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Optical Switching of Chiral Smectic C at Room Temperature†

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New chiral liquid crystal mixtures have been prepared, composed of a material exhibiting a smectic C phase and possessing a negative dielectric anisotropy and of a chiral material with or without, a chiral smectic C phase. These mixtures form room temperature eutectics with a large helical pitch and negative dielectric anisotropy. Uniform unwinding of the helix by an electric field has been obtained over large areas and leads to macroscopic ferroelectric domains of well defined optical properties. Chiral pitches and cell thickness were of the order of 15 μm, which is within the range of standard LCD fabrication techniques. Switching times of 3 ms have been obtained using 20 volts and latching of the switched status has been maintained under sustained AC fields we believe for the first time.

INTRODUCTION

Liquid crystals are now widely used for flat panel displays. The large number of picture elements (pixels) required for these displays has necessitated the use of large multiplex ratios which cannot be achieved with classical electrooptic effects such as the twisted nematic. Several teams of workers have studied fast electrooptic effects, some of which have a bistable long lasting memory,[1–5] i.e. dynamic scattering, cholesteric to nematic transition, smectic A dynamic scattering effect and thermodielectric effect in the smectic A phase. The development of

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non linear thin film devices such as thin film transistors[12,17] (TFT's) and back to back diodes[13] have allowed specific control of each pixel associated with the memory capabilities of these non linear devices. Short response times would also enhance the device performance.

The chiral smectic C, (Sm C*) has been reported[6-10] to have fast electrooptic effects and memory in the case of surface stabilized structures. These effects are based on the switching of permanent electric dipole moments **P** associated with the liquid crystal molecules. These induce a rotation of the optical axis and modify the light transmission between crossed polarisers. The **P**.**E** coupling with the electric field **E** is linear, and we show in this paper that one can make molecular mixtures of controllable chiral pitch that exhibit a Sm C* phase close to room temperature and that can be latched, after unwinding of the helix, in either of or two dipole orientations by a transverse AC electric field, provided the dielectric anisotropy is negative.

The smectic C liquid crystal phase is characterized by a tilt angle θ of the long molecular axis with respect to the normal to the layered structure. A typical value for the mixture reported here is $28 \pm 3°$ at 80°C. The symmetry point group is composed of a two-fold rotation axis parallel to the layers and normal to the long molecular axis, a reflection plane normal to the two-fold axis and a centre of inversion. As pointed out by Meyer et al.,[6] due to the chirality of their molecular structure, some liquid crystal species exhibit a chiral smectic C phase in which only the two-fold rotation axis remains. This allows a non-zero averaged permanent electric dipole moment parallel to the two-fold axis.

However, the chirality itself induces interaction between the molecules of adjacent layers and causes the direction of the long molecular axis to precess along a helical structure, of pitch of the order of a few micrometers the axis of which is normal to the layers. This gives no net polarization on a macroscopic scale.

In order to obtain ferroelectric domains of a given permanent dipole orientation, it is necessary to unwind the helical structure. This allows the dipoles to orient and switching of this orientation with a DC electric field.

MATERIALS AND EXPERIMENTS

The effect of unwinding the chiral smectic helical structure has been theoretically predicted and experimentally demonstrated by several authors.[6-8] In some cases, unwinding of the helix has been achieved

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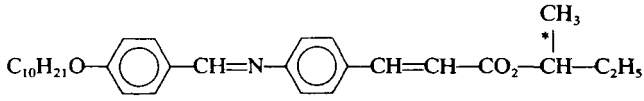
by particular boundary conditions[9-10] and the polarization obtained has given rise to fast electrooptic effects. However so-far this has been achieved with very small thickness of the liquid crystal material, of the order of only a few micrometers, which implies technical difficulties for fabricating displays.

In order to work with standard liquid crystal display thicknesses of the order of 5 to 15 micrometers using addressing fields perpendicular to the panel surface, which eases the display technology, we have synthesized chiral liquid crystal molecules[14,15] which are miscible with non chiral molecules exhibiting a smectic C phase and a negative dielectric anisotropy. This negative dielectric anisotropy is used for latching the molecular orientation by means of an AC electric field as we shall see below.

