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Shear aligned polymer dispersed ferroelectric liquid crystal devices

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Fast switching liquid crystal devices can be produced by forming a dispersion of ferroelectric liquid crystal droplets in a polymer film. Such PDFLCs have been fabricated using a polymerization-induced phase separation technique involving ultraviolet photopolymerization, during which the film was sheared to obtain a uniform orientation of the liquid crystal medium. These birefringence devices show fast response times (sub-millisecond), optimum tilt angle (22·5°), and good contrast (~30:1) at room temperature, using ferroelectric switching. We studied the tilt angles, response times and contrast ratio as a function of voltage and temperature to determine the effects of the preparation parameters on the electro-optic behaviour of these devices. Using a ferroelectric liquid crystal with long helical pitch, such devices appear to be bistable.

1. Introduction

Polymer dispersed liquid crystals (PDLCs) consist of micron-sized droplets of low molar mass liquid crystal dispersed throughout a polymer binder [1]. PDLCs made using nematic liquid crystal operate as scattering devices, becoming transparent when a field is applied. Such materials are ideal for light shutters and projection devices due to their high contrast ratios. Other advantages of PDLCs are that polarizers are not required for operation, thus increasing light transmission and brightness [2]. Thin flexible PDLC devices can be easily fabricated from UV curable polymers and nematic liquid crystals, and quick and easy manufacture reduces production costs making such devices very attractive to the display industry.

Unfortunately some problems in the use of such devices still require consideration. The very distinct threshold characteristics required for high multiplexing rates have not yet been achieved with nematic PDLCs, so high definition displays currently require active matrix addressing. Such displays are expensive and difficult to manufacture. Also hysteresis can make some devices impractical for grey-scale displays.

A different approach must be taken to produce devices for rapid response display purposes. One such route replaces nematic liquid crystals with chiral smectics in a PDLC to produce a ferroelectric device with a high degree of bistability. The voltage characteristics and switching times indicate it may prove suitable for passive matrix addressing.

2. Polymer dispersed ferroelectric liquid crystal devices

The operation of a PDFLC film is based on the ferroelectric effect [3]. In order to achieve this, some particular geometrical arrangement of the liquid crystal molecules is required. The helical winding of the layer directors in the S_C^* phase must be suppressed, and the molecules must be aligned to form a unique director field.

This entrapment and alignment can be produced by forming a PDLC with the ferroelectric material confined within droplets, and applying a mechanical shear to the PDFLC film during phase separation. The droplets in the film will then be elongated by the shearing force, and the smectic liquid crystal inside the droplets will be forced to align in layers in a direction related to that of the shear.

The molecules in the droplets lie at a tilt angle θ to the layer normals when in the S_C^* phase. This generates a spontaneous polarization, P_s, in either the up (π) or down (σ) state. The operation of the PDFLC device is similar to that of a surface stabilized ferroelectric device [4]. The cell

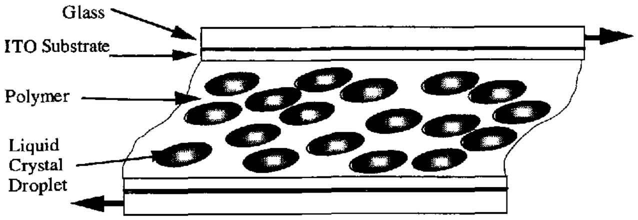

Figure 1. Sheared PDFLC, with droplets elongated in direction of shear.

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is placed between two crossed polarizers, oriented such that its direction of tilt is parallel to one of the polarizers in one switched state. On field reversal, the polarization is driven to the opposite state. This results in a ferroelectric switch by the molecules of 2θ, and resultant intensity change.

