layered smectic structure with the director tilted with respect to the layer normal. The primary difference in the structures of antiferroelectric and ferroelectric liquid crystals lies in the particular orientation of the director with respect to adjacent layers, the ferroelectric and antiferroelectric phase being synclinic (SmC*) and anticlinic (SmC^*_{AFE}), respectively. On application of a sufficiently large electric field, the antiferroelectric structure is transformed to one of two possible ferroelectric states depending on the polarity of the field and the sign of the material's spontaneous polarization. This paper is concerned with the mechanism of switching between the field-induced ferroelectric states, specifically whether layer motion and reorganization is inherent in the switching process. A question of particular interest is whether the switching mechanism differs from that observed in ferroelectric liquid crystals [5], i.e. whether the antiferroelectric nature of the phase is important even though the system is switching between two field-induced ferroelectric states.

Small angle X-ray scattering has proven to be an invaluable tool in the study of smectic liquid crystals as it allows direct measurement of changes in the layer structure and layer spacing in response to external stimuli such as electric fields. The technique was used extensively in developing an understanding of the layer structures and their field-induced modifications in ferroelectric and antiferroelectric devices. The complex structures adopted in SmC* devices have provided some of the biggest challenges to their understanding and commercial employment. X-ray scattering has revealed the chevron structure that is normally adopted in SmC* [6] and SmC^*_{AFE} [7] liquid crystal devices, as well as providing information on the mechanism whereby a sufficiently high field permanently reorganizes the layers in the so-called bookshelf structure [8]. Time-resolved small angle X-ray studies of SmC* devices have demonstrated that during low-field switching the chevron flexes reversibly and rapidly to lower angles to accommodate director motion [5].

There have been relatively few X-ray studies of SmC^*_{AFE} devices. Most have examined the layer geometry or the field-induced chevron-to-bookshelf transition [9–11], although some resonant X-ray scattering experiments have also been carried out that provide information on field-induced changes to the interlayer

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periodic structures in SmC* subphases [12, 13]. One other report of a time-resolved study of an antiferroelectric device has appeared [14]. Our experiment, uniquely, allows us to observe quantitative changes in layer spacing, and hence tilt, that accommodate the switching mechanism, providing a full picture of how the antiferroelectric liquid crystal device switches from one field-induced ferroelectric state to the other. Our data also allow us to make some specific comments about the relaxation mechanism that is part of the switching process.

2. Experimental

The experiment was carried out at the Synchrotron Radiation Source, Daresbury, UK, where the combination of high X-ray flux and rapid response time area detectors allowed the time-resolved experiment to be performed [5]. The device was constructed with 150 μm thick glass substrates, coated on the inner surfaces with an indium tin oxide electrode and nylon alignment layers (with one substrate rubbed) to promote a monodomain structure in the 20 μm thick liquid crystal layer. A 1mm × 1mm X-ray beam of wavelength 1.54 Å is incident on the device. The material under test has been described in detail elsewhere [7]; it is denoted AS573 and exhibits SmC^*_{AFE}, intermediate, SmC* and SmA phases. Using a temperature-controlled environment (stability $\pm 0.5^\circ$C), the device was held below the antiferroelectric phase transition, approximately 20°C below the SmC*–SmA transition, ensuring that no pretransitional phenomena contribute to the switching mechanisms.

As already mentioned, the time-resolved experiment was designed to study the mechanism associated with ferroelectric to ferroelectric switching in the SmC^*_{AFE} phase. The fields involved were greater than those required to induce the chevron-to-bookshelf transition (approximately 14 V for the device used in our experiment), so the device was in a quasi-bookshelf geometry. Specifically, the layer geometry is such that the layers are approximately perpendicular to the substrates but, as would be expected, a small degree of curvature (usually $\sim$ 5–6° in a well aligned device) is retained. This curvature is a remnant of the focal-conic nature of planar cells. A bipolar switching voltage of 23.5 V and period 2 ms was employed, chosen to accommodate the 10 μs time resolution of the X-ray experiment. The total electro-optic response time of the device under these experimental conditions is around 100 μs and the response comprises two distinct regions: an initial 'delay' of around 40 μs during which time there is effectively no change in optical transmission, followed by a conventional electro-optic response with a 10–90% rise time of around 60 μs.

