Nonlinear effects in a real flexoelectric structure

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The flexoelectric effect is considered in a nematic liquid crystal in an electric field above the threshold for the formation of a modulated structure. The dependence of the structure period on the field strength is determined from thermodynamic considerations. The defects produced in the flexostructure are investigated and their main properties determined. It is shown that the required field dependence of a modulated structure is made possible by defect production.

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1. The study of the nonlinear behavior of modulated orientational structures above the threshold of their formation in liquid crystals meets with certain difficulties because of the large number of parameters of such systems. The change of the flexoelectric periodic structure is of particular interest, since this structure exists in a dielectric liquid crystal in a wide range of electric fields above the excitation threshold, in contrast to the dissipative modulated structures, which have in an electrically conducting liquid crystal several instability modes at a slight excess above the threshold of the first mode. This feature of the flexoelectric effect is due to the thermodynamic character of the phenomenon: a modulated structure corresponds at all field values to a minimum of the free energy of a nematic liquid crystal (NMC) and preserves the number of orientational degrees of freedom, even though it is strongly distorted above the formation threshold. A linear theory of the threshold flexoeffect was developed in Refs. 1 and 2. We analyze below the behavior of an NLC above the threshold E_c in strong electric fields $E \gg E_c$.

We recall that a periodic flexostructure has two degrees of freedom, the angles θ and φ that describe the deviation of the director **n** from the initial planar orientation. In the linear approximation the angles θ and φ are the functions

$$\theta=\theta_1 \sin (qy) \cos (\pi z/d), \quad \varphi=\varphi_1 \cos (qy) \cos (\pi z/d),$$

where φ_1 and θ_1 are the amplitudes of the periodic perturbation, and q is the wave number of the structure (in the unperturbed state, an NLC is a layer of thickness d, planarly oriented along the x axis, with boundary conditions $\varphi = \theta = 0$ at $z = \pm d/2$; the z axis coincides with the electric-field direction). The relations between these quantities and the field E (at $E \gg E_c$) are

$$\varphi_1 \sim (E/q)\theta_1, \quad E=E(q)\sim q. \tag{1}$$

We emphasize that it is just the boundary conditions that cause the phenomenon to have a threshold when the threshold values of E_c and q_c correspond to the minimum of the relation $E = E(q)$ and to a stable equilibrium of the system. Above the threshold this $E(q)$ dependence does not satisfy the condition that the free energy $\mathscr{F}$ be a minimum in the nonlinear approximation. The qualitative proportionality (1) does take place, as we shall show, also in the nonlinear case, and the quantity $q = q^*(E)$ corresponds here to the lower harmonic.

2. We consider for simplicity the case when all the elastic constants of the NLC are equal, $K_j = K$, the dielectric anisotropy ϵ_a is zero, and the flexoelectric coefficients satisfy the relation $f_1 = -f_2 = f$. The last condition actually only simplifies the equations, since it is easy to verify that the flexocoefficients enter in all the expressions in the same combination $(f_1 - f_2)$. Since the period $2\pi/q$ of the system is much smaller at $E \gg E_c$ than the thickness d of the NLC layer, we can neglect in first-order approximation the derivatives with respect to the coordinate z in the expression for $\mathscr{F}$. Using the relations

$$n_x=\cos\theta\cos\varphi, \quad n_y=\cos\theta\sin\varphi, \quad n_z=\sin\theta,$$

with allowance for nonlinear corrections up to terms of sixth order in the angles[1] θ and φ, we write down the functional $\mathscr{F}$ (Ref. 1):

$$\mathscr{F}=\int dV\Big\{\frac{1}{2}K\Big(\theta'^2+\varphi'^2-\frac{1}{2}\theta^2\varphi'^2+\frac{1}{24}\theta^4\varphi'^2\Big) +fE\Big[\Big(\varphi-\frac{1}{6}\varphi^3+\frac{1}{120}\varphi^5\Big)\theta'-\Big(\theta-\frac{1}{2}\theta\varphi^2-\frac{2}{3}\theta^3 +\frac{1}{24}\theta\varphi^4+\frac{1}{3}\theta^3\varphi^2+\frac{2}{15}\theta^5\Big)\varphi'\Big]\Big\}. \tag{2}$$

