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Spontaneous induction of the uniform lying helix alignment in bimesogenic liquid crystals for the flexoelectro-optic effect

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Using in-plane electric fields, the electrical induction of the uniform lying helix (ULH) alignment in chiral nematic liquid crystals is reported. This process permits spontaneous induction of the ULH alignment to give an in-plane optic axis, without the need for complex processing. Flexoelectro-optic switching is subsequently obtained by holding the in-plane electrodes at a common voltage and addressing via a third, plane-parallel electrode on a second, or upper, substrate to give a field across the device in the viewing direction. For this device, in optimized bimesogenic materials, we demonstrate full intensity modulation and sub-millisecond response times at typical device temperatures. © *2012 American Institute of Physics.* [doi:10.1063/1.3682305]

Recent work on liquid crystal (LC) devices has focussed on improving the electro-optic response time, using moderate addressing fields, and there is currently significant interest in using faster switching materials and electro-optic effects for, for example, large area flat-panel television displays, holographic projection, and telecommunication phase devices. Existing technology, using conventional nematic LCs, is fundamentally limited by the viscoelastic response with typical "on" and "off" response times of the order of a few milliseconds or more. However, Patel and Meyer showed[1] that the flexoelectro-optic effect in chiral nematic LCs (N*) could be used to generate sub-millisecond response times albeit with weak optical phase modulation and relatively high fields. We showed recently, through design of bimesogenic LCs specifically for this effect,[2–4] that response times substantially less than a millisecond may be obtained using low electric field amplitudes ($<5 \, \mathrm{V}/\mu\mathrm{m}$),[5] with room temperature operation and below.[3]

In order to observe flexoelectro-optic switching in N* LCs, the direction of the applied electric field must be orthogonal to the helical axis. For conventional indium tin oxide (ITO) electrode structures, which are coated on the top and bottom substrates, the zero field LC alignment is in the so-called uniform lying helix (ULH) arrangement where the helical axis lies in the plane of the device.[1] Application of an electric field orthogonal to the helical axis then leads to an in-plane deflection of the optic axis. However, the generation of the uniform ULH texture, necessary for high optical contrast, has proved problematical. Various methods involving combinations of mechanical, thermal, and electric field cycling,[1–7] periodic boundary conditions,[8,9] complex lithographic processes, and electro-hydrodynamic effects using materials with high dielectric anisotropy, $\Delta\varepsilon$,[10] have all been used with varying degrees of efficacy. For example, in the latter case the critical electric field for unwinding of the N* LC to a homeotropic state is relatively low which limits the extent of flexoelectro-optic switching that can be achieved. Bimesogenic compounds, whereby the liquid crystalline moieties are attached end to end via a flexible spacer, avoid these problems and have led to the development of materials that exhibit both low $\Delta\varepsilon$ (<2) and enhanced flexoelectro-optic switching angles of over $\pm80°$.[5,11]

In this letter, we present a method of spontaneously aligning the ULH, in N* LCs optimized for the flexoelectro-optic effect, through purely electrical means at any temperature using a tri-electrode configuration (Fig. 1). An in-plane low frequency field applied across the surface electrodes induces a well-aligned ULH texture. Once this is achieved, the surface electrodes are electrically connected, at the same potential, and using the upper electrode, an electric field is applied across the cell to generate the flexoelectro-optic switching observed through crossed polarisers. In the schematic Fig. 1, the lower surfaces are the inter-digitated ITO electrode arrays with a width w of 3 μm and electrode separation s of 9 μm. The upper electrode is a uniform ITO surface

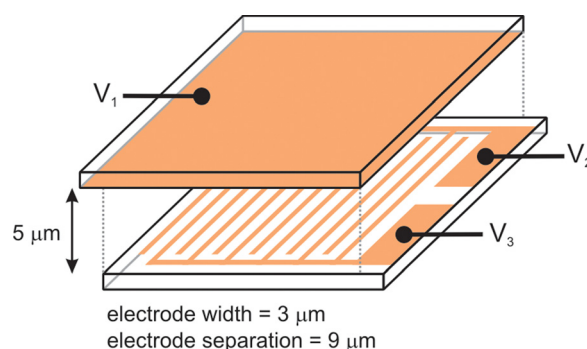FIG. 1. (Color online) Schematic of the electro-optic cell, comprising inter-digitated in-plane ITO-coated electrodes on lower glass substrates and a planar uniform ITO upper electrode, both substrates coated with planar alignment layers (rubbed parallel to the in-plane electrode long axis).

