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Bistable electro-optical switching in the smectic I* phase of a ferroelectric liquid crystal

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The ferroelectric switching behaviour of the highly ordered smectic I* phase has been investigated in the mixture which shows a S*_F phase at room temperature. The bistability was obtained in a 3.5 μm thick cell. Director switching and the reorientation processes have been studied by applying symmetric square and triangular wave pulses. It has been found that an asymmetric switching occurs in the smectic I* phase for low electric fields due to the hexagonal ordering of the molecules in the layer. This asymmetric switching was confirmed by optical microscopy and four stable states have been observed for low electric field. For higher electric fields only one state is stabilized which results in symmetric switching by both methods in the smectic I* phase.

1. Introduction

It is well-known that there are many kinds of chiral smectic phases such as S*_C, S*_F, S*_G, S*_K and S*_H, which are characterized as one dimensional solids within the layer structure [1]. It is also known that some of the smectic phases (highly ordered phases), which show no positional long range order within the layers, but which have a long range orientational order of the bonds. This bond orientational order usually occurs in a hexagonal pattern from which the name hexatics is coined. There is one phase, the smectic I phase, in which the molecules are packed in layers with their long axes tilted with respect to the normal to the layer planes. In a given layer of a domain all of the molecules are tilted in the same direction and the molecules are hexagonally close-packed [2, 3]. The tilt direction is towards the apex of the hexagonal net which is a major distinguishing feature of the smectic I phase in comparison with the other highly ordered smectic phase (e.g. S*_F).

As suggested by Clark and Lagerwall [4] the surface-stabilized device using highly ordered smectic phases has some advantages over the smectic C* phase. The memory effect is strong due to the correlation between the molecules, however, the device is slow because of the rigid structure of the molecules. Therefore, the device based on highly ordered smectic phases may find applications where the storage effect is more important than the speed of the device.

This paper is concerned primarily with the basic understanding of the switching mechanism, due to the hexagonally close-packed molecular structure in the layers, in a thin (3.5 μm), bistable cell of a room temperature ferroelectric liquid crystal mixture (ZLI-4004) which was provided by Merck, Darmstadt. Director switching processes

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have been studied by analyzing the current response for weak and strong electric pulses of a symmetric square and triangular waves. This process has also been studied by polarizing optical microscopy in the smectic I* phase. Recently we have also observed [5] in the same ferroelectric liquid crystal mixture a low frequency dielectric relaxation in both directions of the molecules (parallel and perpendicular to the layer planes) due to the hexagonally close-packed molecular structure in the layers.

2. Experimental

The liquid crystal cells were made of indium tin oxide coated glass plates separated by a $3.5\ \mu\text{m}$ thick spacer. The plates were first coated with adhesion promoter and then with a polyamide nylon solution [6]. The glass plates were baked at about 120°C for one hour and then rubbed unidirectionally. Material was introduced into the cell by capillary action at an elevated temperature to ensure that it was in the isotropic state during filling. The alignment and bistability was found to be excellent in surface treated cells as observed by us [7] in a $3.5\ \mu\text{m}$ thick cell of the S_C^* phase and also by others [8, 9]. The liquid crystal used in this study was a mixture of few compounds and has the phase sequence

$$C < -5^\circ\text{C} \quad S_I^* \quad 43^\circ\text{C} \quad S_C^* \quad 73^\circ\text{C} \quad S_A \quad 75^\circ\text{C} \quad I.$$

The electric pulses of symmetric square and triangular wave, generated by the pulse generator, were applied to a $3.5\ \mu\text{m}$ thick cell to study the director switching mechanism. The generator gives a symmetric pulse of perfect linearity between ($-24\ \text{V} \rightarrow 0\ \text{V} \rightarrow +24\ \text{V}$). This voltage is sufficient to cause a polarization reversal in $3.5\ \mu\text{m}$ film of the ferroelectric mixture in the smectic I* phase at 35°C . The experimental details and the effect of a resistor (R), a capacitor (C) and a polarization reversal device (P) of a ferroelectric liquid crystal cell to a symmetric square and triangular wave pulse has been given elsewhere [6]. Polarizing microscopy (Leitz orthoplan-Pol.) has been used to study the switching at different applied voltages in a bistable cell.

