Electric field driven transformations of orientational structure in chiral nematic systems with large flexoelectricity

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Abstract. Transformation of the orientational structure in a plane-parallel cell of a cholesteric liquid crystal, was studied depending on the magnitude of the voltage applied to the boundary planes of the cell. The equilibrium configuration of the liquid crystal director field was found minimizing the free energy both analytically and numerically. We took into account the orientational elastic energy in the volume and at the cell boundaries, the energy of an inhomogeneous electric field, and the flexoelectric effect. Systems with large flexoelectric coefficient were studied in detail. It was found, that when small voltages are applied to the cell boundaries, a continuous Fredericks transition occurs first, and then, when certain characteristic voltage is achieved, a significant jump-like change in the orientational structure takes place, and this structure gradually changes with the further voltage increase. It is shown that various scenarios of transformation of the orientational structure can be implemented depending on the actual values of the set of material parameters of the system studied. The discovered ability to control the structure of a liquid crystal in a cell can find use in various technical devices.

1. Introduction
Changes in the orientational structure of liquid crystals (LCs) influenced by external electric and magnetic fields has been intensively studied over the past few decades [1–4]. This is primarily due to the applications in designing LC displays, optical switches and other devices, in which the orientational structure of the LC is controlled by weak external fields. Important feature of the electric field is that it is spatially inhomogeneous inside the cell [5–7]. The interest in studying flexoelectricity for the past few decades has grown [8–15], as it provides additional spatial distortion to the external field, hence it certainly affects orientational structure of LC. Threshold effects in LC cells with negative dielectric anisotropy in the presence of flexoelectricity were investigated in our article [16]. In the present work we consider LC with large flexoelectric coefficients in the presence of an external electric field. It allows to obtain exact analytical results for this case. We consider various scenarios of the orientational structure transformation depending on the material constants and on the external field magnitude. For the case of LC with positive dielectric anisotropy we compare the results of numerical simulations with analytical solutions obtained.
2. LC energy model considered

We consider a plane-parallel CLC cell, occupying the region \( 0 \leq z \leq L \) between two plane electrodes with applied voltage \( U \). The averaged orientation of long molecular axes is described by the unit vector director \( \mathbf{n}(z) \),

\[
n_x = \sin \theta \cos \varphi, \quad n_y = \sin \theta \sin \varphi, \quad n_z = \cos \theta.
\]

Here \( \theta = \theta(z) \) and \( \varphi = \varphi(z) \) are polar and azimuthal angles respectively. The orientational free energy in such LC cell can be written in the form [15]

\[
\mathcal{F} = \frac{S_\perp}{2} \left( K_{22} g_0^2 L + \int_0^L \left[ A(\theta)(\theta')^2 + B(\theta)(\varphi')^2 - 2C(\theta)\varphi' \right] dz \right.
\]

\[
+ \sum_{a=1,2} \left[ W_\varphi^{(a)}(\varphi(\alpha)) - W_\varphi^{(a)}(\varphi(0)) \right] - \frac{1}{4\pi} (U - 4\pi \bar{e} J_1)^2 J, \tag{2}
\]

where \( S_\perp \) is the area of the boundary plates, the prime denotes derivative with respect to \( z \).

Angles of easy directions at the boundaries are

\[
\theta^{(1)} = \theta(0), \quad \theta^{(2)} = \theta(L), \quad \varphi^{(1)} = \varphi(0), \quad \varphi^{(2)} = \varphi(L).
\]

We also used the notations

\[
A(\theta) = K_{11} \sin^2 \theta + \cos^2 \theta + 4\pi \bar{e}^2 \sin^2 2\theta / \mathcal{E}(\theta), \quad \mathcal{E}(\theta) = \varepsilon_\parallel + \varepsilon_\perp \cos^2 \theta,
\]

\[
B(\theta) = \sin^2 \theta \left( K_{22} \sin^2 \theta + K_{33} \cos^2 \theta \right), \quad C(\theta) = q_0 K_{22} \sin^2 \theta,
\]

and

\[
J^{-1} = \int_0^L \frac{dz}{\mathcal{E}(\theta)}, \quad J_1 = \varepsilon_\alpha^{-1} \ln \frac{\mathcal{E}(\theta(0))}{\mathcal{E}(\theta(L))}. \tag{3}
\]

Here \( K_{ii} \) are the Frank modulii, \( \varepsilon_\parallel \) and \( \varepsilon_\perp \) are dielectric permittivities along and perpendicular to the director respectively, \( \varepsilon_\alpha = \varepsilon_\parallel - \varepsilon_\perp \) is the dielectric anisotropy, \( \pi / q_0 \) is the helix period, \( \bar{e} = (e_1 + e_3) / 2 \) is the mean flexoelectric coefficient, \( W_{\theta,\varphi}^{(1,2)} > 0 \) are the elastic modulii for the surface anchoring energy of Rapini-Papoular type [17]. It is important to note, that the free energy functional (2) has nonlocal form due to the last term resulting from the electric field and flexoelectricity.

