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J. Robert [a], G. Labrunie [a] & J. Borel [a]

[a] Laboratoire d'Électronique et de Technologie de l'Informatique, Centre d'Étude Nucléaires de Grenoble 38-Grenoble-Gare

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Static and Transient Electric Field Effect on Homeotropic Thin Nematic Layers†

J. ROBERT, G. LABRUNIE and J. BOREL

Laboratoire d'Électronique et de Technologie de l'Informatique
Centre d'Étude Nucléaires de Grenoble
38-Grenoble-Gare

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Abstract—The experiments presented in this paper give a way to the determination of two elastic constants K_{11} and K_{33} and one viscosity coefficient γ_1 of a nematic liquid crystal. ac electric field stresses applied to a thin layer of nematic liquid crystal modify its cooperative molecular alignment.

So, the electrically controlled rotation of molecules induces a variable birefringence as a function of applied voltage and time. The experimental results in relation with the static and dynamic behaviour lead to the determination of these previously stated parameters. Temperature dependence of these parameters is given in the nematic range.

1. Introduction

A liquid crystal in its mesophase is characterized by a long range order. Distortions of orientation may be imposed by external stresses (electric or magnetic fields), by boundary conditions or spontaneously by thermal fluctuations. The bulk elastic properties of liquid crystals are determined by elastic constants.

In this paper, only nematic liquid crystals submitted to ac electric field stresses are investigated. Their behaviour is optically detected through birefringence. The static and dynamic behaviours of this electro-optic effect are considered and they allow the determination of two elastic constants and one viscosity coefficient. The curves giving temperature dependence of these parameters are drawn.

2. Constitutive Equations

We investigate the case of negative molecular dielectric anisotropy, the nematic liquid crystal being sandwiched between two parallel

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and conductive plates and the mean angular position of molecules at rest being perpendicular to the plates.

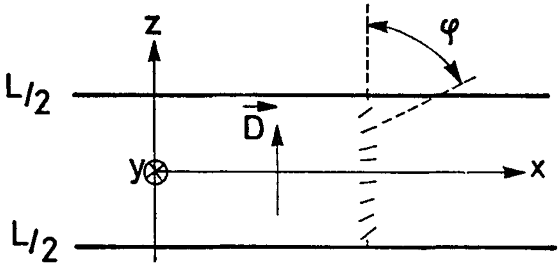

Figure 1. Configuration of a nematic system submitted to an ac electric field in the Freedericksz's geometry.

In this case, Eq. (1) giving the free elastic and electric energy(1) of the system is:

$$F = \tfrac{1}{2}\int_{-L/2}^{+L/2}\left[(K_{11}\sin^2\varphi + K_{33}\cos^2\varphi)\varphi_z^{02} + \frac{\epsilon_a D_z{}^2}{4\pi\epsilon_\parallel{}^2}\,\frac{\sin^2\varphi}{1-(\epsilon_a/\epsilon_\parallel)\sin^2\varphi}\right]\mathrm{d}z \tag{1}$$

ϵ_a = dielectric anisotropy, D = displacement vector.

Static Behaviour

With boundary conditions:

$$z = \pm L/2 \qquad \varphi = 0$$
$$z = 0 \qquad \varphi = \varphi_M \qquad \varphi_z{}^0 = 0$$

The static state is defined by F minimum. The solving of this equation leads to the angular distribution of molecules $\varphi(z)$ in the cell for an applied voltage V.

V and the component along z axis of electrical induction (D_z) are connected by:

$$V = \frac{D_z}{\epsilon_\parallel}\int_{L-/2}^{+L/2}\frac{\mathrm{d}z}{1-(\epsilon_a/\epsilon_\parallel)\sin^2\varphi} \tag{2}$$

$\epsilon_\parallel$ and $\epsilon_\perp$ being the two components of dielectric constant.

