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## Liquid Crystals

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# A highly bistable fast-shear aligned polymer dispersed ferroelectric liquid crystal device

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We present a new shear aligned polymer dispersed ferroelectric liquid crystal material which in this configuration possesses the same electro-optical characteristics as conventional surface stabilized ferroelectric liquid crystals. The sample thickness is  $20\ \mu\text{m}$  as opposed to the  $1\text{--}2\ \mu\text{m}$  used in the normal surface stabilized geometry. The new material shows switching on the  $100\ \mu\text{s}$  timescale and very high bistability ( $G \geq 0.8$ ) minutes after removal of the field. The temperature independent apparent tilt angle is  $25^\circ$  at room temperature, almost equal to the ideal value of  $22.5^\circ$  for many display applications. The material is prepared from a solution of a novel low molar mass organosiloxane ferroelectric liquid crystal in a UV-curable polymer. We study the physical properties of the composite material and give a description of the fabrication technique and its optimization. The material and fabrication technique could be readily used for industrial production of devices including flexible display panels.

## 1. Introduction

In a normal ferroelectric liquid crystal display device the liquid crystal is usually confined in a geometry where at least one dimension is small compared with the pitch of the intrinsic helix of the material. The most widely used geometry is the surface stabilized ferroelectric liquid crystal (SSFLC) arrangement [1] where the liquid crystal is confined between two glass plates. Provided that the gap between the two plates is small compared with the pitch of the helix, the surface forces unwind the helix and create two stable orientations for the molecules which correspond to the intersection between the cone of the helix and the plane of the glass plates. In a uniformly aligned SSFLC device there are thus two distinct stable orientations for the major optic axis of the refractive index indicatrix. Since the molecules of ferroelectric materials have a transverse polarization, the molecules, and therefore the optic axis of the device, can be switched between the two stable orientations with a modest electric field of the appropriate polarity. The optical response time of a SSFLC is typically of the order of microseconds which is two orders of magnitude faster than conventional devices based on the nematic phase. However, because of the small cell gap, typically  $1.6\ \mu\text{m}$ , the industrial production of large area SSFLC devices is not straightforward; the devices are fragile and

susceptible to mechanical shock. In this paper we show how this problem may be overcome.

Molsen and Kitzerow [2] and Leader *et al.* [3] have shown that thicker and rugged devices can be fabricated using polymer dispersed ferroelectric liquid crystals (PDFLCs). The essential uniform alignment of the liquid crystal molecules is achieved by shearing the material during the phase separation that occurs between the liquid crystal and the polymer during photo-induced cross linking. In such a shear aligned PDFLC there are no restrictions in principle on the thickness of the device; the helix in the liquid crystal is always unwound by the confinement in two dimensions within the elongated droplets created by the shearing action. However, the size of the droplets and the quality of the alignment of the smectic layers are determined to a large extent by the relative rates of shearing, phase separation and polymerization of the matrix.

We present in this paper a new type of PDFLC material based on novel organosiloxane ferroelectric liquid crystals [4, 5] which can be shear aligned and yields high quality devices. The electro-optical characteristics of the devices produced are similar to those of the related SSFLC materials, but in samples which are  $20\ \mu\text{m}$  thick. The rate of shearing for the production of the devices is of the order of  $100\ \mu\text{m s}^{-1}$ , far higher than previously reported in the literature [2, 3]. For this reason we have called the device a fast sheared PDFLC (FS-PDFLC).

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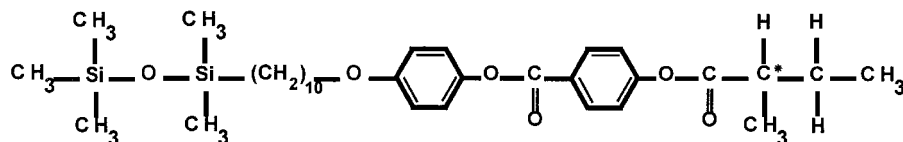


Figure 1. The chemical structure of the  $C_{10}A^*B$  molecule.