a)Authors to whom correspondence should be addressed. Electronic addresses: djg47@eng.cam.ac.uk, aiconess@lgdisplay.com, and hjc37@cam.ac.uk.
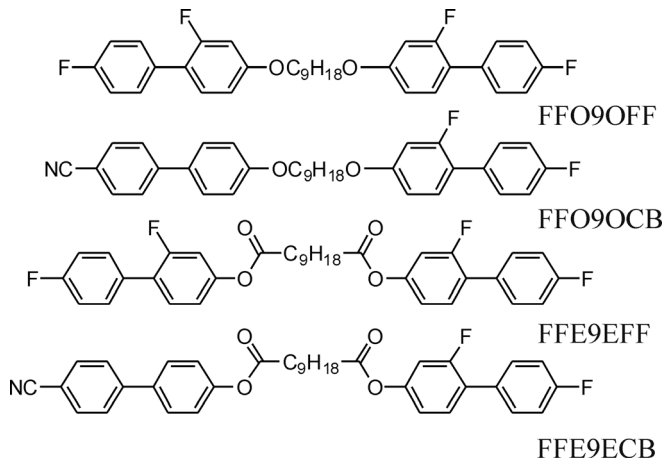
FIG. 2. Chemical structures of the fluorinated bimesogenic compounds used in mixtures.

and the cell gap d is 5 μm. The cell surfaces were pre-treated with antiparallel-rubbed polyimide alignment layers, and the direction of rubbing was parallel to the in-plane electrodes.

The N*LC materials used in the investigation consisted of a mixture of nematic bimesogens, shown in Fig. 2, of equal concentrations by weight. 3.5 wt. % of the high twisting power chiral dopant, BDH-1281 (Merck KGaA),[5,11] was then added to generate the chiral nematic phase. The phase sequence was I-(83 °C) – N*-(35 °C) – smectic LC, as determined from polarised microscopy at a cooling rate of 1 °C/ min. Materials were allowed to mix in the isotropic phase for a period of 24 h before filling into the test cells by capillary action. The microscope-based electro-optic apparatus used in this study is described elsewhere.[5] Electric field amplitudes E for the surface alignment mechanism were calculated from the in-plane applied voltage $(V_2-V_3)/s$ and, for the flexoelectro-optic switching mechanism, from the voltage applied across the cell $(V_1-V_2)/d$, where $V_2 = V_3$. The precise electric field profile, however, for both cases, will be non-uniform within the cell; particularly for the in-plane only mode.[12]

In zero electric field, the N*LC naturally adopts a Grandjean texture due to planar anchoring conditions imposed by the rubbed polyimide alignment layers. A ramping alternating 10 Hz electric field was then applied to the Grandjean texture using the in-plane electrodes (between V_2 and V_3) only (Fig. 3). Above a threshold value, $E_{th} \approx 2$ V$_{rms}$/μm, defects were observed to form above the interdigitated electrodes, and these coalesced over the inter-electrode regions as the field was increased. The texture became more uniform at

$\approx$10 V$_{rms}$/μm and, on field removal, relaxed to the ULH alignment. Between crossed-polarizers, the texture possesses a uniform bright, or dark, state when oriented at 45°, or parallel, to the polarizer transmission axis, respectively (Fig. 3). The frequency of the surface in-plane applied field is important; too high a frequency is not conducive to alignment, yielding a poorly aligned ULH texture. It is emphasized that the excellent ULH alignment was achieved by only using the planar alignment layers and application of an in-plane surface field at constant temperature; no further thermal cycling or mechanical treatment was needed. In this case a ramping electric field was used to illustrate the defect formation process (Fig. 3). In practice, all that is required to induce the ULH texture in these devices is the application of an in-plane low ac frequency electric field of the correct amplitude, e.g., typically 10 Hz, 10 V/μm, respectively. Once the alignment has been induced, polymer stabilization techniques can be employed to further ruggedize the texture.[13–15]

Following the induction of the stable ULH texture, flexoelectro-optic switching was obtained by applying a common voltage to the in-plane electrodes $(V_2 = V_3)$ and addressing across the cell via the third electrode on the top substrate (i.e., between V_1 and common V_2, V_3). Figure 4(a) shows the electro-optical response recorded through crossed polarizers, at 40 °C, when addressing the cell in this configuration. The optical response follows the polarity of the field, as expected for ULH flexoelectro-optic switching (red line). By addressing through an in-plane field only (i.e., no field across the cell gap), at similar field strength, no such modulation (blue line) was observed. In this case, clearly there is no macroscopic rotation of the optic axis in the plane of the device. Figure 4(b) shows the in-plane rotation of the optic axis ϕ, as a function of the applied electric field strength across the upper and lower substrates. The procedure for measuring ϕ is described in detail elsewhere.[2] It is well known[1–8] that ϕ varies according to

$$\tan \phi = \frac{e}{K}\frac{P}{2\pi}E. \tag{1}$$

Here, e and K are the effective flexoelectric and elastic coefficients, respectively, and P is the N* pitch. In accordance with Eq. (1), ϕ is found to vary in a linear fashion at low tilt-angles, reaching 22.5° at $E = 5.5$ V$_{rms}$/μm (Fig. 4(b)). For the mixture considered here ($e/K = 1.8$ C/Nm, $P \approx 320$ nm at 40 °C), the predicted switching angle is $\phi = 26.2°$ for the equivalent electric field strength, and this was confirmed using conventional LC cells with uniform ITO layers on

