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# Anomalous Transient Currents in Ordered Nematic Liquid Crystals

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# ANOMALOUS TRANSIENT CURRENTS IN ORDERED NEMATIC LIQUID CRYSTALS

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Abstract Transient charging currents produced by DC voltage applied to a parallel plate capacitor filled with liquid crystal sample (7CB and 8CB) 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 current curves has been shown to depend on the applied of the external magnetic field. Experimental results have been explained with a simple model based on molecular alignment and quasi-one-dimensional conductivity.

*Keywords: transient currents, alkylocyanobiphenyl, sub-hertz set-up.*

## INTRODUCTION

Recently, much work has been undertaken to interpret transient currents flowing in the nematic liquid crystals (NLCs).[1-3] This is mainly due to the fact that the charge carrier transport processes in NLCs influence the device performances of NLC displays. Transient charging currents induced by a step-voltage application to NLC films have been studied extensively. A peak in the transient charging currents is usually observed and the occurrence of the peak 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 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 maximum. The peak in transient current has also been observed in electro-optic experiment.[4-6] However, the rapid increase of the transient current magnitude after certain time in the nematic phase has not been observed before as we described in our earlier publication.[7]

## EXPERIMENTAL

The alkylcyanobiphenyl liquid crystals used in the present experiment were 7CB and 8CB (BDH Chemical Ltd. K21 and K24) with positive dielectric anisotropy $\Delta\varepsilon \approx 10$ The three terminal parallel plate capacitor was filled with the liquid crystal (LC) sample in the isotropic phase. The area of the cell was 1.54 $cm^2$ and the thickness 60 μm. In an ideal measurement cell with parallel plates of radius $r$ separated by a distance $d$, such that $r>>d$, the electric field vector will always be perpendicular to the plane of the plates. So, in our case $r$=7mm$>>d$=60μm. After temperature equilibrium (a few hours), the dielectric constant of the LC filled sample cell was measured with LCR Meter (HP model 4284A) at 1 MHz. Transient currents at a constant temperature were measured with a low frequency set up developed in our laboratory.[8] The measuring system consists of Keithley 617 Electrometer, personal

computer and expansion boards. Each measuring run was of 24 hours duration consisting of 11 hours (~20000 s) of measuring time and 13 hours of pause. In contrast to the former experiments which were used with step-like voltage pulse, the voltage in our earlier[7] and present measurements has been applied to the sample over the whole measuring period. This is the essential difference between our experiments and earlier investigations.[1-3] We used Neodymium Iron Boron (NdFeB) magnets supplied by MAGNET Co. (California, U.S.A.). Two rectangular magnets 5cm×5cm×2.5cm each, gave magnetic field 0.7T across the sample which was enough to align sample. To confirm so, we measured dielectric constant $\varepsilon_{\perp}$=5.5 and $\varepsilon_{\parallel}$=16.2 at 36°C for 7CB and $\varepsilon_{\perp}$=5.4 and $\varepsilon_{\parallel}$=13.3 at 38°C for 8CB. The measurements compared favorably well with data presented by Bose et al.[9]

## RESULTS AND DISCUSSION

The representative charging currents of NLC for various applied voltages are shown in figure 1 for 8CB cell with homeotropic alignment. A peak in the current transient can be seen for voltage 5V. We note that the results for 7CB are the same within experimental resolutions of our set-up and the peak cannot be observed in the isotropic phase. In figure 2 one can see a prominent peak on the charging curve and pre-jump effect. These experimental results in conjunction with literature, allow us to infer that the current transients are due to the director orientation with the applied electric field. For a material with positive dielectric anisotropy $\Delta\varepsilon=\varepsilon_{\parallel}-\varepsilon_{\perp}>0$ the application of an electric field causes the LC to align in the direction of the filed. Because of the spontaneous fluctuations

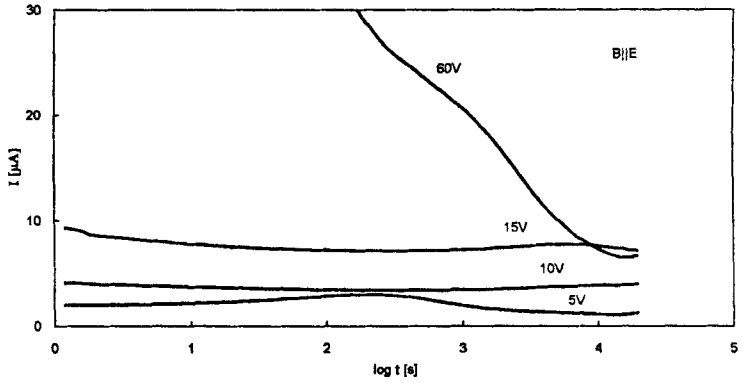


FIGURE 1 Time dependence of the charging currents for various applied voltages for the case of the parallel orientation **B||E**.