Data collection was synchronized with the application of the switching voltage, as described previously [5, 15] and shown in figure 1. Data capture periods of 10 μs were employed, distributed throughout the switching cycle to provide optimum detail in the time regions of interest. The experiment was repeated 500 000 times to allow 5 s of data to accumulate in each time frame, gaining acceptable signal-to-noise levels.

The sample was rotated (rocked) with respect to the direction of the incident X-ray beam and data recorded at three rocking angles, providing details about changes in the layer structure through the depth of the device. An area detector was used to collect the data, allowing several types of information to be deduced simultaneously, as described previously in detail [15], summarised below and illustrated in figure 2.

(1) The reorientation of layers in the depth of the device may be examined via the intensity of the Bragg peak at selected rocking angles. The area detector allows two angular positions separated by twice the Bragg angle, corresponding to the $(0\,0 \pm 1)$ peaks, to be studied simultaneously [16]; each peak appears on the equator of the area detector, as shown in figure 2. It is worth noting that both peaks can be observed simultaneously partly because the Bragg angle is small, approximately 1.5° in this experiment, and partly because of the relative disorder of the smectic layers. Application of the electric field has ensured that the smectic layers are in the bookshelf geometry, so in the static state the Bragg peaks are symmetrical

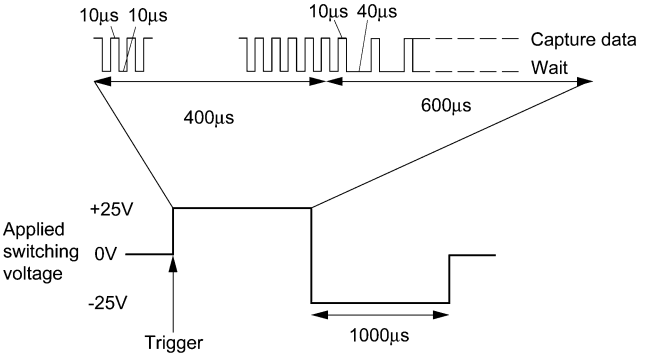

Figure 1. The data capture cycle of the experiment indicating how maximum time resolution was achieved across the switching cycle. The minimum time interval possible for data capture was 10 μs (defined by the apparatus at the SRS, Daresbury), with a 'wait' interval of 10 μs between each capture event. The experiment was repeated 500 000 times, synchronized each time via the external trigger.

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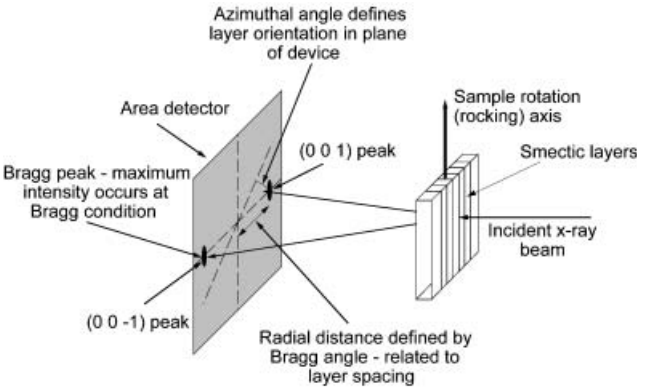

Figure 2. A cartoon illustrating the data that can be obtained from the experiment. Rocking the sample about the axis of rotation provides information about layer motion through the depth of the device, indicated through variations in the intensity of the Bragg peak. A change in position of the peak indicates either a change in layer spacing (radial change) or motion of the layers in the plane of the device (azimuthal rotation of the peak).

about 0°. If the switching field changes the orientation of the smectic layers in the plane of the device, there will be a corresponding change in the intensity of the Bragg peak. Our time-resolved experiment follows such changes through the cycle of the bipolar switching pulse.