In the case of large \( \bar{e} \) and high \( U \) in relation to Frank modules

\[
\frac{\bar{e} U}{K} \gg 1, \tag{4}
\]

where \( K \) is the typical Frank module value in the system considered, the terms \( \propto K_{ii} \) and \( \propto W_{\varphi}^{(1,2)} \) in Eq. (2) can be neglected [15, 16]. Thus, in this case, nematic and cholesteric LCs are described similarly, and the free energy has the following form

\[
\mathcal{F} = \frac{S_\perp}{2} \left( - \frac{1}{4\pi} U^2 J + 2\bar{e} U J J_1 + 4\pi \bar{e}^2 \int_0^L \frac{\sin 2\theta \theta' - JJ_1}{\mathcal{E}(\theta)} dz \right.
\]

\[
+ W_\theta^{(1)}(\theta(0) - \theta_0^{(1)}) + W_\theta^{(2)}(\theta(L) - \theta_0^{(2)}) \right). \tag{5}
\]

Note that in this limit the energy does not depend on the azimuthal angle \( \varphi \).
Our goal is to study how the equilibrium structure of the LC cell (profile \( \theta(z) \)) changes depending on the applied voltage \( U \) for different sets of material parameters. In what follows we restrict ourselves with one of the most commonly used set of easy direction angles \( \theta_0^{(1,2)} = \pi/2 \). It is notable that the free energy (5) in this case is a functional of \( \cos^2 \theta \) only. Thus, it is convenient to use a substitution \( y(z) \equiv \cos^2 \theta(z) + \varepsilon_\perp/\varepsilon_a = \varepsilon(\theta)/\varepsilon_a \). Note that \( y(z) \) is confined: 

\[
y(z) \in \left[ \varepsilon_\perp/\varepsilon_a, \varepsilon_\parallel/\varepsilon_a \right], \quad \forall z \in [0, L].
\]

The free energy as a functional of \( y(z) \) has the form

\[
\mathcal{F} = \frac{S_\perp}{2} \left[ - \frac{1}{4\pi} U^2 J + 2\varepsilon U J I_1 + \frac{4\pi^2}{\varepsilon_a} \int_0^L \frac{(y' + J J_1)^2}{y} dz \right]
\]

\[
+ W^{(1)}_\theta(y(0)) + W^{(2)}_\theta(y(L)) - (W^{(1)}_\theta + W^{(2)}_\theta) \frac{\varepsilon_\perp}{\varepsilon_a}. \tag{6}
\]

In terms of \( y(z) \) we have

\[
J^{-1} = \varepsilon_a^{-1} \int_0^L \frac{dz}{y}, \quad J_1 = \varepsilon_a^{-1} \ln \frac{y(0)}{y(L)}. \tag{7}
\]

In order to find the equilibrium profile \( \theta(z) \), one should obtain function \( y(z) \) minimizing functional (6). Since the function \( y(z) \) is confined, the general necessary condition of the minimum of the free energy is nonnegativeness of its first variation

\[
\delta \mathcal{F} \geq 0, \quad \varepsilon_\perp/\varepsilon_a \leq y(z) \leq \varepsilon_\parallel/\varepsilon_a. \tag{8}
\]

Note that the curve \( y(z) \) must be located inside the rectangle

\[
\begin{cases}
0 \leq z \leq L, \\
\varepsilon_\perp/\varepsilon_a \leq y \leq \varepsilon_\parallel/\varepsilon_a.
\end{cases} \tag{9}
\]

It is of importance, that infinitesimal variations \( \delta y(z) \) is arbitrary, when \( y(z) \in (\varepsilon_\perp/\varepsilon_a, \varepsilon_\parallel/\varepsilon_a) \), while \( \delta y(z) \geq 0 \) if \( y(z) = \varepsilon_\perp/\varepsilon_a \), and \( \delta y(z) \leq 0 \) for \( y(z) = \varepsilon_\parallel/\varepsilon_a \). Condition (8) results in the following Euler-Lagrange equation:

\[
-2yy'' + (y')^2 - J^2 \left( J_1 - \frac{U}{4\pi} \right)^2 = 0, \tag{10}
\]

\[
\varepsilon_\perp/\varepsilon_a < y(z) < \varepsilon_\parallel/\varepsilon_a, \tag{11}
\]

\[
0 < z < L. \tag{12}
\]

