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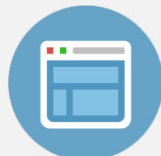
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Asymmetric switching in a ferroelectric liquid crystal device

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The fast multistate switching associated with ferroelectric and antiferroelectric smectic liquid crystals makes them important materials for electro-optic devices, and asymmetric electro-optic responses are of particular interest for some analog switching applications. We report intrinsic asymmetric switching in a ferroelectric liquid crystal device observed using time-resolved small angle x-ray scattering and electro-optic measurements. The experiments reveal a marked variation in the response time depending on field polarity not present in the ferroelectric or antiferroelectric phase. We suggest that this is a consequence of the ferroelectric structure itself and interactions with the device surfaces. © 2008 American Institute of Physics. [DOI: 10.1063/1.3001933]

Liquid crystals are well known for their use in electro-optic devices with ferroelectric and antiferroelectric phases of particular interest because of their fast (microsecond), multistate switching. The most common ferroelectric liquid crystal phase is the chiral smectic-C* (SmC^*) phase¹ in which the rod-shaped molecules exhibit a temperature-dependent tilt angle with respect to the layer normal. An antiferroelectric phase (SmC_A^*) also exists in some materials at lower temperatures and differs from the SmC^* phase in that the tilt direction alternates by $\sim 180^\circ$ from one layer to the next (a two-layer structure). The surface stabilized ferroelectric device,² in which the helical superstructure of the chiral phase is suppressed by the device surfaces, is a typical device geometry. Here, the electro-optic response is equivalent to a rotation of the optic axis of the device through twice the tilt angle and is observed on changing the polarity of the applied electric field. The electro-optic response of the SmC^* and SmC_A^* phases is usually symmetric to fields of opposite polarity, although recently, the ability to induce asymmetric responses has become of increasing importance.^{3,4} Such asymmetry has been obtained by treating the two constraining surfaces of the device differently, or by using specific driving schemes, and has been suggested to have potential applications including inducing analog optical multistability in response to simple bipolar pulses.

Two phases intermediate to the SmC^* and SmC_A^* phases are also known, with structures that have periodicities of three ($\text{SmC}_{\text{FI1}}^*$) or four layers ($\text{SmC}_{\text{FI2}}^*$), and which are ferroelectric and antiferroelectric, respectively. Most of the recent research on the intermediate phases has concentrated on deducing their structures^{5,6} and understanding the factors that control their stability.^{7,8} Since the intermediate phases normally exist over very narrow temperature ranges (2–3 °C), they have not so far been considered as serious candidates for electro-optic devices. This situation has changed recently⁸ with the discovery of materials with intermediate phases stable over as much as 30 °C, opening up a different system for electro-optic switching applications. In this letter we report the asymmetric response of the ferroelectric $\text{SmC}_{\text{FI1}}^*$ phase in a device studied using electro-optics and x-ray scattering. We demonstrate that using this phase offers

a simple way of inducing an asymmetric electro-optic response in a liquid crystal device, which has a potential significance in device design.

The material used here is denoted as AS620 and is described in detail elsewhere.⁹ It exhibits the SmC^* , $\text{SmC}_{\text{FI1}}^*$, and SmC_A^* phases as revealed via resonant-scattering x-ray experiments.¹⁰ The sample was contained in devices with gap thicknesses of about 20 μm , constructed using 150- μm -thick glass substrates that were coated on their inner surfaces with indium tin oxide electrodes and nylon alignment layers. The alignment layers were rubbed on one surface only to obtain excellent monodomain planar alignment of the liquid crystal. The electric-field response of the device was studied using standard electro-optic and polarization measurements employing current reversal methodology,¹ providing information on the field-induced reorientation of the optic axis and the spontaneous polarization (P_s) of the system. Time-resolved small angle x-ray scattering (SAXS) was also employed, probing changes in the layer structure in the device on a microsecond time scale (with a time resolution of 10 μs). Details of the SAXS experiment have been described previously.^{11,12} An advantage of employing three experiments is that the optical and electrical responses can be examined alongside the reorientation of the smectic layers, providing further information about the switching.¹¹

In the absence of an electric field, the samples had a chevron structure caused by buckling of the smectic layers as the molecular tilt increases with lowering temperature.¹³ To make the switching experiments in the different phases comparable, fields were chosen to be above the threshold for the transition from the chevron to bookshelf layer structures at each temperature by approximately the same amount.¹⁴ Switching was between the field-induced ferroelectric states. The response times were chosen to be greater than 50 μs to allow sufficiently detailed information about the switching to be gleaned from the experiments. Consequently, bipolar pulses with amplitudes 0.2 and 0.3 V/ μm above the chevron to bookshelf threshold were chosen for $\text{SmC}_{\text{FI1}}^*$ and SmC_A^* phases, respectively.

The electro-optic response and P_s curves were determined in both phases of interest. The former comprised the well-known “delay” following application or reversal of the field during which time there is no discernable change in the optic axis, followed by a significant response corresponding

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TABLE I. The experimental responses in SmC_A^* and SmC_{FI}^* phases.

