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Preliminary communication

Local Fréedericksz transitions at a nematic liquid crystal/ thin film oxide ferroelectric interface

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The electro-optical properties of a novel device comprising nematic liquid crystals and a thin film oxide ferroelectric (OFE) substrate are reported. The OFE was the lead zirconate–lead titanate, PZT, system with the molar composition 30:70, respectively. The first evidence of the interaction of nematic liquid crystals with the spontaneous polarisation (P_s) of an OFE is presented. Coupling of the ferroelectric P_s from poled grains (5–10 μm in diameter) with the liquid crystal results in local Fréedericksz transitions, allowing the grain structure of the substrate to be visualized. Further, this novel device structure allows the director tilt and anchoring energy of commercially available nematic materials to be quantified.

The interaction of ferroelectric substrates with nematic liquid crystals (NLCs) has long been established [1–5]. The interaction mechanism is believed to involve the coupling of the spontaneous polarisation (P_s) of the crystalline surface with the liquid crystal. In general this interaction is poorly understood although it is thought to depend on several factors such as the dielectric anisotropy of the liquid crystal, $\Delta\epsilon$, the anchoring energy of the surface and the symmetry of the ferroelectric substrate [1, 6].

The understanding of NLC surface interactions is fundamental to display device applications as indicated by the growing trend in surface alignment based electro-optical effects. For example, the flexoelectrically controlled, surface bistable NLC display of Barberi *et al.* [7] is based on an asymmetric surface anchoring effect facilitated by controlled grazing evaporation of SiO. More recently, a device which exploits the voltage-dependent anchoring

of an NLC on a grating surface has been realized [8]. The interaction between a ferroelectric polymer, poly(vinylidene fluoride–trifluoroethylene) [PVF₂–TrFE (30:70)], and an NLC has only just been reported [9]. Previous work on liquid crystals on ferroelectric crystal surfaces has been limited to substrates of the order of between 0.05 and 10 mm in thickness. This paper describes the interaction between NLCs and an oxide ferroelectric substrate of submicron thickness. A measure of the anchoring energy is obtained, hence providing an insight into the surface forces involved in the interaction.

The novel device presented here utilizes a thin film (<0.5 μm thick) of the oxide ferroelectric Pb(Zr_{0.30}Ti_{0.70})O₃ (PZT 30/70) to orient an NLC. It is demonstrated for the first time, that poling of a PZT (30/70) film induces local Fréedericksz transitions in NLCs above those grains whose ferroelectric domains are oriented collinearly with the poling field direction. Using well known equations to describe the director profile through a nematic device, as well as a knowledge of the elastic constants of the

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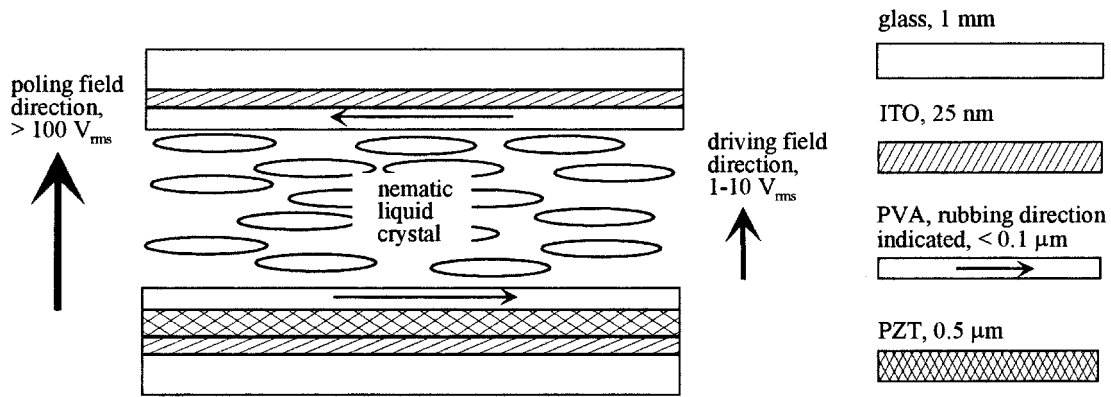


Figure 1. Schematic of the oxide ferroelectric/NLC device. The large vertical arrow indicates the poling field direction, only present on first application, whereas the smaller vertical arrow indicates the normal driving field for NLC devices. Also shown is the antiparallel arrangement of alignment layers as indicated by the horizontal arrows.

