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Surface Evanescent Field Characterisation of Antiferroelectric Liquid Crystals

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In this paper we report a study of the dynamics of antiferroelectric liquid crystals by using simultaneous total internal reflection and transmission. In devices having both substrates coated with a polyimide, but only one rubbed, we found that the optic axis of the liquid crystal region close to the unrubbed substrate deviates from the rubbing direction less than the bulk. In switching experiments we find that this region is also slower than the bulk and it is virtually free from the undesired pretransitional effect. We also found an asymmetry in the switching of the surface region and we use a simple model to account for it.

Keywords: antiferroelectric liquid crystals; total internal reflection; surface evanescent field; high quality alignment

INTRODUCTION

The use of surface evanescent fields to probe the near surface region in smectic liquid crystals is a powerful investigation technique for liquid crystal devices where a complex structure is present. It has proven effective for the understanding of the internal structure in SmC* liquid crystals [1], [2], [3], [4]. However its use for more complex smectics such as SmC_A* has been quite limited [4], [5] due to the difficulties in obtaining the necessary high quality and uniform alignment. Hence a sufficient understanding of the complex internal structure of antiferroelectric liquid crystals (AFLCs) in thin devices is yet to be developed. For instance, it is still quite unclear whether AFLCs in thin cells [2-3 μm] operate in a fully surface stabilised configuration as is the case for long pitch SmC* materials in the surface stabilised configuration demonstrated by Clark and Lagerwall [6]. It is reasonable to expect only a partial suppression of the helix in thin cells. To our knowledge this has not been clarified so far. An investigation into whether a deformed helicoidal structure exists in the bulk (i.e. far from the alignment layer) is of fundamental importance. The much debated pretransitional effect [7] could be, in fact, due to an incomplete surface stabilisation. This effect, which we have found is accompanied by both a rotation of the optic axis and increase of the optical birefringence [7] is highly undesired in passively addressed AFLC displays because it is responsible for partial transmission when a holding voltage and crosstalk data are applied.

In our recent work we have investigated the problem of alignment of AFLC, and succeeded in producing high quality devices [8], [9]. These are suitable for accurate investigation of the polarised light transmitted

through the device as well as the polarisation conversion of a totally internally reflected beam (whilst working above the critical angle condition) by the coupling with the evanescent field penetrating into the liquid crystal close to the substrate. The comparison of these two signals in static and dynamic conditions (with an applied voltage waveform) may disclose whether or not the AFLC is fully surface stabilised.

EXPERIMENTAL

We use high quality alignment devices filled with the commercial mixture CS4000. According to the manufacturer, this mixture exhibits the following phase sequence: Iso- 100.6 °C - SmA* - 83.5 °C - SmC* - 82.2 °C - SmC_A*- (-10.1 °C) – mp. However, when the mixture is sandwiched in thin cells, we observe only one phase transition between the smectic phases, at 81 °C [10]. One of the substrates is a conventional Indium Tin Oxide (ITO) coated glass plate. The second electrode is a high index prism, which is also coated with ITO. The thickness of the (empty) device used for this work is $2.0 \pm 0.1 \mu\text{m}$. The alignment is produced by spin-coating a polyimide on both substrates and strongly rubbing only one of them. After rubbing, the polyimide thickness is $30 \pm 5 \text{ nm}$. No blocking layers are used. The optical set-up is illustrated in Figure 1. The refractive index of the prism is 1.778 and its the strain birefringence is negligible. In these experiments, the penetration depth of the evanescent field in the liquid crystal is $\approx 100 \text{ nm}$ (5% of the cell thickness).