The prime denotes here differentiation with respect to y; $\theta(y, z)$ and $\varphi(y, z)$ can assume values of the order of unity. The dependences of the angles θ and φ on the coordinate z are substituted in the form of the expansions

$$\begin{aligned}\theta(y,z)&=\theta(y)\cos\frac{\pi z}{d}+T(y)\cos\frac{3\pi z}{d}+\dots,\\ \varphi(y,z)&=\varphi(y)\cos\frac{\pi z}{d}+\Phi(y)\cos\frac{3\pi z}{d}+\dots.\end{aligned} \tag{3}$$

We solve the problem first by confining ourselves to the first harmonics in this expansion. We calculate next the corrections due to allowance for the following harmonics and verify that they are small.

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Integrating in (2) with respect to z, we obtain the expression

$$\mathcal{F}=dL\int dy\left\{\frac{1}{2}K\left(\frac{1}{2}\theta'^2+\frac{1}{2}\varphi'^2-\frac{3}{16}\theta^2\varphi'^2+\frac{5}{384}\theta^4\varphi'^2\right)\right.$$
$$+fE\left[\left(\frac{1}{2}\varphi-\frac{1}{16}\varphi^3+\frac{1}{384}\varphi^5\right)\theta'-\left(\frac{1}{2}\theta-\frac{3}{16}\theta\varphi^2\right.\right.$$
$$\left.\left.\left.-\frac{1}{4}\theta^3+\frac{5}{384}\theta\varphi^4+\frac{5}{48}\theta^3\varphi^2+\frac{1}{24}\theta^5\right)\varphi'\right]\right\}, \quad (4)$$

where L is the longitudinal dimension of the NLC layer. Variation of the functional (4) with respect to the variables θ and φ leads to a system of equations for the functions $\theta(y)$ and $\varphi(y)$. We seek the solution of the considered nonlinear problem in the form

$$\theta(y)=\theta_1\cos(qy)+\theta_3\cos(3qy)+\ldots,$$
$$\varphi(y)=\varphi_1\sin(qy)+\varphi_3\sin(3qy)+\ldots. \quad (5)$$

We shall verify below that the minimum of the functional $\mathcal{F}$ corresponds to the values $q\sim q(E)=2fE/K$, $\varphi_1\sim\theta_1$, $\varphi_3\sim\theta_3\ll\theta_1$. The last inequality justifies the assumption (5), while the approximate equalities obtained from consideration of the preceding approximation in the expansion of $\mathcal{F}$ in powers of θ and φ can be used in the nonlinear corrections contained in expression (4) and in the corresponding equations. After substituting expansions (5) in these equations we arrive thus at the algebraic relations

$$9\theta_3-3\varphi_3=0,\quad 9\varphi_3-3\theta_3-\frac{3}{16}\theta_1^3=0,$$
$$-\theta_1+\frac{q(E)}{q}\varphi_1-\frac{3}{8}\theta_1^3-0.071\theta_1^5=0,$$
$$-\varphi_1+\frac{q(E)}{q}\theta_1-\frac{3}{16}\theta_1^3-0.080\theta_1^5=0.$$

The solution of these equations is written in the form

$$\varphi_3\approx\frac{3}{128}\theta_1^3,\quad \theta_3\approx\frac{1}{128}\theta_1^3,$$
$$\varphi_1\approx\theta_1+\frac{3}{32}\theta_1^3-0.018\theta_1^5,\quad \frac{q(E)}{q}\approx1+\frac{9}{32}\theta_1^2+0.062\theta_1^5. \quad (6)$$

The density of the free energy (4) is then

$$F=\frac{1}{V}\mathcal{F}\approx\frac{1}{4}Kq^2(E)\left(-\frac{9}{64}\theta_1^4+0.055\theta_1^6\right). \quad (7)$$

