

## Time-resolved sign-dependent switching in a hybrid aligned nematic liquid crystal cell

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**Abstract.** An optical waveguide technique is used to determine the director tilt profile across a hybrid aligned nematic (HAN) liquid crystal cell, in which the optical response is dependent on the sign of the applied voltage. Two physical models are shown that fit the equilibrium experimental data, but with alternative explanations for this sign dependence. Models with either a flexoelectric coefficient of  $2.25 \times 10^{-11} \text{ C m}^{-1}$  or a bound surface charge of  $12.2 \mu\text{C m}^{-2}$  are shown that fit this equilibrium data. In an attempt to resolve this degeneracy sign-dependent switching data are analysed. However, neither model can explain these switching data, which are affected by slow transients of  $\sim 100 \text{ ms}$  which are believed to be due to the motion of free ions in the liquid crystal. From the form of these slow transients, it is suggested that the equilibrium position of the ions is next to a cell substrate.

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## 1. Introduction

Nematic liquid crystals consist of anisotropic ‘rod like’ molecules that tend to locally orient in a direction called the director. They are commonly used for liquid crystal displays (LCDs), in which liquid crystal is typically sandwiched in a layer a few microns thick between two glass substrates, with suitable surface treatments constraining the director at the boundaries to provide the desired director profile across the cell. An appropriate electric field applied across the liquid crystal causes the director to realign, providing a convenient mechanism to control the optical properties of the device. In conventional devices, upon removal of the applied field the director elastically relaxes back to its zero applied field state.

Bistable LCDs, in which there are two stable director profiles without an applied field, offer some advantages over these conventional monostable displays. Firstly, they allow high-resolution multiplexed displays without requiring a thin film transistor (TFT) to control every pixel. Secondly, with infrequently updated images they offer lower power consumption. In some bistable displays, such as the zenithal bistable device (ZBD) [1] and the post-aligned bistable nematic (PABN) [2], the bistability is provided by a micro-structured surface: a blazed grating and posts, respectively. Experimentally, these bistable displays show switching between the two states which is dependent on the sign of the applied voltage.

Flexoelectricity [3], a characteristic of asymmetric molecules with a permanent dipole, has been suggested as a possible mechanism for this switching in both these devices. In a simplified one-dimensional (1D) model, numerical studies have shown bistable switching can result from flexoelectricity, given a large flexoelectric coefficient [4]. Further models have also shown the role flexoelectricity plays in switching ZBDs [5]–[7]. Later experimental work on ZBDs shows that the bistable switching at the grating is independent of the alignment condition at the opposite substrate [8]. This implies that the ZBD switching mechanism is controlled by some surface polarization of the grating [9], which Jones *et al* suggest is governed by flexoelectric, order electric, dielectric and ionic effects.

Two mechanisms for surface polarization in liquid crystals are discussed by Sonin [10]. Firstly, ionic adsorption, in which ions of one polarity are preferentially adsorbed onto a surface from the bulk liquid crystal, to give a sheet of surface charge. Secondly, preferential alignment of dipolar liquid crystal molecules at a surface that gives homeotropic (perpendicular) director alignment.

A variety of experimental studies into sign-dependent switching in liquid crystals have been performed [11, 12], often to measure the flexoelectric coefficient. Many of these have examined the hybrid aligned nematic (HAN) cell, in which one substrate is treated so that it aligns the director planar (parallel), with the other substrate treated to give homeotropic alignment. We study a HAN cell using the fully leaky guided mode technique [13], which allows the measurement of the director profile in the cell.

Similar techniques have been used to study sign-dependent effects in HAN cells previously [11, 12]. In both these studies, the authors considered director profiles once the cell had reached static equilibrium; the switching dynamics were not measured. Both of these papers attribute the observed sign-dependent effect to a combination of flexoelectricity and surface polarization. In section 4, we present results from a similar study of the static director profiles for our cell. However, as we discuss more fully below, we find that the static director profiles measured do not allow us to distinguish between flexoelectricity and surface polarization as the sign-dependent mechanism.

Therefore, in section 5 we analyse some further data, taken during the switching process, which allows us to work towards resolving this degeneracy presented in section 4. These time-resolved director profiles reveal a significant difference in switch-on depending on the sign of the applied voltage and also show switching occurs on slower timescales than expected. This switching could not be explained by including Ericksen–Leslie dynamic theory [14] in the sign-dependent model used to successfully describe the static data. We make the assumption that this ‘slow’ switching is due to free-ion motion in the cell and finally suggest, based on the sign dependence of the switching profiles, that the 0 V equilibrium position of such ions must be next to a cell substrate.

## 2. Theory

Throughout a HAN cell the director,  $\underline{n}$ , may be considered to be parallel to the  $x$ – $z$ -plane. It can therefore be described by a single angle, the tilt angle  $\theta(z)$ , measured from the  $z$ -axis. We define  $d$  as the thickness of the cell and assume that there are no space charges within the cell, and that a surface charge density,  $\sigma$ , is bound to the homeotropic surface (at  $z = 0$ ). A dc potential of  $V$  volts is applied across the cell, such that the electric field,  $\underline{E} = (0, 0, E)$ , is parallel to the  $z$ -axis. Given these assumptions,  $\theta$  can be found by solving the Euler–Lagrange equation:

$$\begin{aligned} & (k_{11} \sin^2 \theta + k_{33} \cos^2 \theta) \frac{\partial^2 \theta}{\partial z^2} + \frac{1}{2} (k_{11} - k_{33}) \sin 2\theta \left( \frac{\partial \theta}{\partial z} \right)^2 \\ & + \frac{1}{2} \sin 2\theta \left( (e_s - e_b) \frac{\partial E}{\partial z} - \varepsilon_0 (\varepsilon_{\parallel} - \varepsilon_{\perp}) E^2 \right) = 0, \end{aligned} \quad (1)$$

