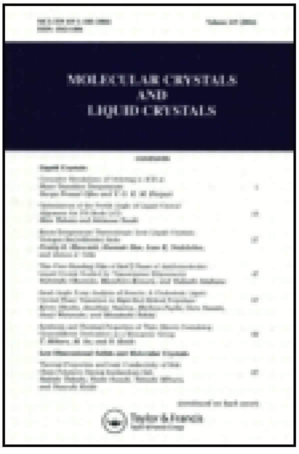

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Determination of Rotational Viscosity and Pretilt Angle in Nematic Liquid Crystals from Transient Current: Influence of Ionic Conduction

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Determination of Rotational Viscosity and Pretilt Angle in Nematic Liquid Crystals from Transient Current: Influence of Ionic Conduction

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A method is described for the measurement of the rotational viscosity γ_1 in nematic liquid crystal cell assemblies. Theoretical formulas to determine γ_1 from transient current are developed by considering both director rotation and conduction of impurity ions. The pretilt angle and the product of the drift mobility and density of the ions are also obtained with the method. Experimental results for 4-pentyl-4′-cyano biphenyl cells with or without polyimide alignment layers are reported.

Keywords: Nematic liquid crystal, rotational viscosity, pretilt angle, impurity ion, transient current

1. INTRODUCTION

Nematic liquid crystals (NLCs) are currently used for display devices. Measurement of the rotational viscosity γ_1 and pretilt angle θ_0 in device configurations is particularly important in establishing efficient device design; γ_1 dominates the response times of NLCs to an externally applied field, and θ_0 governs the viewing characteristics,[1] creation or elimination of disclinations,[2] and response times.

We have developed an accurate and simple method for the measurement of γ_1 in previous papers.[3–5] The method is based on the analysis of a displacement current peak due to director rotation in NLC cells excited by a *dc* step voltage pulse.[6] The major advantages over conventional methods[7–18] are that γ_1-values as well as θ_0-values can be measured in actual device configurations with inexpensive instruments. Furthermore, the displacement current peak directly yields the γ_1-values, whereas electro-optic[13–15] or electro-capacitive[16–18] methods require the differentiation of transmittance or capacitance with respect to time, respectively, in order to measure γ_1 with the same accuracy as the present method. Applicability of this method is, however, restricted to the cell in which the conduction current is much smaller than the displacement current.

NLC materials are conductive, i.e., always contain impurity ions. These ions originate from more or less dissociated impurities as well as from the spontaneous dissociation of the NLC molecules themselves. Since the impurity ions significantly influence the NLC devices, the transport mechanism of the impurity ions have been studied with transient current techniques.[19–25] For NLC materials containing relatively large amounts of impurity ions, the influence of the ionic conduction current on the analysis in the present method must be considered.

In this paper, we derive theoretical expressions, which enable us to obtain γ_1 from the transient current, by considering director rotation and conduction current, and apply the analysis to the cell containing impurity ions whose density is not negligible. We can determine γ_1, θ_0, and the product of the drift mobility and density of the ions by means of the present technique.

2. THEORY

We consider the one-dimensional dynamic behavior of the director $\mathbf{n} = (\sin\theta, 0, \cos\theta)$ and impurity ions in NLCs along the z-axis which is in the direction of an externally applied electric field, where θ is the tilt angle between the director and the z-axis. Before the electric field application, uniform distributions of the director and of the positive and the negative ions with the same density n are assumed. Neglecting the elastic and the flow terms, we express the director motion as[26]

$$\gamma_1 \frac{d\theta(t)}{dt} = -\frac{1}{2}\varepsilon_0 \Delta\varepsilon E^2 \sin 2\theta(t), \tag{1}$$

where ε_0 is the dielectric permittivity of vacuum, $\Delta\varepsilon$ is the dielectric anisotropy, $E(=V/L)$ is the applied electric field, V is the applied voltage, and L is the cell thickness. Integration of Equation (1) yields

