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THRESHOLD FLEXOELECTRIC EFFECT IN NEMATIC LIQUID CRYSTAL

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Résumé. — Une résolution complète du problème d’instabilité flexoélectrique est donnée dans le cas d’un échantillon d’épaisseur finie et sans approximations sur les constantes élastiques.

Abstract. — Total solution of flexoelectric instability problem is given for finite thickness sample and real elastic moduli.

There is a linear coupling between electric polarization and orientational deformation in nematic liquid crystal [1]. The appropriate term in the free energy of liquid crystal under the influence of electric field \( \mathbf{E} \) can be written in accordance with symmetry properties as

\[
\delta F = - \int \left[ \varepsilon_1 (\mathbf{E} \cdot \mathbf{n}) \text{div} \mathbf{n} + \varepsilon_2 \mathbf{E} \cdot (\mathbf{n} \times \mathbf{n}) \right] d^3 \mathbf{r},
\]

where \( \mathbf{n}(\mathbf{r}) \) is director, \( \varepsilon_1 \) and \( \varepsilon_2 \) are flexoelectric moduli. This coupling results in appearance peculiar flexoelectric effect which is a periodic distortion of initial planar orientation. Meier [1] had shown that the infinite liquid crystal must be disturbed and the perturbation must be periodic along the director orientation in the absence of electric field. This deformation is not the threshold one and its period is inversely proportional to electric field strength.

It was shown in the paper [2] that another type of flexoelectric instability is possible in nematic liquid crystal layers of finite thickness with a homogeneous orientation on the walls. New instability arising at certain threshold voltage is characterized by small director deviations from \( X \) axis in two planes: on angle \( \theta \) in \( XZ \) plane and on angle \( \varphi \) in \( XY \) plane (see Fig. 1). The instability results in arising of the periodic along \( Y \) axis domain structure which is parallel to \( X \) axis. In the case of small deformations \( |\varphi|, |\theta| \ll 1, n_x = 1, n_y = \varphi, n_z = \theta, E_x = E_y = 0, E_z = E \) and equal elastic moduli \( K_{11} = K_{22} = K \) the solution corresponding to the boundary conditions \( \theta = \varphi = 0 \) at \( z = \pm d/2 \) takes the form:

\[
\begin{align*}
\theta &= \theta_0 \cos(qy) \cos(nz/d), \\
\varphi &= \varphi_0 \sin(qy) \cos(nz/d).
\end{align*}
\]

The instability appears at the threshold voltage \( U_o \) and the threshold wave vector \( q_o \):

\[
U_o = \frac{2 \pi K}{|e^*| (1 + \mu)}, \quad q_o = \frac{\pi}{d} \sqrt{\frac{1 - \mu}{1 + \mu}}
\]

where

\[
\mu = (\varepsilon_a K/4 \pi e^*^2), \quad e^* = e_1 - e_2,
\]

\( \varepsilon_a \) is dielectric anisotropy, \( d \) is the layer thickness. Similar flexoelectric instability can exist under the influence of constant as well as alternating electric field. Frequency dependence of the threshold is analysed in reference [2].

Such type of instability seems to be observed for the first time by Vistin [3]. Perhaps this instability can be observed in alternating field at temperatures when the Williams domains do not exist because of negative sign of conductivity anisotropy [4]. Arising of this flexoelectric instability in constant field and the dependence of threshold voltage and wave vector upon dielectric anisotropy were investigated in reference [5] in details using \( n \)-butyl-\( n' \)-methoxyazoxybenzol doped by 2-3-dicyano-4-aminoxyphenyl ester of amlyoxybenzoic acid or \( n' \)-cyanophenyl ester of n-heptylbenzoic acid for changing of dielectric anisotropy. The experimental data are in good agreement with the theory and allow one to define \( K \) and \( e^* \) for BMAOB taking

\[
\begin{align*}
\varepsilon_a < 0 & \quad (K = 6.5 \times 10^{-7} \text{ dyne}), \\
e^* &= 1.7 \times 10^{-4} \text{ CGS units}.
\end{align*}
\]
The equations (3) were received for equal moduli $K_{jj}$ so only average values of $K_{jj}$ and $e^*$ were determined in reference [5].

