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Transient currents in nematic liquid crystals

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Transient currents induced by the polarity reversal of a dc voltage applied to a liquid-crystal cell have been experimentally studied. For voltages less than a characteristic value, the time occurrence of the peak current decreases with decreasing applied voltage, which is contrary to behavior expected for a normal carrier-transport process. To explain the experiments, a simple model based on the time variation of an applied voltage and the internal electric field induced by the adsorbed electric charges on the cell substrate has been proposed. The results from the model agree reasonably well with the experimental observations.

I. INTRODUCTION

Studies of transient currents induced by the switching or polarity reversal of an applied dc voltage in nematic liquid crystals (NLC's) are of both practical and fundamental interest. At first a normal charging current appears within about 1 msec, which is limited by the response time of the sample and measuring circuit, and then a peak current is observed. In recent years, there have been extensive studies[1-7] on such transient currents. The occurence of peaks in the transient current as a function of time has been explained as originating in the space-charge-limited current,[8,9] the carrier mobility distribution dependent on nematic director orientation[5,6] and the electric double-layer model.[1,7] Experimental results on the transient currents accompanying the application of a dc voltage above a characteristic voltage V_c, whose physical meaning is discussed in the following section, slightly larger than the threshold voltage for the Freedericksz transition have been given by the authors in previous publications.[5,6] In these studies, the mechanism of the peak occurrence is also discussed theoretically based on the spatial distribution of the carrier mobility in NLC's dependent on the director orientation and the presence of the adsorbed charge layers on the substrate. In these studies, for applied voltages above V_c, the time of occurrence of the peak current decreases with increasing applied voltage, as might be expected from the higher drift velocities at higher fields. The transient currents accompanying the application of a dc voltage below V_c has not been extensively studied experimentally and theoretically. For applied voltages below V_c, an unusual phenomena appears as the time occurrence of the peak current decreases with decreasing applied voltage. In this paper, this unusual phenomenon of the transient currents in NLC's is described based on the experimental results and a simple theoretical model.

II. EXPERIMENT

The nematic liquid-crystal material was introduced between two pieces of glass with transparent In_2O_3 electrodes on their surfaces. The area of these cells was typically 2 cm^2. The thickness of the cell was 6.8 μm. The material which we have investigated is 4-cyano-4′-5-alkyl-bipheneyl (5CB: BDH Chemical Ltd. K15), which has a positive dielectric anisotropy $\Delta\epsilon = 10$. Provisions were made for maintaining the cell at a constant temperature. Transient currents were observed through a series load resistor (1 kΩ) by means of a digital storage oscilloscope (Iwatsu DS-8621). The oscilloscope downloads the measured data to a microcomputer. The pulse voltage (0.1 Hz pulse frequency, $\sim\pm0.5-10$ V voltage) was applied to the cell. The measurements were repeatedly carried out for three different samples prepared under the same conditions.

Typical transient current shapes induced by the polarity reversal of a dc voltage are shown in Fig. 1 as a function of time for various applied voltages less than V_c and also for 3.0 V, which is greater than V_c. At first a normal charging current appears within about 100 μsec, and then a peak of transient current is observed. The quantitative identification of the peak in the transient current is a rather subtle business from the observation of the oscilloscope, but the peak position can be determined by digital processing of the current shape information. The I-V characteristics of the peak current caused by the polarity reversal of a dc voltage are shown in Fig. 2 for various temperatures. As can be seen in this figure, the polarity

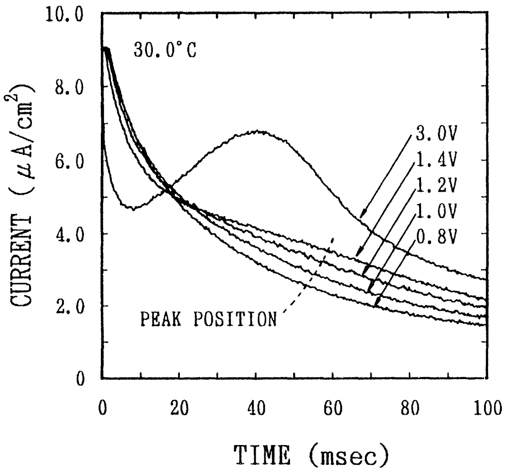

FIG. 1. Transient current shapes induced by the polarity reversal of a dc voltage applied to a NLC cell for voltages less than a characteristic value and a voltage of 3 V at a constant temperature of 30 °C.