## 2. Materials and device fabrication

The starting mix for the fabrication of the FS-PDFLC is a one phase solution of one of the low molar mass ferroelectric organosiloxane liquid crystals synthesized in this laboratory [4–6] in a UV-curable optical adhesive (NOA-65 from Norland Inc., New Brunswick, NJ, USA [7]). The solution does not spontaneously phase-separate at room temperature. The phase separation is induced by photo-polymerization; a process that affords a good control of the rate of the phase separation under UV irradiation [8].

The particular liquid crystal used in the device presented in this paper is the  $C_{10}A^*B$  organosiloxane described by Newton *et al.* [6]. A schematic of the  $C_{10}A^*B$  molecule is given in figure 1 and some relevant physical properties of the material are summarized in the table. The composition of the mix for the fabrication of the FS-PDFLC device is a 1:2 weight to weight solution of the liquid crystal in the uncured prepolymer. Details of the computer controlled apparatus used for optimizing the rates of shearing and photo-polymerization are given in a separate publication [9].

For the fabrication of the FS-PDFLC, the solution is placed between two glass plates coated with ITO transparent electrodes. The glass plates are kept apart by glass sphere spacers  $20\ \mu\text{m}$  in diameter. The shear force is applied to the lower plate simultaneously with the UV illumination. The optimum parameters for the shearing-curing process at room temperature are  $7\ \text{mW cm}^{-2}$  of UV radiation power density at the surface of the PDFLC material and a shear rate of  $60\text{--}80\ \mu\text{m s}^{-1}$ . The total curing time is five seconds and the total shearing displacement of the order of  $375\ \mu\text{m}$ . A typical polymer matrix obtained by this technique is shown in figure 2. This shows the device viewed between parallel polarizers at a temperature above the clearing point of the liquid crystal. The parallel river-like grooves containing the liquid crystal are clearly visible due to the slight refractive index mismatching at this elevated temperature. The cross section width of the grooves is of the order of  $2\ \mu\text{m}$ .

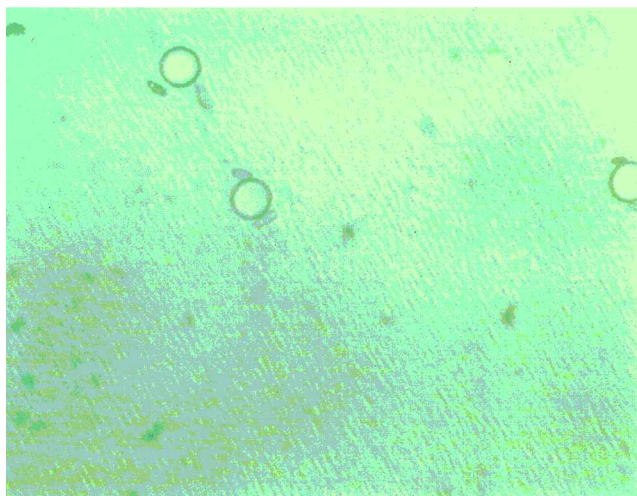


Figure 2. The FS-PDFLC device viewed between parallel polarizers at a temperature above the clearing point of the liquid crystal material (see the text).

## 3. Device characterization and performance

Many applications of ferroelectric liquid crystal devices rely on the optical bistability of a surface stabilized geometry. The degree of bistability depends on the competition between the surface forces which unwind the helix and the elastic forces in the bulk of the material which tend to restore the helix. In a perfectly bistable device, the surface forces dominate completely the elastic forces. In a real device, however, there is always some degree of relaxation towards the restoration of the helix after the switching electric field has been removed. A partial restoration of the helix degrades the quality of the optical contrast between the *on* and the *off* states of the device.