The maximum difference between the transmitted light intensity for the two orientations of the director can be expressed as

$$\Delta I = I_0(T_\pi - T_\sigma)\sin^2 4\theta \qquad (1)$$

where T_π and T_σ are functions representing the transmission in the π and σ states in terms of the refractive indices, sample thickness, and the wavelength. These functions are similar to the $\sin^2(\Delta n\pi d/\lambda)$ term in the conventional SSFLC device, but are convoluted by refractive index differences between the liquid crystal and polymer matrix, shape, structure factors etc. They will be empirical functions for each PDFLC device and its method of preparation.

3. Fabrication of PDFLCs

PDFLCs were manufactured using Norland Optical Adhesive NOA65 and the liquid crystal mixture SCE3 (Merck Ltd, U.K.), which exists in its chiral smectic C phase at room temperature (see below).

Phases of SCE3 K $<$ 0°C S_C^* 74·03°C S_A 107·15°C N* 138·54°C I

The liquid crystal was mixed with NOA65 in the weight ratio 1:2, and heated to above the isotropic clearing temperature to achieve good mixing. This mixture whilst still isotropic and fluid was introduced by capillary action into a prepared ITO coated glass cell, (20 μm spacer beads). The temperature was controlled throughout by a Linkam microscope hot stage.

The mixture was phase separated by a combination of thermal and UV photopolymerization induced separation processes to produce a dispersion of spherical droplets containing the smectic liquid crystal, within the polymer matrix. The cure temperature and UV power both determine the size and distribution of droplets formed [5]. An example of the droplet distribution produced is shown in figure 2. After the droplets had been formed; the liquid crystal was taken into the S_A phase and a shear force was applied to the cell at a predetermined rate of 3·5 μm s^{-1} to produce a uniformly oriented device. UV exposure was carried out during and after the shearing process, to complete the photopolymerization, and cure the polymer matrix as it cooled to room temperature [6]. A sheared device is shown in figure 3.

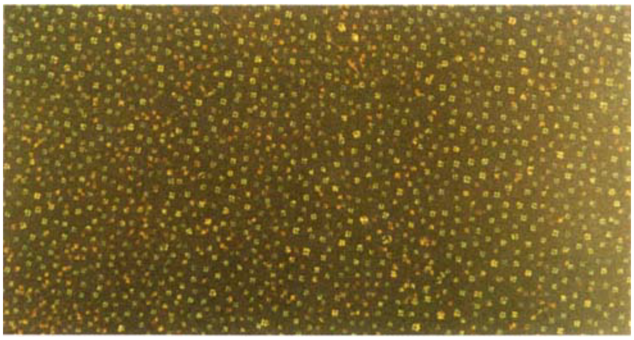


Figure 2. Droplets of SCE3 in NOA65 cured at 74°C with 4 μW cm^{-2} UV. Dark spheres are 20 μm spacers. Average droplet diameter 4 μm.

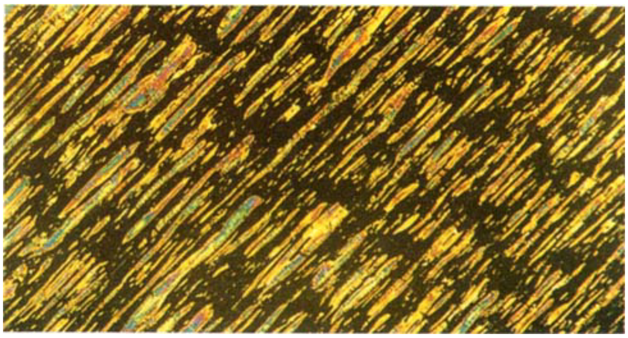


Figure 3. Same cell after shearing at 3·5 μm s^{-1}. Droplets elongated in direction of shear. Length varies from 10 μm to $>$ 100 μm.

4. Experimental

All optical microscope observations were carried out using an Olympus BH-2 microscope with Linkam hot stage.