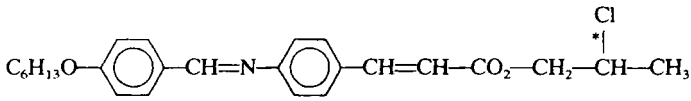
Our chiral plus non-chiral liquid crystal mixtures allow the preparation of long pitch chiral smectic C materials which favour electrical unwinding of the helical structure.

2.1 Materials

In addition to possessing a chiral smectic C phase a "good candidate" has to combine many properties such as, a great chemical stability, a convenient temperature range for the liquid crystalline phase, a long pitch of the helix in the chiral smectic C phase, desirable surface alignment properties, a negative dielectric anisotropy and a high polarization. As a matter of fact, none of the ferroelectric liquid crystals described by us in our previous publication[15] or in the literature,[16,17] actually possesses these qualities. The experimental investigations have almost exclusively concerned DOBABC and HOBACPC (p-decyloxybenzylidene p'-aminocinnamate of S (-) methyl 2 butyl) and p-hexyloxybenzylidene p'-aminocinnamate of R (-) chloro 2 propyle):



DOBABC



HOBACPC

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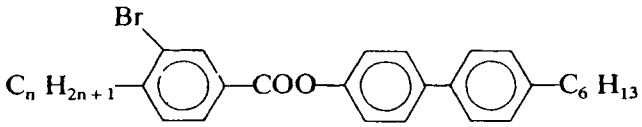
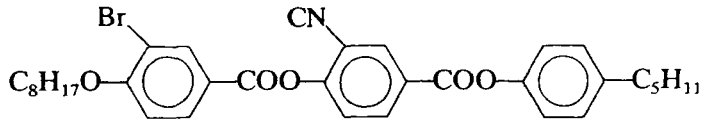
These compounds melt at around 70°C and have a low chemical stability. Efforts have been made to obtain room temperature ferroelectrics[16,18] but the pitch of the helix in the chiral smectic C phase is unfortunately too short (~3 μm) to work with standard liquid crystal display thicknesses of the order of 5 to 15 micrometers and the negative dielectric anisotropy is too weak. So, in order to obtain the required properties for our materials, we have synthesized[15] chiral liquid crystal molecules (**B**) which are miscible with non chiral molecules (**A**) exhibiting a smectic C phase and which allow the preparation of long pitch chiral smectic C mixtures. **A** belongs to the four following series:

① 

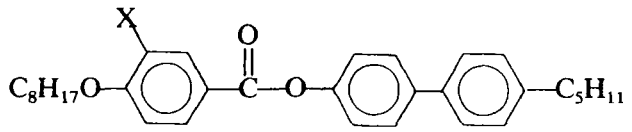
(A1 series)

n	K		S_C		N		I	
6	•	42°C	•	86°C	•	109°C	•	**(A1–6)**
7	•	56°C	•	84.5°C	•	109°C	•	**(A1–8)**
8	•	116°C	•	120°C	•	144°C	•	**(A1–8)**

② 

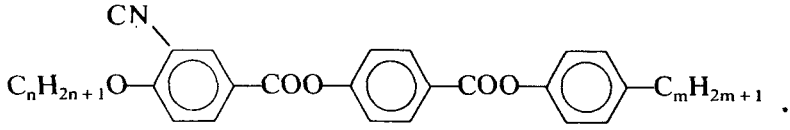
(A2)

K		S_C		S_A	N		I
•	89°C	•	93.5°C	—	•	113°C	•

③ 

(A3)

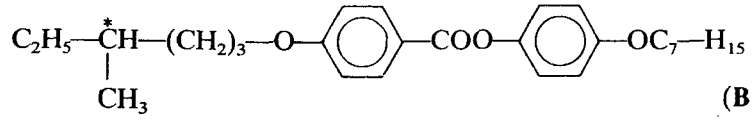
X	K		S_C		S_A		N		I	
Br	•	111°C	•	112°C	—		•	148°C	•	(A3Br)
CN	•	99°C	•	158°C	•	159°C	—		•	(A3CN)

④ 

	K		S_C		S_A		I
n = 8 m = 5	•	82°C	•	100°C	•	167°C	•

All these compounds have a negative dielectric anisotropy ($\Delta\epsilon \simeq -1$).