$$\theta(t) = \tan^{-1}\left[\tan\theta_0 \exp\left(-\frac{\varepsilon_0 \Delta\varepsilon E^2}{\gamma_1} t\right)\right], \tag{2}$$

where θ_0 is the initial tilt angle and identical to the pretilt angle. The displacement current I_d due to the director response to the applied electric field is given by

$$I_d(t) = SE\frac{d}{dt}\varepsilon(\theta(t)), \tag{3}$$

where S is the area of the electrodes, ε is the dielectric permittivity of NLCs,

$$\varepsilon(\theta(t)) = \varepsilon_0[\varepsilon_\perp + \Delta\varepsilon \cos^2\theta(t)], \tag{4}$$

and $\varepsilon_\perp$ is the dielectric constant normal to the director. The conduction current I_c due to the drift of a single species of monovalent mobile ions in a medium sandwiched between two electrodes (at least one of the electrodes is blocking) is given by

$$I_c(t) = Sqn\mu E\left(1 - \frac{t}{t_0}\right), \tag{5}$$

in $0 \le t \le t_0$ under the small signal condition, where q is the electronic charge, t_0 $(= L/\mu E)$ is the ion transit time, and μ is the drift mobility of the ions, whose anisotropy is neglected. Using Equations (1)–(5), we have the total current $I(t)$ flowing in an NLC cell,

$$I(t) = I_d + I_c$$

$$= \frac{S[\varepsilon_0 \Delta\varepsilon \sin 2\theta(t)]^2 E^3}{2\gamma_1} + Sqn\mu E\left(1 - \frac{t}{t_0}\right) \tag{6}$$

$$= \frac{2S(\varepsilon_0\Delta\varepsilon)^2 E^3}{\gamma_1}\left\{\frac{\tan\theta_0 \exp\left[-\frac{\varepsilon_0\Delta\varepsilon E^2}{\gamma_1}t\right]}{1 + \tan^2\theta_0 \exp\left[-\frac{2\varepsilon_0\Delta\varepsilon E^2}{\gamma_1}t\right]}\right\}^2$$

$$+ Sqn\mu E\left(1 - \frac{t}{t_0}\right). \tag{7}$$

We find from Equations (1) and (6) that the current has a peak at $\theta = 45^\circ$ in much shorter time range than that of t_0,[23,25,27] $(1 \gg t/t_0)$. The peak current I_p which is the current at the peak is

$$I_p = \frac{S(\varepsilon_0\Delta\varepsilon)^2 E^3}{2\gamma_1} + Sqn\mu E. \tag{8}$$

The peak time t_p at which the current has the peak is obtained from Equation (2) as

$$t_p = \frac{\gamma_1 \log(\tan\theta_0)}{\varepsilon_0\Delta\varepsilon E^2}, \tag{9}$$

where $\theta_0 > 45^\circ$ is necessary to observe the current peak. Note that θ_0 does not contribute to I_p but to t_p. These two equations [Equations (8) and (9)] are valid for $t_p \ll t_0$ and $\xi \ll L$, where $\xi[= L\sqrt{K_1/(\varepsilon_0\Delta\varepsilon)}/V]$ is the coherence length for the strong anchoring condition and characterizes the elastic deformation region in the vicinity of the electrodes,[28] and K_1 is the splay elastic constant. The conditions $t_p \ll t_0$ and $\xi \ll L$ are identical to $V \gg \mu\gamma_1 \log(\tan\theta_0)/(\varepsilon_0\Delta\varepsilon) \equiv V_{c1}$ and $V \gg \sqrt{K_1/(\varepsilon_0\Delta\varepsilon)} \equiv V_{c2}$, respectively.