self-consistently with Gauss’ law for  $E$ . Here  $k_{11}$  and  $k_{33}$  are the splay and bend elastic constants,  $\varepsilon$  refers to the relative dielectric permittivity (with subscripts denoting parallel and perpendicular components) and  $(e_s - e_b)$  is the flexoelectric coefficient (with the subscripts denoting splay and bend, respectively), adopting the Rudquist sign convention [15]. We split  $E$  into two parts, the first due to the applied field and the flexoelectric polarization and the second due to surface charges

$$E = -\frac{\partial v}{\partial z} + \frac{\sigma}{2\varepsilon}. \quad (2)$$

Only the first term, which defines a partial electric potential,  $v$ , appears in Gauss’ law

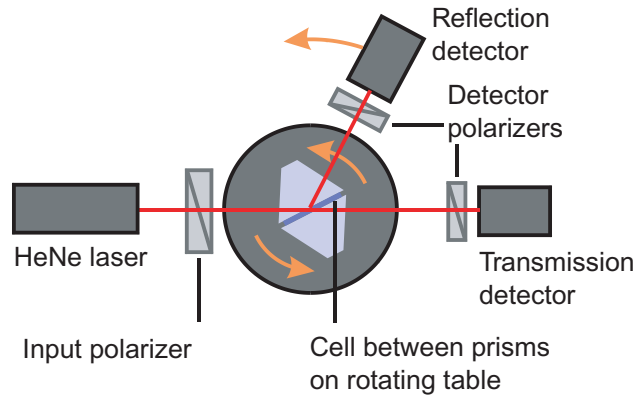
$$\frac{\partial}{\partial z} \left( -\varepsilon \frac{\partial v}{\partial z} + (e_s - e_b) \sin 2\theta \frac{\partial \theta}{\partial z} \right) = 0, \quad (3)$$

because the second term contributes only a constant,  $\sigma$ , to the electric displacement. Then  $v$  satisfies (3) where the local relative permittivity

$$\varepsilon = \varepsilon_0 \left( (\varepsilon_{\parallel} - \varepsilon_{\perp}) \cos^2 \theta + \varepsilon_{\perp} \right) \quad (4)$$

depends upon  $\theta(z)$ . We solve these equations numerically, together with the boundary conditions  $\theta(0) = \theta_0$ ,  $\theta(d) = \theta_d$ ,  $v(0) = V$  and  $v(d) = 0$ . Dependence on the sign of  $V$  (and thus  $E$ ) enters these equations through either the flexoelectric effect or surface charges. If  $(e_s - e_b)$  is nonzero, then  $E$ , and hence the gradient of  $E$ , will depend upon the sign of  $E$ . As a result, the term

$$(e_s - e_b) \frac{\partial E}{\partial z} \quad (5)$$



**Figure 1.** The fully leaky guided mode technique.

may be either negative or positive, and so reinforce or oppose the dielectric term

$$-\varepsilon_0 (\varepsilon_{\parallel} - \varepsilon_{\perp}) E^2, \quad (6)$$

which is always positive for negative dielectric anisotropy materials.

On the other hand, if  $\sigma$  is nonzero, the total electric field may be enhanced (when  $\sigma$  and  $V$  act in the same sense) or reduced (otherwise).

### 3. Experiment

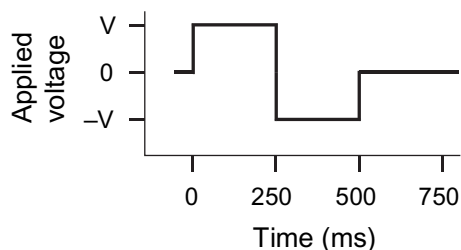
The HAN cell studied consists of two ITO-coated glass substrates separated by microspheres to form a cell gap,  $d$ , of three microns. One substrate was treated with a commercially available polyimide, Nissan 150, rubbed to promote planar alignment. The other substrate was treated with a proprietary Merck treatment to promote homeotropic alignment. A negative dielectric anisotropy nematic liquid crystal, ZLI 4788-000 [Merck], was capillary filled into the cell gap at 95 °C, a temperature above the nematic-isotropic transition. This liquid crystal is of particular interest as is known to switch in a PABN device [2].

Many detailed experimental studies of equilibrium director profiles have been performed using the fully leaky guided mode technique [13]. A more recent development of this technique allows time-resolved measurements to study the cell switching [16]. As shown in figure 1, the cell is optically matched between the two glass prisms. This cell and prism assembly is mounted on a rotating turntable such that a collimated laser beam is incident upon the homeotropic surface of the cell. As the table is rotated and a switching voltage is applied to the cell, eight sets of optical intensities are recorded

$$R_{pp}, R_{ps}, R_{sp}, R_{ss}, T_{pp}, T_{ps}, T_{sp}, T_{ss}.$$

Here, R and T refer to the intensity of light reflected and transmitted, respectively, whereas the first and second subscript letters describe the orientation of the input and detector polarizers, respectively. Thus the recorded experimental data consist of time-dependent intensities as a function of angle-of-incidence.

To interpret the experimental data, we use a multilayer optical model of the cell, based on the Berreman  $4 \times 4$  matrix approach [17]. The director profile of the cell may be deduced by



**Figure 2.** Voltage waveform applied to the cell.

dividing the modelled liquid crystal into many sublayers, and then adjusting the parameters in the optical model until the output fits the data satisfactorily. Alternatively, we can find values of  $k_{11}$ ,  $e_s - e_b$ ,  $\sigma$ , and so on that, once (1) and (2) are solved and the resulting reflection and transmission coefficients computed, minimize the discrepancy between model and experiment. Recent work describes the challenges and progress with this fitting process [18, 19].

