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Anomalous charging and discharging processes in liquid crystals

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Transient charging and discharging currents produced by a DC voltage applied to a parallel plate capacitor filled with a liquid crystal sample have been studied experimentally. A discontinuous jump of the current magnitude in the nematic phase has been observed for the first time. The shape of the charging and discharging current curves has been shown to depend on the run numbers. Experimental results have been explained with a simple model based on molecular alignment.

1. Introduction

Recently, much work [1, 2] has been undertaken on transient charging and discharging currents induced by a step-voltage application to nematic liquid crystal films. A peak in the transient charging and discharging currents has been observed above and below the characteristic applied voltages V_c. The characteristic applied voltage V_c can be defined as the voltage for which the time of the peak position of the charging current against the applied voltage is a maximum. The occurrence of peaks in transient current as a function of time has been explained in terms of space charge-limited current, the carrier mobility distribution due to nematic director orientation and ionic transport effects. There are two cases: for applied voltages above V_c the time of occurrence of the peak current decreases with increasing applied voltage, and for applied voltages below V_c the time occurrence of the peak current decreases with decreasing applied voltage. The peak in transient current has also been observed in electrooptic experiments [3–5]. However, the rapid increase of the transient current magnitude after a certain time in the nematic phase has not been observed before. In this paper, unusual charging and discharging currents in liquid crystals (8CB) have been investigated as a function of the applied voltage, temperature and time.

2. Experimental

The alkylcyanobiphenyl liquid crystal used in the present experiment was 8CB (BDH Chemicals Ltd. K24) with positive dielectric anisotropy $\Delta\varepsilon = 10$. The three terminal parallel plate capacitor was filled with the liquid crystal (LC) sample in the isotropic phase. The area of the cell was $1{\cdot}13\,\mathrm{cm}^2$ and the thickness $100\,\mu$m. After temperature equilibrium (a few hours), the dielectric constant of the LC filled sample cell was measured with a LCR Meter (HP model 4284 A) at 1 MHz. The measurement at 25°C was $\varepsilon_{\mathrm{meas}} = 13{\cdot}3$ which compares very favourably with $\varepsilon_{\parallel} = 13{\cdot}5$ at 25°C for 8CB [6]. This would seem to prove that the sample was oriented homeotropically (almost parallel to the applied electric field). We used a low frequency set-up developed in our

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laboratory [7] to measure the charging and discharging currents. The measuring system consists of a Keithley 617 Electrometer, personal computer and expansion boards. In contrast to the former experiments which were used with a step-like voltage pulse, the voltage in our measurement has been applied to the sample over the whole measuring period. This is an essential difference between the present and former experiments [1, 2].

3. Results

The charging currents of the LC cell in the nematic phase are shown in figure 1 for three successive runs. As we can see, the shape of the current curves depends on the run number: the current magnitude increases with the run number. But the most striking feature we observe in figure 1 is the sudden jump of the current magnitude after a certain time. This phenomenon has never been observed in liquid crystals. The increasing rate of the current depends on the run number and that for the third run is of the order of 100; increasing from ~2 μA to ~200 μA. The corresponding discharging currents are shown in figure 2. A prominent peak in the current for a first run can be seen around 40s. The current shapes for the second and third runs are similar but a difference between the first run and the second and third runs is clearly visible. It should be pointed out that such a difference has never been observed in liquid crystals. It should also be noted that in the second and third runs two peaks are observed.

The time dependence of the discharging currents for different choices of the applied voltages in the nematic phase is shown in figure 3. In our measurements, the peak of the transient current in the nematic phase cannot be observed for the applied voltages less than $V_{\min} = 1{\cdot}5$ V. These current shapes have been observed in the previous experiments [1, 2] but the time occurrence of the peak is much longer (~40 s) than in the case of LC film measurements (~40 ms). Based on our data and those presented by Sugimura and his colleagues [2] we have roughly estimated that the peak time scaling factor is proportional to the square of the cell thickness, d^2. In both, nematic and smectic phases, the peak current has been observed for applied voltages below V_c as the time occurrence