Inside the interval \( (0, L) \) there can exist regions, where \( y(z) \) achieves limiting values

\[
y(z) = \varepsilon_\perp/\varepsilon_a \text{ or } y(z) = \varepsilon_\parallel/\varepsilon_a, \tag{13}
\]

and the condition (11) is not satisfied. In such regions Eq. (10) should be replaced with the following inequality

\[
\left( -2yy'' + (y')^2 - J^2 \left( J_1 - \frac{U}{4\pi} \right)^2 \right) \frac{\delta y}{y} \geq 0. \tag{14}
\]

Necessary criteria of the free energy minimization also include the following boundary conditions

\[
\begin{align*}
-\left( J - \frac{U}{4\pi} \right) \delta y(0) & \geq 0, \tag{15} \\
\left( J - \frac{U}{4\pi} \right) \delta y(L) & \geq 0. \tag{16}
\end{align*}
\]
where
\[ g_i = \frac{\varepsilon_a W_i}{8\pi\bar{e}^2}, \quad i = 1, 2. \] (17)

Note that inequalities (14)–(16) are analogues of Karush–Kuhn–Tucker conditions (see e.g. [18]) for the Euler-Lagrange equation and boundary conditions.

It is important to note that the search of the equilibrium LC cell orientational structure reduced to an unusual mathematical problem: Euler-Lagrange equation (10) is actually an integro-differential equation also containing the sought-for function values at the boundaries. Each of the boundary conditions (15) and (16) are actually inequalities and also include integral term \( J \) depending on the distribution of \( \theta(z) \) inside the volume and both values \( y(0) \) and \( y(L) \) in term \( J_1 \). Finally, in the case (13), Euler-Lagrange equation transforms into the inequality (14).

The sufficient condition of the free energy minimization can be obtained by complementing Eq. (8) with the condition of positiveness of the second variation of the free energy
\[ \delta^2 F > 0, \quad \varepsilon_\perp/\varepsilon_a < y(z) < \varepsilon_\parallel/\varepsilon_a. \] (18)

In the regions, where \( y(z) \) achieves its limiting values (13), the sufficient condition inside the LC-cell is provided by Eq. (14).

In order to solve Euler-Lagrange equation (10), we introduce a notation
\[ a = J(J_1 - \frac{U}{4\pi\bar{e}}). \] (19)

Since \( J(J_1 - U/(4\pi\bar{e})) \) is independent on \( z \), we obtain parametric equation
\[ -2yy'' + (y')^2 - a^2 = 0. \] (20)

Boundary conditions (15) and (16) also include this parameter. There are three different solutions of Eq. (20)
\[ y(z) = \pm az + b, \] (21)
\[ y(z) = \frac{c}{4} (z + b)^2 - \frac{a^2}{c}, \] (22)

where \( b \) and \( c \) are arbitrary constants. Thus the formula (19) is actually a self-consistency condition on the parameter \( a \), since \( J \) and \( J_1 \) depend on \( a \). It turned out, that the condition (19) for the linear solution (21) results in the equation
\[ \frac{JU}{4\pi\bar{e}} = 0, \]

which can only be satisfied in the trivial case \( U = 0 \). That is why in what follows we consider only parabolic solution (22) with regard to the condition (13). This condition in combination with Eqs. (13), (15) and (16) allows us to finally find parameters \( a, b \) and \( c \). It can be seen, that the boundary conditions (15) and (16) can be satisfied only when
\[ y(0) = \mu_0, \quad \mu_0 = \varepsilon_\perp/\varepsilon_a \text{ or } \mu_0 = \varepsilon_\parallel/\varepsilon_a, \] (23)
\[ y(L) = \mu_L, \quad \mu_L = \varepsilon_\perp/\varepsilon_a \text{ or } \mu_L = \varepsilon_\parallel/\varepsilon_a, \] (24)

so the boundary conditions can be easily verified accounting for the sign of the expressions within square brackets. Thus, in order to find parameters \( a, b \) and \( c \) in Eq. (22), we should use
self-consistence condition (19) and Eqs. (23), (24). We do not give explicit expressions for these parameters as they are too cumbersome.

In order to illustrate obtained results, we carried out numerical and analytical calculations of dependencies $y(z)$ and $\theta(z)$. In what follows, we restrict ourselves with the case $\varepsilon_a > 0$. The numerical algorithm is based on the technique proposed for finding the equilibrium $\theta(z)$ profiles in [7] and used in [15, 16]. The following set of parameters was used: $L = 0.006 \text{ cm}$, $\varepsilon_\perp = 7.2$, $\varepsilon_\parallel = 16.2$, $\varepsilon_a = 9$, $\bar{\varepsilon} = 0.01 \text{ statC/cm}$. It was found that depending on the value of $g_2L$, there can occur several scenarios of the orientational structure transformation. In Fig. 1 the structural transformation of LC is shown in the case of relatively weak surface anchoring energy $W_\theta^{(2)}$, so the following inequality is satisfied

$$g_2L < \frac{2}{\kappa}, \quad (25)$$

where $\kappa = \sqrt{\varepsilon_\parallel/\varepsilon_a}$. Note that curve 1 corresponds to the area of relatively low voltages $U$, and