The mean value of the refractive index (light being perpendicular to the plates) is so determined by the relation:

$$\langle n\rangle = \frac{n_0 n_e}{L}\int_{-L/2}^{+L/2}\frac{\mathrm{d}z}{\{n_e{}^2\cos^2[\varphi(z)] + n_0{}^2\sin^2[\varphi(z)]\}^{1/2}}. \tag{3}$$

Using a computer, we have drawn a set of curves $\langle n \rangle$ versus V for different values of the parameter

$$\eta = \frac{K_{11} - K_{33}}{K_{33}}$$

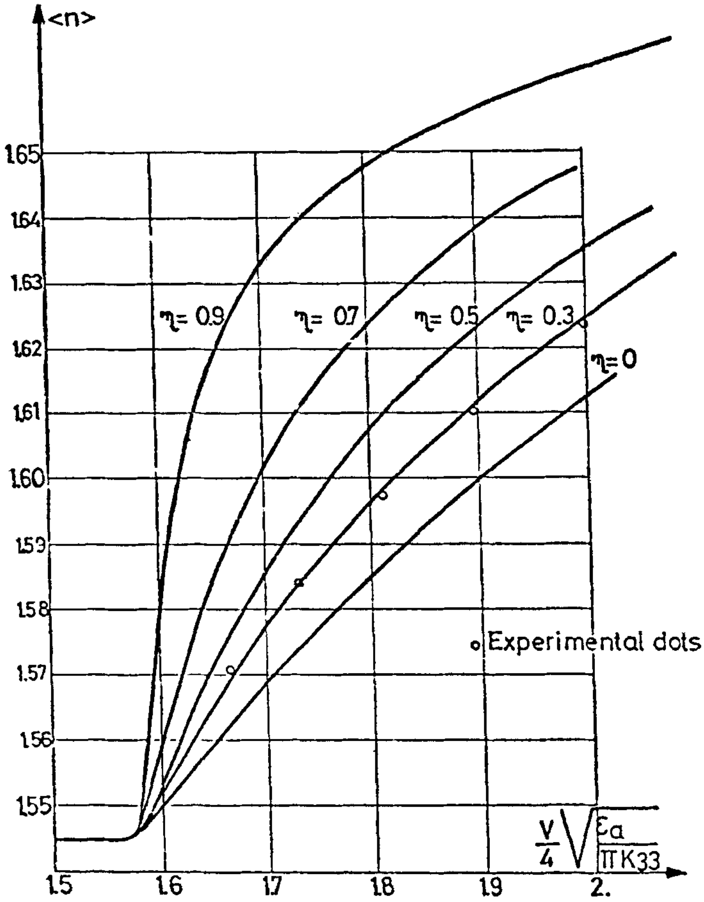

Figure 2. Theoretical curves in the static case for different values of parameter η and experimental dots.

when $n_0 = 1.545$

$n_e = 1.755$

$\epsilon_{\parallel} = 4.72$

$\epsilon_{\perp} = 5.28$

Approximate calculations can be achieved when φ and φ_M are small angles.

$$\frac{\langle n\rangle}{n_0} \simeq 1+\frac{n_e{}^2-n_0{}^2}{4n_e{}^2}\cdot\frac{V^2+(4\pi^3 K_{33}/\epsilon_a)}{[\frac{2}{3}-(\epsilon_a/\epsilon_\parallel)]V^2-\eta(4\pi^3 K_{33}/\epsilon_a)} \tag{4}$$

when $\epsilon_a < 0$ (negative dielectric anisotropy) the threshold voltage is given by:

$$V_{\text{th}} = 2\pi\sqrt{\pi\frac{K_{33}}{|\epsilon_a|}}$$

The fitting between theoretical curves and experimental dots leads to the determination of elastic constants K_{11} and K_{33}.