3. Results and discussion

In a thin cell of smectic C* liquid crystal the director is constrained to lie parallel to the cell boundaries with the smectic layers being aligned more or less perpendicular to the cell. Applying electric pulses of appropriate polarity above a certain threshold voltage we can switch between up and down states. The director is free to take any position, depending upon the tilt angle (θ), on a cone around the smectic layer normal as indicated in figure 1. The situation is different in highly ordered smectic phases (S_I^* , S_F^* , S_J^* , S_K^* etc.) as the molecules are hexagonally close-packed in the smectic layers, although, the molecules are packed in layers with their long axes tilted with respect to the normal to the layer planes as in the smectic C* phase.

In highly ordered phases the director takes definite positions on the cone because of the molecular hexagonal close-packing and does not change continuously as for the S_C phase. Uemura *et al.* [10] have studied how the continuous change of the tilt angle in the S_C phase due to the electric field is modified in highly ordered smectic phases. The relation between the hexagonal structure and the tilting direction of the director in the smectic I phase is shown in figure 2. It also shows the positional relationship between the hexagons and the bounding surfaces; one of the sides is parallel to the bounding surfaces in the S_I phase. When an electric field varies slowly, the director

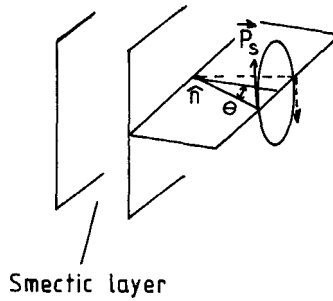


Figure 1. Geometry for a bistable ferroelectric liquid crystal cell. The orientation of the director \hat{n} and the spontaneous polarization P_s of the molecules in the up and down states are shown.

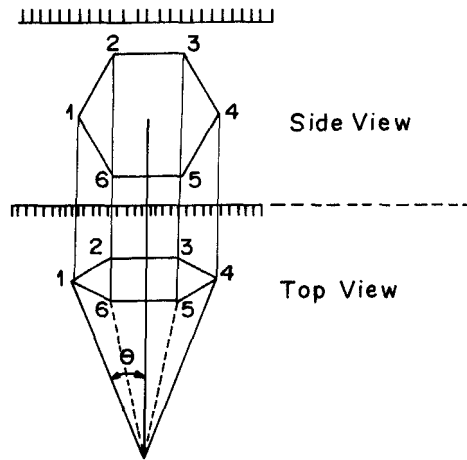


Figure 2. Side and top views of the hexagonal structure in the smectic I phase. Here θ is the director tilt angle which can take only six positions on the hexagon.

jumps discontinuously from (1) $\theta = 0$ to (4) $\theta = \pi$ via (2) $\theta = \pi/3$ and (3) $\theta = 2\pi/3$ or (6) $= -\pi/3$ and (5) $\theta = -2\pi/3$. Four pairs of extinguishing directions can be seen under a polarizing microscope. Two of them correspond to the (1) and (4) states and out of the remaining four positions only two (2 or 6 and 3 or 5) are seen under the polarizing microscope as the projected directions of (2) and (3) coincide with those of (6) and (5), respectively (see figure 2).

It is well known that in a bistable thin ferroelectric liquid crystal cell, prepared by a rubbing technique, the smectic layers are not exactly perpendicular but are tilted with respect to the surface of the glass plates which is called the pretilt of the director or tilted smectic layers. This pretilt which is constant in a particular cell, exists even in the highly ordered smectic phases e.g. S_1^* and it has been confirmed in the present study, by polarizing microscopy at a low electric field. It is worth mentioning here that the switching mechanism in the S_C^* and S_1^* phases is similar except because in the S_C^* phase the director is free to take any position on the surface of the cone (see figure 1) and in the S_1^* phase the director can take only six positions corresponding to the six edges of the hexagon (see figure 2).

Initially, we concentrate on the results obtained by applying symmetric square and triangular wave pulses to the $3.5 \mu\text{m}$ cell and then draw a comparison with those