From Equation (8) we have

$$\frac{I_p}{E^3}=\frac{S(\varepsilon_0\Delta\varepsilon)^2}{2\gamma_1}+Sqn\mu\frac{1}{E^2}. \tag{10}$$

Equations (9) and (10) relate the peak time and peak current in the transient current to the physical quantities (γ_1, $\Delta\varepsilon$, θ_0 and $qn\mu$). If one of three quantities (γ_1, $\Delta\varepsilon$ and θ_0) is known, $qn\mu$ and the other two quantities can be determined according to the present theory. Since $\Delta\varepsilon$ can accurately and easily be obtained from dielectric measurement, γ_1-values can be determined from the extrapolation of I_p/E^3 to $1/E^2=0$. We can also determine $qn\mu$-products and θ_0-values from the slope of I_p/E^3 vs $1/E^2$ and of t_p vs $1/E^2$, respectively.

3. EXPERIMENT

The NLC material used in the present experiment was 4-pentyl-4′-cyano biphenyl (5CB) with positive dielectric anisotropy. The NLC was introduced between two pieces of glass with a transparent In_2O_3 electrode, whose area was 2 cm^2. Three cells with different thicknesses (5.3, 12 and 44 μm) were prepared. Polyimide alignment layers (100 nm thickness) were coated on the substrate surfaces of the cells with 5.3 and 44 μm in thickness. All surfaces were unidirectionally rubbed in an antiparallel manner to produce homogeneous alignment.

A *dc* voltage pulse was applied to the cell at constant temperatures, where 5CB is in the nematic phase. Transient currents were observed through a series load resistor by means of a wide-band pre-amplifier and a digital storage oscilloscope. The measured data were transferred to a micro-computer for the analysis and the storage.

4. RESULTS AND DISCUSSION

Current transients of the 5CB cell with 12 μm in thickness excited by various *dc* voltage pulses at 303 K are shown in Figure 1. A remarkable current peak in the transients due to director rotation is observed. The current baselines caused by ionic conduction are too high to be neglected while those in ZLI-2293 (Merck) are negligible as reported in References 3–5. Thereby, we must take account of the influence of the ionic conduction current. The expression of the conduction current [Equation (5)] is applicable to the 5CB cells because both the In_2O_3 electrode and the polyimide layer block carrier injection,[24,29] and the anisotropy of the drift mobility of the ions in 5CB is small.[30]

Plots of I_p/E^3 vs. $1/E^2$ and t_p vs. $1/E^2$ for the three cells at 303 K are shown in Figures 2 and 3, respectively. Using the physical quantities of a 5CB cell at 303 K such as $\gamma_1=0.063$ Pa·s, $\theta_0=83.5°$ (these two quantities are measured in this work), $\mu=3.5\times10^{-6}$ cm^2/(V·s),[25] $\Delta\varepsilon=9.7$,[11] and $K_1=5.1\times10^{-12}$ N,[11] V_{c1} and V_{c2} are estimated to be 0.6 V and 0.2 V, respectively. It is obvious that V_{c1} determines the lower

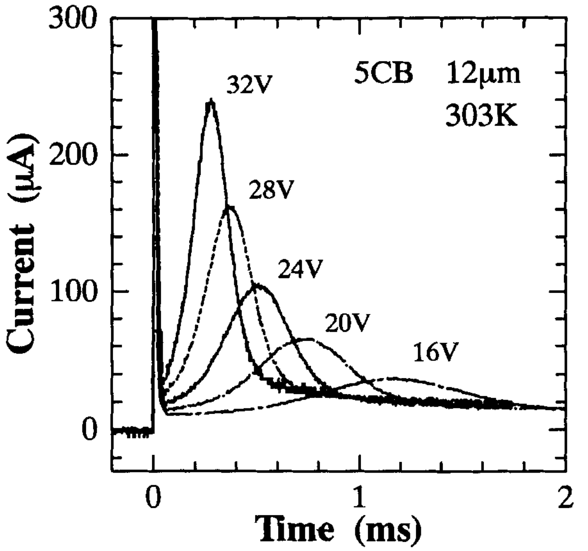

FIGURE 1 Transient current shapes in the 5CB cell with 12 μm in thickness excited by various *dc* voltage pulses at 303 K.

voltage limit of the experiment and is much smaller than the applied voltages used here. There exists the upper voltage limit for the observation of the current peak as well; the initial charging current affects the current peak because the peak time becomes very short above the upper voltage limit. We find that experimental data points lie on the straight lines in Figures 2 and 3, indicating the validity of the present analysis.