The electro-optical measurements were performed using an apparatus in which the PDFLC cell was held in a rotating, temperature-controlled mount between crossed polarizers. The transmitted intensity of a helium–neon laser through the device was detected by a photodiode and measured on a digital oscilloscope. The driving voltage was supplied to the device from an amplified, programmable, function generator, and was also recorded on the digital oscilloscope [6].

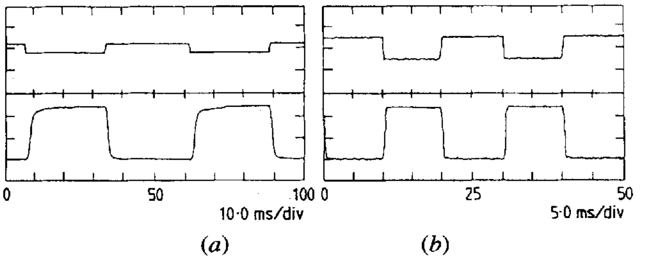
4.1. *Measurements*

4.1.1. *Electro-optic characteristics*

These devices show good electro-optic properties at room temperature. A 20 μm thick film can be switched by a 10 $V_{p\text{-}p}$ square wave, and showed an almost perfect square shape response when driven at 30 $V_{p\text{-}p}$, (see figure 4).

4.1.2. *Tilt angle*

Tilt angle was measured by recording the transmitted



(*a*) (*b*)

Figure 4. The electro-optic curves of the PDFLC film driven by a square wave at different voltages at a temperature of 40°C. The upper trace shows the applied square wave whilst the lower trace shows the electro-optic response. (a) $V_{P\text{-}P} = 10\,V$ and (*b*) $V_{P\text{-}P} = 30\,V$.

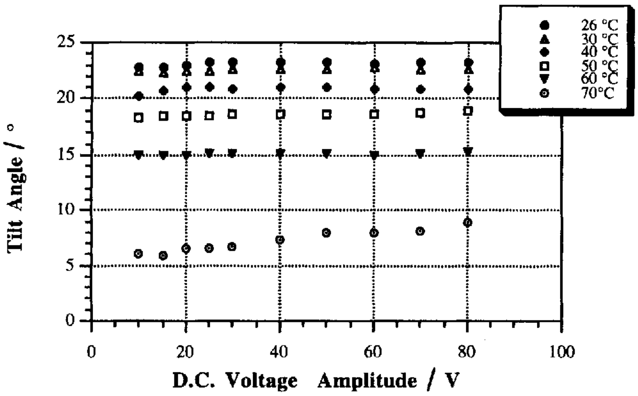


Figure 6. Voltage dependence of tilt angle in SCE3 PDFLC using 50 Hz square wave signal.

intensity as the device was rotated between crossed polarizers. When a switching d.c. field is applied, the phase of the output shifts by the induced tilt change of 2θ. This shift in phase is measured by recording the transmitted intensity of both states over 180° of rotation, and recording the minimum of both transmission curves.

The tilt angle was measured for the PDFLC with a square wave voltage of 15 V amplitude, and compared to pure SCE3. The result is shown in figure 5. It can be seen that the tilt of the molecules in the droplets is only slightly (~ 1·5°) smaller than in an SSFLC device. The actual cause of this may be the presence of small amounts of polymer in the droplets due to the incomplete phase separation of the polymer and liquid crystal, which would interfere with the tilt of the molecules. There could also be additional surface effects due to confinement of the smectic layers in three dimensions within the droplet. The device has a tilt angle of ~ 22·5° at room temperature.

The tilt angle showed no significant variation with applied voltage within the S_C^* phase above about 5 V (0·25 V μm^{-1}) (see figure 6). Within 4°C of the S_C^*–S_A transition (~ 70°C), the tilt had a small field dependency. The electro-optic switching was ferroelectric with possibly a weak electroclinic component at higher field strengths. The tilt showed a clear electroclinic dependence with voltage above the S_C^*–S_A transition.

