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Nonlinear Optical Response of Nematic Liquid Crystal on Varying Pressure Difference in the Presence of Electric Field

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We are presenting the new results of an optical investigation of an initially homeotropic liquid crystal layer under action of sinusoidal pressure difference. The experiments were carried out by using the striped liquid crystal cell and the nematic mixture with a positive dielectric permittivity anisotropy. The non-linear optical response in the presence of stabilizing electric field (U) was studied and analysed.

Keywords: pressure difference; electric field; Poiseuille flow

INTRODUCTION

It is well known, that nematic liquid crystals (NLC) show Non-Newtonian behavior in shear flows due to a connection between a velocity gradient and an orientation described by a director. This leads to a number of peculiarities which don't take place in isotropic liquid^[1]. In spite of numerous works devoted to the stationary flows of different types only a few researches dealt with oscillating flows^[2, 3] especially with unsteady Poiseuille flows^[4, 5]. The latter differ sufficiently from the stationary ones. In particular, the flow induced orientation becomes unstable even at homeotropic boundary conditions under the threshold amplitude of vibrations, which depends on a

frequency^[4]. Recently^[4, 6] a rather full description of behavior of nematics under an oscillating Poiseuille flow was fulfilled and compared with a rare experimental results, obtained at sound frequency^[5].

In this paper we present the experimental results on a behavior of initially homeotropic layer of nematic liquid crystal distorted by a dynamic pressure gradient under an action of stabilizing electric field. The experiments were carried out using the stripped liquid crystal cell, described earlier^[7]. Different values of electric voltage can be applied to the separate stripes and so to create a stepped inhomogeneity in the direction of the flow (fig.1.).

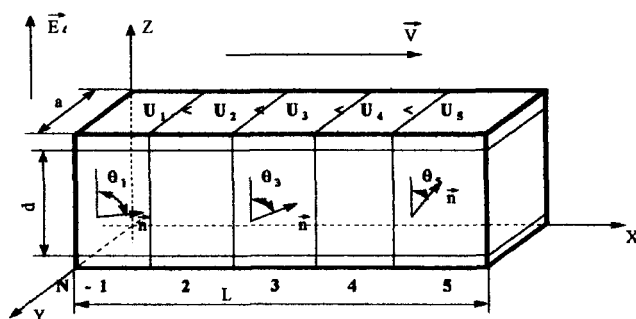


FIGURE 1 Geometry of experiment.

The pressure difference varying with time by the law:

$$\Delta p = \Delta p_m \cos \omega t, \quad \omega = 2\pi/T \quad (1)$$

was applied to the open sides of sandwich like cell.

The alternating pressure difference induces a periodical motion of the anisotropic liquid in a rectangular channel $[L \times a \times d = 35 \times 15 \times 0.115 \text{ mm}]$. The flow induced motion of the director was registered by polarized laser beam ($\lambda = 0.63 \mu\text{m}$) passing, normally to the surfaces of

the cell and the analyzer (the angle (α) between the vector \vec{E}_ℓ of the light wave and the flow direction is equal to $\pi/4$). So the expressions for a phase difference (Φ) between ordinary (o) and extraordinary (e) rays and for the light intensity can be written as follows:

$$\Phi = \frac{2\pi}{\lambda} \int_{-d/2}^{d/2} \Delta n dz \quad (2)$$

$$I = I_0 \sin^2 \frac{\Phi}{2} \quad (3)$$

$$\text{there } \Delta n = n(\theta) - n_o \quad (4)$$

$$n(\theta) = n_o \left[1 - \frac{n_e^2 - n_o^2}{n_e^2} \cos^2 \theta \right]^{-1/2} \quad (5)$$

n_o and n_e - refractive indexes of ordinary and extraordinary rays, I_0 - the input light intensity, θ - the angle of an orientation.

Some experimental dependencies $I(t)$ at a relatively low pressure differences amplitude and different electric voltages (U) applied to the central stripe of the cell are shown in figure 2 (the voltages on the other stripes are proportional to the number of the stripe N). One can see a sufficiently non-linear optical response in the absence of an electric field. The stabilizing electric field effectively suppresses the non-linearities of the dependencies $I(t)$. At high enough voltages time dependencies $I(t)$ are rather simple and the electric field decreases the amplitude of an optical response (fig. 2). Using the changes of an optical intensity one can restore the dependence $\Phi(t)$ according to the expression (3). The obtain results are shown in fig.2. It is obvious that the time dependencies $\Phi(t)$ can be expressed by relatively simple laws at variations of electric voltages in a wide range. In particular the frequency of changing of $\Phi(t)$ is twice then that of pressure and it's amplitude decreases monotonically on increasing of electric voltage. It makes possible to describe the results mentioned above with the help of linearized hydrodynamic equations. For this experiment geometry they can be expressed as ^[7]:

$$\rho \frac{\partial V_X}{\partial t} = -\frac{\partial p}{\partial X} + \eta_2 \frac{\partial^2 V_X}{\partial Z^2} + \alpha_2 \frac{\partial^2 \theta}{\partial Z \partial t} \quad (6)$$