B is one of the following three molecules:

① 

(B1)

K		S_C^*		N*		I
•	30°C	•	46°C	•	62.5°C	•

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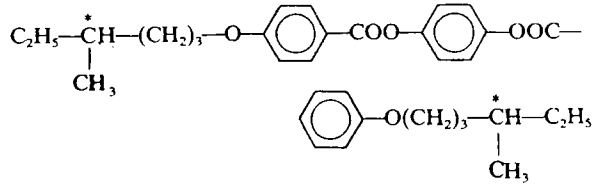

(B3)

TABLE

K_1		K_2		S_C^*		N^*		I
•	97°C	•	99°C	•	110°C	•	165°C	•

Typical N*, Sm A, Sm C* textures appear in the photographs of Figures 2 to 6 and an example of the phase diagram of a mixture is shown in Figure 1. The mixing of the two components (**A** + **B**) allows the preparation of Sm C* mixtures of adjustable helical pitch (Figures 5 and 6), with both a mesomorphic range close to room temperature and a negative dielectric anisotropy.

These materials were synthesized by standard procedures[15] from S (-) *2*-methyl *1* butanol and hydroquinone on *4*-hydroxy benzoic acid. They were purified by column chromatography over silica-gel (70-230 mesh) using toluene as eluant. All chemical intermediates were analysed by NMR (Bruker WP 80 SY) and IR (Perkin Elmer 283 B). The transition temperatures were determined by differential scanning calorimetry (DSC 4 Perkin Elmer) and the liquid crystal phases determined by the contact method and optical microscopy using a Zeiss polarising microscope in conjunction with a Mettler FP 5 hot stage.

2.2 Experiments

The electro-optical cells used in our experiments were composed of two parallel indium-tin oxide (ITO) coated glass plates covered with a thin layer[10] of SiO or polyimide which induced planar alignment of the molecules. The electric field induced across the ITO electrodes was perpendicular to the plates and parallel to the smectic layers. Slow cooling of the samples across the Sm C* transition temperature (Curie point) under a D.C. electric field prevented winding of the helical structure and yielded well aligned ferroelectric monodomains, the orientation of which depended on the direction of the D.C. field. As mentioned earlier in this paper, our liquid crystal mixtures had a negative dielectric anisotropy which not only helped unwinding the