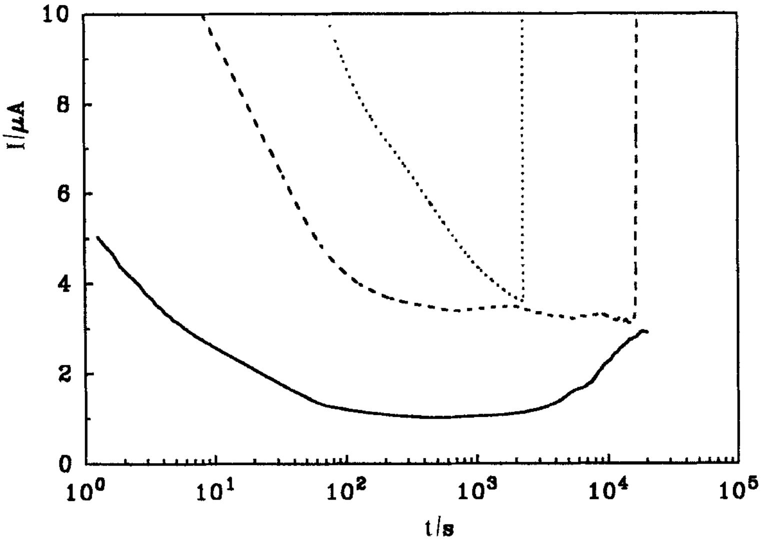

Figure 1. Time dependence of the charging current in the nematic phase (——, first run; - - -, second run; · · · ·, third run).

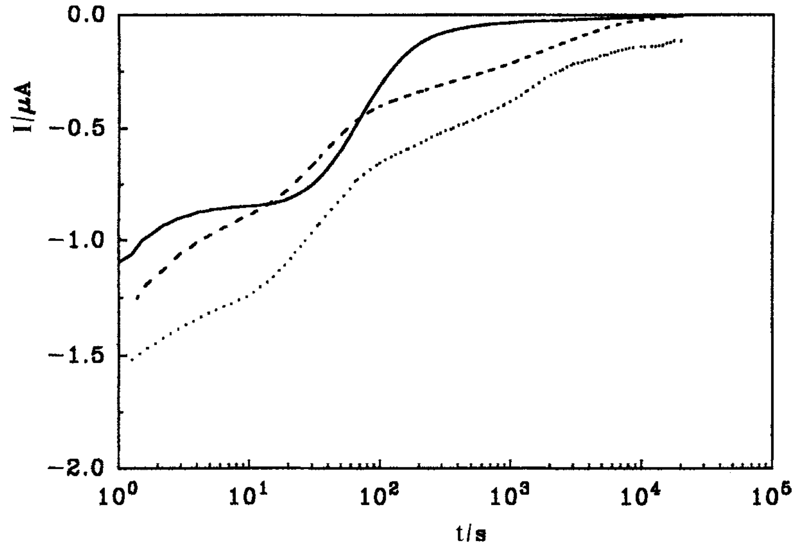

Figure 2. Time dependence of the discharging current in the nematic phase (——, first run; - - -, second run; ····, third run).

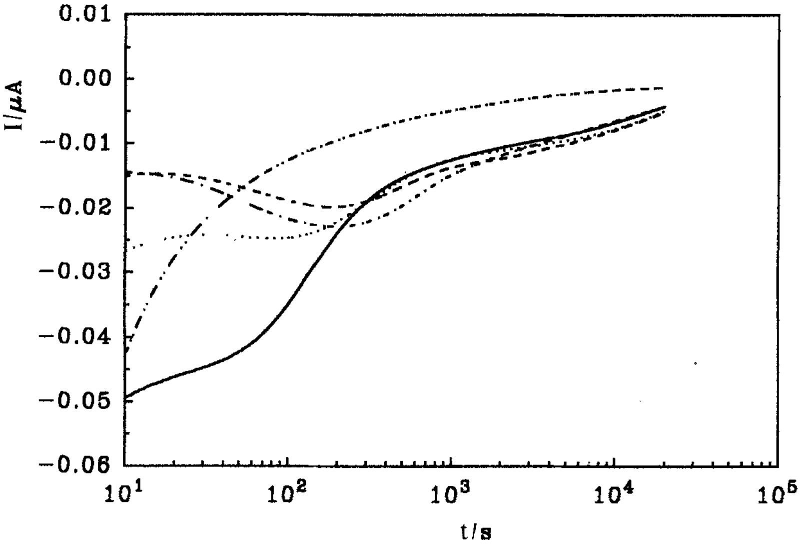

Figure 3. Time dependence of the discharging currents for various applied voltages in the nematic phase (—··—, 1 V; ——, 5 V; ····, 10 V; - - -, 40 V; —·—, 100 V).

of the peak current decreases with decreasing applied voltage. We have measured the dielectric constant of the LC filled sample cell after the first, second and third runs. They are 13·3, 13·09 and 13·1, respectively. Additionally, we have measured the dielectric constant a few hours after we stopped the experiment. The value of the dielectric constant was similar to the last value measured. This indicated that the final state was stable at least for a few hours.