$$k_{33} \frac{\partial^2 \theta}{\partial Z^2} = \alpha_2 \frac{\partial V_X}{\partial Z} + \gamma_1 \frac{\partial \theta}{\partial t} + \epsilon_0 \Delta \epsilon E^2 \theta \quad (7)$$

where: $\eta_2 = \frac{1}{2}(\alpha_4 + \alpha_5 - \alpha_2)$ - the shear viscosity coefficient, ρ - the density, V_X - the flow velocity, E - the electric field strength, α_i - the Leslie's coefficients, in particular α_2 - the Leslie's coefficient, which provides a coupling between the velocity [$V_X(Z)$] and the orientation $\theta(Z)$, γ_1 - the rotational viscosity coefficient, k_{33} - the Frank's module.

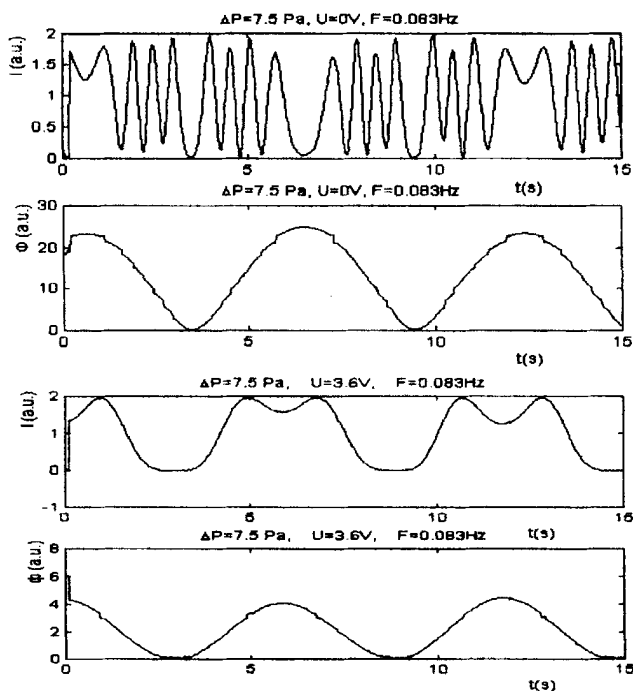


Figure 2 The time dependencies of the intensity I and the phase difference Φ at different voltages.

The first inertial term in equation (6) can be neglected up to frequencies about $\sim 1\text{kHz}$ ^[51] (for the parameters of our experiment). At this assumption the solution of the system for strong boundary anchoring can be written as follows :

$$\theta(\tilde{z}, \tilde{t}) = \theta_r(\tilde{z}) \cos \tilde{t} + \theta_i(\tilde{z}) \sin \tilde{t} \quad (8)$$

where $\tilde{t} = \omega t$ and $\tilde{z} = \frac{z}{d}$ - dimensionless parameters ;

$$\theta_r(\tilde{z}) = -\frac{\Delta p_m}{\Delta X} \frac{d}{\eta_2 \omega \left[\left(\frac{\omega_E}{\omega} \right)^2 + m^2 \right]} \cdot \left\{ \left(\frac{\omega_E}{\omega} \right) \left[\tilde{z} + \frac{1}{2} \right] \cdot \right. \\ \left. \cdot \frac{\cos k_2 \left(\tilde{z} + \frac{1}{2} \right) \cosh k_1 \left(\tilde{z} - \frac{1}{2} \right) - \cos k_2 \left(\tilde{z} - \frac{1}{2} \right) \cosh k_1 \left(\tilde{z} + \frac{1}{2} \right)}{(\cosh k_1 - \cos k_2)} \right\} - m \cdot \quad (9)$$

$$\left[\frac{1}{2} \frac{\sin k_2 \left(\tilde{z} - \frac{1}{2} \right) \sinh k_1 \left(\tilde{z} + \frac{1}{2} \right) - \sin k_2 \left(\tilde{z} + \frac{1}{2} \right) \sinh k_1 \left(\tilde{z} - \frac{1}{2} \right)}{(\cosh k_1 - \cos k_2)} \right]$$

$$\theta_i(\tilde{z}) = -\frac{\Delta p_m}{\Delta X} \frac{d}{\eta_2 \omega \left[\left(\frac{\omega_E}{\omega} \right)^2 + m^2 \right]} \cdot \left\{ \left(\frac{\omega_E}{\omega} \right) \left[\frac{1}{2} \right] \cdot \right. \\ \left. \cdot \frac{\sin k_2 \left(\tilde{z} - \frac{1}{2} \right) \sinh k_1 \left(\tilde{z} + \frac{1}{2} \right) - \sin k_2 \left(\tilde{z} + \frac{1}{2} \right) \sinh k_1 \left(\tilde{z} - \frac{1}{2} \right)}{(\cosh k_1 - \cos k_2)} \right\} + m \cdot \quad (10)$$

