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Effect of Electrical Properties of Orientation Films on Switching Current and EO Performance of FE-LCD

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Simultaneous observation of the temporal behaviors of the switching currents, and the electrooptic (EO) response, in ferroelectric liquid crystal (FLC) cells prepared by using molecular orientation films of rubbed polyimide (PI) and PI Langmuir-Blodgett films has been carried out. The effect on these phenomena of doping of a charge-transfer complex into the FLC medium also has been investigated. The behavior has been interpreted by investigating the effect of the internal depolarization fields originating from accumulated charges on the nonconductive orientation films such as ordinary rubbed PI.

In the case where PI-LB films were used, the switching current and the EO response were shown to start exactly at the instant when the applied triangular voltage waveform reversed polarity, whereas in the case of insulating orientation layers there occurred a phase advance both in the switching current and EO response. Through this study, the magnitude of the depolarization field was estimated.

Keywords: *ferroelectric LC, switching current, LB film, polymode, charge-transfer complex*

1. INTRODUCTION

The observation and analysis of the switching current passing through a ferroelectric liquid crystal (FLC) cell for applied-triangular or square-voltage waveforms are useful to characterize its electrical properties.^{1,2}

The present research work has been done with the aim of knowing how the electrical properties of orientation films and of the FLC medium affect the temporal behavior of the switching currents and the EO responses for triangular-voltage waveforms, and of evaluating the magnitude of the depolarization fields that degrade the memory capability in FLC EO devices.

We adopted two kinds of orientation layers; one was made of ordinary rubbed polyimide (PI) films and the other was polyimide (PI) Langmuir-Blodgett film.³ The basic FLC material used in this work was ZLI-3654 (E. Merck), either doped with a charge-transfer-complex CTC (TMTTF-ODTCNQ),⁴ or undoped. For a

comparison, other FLC materials were also examined to investigate the effect of the value of spontaneous polarization; actually they were CS-1014 (Chisso, $P_s = 5 \text{ nC/cm}^2$) and Felix-002 (Hoechst, $P_s = -42.4 \text{ nC/cm}^2$).

The variations of the initiation times of both the switching currents and the EO responses for triangular-voltage waveforms were shown to depend on the electrical properties of the orientation films, the ionic properties of the FLC medium, and the frequency of the applied voltages and their amplitudes.

Experimental data have been analyzed by adopting a formula developed by Chieu and Yang⁵ for depolarization field, the magnitude which was evaluated.

2. EXPERIMENTAL

The FLC materials ZLI-3654 (E. Merck), both doped with CTC (TMTTF-ODTCNQ, from Japan Carlit) and undoped, were used in surface-stabilized ferroelectric liquid crystal (SSFLC) cells.⁶ Orientation layers were made of ordinary rubbed PI (RN-305, Nissan Chem. Ind.) films and those of the Y type PI-Langmuir Blodgett films of five-stacking layers.

Ordinary triangular and modified waveforms of frequencies from 0.02 Hz to 200 Hz were applied to the samples and the temporal behavior of the switching current was observed simultaneously with the EO response.

3. RESULTS AND DISCUSSIONS

3.1 A Sample with PI-LB Orientation Films and with Undoped-FLC

Figure 1 represents the result of the EO characteristic and the switching current for a triangular waveform applied to a cell using PI-LB orientation films and undoped FLC material.

It is clearly recognized that the starting times of both the EO response and the reverse current coincide exactly with the instant when the voltage waveform changes its polarity. These effects may be interpreted by assuming that no depolarization field exists. However, it is noteworthy to point out that this coincidence is seen in particular for a very low frequency as shown in Figure 2; with the increase of the frequency there occurs a delay time in the starting point and it increases with the frequency. The ordinate shows a relative delay time $\Delta t_{\text{del}}/T$, where T stands for a quarter of the full period of the waveform. Both the delay and the advancement occur within a quarter of the full period. This kind of small amount of frequency-dependent delay may be interpreted as an effect of the viscosity as described in a previous study.¹ For this reason, most of the measurements in this work were done at a very low frequency to avoid this effect. Anyhow, we take the data of Figure 1 as a standard in this work, since other samples showed a deviation from the data of Figure 1, as will be described in the following subsections. Almost the same data as those of Figure 1 were obtained for a sample prepared by performing rubbing on bare ITO glass plates.