(2) Motion of the layers in the plane of the device is apparent from measurement of the angular position of the Bragg peak on the detector, while the angular width of the peak on the detector (analogous to the spread of χ in [10, 14]) provides information about the in-plane distribution of the layers (this can be interpreted as layer curvature). In our experiments, such measurements have an accuracy of $\pm 0.1°$.

(3) Changes in the smectic layer spacing, d_{AFE}, are deduced from measurement of the Bragg angle (the position of the peak on the area detector with respect to the straight-through position), with a relative accuracy of ± 0.02 Å. Associated values of steric tilt angle are calculated using [17] $\cos^{-1}(d_{AFE}/d_{SmA})$, where d_{SmA} is the layer spacing value determined previously for this material using powder pattern experiments at the SmA–SmC* transition.

3. Results

3.1. *Changes in Bragg peak intensity (probing layers through the depth of the device)*

We consider first the changes in the intensity of the $(0\,0 \pm 1)$ Bragg peaks, measured at three rocking positions of the sample. The data are shown for each of the Bragg peaks in figure 3, while the positions studied are indicated schematically on the rocking curves in figure 4. As these diagrams are fairly complicated, it is helpful to describe exactly how they relate to one another. When the sample is rocked to position 1

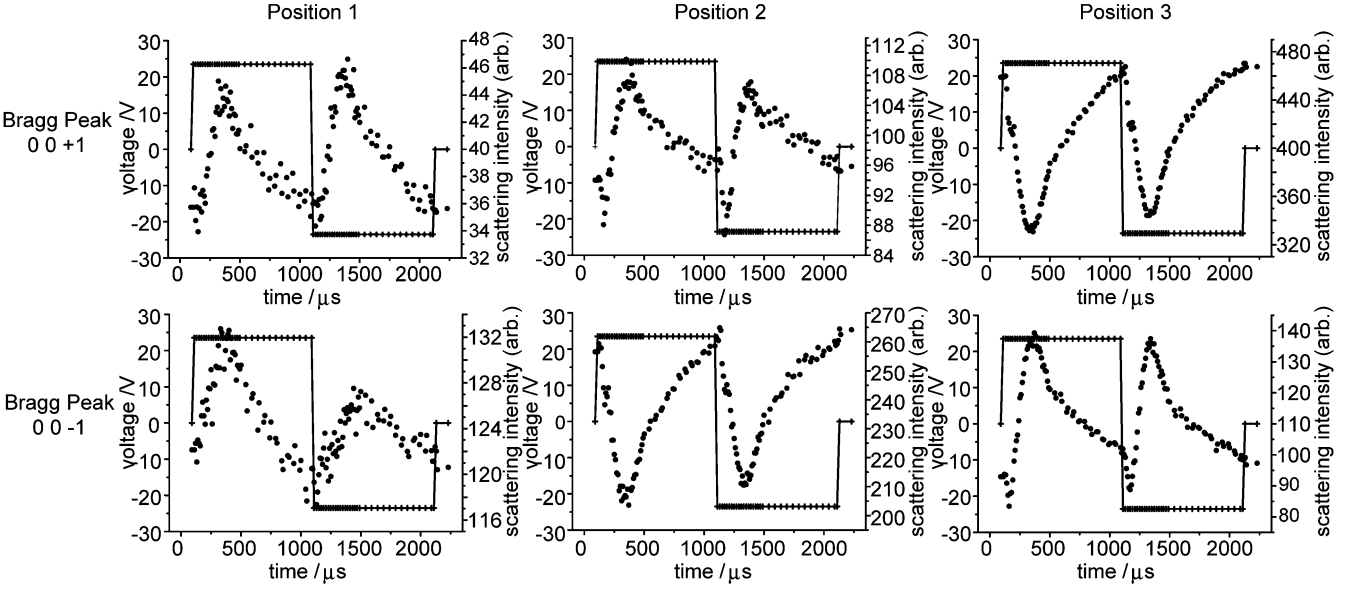

Figure 3. The integrated Bragg peak intensity as a function of time throughout the switching cycle for three different rocking positions 1, 2 and 3 (figure 4). The bipolar pulse employed is also shown, with data capture points indicted by lines on the pulse.

