Application of the flexoelectric effect in liquid crystals to create acousto-optic transducers

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Abstract. The flexoelectric effect in nematic liquid crystals was studied using acoustic modulation spectroscopy. A cell consisting of three plates separated by spacers was used. The movable plate made shear oscillations in the plane of the cell. We studied thin layers of liquid crystals with homeotropic orientation of molecules with a thickness of from 20 to 100 microns. The frequency of exposure is 1 kHz. In the framework of the de Gennes model, near the phase transition liquid crystal - isotropic phase, the resulting signal has a flexoelectric nature and is due to a dipole mechanism. In this work, the temperature dependences of the flexosignal, the temperature effect on the value of the average constant angle of inclination \(\theta_c\) and the amplitude of the oscillations of the director \(\theta_d\) of a liquid crystal are investigated. The dependences of flex coefficients \(e_{ij}\) on temperature are obtained. It was found that dipole and quadrupole mechanisms contribute to the observed molecular orientational polarization. The results of this study are proposed to be used to create liquid crystal transparencies of the new generation, as well as acousto-optic transducers for automated vibration control systems, since the use of liquid crystals provides a number of advantages (lightweight construction, low power consumption and overall dimensions, low cost).

1. Introduction
Molecular order in liquid crystals (LC) leads to anisotropy of its physical properties, which determine the high sensitivity of such systems to temperature and impurity concentrations, as well as to the action of electric and magnetic fields, elastic stresses and viscous flow. Most of the effects observed in LCs associated with the features of their structural transformations have no analogues in solids and in isotropic liquids. These effects include the piezoelectric effect. By definition, the direct piezoelectric effect in solid crystals is the occurrence of macroscopic volume polarization under the action of mechanical stresses or deformations. In liquid crystals, most of which have a center of symmetry, such a mechanism is impossible. However, as Mayer [1] showed, a special piezoelectric effect is realized in a nematic medium, which is called flexoelectric (flexo-bending) [2]. It consists in the appearance of electric polarization as a result of the deformation of a bend or twisting of the director orientation lines in space due to the interaction of dipole molecules having an anisotropic geometric shape.

The relevance of studying the flexoelectric effect in liquid crystals [3, 4] is associated with the prospects of practical application in engineering, for example, to create displays of a new generation. The use of liquid crystals means economic efficiency, simplicity, convenience, small dimensions of devices and low power consumption. The article studies the flexoelectric effect in the vicinity of the transition of a nematic liquid crystal - an isotropic liquid.
2. Experimental technique
For an experimental study of the flexoelectric effect, an installation was designed and assembled, a block diagram of which is shown in figure 1.

Figure 1. Installation for the study of the flexoelectric effect: 1 - source of sound oscillations, 2 - stage, 3 - polarization microscope with photometric attachment, 4 - differential thermocouple, 5 - microvoltmeter of direct current, 6 - millivoltmeter of direct current, 7 - selective amplifier, 8 - analog-to-digital converter, 9 - computer.

The transmitted or reflected light wave from the cell is recorded by a spectrophotometric attachment, and the constant component of the light flux is then recorded by a millivoltmeter of direct current. The variable components of the harmonics were measured by a selective amplifier, then the signal was fed to an analog-to-digital converter, then to a computer. The frequency of exposure was in the frequency range from 20 Hz to 20 kHz [3, 4]. The cell with the LC was placed on the object table of the microscope. In our case, we studied the effect of shear oscillations on the liquid crystal film. To create an acoustic contact, the source of vibrations was connected to the cell by a waveguide. An experimental study of the flexoelectric effect was carried out on a sandwich cell. The cell is assembled from three transparent glass plates with conductive coatings of chromium, two of them were made of a glass slide (20×30×2 mm), the movable middle plate was made of a cover glass with a size of 24×24 mm and a thickness of 100 μm. The cell was a capacitor with some part of the free space in which the LC was located. Nematic LC films with a thickness of \( h = 20-100 \) μm were considered in the work: n-methoxybenzilydenene-n-butylaniline (MBBA); heptylbenzoic acid cyanophenyl ester (HACE); nitrophenyloctyloxybenzoate (NPOOB), 4-octyl-4-cyanobiphenyl (OCB), butetil heptanooyloazoxybenzene (BHAOB); a mixture of LC on the basis of two ringed ether (TRE).

Consider the effects of polarization that occur in the isotropic phase of a nematic liquid crystal (NLC) when subjected to vibrational oscillations of a plate. A viscous wave arises in the sample, which quickly fades out. The attenuation \( \delta \) is determined by the viscosity \( \eta \), the density \( \rho \) of the liquid and the oscillation frequency of the plate \( \omega \): \( \delta \sim \left[ \frac{\eta}{\rho \omega} \right]^{1/2} \) [5]. In NLC, such vibration leads to the appearance of an alternating electrical signal, as a result of the occurrence of the flexoelectric effect [6]. Since in the isotropic phase, the flux induces the appearance of the order parameter, the flexoelectric polarization should occur. At a large absolute value of the vibrational velocity \( v \), the propagating elastic-viscous
wave begins to influence the orientation and the order parameter $S$ [7], which is similar to the Maxwell effect [8] - the orientation of anisotropic molecules in the flow.