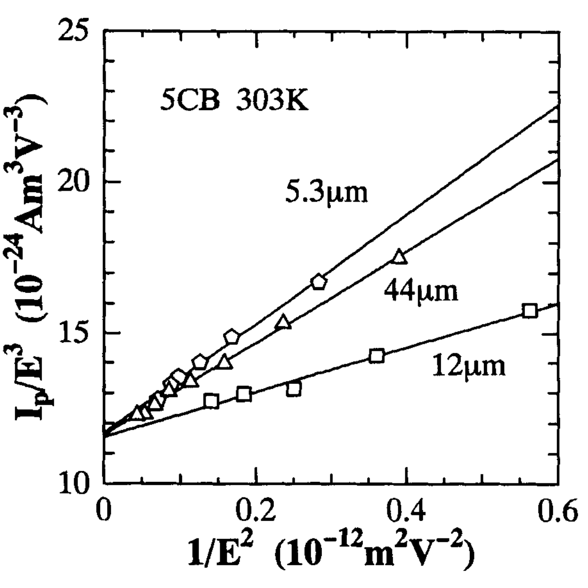

FIGURE 2 A plot of I_p/E^3 vs. $1/E^2$ for the 5CB cells at 303 K.

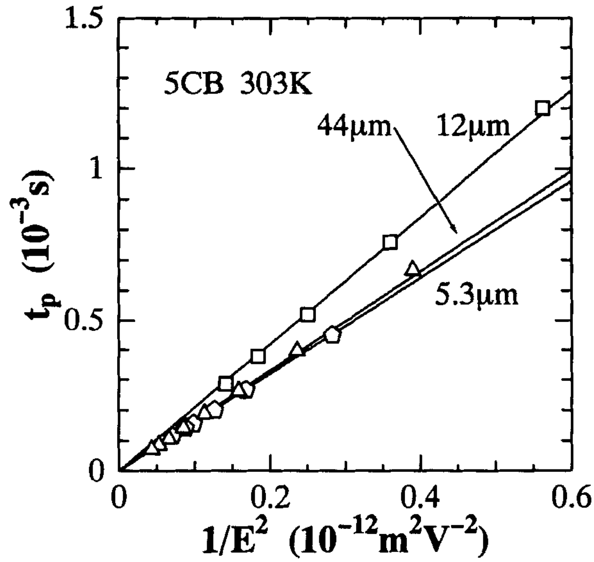

FIGURE 3 A plot of t_p vs. $1/E^2$ for the 5CB cells at 303 K.

We use $\Delta\varepsilon$-values reported in the literature[11] to determine γ_1-values of 5CB. The extrapolation of I_p/E^3 to $1/E^2 = 0$ in Figure 2 gives the values of $S(\varepsilon_0\Delta\varepsilon)^2/(2\gamma_1)$, from which γ_1 can be determined. The $qn\mu$-products and the θ_0-values are also obtained from the slopes of the plot of I_p/E^3 vs $1/E^2$ in Figure 2 and of the plot of t_p vs $1/E^2$ in Figure 3, respectively. These results are listed in Table I. The density n of the impurity ions incorporated in the cells with polyimide layers relatively large for the cell without the layers, as manifested from Table I. The conductivity of 5CB is reported[24,29,30] to be in the range 10^{-9}–10^{-10} S·cm^{-1}, which is consistent with our results. It is evident from Table I that (1) γ_1-values are identical for the cells with or without polyimide alignment layers, (2) the γ_1-values are independent of the cell thickness, and (3) the θ_0-values are independent of the cell thickness as long as the surface treatment is the same. The fact (1) shows that the difference in anchoring energy influence the lower voltage limit of the measurement due to the elastic deformation, but does not the measured values of γ_1.

