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## The Interaction of Poled Thin Film Ferroelectrics with Nematic Liquid Crystals

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Nematic liquid crystals (NLCs) have been widely used to image the static and dynamic domain structure that occurs in poled ferroelectric crystals. The interaction mechanism is influenced by both the coupling of the ferroelectric polarisation with the NLC and the effect on the surface anchoring of the NLC director due to the ferroelectric substrate. This paper reports the formation and observation of poled structures in thin film ferroelectric ceramic systems incorporated in a novel liquid crystal device architecture. Hybrid display devices combining thin film oxide ferroelectrics (OFEs) with NLCs are described. The OFEs are of the lead zirconate-lead titanate,  $\text{Pb}(\text{Zr}_{0.30}\text{Ti}_{0.70})\text{O}_3$  (PZT 30/70) system, are under  $1\text{ }\mu\text{m}$  thick, transparent and are combined with commercially available NLCs. Visualisation of poled domains within the OFE is demonstrated. The mechanism of coupling the polarisation of the OFE with the orientation of the NLC is described and preliminary results are discussed.

**Keywords:** ferroelectric thin films; PZT; nematic; birefringence

## INTRODUCTION

It well known that nematic liquid crystals (NLCs) can be used to observe the domain structures of ferroelectric crystals, where the thickness of the crystal substrate is 0.5 to 10 mm thick.<sup>[1]</sup> This paper reports the extension of this technique to thin film oxide ferroelectric (OFE) ceramic films of thickness less than 0.5  $\mu\text{m}$ . The OFE films investigated were the  $\text{Pb}(\text{Zr}_{0.30}\text{Ti}_{0.70})\text{O}_3$  (PZT 30/70) system with commercially available NLCs. Preliminary results are presented and the coupling of poled regions PZT with NLCs is discussed.

## THEORY

To investigate the interactions between OFEs and NLCs the hybrid device depicted in Fig.1 was considered. A planar alignment of the NLC director was assumed for the following theoretical description.

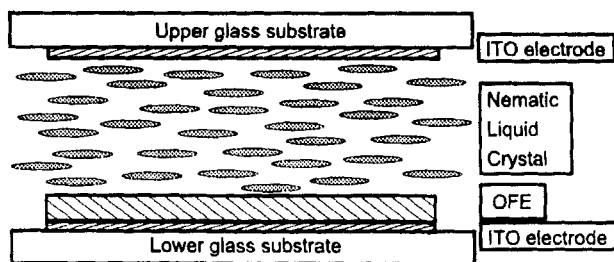


FIGURE 1 Schematic representation of the OFE/NLC device.

Modelling the NLC and OFE layers as two dielectrics in series results in Eqn. (1) for the voltage appearing across the OFE (at 1-5 kHz).

$$|V_{OFE}| = V_0 / \left[ 1 + \frac{\epsilon_{OFE} t_{LC}}{\epsilon_{LC} t_{OFE}} \right], \quad (1)$$

where  $V_0$  is the driving voltage,  $\epsilon$  is the dielectric constant, and  $t$  is the layer thickness, with the appropriate subscript for the liquid crystal (LC) and OFE layers. If  $|V_{OFE}|$  is of the order of the coercive field,  $5 \text{ V}\mu\text{m}^{-1}$  for PZT (30/70), then the OFE layer will be poled and its influence on the overlying NLC layer can be observed. Typically the PZT layer was  $0.3 \mu\text{m}$  thick so that  $|V_{OFE}|$  was  $1.5 \text{ V}_{\text{rms}}$ . The appropriate value of  $\epsilon_{LC}$  to substitute into Eqn. (1) is the parallel component of the dielectric constant,  $\epsilon_{//}$  (26 and 19 for K15\* and E7\*, respectively) as is demonstrated by the following cases. For a device comprising  $5 \mu\text{m}$  K15 and  $0.3 \mu\text{m}$  PZT ( $\epsilon_{OFE} = 230$ ),  $V_0$  is calculated to be  $223 \text{ V}_{\text{rms}}$ . Similarly,  $V_0$  is  $304 \text{ V}_{\text{rms}}$  for  $5 \mu\text{m}$  E7 and  $0.3 \mu\text{m}$  PZT. The corresponding voltages across the LC layers are  $221.5 \text{ V}_{\text{rms}}$  (K15) and  $302.5 \text{ V}_{\text{rms}}$  (E7), well above the threshold for the NLCs ( $\sim 1 \text{ V}_{\text{rms}}$ ) confirming that  $\epsilon_{//}$  is the appropriate dielectric constant to use.