![Figure 1](image)

**Figure 1.** Profiles $\theta(z)$ (in the left figure) and dependencies $y(z)$ (in the right figure) obtained in the case of weak anchoring, and inequality (25) is satisfied. Surface anchoring modules are $W_\theta^{(1)} = 0.0025 \text{ erg/cm}^2$, $W_\theta^{(2)} = 5 \times 10^{-4} \text{ erg/cm}^2$ for each curve. Applied voltage corresponding to the lines: 1 – $U = 0.04 \text{ V}$, 2 – $U = 0.05 \text{ V}$, 3 – $U = 1.5 \text{ V}$, 4 – $U = 4.67 \text{ V}$, 5 – $U = 7.5 \text{ V}$, 6 – $U = 15 \text{ V}$.

the criterion (4) is not satisfied for it. Nevertheless, it is still close to the real profile, which can be calculated using the full free energy model (2). We can also see a significant jump-like transformation of the structure between lines 1 and 2. This transformation takes place, when the voltage

$$\tilde{U}_1 = \frac{8\pi \bar{\varepsilon}}{\varepsilon_a} \ln \left( 1 + \frac{g_2L}{2} \right) \quad (26)$$

is achieved. We should mention, that for the parameters used in calculation, $g_2L \ll 1$, hence in this case $\tilde{U}_1 \approx W_\theta^{(2)} L/(2\bar{\varepsilon})$. Thereafter the profile changes gradually, and at voltage

$$U_1 = \frac{8\pi \bar{\varepsilon}}{\varepsilon_a} \ln \left( \frac{\kappa + 1}{\kappa} \right), \quad (27)$$

in the volume of the cell there appears saturated region, where $\theta = 0$. With the further increase of voltage, saturation propagates into the volume of the cell (lines 5-6 in Fig. 1).
If the surface anchoring satisfies the inequalities

\[ \frac{2}{\kappa} < g_2 L < \frac{4}{\kappa - 1}, \]  

(28)

the transformation scenarios changes, as it is shown in Fig. 2. The low-voltage transformation is analogical to this in the previous case, but when the voltage \( \tilde{U}_1 \) is achieved, jump-like transformation from curve 3 to 4 takes place. It is important to note, that right after this transformation, there appears saturated region. As the voltage continues to grow, this zone gradually expands.

Finally, if the surface anchoring energy is strong enough to satisfy the following inequality

\[ g_2 L > \frac{4}{\kappa - 1}, \]  

(29)

then the last scenario of the orientational structure transformation takes place. It is shown in Fig. 3. The low-voltage transformations are similar to these in the previous cases. However, as the voltage rises and reaches the value

\[ U_2 = \frac{8\pi \tilde{\varepsilon}}{\varepsilon_a} \ln \left( \frac{\kappa + 1}{\kappa - 1} \right), \]

there appears a region of saturation in the middle of the cell (see lines 4-5 in Fig. 3). This region continues to expand with the increase of voltage until \( \tilde{U}_2 \) reaches the value

\[ \tilde{U}_2 = U_2 + \frac{8\pi \tilde{\varepsilon}}{\varepsilon_a} \left( \frac{g_2 L}{2} \cdot \frac{\kappa}{\kappa - 1} \cdot \frac{\varepsilon_\perp}{\varepsilon_\parallel} - 2 \right). \]  

(30)

When it is achieved, a jump-like transformation takes place (in the presented set of parameters, \( U_2 = 19.68 \) V, see lines 5-6 in Fig.3). It is interesting to note, that the saturated region is larger in the resulting configuration (compare profiles 5 and 6 in Fig. 3). Thereafter the saturation region gradually expands, as the voltage increases (lines 6-7 in Fig. 3).
Figure 3. Profiles $\theta(z)$ (in the left figure) and dependencies $y(z)$ (in the right figure) obtained in the case of weak anchoring, and inequality (29) is satisfied. Surface anchoring modules are $W_{\theta}^{(1)} = 1.75 \text{ erg/cm}^2$, $W_{\theta}^{(2)} = 0.7 \text{ erg/cm}^2$ for each curve. Applied voltage corresponding to the lines: 1 – $U = 5 \text{ V}$, 2 – $U = 15 \text{ V}$, 3 – $U = 16 \text{ V}$, 4 – $U = 16.5 \text{ V}$, 5 – $U = 19.65 \text{ V}$, 6 – $U = 19.75 \text{ V}$, 7 – $U = 25 \text{ V}$.

The obtained results show, that in the case of high flexoelectric coefficients, there are several scenarios of the orientational structure transformation, which depend on the parameters of the system. The results also demonstrate the possibility of different saturation and jump-like transformations in such LC cells, which can be used to design switching devices of new type.

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