The degree of bistability can be quantified by the bistability number  $G$  introduced by Molsen and Kitzerow [2],

$$G = \left| \frac{\Delta I_m}{\Delta I_v} \right| = \left| \frac{I_m^{\text{on}} - I_m^{\text{off}}}{I_v^{\text{on}} - I_v^{\text{off}}} \right|$$

Table 1. The characteristics of  $C_{10}A^*B$  measured in the FS-PDFLC device and in a  $3.3\ \mu\text{m}$  thick SSFLC cell. The quantities quoted are the  $\text{SmC}^*$  to isotropic transition temperatures and the response time (at  $15\ \text{V}_{\text{pp}}\ \mu\text{m}^{-1}$ ), tilt angle and spontaneous polarization measured at room temperature ( $25^\circ\text{C}$ ). In both configurations the crystallization temperature is below  $-5^\circ\text{C}$ .

	Clearing temperature/ $^\circ\text{C}$	Response time/ $\mu\text{s}$	Apparent tilt angle/degree	Apparent $P_s/n\text{C cm}^{-2}$
FS-PDFLC	49	155	25	5
SSFLC	52	190	35	15

where  $I_v^{\text{on}}$ ,  $I_m^{\text{on}}$  and  $I_v^{\text{off}}$ ,  $I_m^{\text{off}}$  are respectively the intensities of the light transmitted through the device in the *on* state and that transmitted in the *off* state.  $\Delta I_v$  is measured in the presence of the switching field and  $\Delta I_m$  is measured a sufficiently long time after the switching field has been removed. Figure 3 shows a typical electro-optic response used for the measurement of the bistability number. For the measurements, the device is placed between crossed polarizers with the normal to the smectic planes at an angle of  $22.5^\circ$  from the direction of the polarization of the incident light. An ideal, perfectly bistable device in which the helix is completely restored in the absence of the switching field has a bistability number equal to 1 and a device in which the helix is completely restored in the absence of the switching field has a bistability number equal to 0.

The degree of bistability obtained in the FS-PDFLC devices is high, the bistability number being typically greater than 0.8 which compares favourably with the values of the order of 0.7 obtained previously [2]. The quality of the contrast between the two stored optical states is illustrated in figures 4 and 5 which show the

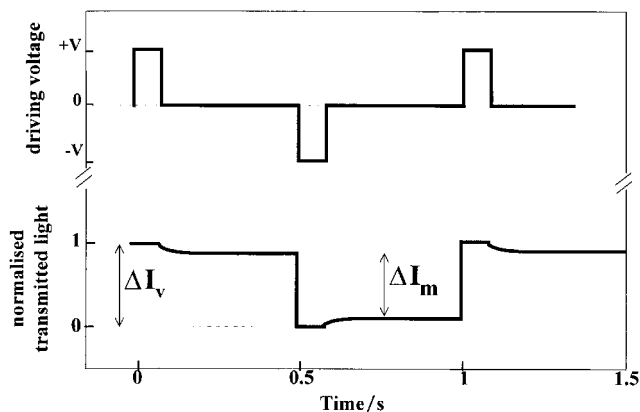


Figure 3. The electro-optic response of the FS-PDFLC device to a series of bipolar pulses. The normalized intensity of the transmitted light is given in arbitrary units for the measurement of the dimensionless bistability number  $G$ .



Figure 4. The FS-PDFLC device at room temperature in the *off* state with no field applied.

two relaxed states of the device viewed between crossed polarizers, in the latter case a few minutes after the switching field has been removed. We quantify the contrast by the ratio of the *on* to *off* intensity of the transmitted light, with the origin for the measurements of absolute intensities of transmitted light defined as the intensity measured between crossed polarizers in the absence of the specimen. The contrast ratio for the specimen in figures 4 and 5 is 10:1. We will discuss how this may be improved below.

#### 4. Discussion

Unlike Molsen and Kitzerow [2], who did not observe a threshold for ferroelectric switching, we observe a threshold field at  $0.7 \text{ V } \mu\text{m}^{-1}$ . Figure 6 shows the intensity of the transmitted light through the FS-PDFLC device as a function of the applied electric field. Another noteworthy difference is the order of magnitude greater shear rates used in the present work, with an optimum bistability being observed at shear rates of  $60\text{--}80 \mu\text{m s}^{-1}$  and only a gradual decrease in  $G$  at higher shear rates. The mechanisms responsible at the molecular level for the different behaviour of the organosiloxane based FS-PDFLC devices are still being investigated. A systematic study of FS-PDFLCs using different organosiloxane molecules is currently in progress. It would appear that the siloxane head group of the liquid crystal molecule plays an important role in the stability of the starting mix at room temperature and in the rate of phase separation. This in turn affects the shape and size of the droplets and the content of polymer within the liquid crystal droplet. Further, it would appear that 'molecular' phase separation [10], between the silicon rich and mesogen rich regions within the droplets, occurs and after curing helps to re-establish the bookshelf geometry. To complete the characterization of the