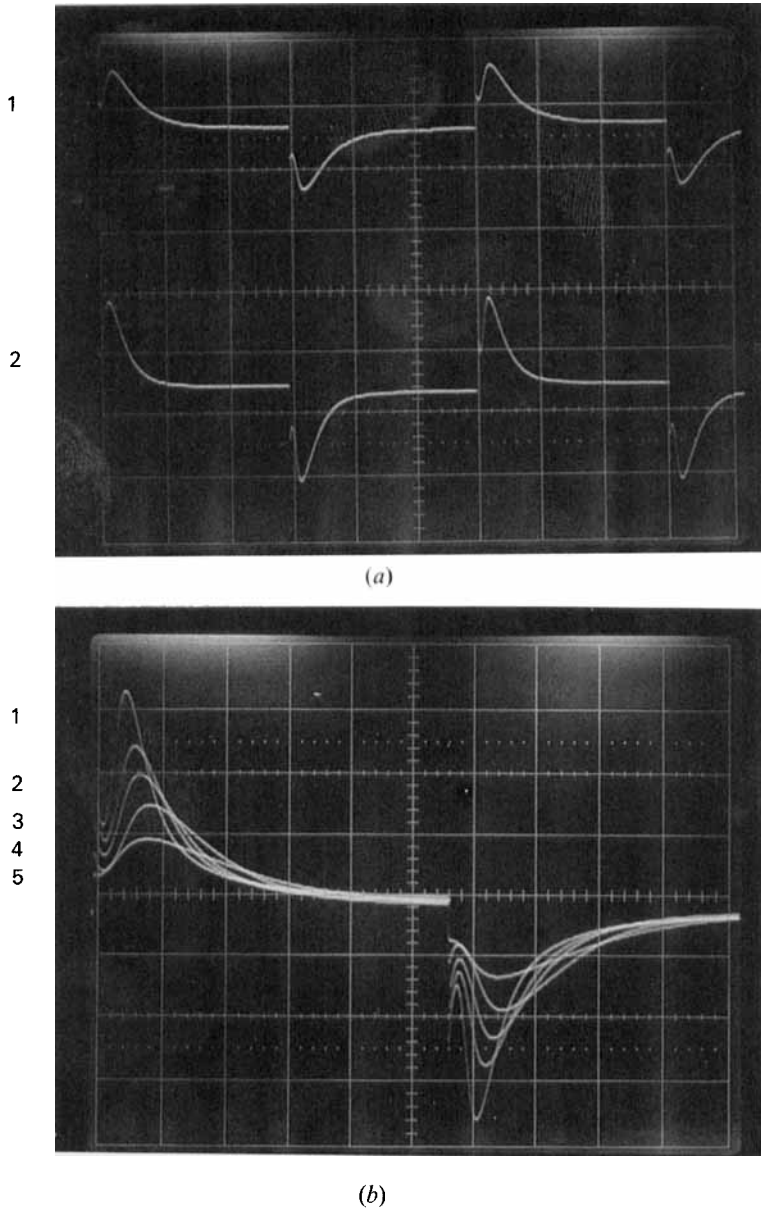


Figure 3. Oscillographs of the symmetric square wave pulses for a $3.5 \mu\text{m}$ thick cell in the S_C^* phase at 56°C for different applied voltages (applied frequency = 92 Hz). (a) (1) $V_{pp} = 6 \text{ V}$ and (2) $V_{pp} = 10 \text{ V}$ and (b) (1) $V_{pp} = 20 \text{ V}$, (2) $V_{pp} = 16 \text{ V}$, (3) $V_{pp} = 12 \text{ V}$, (4) $V_{pp} = 10 \text{ V}$ and (5) $V_{pp} = 6 \text{ V}$.

obtained by polarizing microscopy, particularly in the S_I^* phase. Figure 3 illustrates the oscilloscope trace at different voltages of the symmetric square wave of a $3.5 \mu\text{m}$ cell in the smectic C^* phase at 56°C . As the figure shows the response to a symmetric square wave, symmetrical about zero, is also symmetrical and the humps which represents the reorientating director are equally delayed by the response time [11]. It is also clear from the figure 3 (a) that switching of the director in the smectic C^* phase

is symmetric for low ($V_{pp} = 6$ V) and high ($V_{pp} = 20$ V) applied voltages of square wave pulses but the position of the hump shifts along the abscissa for lower voltages (see figure 3(b)), indicating a slower response time for lower voltages, which is consistent with the other data.