## EXPERIMENTAL

Hybrid devices were constructed according to Fig. 1 where the PZT layer was deposited onto ITO glass by a sol gel-spin coating procedure,<sup>[2]</sup> with thickness between  $0.15$  and  $0.50 \mu\text{m}$ . Surface alignment of the NLC was achieved by use of an aqueous solution of polyvinyl alcohol (1 % v/v) and rubbing unidirectionally. The thickness of the NLC layer was between  $4$  and  $10 \mu\text{m}$  (controlled with the introduction of polyethyleneterephthalate spacers between

\* Supplied by Merck, Darmstadt, Germany

the substrates) measured before filling by spectrophotometry. The edges of the devices were glued, leaving a small hole to enable filling. The devices were filled with NLC in vacuum to avoid the presence of air and the filling hole glued. Electrodes were attached to the exposed ITO edges of both substrates with indium solder. Electric fields were applied to the devices with a function generator and amplifier, the output being monitored with a multimeter. With this set-up voltages of up to 600 V<sub>rms</sub> were available yielding electric fields of up to 150 V $\mu\text{m}^{-1}$ . Devices were observed using a polarising microscope with the NLC alignment direction at 45° to the crossed polarisers.

## RESULTS

### K15/PZT Devices

Plate 1 shows a device comprising 4.2  $\mu\text{m}$  K15 and 0.26  $\mu\text{m}$  PZT after a poling field of 208 V<sub>rms</sub> (at 1 kHz) has been applied. The poled PZT grains can be visualised as purple regions on a blue background in Plate 1. The blue background arises from the birefringence,  $\Delta n$ , of the NLC. Note that the contribution to  $\Delta n$  from the PZT is negligible since the thickness of the film is 0.26  $\mu\text{m}$  and  $\Delta n$  of PZT is of the order of 0.01, both much smaller than that of the liquid crystal ( $\Delta n = 0.212$  for K15, at 589 nm, 20°C).

### E7/PZT Devices

Plate 2 shows a device comprising 8.9  $\mu\text{m}$  E7 and 0.26  $\mu\text{m}$  PZT after a poling field of 538 V<sub>rms</sub> (at 1 kHz) has been applied. The poled grains in Plate 2 are much larger than those for the K15 device, shown in Plate 1. The larger grains provide a clearer visualisation of the poled regions of the PZT film.

40  $\mu\text{m}$

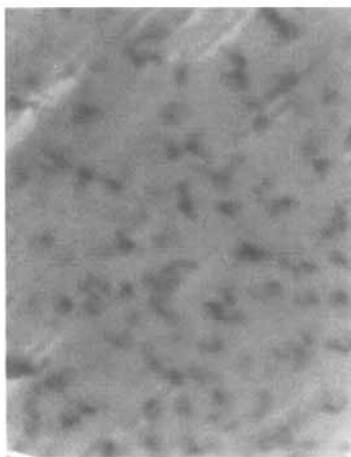


PLATE 1. K15, no voltage

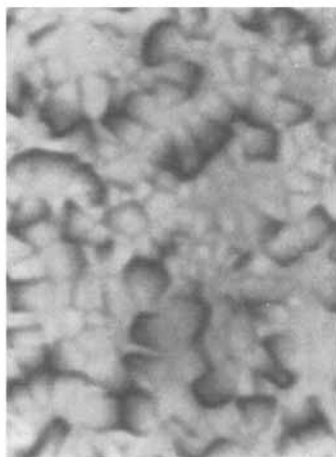


PLATE 2. E7, no voltage



PLATE 3. K15, 0.71  $V_{\text{rms}}$

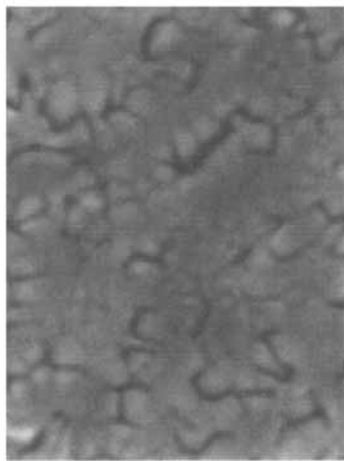


PLATE 4. E7, 1.03  $V_{\text{rms}}$

(See color plate XV at the back of this issue)