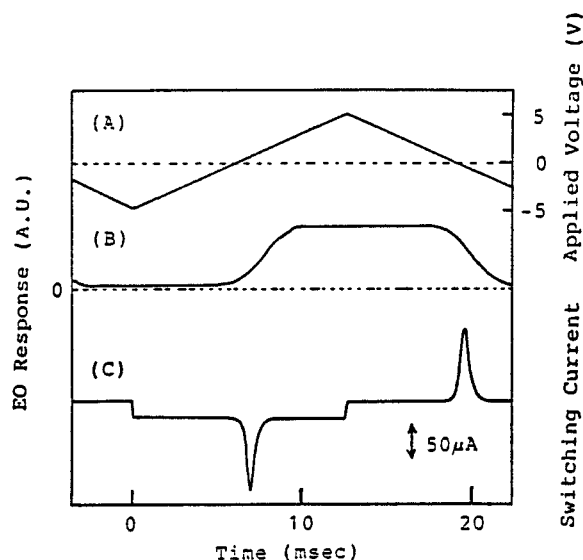


FIGURE 1 The EO response and switching current for a triangular waveform. FLC, ZLI-3654, is aligned with PI-LB films. (A) Applied Voltage, (B) EO response, and (C) current.

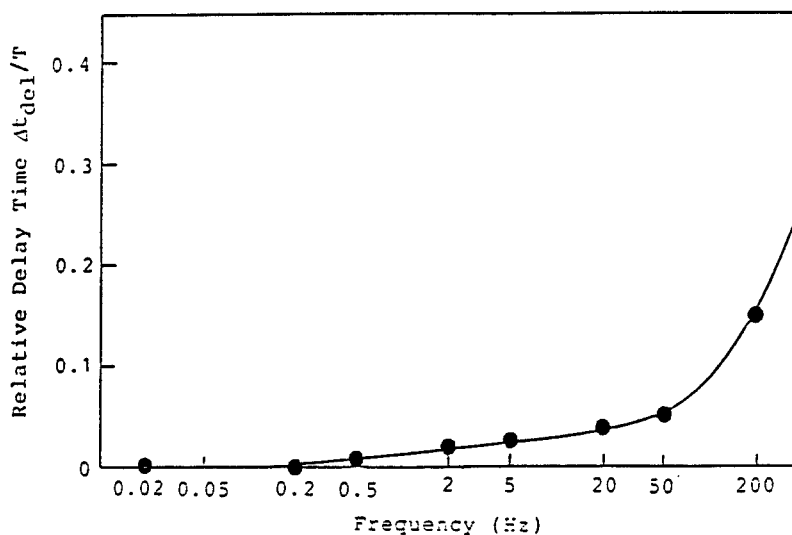


FIGURE 2 Relative delay time versus frequency of the triangular waveforms at 10 V p-p. For the sample with undoped FLC and PI-LB films.

3.2 A Sample with Rubbed PI Films and with Undoped FLC

The temporal behavior of both the EO response and the reverse currents for a sample with rubbed PI films and undoped FLC are shown in Figure 3; two different remarkable features are seen in this figure when compared with Figure 1; one is the advance in the initiation of both the EO response and the reverse current, and the other is a modification in the base line of the current. The latter is thought to