Figure 4 shows the oscilloscope trace for different voltages of symmetric square wave pulses in the smectic I^* phase at 36°C . It is interesting to note that in the smectic

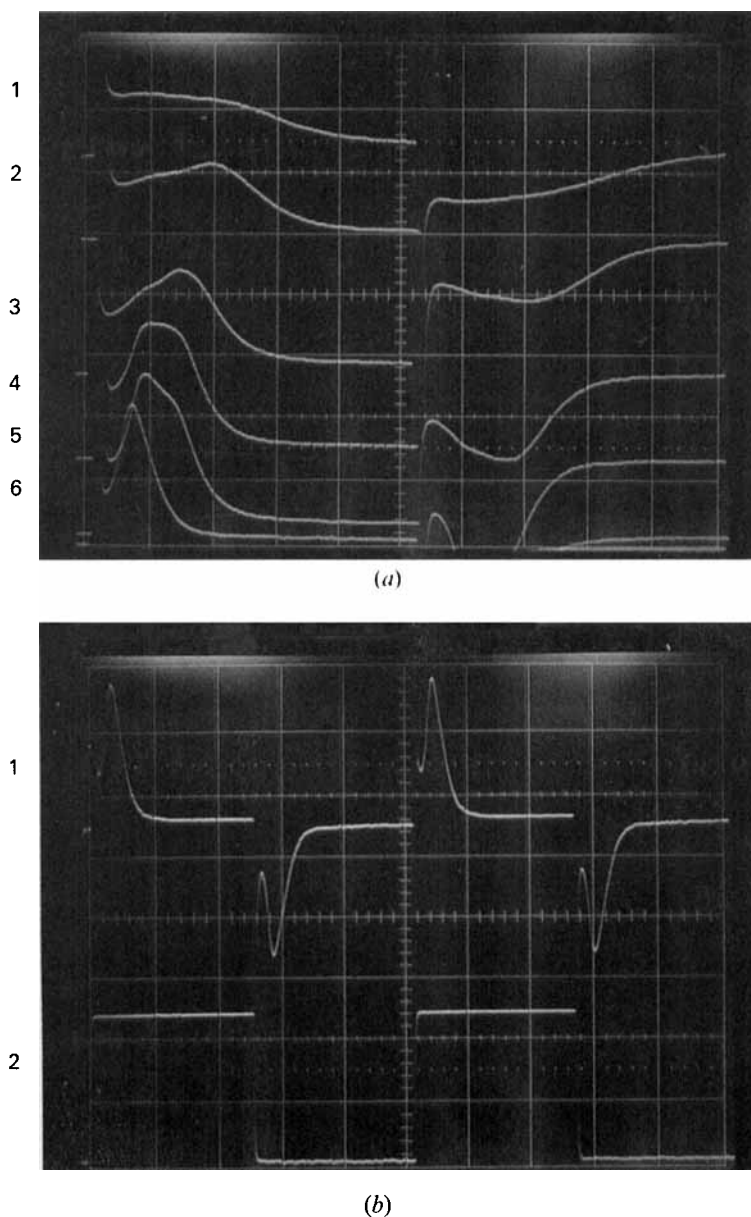


Figure 4. Oscilloscope of the symmetric square wave pulses for a $3.5\ \mu\text{m}$ thick cell in S_I^* phase at 36°C for different applied voltages (applied frequency = 10 Hz). (a) (1) $V_{pp} = 12$ V, (2) $V_{pp} = 14$ V, (3) $V_{pp} = 16$ V, (4) $V_{pp} = 19$ V, (5) $V_{pp} = 21$ V and (6) $V_{pp} = 23$ V and (b) (1) $V_{pp} = 23$ V and (2) applied square wave.

I^* phase switching is a complex phenomenon and not a symmetric one as observed in the S_C^* phase (see figure 3). For square wave pulses with a low applied voltages (about 15 V) it appears that more than one state is stabilized (see curve 2 and 3 in figure 4(a)) which results in an asymmetric behaviour on the left hand side of the square wave. As the applied voltage is increased (about 18 V) the position of the peak shifts to the right hand side of the square wave which may be due to the fact that other states are stabilized at the cost of previous ones (see curve 4 and 5 of figure 4(a)). At very high voltages (about 23 V) symmetric switching (see figure 4(b)) was seen, indicating that only one state is stabilized as in the S_C^* phase and the other three states are not stabilized. Similar behaviour has been observed by Goodby *et al.* [12] in a thick cell of HOBACPC ((R)-4-*n*-hexyloxybenzylideneamino 2'-chloropropylcinnamate) in the smectic J^* phase when a square voltage is applied. The switching occurs in an asymmetric manner and evidence was found of four different stable optical states. However, for thinner samples they have not observed these different stable states. The evidence for different stabilized states at lower applied voltages has also been observed by us under a polarizing microscope which is to be discussed later.