From this we obtain a value $\theta_1=\theta_1^*$ that satisfies the condition $\partial\mathcal{F}/\partial\theta_1=0$, for the minimum of the free energy, as well as the corresponding value $q=q^*(E)$:

$$|\theta_1^*|\approx1.304,\quad q^*(E)\approx0.603q(E),\quad F(q^*)\approx-0.037Kq^2(E). \quad (8)$$

We now verify that it is indeed possible to retain in (3) only the first harmonics in the coordinate z; to do this we must estimate the values of $T(y)$ and $\Phi(y)$, using relations (6)–(8) as the zeroth approximation. Substituting (3) in the free energy (2) we obtain a functional that depends on the four variables $\theta(y)$, $\varphi(y)$, $T(y)$, $\Phi(y)$.

Assuming that $T\sim\Phi\ll\theta$, we confine ourselves in this functional only to the terms quadratic in T and Φ. The variational equations are then linear in these variables and we readily obtain

$$T(y)=T_1\cos(qy)+\ldots,\quad \Phi(y)=\Phi_1\sin(qy)+\ldots,$$

with

$$T_1\approx-\frac{3}{26}\theta_1-0.007\theta_1^3,\quad \Phi_1\approx-\frac{3}{26}\theta_1+0.021\theta_1^3.$$

It can be seen that allowance for the next higher harmonics in the coordinate z cannot change qualitatively the results (6)–(8).

We have thus found an approximate solution of the nonlinear problem and described the modulated flexoelectric structure of the NLC above the instability threshold at $E\gg E_c$. The spatial period $Y=2\pi/q^*(E)$ of this stripe structure is usually proportional to the electric field if $E\gg E_c$, i.e., $Y\approx\pi K/0.6fE$. [We note that the $Y(E)$ dependence is similar to the $X(Y)$ dependence corresponding to Meyer's orientational structure,[3] but in contrast to the latter the deflection angles θ and φ are bounded in the structure considered here.]

These qualitative results do not change if $\epsilon_a\neq0$ and $f_1+f_2\neq0$, provided that the condition $|\varepsilon_aK/4\pi(f_1-f_2)^2|<1$ is satisfied.

3. The director-field distortions given by relations (5) can be directly obseved in polarized light as a system of opaque stripes parallel to the x axis, with a period $2\pi/q$ along the y axis.[5] The described flexostructure can thus be regarded as a special diffraction grating with variable period, $q=q^*(E)$. This phenomenon can find practical use (see, e.g., Ref. 4). It is therefore important to investigate this structure in greater detail, particularly the defects observed in the system of the stripe domains.[5] Defects outwardly similar to edge dislocations in crystals appeared already at $E\gtrsim E_c$. In stronger fields the number of defects increases greatly; they combine into clusters that tend to localize the field of the domain-structure distortions.

We determine now the distortion energy of the described flexostructure. To this end we introduce the domain-distortion field $u(x,y)$, reckoned along the y axis, and express in its terms the NLC director inclination angles $\theta(\mathbf{r})$ and $\varphi(\mathbf{r})$:

$$\theta=[\theta_1\cos q(y-u(x,y))+\theta_3\cos 3q(y-u(x,y))+\ldots]\times\cos(\pi z/d),$$
$$\varphi=[\varphi_1\sin q(y-u(x,y))+\varphi_3\sin 3q(y-u(x,y))+\ldots]\times\cos(\pi z/d). \quad (9)$$

Substituting these functions in the general expression for the NLC free energy, with flexoelectric terms, and integrating with respect to the coordinate z, we obtain

$$\frac{1}{d}\mathcal{F}=\int dx\,dy\left\{\tilde{\Psi}(y-u)\left(1-2\frac{\partial u}{\partial y}+\left(\frac{\partial u}{\partial x}\right)^2+\left(\frac{\partial u}{\partial y}\right)^2\right)\right.$$
$$\left.-\tilde{R}(y-u)\frac{\partial u}{\partial x}-\tilde{\Phi}(y-u)\left(1-\frac{\partial u}{\partial y}\right)\right\},$$