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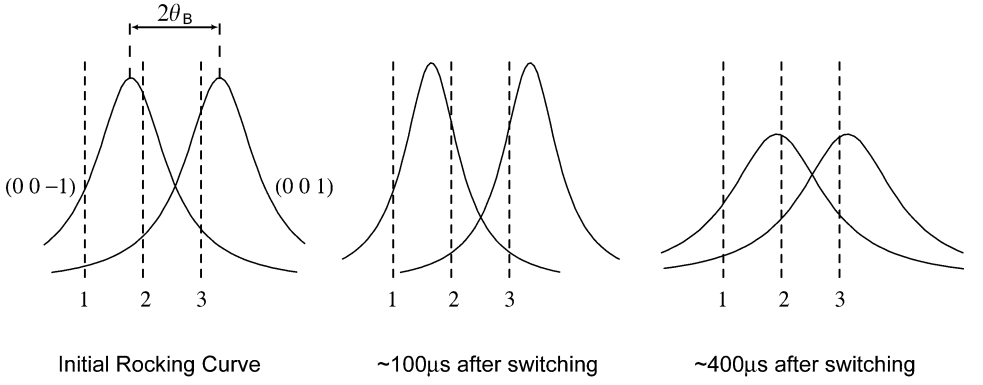

Figure 4. A cartoon illustrating the time dependence of the rocking curves following a voltage change. The two peaks correspond to rocking curves generated by the (0 0 −1) and (0 0 1) Bragg peaks, and are thus separated by twice the Bragg angle, $2\theta_B$. In our experiment, a single rocking angle is selected (position 1, 2 or 3) and the (0 0 ±1) Bragg peaks appear 180° apart on the equator of the detector with different intensities at all angles other than 0°. The intensity data of figure 3 are the quantitative values of intensity for the (0 0 ±1) Bragg peaks. Hence, the cartoon above indicates that initially at position 1 the intensity of the (0 0 −1) peak is much higher than that of the (0 0+1) peak, while the first points in figure 3 at position 1 give the relevant quantitative data.

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shown in figure 4, the initial intensity of the (0 0 −1) peak is larger than that of the (0 0+1) peak. This is shown schematically by the intersection of the line corresponding to position 1 with the rocking curves in figure 4, and quantitatively in figure 3 where the initial Bragg peak intensities of the (0 0 −1) and (0 0+1) peaks are approximately 120 and 36 (in arbitrary units) respectively. Conversely, at position 3 the intensity of the (0 0+1) peak is initially much larger (460 units) than that of the (0 0 −1) peak (~90 units). This again is reflected in the intersection of the line with the Bragg peaks in the initial rocking curve of figure 4. As the switching cycle progresses, the measured intensity of the peaks changes in each of the three positions studied and this is interpreted in terms of changes in the smectic layer structure.

Several features are apparent from the Bragg peak intensity data of figure 3. The initial response to a voltage change exhibits no delay, and rapid changes occur in the first few tens of microseconds, a time scale comparable to the electro-optic response time. It is noteworthy that in contrast to the X-ray data, the electro-optic response shows an effective delay of around 40 μs immediately following the application of a voltage. A longer time scale relaxation follows in the Bragg peak intensity after around 400 μs. Further, changes appear to occur co-operatively, i.e. a reduction in intensity of a peak at one rocking angle is to some extent compensated by an increase at another, indicating that the layers remain largely intact. The data from the three different angles allow a picture to be constructed of the layer motion in the depth of the device. The data are not consistent with a simple shift in the angle of the bookshelf structure through the depth of the device, but are congruent with an initial narrowing of the rocking peaks immediately after switching (within the first 100 μs), followed by broadening, as shown in figure 4, before finally relaxing to the original state. Such data can be interpreted as a rapid decrease in layer curvature in the depth of the device (there are contributions to the Bragg peak from fewer directions), followed by an increase, before relaxation to the initial state.