We show dependence of I_p/E^3 and t_p on $1/E^2$ as a function of temperature for the 12 μm-thickness cell in Figures 4 and 5, respectively. We find that the values of I_p/E^3 and t_p at various temperatures are also proportional to the $1/E^2$-values. Obtained

TABLE I

The rotational viscosity γ_1, the product of the density and drift mobility of the ions $qn\mu$, and the pretilt angle θ_0 for the 5CB cells measured at 303 K.

L(μm)	Alignment layers	γ_1(Pa·s)	$qn\mu$ (S·cm^{-1})	θ_0(°)
5.3	Polyimide	0.063	9.1×10^{-10}	83.5
12	–	0.064	3.7×10^{-10}	86.5
44	Polyimide	0.063	7.6×10^{-10}	83.9

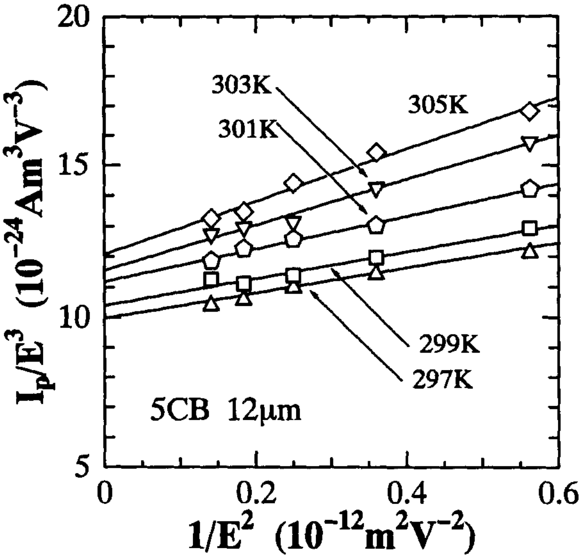

FIGURE 4 A plot of I_p/E^3 vs. $1/E^2$ for the 12 μm-thickness 5CB cell at various temperatures.

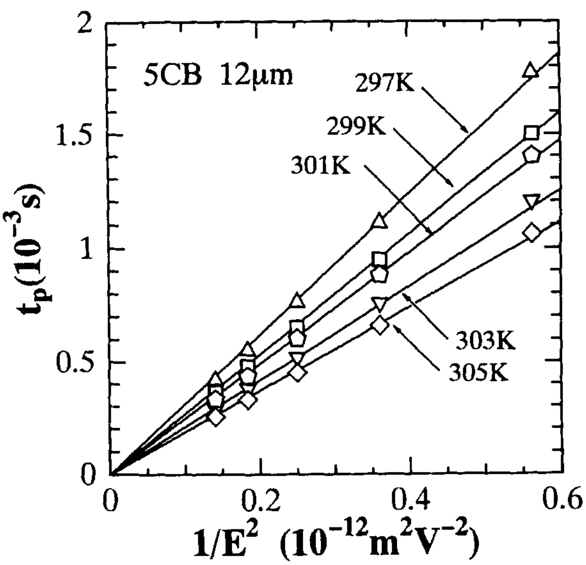

FIGURE 5 A plot of t_p vs. $1/E^2$ for the 12 μm-thickness 5CB cell at various temperatures.

values of γ_1, $qn\mu$ and θ_0 in the same manner are shown in Figures 6 and 7. The activation energies for γ_1 and $qn\mu$ measured with this method are estimated from Figure 6 to be 0.74 and 0.77 eV, respectively. From Figure 7 we find that θ_0-values does not greatly change in the temperature range. The data of γ_1 are in excellent agreement