where $\tilde{\Psi}$, $\tilde{R}$, and $\tilde{\Phi}$ are polynomials of sines and cosines of $q(y-u)$. To separate the nonlinearity with respect to $u(x, y)$ in explicit form, we make the substitution $y' = y - u(x, y)$. We also note that since the scale of the change of the functions $\tilde{\Psi}$, $\tilde{R}$, and $\tilde{\Phi}$ with respect to y' is at any rate less than $2\pi/q$, and that of the function $u(x, y')$ exceeds this period of the domains, these functions can be integrated with respect to dy' separately. We confine ourselves thereby to relatively long-wave distortions of the domains. Taking this remark into account, we obtain ultimately

$$\frac{1}{d}\mathcal{F}\approx\frac{V}{d}(\Phi+R)+\int dx\,dy\left\{\Psi\left(\frac{\partial u}{\partial y}\right)^2\right.$$
$$\left.+\Phi\left(\frac{\partial u}{\partial x}\right)^2+2\Phi\left(\frac{\partial u}{\partial x}\right)^2\frac{\partial u}{\partial y}+3\Phi\left(\frac{\partial u}{\partial x}\right)^2\left(\frac{\partial u}{\partial y}\right)^2\right\}, \tag{10}$$

$$\Psi=\frac{q}{2\pi}\int\limits_{-\pi/q}^{\pi/q}dy'\tilde{\Psi}(y')\approx\frac{1}{8}K_1q^2\varphi_1^2$$
$$+\frac{1}{8}K_2q^2\theta_1^2+\frac{3}{128}(K_3-K_2)q^2\varphi_1^4$$
$$+\frac{3}{128}(K_2-K_1-3K_3)q^2\varphi_1^4\theta_1^4+\ldots,$$

and the remaining parameters are similarly determined:

$$\Phi\approx\frac{1}{8}K_3q^2(\varphi_1^2+\theta_1^2)+\frac{3}{128}(K_1-K_3)q^2(\varphi_1^4+\theta_1^4)$$
$$+\frac{3}{128}(8K_2-2K_1-9K_3)q^2\varphi_1^4\theta_1^4+\ldots,$$
$$R\approx-\frac{1}{4}(f_1-f_2)Eq\varphi_1\theta_1+\frac{3}{128}(f_1-f_2)Eq\varphi_1^3\theta_1$$
$$+\frac{3}{64}(f_1-f_2)Eq\varphi_1\theta_1^3+\ldots.$$

The unperturbed free-energy density $\mathcal{F}_0/V=(\Phi+R)$ of the modulated flexostructure coincides with expression (7). The energy (10) acts as the "elastic" energy of the two-dimensional crystal made up of the flexoelectric domains. It reflects the symmetry properties of any stripe domain structure (point group D_{2h}). The components of the stress tensor of this effective crystal are

$$\sigma_{22}=\sigma_{yy}=2\Psi\,\partial u/\partial y,\quad \sigma_{12}=\sigma_{xy}=2\Phi\,\partial u/\partial x,\quad \sigma_{12}=\sigma_{21}. \tag{11}$$

We consider now an isolated defect, named hereafter dislocation, in such an effective crystal. We define the Burgers vector in the usual manner:

$$\oint\nabla u_D\,dl=mb, \tag{12}$$

where $b = 2\pi/q$ and m is an integer.

We confine ourselves in the energy (10) to terms quadratic in u. To satisfy condition (12) we must therefore introduce in the Euler equation a source in the form of a cut, such that a field crossing it acquires an energy mb. For a dislocation at the point $\mathbf{r} = 0$

$$\left(\frac{\partial^2}{\partial y^2}+\frac{\Phi}{\Psi}\frac{\partial^2}{\partial x^2}\right)u_D(\mathbf{r})=-mb\frac{\partial}{\partial y}\delta(y)\Theta(x),$$

where $\mathbf{r}$ is the two-dimensional radius vector, $\delta(y)$ is a delta function, and $\Theta(x)$ is the step function; $\Theta(x) = 1(x>0)$, $\Theta(x) = 0(x<0)$. This equation has in coordinate space the solution

$$u_D(\mathbf{r})=-\frac{mb}{2\pi}\operatorname{arctg}\frac{\Psi^{1/2}x}{\Phi^{1/2}y}+\frac{mb}{4}\operatorname{sign}(y), \tag{13}$$

from which it is easy to obtain the stress tensor and the dislocation energy:

$$E_D=\frac{m^2b^2}{2\pi}(\Psi\Phi)^{1/2}\ln\frac{L}{a}, \tag{13'}$$

where a is the cutoff radius.