3.2. *Changes in the angular orientation of the Bragg peak (motion of the layers in the plane of the device)*

As mentioned in point (2) above, our experiment also allows the layer structure in the plane of the device to be examined. Figures 5 (*a*) and 5 (*b*) show the time dependence of the relative angle of the centre of mass of the Bragg peak on the area detector, and its angular width, respectively. Both data sets show that distinct changes occur within the first 100 μs following voltage change. A rapid rotation of the layer normal by 0.5° in the plane of the device (figure 5) occurs at the same time as a reduction in the in-plane layer curvature apparent from a reduction in the angular width of the peaks of approximately 1.7° from the initial value of 5.8°, figure 5.

3.3. *Changes in Bragg angle (steric tilt angle)*

The variation of layer spacing in the device across the switching cycle is shown in figure 6, together with the change in steric tilt angle. There is a 0.1 Å increase in the layer spacing observed when the field is first applied. As explained previously, this change in layer spacing can be associated with a change in the steric tilt angle of

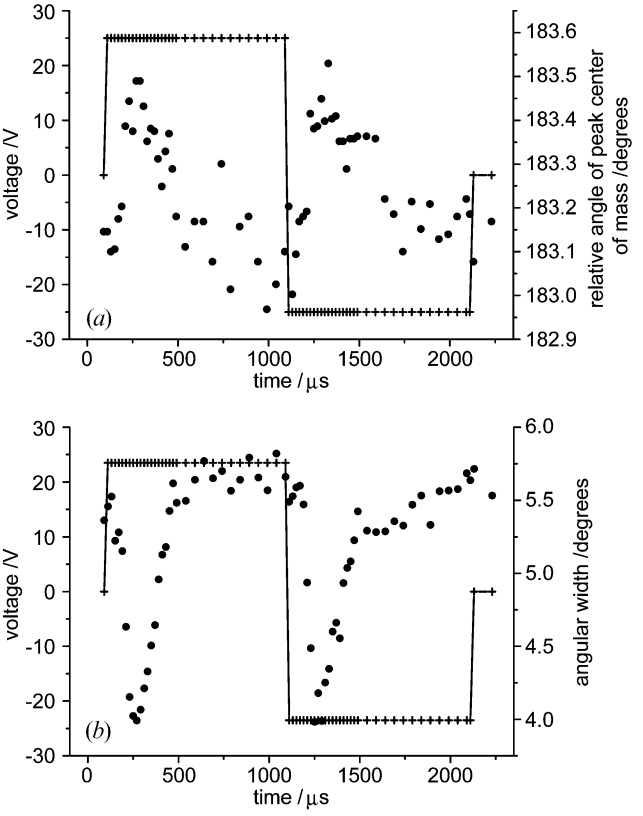

Figure 5. The relative angle of the centre of mass of the Bragg peak on the area, and the change in the angular width of the diffraction peak, as a function of time over the switching cycle.

the system. For this material, using the values noted in point (3) above, it can be seen that a 0.5° decrease in the director tilt angle occurs, corresponding to the field-induced change in layer spacing. Further, the layer spacing is rather temperature-insensitive for this material (enantiomer of AS666 [17]) around the temperature selected for this experiment and an increase in tilt angle is associated with an effective increase in temperature, i.e. a shift towards the ferroelectric phase. Interestingly, the 0.5° change in tilt observed corresponds reasonably well with the change that would be required to shift this system into the ferroelectric state.