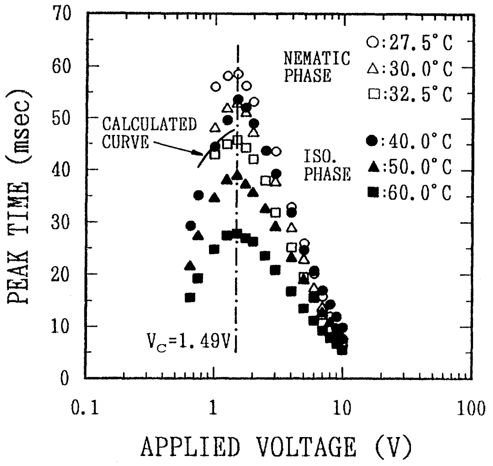

FIG. 3. Time occurrence of the peak in the current as a function of the applied voltage for various temperatures in the nematic and the isotropic phase. The solid line shows the numerical calculation result.

reversal current is nearly proportional to the square of the voltage at high voltage and to the first power of the voltage at low voltage. These results are well known[6,10] in the field of the transient current in NLC's and are beyond the scope of this paper. Figure 3 shows the time occurrence of the peak current as a function of applied voltage for various temperatures in the nematic and the isotropic phases. In our measurement system, the peak of transient current in the nematic phase cannot be observed for applied voltages less than 0.75 V. In both nematic and isotropic phases, expected results are obtained for the applied voltage above V_c as the time occurrence of the peak current increases with decreasing applied voltage. For applied voltages less than V_c, however, the time occurrence of the peak current decreases with decreasing applied voltage. This behavior, which occurs for the applied voltages insufficient to cause director rotation, is contrary to behavior expected for the normal carrier transport process in a typical NLC.

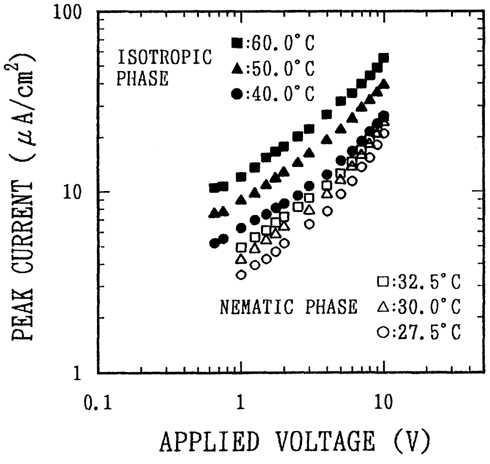

FIG. 2. *I-V* characteristics of the peak in the transient current caused by the polarity reversal of a dc voltage applied to a NLC cell for various temperatures.

III. RESULTS AND DISCUSSION

If the occurrence of a peak in the transient current is due to charge drift of ionic and/or electron carriers in the NLC layer dependent on the external applied electric field, the peak would be normally expected to occur at shorter times for higher voltages following the well-known relationship

$$v = \mu E , \tag{1}$$

where v is carrier velocity, μ is carrier mobility, and E is electric-field intensity. The experimental results as shown in Fig. 3, however, are not consistent with Eq. (1) for the applied voltage below V_c. These unexpected results on the time occurrence of the peak current are the main subject of our investigation. This behavior in NLC's has not been previously reported to the best of our knowledge.

One simple model for the mechanism of the occurrence of the peak in the transient current induced by the polarity reversal of a voltage below V_c is proposed. It is clear that the occurrence of the peak in the transient current will not be due to the space-charge-limited current because of the lack of agreement with Eq. (1) and Child's law in Fig. 2. Also the occurrence of the peak does not originate in the spatial distribution of charge carrier mobility in the bulk as reported in Refs. 5 and 6, because the director rotation of NLC's does not occur for voltages below the threshold voltage for the Freedericksz transition and in the isotropic phase. For simplicity we do not consider the effects based on electrohydrodynamic instabilities[11] and liquid flow.

There have been many reports[6,12–14] on the existence of adsorbed charge layers which are formed by immobilized charges with the same sign on each substrate interface. The total amount of charge Q per unit area is introduced by impurities in NLC's and/or the electric decomposition of liquid-crystal molecules, and satisfies overall charge neutrality in the bulk as follows:

$$Q = Q^{+} = Q^{-} , \tag{2}$$

where $Q^{+} = \bar{\rho}^{+} d$ and $Q^{-} = 2\sigma^{-} + \bar{\rho}^{-} d$ (d is film thickness, σ^{-} is adsorbed charge density per unit area, and $\bar{\rho}^{+}$ and $\bar{\rho}^{-}$ are the average volume density of the positive and negative charge, respectively). The adsorbed charge layers exist with or without the external applied voltage and produce an asymmetric electric field. The internal electric field $E_s(z)$ originates in these adsorbed charge layers and exists spontaneously in the vicinity of the substrate surfaces. The z axis is taken as normal to the substrate surface, and each substrate surface is located at $z = d/2$ and $z = -d/2$. The internal electric field $E_s(z)$ is obtained by the numerical calculation of Poisson's equation as

$$\frac{\partial E_s(z)}{\partial z} = \rho^{+}(z)/(\epsilon\epsilon_0) , \tag{3}$$