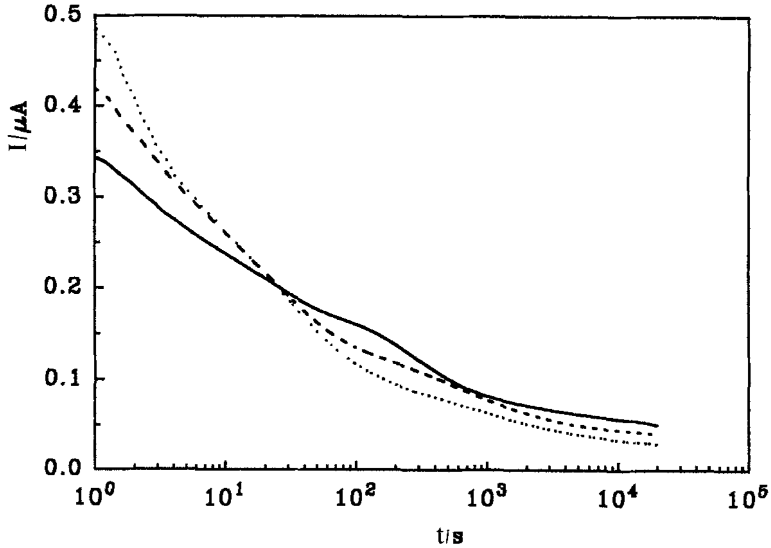

Figure 4. Time dependence of the charging current in the smectic A phase (——, first run; - - -, second run; ····, third run).

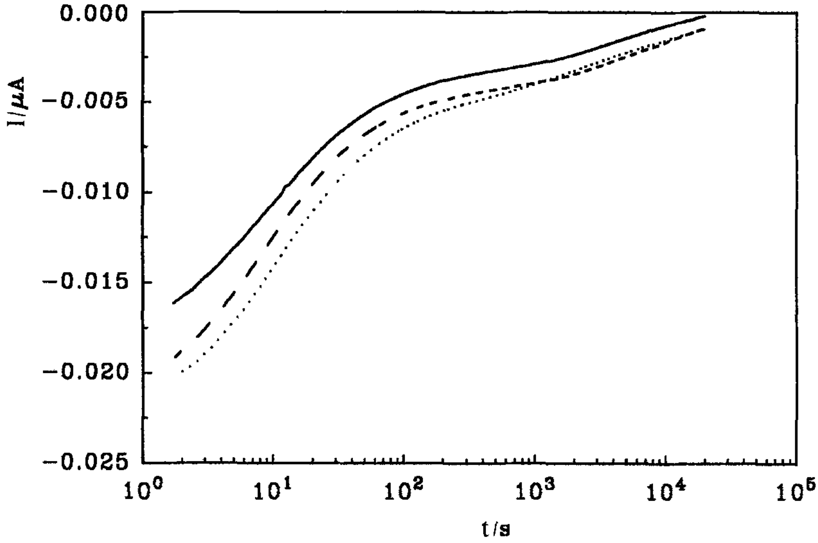

Figure 5. Time dependence of the discharging current in the smectic A phase (——, first run; - - -, second run; ····, third run).

The charging and discharging measurements have also been carried out on the smectic A phase. The results are shown in figures 4 and 5. Charging current shapes (see figure 4) are different in the first, second and third runs. We recognize the same phenomenon in the case of the discharging current (see figure 5). Contrary to the nematic phase measurements, the rapid increase of current is not observed in the smectic phase.