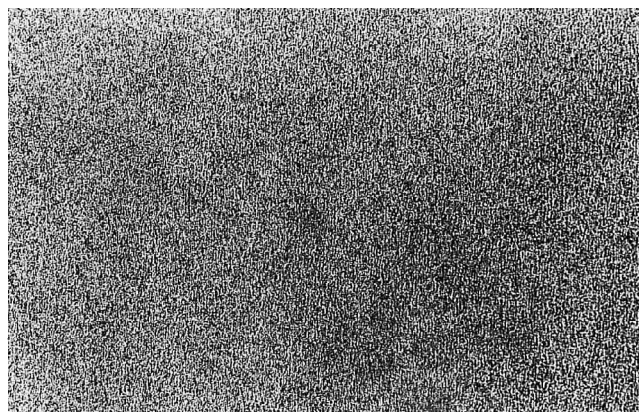


Figure 5. The FS-PDFLC device at room temperature in the *on* state with no field applied. The device has been switched with a short d.c. pulsed electric field.

Figure 6. The electro-optic hysteresis curve for the optimized FS-PDFLC device. The threshold field for switching is  $0.7 \text{ Vpp } \mu\text{m}$ .

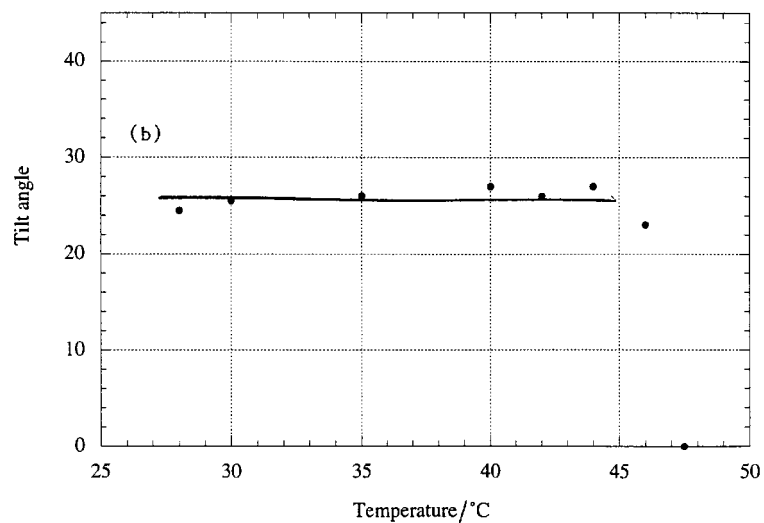
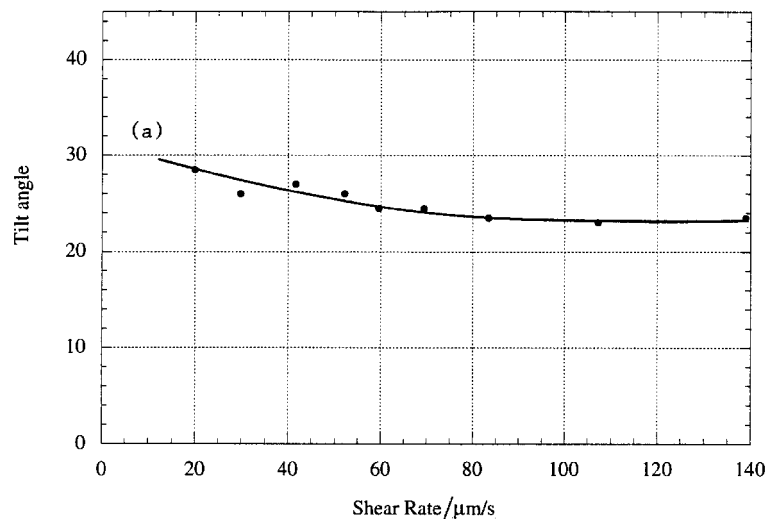
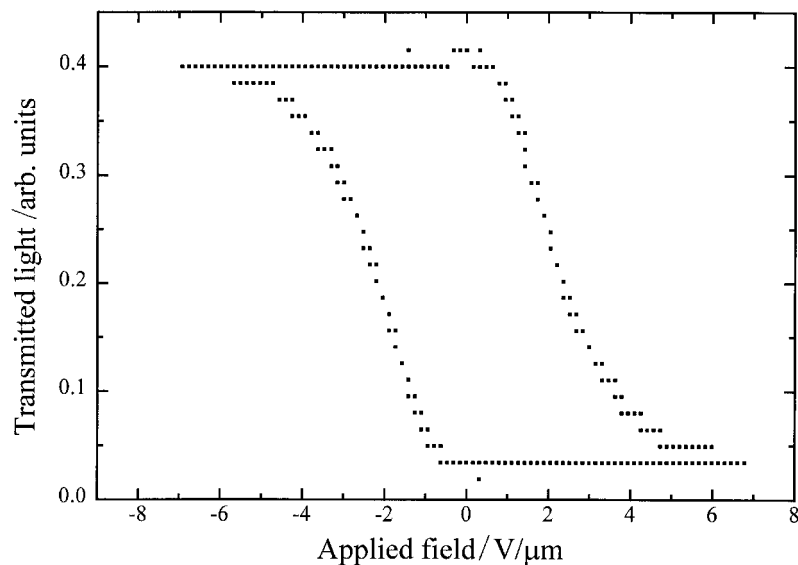