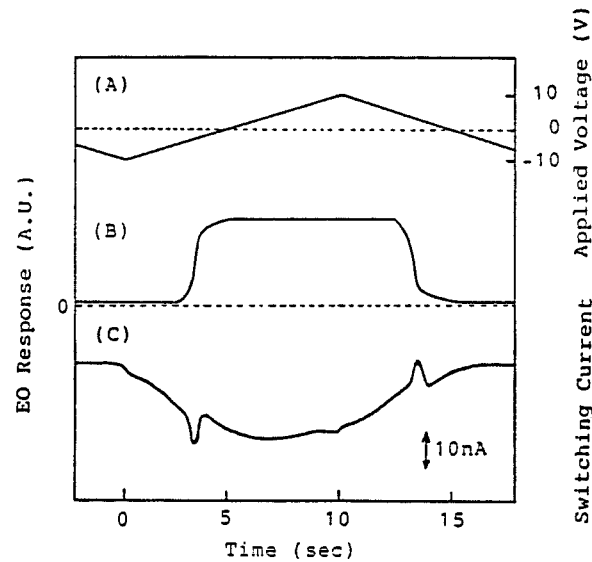


FIGURE 3 The EO response and switching current for a triangular waveform at 0.05 Hz. For a sample with undoped FLC and rubbed PI films. (A) applied waveform, (B) EO response, and (C) current.

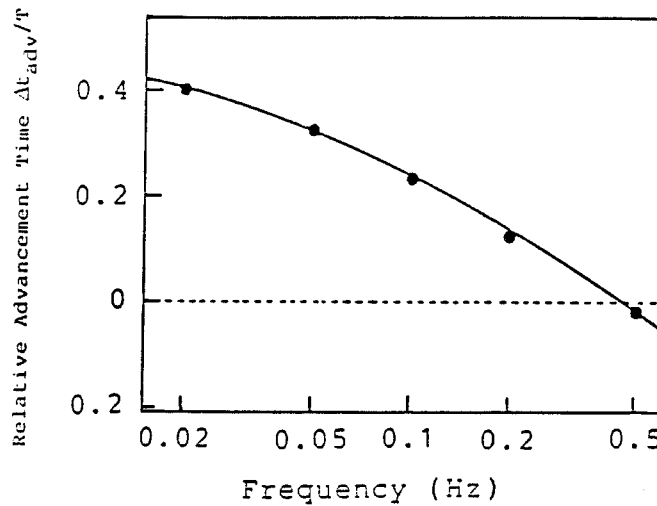


FIGURE 4 The relative advancement versus the frequency at 20 V p-p. For the sample with undoped FLC and rubbed PI films.

originate from the effect of ionic impurities even though we did not intend to dope any impurity. Again, the relative shift in the starting time of the EO response or that of the current plotted against the frequency was examined and the result is shown in Figure 4 as a plot of the ratio $\Delta t_{adv}/T$. Contrary to the data shown in Figure 2, there occurs an advance in these events which shows a frequency dependence. There is a special value of the external applied voltage, $V_{0(rev)}$, which

initiates the reverse current. Even though external voltage $V_{0(\text{rev})}$ is applied, the effective internal field is zero owing to the cancellation by the internal depolarization field. The frequency dependence of the $V_{0(\text{rev})}$ is shown in Figure 5. The value of $V_{0(\text{rev})}$ decreases with the increase of the frequency; at about 0.4 Hz, the $V_{0(\text{rev})}$ becomes zero. These phenomena may be explained by taking account of the effects of the ions, which tend to decrease with the frequency, and the effects of the viscosity.

It is also shown that the relative advance time $\Delta t_{\text{adv}}/T$ depends on the value of the V_0 (expressed in p-p), as shown in Figure 6, in a low-frequency limit. The

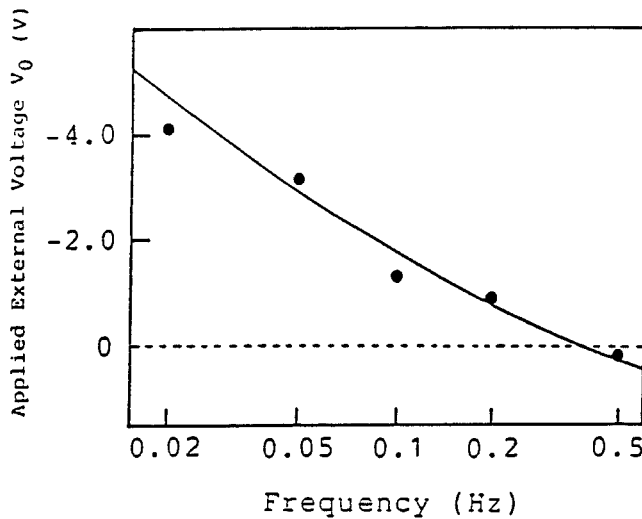


FIGURE 5 A plot of the applied external voltage V_0 at which the reversal current initiates versus the frequency at 20 V p-p. For the sample with undoped FLC and rubbed PI films.