It is interesting to point out here that in the S_C^* phase switching is very fast ($\approx 100 \mu\text{s}$ at 45°C) as compared with the S_I^* phase ($\approx 3 \text{ ms}$ at 40°C) which is due to the rigid structure of the molecules in the S_I^* phase and this rigidity is brought about by increased correlation between the molecules both within and between the layers. Nevertheless, it is possible to switch the sample between stable states in a bistable manner at high electric fields in a $3.5 \mu\text{m}$ thick cell. The asymmetric switching behaviour in the S_I^* phase for lower voltages is because of the hexagonal structure within the smectic layer.

When a symmetric triangular pulse is applied to the cell in the S_I^* phase the asymmetric switching behaviour due to the polarization reversal current was not

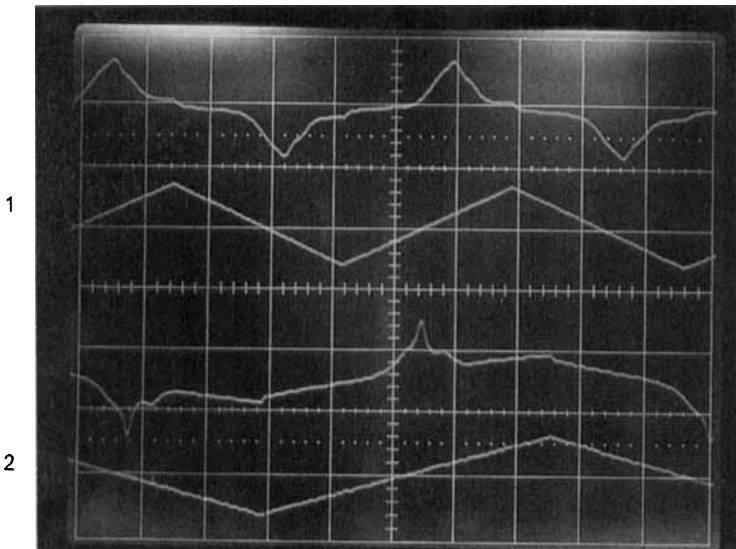


Figure 5. oscillographs of a polarization current response of a symmetric triangular pulse for a ferroelectric liquid crystal cell in the S_I^* phase at 36°C . (1) applied frequency of 22 Hz and (2) applied frequency of 1 Hz.

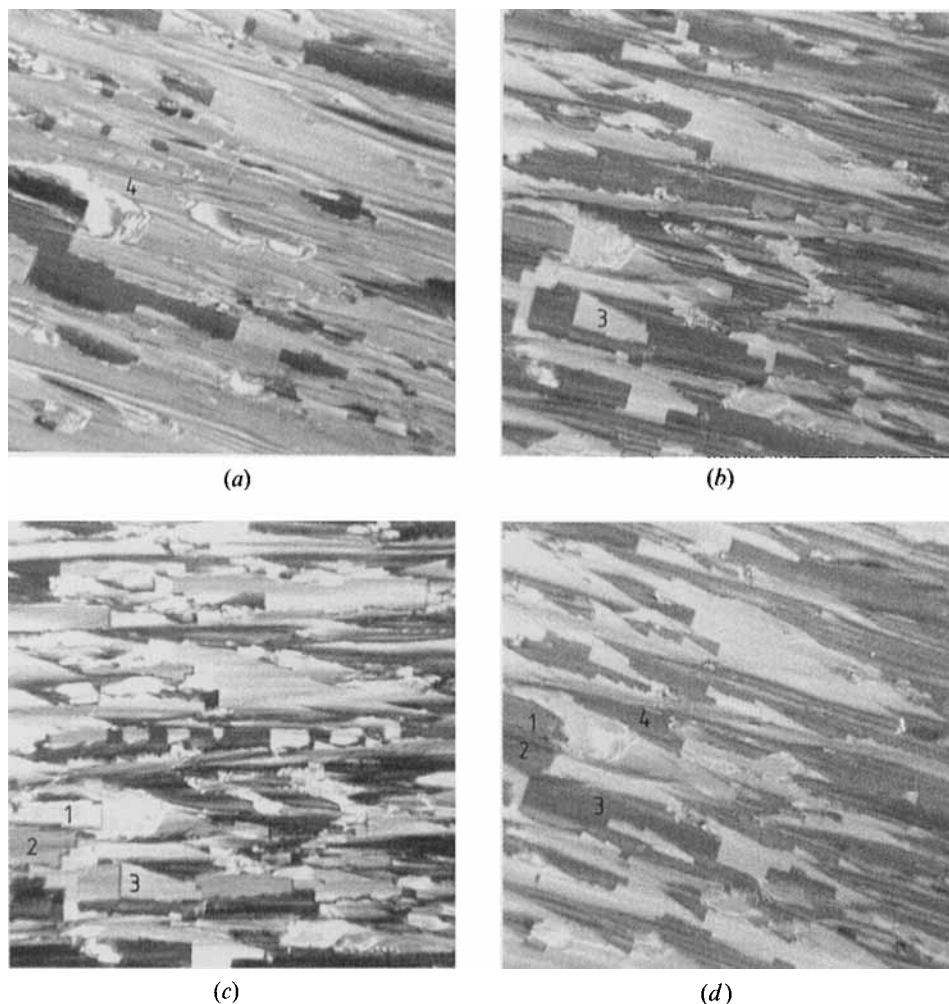
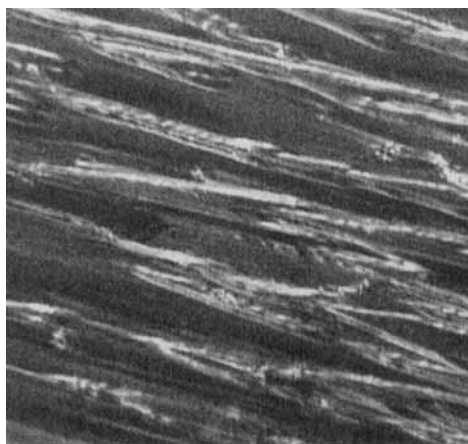