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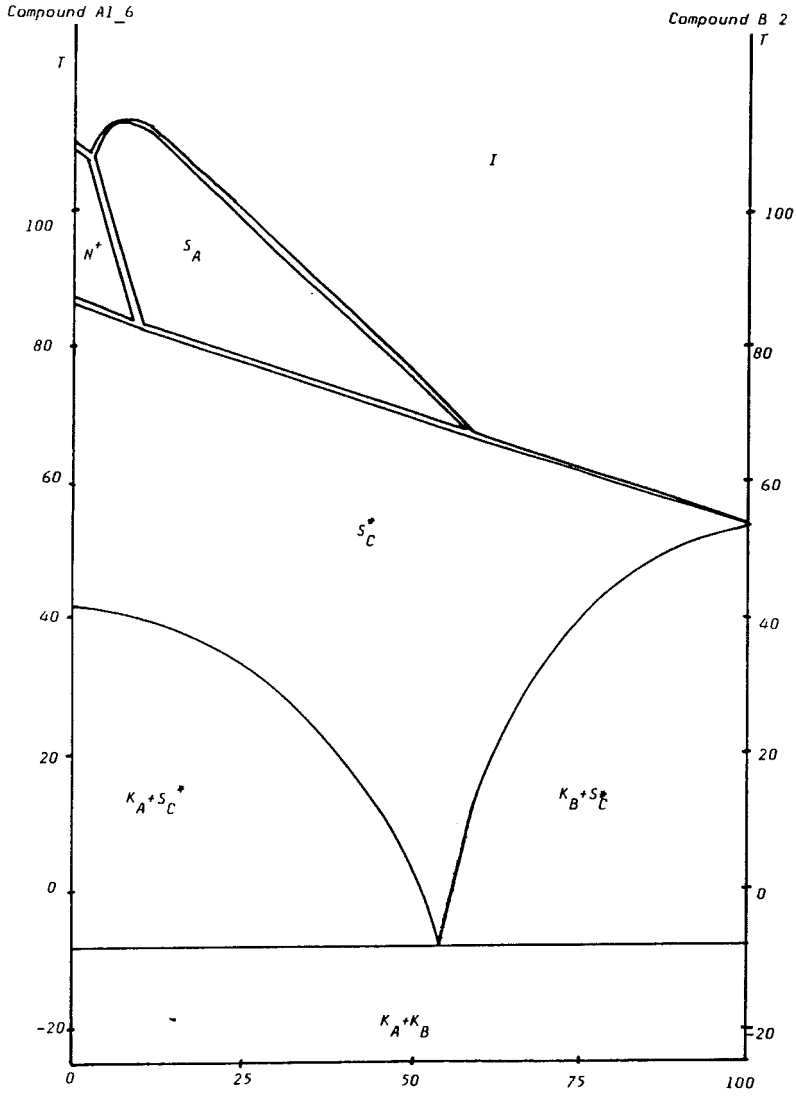

FIGURE 1 Phase Diagram between A1-6 and B-2

helix but also was used to latch the switched Sm C* states with AC electric field.

In the case of our samples, the chiral smectic C helical axis was oriented parallel to the cell glass plates. Taking the z axis of the reference frame parallel to this helical axis and assuming infinite

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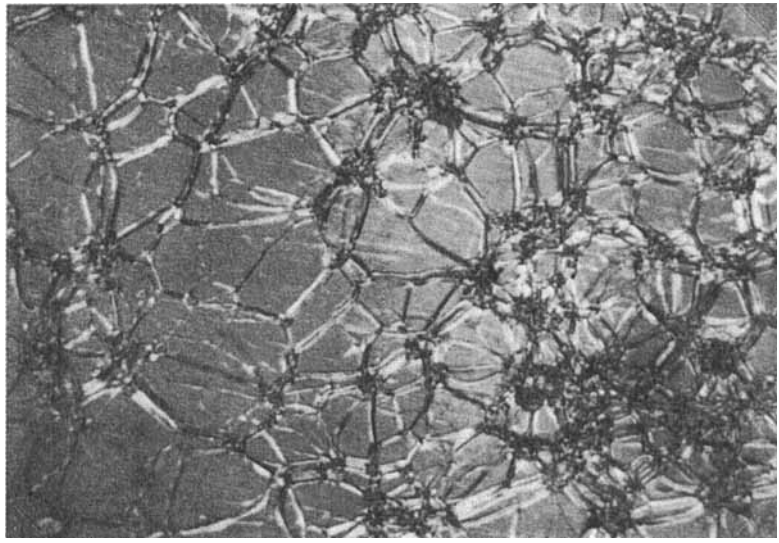

FIGURE 2 Cholesteric texture with oily streaks (planar texture) with **B1** material; T = 55°C; crossed polarizers; (X 250). See Color Plate XXXIX, located in this issue.