of the alignment in the nematic medium and the electric field there is a competition which can be described by the one-dimensional torque balance equation in the form[10]

$$k\frac{\partial^2\theta\langle z,t\rangle}{\partial z^2}-\frac{1}{2}\cdot\varepsilon_0\cdot\Delta\varepsilon\cdot E^2\langle z,t\rangle\cdot\sin\langle 2\theta\langle z,t\rangle\rangle-\gamma\frac{\partial\theta\langle z,t\rangle}{\partial t}=0 \qquad (1)$$

where $k$ is the average elastic constant, $\varepsilon_0$ is the permittivity of free space, $E\langle z,t\rangle$ is the electric field at position along the $z$ axis which is a normal direction to the plate surface, $\gamma$ is the rotational viscosity, and $\theta\langle z,t\rangle$ is the director tilt being measured from the $z$ axis. As one can see the competition is space and time dependent. So, it seems clear that system needs time to develop new molecular structure. This new structure finally allows the rapid increase of the transient current magnitude as it is shown in figure 3. After second run the sudden jump of the current magnitude is visible. The increasing rate is of he order of 1000; increasing from ~2μA to ~3mA.

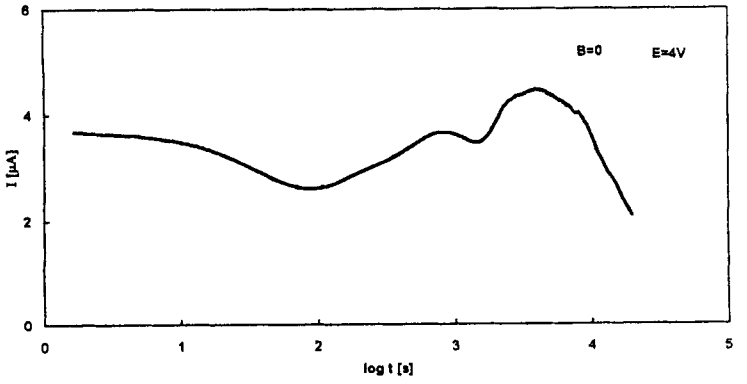


FIGURE 2 Time dependence of the charging current for voltage 4V for the case $\underline{\mathbf{B}}$=0.

The same phenomenon is shown in figure 4. The important difference one can notice is the shorter period ($1.3 \cdot 10^3$s) before jump in the case of measurement with the magnetic field $\underline{\mathbf{B}} \| \underline{\mathbf{E}}$ than without the magnetic field ($4.3 \cdot 10^3$s). In an earlier paper[7] we have proposed expression which described the occurrence of the peak. We have assumed that the applied voltage V produces across the cell a uniform electric field E which can be written as

$$E = \frac{V}{d} = \frac{Q+P}{C \cdot d} \qquad (2)$$

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,

$$P \propto \exp\left\langle \frac{t}{A\sqrt{\left\langle \left\langle \tau_c - t \right\rangle^2 + \left\langle \tau_c + t \right\rangle^2 \right\rangle}} - \frac{t}{\tau_0} \right\rangle \qquad (3)$$

where t represents time in seconds and $\tau_0$ is the relaxation time of the system when the applied voltage is less then $V_{min}$, $\tau_c$ is the time constant proportional to the time of the peak position and A is the constant proportional to the characteristic voltage $V_C$.

To confirm that electric field acting on NLC can distort the mean direction of the molecules and thus alter, for example, the electrical and the optical[11] properties, we measured charging and discharging currents for three successive runs. Results shown in figure 5 were taken with the magnetic field perpendicular to the electric one ($\underline{\mathbf{B}} \perp \underline{\mathbf{E}}$). As we can see the first charging curve goes below zero. It means that the electric field reorients molecules which have been aligned by magnetic field $\underline{\mathbf{B}} \perp \underline{\mathbf{E}}$ in the direction of itself. New orientation of the director $\underline{\mathbf{n}}$ parallel to the electric field minimizes the elastic and the dielectric energy. So, the NLC gives energy to the measuring system. The same tendency is visible for discharging current (Fig.5b). Additionally, from figure 5 one can draw conclusion that the competition between the electric field and the magnetic one in the presented case is the long term behavior. Just after ~40000s (one charging run and one discharging run) charging curve shows normal charging shape. We measured charging currents for homogeneous alignment (director parallel to the plate surface).