$$\left[\tilde{z} + \frac{1}{2} \frac{\cos k_2 \left(\tilde{z} + \frac{1}{2} \right) \cosh k_1 \left(\tilde{z} - \frac{1}{2} \right) - \cos k_2 \left(\tilde{z} - \frac{1}{2} \right) \cosh k_1 \left(\tilde{z} + \frac{1}{2} \right)}{(\cosh k_1 - \cos k_2)} \right]$$

$$k_1 = \sqrt{\frac{1}{2} \left[\frac{\omega_E}{\omega_0} + \sqrt{\left(\frac{\omega_E}{\omega_0} \right)^2 + m^2 \left(\frac{\omega}{\omega_0} \right)^2} \right]} \quad (11)$$

$$k_2 = \frac{\frac{1}{2} m \left(\frac{\omega}{\omega_0} \right)}{\sqrt{\frac{1}{2} \left[\frac{\omega_E}{\omega_0} + \sqrt{\left(\frac{\omega_E}{\omega_0} \right)^2 + m^2 \left(\frac{\omega}{\omega_0} \right)^2} \right]}} \quad (12)$$

$$m = 1 - \frac{(-\alpha_2)}{\eta_2} \cdot \frac{1}{1-\lambda}; \quad \lambda = \frac{\alpha_3}{\alpha_2};$$

The degree of influence of an electric field on the solutions obtained above depends on relative values of three frequencies: ω - the frequency of external force, $\omega_0 = \frac{k_{33}}{d^2 \gamma_1}$ - the frequency of orientational relaxation in the absence of the electric field and $\omega_E = \frac{\epsilon_0 \Delta \epsilon E^2}{\gamma_1}$ - the analogous frequency in the presence of the electric field.

In the case of very low electric voltages ($\omega_E \ll \omega_0$) two wave vectors k_1 and k_2 , which describe the spatial changes of the orientation, become equal to each other:

$$k_1 \approx k_2 \approx k \approx \sqrt{\frac{1}{2} m \frac{\omega}{\omega_0}}$$

This result is the same as obtained earlier^[6] in the absence of the electric field and means that the thickness of boundary layers ($\delta = k^{-1}$) decreases with increasing of ω .

On the other hand in the limit of strong fields ($\omega_E \gg m\omega$) the form of a spatial inhomogeneity of the orientation and the thickness of

layers boundary don't depend on frequency of vibrations ($k_2 \cong 0$,

$$k_1 \approx \sqrt{\frac{\omega_E}{\omega_0}} \approx \left(\frac{U}{U_\Phi} \right)^2, \quad \text{where} \quad U_\Phi = \sqrt{\frac{k_{33}}{\epsilon_0 \Delta \epsilon}} \cdot \pi).$$

As far as the amplitude of orientational oscillations is concerned it decreases with increasing the frequency (ω) and strength (E) of the electric field.

It is clear, that an electric field's application, effectively decreases a value of $\theta_i(\tilde{Z})$ and so the phase shift between the pressure oscillations and of the director. It is of interest that the experimental dependencies of Φ on Δp obtained at different voltages are in an agreement with the linear theory predictions (fig. 3), at least in the range $0 < \Phi < 10$ rad. The data, presented in this figure, are obtained from dynamic, response of a nematic accounting the phase shift between $\Phi(t)$ and $\Delta p(t)$. The difference between the experimental results and the theoretical law ($\Phi \sim (\Delta p^2)$) at large values of Φ can be due to different reasons. The main one is a possible influence of a nonlinear behavior of an orientational structure which takes place at high pressure difference and weak electric fields.

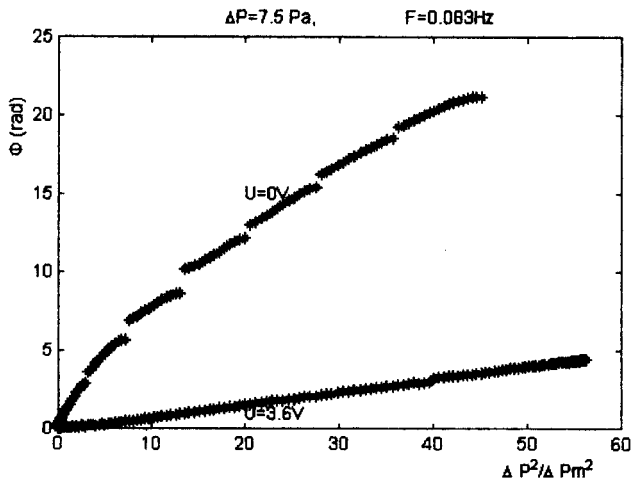


FIGURE 3 The dependence of Φ on the pressure difference Δp

For the striped cell used by us it can lead to the difference between the real dependencies $G(t)$ applied to different stripes and the harmonic law (1) due to the influence of the effective viscosity on the orientation of a liquid crystal.

Some interesting experimental results on a non-linear behavior of an orientation under Poiseuille flow will be presented later.

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