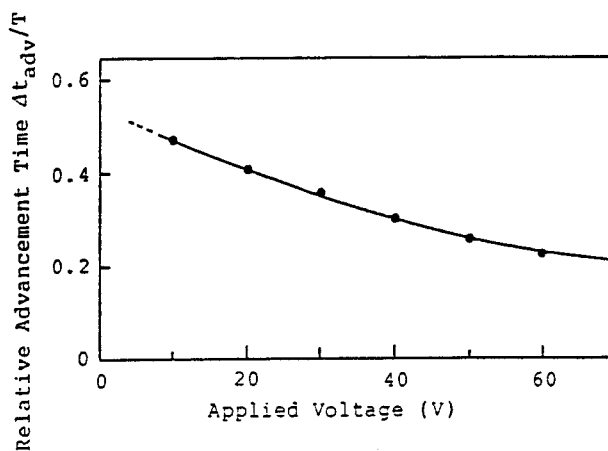


FIGURE 6 The relative advancement time versus the amplitude of the applied triangular waveform at 0.02 Hz. For the sample with undoped FLC and rubbed PI films.

advance time decreases with the external voltage V_0 and shows a saturation in the high voltage region.

The external applied voltage $V_{0(\text{rev})}$, corresponding to the zero internal effective field, shows a sublinear increase with the amplitude of the $V_0(\text{p-p})$ as can be seen in Figure 7. This nonlinearity may be interpreted by taking account of a nonlinear behavior of the ions in the FLC and also that of the induced charge in the orientation layers due to the orientation of the spontaneous polarization P_s .

Figure 8 compares the behaviors in the EO performances of a cell operated with waveforms having different voltage amplitudes.

Figure 9 shows behavior of the current for a modified waveform accompanying the flat portions in both the top and bottom parts in addition to ordinary triangular waveform. The slope in the current which is attributable to the ionic effect tends

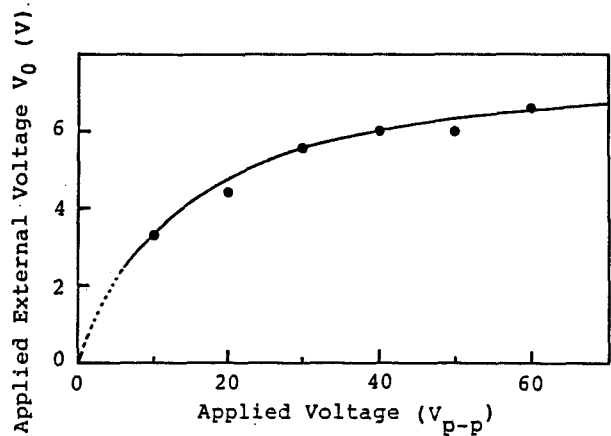


FIGURE 7 The dependence of the applied external voltage V_0 at which the initiation of the reversal current occurs on the amplitude of the applied triangular waveform at 0.02 Hz for the sample with undoped FLC and rubbed PI films.