As with previous studies [11, 12], a square wave voltage was applied to the cell, of low enough frequency to be considered ‘dc’. A slightly different voltage form was used in this study, the bipolar pulse shown in figure 2. Eight voltages were applied: 1, 2, 3 and 8 V, with both positive and negative polarity, with the planar substrate held at ground. We define the polarity of the bipolar pulse as the polarity of the first half of the pulse.

In the static results section below, only data at 249 ms for  $\pm 2$ ,  $\pm 3$  and  $\pm 8$  V bipolar pulses are considered. We neglect the  $\pm 1$  V data, as it has not reached equilibrium during this time. Previous work [11, 12] has only considered such equilibrium data; in section 5, later in this paper we analyse the switching during the bipolar voltage pulses.

#### 4. Static results

The cell used in this study had already been well characterized for an applied ac voltage ( $3 V_{\text{rms}}$  10 kHz sine wave), following the method detailed by Cornford *et al* [19], by utilizing a Ericksen–Leslie physical model [14] to describe the director profile. Thus many physical parameters of the cell were well known, as shown in table 1; these were then used as the starting point for the dc fits.

However, these ac data were fitted using a model assuming no flexoelectricity or surface polarization. Flexoelectricity is known to have a minor effect on ac director profiles, which can effectively be compensated for by small adjustments of other materials parameters: Brown and Mottram [20] highlight the degeneracy between the elastic constant  $k_{33}$  and flexoelectricity. Therefore, when fitting the dc experimental data, the elastic constants and dielectric anisotropy were allowed to vary slightly from their values determined from the ac data; the final fitted values are given in table 2. All other parameters were locked at the values determined from ac fitting as listed in table 1.

Two fits to the experimental data were begun, each with different starting conditions:

- (a) Surface polarization,  $\sigma$ , variation only; flexoelectric coefficient locked to zero.
- (b) Flexoelectric coefficient,  $(e_s - e_b)$ , variation only; surface polarization locked to zero.

**Table 1.** Parameters determined from fitting ac data with a physical model, adopting the Helfrich notation [14] for shear viscosities.

Liquid crystal parameter	Parameters from ac fit
Refractive indices	
Perpendicular	1.490
Parallel	1.656
Thickness	3.016 $\mu\text{m}$
Splay elastic constant, $k_{11}$	13.7 pN
Bend elastic constant, $k_{33}$	21.1 pN
Elastic ratio, $k_{33}/k_{11}$	1.54
Parallel permittivity, $\varepsilon_{\parallel}$	5.5
Perpendicular permittivity, $\varepsilon_{\perp}$	10.3
Dielectric anisotropy, $\Delta\varepsilon = \varepsilon_{\parallel} - \varepsilon_{\perp}$	-4.8
Rotational viscosity, $\gamma_1$	367 $\text{mPa s}^{-1}$
Shear viscosity, $\eta_1$	353 $\text{mPa s}^{-1}$
Shear viscosity, $\eta_2$	8 $\text{mPa s}^{-1}$
Shear viscosity, $\eta_{12}$	-10 $\text{mPa s}^{-1}$

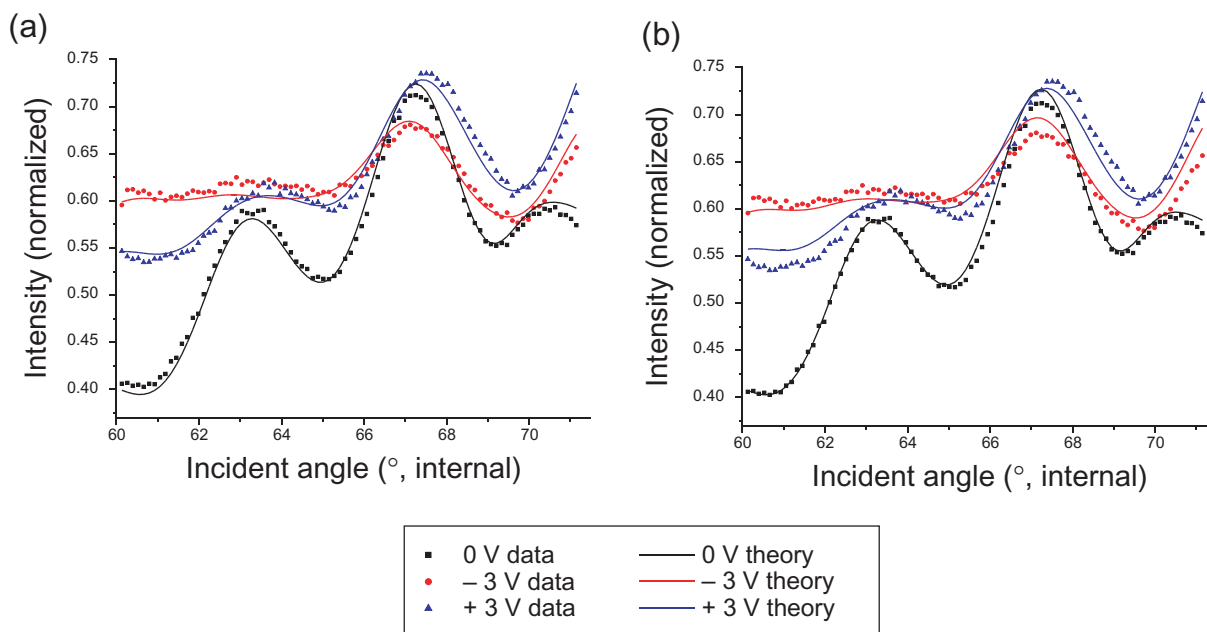
**Table 2.** Parameters determined from fitting equilibrium dc data with a physical model.