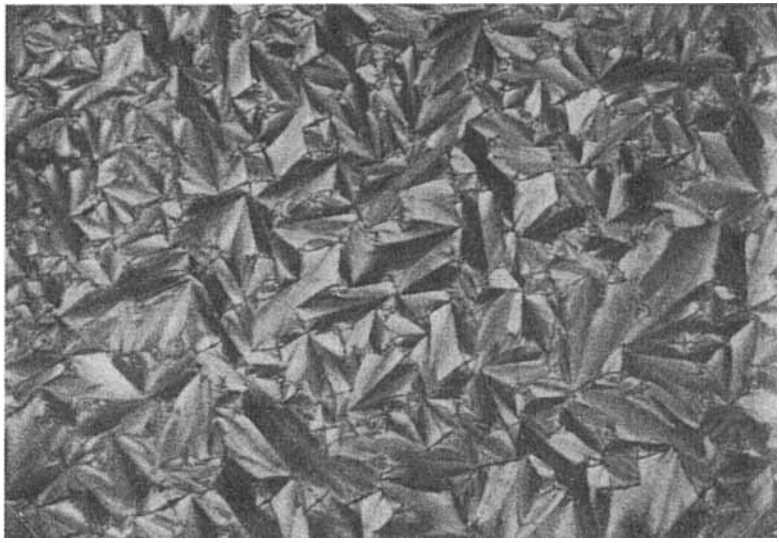

FIGURE 3 Smectic A fan-shaped texture with **A3** CN material T = 158,5°C; crossed polarizers (X 250). See Color Plate XL, located in this issue.

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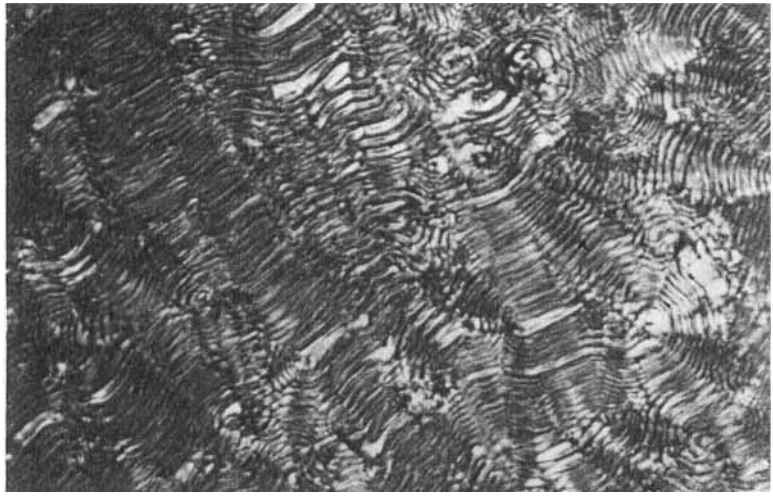

FIGURE 4 Chiral smectic C, fan-shape texture with equidistant lines, for **B1** material T = 42°C; crossed polarizers (X 250); small pitch (3 μm). See Color Plate XLI, located in this issue.

FIGURE 5 Chiral smectic C, fan-shape texture with equidistant lines, for 50% wt **A1**-6 + 50 % wt **B2** T = 55°C; crossed polarizers (X 250); large pitch (10 μm). See Color Plate XLII, located in this volume.

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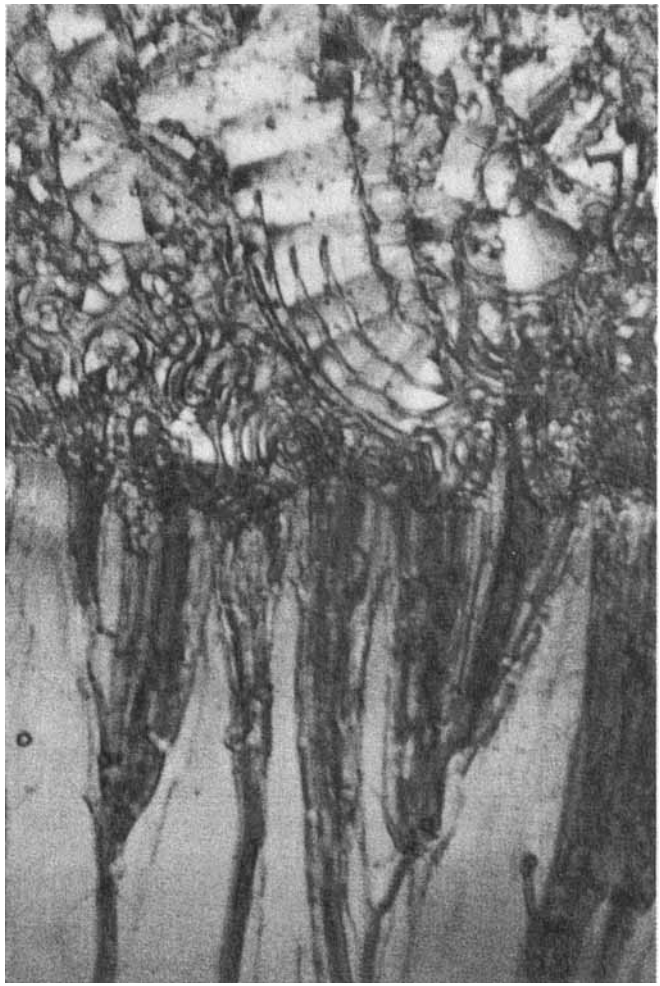