De Genne proposed a model for describing pre-transition phenomena in a stream based on the Landau theory of phase transitions, for example, the appearance of birefringence in the isotropic phase [2]. The observed signal behind the phase transition has a flexoelectric nature and is due to the dipole mechanism. According to [9], the contribution of the flexoelectric mechanism to the free energy of a crystal $\delta F$ can be written as follows:

$$\delta F \sim e_{10}E_{a}S_{a,\beta} \frac{\partial S_{\beta m}}{\partial x_{m}} + e_{30}E_{a}S_{m,\beta} \frac{\partial S_{\beta a}}{\partial x_{a}},$$

where $S_{a,\beta}$ is the order parameter tensor, $e_{10}$ are the proportionality coefficients, which are weakly dependent on temperature. Where flexography takes the form: $P_{a} = e_{10}E_{a}S_{a,\beta} \frac{\partial S_{\beta m}}{\partial x_{m}} + e_{30}S_{m,\beta} \frac{\partial S_{\beta a}}{\partial x_{a}}$.

The dependence of $P_{a}$ on temperature and velocity gradient is determined from the equations of isotropic nematodynamics. The general system of equations [9]:

$$\begin{bmatrix}
I_{a,\beta} \\
\varphi_{a,\beta}
\end{bmatrix} = \begin{bmatrix}
\eta \\
\mu
\end{bmatrix} \times \begin{bmatrix}
d_{a,\beta} \\
R_{a,\beta}
\end{bmatrix},$$

where $I_{a,\beta}, \varphi_{a,\beta}$ are the generalized forces, $d_{a,\beta}, R_{a,\beta}$ are the flows, $I_{a,\beta}$ is the viscous stress tensor. Since $\varphi_{a,\beta} \approx -\delta F / \delta S_{a,\beta} = -A(T)S_{a,\beta}$, $d_{a,\beta} = \frac{1}{2} \left( \frac{\partial \varphi_{a,\beta}}{\partial \beta} + \frac{\partial \varphi_{\beta,\alpha}}{\partial \alpha} \right)$, $R_{a,\beta} = \frac{\partial S_{a,\beta}}{\partial t}$, the equation of isotropic nematodynamics transforms to the form (the speed $v_{z}$ lies in the plane of the oscillating plate, $OZ$ is perpendicular to $v_{z}$ and collinear to the normal vector to the surface):

$$-A(T)S_{zc} = \mu \frac{\partial \varphi_{z}}{\partial \zeta} + \nu S_{\varphi z}, \quad \rho \frac{\partial \varphi_{z}}{\partial t} = \eta \frac{\partial \varphi_{z}}{\partial \zeta} + 2\mu - \frac{\partial S_{\varphi z}}{\partial \zeta},$$

from here

$$S_{zc} \left[ A(T) + i\nu \omega \right] = \mu \frac{\partial S_{z}}{\partial \zeta} = \mu G, \quad A(T) = A_{0}(T - T_{N}^{*}),$$

$$S_{\varphi z} = \frac{\mu Ge^{i\phi}}{\sqrt{A^{2}(T - T_{N}^{*})^{2} + \nu^{2} \omega^{2}}}, \quad \tan \varphi = \frac{\nu \omega}{A(T - T_{N}^{*})},$$

or considering that $\omega \sim \exp(-\lambda z)$, $\lambda \sim \rho \omega \eta^{1/2}$, and setting $A(T) >> \nu \omega$, we have

$$P_{x} = e_{10}S_{xc} \frac{\partial S_{zx}}{\partial \xi} + e_{30}S_{x} \frac{\partial S_{zx}}{\partial \xi} \approx 0, \quad \text{t.k.} \frac{\partial S_{zx}}{\partial \xi} = 0,$$

$$P_{z} = e_{10}S_{zx} \frac{\partial S_{zx}}{\partial \xi} + e_{30}S_{z} \frac{\partial S_{zx}}{\partial \xi} \approx e_{10}(T - T_{N}^{*})^{-2} \lambda^{3} S_{o}^{2}.$$  \hspace{1cm} (1)

Far from the phase transition, when the parameter of the nematic order $S$ is significantly greater than the induced additive $\Delta S \approx \frac{\mu}{2a(T - T^{*})} \frac{\partial \varphi_{z}}{\partial \zeta}$ [7].

3. Results and its discussion

The dependence of the first harmonic of the signal $U_{10}$ for n-methoxybenzylidene-n-butylaniline (MBBA) is presented in figure 2. As the $T_{N}$ phase transition to an isotropic state approaches the temperature, a significant change in the flexo signal behavior is detected. At the phase transition point, the signal size does not abruptly drop to zero, which is typical of the pre-transition behavior of most NLC parameters, but decreases according to a power law from the type temperature $(T - T^{*})^{-n}$, where $T^{*}$ is the phase transition temperature. Analysis of the temperature change after the phase transition
shows that \( n \approx 2 \). A similar relationship \( \omega \sim U \) occurs in the case of the study of a mixture of two ringed ether (TRE). The effects observed in MBBA and TRE have typical features and are characteristic of other compounds with NLC-phase. It should be noted that the phase-transition at \( T > T_c \) shows the dependence of the flexoelectric signal, predicted by the theory of De Gennes, and the magnitude of the flexoelectric polarization: \( P \sim (T-T_c)^2 \). The ratio of the longitudinal and transverse components of the polarization \( P_z/P_x \) is 1.2 and 1, respectively, for MBBA and TRE, and is in good agreement with the values of dielectric constant.