The purpose of the present paper is the investigation of threshold characteristics of flexoeffect using real values of elastic moduli. In the case of small deformations $|\varphi|, |\theta| \ll 1$ the free energy of nematic can be written as

$$F = \frac{1}{2} \int \left\{ K_{11} \left( \frac{\partial \varphi}{\partial y} + \frac{\partial \theta}{\partial z} \right)^2 + K_{22} \left( \frac{\partial \theta}{\partial y} - \frac{\partial \varphi}{\partial z} \right)^2 - \frac{e_a E^2}{4 \pi} \theta^2 - 2 e_1 E \left( \frac{\partial \varphi}{\partial y} + \frac{\partial \theta}{\partial z} \right) \varphi - 2 e_2 E \left( \frac{\partial \theta}{\partial y} - \frac{\partial \varphi}{\partial z} \right) \varphi \right\} d^2 r.$$  

(4)

Minimizing the free energy $F$ with respect to $\varphi$ and $\theta$ one can receive the following equations for stationary orientation of director

$$K_{11} \frac{\partial^2 \varphi}{\partial y^2} + K_{22} \frac{\partial^2 \varphi}{\partial z^2} - e^* E \frac{\partial \varphi}{\partial y} + (K_{11} - K_{22}) \frac{\partial^2 \theta}{\partial y \partial z} = 0,$$

$$K_{22} \frac{\partial^2 \theta}{\partial y^2} + K_{11} \frac{\partial^2 \theta}{\partial z^2} + e^* E \frac{\partial \theta}{\partial y} + (K_{11} - K_{22}) \frac{\partial^2 \varphi}{\partial y \partial z} + \frac{e_a E^2}{4 \pi} \theta = 0.$$  

(5)

Let us take the solution satisfying to boundary conditions $\varphi = \varphi_0 \sin (qy) \exp (ipz)$, $\theta = \theta_0 \cos (qy) \exp (ipz)$.

(6)

Inserting (6) into (5) and taking into account the condition of nontriviality of $\theta_0$ and $\varphi_0$ values one can write the following equation for characteristic numbers $\rho$ :

$$s^4 + \left( 2 - \mu \xi^2 \right) s^2 + \left[ 1 - \frac{\xi^2}{\lambda} (1 + \mu) \right] = 0$$  

(7)

where $s = p/q$, $\mu = (e_a K_{11}/4 \pi e^2)$, $\lambda = K_{22}/K_{11}$, $\xi = e^* E/K_{11} q$.

Equation (7) has 4 roots : $\pm s_1, \pm s_2$. So one get the complete solution of equation (5) in the form :

$$\varphi = \sin (qy) \sum_{i=1}^{2} \left( a_i \cos (qs_i z) + b_i \sin (qs_i z) \right),$$

$$\theta = \cos (qy) \sum_{i=1}^{2} \left[ a_i (\beta_i \cos (qs_i z) - \gamma_i \sin (qs_i z)) + b_i (\beta_i \sin (qs_i z) + \gamma_i \cos (qs_i z)) \right],$$

where $\lambda = (qd/2)$. Equation (9) is the dispersion equation connecting field strength $E$ with wave number $q$. The dependence $E = E(q)$ was calculated numerically for different values of $\mu$ and $K_{22}/K_{11}$. The instability threshold is determined as a minimum $E_c = E(q_c)$ on the curve $E(q)$. Critical potential difference $U_c = E_c d$ does not depend on layer thickness $d$.

Dependence of threshold characteristics on parameter $K_{22}/K_{11}$ is different for negative and positive $e_a$. The values $U_c$ and $q_c$ increase almost linearly with increasing of ratio $K_{22}/K_{11}$ (Fig. 2) for $e_a < 0$. At $e_a > 0$ the threshold voltage increases up to Fredericks transition threshold $U_p = \pi (4 \pi K_{11}/e_a)^{1/2}$ and wave vector decreases to zero (see Fig. 3). At $K_{22} = 0$ solution does not exist. Calculations were performed for the following values of liquid crystal parameters : $K_{11}, K_{22}, e_a, e^*, q_c, d$.