NLCs, enables the maximum director tilt in the centre of the device to be quantified. More importantly, this methodology allows estimates of the surface anchoring energy of NLCs to be obtained.

The devices were constructed according to the schematic shown in figure 1 where the PZT (30/70) layer was deposited by sol-gel/spin-coating [10] with thickness in the range 0.15 to 0.50 μm . Antiparallel rubbed polyvinylalcohol (PVA) alignment layers were applied to the faces of the device and spacers included, resulting in a thicknesses of the NLC layer between 4 and 10 μm ($\pm 0.3 \mu\text{m}$). The commercially available nematic material K15 and the nematic mixture E7 were both employed \dagger . Sinusoidal voltages of up to 600 V_{rms} and frequencies between 1 and 5 kHz were applied to the devices which were held in a hot stage controlled to $\pm 0.1^\circ\text{C}$. The electro-optic properties of the devices were observed with a polarizing microscope with the device alignment direction at 45° to that of the crossed polarizers.

In order to produce an interaction between the OFE film and the NLC, an electric field of the order of the coercive field must be applied across the OFE to pole it. To a first approximation, the device can be modelled electrically as two dielectrics in a series circuit where each layer has associated with it a capacitive and a resistive element. It can be shown that at high frequency the voltage across the OFE (PZT) is

$$|V_{\text{OFE}}| = V_0 \left/ \left[1 + \frac{\varepsilon_{\text{OFE}} t_{\text{NLC}}}{\varepsilon_{\text{NLC}} t_{\text{OFE}}} \right] \right. \quad (1)$$

where ε is the dielectric constant and t the thickness of the layer, the subscript defining the layer type. V_0 is the driving voltage. The coercive field of PZT (30/70) is approximately $5 \text{ V } \mu\text{m}^{-1}$, ε_{PZT} is 230, and ε_{\parallel} for K15 is

\dagger Business Unit Liquid Crystals, E. Merck, Darmstadt, Germany.

26. Hence V_0 must be 224 V_{rms} in order for the PZT to be poled in a device comprising 5 μm of K15 and 0.5 μm of PZT. The parallel component of the NLC dielectric constant is the appropriate one to include in the calculations since the poling field gives a voltage much greater than the threshold voltage across the device and hence homeotropic alignment of the nematic director. For a device including 5 μm of E7, ε_{\parallel} is 19, lower than for K15, and V_0 must be increased to 305 V_{rms} if the PZT (0.5 μm) is to be poled.

Evidence for the interaction of the poled regions of the PZT film with an overlying layer of NLC is presented in figure 2 where a PZT (0.26 μm)/E7 (8.87 μm) device has been subjected to a poling voltage of 640 V_{rms} (at 1 kHz). The poled grains of the PZT are visualized as a change in the birefringence of the NLC from the original pink to a green colour. The transparent PZT film does not contribute to the optical retardation of the device, since both the birefringence and layer thickness are an order of magnitude smaller than those of the liquid crystal. Setting the alignment direction to that of one polarizer causes extinction of both poled and unpoled grains, implying that the nematic director has tilted out of the plane of the substrate without a twist. Similar effects to those presented in figure 2 have also been observed for K15/PZT devices after application of the appropriate poling field. The phenomenon depicted in the figure 2 may be interpreted in terms of the surface shown in figure 3, which illustrates the PZT after a poling field has been applied. It is well known that the deposition of PZT (30/70) onto ITO coated glass yields a distribution of c -axis (polarization vector) orientations. Thus a poling field perpendicular to the substrate will pole only those ferroelectric domains which are oriented along the field direction. For those domains which satisfy this criterion, a bulk spontaneous polarisation (\mathbf{P}_s) of the grain will result. Typical values of the \mathbf{P}_s for PZT