	SmC_A^* phase		SmC_{FI}^* phase	
	+ve polarity	-ve polarity	+ve polarity	-ve polarity
Electro-optic delay (μs)	28 ± 3	27 ± 3	27 ± 3	41 ± 3
Electro-optic 10%-90% response (μs)	20 ± 3	24 ± 3	22 ± 3	32 ± 3
Total, $=\tau_{\text{optical}}(\mu\text{s})$	48 ± 3	51 ± 3	50 ± 3	73 ± 3
P_s current pulse height (mA)	4.03 ± 0.05	4.14 ± 0.05	1.72 ± 0.02	1.55 ± 0.02
P_s current pulse FWHM (μs)	22 ± 1	21 ± 1	50 ± 1	54 ± 1

to the rotation of the optic axis by twice the tilt angle. The associated current pulse, which is proportional to the P_s change, was also measured. The data are given in Table I. It is clear that there is a marked asymmetry in the optical and electrical responses in the ferroelectric phase for each different polarity.

For the x-ray experiments, the device was positioned such that the layers satisfied the Bragg condition for the bookshelf geometry, and changes in the intensity and position of the Bragg peak were monitored during the application of the bipolar pulse (Figs. 1 and 2). The data show that the smectic layers are reoriented (Fig. 1) and that the layer spacing changes (Fig. 2) immediately following the application or reversal of the field in both SmC_{FI}^* and SmC_A^* phases. The dynamics of the changes in the smectic layer orientation and spacing during the switching cycle are similar to those described in the SmC_A^* phases of other materials,¹¹ although there are no other reports of dynamic x-ray data for the SmC_{FI}^* phase. A key difference between these data and those reported for the SmC_A^* phase is the marked slowing of the response of the SmC_{FI}^* phase for one field polarity, which is seen to occur by a factor of around 50%. The fact that this difference in the response time can be observed irrespective of the measurement technique implies that there is a clear difference in one of the switching cycles in the SmC_{FI}^* phase, which is not apparent in the antiferroelectric phase. We note that no asymmetry is observed in switching of the ferroelectric phase.

We suggest a qualitative explanation for our observations based on the possibility that an intermediate ferroelectric state can be formed during the switching cycle. First, we note several points that are relevant to our argument:

- (1) In contrast to other work,^{3,4} the entirely symmetric responses we observe in the SmC^* and SmC_A^* phases suggest that the asymmetry in the surface treatment of the device cannot alone be the cause of the asymmetric response in the SmC_{FI}^* phase. Indeed, the surfaces have identical alignment layers, although only one is rubbed.
- (2) We can assume that a chevron structure is not present during switching; the Bragg peak intensity changes (Fig. 1) confirm the retention of a bookshelf structure during switching since the formation of a chevron structure would be accompanied by a very significant reduction in the scattered intensity as the layers rotate away from the Bragg condition (a 1° rotation decreases the scattered intensity by at least a factor of 5). Further, a previous study¹⁴ indicated that the field-induced bookshelf (or quasibookshelf) geometry is retained in this material on removal of the applied field.
- (3) In the absence of an applied field, the molecular orientation in the SmC_{FI}^* phase is in the form of a biaxial distorted-clock structure,⁵ which texture observations confirm is retained, at least to some extent, in the device.

Since the switching response in the SmC_{FI}^* phase is asymmetric, we propose that the formation of an intermediate switching state is favorable only during one half of the switching cycle. We explain how this might occur as follows.

Consider first the possible director configurations that the SmC_{FI}^* phase can adopt in a bookshelf geometry [Figs. 3(a) and 3(b)]. These diagrams show schematically the director orientation of the three successive layers in the structure (in reality these would occur behind one another, not adja-

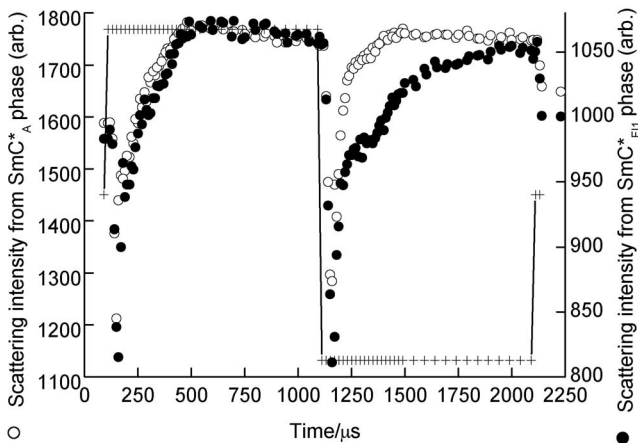


FIG. 1. Integrated intensity of the Bragg peak as a function of time during the application of the bipolar pulse in the SmC_{FI}^* (●) and SmC_A^* (○) phases. The bipolar pulse (2 ms duration) is superimposed on the data (+).