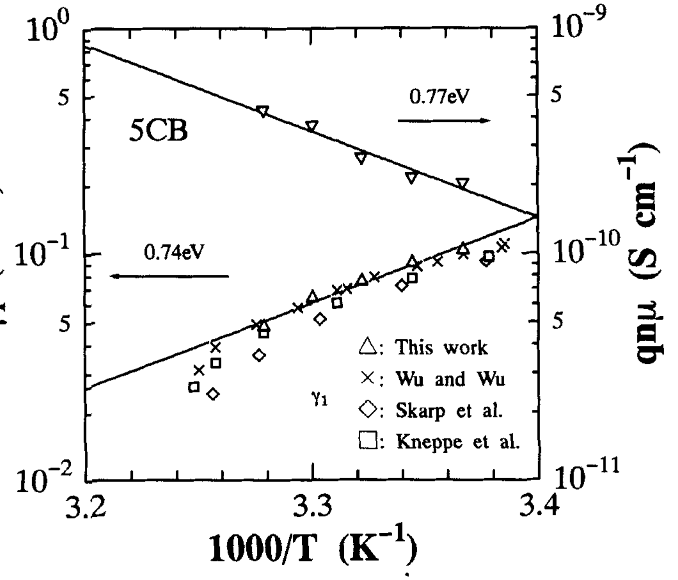

FIGURE 6 Temperature dependence of γ_1 and $qn\mu$ for the 5CB cell with 12 μm in thickness. △, ◇, □ and × represent the data points of γ_1 measured in this work, by Skarp *et al.* (Reference 11), by Kneppe *et al.* (Reference 12), and by Wu and Wu (Reference 15), respectively. ▽ represents the data points of $qn\mu$ measured in this work. The activation energies of γ_1 and $qn\mu$ are estimated to be 0.74 and 0.77 eV, respectively.

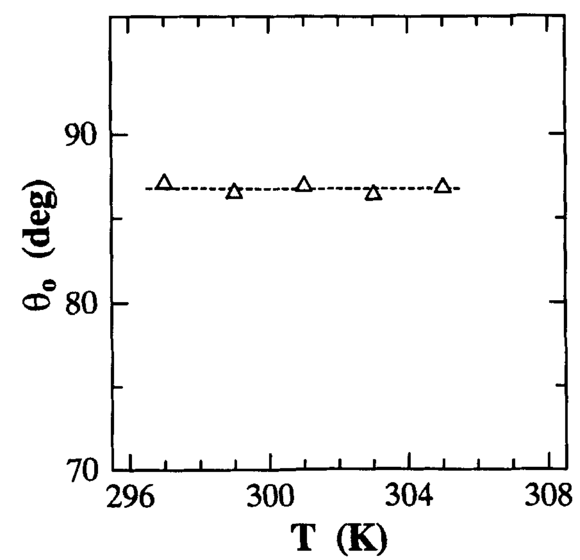

FIGURE 7 Temperature dependence of θ_0 for the 5CB cell without polyimide alignment layers.

with the electro-optic data[15] in a device configuration but slightly deviate from the data using the Poiseuille flow and torsional flow methods[11] or the rotating magnetic field method.[12] Thus we stress that the measurement of γ_1 in device configurations is essential for the estimation of the response times of NLC devices.

5. CONCLUSIONS

We have derived the expressions of transient current in NLCs by taking account of the director response to the electric field and the influence of the ionic conduction current. The peak time and peak current in the transient current are related to the physical quantities (γ_1, $\Delta\varepsilon$, θ_0 and $qn\mu$) by the two equations [Equations (9) and (10)]. Since $\Delta\varepsilon$ is accurately obtained from dielectric measurement, the other three quantities can be determined. We have measured the transient current of 5CB cells containing impurity ions. The γ_1-values of 5CB cells for the different surfaces and thicknesses are obtained from the analysis of the current, and are found to be the same and in excellent agreement with the values using the electro-optic method. We also find that the θ_0-values of 5CB cells obtained from this method are dependent on the surface treatment but not on the cell thickness. The activation energies for γ_1 and $qn\mu$ are estimated to be 0.74 and 0.77 eV, respectively.

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