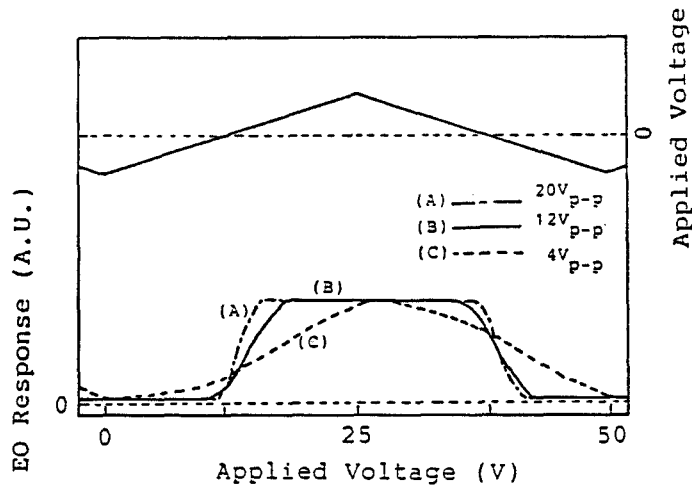


FIGURE 8 The EO responses of the sample with undoped FLC and rubbed PI films for various voltages at 20 Hz: (A) 20 V p-p; (B) 12 V p-p; and (C) 4 V p-p.

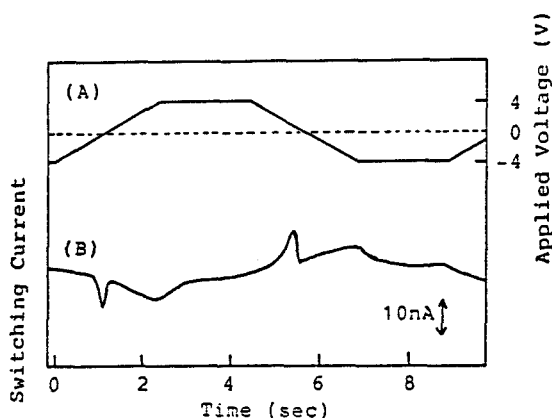


FIGURE 9 The switching current for a modified waveform accompanying flat parts both in its top and bottom.

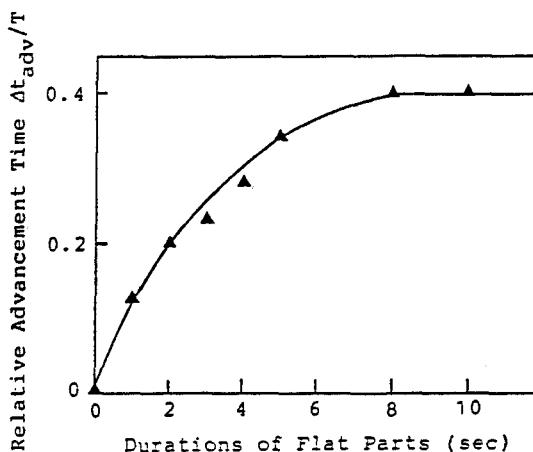


FIGURE 10 The relative advancement time versus the durations of the flat parts in Figure 9.

to decrease after the flat part starts. This may suggest that progress of the continual accumulation of the charges is taking place in the interfacial region, but the ionic current decreases as shown in the figure. By introducing the flat parts in the waveform, the advance due to the accumulated charge is enhanced as shown in Figure 10.

In order to identify which kind of charge gives rise to phase advance in the reversal current as shown in Figures 3, 4, and 6, we examined two other additional FLC materials besides ZLI-3654: CS-1014 (CHISSO) with $PS = 5 \text{ nC/cm}^2$, and Felix-002 (Hoechst AG) with $PS = -42.4 \text{ nC/cm}^2$. The former did not show such a phase shift as the FLC material ZLI-3654 did, and the latter showed rather a fairly large phase delay owing to its high viscosity reaching 1000 mPas. From these facts, the major origin for the generation of the phase advancement is thought to be attributable to the depolarization field originating from the charge induced in the PI orientation films due to the switching of the FLC.

3.3 A Sample Using a CTC Doped FLC but Adopting Ordinary Rubbed PI Orientation Films

The EO performance and the switching current of a sample with the FLC doped with the CTC of 0.04 wt% is shown in Figure 11. There occurs an advance of a quarter cycle (phase shift of 90 degrees) in this particular sample. The effect of the ionic charges is largely enhanced; a tendency found as the FLC material doped with increasing amounts of the CTC, up to 0.04 wt%. The frequency dependence of the relative advance time ratio $\Delta t_{adv}/T$ is shown in Figure 12. In the high frequency region, say above 100 Hz, the effect of the CTC ions tends to disappear; this is thought to be attributable to a finite mobility of the CTC ions. This may be in agreement with the occurrence of the inverted bistability in this system for a pulse voltage whose width exceeds 200 μ s.⁴

The EO response and the reverse current for a modified waveform accompanying flat portions at zero level is shown in Figure 13. After ceasing the external field the effects continue to remain due to the depolarization field.