4. Discussions

One simple model for the mechanism of the occurrence of the peak in the transient current induced by the polarity reversal on an applied DC voltage is proposed by Sugimura and his colleagues [2]. According to the model of Sugimura, below the characteristic voltage, V_c, the time occurrence of the peak current is dominated by the competition of the internal and external electric fields. As we can see (see figure 3) in our experiment, the time occurrence of the peak current decreases with decreasing applied voltage. On the other hand, an external electric field E applied to a uniformly oriented nematic liquid crystal can distort the mean direction of the molecules (the director **n**) and thus alter, for example, the optical properties of the nematic liquid crystal [8]. These polar surface instabilities were recently observed in homeotropic layers of the nematic liquid crystal with positive dielectric anisotropies under the action of vertical DC electric field [9]. This effect is manifested by the appearance of rounded domains due to static deformation of the director [9–11].

We have assumed that the applied voltage V produces across the cell a uniform electric field E which can be written as

$$E=\frac{V}{d}=\frac{Q+P}{Cd}, \tag{1}$$

where d is the cell thickness, Q the density of the free charge on the electrodes, C the capacitance per unit area of the filled cell (capacitor) and P the density of the polarization charge. Generally, the density of the polarization charge P is a function of the electric field strength and time,

$$P\propto\exp\left(\frac{t}{A\sqrt{[(\tau_c-t)^2+(\tau_c+t)^2]}}-\frac{t}{\tau_0}\right), \tag{2}$$

where t represents time in seconds and τ_0 is the relaxation time of the system when the applied voltage is less then V_{min}, τ_c is the time constant proportional to the time of the peak position and A is the constant proportional to the characteristic voltage V_c. In figure 1, we can see that after the first run, the sudden jump in the current is not observed. The flow of current was not smooth because the orientation of the sample over one run period ($\sim$20 000 s) was not complete. We can already notice the sudden jump in the current starting from the second run. We believe that the time period necessary for the external field to align the molecules was too short for the first run but enough for the subsequent runs. Because of this new molecular order, the transport of charge is facilitated in the bulk of the sample and the increase of the current magnitude is observed.

This explanation can be supported by the data presented by Lavrentovich and his colleagues [12]. Applying the vertical DC electric field ($U=3$ V) to a homeotropically oriented 60 μm thick nematic (5CB) layer, they have observed polar ordering induced by surface instability. It seems our observation of the rapid increase of the current due to the change of the mean direction of the molecules is caused by the polar ordering. This conclusion is supported by the fact that the dielectric constant values before and after the first run were different. For phenomenological interpretation of the present data we want to point out that our value of the applied electric field $E=5\text{ V }100\,\mu\text{m}^{-1}$ was very similar to that of Lavrentovich *et al.* [12].

However, the increase in current has not been observed in the smectic phase. There are two possible ways to explain this fact. Probably, the applied electric field is unable to produce new order in the case of the smectic phase in the same time range as in the case of the nematic phase. Since our charging time period is only 20 000 s, the current increase would be out of our measuring time range. Another possibility is that the smectic layers are able to restrict smooth current flow. From the molecular ordering point of view, the surface between neighbouring smectic layers is something like an incommensurate surface. Therefore, it seems natural to suppose that the increase of the current in the nematic phase is much larger than that in the smectic phase even though the latter has a more aligned local structure.

One possible interpretation of the occurrence of the second peak observed in our experiment (see figure 2) might be based on the molecular process. Using the Fourier transform of our data we have found out that the origin of this peak is related to the molecular reorientation. As has been mentioned in the literature, in the case of cyanobiphenyls we can observe the appearance of rounded domains of molecules [9–11]. It is possible that we observe some kind of reorientation of these domains.

If the current increases in the nematic phase is due to a new molecular structure, it might be worthwhile to point out that this phenomenon appears in our experiment without any additional aligning agent as for example a magnetic field. It may be quite possible to make a device to control current flow by means of the molecular ordering. Although, we have discussed the molecular ordering produced by the electric field, it is also possible (as mentioned earlier) to order the molecular structure by using a strong magnetic field. Work is in progress in our laboratory to this end.

5. Conclusion

We have studied the unusual behaviour of the charging and discharging currents induced by a DC voltage applied to a 8CB LC filled sample cell. The molecular ordering influences the charging and discharging processes in the nematic phase. The unusual behaviour of the charging current has been qualitatively explained by a simple model based on a time dependent ordering effect. Knowledge of the mechanism of such a phenomenon could be important for proper understanding of the current flow through LCs as well as for practical applications, which, for example, may be an active matrix-driven LC display.

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