Dynamic Behaviour

The elastic and electrical couple per volume unit is:

$$\Gamma_1 = -\Bigg[(K_{11}\sin^2\varphi+K_{33}\cos^2\varphi)\varphi_z^{00}+(K_{11}-K_{33}) \cdot\sin\varphi\cos\varphi\,\varphi_z^{02}-\frac{\epsilon_a D_z{}^2(t)}{4\pi\epsilon_\parallel{}^2}\cdot\frac{\sin\varphi\cos\varphi}{[1-(\epsilon_a/\epsilon_\parallel)\sin^2\varphi]^2}\Bigg] \tag{5}$$

If we assume that the orientation of the unit vector **L** is changed with a speed **du/dt** without gravity center movement, viscosity couple may be written:

$$\Gamma_2 = \gamma_1\left(\mathbf{u}\times\frac{\mathbf{du}}{\mathbf{dt}}\right) = \gamma_1\cdot\frac{\mathrm{d}\varphi}{\mathrm{d}t} \tag{6}$$

at equilibrium

$$\Gamma_1+\Gamma_2 = 0 \tag{7}$$

The mathematical treatment of this equation is rather complex: so, approximate formulas are deduced assuming the angular distribution in the cell being $\varphi(z,t) = \varphi_M(t)\cos\pi(z/l)$ for small values of φ.

$$-\left(\pi^2 K_{33}+\frac{\epsilon_a V^2}{4\pi}\right)\varphi_M-\left[\pi^2(K_{11}-K_{33})-\frac{\epsilon_a V^2}{4\pi}\left(\frac{2}{3}-\frac{\epsilon_a}{\epsilon_\parallel}\right)\right]\varphi_M{}^3 = L^2\gamma_1\frac{\mathrm{d}\varphi_M}{\mathrm{d}t} \tag{8}$$

This equation leads to the approximate result:

$$\frac{\langle n\rangle-n_0}{n_0} \simeq \frac{n_e{}^2-n_0{}^2}{4n_e{}^2}\cdot\frac{V^2-V_{\text{th}}^2}{[\frac{2}{3}+(|\epsilon_a|/\epsilon_\parallel)]V^2+\eta V_{\text{th}}^2}\cdot\frac{1+\tanh(t-t_0/T)}{2} \tag{9}$$

with

$$\frac{1}{T} = \frac{|\epsilon_a|}{4\pi L^2 \gamma_1} \cdot (V^2 - V_{\mathrm{th}}^2) \tag{10}$$

and t_0 is an integration constant function of thermal fluctuations.⁽²⁾

The experimental value of the time constant permits the determination of viscosity coefficient γ_1.

3. Experiments

The nematic liquid crystal used is MBBA (*p*. Metoxybenzylidene-*p*-*n*-butylaniline) and we have studied the cooperative molecular alignment of this nematic compound submitted to an ac electric field by birefringence measurement. MBBA is sandwiched between two conducting reflecting (slightly transparent) electrodes. The cell is made of two optically flattened glass plates coated with aluminium (300 Å) and indium oxide. Indium oxide coating suitably cleaned and rubbed gives, without use of surfactant deposited on the plates or mixed into the liquid crystal, an homeotropic structure to MBBA. Distance between the two plates is fixed by mylar spacers.

In order to measure the indices, the two glass plates are inclined at a small angle (one half minute) and an interference fringes pattern can be observed. Electric field necessary to tilt molecules has to be alternating or pulsed in order to avoid ion migration and limit layer formation (Fig. 3). Mean value of refractive index as a

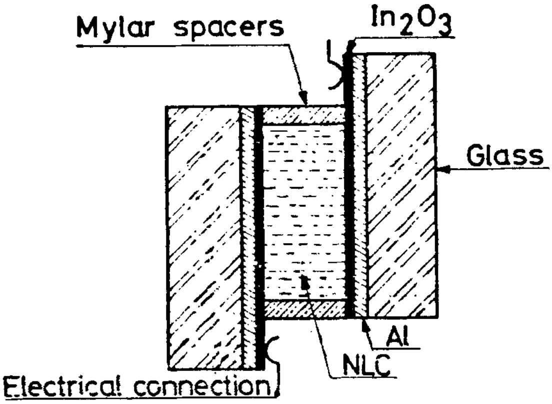

Figure 3. Cut view of liquid crystal cell.

function of time and voltage is measured by detecting fringes in front of a photomultiplier detector or a telescope. A drawing of the experimental set-up is given on Fig. 4, and Fig. 5 shows the detected optical signal.