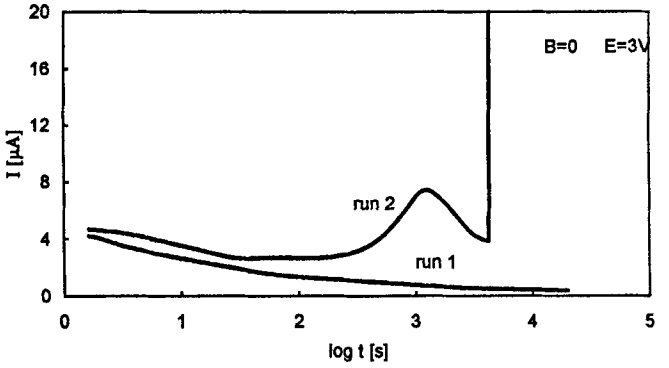


FIGURE 3 Time dependence of the charging currents for two successive runs for voltage 3V for the case **B**=0.

Aligned samples were prepared by slowly cooling from the isotropic phase into the nematic phase in the presence of a magnetic field. During measuring period magnetic filed was applied to the sample. The results are shown in figure 6. No current jump is observed. However, prominent peaks related to the reorientation of director are visible. Based on the data presented in figures 3, 4 and 6 we attempt to propose one possible interpretation of the occurrence of the sudden jump of the transient current magnitude. Because the voltage in our experiment has been applied to the sample over the whole measuring period, current increase may be explained by conductivity increase. For direction perpendicular to the plate surface homogeneous alignment NLC may be considered as stocked molecular quasi crystal which tends to be an insulator. This is because the overlap of completely full or empty $\pi$-orbitals is weak and this leads to a band structure with a characteristically large band gap[12]. So, in this

case we did not observe the sudden jump of the transient current magnitude.

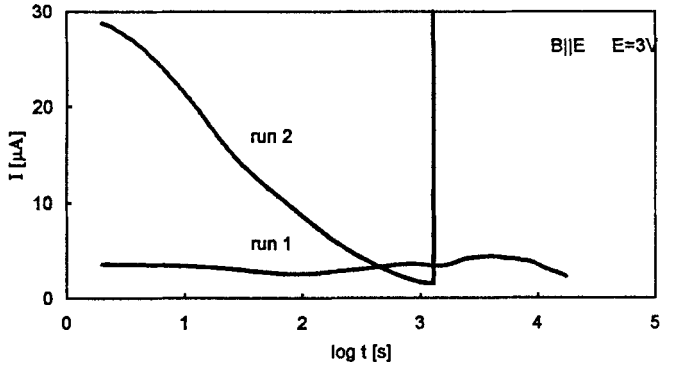


FIGURE 4 Time dependence of the charging currents for two successive runs for voltage 3V for the case of the parallel orientation **B||E**.

The principal structural requirement for quasi-one-dimensional conductivity is that the system possesses linear molecular chains along which charge transport can occur.[12] Homeotropically oriented sample fulfills this condition. Boden et al.[12] have argued that the conductivity of such a system as LC is generally associated with charge transport occurring via a hopping or tunneling mechanism. However, for aligned samples conductivity is independent of frequency and takes place by hopping between localized sites - potential wells. They have analyzed the measurements using the Scher and Lax model.[13] The frequency dependent diffusion coefficient $D(\omega)$ is given by

$$D\langle\omega\rangle = \frac{\Delta^2_{rms}}{2d} \cdot \frac{i\omega\Psi\langle i\omega\rangle}{1-\Psi\langle i\omega\rangle} \qquad (4)$$

where $\Delta^2_{rms}$ is the mean-square displacement of the diffusing charge carriers which is proportional to time, d is the dimensionality of the system, and $\Psi\langle i\omega\rangle$ is the Laplace transform of $\psi(t)$, the distribution function describing the probability for a hop to occur at time $t$ after the preceding one. $\psi(t)$ determines the frequency dependence of the conductivity.