Model	Flexoelectric ( $\times 10^{-11} \text{ C m}^{-1}$ )	Surface charge ( $\mu\text{C m}^{-2}$ )	$k_{11}$ (pN)	$k_{33}$ (pN)	$\Delta\varepsilon$
a	0 (locked)	12.2	13.8	21.3	-4.77
b	2.25	0 (locked)	13.5	20.7	-4.92

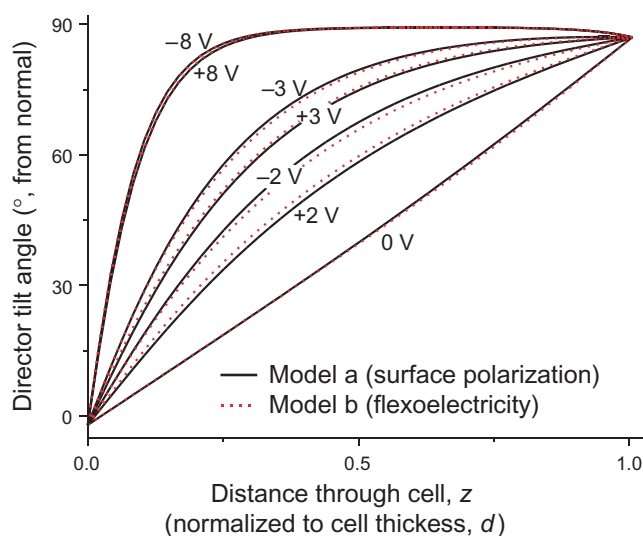
Fits to the experimental data from both physical models, as the examples in figure 3 show, are good. Fits to model b (flexoelectricity only) are visibly slightly worse than model a (surface charge only). However, the difference is rather small and ruling out the flexoelectric model based on these fits cannot be justified.

Figure 4 shows the director profiles resulting from the two physical model fits. In both models, the director profiles are comparable, showing clear sign dependence at  $\pm 2$  and  $\pm 3$  V and much smaller sign dependence at  $\pm 8$  V. But while the director profiles are similar between the two fitted models, differing by  $\approx 2^\circ$  at most, the modelled sign-dependent physical mechanisms are different. From this static experimental data, it does not appear possible to distinguish between flexoelectricity and surface polarization.

Further modelling shows that it is possible, not too surprisingly, to fit the experimental data using a model that combines both flexoelectricity and surface polarization. There are many possible combinations of flexoelectricity and surface polarization that have director profiles that are the same to within small fractions of a degree, and hence give almost identical optical responses. For example, a flexoelectric coefficient of  $2.15 \times 10^{-11} \text{ C m}^{-1}$  and a surface charge of  $3.0 \mu\text{C m}^{-2}$  fits the experimental data as well as the models shown in figure 4. It is even possible to explain the data with a slightly negative flexoelectric coefficient ( $-0.5 \times 10^{-11} \text{ C m}^{-1}$ ) that is compensated for by a larger surface charge value ( $14.4 \mu\text{C m}^{-2}$ ).



**Figure 3.** (a) Surface polarization and (b) flexoelectricity model fits (lines) to  $T_{ps}$  experimental data (points) for three applied voltages.



**Figure 4.** Fitted director profiles for surface polarization (solid) and flexoelectric (dots) physical models, for varying applied voltages.

Mazulla *et al* [11] and Jewell *et al* [12] have previously made measurements of sign-dependent effects using similar optical experiments, with HAN cells filled with 5CB and E7 respectively. In both cases, they argued that the observed sign dependence of the optics was caused by a combination of flexoelectricity and surface polarization, as flexoelectricity alone was unable to account for the magnitude and shape of the observed sign-dependent director profiles. In both papers, the influence of mobile ions is argued to be negligible.

Both papers show that flexoelectricity alone cannot explain their experimental data; Mazulla *et al* comment that a model with an intermediate flexoelectricity coefficient,  $3 \times 10^{-11} \text{ C m}^{-1}$ , did not show sufficient difference between director profiles of opposite voltage polarity to explain their experimental observations. Both comment that the director profiles become ‘s’ shaped if the flexoelectric coefficient is made large ( $\geq 5 \times 10^{-11} \text{ C m}^{-1}$ ) and that this is not seen experimentally. Hence, to explain their data, they add surface polarization to the model and assume a rather smaller flexoelectric coefficient of about  $1 \times 10^{-11} \text{ C m}^{-1}$ .

However, modelling shows that the ‘s’ shape observed in the 0 V profile with larger values of flexoelectricity is damped when free ions are included in the model, while essentially maintaining the separation of the  $\pm V$  profiles [21]. Free ions are likely to be present in small concentrations in any cell, so it may be possible that a model including both ions and flexoelectricity can explain their data, without requiring surface polarization. Alternatively, it would be interesting to see if their data could be matched by a surface polarization only model; neither paper comments on this possibility.