FIGURE 6 Contact between a cholesteric (**B1**) and smectic C (**A1-6**); $T = 60°C$; crossed polarizers (X 250). Chiralization lines appear at the contact boundary. See Color Plate XLIII, located in this issue.

length along the z axis, one can derive the following expression for the distorsion energy.[19,20]

$$F_d = \frac{1}{2} K_1 (\mathbf{div}\ \vec{n})^2 + \frac{1}{2} K_2 (\vec{n}.\mathbf{Curl}\ \vec{n} + q_0)^2 + \frac{1}{2} K_3 (\vec{n}\ \mathbf{x}\ \mathbf{Curl}\ \vec{n})^2$$

where: $\vec{n}$ is the molecular order director

K_1, K_2, K_3 are elastic constants (splay, twist, bend)

$q_0 = d\phi/dz$ equilibrium uniform twist

and the following expression for the action of the electric field on the system:

$$F_{dip} = \vec{P}.\vec{E}$$

$$F_{diel} = -\frac{1}{8} \Delta\epsilon\ (\vec{E} \cdot \vec{n})^2$$

$$F = \int_{-\infty}^{+\infty} \left[\frac{1}{2} K_0\, \theta^2 \left(\frac{d\phi}{dz} - q_0 \right)^2 + \left(\frac{\Delta\epsilon}{8\pi} \right) E^2\, \theta^2\, \mathrm{Cos}^2\, \phi \right.$$

$$\left. - P_0\, \theta\, E\, \mathrm{Cos}\, \phi \right] dz$$

where $\Delta\epsilon$ dielectric anisotropy

E electric field

θ molecular tilt angle

ϕ azimuthal angle

P_0 permanent dipole moment

K_0 twist elastic constant

$q_0 = \dfrac{2\pi}{Z}$

Z_0 helical pitch

For DC electric field, the threshold field is given by:

$$E_{DC} = 2\,\pi^2 \frac{K_2}{Z^2 P}\ (\text{SI units})$$

Numerical application

Although $\Delta\epsilon$ and P_0 have to be measured accurately, typical values obtained with compounds similar to those that we used are a good

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approximation:

$$K_2 = 5 \times 10^{-12} \text{ J/m}$$

$$\theta = 25°$$

$$|\Delta\epsilon| = 10^{-11} \text{ C/m}$$

$$P = 2.10^{-5} \text{ C/m}^2$$

$$Z = 15 \ \mu\text{m}$$

which leads to $E_{DC} = 2.10^4$ V/m.

Switching of the domains was induced by a reversible electric field. The switching times associated with the switching of the optical axis are shown in Figure 8 for temperatures ranging from 30°C to 45°C. The switching times extend from 3 to 10 ms for 15 μm thick samples under +20 V or −20 V.