However, one should note a possibility that the voltage-induced rotation of the layer normal could effect the position of the Bragg peak. Whilst comparisons between the data from the different rocking positions could confirm or discount this possibility, the signal to noise ratios obtained at the other two positions were not sufficient to provide a definitive answer. It is also worth noting that a small asymmetry is observed in all the data that may be attributed to the fact that either our wave form is slightly asymmetric (there is a short period of zero field before the application of the bipolar pulse that goes positive, but none on the negative switch), or our device is slightly asymmetric (one side only is rubbed).

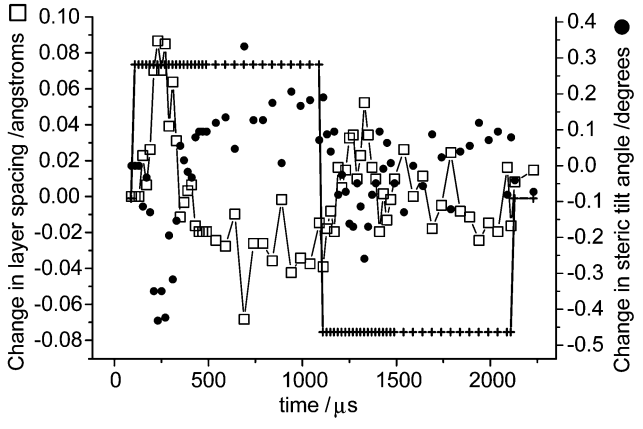

Figure 6. Layer spacing and steric tilt changes during the course of a switching cycle.

4. Discussion

The data can be summarized as follows, providing an insight into the ferroelectric to ferroelectric switching mechanism in the antiferroelectric liquid crystal device. In the first 100 μs following the application of a voltage or field reversal, several changes occur concurrently to the smectic layers. There is no equivalent of the electro-optic 'delay' in the X-ray data, indicating that although there is clearly an immediate response to the field observed through the X-ray scattering from the layers, the initial response has little influence on the director. The overall picture indicates that, in the 100 μs, the layer structure through the depth of the device decreases in curvature. Further, the layer curvature in the plane of the device also decreases, while the layer normal rotates. This initial change in geometry is shown schematically in the middle diagram figure 7. After this initial motion (essentially at the end of the electro-optic response), the layer structure in the depth of the device *increases* in curvature. Conversely, the in-plane curvature continues to decrease and the layer normal continues to rotate, reaching a minimum and maximum, respectively, after approximately 250 μs. During this intermediate time period the changes in layer spacing, and thus steric tilt angle, are also seen to reach their maximum values. After 250 μs, all these motions begin to relax back to their initial positions. However, the layer curvature in the depth of the device continues to increase until that too reaches a maximum value at about 400 μs (shown in the third diagram of figure 7) when it also begins to relax back.

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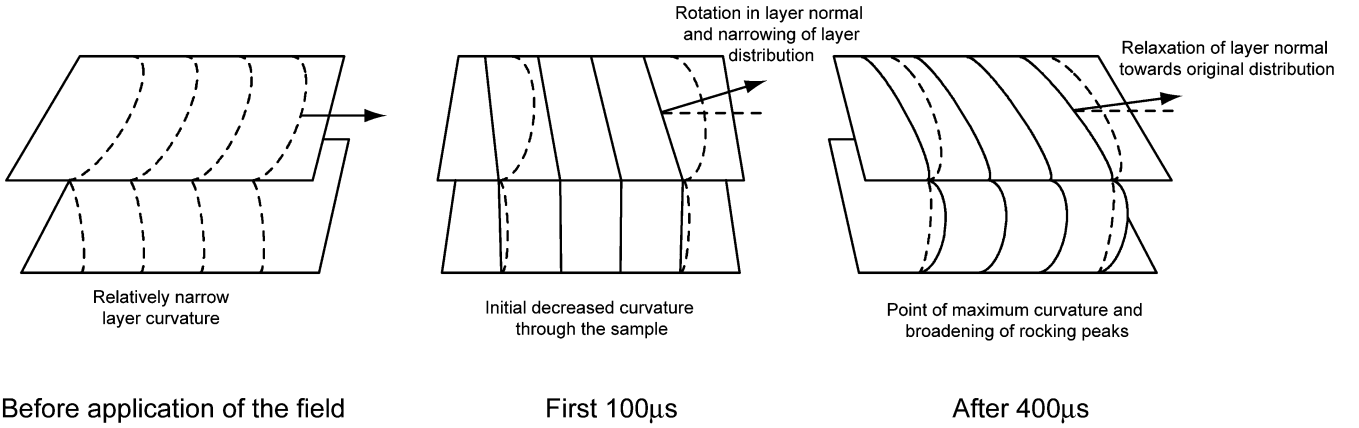