where $\rho^{+}(z)$ is the distribution function of the diffused charge density compensating the surface adsorbed charges, ϵ_0 is permittivity of free space, and the dielectric constant ϵ is defined by the independent quantity of the director orientation. For simplicity the distribution function of the internal electric field is assumed to be exponential and is written

$$E_s(z) = (\sigma^{-}/\epsilon\epsilon_0)\exp[-z/L_D(z)] , \tag{4}$$

where $L_D(z)$ is the thickness of the compensating diffused charge layer and has the relation

$$[L_D(z)]^2 = \epsilon k_B T/[e\rho^{+}(z)] .$$

(k_B is the Boltzmann constant, T is the absolute temperature, and e is the elementary electric charge.) $\rho^{+}(z)$ satisfies the relation

$$\bar{\rho}^{+} d = \int_{-d/2}^{d/2} \rho^{+}(z) dz . \tag{5}$$

Further details on the derivation of $E_s(z)$ are given in Ref. 6.

The external electric-field intensity across the cell accompanying the polarity reversal of the applied voltage changes with time. Figure 4 shows the equivalent circuit of the NLC cell used to calculate the time variation of the electric-field intensity, where R_1 and C_1 are the resistance and the capacitance of the bulk of the NLC, respectively, and R_2 is the contact resistance between the electrode and the NLC. It is assumed for simplicity that the electric field produced by the applied voltage is uniform across the cell. The time variation of the electric-field intensity $E_T(t)$ accompanying the polarity reversal of the applied voltage V is presented as

$$E_T(t) = (V/d)[1 - 2\exp(-t/\tau_c)] , \tag{6}$$

where d is the cell thickness and τ_c is the time constant of the equivalent circuit as presented by

$$\tau_0 = C_1 R_1 R_2/(R_1 + R_2) . \tag{7}$$

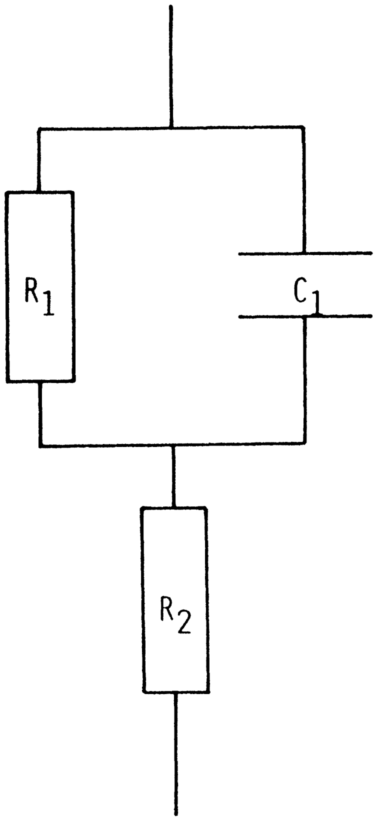

FIG. 4. Equivalent circuit of the NLC cell used to calculate the time variation of the electric-field intensity, where R_1 and C_1 are the resistance and the capacitance of the bulk, respectively, and R_2 is the contact resistance between the electrode and the NLC's.

The relationship between a time and position of the electric-field intensity across the cell is given by

$$E(z,t) = E_s(z) + E_T(t) . \tag{8}$$

Now we can calculate the transient current for the small signal case. Then the surface density ρ_s of the drift carrier sheet is small compared to the total amount of charge Q in the bulk. The drift of the carrier sheet across the cell causes negligible field perturbations. The transient current density $J(z,t)$ accompanying the transport of the carrier sheet across the cell is generally given by[15]

$$J(z,t) = (\rho_s/d)\mu E(z,t) , \tag{9}$$

in which the diffusion current term is neglected for simplicity. The carrier density will be continuously decreased by the loss of carriers due to recombination. The carrier recombination can be characterized by a bulk lifetime τ_t which is independent of the carrier density. Then the drift carrier density is given by

TABLE I. Representative values of significant parameters.

Name	Symbol	Representative value	Source
Film thickness	d	6.8 μm	a
Cell resistance	R_1	200 MΩ	a
Cell capacitance	C_1	3.18 μF	a
Contact resistance	R_2	6 kΩ	b
Mobility	μ	2.5×10^{-10} m^2/V sec	c
Total charge density	Q	2×10^{-4} C/m^2	c
Adsorbed charge density	σ^-	0.8×10^{-4} C/m^2	c
Carrier sheet density	ρ_s	1×10^{-5} C/m^2	c
Dielectric constant	ϵ	12.1	c
Carrier lifetime	τ	0.05 sec	d

[a] Measured value in this work.

[b] Estimated value from the results reported in Refs. 12 and 16.

[c] Reported value in Ref. 6 corresponding to the material studied.