## DISCUSSION

Plates 1 and 2 provide evidence of the interaction of poled grains of PZT with both NLCs investigated. The change in birefringence of the NLC evident above these grains corresponds to an out of plane director tilt (no twist was observed). By applying an electric field to the poled devices to observe the Fredericks transition, the background birefringence of the NLC can be matched with that above the poled grains. This is demonstrated in Plates 3 and 4 for K15 and E7, respectively. The voltage in Plate 3 is  $0.71 V_{rms}$  (1 kHz),  $0.03 V_{rms}$  above that for the Fredericks threshold transition ( $V_{th} = 0.68 V_{rms}$ , measured experimentally). The voltage in Plate 4 is  $1.03 V_{rms}$  (1 kHz),  $0.10 V_{rms}$  above that for  $V_{th}$  ( $V_{th} = 0.93 V_{rms}$ , measured experimentally). This 'bias' voltage above the poled PZT grains,  $0.71$  and  $1.03 V_{rms}$  for K15 and E7 respectively, allows the maximum director tilt,  $\theta_{max}$ , at the centre of the device to be obtained. The voltage dependency of  $\theta_{max}$  is given by<sup>[31]</sup> Eqn. (2) which can be solved numerically, as is shown in Fig. 2(a),

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

where  $\kappa = (K_{33} - K_{11})/K_{11}$  and  $K_{11}$  and  $K_{33}$  are the splay and bend elastic constants, respectively. For K15,  $K_{11} = 6.0$  pN and  $K_{33} = 10.5$  pN while for E7,  $K_{11} = 11.1$  pN and  $K_{33} = 17.1$  pN. The value of  $V/V_{th}$  measured by matching birefringence as described is  $1.04$  for K15 resulting in a value for  $\theta_{max}$  of  $17^\circ$  while that for E7 ( $V/V_{th} = 1.11$ ) is  $31^\circ$ . The director profile above the poled grains can be calculated using Eqn. (3) as is shown in Fig. 2(b), where  $d$



is the thickness of the NLC, while  $z$  is the distance through the device.

$$\frac{z}{d} \frac{V}{V_{th}} = \frac{1}{\pi} \int_0^{\theta(z/d)} \left( \frac{\cos^2 \phi + \frac{K_{11}}{K_{33}} \sin^2 \phi}{\sin^2 \theta_{max} - \sin^2 \phi} \right)^{1/2} d\phi \quad 0 \leq (z/d) \leq 1/2 \quad (3)$$

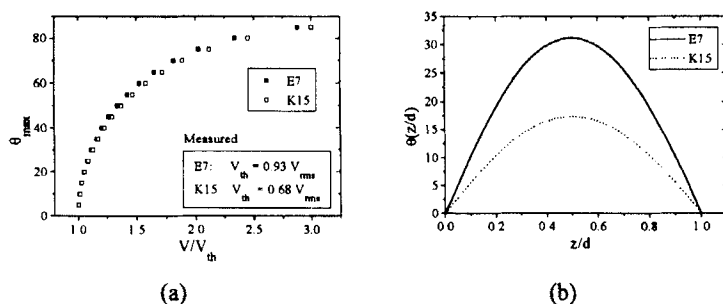


FIGURE 2 (a) Voltage dependency of  $\theta_{max}$  calculated from Eqn (2), and (b) the director profile above poled PZT grains for the devices studied.

Fig. 2(a) demonstrates that close to threshold, the uncertainty in  $\theta_{max}$  is greatest. Nonetheless, the uncertainty associated with the measurements of  $\theta_{max}$  is less than the difference between the K15 and E7 data. The above theoretical treatment does not include a surface energy term, and assumes a symmetric distortion of the director with no surface pretilt. Since  $\theta_{max}$  for E7 ( $31^\circ$ ) was found to be nearly twice that of K15 ( $17^\circ$ ) and the thickness of the E7 layer is also about twice that of the K15 layer, it is clear that surface interactions are important. In addition to the inclusion of surface anchoring

terms, in a more detailed analysis, it is appropriate to also consider the possibility of non-planar alignment being induced at the OFE substrate through flexoelectric interactions at the poled substrate.

## CONCLUSIONS

The technique of visualising poled domains in solid ferroelectric systems using nematic liquid crystals has been demonstrated to be useful for thin films of oxide ferroelectric ceramics. A first order approximation of the director profile above poled grains in the OFE for two different device thicknesses has been described. Preliminary results indicate that surface anchoring phenomena are significant in the interaction between the NLC and OFE. Further work is underway to establish the significance of surface forces (including possible flexoelectric contributions) and to incorporate them in the theoretical model.

## Acknowledgements

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