Figure 7. The apparent tilt angle of the FS-PDFLC as a function of (a) shear rate at  $28^\circ\text{C}$ , and (b) temperature for a shear rate of  $60 \mu\text{m s}^{-1}$ .

FS-PDFLC device it is essential to assess to what extent the intrinsic characteristics of the liquid crystal material are affected in the FS-PDFLC geometry. In the table the characteristics of  $C_{10}A^*B$  measured in the optimized FS-PDFLC device are compared with those measured for the same material in a  $3.3\ \mu\text{m}$  thick SSFLC.

The optical response time is defined as the time between the 10% to 90% change in the intensity of the transmitted light when the device is switched between the two stable states. The driving voltage for the measurements quoted in the table is a low frequency square wave of  $15\ \text{V}_{\text{pp}}\ \mu\text{m}^{-1}$ . The response times are of the same order in both devices, with the FS-PDFLC being marginally faster.

The transition temperatures were measured with a polarized light microscope equipped with a temperature regulated heating stage stable to within  $0.1^\circ\text{C}$ . The clearing temperature observed in both confined geometries is close to that measured by DSC ( $53^\circ\text{C}$ ) in a bulk  $C_{10}A^*B$  specimen.

The apparent tilt angles measured in the FS-PDFLC as a function of shear rate and temperature are shown in figure 7. The tilt angle was determined by optical microscopy as half the angle between the two positions of optical extinction when the sample is switched between crossed polarizers.

Firstly, the apparent tilt angle decreases gradually with increasing shear rate being optimal above  $60\ \mu\text{m}\ \text{s}^{-1}$ . Secondly the tilt angle is  $\sim 25^\circ$  at room temperature and independent of temperature over the entire  $\text{SmC}^*$  range. This value is close to the ideal of  $22.5^\circ$  for many applications. A temperature independent tilt angle is important for defining the axis of the polarizer during device fabrication. A change of tilt angle with temperature would lead to a reduced or varying optical contrast. It is interesting to note that the apparent tilt angle is reduced by some  $10^\circ$  in the FS-PDFLC as compared with that in the SSFLC geometry. This is presumably due to wall curvature and the greater percentage of polymer surface available to stabilize this geometry. Figure 8 shows the temperature dependence of the apparent spontaneous polarization  $P_s$ , measured by the current pulse technique [11]. In the FS-PDFLC cell the measured  $P_s$  is one third of that in the SSFLC cell. The reduction in  $P_s$  corresponds almost exactly to the concentration of liquid crystal material. This implies that the phase separation in the FS-PDFLC is almost complete and that the local value of  $P_s$  is the same in both geometries. The fortuitous reduction in the apparent tilt angle must therefore mainly be attributed to wall curvature at the droplet edges. Model geometries and experiments are being constructed to examine the subtle interface phenomena. The progressive drop in the