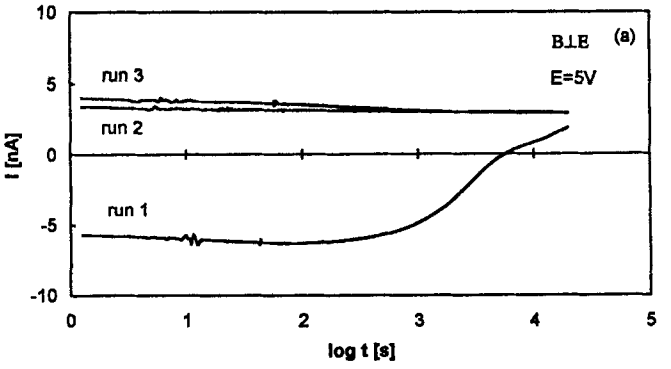


FIGURE 5a Time dependence of the charging currents for three successive runs for voltage 5V for the case of the perpendicular orientation **B⊥E**.

After a few calculation steps they arrived at the following relationship for the long term (low frequency) limit of the frequency dependent conductivity,

$$\sigma\langle\omega\rangle = \frac{ne^2}{kT}\cdot\frac{\Delta^2_{rms}}{2d}\cdot\lambda = \text{const} \qquad (5)$$

where n is the concentration of charge carriers and the electronic charge. That is, at long term, the system behaves as though it were an ordered system with just a single effective transfer rate between sites.

In practice, LCs always contain impurity ions. These ions can originate from dissociated impurities, and

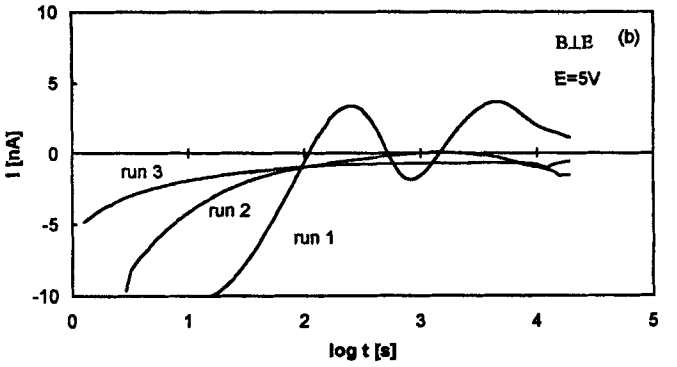


FIGURE 5b Time dependence of the discharging currents for three successive runs for voltage 5V for the case of the perpendicular orientation **B⊥E**.

also from the spontaneous dissociation of the LC molecules themselves. On the other hand Lavrentovich et al.[14] have observed polar ordering in oriented 60μm thick nematic (5CB) system. Taking into account above and the additional information that for cyanobiphenyls[7,15-17] one can observe strong dipole-dipole interactions leading to the formation of dimers, we would like to propose quasi-one-dimensional conductivity mechanism as responsible for jump of the current magnitude. The dimers spreading along the plate normal can be treated as localized sites. Hopping takes place among these localized sites. To develop this structure NLC system needs time which agrees with our observations.

## CONCLUSION

We have studied unusual behavior of the charging currents induced by the DC voltage applied to the NLC

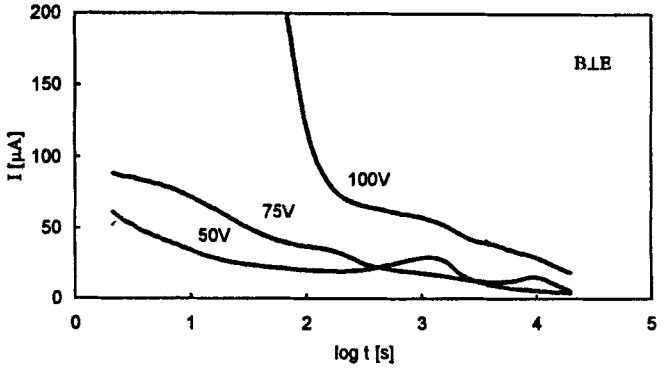


FIGURE 6 Time dependence of the charging currents for various applied voltages for the case of the perpendicular orientation **B⊥E**

cell. The molecular ordering influences the charging processes. The unusual behavior of the charging current has been qualitatively explained by a simple model based on time dependent ordering effect. Knowledge of the mechanism of such a phenomenon could be important for proper understanding of the current flow through LC as well as for practical application, which, for example, may be an active matrix-driven LC display.

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