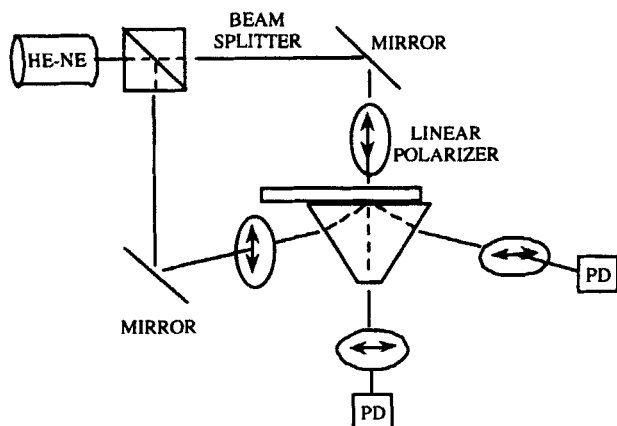


FIGURE 1 Experimental set-up for simultaneous total internal reflection and transmission measurement.

The devices have only one substrate rubbed. In all such devices the optic axis orientation of greatest extinction between crossed polarisers deviates from the rubbing direction. We have also found that a layer rotation occurs under the application of a DC-free symmetric triangular waveform [8], [9]. Such an effect is enhanced when the temperature is increased while in the AFLC phase. With the present combination of polyimide and liquid crystal the rotation of the effective optic axis as observed for the transmission signal is 13.7° . In these experiments we focus our attention on the region near the unrubbed surface. Interestingly we observed that the 100 nm of liquid crystal near the surface appears to be in the antiferroelectric state (AF) even in the freshly filled device. We also found that the effective deviation from the rubbing direction of the surface region is only 4.9° , significantly

less than in the bulk, but in the same direction. A preliminary evaluation of the voltage induced layer rotation shows that the surface rotates less than the bulk.

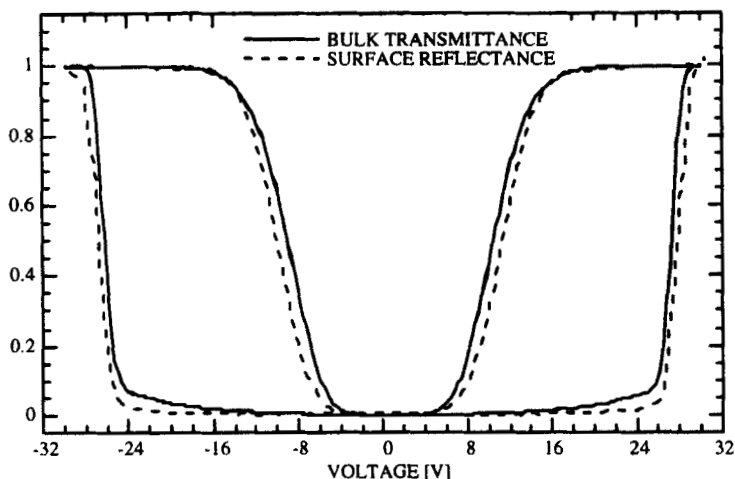


FIGURE 2 Experimental investigation of tristable switching. The depolarisation reflection signal (dashed line) shows that the region near to the surface tends to stay in the AF state and is virtually free from pretransitional transmission.

Switching behaviour

The experiments were carried out at room temperature. When a triangular waveform is applied to the device we find interesting

differences between the surface and the bulk regions. The AF to F switching threshold is slightly increased at the surface while the onset of switching in the F to AF is not significantly affected. More interestingly the pretransitional effect has virtually disappeared in the near surface region. These results are illustrated in Figure 2.

It seems reasonable to attribute the reduction of the pretransitional regime to a stronger stabilisation effect produced by the surface.

When we drive the device with a step voltage of amplitude sufficient to switch homogeneously from AF to F [11] we observe that the bulk response is nearly twice as fast as the surface as shown in Figure 3.