There was evidence of bistability in these systems, as the switched molecules were observed to remain in their aligned state after the switching voltage had been removed. Further measurements were made to determine the extent of this tilt and its duration.

4.1.3. *Bistability*

The observation of the stability of a switched alignment after a pulse led to an investigation of the bistability of the devices in general. This was done by application of a procedure to measure the bistability number of these systems as defined in the work of Kitzerow [7]. The response and relaxation of the system to a sequence of fast negative and positive going pulses were recorded, and the relaxation after each pulse was measured. The response is shown in figure 7. The molecules do show a small relaxation after a short alignment pulse, but do not relax back to a central state as would be expected if the switching mechanism were based on helical unwinding [8]; instead they show an intermediate relaxed state until they are

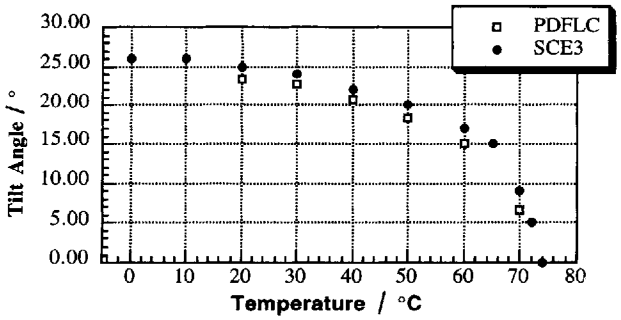


Figure 5. Temperature dependence of tilt angle of a 20 μm thick film of PDFLC containing SCE3 compared to that of pure SCE3 in a 2 μm thick SSFLC device.

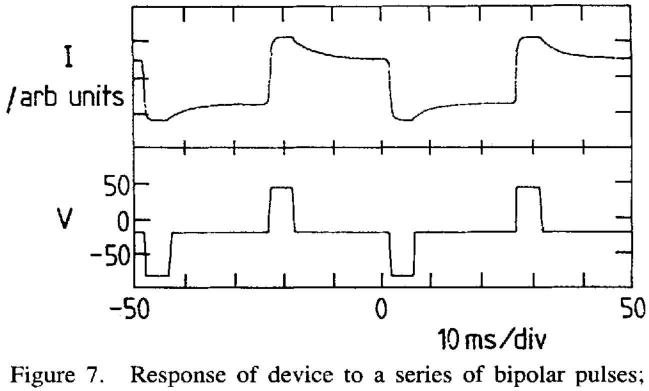


Figure 7. Response of device to a series of bipolar pulses; showing intermediate relaxed states. $G = 0{\cdot}5$.

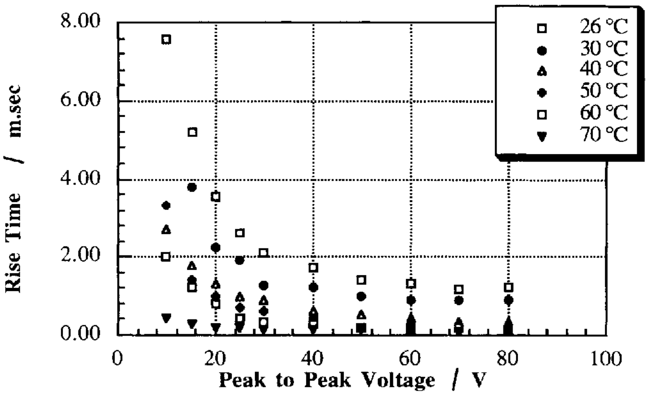


Figure 8. Variation of response time with a peak to peak voltage. 100 Hz square wave. Sample thickness 20 μm.

switched by the following pulse. The bistability number is defined as

$$G=\frac{\Delta I_{\mathrm{m}}}{\Delta I_{\mathrm{v}}} \tag{2}$$

where ΔI_{m} is the intensity difference in the field off states, and ΔI_{v} is the intensity difference during the applied pulse; a perfect bistable system would give a G of 1, and one with helical unwinding a G of 0.