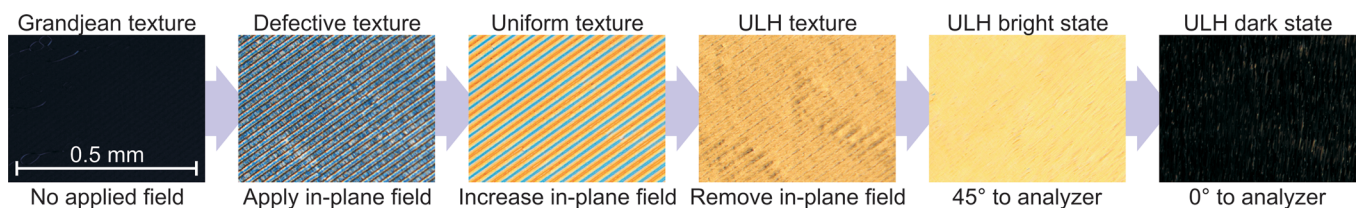FIG. 3. (Color online) Schematic diagram of the uniform lying helix texture induction process using interdigitated electrodes. Application of low frequency, in-plane (V_2, V_3) fields induce defects, which relax into the ULH alignment after the removal of the field. Flexoelectro-optic switching is achieved by applying a voltage across the connected in-plane electrodes ($V_2 = V_3$) and the top substrate (V_1).
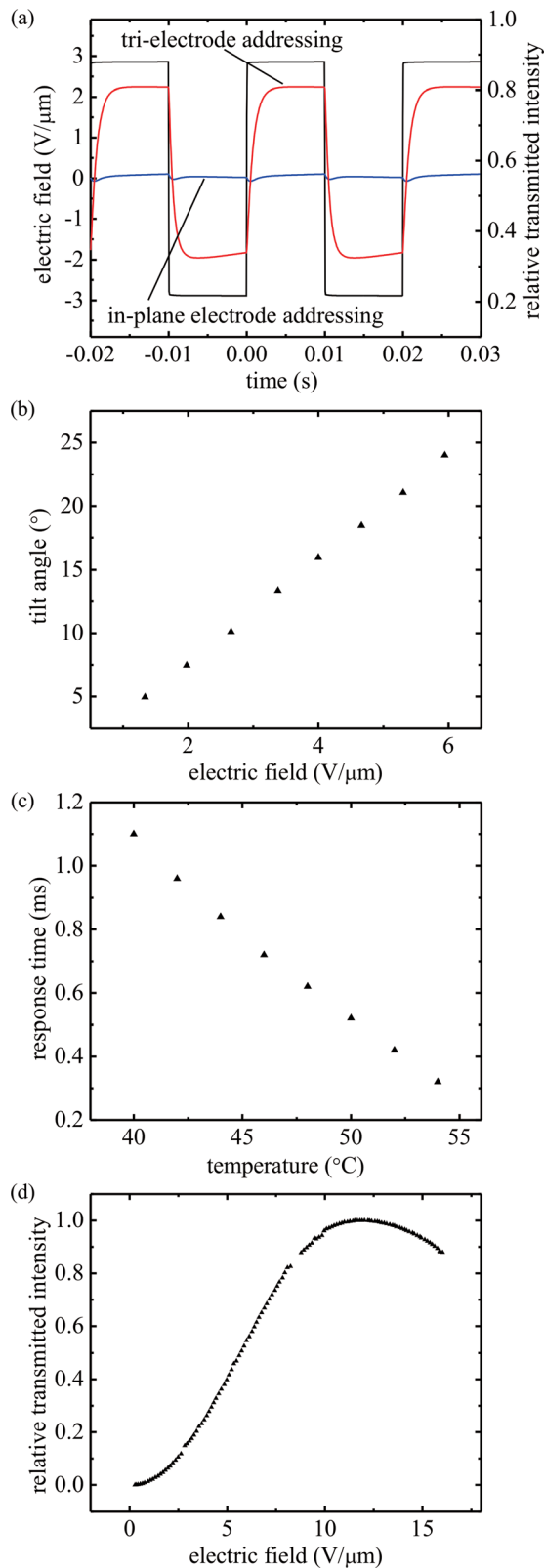
FIG. 4. (Color online) Electro-optic characterization of the device measured at 40 °C: (a) the relative transmitted intensity recorded with application of electric field (2.8 V$_{rms}$/μm), 50 Hz signal (black line plotted on primary axis) (i) between the substrate in-plane electrodes only (blue line, plotted on the secondary axis) and (ii) addressing between the lower in-plane electrodes and the upper plane-parallel electrode (red line, plotted on secondary axis), both with crossed polarizers. (b) The tilt angle of the optic axis as a function of the applied electric field. (c) The temperature dependence of the exactly superposable rise and decay response times. (d) The dependence of the transmission on the applied electric field showing full intensity modulation, through crossed polarizers.
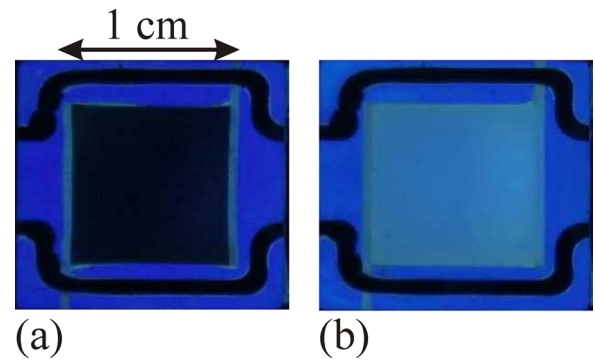