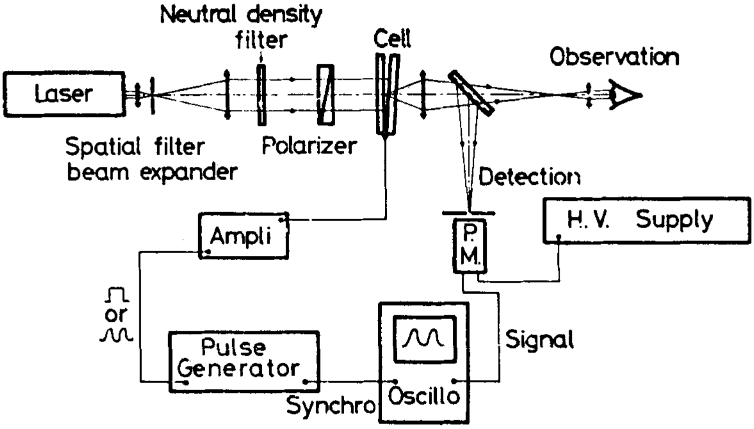

Figure 4. Experimental measurement set-up.

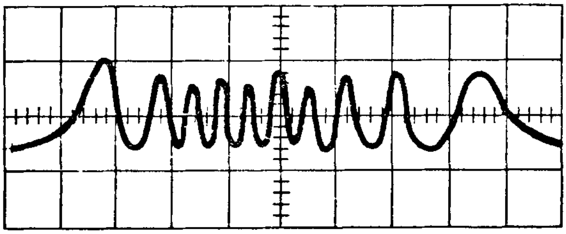

Figure 5. Detected optical signal when pulsed ac electric field is applied to the cell.

4. Experimental Results

Static Behaviour

To determine the temperature dependence of the two elastic constants K_{11} and K_{33} and viscosity coefficient γ_1, we have to know temperature dependence of dielectric anisotropy and indices. The first one has been given by Diguet *et al.*⁽³⁾ and the second one according to ordinary index n_0 by Haller *et al.*⁽⁴⁾

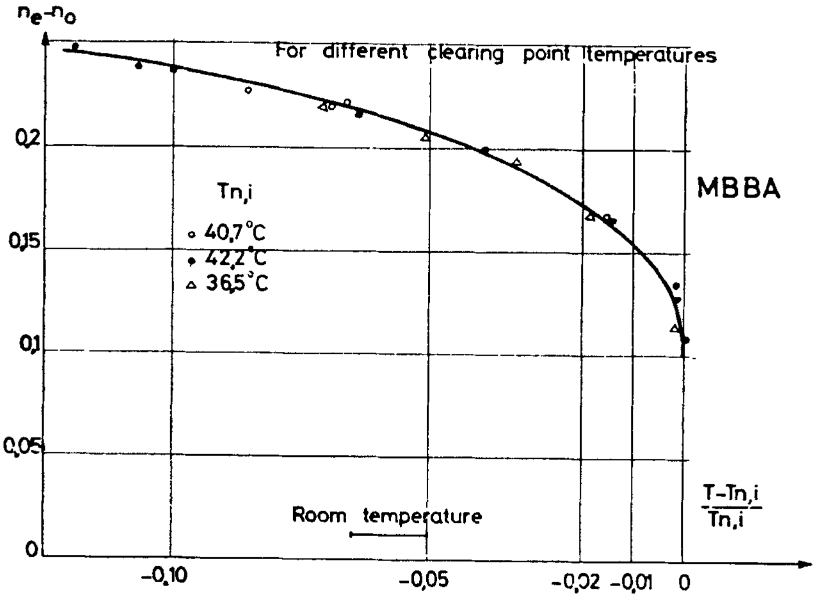

Figure 6. Birefringence versus reduced temperature.