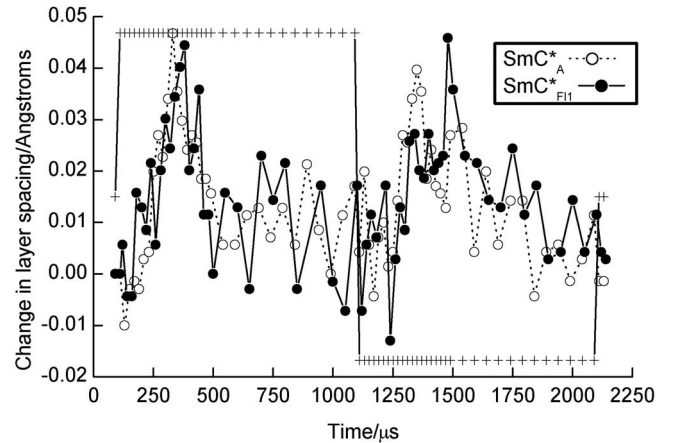


FIG. 2. The change in smectic layer spacing during the switching cycle in the SmC_{FI}^* (●) and SmC_A^* (○) phases. The solid lines are a guide for the eyes.

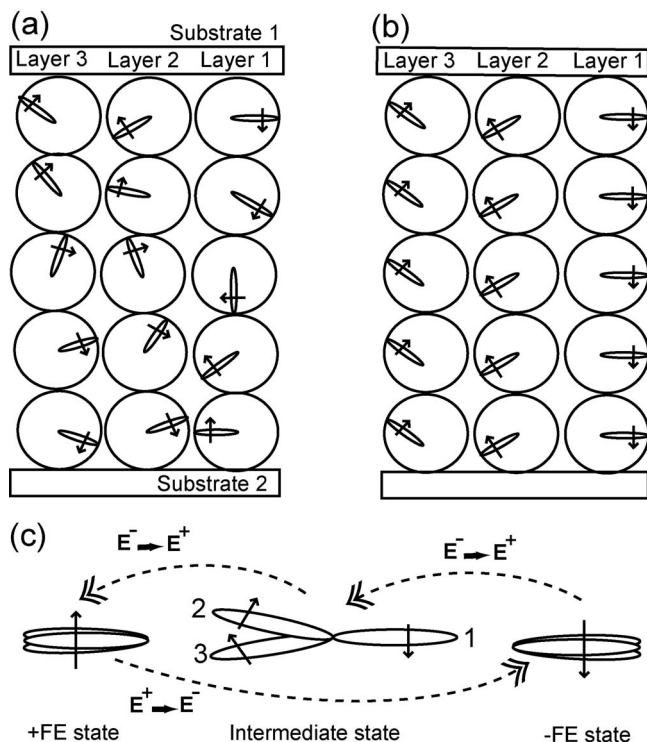


FIG. 3. Schematic illustrations of the repeat unit of the three-layer ferroelectric phase with the sample adopting (a) splayed director orientation and (b) uniform director orientation. The view is along the layer normal with the ellipses representing projections of the director and the arrows representing dipoles. The layers in the three-layer structure would not normally be adjacent but would lie one behind the other. (c) A possible switching mechanism in the ferroelectric phase. The solid arrows again indicate the dipole associated with each layer and the dashed arrows indicate the preferred route during the switching cycle. Note that the total contribution to polarization from the \pm FE states is $\pm P_s$, respectively, and therefore the contribution from the intermediate state shown is $+P_s/3$.

cent as shown) where (a) polar surface anchoring^{1,15} is dominant so that the net P_s points into the surface at each substrate and (b) where this condition is not held. The structure in Fig. 3(a) will form only at the expense of a large elastic distortion within the bulk of the device, and hence is not favored, while the bulk structure in Fig. 3(b) is uniform and so is potentially more stable. We now consider the switching of the system between two ferroelectric states, Fig. 3(c), which also indicates the possible formation of an intermediate three-layer structure. As already discussed, the only possibility for such a structure would be that shown in Fig. 3(b), and we propose that this would only form during the part of the switching cycle where the total P_s has the same sign as the applied field. We note that there are two equivalent versions of the structure in Fig. 3(b), one with the net P_s pointing up (as shown) and the other with P_s pointing down. We suggest that the slight asymmetry in our device (one surface rubbed, the other not) is sufficient to select between one of the two equivalent possibilities. Thus the path with the upper arrows in Fig. 3(c) is selected during one half of the switch-

ing cycle, providing a possible asymmetry in the switching cycle, as the upper path would take longer than the lower path. This proposed intermediate switching state is consistent with the model in Ref. 16, which shows that the three-layer structure is more stable than the SmC* structure at low fields but less stable at higher fields, allowing for the two-stage process that causes the asymmetric response.

In summary, we report a previously unobserved asymmetric response to electric fields in the three-layer intermediate smectic phase. We offer an explanation of this asymmetry in the context of the interaction of the phase structure with the device substrates during switching, such that in one switching direction the system passes through an intermediate state. The asymmetry is primarily a consequence of the structure of the phase itself and serves to amplify an extremely small difference in the construction of the device. This mode of fast switching in liquid crystals offers a different route to inducing an asymmetric response in ferroelectric liquid crystal devices, which is particularly exciting given the advent of wide intermediate phase mixtures.

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