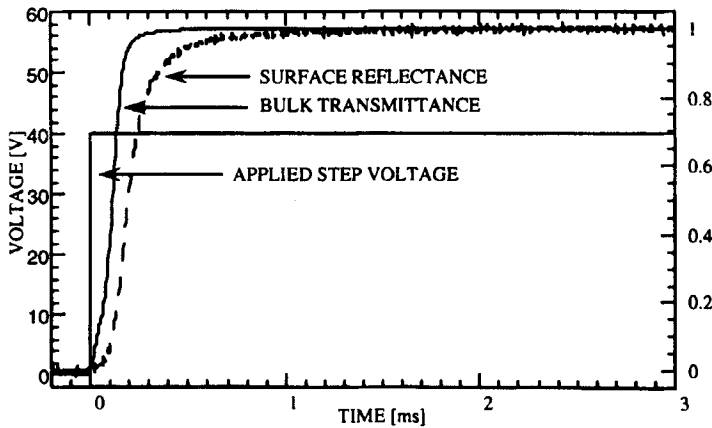


FIGURE 3 Step response. The bulk (plain line) leads the surface (dashed line) : the 10-90% rise time is 140 μ s and 270 μ s for the bulk and region near the surface, respectively.

Within a range of frequencies of the triangular waveform, lower than the frequency for which direct F to F switching occurs, two dark states are usually observed in each half of the voltage cycle, separated by an overshoot (Figure 4). In the present experiment we have found that this effect is weaker in the surface region when compared with the bulk. There also exists a strong asymmetry in the surface switching, whilst the bulk behaviour is symmetric, as seen in Figure 4.

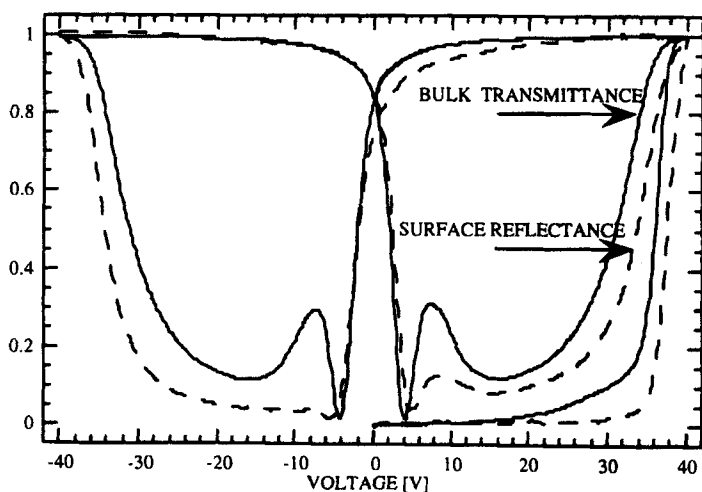


FIGURE 4 Switching under the application a 50 Hz triangular waveform. Bulk transmission (plain line) and reflection (dashed line) are shown from the initial application of the driving voltage.

THEORY OF ASYMMETRIC SWITCHING

In order to describe differences between the switching behaviour of bulk and surface and the asymmetry of the surface, we use a simple two layer, zero-dimensional model [12], [13]. We define θ as the cone angle and ϕ_1 and ϕ_2 as the azimuthal angles of the directors in the two layers (see Figure 5). With these definitions the directors can be expressed as

$$\begin{aligned} \mathbf{n}_1 &= (\sin\theta \cos\phi_1, \cos\theta, \sin\theta \sin\phi_1) \\ \mathbf{n}_2 &= (\sin\theta \cos\phi_2, \cos\theta, \sin\theta \sin\phi_2) \end{aligned}$$

and the spontaneous polarisation vectors as

$$\begin{aligned} \mathbf{P}_{s1} &= P(-\sin\phi_1, 0, \cos\phi_1) \\ \mathbf{P}_{s2} &= P(-\sin\phi_2, 0, \cos\phi_2) \end{aligned}$$

Therefore when $\phi = 0$ or $\phi = \pi$ the director is parallel to the glass surfaces. We use the following expression for the free energy:

$$\begin{aligned} E = & -\frac{V}{d} P(\cos\phi_1 + \cos\phi_2) + \gamma \cos(\phi_1 - \phi_2) \\ & + \mu_{np}(\sin^2\phi_1 + \sin^2\phi_2) + \mu_p \left(\sin^2\frac{\phi_1}{2} + \sin^2\frac{\phi_2}{2} \right) \end{aligned}$$

where these terms represent, respectively: (i) the interaction between the spontaneous polarisation and the applied electric field; (ii) the antiferroelectric ordering; (iii) a non-polar surface interaction term,

stabilising the director in the $\phi = 0$ or $\phi = \pi$ states; (iv) a polar surface interaction term which stabilises the $\phi = 0$ state when $\mu_p > 0$.