4. GENERAL DISCUSSIONS AND EVALUATION OF THE DEPOLARIZATION FIELD

According to Chieu and Yang,⁵ the effective field in a FLC medium is given as

$$E_{eff} = \frac{V_0}{d_1 + 2 d_2(\epsilon_1/\epsilon_2)} - \frac{2 d_2/\epsilon_2}{(d_1/\epsilon_1) + (2 d_1/\epsilon_2)} \frac{\sigma_t}{\epsilon_1} \quad (1)$$

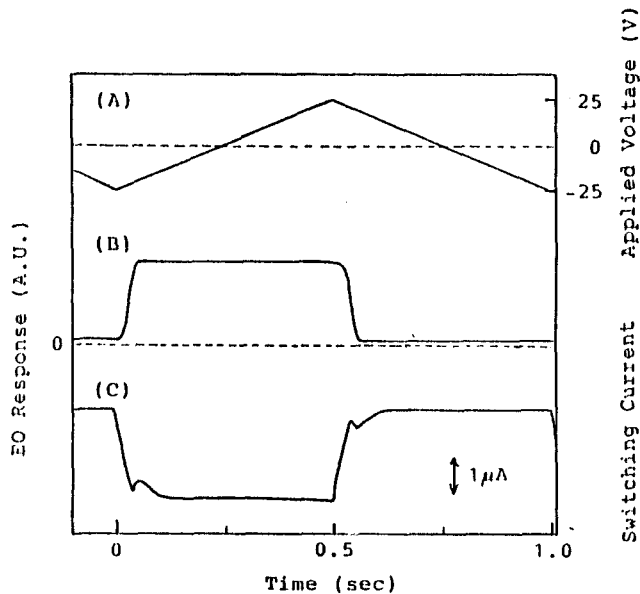


FIGURE 11 The EO response and switching current for a triangular waveform at 1 Hz. For the sample with CTC doped FLC and rubbed PI films.

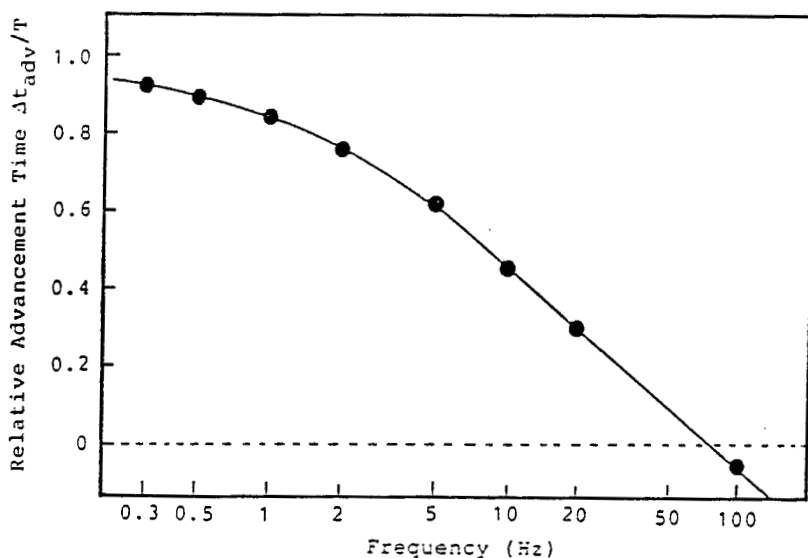


FIGURE 12 The relative advancement versus the frequency at 50 V p-p. For the sample with CTC doped FLC and rubbed PI films.