## 5. Dynamic results

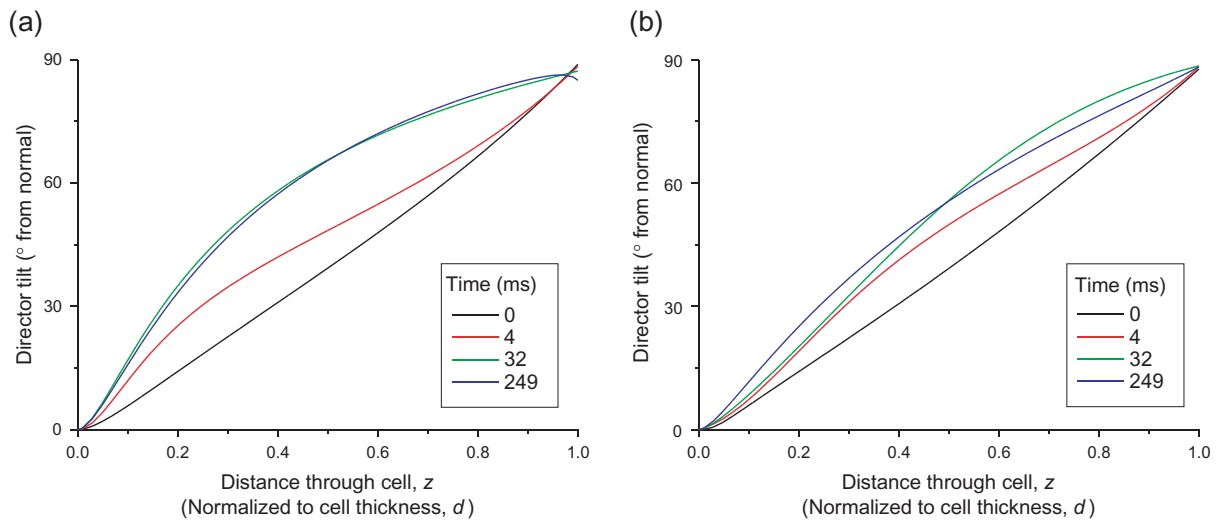
To fit the switching data a model was developed based on an Ericksen–Leslie dynamic model [14, 19] combined with the sign-dependent terms (flexoelectricity and surface polarization) as described in section 2 above. However, despite considerable effort and allowing a very large variation in parameters, the model never gave a satisfactory fit to the experimental data. This was unexpected: the Ericksen–Leslie model provided a good explanation of ac switching data [19] and as seen above the sign-dependent model was able to fit the equilibrium dc data. The failure of this model to fit the data was due to ‘slow’ transients observed experimentally—discussed further below—which were not evident with a large enough amplitude or over a long enough timescale in the model, even when the model included flow.

Therefore in this section, we present results in which the experimental data have been fitted using a mathematical function—in this case, a 3rd order rational Bezier function—to describe the director profile, rather than a physical model. This yields an ‘image’ of the director profiles, allowing the switching process to be explored without requiring a physical model. A future paper will analyse this data with the aid of a suitable physical model [22].

By using a Bezier function description of the director profile, very good fits to the experimental data were obtained—comparable to the physical model fits shown in figure 3. At least 46 time steps were fitted for each bipolar voltage pulse, with times concentrated after changes in applied voltage. This analysis yielded unexpectedly complex director profiles, examples of which are shown in figure 5. Further details are shown by Taphouse [21].

There is a notable difference between the switch-on from 0 V to negative and positive voltages. In the switch-on to  $-2 \text{ V}$ , shown in figure 5(a), by 32 ms the director has essentially reached the 249 ms equilibrium profile. At 4 ms, the director profile is switching most rapidly near  $z = 0.25d$ . This characteristic profile shape is similar to that observed during the ac switch-on [19, 21], and is caused by flow. Without flow the director in this region of the cell would switch more slowly.

Conversely, the switch-on to  $+2 \text{ V}$  behaves very differently, as shown in figure 5(b). After 32 ms of switching, the director profile has not reached the equilibrium 249 ms position. At  $z = 0.2d$ , the 32 ms director tilt angle is only slightly larger than at 4 ms; the cell is switching