Figure 2. Temperature dependences of the first harmonic \( U_{1\omega} \) (solid lines) and the square root of the reciprocal of the signal (dotted lines).

Figure 3. Temperature dependences of the angles of deviation \( \bar{\theta}_d \) and constant tilt of the director \( \bar{\theta}_c \).

According to figure 2, the behavior of the magnitude of polarization in the isotropic phase is typical for any NLC and is described by formula (1). However, in the NLC phase, the behavior of the flexoelectric polarization depends on the amplitude of the shift. For example, in MBBA, at small shear amplitudes, the classical behavior described by formula (1) takes place. When \( T > T_c \), the polarization is proportional to \( (T-T_c)^2 \), and in the vicinity of the phase transition undergoes a jump, is determined by the temperature dependence of the order parameter far from \( T_c \). In this situation, only the polarization component along the \( Z \) axis is nonzero. At large shear amplitudes, the stationary director slope appears, which depends on the amplitude and temperature. This will result in the appearance of the second component of polarization \( P_x \), which is also preserved in the isotropic phase. The dependences of the values of the stationary angle of deviation on temperature for the three-component mixture of TKE are illustrated in figure. 3. In figure 3, two opposite situations are also shown. In one case for MBBA, the effect of flexoelectric polarization determines the dependence of the order parameter on temperature, when the director’s stationary orientation determines the behavior of the magnitude of the induced polarization in the vicinity of the NLC phase transition — an isotropic liquid. In the second situation, the elastic effects on which the director deviation is imposed are significant. Moreover, the average director angle under the action of a shear wave is defined as \(< \theta > \sim \nu \delta / K_{33} \), where \( K_{33} \sim S^2 \), and the depth of penetration of a viscous wave is \( \delta \sim (T_c-T)^{1/2} \). However, in the isotropic phase, the temperature behavior of the induced polarization does not depend on elasticity and is the same for both types of compounds. Such a change in the behavior of the induced flexoelectric polarization with the appearance of a stationary director slope in the flow indirectly indicates a change in the type of phase transition. For more convincing evidence of such a statement, it is necessary to study the thermodynamic characteristics of a NLC when a transverse viscous wave of high intensity appears in it. Figure 2 shows the dependences of the root of the reciprocal of the shift-induced signal on temperature \( (T-T_n^*) \), confirming the assumption made earlier about the flexoelectric nature of the potential difference.
observed during the phase transition. Knowing the temperature changes of the quantities in the formula for $U_{10} \approx e_{33} c \theta (\lambda h^2) \exp(i \omega t)$, we obtain the dependences of the flexographic coefficients $e_{33}$ for the substances under study (figure 4).

$$e_{33} \approx 10^5 \text{SGSE/cm}$$

Figure 4. Temperature dependences of the flexoelectric coefficients $e_{33}$.

It can be noted that the behavior of the temperature dependence of the LC flexoelectric coefficients in the nematic phase $e_{33}(T)$ is described by the general dependence on the order parameter $S$: $e \sim \alpha S + \beta S^2$. All values of weight coefficients differ significantly in different NLCs, but they are consistent with the data obtained for the $e_{11}$ modules by the method of bending vibrations (table 1).

Table 1. Values of weighting coefficients for liquid crystals.

| Liquid crystals | $\alpha$ | $\beta$ |
|-----------------|---------|--------|
| MBBA           | -0.1   | -0.9   |
| BHAOB          | -0.3   | -0.7   |
| HACE           | -1.0   | -      |
| NPOOB          | -1.0   | -      |
| OCB            | -1.0   | -      |

For example, in MBBA $\alpha \approx 0.1$ and $\beta \approx 0.9$, which indicates the dipole mechanism of flexoelectric polarization. For the HACE $\alpha \approx 1$ and $\beta \approx 0$, this indicates the quadrupole character of the flexoelectric coefficients of nematics of this class.

4. Conclusion

The article investigated the temperature dependences of the flexosignal induced by shear oscillations, studied the effect of temperature on the average constant angle of inclination $\theta_c$ and the amplitude of the director oscillations $\vec{d}_d$. The temperature dependences of flexoelectric coefficients $e_{33}$ were obtained, which clearly demonstrate the role of the dipole and quadrupole mechanisms in the formation of the bulk orientational polarization of the molecules of the substances under study. The calculation results are consistent with the data obtained for flexoelectric coefficients $e_{11}$ by the method of bending vibrations. The results can be used to develop a new generation of liquid crystal displays, as well as to create acousto-optic transducers for automated vibration control in various industries.

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