<table>
<thead>
<tr>
<th>$\cos (\lambda s_1)$</th>
<th>$\cos (\lambda s_2)$</th>
<th>$0$</th>
<th>$0$</th>
<th>$\sin (\lambda s_1)$</th>
<th>$\sin (\lambda s_2)$</th>
<th>$\gamma_1 \cos (\lambda s_1)$</th>
<th>$\gamma_2 \cos (\lambda s_2)$</th>
<th>$- \beta_1 \sin (\lambda s_1)$</th>
<th>$- \beta_2 \sin (\lambda s_2)$</th>
</tr>
</thead>
</table>

(9)

![Fig. 2. Dependence of threshold voltage $U_c$ (curve 1) and threshold wave vector $q_c$ (curve 2) upon $K_{22}/K_{11}$ at $e_a = -0.1.$](image-url)
The comparison of theoretical and experimental results allows to determine the value
\[
e^* = e_1 - e_2 = 1.7 \times 10^{-4} \text{ CGS units}.
\]

We considered the dependence of threshold characteristics upon surface coupling energy corresponding to polar (angle \(\theta\), energy density \(W_\phi\) [6]) as well as to azimuth (angle \(\phi\), energy density \(W_\phi\) [7]) director deviations. In the case of symmetric boundary conditions \(W_\phi = W_\phi = 0\) or \(W_\phi \sim W_\phi \rightarrow \infty\) the flexoelectric structure arises at the same threshold conditions. In the case of nonsymmetric boundary conditions \(W_\phi > W_\phi\) or \(W_\phi > 0\) the effect can arise only when \(W_\phi > W_\phi > K_{ij}/d\) or \(W_\phi > 0\) that is in qualitative agreement with results of reference [7].

These results show that the difference \(e_1 - e_2\) can be found with high accuracy from the comparison of theoretical and experimental dependences of flexoelectric threshold upon dielectric anisotropy (see Fig. 4). Observation of this effect in substances having smectic and nematic phases is of particular interest. In this case the threshold voltage must increase proportionally to \(K_{22}\) near the temperature of phase transition to smectic phase if \(e_\phi < 0\). In the same temperature range the flexoeffect for substances with \(e_\phi > 0\) must turn to Fredericks effect which is followed by sharp increasing of the period of modulated structure.

It must be noted that described structure is electrically polarized since there is a component of macroscopic polarization along Z axis \(\delta P_z \sim \epsilon_{1,2} \theta_0 \phi_0\), where \(\theta_0\) and \(\phi_0\) are the amplitudes of the director deviations. At \(U > U_e\) the amplitudes \(\theta_0\) and \(\phi_0\) are proportional to \((U - U_e)^{1/2}\) i.e.
\[
P_z \sim e_{1,2} \epsilon_e (U - U_e)/U_e.
\]

At \(U \gg U_e\) we have \(\theta_0 \sim \phi_0 \sim I\) and \(q \sim e_{1,2} E K^{-1}\), i.e. \(\delta P_z \sim \epsilon_{1,2} \theta_0 \phi_0\). Such change of polarization \(P_z(E)\) under the influence of low frequency electrical field can result in hysteresis phenomena connected with a long time of orientational relaxation \(\tau \sim (\gamma/K^2)\) where \(\gamma\) is a viscosity of substance. Thus some hysteresis loop can be observed at frequencies \(f \sim 10\) Hz. In reference [8] some oscillograms of dielectric hysteresis loop type were observed but perhaps they are caused by nonlinearity of voltampere characteristics [10]. Linear dependence \(q \sim E\) at \(U > U_e\) was observed in reference [8, 9].

Analytic and numerical calculations show that the existence of tilt orientation results in increasing of threshold \(U_e \sim \cos \theta_0\) where \(\theta_0\) is the tilt angle, i.e. the effect disappears for homeotropic orientation.

References