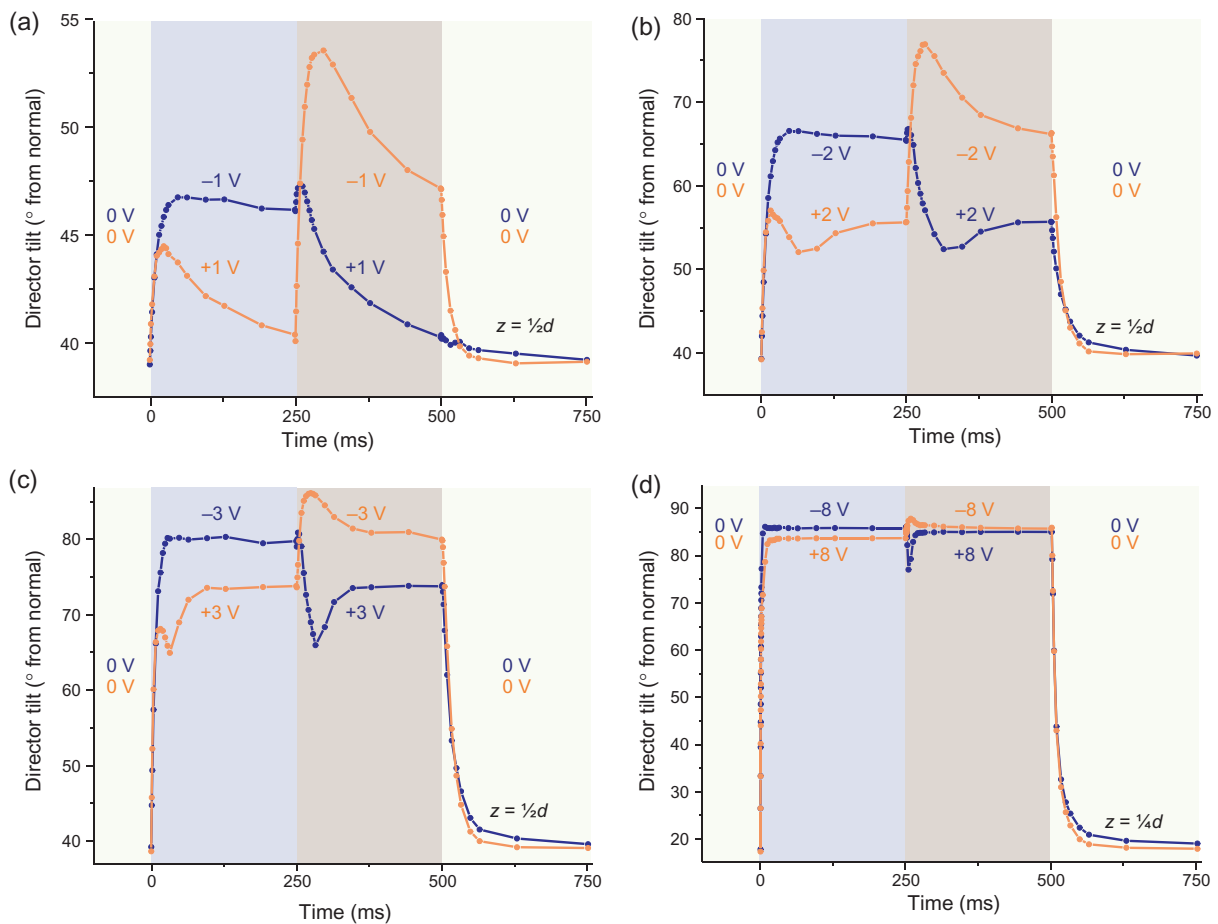


**Figure 5.** Director profiles during (a) switch-on to  $-2$  V from  $0$  V and (b) switch-on to  $+2$  V from  $0$  V.

extremely slowly in this region between these times. Whereas at  $z = 0.8d$ , the 32 ms tilt is already larger than the equilibrium 249 ms tilt angle; here the tilt appears to ‘switch-off’ after 32 ms, as the tilt angle unexpectedly decreases to the 249 ms value even though the voltage is still being applied. This switch-on to  $+2$  V is unlike that observed for ac switch-on [19], in which the director profiles switch-on with comparable shape and rate to those shown in figure 5(a) for switch-on to  $-2$  V.

Figure 6 details the tilt during switching, for all eight applied bipolar pulses, at  $z = 0.5d$  for (a)–(c) and  $z = 0.25d$  for (d). When examined with figure 5 (equivalent graphs for the other voltages shown in [21]), the switching behaviour can be summarized:

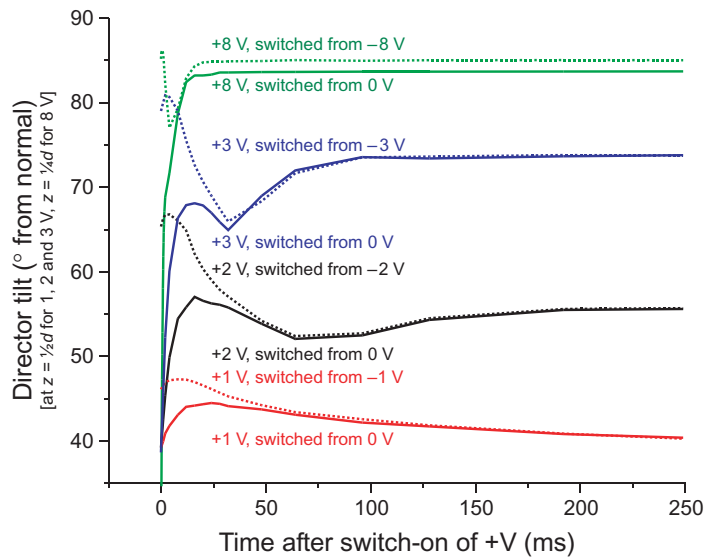
1. Equilibrium negative voltages produce more planar director profiles than positive voltages, irrespective of whether the negative voltage was applied before or after the positive voltage.
2. For all voltages, the shape and rate of the switch-on director profiles from  $0$  V to  $-V$  are comparable with ac switch-on studies [19, 21]; tilt angles increase monotonically with time, with the faster switch-on towards the homeotropic end (near  $z = 0.25d$ ) providing evidence of flow.
3. For all voltages, the switch-on from  $0$  V to  $+V$  was anomalous compared with the ac switch-on. This is most apparent from the distinctive dip observed in the switch-on from  $0$  V to both 2 and 3 V, shown in figure 6(b) and (c). Switch-on of the director profile is rapid over the first  $\approx 30$  ms, after which the director profile starts ‘switching off’, (i.e. becoming more like the  $0$  V profile) even though a voltage is still being applied. With  $+2$  V and  $+3$  V, after this ‘switching off’ period, the director profile then continues to ‘switch-on’ i.e. become more planar with time. Switching from  $0$  to  $+V$  is clearly slower than from  $0$  to  $-V$ : for  $+1$  V, switching was still occurring after 250 ms, compared with  $\approx 64$  ms to reach equilibrium with  $-1$  V applied.
4. When the polarity of the applied voltage is reversed, non-monotonic behaviour of tilt angle with time is observed regardless of the sign of the first half of the bipolar pulse. The time constant associated with this switching behaviour decreases with increasing voltage.