The devices produced with SCE3 gave a bistability number of 0·45–0·55, indicating the system to be at least partially bistable. This figure is comparable to one reported by Molsen and Kitzerow for their systems produced with a low shear such that the droplets had barely deformed from the spherical. However, in our case, the system contained massively elongated droplets, with aspect ratios in excess of 25:1. They do not show the helical unwinding predicted [8] if the dimensions of the droplets exceeded the pitch of the chiral helix of the S_C^* phase. SCE3 has a long pitch of $>30\,\mu$m, and can therefore be expected to be stabilized in droplets elongated far from their 4 μm spherical diameter. However the wide spread of droplet lengths observed could indicate that some droplets contain constrained helices whilst others are free to unwind.

Measurements made by minimizing the intensities for the two relaxed states by rotation of the sample stage gave a relaxed tilt of 13° as compared with 22·5° at the fully switched state.

4.1.4. *Response times*

Response times were measured via the digital oscilloscope with the cell driven by a 100 Hz square wave signal, as a function of both temperature and voltage (see figure 8). The switching time was defined as the time taken for the transmission to vary from 10 to 90 per cent as the field reversal took place.

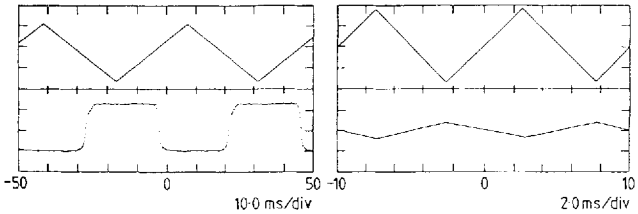
Clearly the material has a fast response to the external field. At room temperature, the response was ~2 ms for a 30 V peak to peak signal. Above 40°C the response is sub-millisecond for the same voltage; although this is not as fast as the times reported for a PDFLC device showing helical unwinding [7]. The response times show a dependence upon voltage and temperature. The strong voltage dependence of the switching times at low voltages is also indicative of bistable behaviour. The voltage response appears to follow an inverse field dependence, as in the case of a simple ferroelectric, i.e.

$$\tau_{\mathrm{s}}=\frac{\gamma\sin^2\theta}{P_{\mathrm{s}}\cdot E}. \tag{3}$$

However, it is not simply enough to use bulk measurements of P_{s} and γ to calculate a response time, as both will be affected by the confinement of the liquid crystal. Measurements of P_{s} and γ should be made to relate these values to those in a pure SSFLC device. Thought must also be given as to the actual field across the droplet before a true comparison with typical SSFLC switching times may be made. It is clear, however, that at moderate fields (i.e. $\sim 2\,\mathrm{V}\,\mu\mathrm{m}^{-1}$), sub-millisecond switching may be easily obtained in this PDFLC device.

It is possible to demonstrate the change from ferroelectric to electroclinic switching as a function of temperature using a triangular driving voltage—see figure 9. Using a 50 $\mathrm{V_{rms}}$ triangular wave the switching is clearly ferroelectric at 30°C (i.e. within the S_C^* phase), whilst at 87°C, some 12°C into the S_A phase, the switching is electroclinic with the optical response following the applied field. The effect of constraining the liquid crystal in a polymer matrix does not appear to affect the switching behaviour above the S_A transition anymore than in the S_C^* phase.

The contrast ratio (defined as $I_{\max}$–$I_{\min}/I_{\min}$) was measured on the apparatus. $I_{\min}$ occurred when one of the tilt directions of the aligned droplets was parallel to the polarizer, and $I_{\max}$ corresponded to the other switched state obtained on field reversal. The intensities were measured using the photodiode attachment and contrast ratio determined as a function of both temperature and applied voltage (see figure 10).