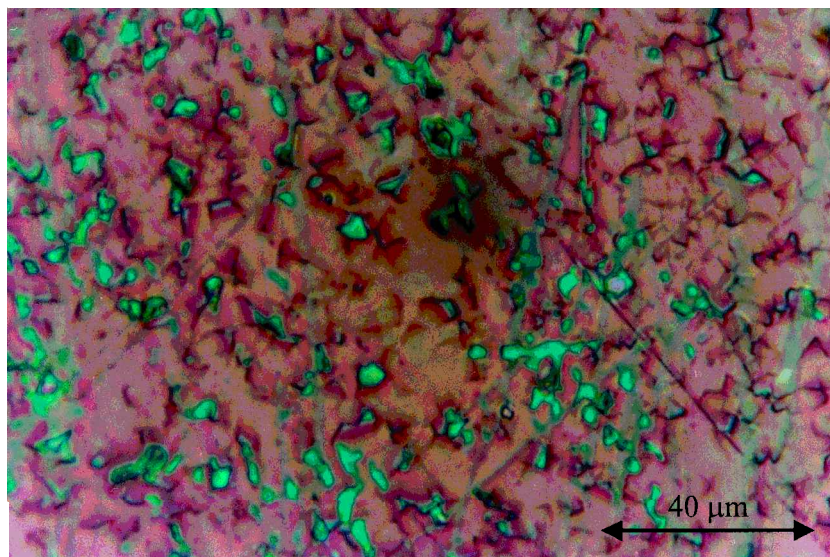


Figure 2. Photomicrograph taken for a device comprising $8.87\ \mu\text{m}$ of E7 and $0.26\ \mu\text{m}$ of PZT after a poling voltage of $640\ V_{\text{rms}}$ (at $1\ \text{kHz}$) has been applied; crossed polarizers, 30°C . See the text for details.

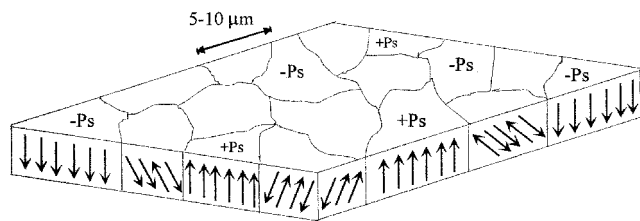


Figure 3. Topological model showing the grain structure of the PZT in a device after a poling field has been applied (the ITO and alignment layers are not shown). Note that it is the leakage field created above the poled PZT grains, arising from the $+P_s$ and $-P_s$ regions, which couple with the NLC.

(30/70) films are between 30 and $35\ \mu\text{C cm}^{-2}$ (compared with between 10 and $150\ \text{nC cm}^{-2}$ for ferroelectric liquid crystals). The leakage field due to the polarization of the PZT substrate couples with the positive $\Delta\epsilon$ of the NLC inducing a local Fréedericksz transition above the poled grains. Since the Fréedericksz transition depends on the magnitude of the applied voltage, not on its direction, the NLC is unable to distinguish between those grains possessing $+P_s$ and $-P_s$.

The existence of the P_s can be thought of as a 'bias' voltage across the NLC layer. The magnitude of this bias voltage was measured by applying a driving voltage (much smaller than the poling voltage) to the devices to match the birefringence colours above unpoled PZT grains with those where poling has taken place. The driving voltage needed to achieve a match for K15 was $0.71\ V_{\text{rms}}$ and for E7 was $1.03\ V_{\text{rms}}$, greater than the experimentally measured threshold voltages, V_{th} , of 0.68 and $0.93\ V_{\text{rms}}$ for K15 and E7, respectively, as expected. The maximum angle of director distortion occurring in the centre of the device, θ_{max} , can be calculated from these measurements by solving equation (2) numerically