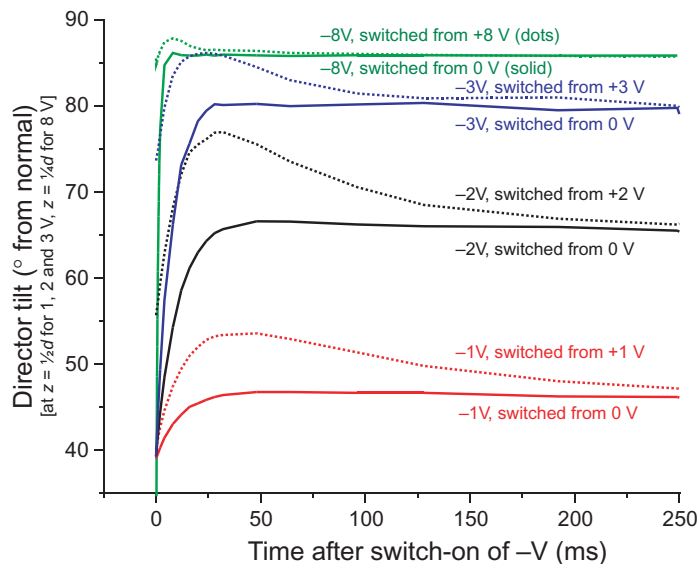
**Figure 6.** Director tilt during switching with (a) 1 V, (b) 2 V, (c) 3 V and (d) 8 V bipolar voltage pulses. Tilt shown at  $z = 0.5d$  for (a)–(c) and  $z = 0.25d$  for (d). Points show experimental data, and lines are a guide for the eye.

5. Switch-off director profiles are not dependent on the polarity of the bipolar pulse, aside from the different profiles at a time of 500 ms, and show a monotonic return to the 0 V equilibrium profile. Further the switch-off director profiles are similar in shape and switching speed to that observed with ac switch-off [21].

The time constants of switching are of particular interest. From ac switching studies on the same cell [21], switch-on—which is driven by  $3 V_{\text{rms}}$ —is complete in  $\approx 30$  ms, whereas switch-off—which relaxes due to elastic forces—takes about twice this time [21]. In the data presented in figure 6, an additional, larger, time constant is needed to describe the ‘dc’ switch-on data. For the 1 V data, the director has not reached equilibrium 250 ms after a voltage change and there are clearly transients of  $\approx 100$  ms in the 2 and 3 V data. Switching between the two applied voltage polarities is a driven process—the voltage changes sign, but not magnitude. It is therefore expected to occur on a similar timescale as with ac switch-on, i.e.  $\approx 30$  ms. These unexpectedly slow timescales observed experimentally are characteristic of ion motion in the cell.



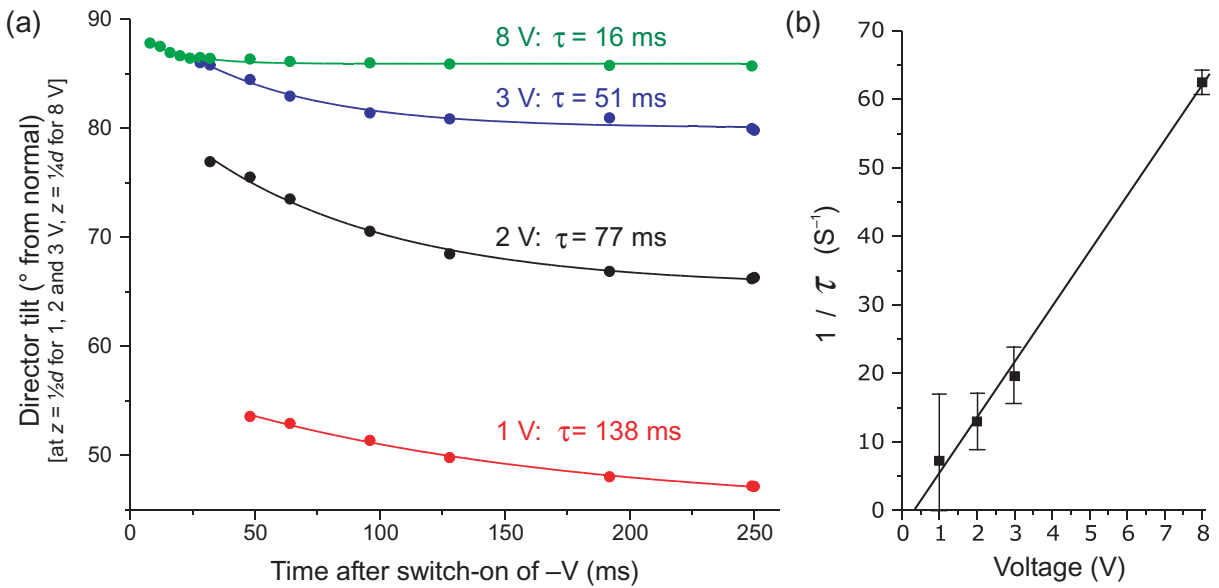
**Figure 7.** Switching to a positive voltage, from 0 V (solid) and  $-V$  (dots).



**Figure 8.** Switching to a negative voltage, from 0 V (solid) and  $+V$  (dots).

Further examination of the director tilt as a function of time (as in figure 6) shows an interesting asymmetry in switching. When a positive voltage is applied, regardless of whether the cell is switching from 0 V (i.e. 1st half of a positive bipolar pulse) or from a negative voltage (i.e. 2nd half of a negative bipolar pulse), the director tilt angle during switching is the same, discounting the initial  $\approx 30$  ms of switching. This is clearly shown in figure 7; the ‘longer-term’ dynamics of switching to a positive voltage are independent of the previous voltage.