Cell thicknesses used in our experiments do not allow surface stabilized latching of the Sm C* molecules, due to the competition between surface anchoring and torque elastic energy. To overcome this and obtain a memory effect, independent of the electrical addressing scheme, we have tried and succeeded in using, and AC electric field perpendicular to the cell plates. Given the negative

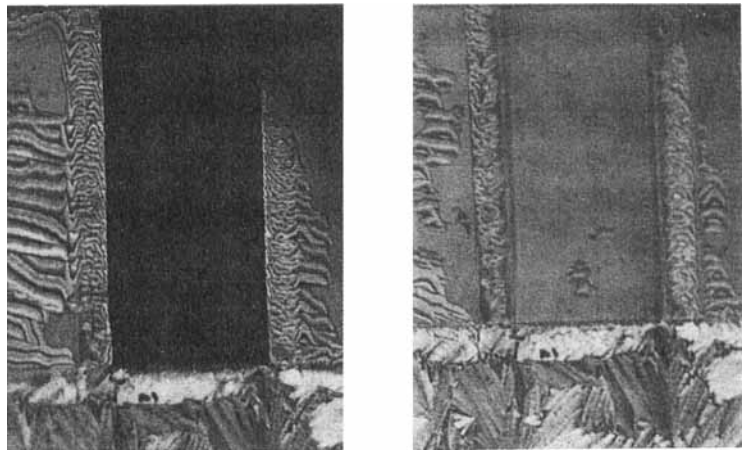

FIGURE 7 Electrooptic switching of Sm C* and AC latching of OFF and ON states in a matrix array. (picture element 0,4 × 0,9 mm). See Color Plate XLIV, located in this issue.

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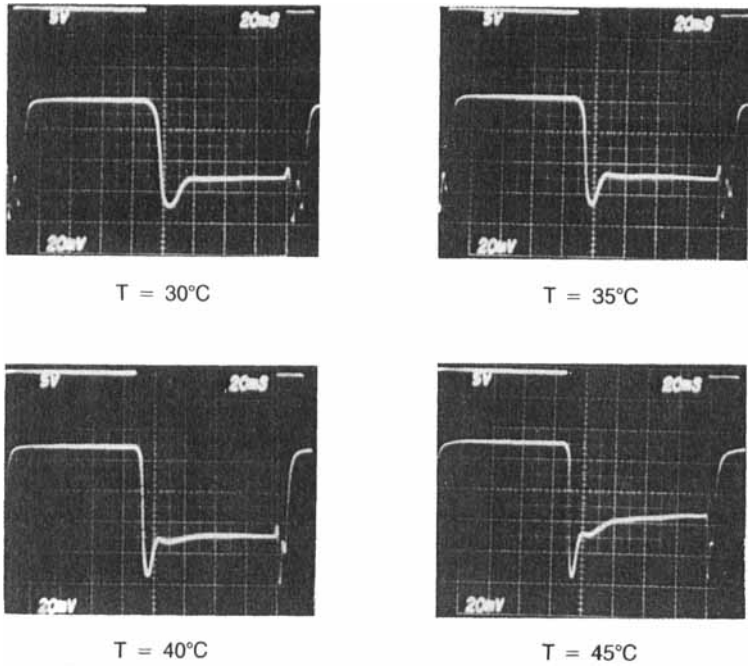

FIGURE 8 Sm C* switching response curves at four different temperatures.

dielectric anisotropy of our mixture, onset of this A.C. field when the D.C. switching pulse is turned off holds the molecules parallel to the plates in either of the two switched states. This is shown on the two photographs of Figure 7 where one can see a portion of a matrix array where a picture element 0.9 × 0.4 mm is switched on and off and held in either state by a 100 V, 5 kHz A.C. electric field. This opens the way to large multiplexing capabilities. Improvement of the materials to obtain larger values of the negative $\Delta\epsilon$ and thicknesses of 10 μm would allow a decrease in the amplitude of the A.C. field.

CONCLUSION

We have demonstrated in this work that it is possible to obtain Sm C* mixtures of adjustable helical pitch of the order of standard LCD thicknesses (10 to 15 μm).

Materials of low viscosity and high permanent polarization are desirable in order to improve switching time and lower the latching

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AC field. The memory effect obtained with the AC field is a first step toward multiplexing of a Sm C* panel.

Acknowledgments

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