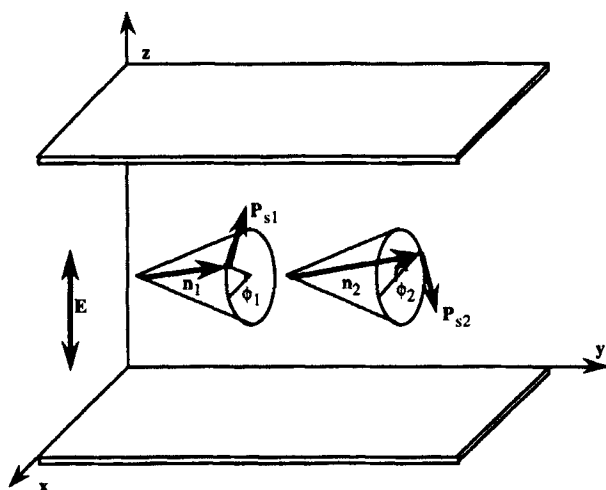


FIGURE 5 Definition of the director orientation used.

We will use this zero-dimensional model to describe two regions, one near to the surface and one in the bulk of the cell. Near to the cell surface we assume that a polar interaction between the molecular dipole and the alignment layer results in a non-zero polar energy term ($\mu_p \neq 0$) whilst in the bulk this surface interaction is negligible and $\mu_p = 0$. In this case we find a qualitative agreement between theory and

experiments, i.e. whilst the bulk transmission curve is symmetric the surface reflection is asymmetric (Figure 6).

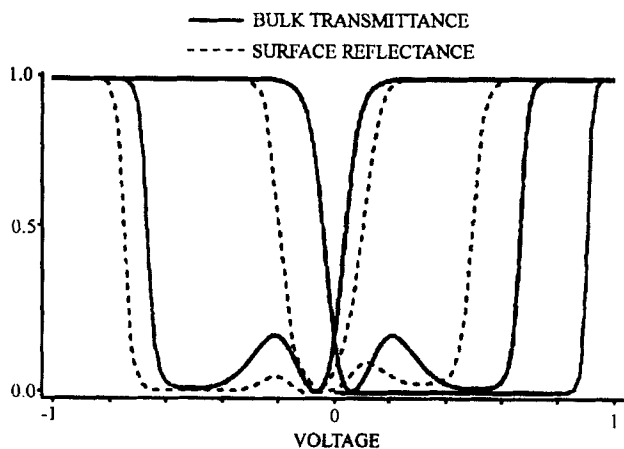


FIGURE 6 Simulation results. The asymmetry in the region near the surface can be described by a non-zero polar surface interaction, not acting on the bulk.

A more detailed theoretical analysis will be conducted at a later date in order to quantitatively fit a theoretical model to the experimental results.

CONCLUSIONS

We have investigated, by simultaneous total internal reflection and transmission of polarised light, the internal structure of antiferroelectric liquid crystal devices. We have found that significant differences exist between the bulk and near surface region. First we observed that the tendency for layer rotation is reduced on the unrubbed surface. More importantly we measured a much reduced pretransitional regime at the surface. We interpret these results in terms of a surface stabilisation effect, but we argue that only a thin region near the surface is fully surface stabilised. This stabilisation favours either the AF or the F state and disfavours intermediate configuration of the directors. This stabilisation is also thought to slow down switching and hamper layer rotation.

Furthermore we observed that there exists a range of frequencies where the surface switching is asymmetric (while the bulk is not). This can be satisfactorily described in terms of a simple model where both a polar and a non-polar surface interaction terms are considered near the surface while only a non-polar term is considered for the bulk. This appears reasonable as the polar interaction can be attributed to the polymer alignment layer. However, in the middle of the sample, the net polar interaction due to the surfaces (of equal and opposite polarisation) is zero. On the other hand the non-polar interaction, being steric in nature, does not.

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