In contrast, as shown in figure 8, the ‘longer-term’ dynamics of switching to a negative voltage are dependent on the previous voltage. When switching from 0 V to a negative voltage, it is clear that switching is completed in  $\approx 30$  ms, although as expected, switching is faster with higher voltages. Conversely, when switching to a negative voltage from a positive one, the director tilt switches over a rather longer timescale, again depending on voltage.



**Figure 9.** (a) First-order exponential decay (lines) fitted to director tilts (points) at selected times yields time constants,  $\tau$ ; (b)  $1/\tau$  against voltage (points) with a linear best fit line.

Figure 9(a) shows the voltage dependence of this slow switching time constant when switching from a positive to a negative voltage, determined by fitting a first-order exponential decay to the time-dependent director tilt. The time constant,  $\tau$ , of this ‘switching off’ is inversely proportional to the applied voltage,  $V$ . This is consistent with ion motion across the cell gap,  $d$ , as given by [23]

$$\tau = \frac{d^2}{\mu V}. \quad (7)$$

Then from the gradient of the linear best fit in figure 9(b), the ionic mobility,  $\mu$ , can be estimated as  $7 \times 10^{-11} \text{ m}^2 \text{ V}^{-1} \text{ s}^{-1}$ . This is comparable to values reported in the literature for mobile ions in other liquid crystals: Mada and Osajima [23] quote a mobility of  $2.5 \times 10^{-11} \text{ m}^2 \text{ V}^{-1} \text{ s}^{-1}$  and Stojmenovik *et al* [24] give a value of  $2.1 \times 10^{-10} \text{ m}^2 \text{ V}^{-1} \text{ s}^{-1}$ .

We have shown experimentally that switching to dc voltages is slower than expected, when compared with ac switching. We suggest that these ‘slow’ transients are caused by the movement of charged ions across the cell and now discuss important implications that follow from this. When a positive voltage is applied to the cell, regardless of the voltage history, slow transients are observed: our hypothesis is that the ions are able to move. However, when the cell is switched from 0 V to a negative voltage, no ‘slow’ switching transients are observed: our hypothesis is that the ions are not able to move.

This behaviour could be explained if all the mobile ions in the cell were positively charged, and at 0 V were positioned next to the homeotropic cell boundary. When a negative voltage is applied, the ions would not move significantly, as the electric field would act to push them towards the cell substrate. Conversely, a positive voltage would force the ions away from the cell boundary into the bulk liquid crystal. As the ions ‘slowly’ moved across the cell, they would change the electric field distribution of the cell, perhaps leading to the slow switching transients observed, until they reached some equilibrium.

Alternatively, negative ions could be located right next to the planar alignment surface, or there could be a mix between these two situations. From the data, it is not obvious how to distinguish between these possibilities, although it is likely that positive and negative charges have different mobilities and hence transit times. If the cell contained both positive and negative ions plated on opposite substrates, then there would probably be two ‘slow’ ionic time constants, but this is not immediately evident. Regardless of the sign of mobile ionic charge, this explanation of the observed behaviour requires that the 0 V equilibrium position of mobile ions is next to a substrate.

## 6. Conclusions

The optics of a HAN cell filled with a negative dielectric anisotropy liquid crystal, ZLI 4788-000, are dependent on the sign of the applied voltage. Careful use of the fully leaky guided mode technique, in which angle-dependent optical intensity data is analysed, yields the director profile of the liquid crystal cell, thus allowing detailed study of this sign dependence. Prior to examining the sign-dependent voltage behaviour, we characterize the cell with applied ac voltages.

Experimentally, with a negative voltage applied (planar substrate held at ground) the equilibrium director profile is more planar than when a positive voltage is applied. Two physical models were considered to explain these sign-dependent director profiles: flexoelectricity and surface charge. It was found that both these models provide a satisfactory fit to the experimental optical data, either with a flexoelectric coefficient of  $2.25 \times 10^{-11} \text{ C m}^{-1}$  or a bound surface charge of  $12.2 \mu\text{C m}^{-2}$ . Further, the data can be explained by certain combinations of the two effects. Thus there is degeneracy between flexoelectricity and surface polarization that cannot be resolved by just measuring the equilibrium director profiles of this cell.

Understanding the director profiles during switching provides a possible path to resolving this difficulty. Rather unexpectedly however, simply adding the Ericksen–Leslie switching theory to the physical model successfully used to fit the static data did not yield satisfactory fits to the dynamic data. In the absence of a suitable physical model to generate director profiles to fit the dynamic data, we used a simple mathematical function. This allowed the director profile to be ‘imaged’ during switching.

Then the measured director profiles show switch-on from 0 V proceeds rather differently depending on the sign of the applied voltage. With a negative voltage, switch-on is as expected from ac switching studies, in which the director profile changes monotonically with time, with faster switching near the homeotropic boundary providing evidence of flow. Switch-on to positive voltages is much slower and anomalous in form; in some cases the director ‘switches off’ (becomes more like the 0 V state) even though a voltage was being applied.

We suggest that this slower ‘dc’ switching is because of mobile ion motion across the cell, yielding an estimate for the ion mobility of  $7 \times 10^{-11} \text{ m}^2 \text{ V}^{-1} \text{ s}^{-1}$ , which is typical for ions in liquid crystals [23, 24]. Further, the dependence of the switch-on dynamics to both applied voltage polarity and previous voltage history suggests that the 0 V equilibrium positions of the mobile ions are next to a cell substrate, such that a negative applied voltage forces the ions to remain at the cell substrate. In a future paper [22], we present a model that includes ion dynamics to explore the physical reasons for this behaviour.

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