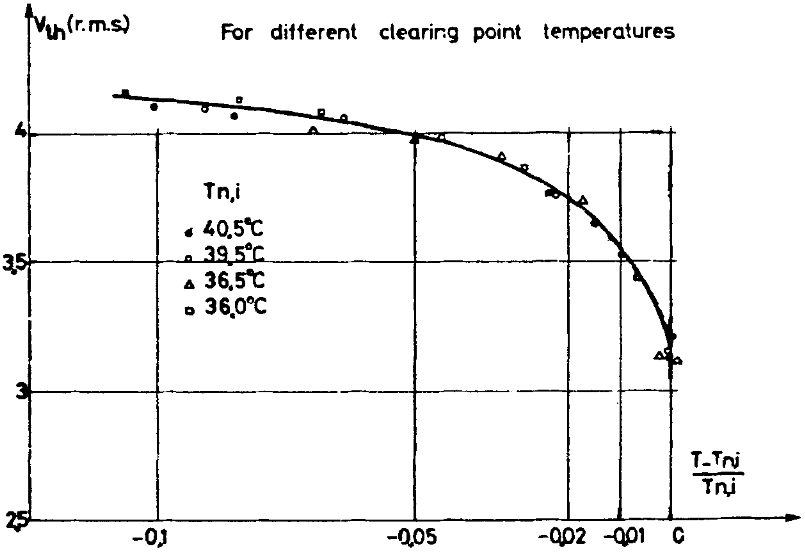

Figure 7. Threshold voltage versus reduced temperature.

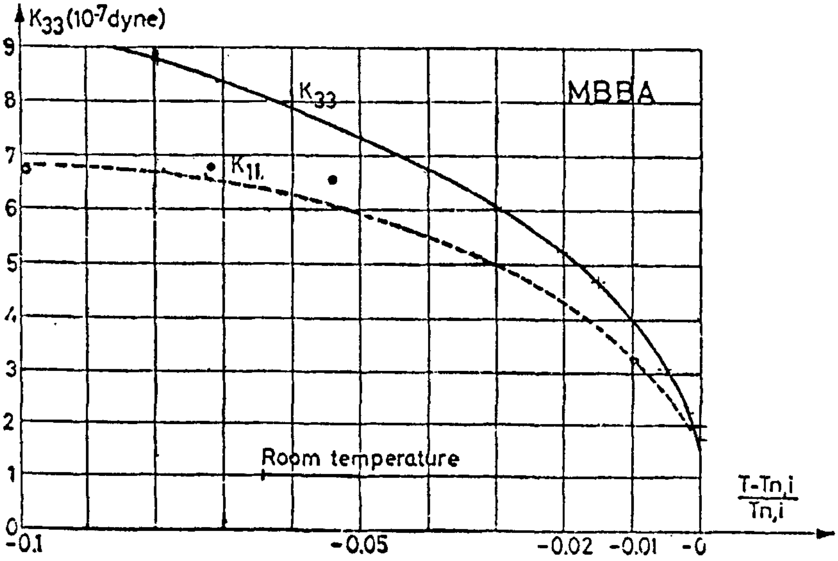

Figure 8. Elastic constants K_{11} and K_{33} versus reduced temperature.

In order to determine for each temperature K_{11} and K_{33}, we have to measure birefringence $n_e - n_0$ and to draw the curve $\langle n \rangle$ as a function of applied voltage V. K_{33} is deduced from threshold voltage and K_{11} from the slope of the curve $\langle n \rangle$ versus V in the vicinity of threshold voltage. The accuracy on K_{33} parameters is dependent on accuracy on threshold voltage. This last one may be reached with an accuracy better than 5%. For K_{11}, accuracy is approximately 10%.