[11]:

$$\frac{V}{V_{\text{th}}} = \frac{2}{\pi} \int_0^{\pi/2} d\psi \left(\frac{1 + \kappa \sin^2 \theta_{\text{max}} \sin^2 \psi}{1 - \sin^2 \theta_{\text{max}} \sin^2 \psi} \right)^{1/2} \quad (2)$$

where $\kappa = (K_{33} - K_{11})/K_{11}$ (K_{11} and K_{33} are the splay and bend elastic constants, respectively). The values used for K_{11} were $6.0\ \text{pN}$ (K15) and $11.1\ \text{pN}$ (E7) while those used for K_{33} were $10.5\ \text{pN}$ (K15) and $17.1\ \text{pN}$ (E7). The ratio V/V_{th} measured by the method described above was found to be 1.04 for K15 ($4.70\ \mu\text{m}$ device) and 1.11 for E7 ($8.87\ \mu\text{m}$ device). Both devices had a PZT layer thickness of $0.26\ \mu\text{m}$. The data imply a θ_{max} of $17 \pm 5^\circ$ for K15 and $30 \pm 5^\circ$ for E7. The model used assumes: (i) an infinite surface energy term, (ii) a symmetric distortion of the nematic director and (iii) no pretilt at the substrates. While the different $\Delta\epsilon$ of the NLCs would be expected to result in different voltages for the same leakage fields in both cases, the large difference in θ_{max} implies that significant surface interactions are occurring. This will be the subject of a future publication, though it is possible to make some comments as follows. As the nematic director is distorted only in the plane perpendicular to the substrate, then the surface anchoring energy, $W\theta$, may be calculated from [11],

$$W\theta = \frac{\pi^2 d(k_{11} + k_{33})}{2d_B^2} \quad (3)$$

where d_B is the thickness of the domain wall and d is the thickness of the nematic layer. Estimations of d_B for the K15 and E7 devices are 2 and $5\ \mu\text{m}$, respectively, resulting in anchoring energies of $\sim 9 \times 10^{-2}$ and $5 \times 10^{-2}\ \text{erg cm}^{-2}$, respectively. These surface energies compare well with those measured for K15 and other nematic materials in the literature [11]; these range

between 10^{-4} and 1 erg cm^{-2} . Of particular relevance to the results presented here is the measurement of the anchoring energy of MBBA on a cleavage surface of the ferroelectric crystal triglycine sulphate (TGS) by Gonyakov *et al.* [12] which they reported to be $2.1 \times 10^{-2} \text{ erg cm}^{-2}$. It is not unreasonable to imagine that the surface \mathbf{P}_s contributes to the anchoring energy for both the ferroelectric crystal and the thin film oxide ferroelectric systems. If, as seems likely, it is a surface electric field which couples electrostatically with the NLC director, then one would expect this field to be proportional to the ratio of the \mathbf{P}_s to the dielectric constant, ϵ , of the ferroelectric substrate, as these factors will determine the electrostatic field at the surface of the ferroelectric due to the spontaneous polarization. For TGS, the \mathbf{P}_s and ϵ are $2.8 \mu\text{C cm}^{-2}$ and 43, respectively [13], while these quantities have been measured in the present work as being $40 \mu\text{C cm}^{-2}$ and 230 for the PZT (30/70) films used. From these figures, the ratios \mathbf{P}_s/ϵ are 0.065 for TGS and 0.174 for PZT (30/70). These are in good comparison with the ratios for the anchoring energies of the NLC for the respective substrates. Clearly, this is a preliminary result, but appears to provide further evidence that the physical interaction which is causing the alignment of the liquid crystal at the surface of the ferroelectric is an electrostatic one associated with the spontaneous polarization in the ferroelectric.

In conclusion, it has been demonstrated for the first time that coupling of PZT OFE films with NLCs is possible. The interaction mechanism of the PZT with the NLCs is thought to involve the leakage field above poled PZT grains possessing either a $+\mathbf{P}_s$ or a $-\mathbf{P}_s$. Since a number of assumptions have been made, further work is required in order to elucidate a more accurate description of the coupling process. Nonetheless, results so far indicate anchoring energies close to those of ferroelectric crystal/NLC systems, implying significant surface influences in devices such as the one described

here. Monostable switching has been demonstrated and the potential of hybrid devices including thin film OFEs will be the subject of future investigations.

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