FIG. 5. (Color online) Photographs of the electrically induced ULH aligned texture, between crossed polarizers, on a cold-cathode fluorescent backlight. (a) The "off" state with the ULH optic axis positioned along the analyzer direction and (b) the "on" state with the application of an $E = 11$ V$_{rms}$/μm, 50 Hz electric field.

both upper and lower substrates. The slight reduction ($\approx 4°$) in the "effective" value of ϕ measured with the tri-electrode geometry is believed to be due to the slight non-uniformity of the field profile, due to the substrate bearing the in-plane gap electrodes.

Response times for 10% to 90% (rise) and 90% to 10% (fall) changes in light transmission for the ULH device are shown in Fig. 4(c) as a function of temperature for $E = 5.5$ V$_{rms}$/μm, which is the field needed to deflect the optic axis through $\phi = \pm 22.5°$. The rise and decay curves were symmetrical for each pulse and the response times (rise and decay) were numerically equal to each other. The response times decreased from $\tau = 1.1$ ms at 40 °C to $\tau = 500$ μs at 50 °C, due to the Arrhenius-like decrease of the viscosity with increasing temperature.

To demonstrate intensity modulation, around the zero field "off" state, the ULH optic axis is positioned (between crossed polarizers) in the same direction as the transmission axis of one of the polarizers to give zero light transmission. The average transmission, as the optic axis switches reversibly through $\pm \phi$ on field reversal is then proportional to $\langle \sin^2(2\phi) \rangle$. This is shown in Fig. 4(d), where the full average intensity modulation occurs at $\phi = 45°$, corresponding to $E = 11.8$ V$_{rms}$/μm. As expected, Fig. 4(d) does not exhibit a threshold, because the underlying flexoelectric effect itself is threshold-less (Eq. (1)). The absolute maximum light intensity transmitted is modulated by $\sin^2(\pi \Delta n d/\lambda)$, where Δn is the effective birefringence of the ULH structure and λ is the wavelength of light. Reducing the magnitude of the driving field, or more practically the voltage, may be achieved using a number of different methods. Firstly, the cell gap could be reduced by using materials with greater birefringence, leading to a reduction in the voltage required. Secondly, the addressing scheme could be modified, since the switching is electrically symmetric (no dc field bias) to use one $\phi = 22.5°$ switched state as the dark state, which requires one polarizer axis to be at 22.5° to the in-plane electrode long axis; this then halves the drive voltage requirements. Thirdly, through continued material improvements, an increase in the e/K parameter would further lower the effective field required.

Figure 5 shows the excellent uniform alignment and optical contrast of the tri-electrode induced ULH texture, of a large area cell, between crossed polarizers in the "off" (no

electric field) and "on" states (with an applied electric field of $E = 11\,\mathrm{V_{rms}}/\mu m$ at $f = 50\,\mathrm{Hz}$. The alignment is fully retained after switching the sample many hundreds of times. Contrast ratios of $\approx 50{:}1$ were obtained.

In summary, these results show that it is possible to induce stable ULH alignment in a chiral nematic liquid crystal using in-plane electric fields at ambient temperatures. ULH flexoelectro-optic switching is subsequently obtained by addressing via a third electrode across the device. The possibility of having a wholly electrical induced alignment is potentially important for practical application and manufacturability of next-generation devices employing the chiral flexoelectro-optic effect.

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