$(T - T_{\mathrm{N.I}})/T_{\mathrm{N.I}}$ is the chosen reduced temperature parameter where $T_{\mathrm{N.I}}$ is the absolute temperature of nematic–isotropic transition. In agreement with Haller, the curves as a function of the previous reduced temperature are independent of transition temperature $T_{\mathrm{N.I}}$. So, the experimental dots are plotted for different transition temperatures and the birefringence, the threshold voltage, K_{11} and K_{33} are given on Figs. 6, 7 and 8 versus previously defined reduced temperature.

Dynamic Behaviour

The input voltage applied to the cell is a high frequency modulated pulsed voltage. So, the mean value of applied voltage is zero and the migration of disturbing ions does not occur.

Theoretical computations prove that, for small values of birefringence, the variation $\langle n \rangle(t)$ is an exponential law characterized by a time constant. This time constant can be computed using the experimental rise time or decay time. When the applied voltage is much larger than threshold voltage, rise time τ_r is connected to viscosity γ_1 by the relation:

$$\frac{1}{\tau_r} = \frac{1}{\gamma_1 L^2} \cdot \frac{\epsilon_a V^2}{4\pi}$$

The decay time τ_d is given by:

$$\frac{1}{\tau_d} = \frac{\pi^2 K_{33}}{\gamma_1 L^2}$$

For each temperature, the curves giving the mean value of birefringence versus time for different applied voltages lead to the

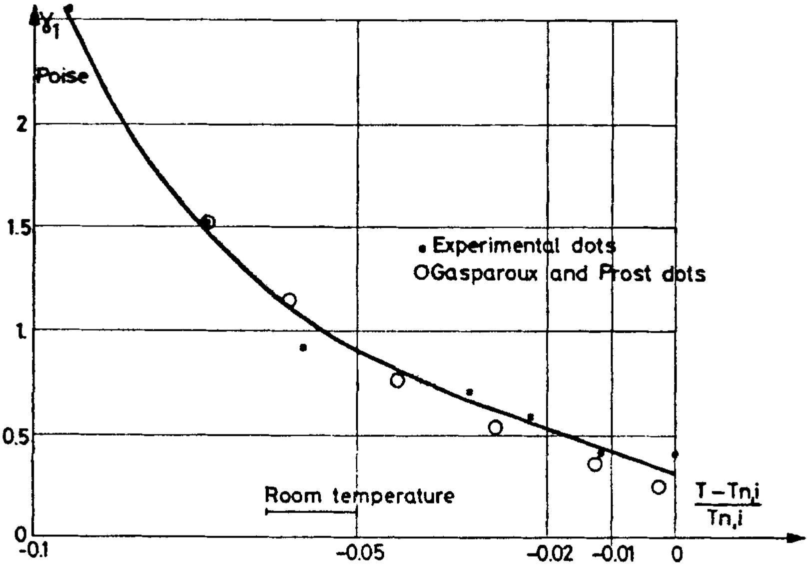

Figure 9. Viscosity coefficient compared with Gasparoux and Prost results versus reduced temperature.

determination of one viscosity coefficient γ_1; the reduced temperature dependence of γ_1 is given on Fig. 9.

5. Concluding Remarks

The accuracy of the determination of γ_1 is connected with the measurement of L and this value has not been measured with a good accuracy. But the relative variations of γ_1 versus reduced temperature are obtained very accurately. The comparison with the values given by Gasparoux and Prost(5) from susceptibility measurements in rotating magnetic field is in fair agreement.

Optical effect during transient time does not present any scattering of light; so, it seems that no noticeable fluid movement occurs and that the " back flow " effect is negligible. This effect has been neglected in this paper.

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