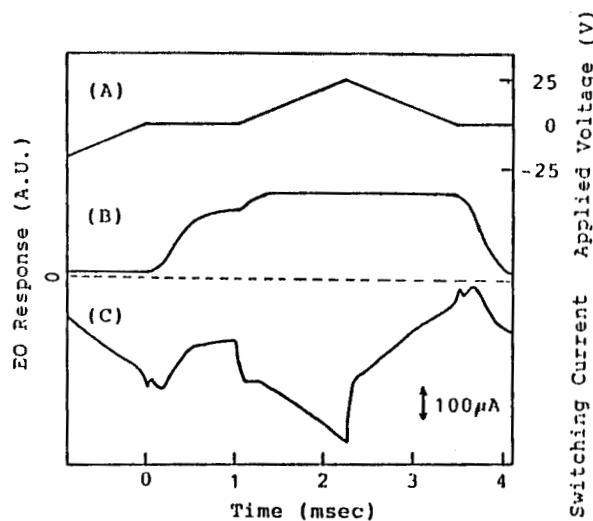


FIGURE 13 The EO response and switching current for a modified triangular waveform at 200 Hz accompanying flat parts at the base line. For the sample with CTC doped FLC and rubbed PI films.

where d_1 , d_2 , ϵ_1 , ϵ_2 stand for the thicknesses and the dielectric constants of the LC medium and the orientation layers, respectively. The mobility of CTC dissolved into an FLC host was evaluated as $\mu = 5.0 \times 10^{-7} \text{ cm}^2/\text{V} \cdot \text{sec}$ by B. Y. Zhang *et al.*⁷ The total charge σ_t comprises the following three parts: $\sigma_t = \sigma_i + \sigma_c + \sigma_s$, corresponding to the ionic charges accumulating on the orientation films originated from the impurity ions, those of CTC, and the induced charge in the orientation films by spontaneous polarization, where σ_s is equal to P_s .

Both the switching currents and the EO responses start at the instant when the internal effective field given by Equation (1) changes its polarity, since no threshold field exists for a low frequency field.

The value of V_0 for giving $E_{\text{eff}} = 0$ under the exist of the surface charges σ_i is

$$V_0 = \frac{2 d_2}{\epsilon_2} \sigma_i \quad (2)$$

In a sample with LB films, the observed V_0 is zero probably due to σ_i which will be discharged in terms of the outer circuit owing to the finite conductivity of the ultrathin films. It is also interesting to point out that V_0 vanishes when d_2 is infinitesimally small ($d_2 \ll d_1$) as exemplified by LB films of 5 molecular layers or less, even though σ_i exists.

For a sample with the nonconductive (rubbed PI) films there occurs the accumulation of the charges σ_i , but the degree of the accumulation depends on the field duration. This effect is thought to be the origin of the frequency and voltage dependence shown in Figures 4 and 5. The occurrence of the advance in the phase for the EO and the current characteristics seems to be contradictory to causality, but these phenomena are also explained by taking account of the accumulation of the surface charges that generate the depolarization field.

The maximum value of the depolarization field of the undoped FLC is estimated to be 4 volts, and that for the CTC doped FLC is 20 volts for $V_0 = 25$ volts. By substituting $d_2 = 50$ nm and $\epsilon_2 = \epsilon_0 \times 5.5$, one obtains σ_i is about 30 nC/cm² for the former case, which is in agreement with the value of Ps of the FLC material (ZLI-3654).

It is considered that the depolarization field originating from the surface charges takes a part in degrading the memory capability in an SSFLCD, as already discussed by Ikeno *et al.*⁸

5. CONCLUSIONS

The reverse current and the EO response for a triangular voltage waveform are shown to start at the instant when the polarity of the waveform changes its polarity, in particular for a sample with PI-LB films. A sample with ordinary rubbed polyimide orientation films, on the other hand, showed an advance in the phase. Furthermore, for a CTC doped FLC with rubbed PI films, the advance of at most 90 degrees was observed. These effects may be explained by assuming the existence of the depolarization field originating from the accumulation of charges in the interfacial regions. For a sample with pure FLC material and nonconductive PI orientation films, the depolarization is thought to originate from the induced charge in the orientation films due to the switching of the FLC with a fairly large value of spontaneous polarization. An analysis of the observed result shows the validity of this model.

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