(*a*) (*b*)

Figure 9. Switching with a 50 $\mathrm{V_{rms}}$ triangular wave. The uppermost trace shows the applied wave form, whilst the lower shows the optical response viewed through crossed polarizers. (*a*) 30°C bistable switching and (*b*) 87°C electroclinic switching.

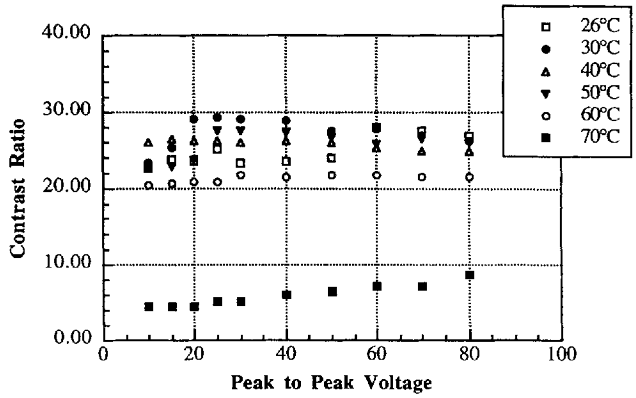


Figure 10. Variation of contrast ratio with voltage for an SCE3 PDFLC.

The device shows a CR of ~25–30 at room temperature. This compares reasonably with commercial SSFLCs with a CR of ~40 measured under the same experimental conditions. The contrast ratio shows a dependence on tilt angle as it varied with temperature and voltage, showing a maximum at ~30°C where the tilt angle is 22·5°. Above and below this angle, the transmission for I_{max} decreases as the switching angle is then slightly greater or smaller than 45°.

5. Discussion

It has been possible to fabricate a ferroelectric PDLC device, using a commonly available room temperature mixture SCE3, showing fast (sub-millisecond) switching at moderate field strengths ($\sim 2\,\mathrm{V}\,\mu\mathrm{m}^{-1}$) and good contrast (~25–30) at ambient temperature. As an alternative ferroelectric device, it has advantages in that such devices are far thicker than a SSFLC (20 μm as opposed to 2 μm) and far more rugged. As a display device, it suffers from the need for polarizers which reduces its brightness and contrast. The switching times and threshold characteristics also need to be improved for video frame rate passive matrix addressing. As a bistable device, it is only suitable for displays operating in black and white. A ferroelectric device based on helical unwinding would allow linear variation of transmitted intensity and therefore be suitable for grey scale applications.

There is considerable work to be done with these devices to realize their full potential and to establish the arrangement of the liquid crystal material in the elongated droplets and the exact mechanism of switching. The production of the droplets and the influence of phase separation rate, UV intensity, and shear rate on the dispersion produced must be studied in more detail. The study of the alignment of the molecules within the droplets is essential before the device can be fully understood. Other work has proposed that the material may adopt a chevron-like arrangement within the droplet [7]. This needs to be investigated for the case of these droplets. A more detailed study of the droplets in the S_A and nematic phases would also aid our understanding. Other work has also suggested that the same materials can be used to prepare devices that are either bistable or show helical unwinding [9]. This depends on the relative sizes of the S_C^* helical pitch and droplet dimensions. SCE3 has a long pitch, $>30\,\mu\mathrm{m}$, and it therefore will remain constrained in quite large droplets. It is proposed to study the influence of particle elongation on the characteristics of the switching produced, and relate this further to the liquid crystalline arrangements inside the droplets. We have observed partial bistability in these materials and are currently measuring how this is influenced by the preparation procedures and ferroelectric properties.

Finally, it is proposed to study the effect of polymer: liquid crystal concentration, and rate of phase separation on the formation of the droplets, the tilt angles obtained and also the switching characteristics of the devices fabricated using new siloxane containing liquid crystals [10] that have temperature independent tilt angles and very fast (i.e. microsecond) response times. From these studies we hope to determine the optimum composition and phase separation rate needed for good device production.

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