Figure 6. Optical photographs of a $3.5\ \mu\text{m}$ thick cell in the S_I^* phase at about room temperature (applied voltage = $3.5\ \text{V}$). The turn table is rotated by (a) 0° , (b) 9° , (c) 20° and (d) 11° successively.

observed as there was a clear asymmetric switching observed in square wave. Figure 5 illustrates the current response due to polarization realignment by a symmetric triangular wave for the $3.5\ \mu\text{m}$ thick cell at 36°C . However, it is worth mentioning here that at 1 Hz frequency the double peak (see curve 2 of figure 5) due to polarization reversal is not caused by asymmetric behaviour but due to the different director reorientation of the surface and bulk director which has been observed by us [7] and by Bawa *et al.* [6]. The two peaks have also been observed in the S_C^* phase in the present mixture for the $3.5\ \mu\text{m}$ thick cell.

Figures 6(a-d) are the optical photographs of the smectic I^* phase in a $3.5\ \mu\text{m}$ thick cell which have been taken at about room temperature by applying low electric fields (about $3.0\ \text{V}$). First a $3.0\ \text{V}$ electric field was applied for a few seconds to obtain the switching in the cell and then the field was removed. All of the photomicrographs were taken in the bistable state. Photographs 6(b), (c) and (d) have been taken after

rotating the turntable of the polarizing microscope by 9, 20 and 11° successively. Four states are clearly noticeable as indicated in the photographs which correspond to the four states shown in figure 2. The tilt angle (turn table rotation) for the corresponding four states agree very well with the theoretical calculations which has been done for the smectic I phase [10, 13]. It is worth noting that four states of switching has also been observed [10, 12] for 8SI* ((S)-4-(2-methylbutyl)phenyl-4'-*n*-octylbiphenyl-4-carboxylate) and HOBACPC in the smectic J* phase by applying a low electric field under a polarizing microscope. For higher electric fields (above 20 V) four state switching is not observed (see figure 7(a) and 7(b)).



(a)



(b)

Figure 7. Optical photograph of a ferroelectric liquid crystal cell in the S_1^* phase at about room temperature at (a) = +23 V and (b) $V = -23$ V.

From figure 4(a) it is also clear that the switching is symmetric for higher electric pulses of symmetric square wave, indicating that only one state is favoured (most probably state number 1 in figure 6). However, at low voltages there is no clear indication of four states (see figure 4(a)) by applying the symmetric square wave in

contrast to the clear indications of four states observed by optical microscopy (see figure 6) although we can very clearly sense (see figure 4 (a)) curve 2, 3, 4 and 5 that there is complex phenomenon taking place for lower voltages of square wave pulse in the smectic I^* phase due to the hexatic close-packing of the molecules in the layers.

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