[d] Assumed value for the adjustable parameter of the time occurrence of peak current to the experimental results in this work. This value does not essentially affect the singular phenomenon.

$$\rho_s(t)=\rho_s\exp(-t/\tau_t)\ . \tag{10}$$

Further discussion of the carrier recombination center is beyond the scope of this simple model. Various characteristics of the transient current are obtained by numerical calculations based on the simple model described above. The physical quantities used in these calculations are $R_1=200$ MΩ and $R_2=6$ KΩ. Other physical quantities used in these calculations are listed in Table I, and the origins of these values are also explained in its caption. The results of the numerical calculations using Eqs. (2)–(10) for the transident current shapes are shown in Fig. 5 for various applied voltages less than V_c. The time occurrence of the peak in these transient currents decreases with decreasing applied voltage. This unusual behavior of the time occurrence of the peak in the transient current is mainly determined by the contact resistance, as is apparent in Eqs. (6)–(8). The solid line in Fig. 3 also shows the numerical result of the time occurrence of the peak in the transient current as a function of the applied voltage. These numerical results are in agreement with the experimental results as shown in Figs. 1–3.

Both the spontaneous internal electric field and the time variation of the external electric field induced by the polarity reversal of the voltage also affect the transient current of the applied voltage above V_c. However, applied voltages above V_c induce the changes in the mobility distribution originating in the director rotation. Now we can define the characteristic voltage V_c as the critical voltage for changing the mechanism dominating the occurrence of the peak in the transient current. Above V_c the mechanism is dominated by the mobility distribution and below V_c it is dominated by the time variation of the external electric-field intensity based mainly on the time constant of a NLC cell. From our experimental results, the value of the characteristic voltage is 1.49 V. In our simple model, however, the characteristic voltage cannot be theoretically derived.

A similar unusual phenomenon in the transient currents is observed for NLC cells containing different substrate surfaces with and without polyimide layers which act on the treatment layers for the molecular alignment and as blocking layers for the charge injection from the electrodes. Thus the electrochemical effects in the vicinity of the substrate surface are not considered to be the origin of the unusual behavior phenomenon. In our simple model, we have not identified the origin of the charge carriers either as electrons or ions. However, it is not necessary to consider the origin of the charge carriers in order to apply our simple model. Finally, we must make special mention of the internal electric field that originates in the vicinity of the substrate surface. In these regions there would occur dipole-dipole association and/or selective absorption of nematic molecules on the substrate as reported in Refs. 12–14, 16, and 17.

The unusual behavior of the occurrence of the peak in

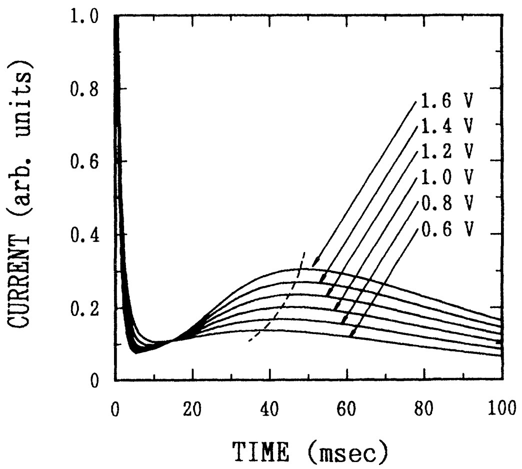

FIG. 5. Numerical calculation results on the transient current shapes for a series of values of the applied voltage. The dashed line presents the peak positions of each current shape.

the transient current has been observed for particular NLC materials in the nematic and isotropic phases. If our model is correct, this effect originates in the competition of internal and external electric fields as shown in Eq. (8). The requirement for the occurrence of the unusual behavior is that spontaneous internal electric fields exist in the vicinity of the substrate surfaces and the sample has a long time constant as shown in Eq. (7). Thus this behavior will occur for other types of liquid crystals and for many types of organic materials with similar field distributions in the vicinity of the interface between a material and electrodes, and similar transport properties.

IV. CONCLUSION

The unusual behavior of the occurrence of the peak in the transient current induced by the polarity reversal of a dc voltage applied to a NLC cell has been studied. A peak in the transient current has been observed for polarity reversal of an applied voltage below the threshold voltage for the Freedericksz transition and in the isotropic phase without director rotation. Contrary to behavior expected for a normal carrier transport process, the time occurrence of the peak in the transient current has decreased with the decrease of an applied voltage below the characteristic voltage V_c. In our experiments, the characteristic voltage of 1.49 V has been estimated. The unusual behavior has been quantitatively explained by a simple model based on the time variation of an external applied voltage after the polarity reversal and the internal electric field which is induced by the adsorbed electric charges on the cell substrate. The results from the model agree reasonably well with the experimental observations.

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