Figure 7. A cartoon illustrating much of the change in layer structure that occurs during a ferroelectric to ferroelectric switching cycle in an antiferroelectric liquid crystal device. The dashed lines on the left hand diagram indicate the initial state. Changes in layer spacing are not shown, and the full relaxation phenomenon occurs on a longer time scale than that indicated here. It should be noted that the cartoon illustrates what is happening on a microscopic domain level and that gross rotation of the layers across a device is not suggested.

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The changes in layer curvature in the depth of the device are apparent to some extent in the data of Takahashi *et al.* [14] for a different device and driving wave form. Reorientation from one ferroelectric state to the other is clearly mediated by a significant change in the layer structure, of quite a different form to that observed in ferroelectric devices [5, 8]. One possible explanation for this very different switching mechanism is that adjacent antiferroelectric layers take different reorientation paths during switching. It is not unreasonable to find that switching in an antiferroelectric liquid crystal device differs from that in a ferroelectric device, even though the switching is from a ferroelectric to a ferroelectric state. The visco-elasticity is expected to be different in the two phases, and the different static responses of the phases to electric fields have been reported [7, 9] and discussed theoretically [18] elsewhere. Nonetheless, a more detailed theoretical description is warranted, now that the physical mechanism, mediated by a change in layer spacing, is clear.

Finally, we note that the graphs have a common feature: a rapid change occurs on application or reversal of the voltage, followed by a smooth and relatively rapid relaxation to the initial state. The time sequences (an initial response and sharp, but smooth relaxation with no evidence of oscillation) are reminiscent of van der Pol systems [19], characterized by non-linear damping terms. This observation may provide a further insight into the visco-elasticity of the antiferroelectric phase, although measurements must be made for different applied voltages and temperatures before it is clear whether the temporal data can be analysed quantitatively to access visco-elastic parameters.

5. Conclusions

Time-resolved X-ray scattering has been used to probe the smectic layer motion during switching between the induced ferroelectric states in an antiferroelectric device held in a field-induced bookshelf geometry. It is clear that there is significant motion of the layers during the switching process, and that this motion is both fast (occurring on a microsecond time scale) and fully reversible. Equally rapid layer reorientation has been observed previously in similar experiments on surface-stabilized ferroelectric devices [5], although then the motion was related to chevron flexing. The 40 μs 'delay' observed in the electro-optic response immediately after the field is applied is not apparent in the X-ray data — layer reorganization occurs immediately after the field is applied or changed. Interestingly, the data for this antiferroelectric device show that, in addition to a reorganization of the layers in both the plane and depth of the device that occurs as part of the switching process, there is a measurable change in layer spacing, not observed previously. The layer spacing change is equivalent to a decrease in tilt angle of around 0.5°. It is tempting to question whether this could be a signature of a 'de-Vries' [20] or electroclinic effect, although the phase structure is clearly antiferroelectric. This explanation can, however, be rejected since the change in layer spacing occurs only on changing the applied field, and is not retained as the field is maintained. It is clearly part of the switching mechanism in the system and perhaps provides information for theoreticians to determine a full picture of switching processes in antiferroelectric liquid crystal devices.

This paper is dedicated to Sven T. Lagerwall at the occasion of his 70th birthday.

Acknowledgements

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