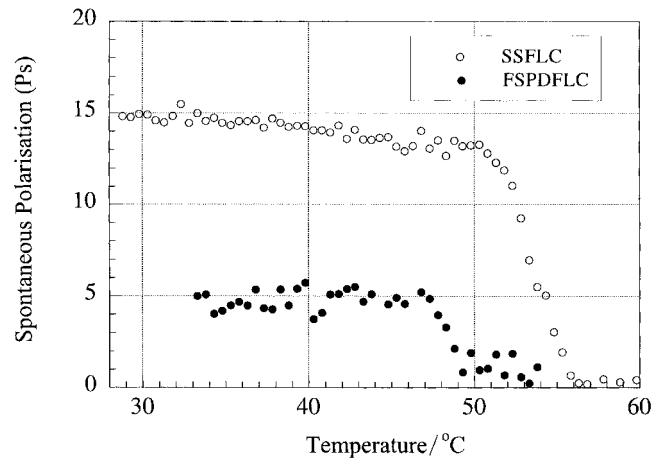


Figure 8. The apparent spontaneous polarization of  $C_{10}A^*B$  as measured in a SSFLC cell and in the optimized FS-PDFLC matrix.

measured  $P_s$  above the onset of the clearing temperature is due to a small biphasic range; the local spontaneous polarization remains constant.

## 5. Conclusions

The FS-PDFLC devices presented are highly bistable with a good contrast and are consistently reproducible. The devices can be heated above the clearing point of the liquid crystal material and cooled again without any significant loss of bistability, alignment, electro-optic response or contrast. The tilt angle and the temperature range of the ferroelectric phase, as well as the temperature invariance of both the  $P_s$  and the tilt angle make the device ideal for room temperature applications. The use of glass plates to support the PDFLC material is not an essential requirement. The fabrication process is readily adapted for a flexible substrate using photo-etched spacers, which when combined with the fast rate of shear at room temperature should enable the industrial production of large area flexible panels by roll to roll processes. Such processes typically take place at a rate of a few metres per minute which is entirely comparable with the fabrication shear rates ( $60\text{--}80\ \mu\text{m}\ \text{s}^{-1}$ ). As mentioned above, bistability was achieved in samples as thick as  $20\ \mu\text{m}$  rather than  $1\text{--}2\ \mu\text{m}$  in conventional SSFLC devices. Reduction of the thickness, in conjunction with adjustment of the birefringence of the ferroelectric material, will allow the optical contrast to be optimized so that the device operates as a  $\lambda/4$ , or indeed  $\lambda/2$ , waveplate. Clearly the use of cells thicker than  $1\text{--}2\ \mu\text{m}$  leads to simpler and less stringent production processes. Further the bistability in samples as

thick as 20  $\mu\text{m}$  suggests that such FS-PDFLC should make for interesting dye guest–host ferroelectric displays. The increased optical path length should give good colour contrast, whilst the possibility of single or zero polarizer devices [12] should lead to greatly improved brightness.

Finally, in the present devices we have used commercially available optical adhesives and these have allowed proof of concept experiments to be carried out. It is envisaged that future work will be carried out using other photo-crosslinkable polymer systems in which the refractive index of the host polymer may also be varied to improve further the contrast and also allow different liquid crystal/polymer interface phenomena to be studied to optimize or minimize, for example, the effects of anchoring energy, surface tilt and wall curvature.

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