It can be seen that the considered defects in the modulated flexoelectric structure are analogs of edge dislocations in an anisotropic solid with zero Poisson coefficient [see Eq. (11)]. The interaction forces between two dislocations $m(\mathbf{r}_1) = m_1\delta(\mathbf{r}_1)$ and $m(\mathbf{r}_2) = m_2\delta(\mathbf{r}_2 - \mathbf{r})$ are of the form

$$f_x=m_1m_2\frac{b^2d}{\pi}(\Psi\Phi)^{1/2}\frac{x}{x^2+(\Phi/\Psi)y^2},$$
$$f_y=m_1m_2\frac{b^2d}{\pi}\Phi\left(\frac{\Phi}{\Psi}\right)^{1/2}\frac{y}{x^2+(\Phi/\Psi)y^2}. \tag{14}$$

It can be seen from these expressions that it is expedient to arrange the dislocations on one line so as to form a wall either along the y axis or along the x axis. The dislocations move apparently only along the x axis, since there is no orientational modulation in this direction. For the same reasons, dislocations with opposite signs form walls mainly along the y axis.

4. We shall show now that the mechanism whereby the period of the considered flexoelectric modulated structure is decreased with increasing field is the formation of dislocations. We note first that in an ideal (defect-free) system of domains even a small change of the period changes, as can be easily verified, the direction of the director by angles of the order of $2\theta_1^*\sim\pi$ in practically the entire NLC volume. This process requires a macroscopically high energy. If, however, an increase of the electric field from E_0 to E leaves the flexostructure period equal to $b_0 = 2\pi/q^*_0$, where $q^*_0 = q^*(E_0)$, this means the appearance of a distortion $u = y(q^*(E) - q_0^*)/q^*(E)$ and of stresses

$$\sigma_{yy}=\sigma\approx0.8f^2K^{-1}\theta_1^{*2}E(E-E_0). \tag{15}$$

These distortions can be completely offset by creation of $N_0\sim 0.6fL(E - E_0/\pi K$ dislocations, which leads to the appearance of additional domains and to a change of the flexostructure period to $b = 2\pi/q^*(E)$ [see (8)]. When N_D dislocations appear in the NLC volume, the period of the modulated structure becomes equal to

$$b_{eff}=\frac{2\pi}{q_{eff}}\approx\frac{L}{0.6fE_0L/\pi K+N_D}. \qquad (16)$$

We consider now a dislocation-creation mechanism similar to the Frank-Read source in crystals.[6] A dislocation line with end points secured at the boundaries $z=\pm d/2$, located on the boundary of the NLC layer, exerts no influence on the flexostructure domain period. It can be produced by fluctuation at any inhomogeneity of the director distribution on the boundary. Since the Burgers vector (12) of the considered defects is always parallel to the y axis, this dislocation is acted upon by a force only in the x direction, due to the stresses (15). This bends the dislocation line. We note that since the director orientation is fixed on the boundaries $z=\pm d/2$, the dislocation-line segments located along this boundary vanish. Therefore, when the line curvature radius reaches the critical value $\sim d/2$ the dislocation "breaks away" from the pinned ends and moves into the interior under the influence of the force acting on it, altering thereby the period of the modulated structure. Thus, each such source produces a group of dislocations along the x axis; the frontal dislocation in this group can be stopped by the opposite boundary of the NLC or by some defect in the volume, at a macroscopic distance $\sim L$ from the source. The reverse (blocking) action of a similar group on the source is given by the expression $\sigma_{\rm rev}\sim-\sigma l(2L-l)$, where l is the length of the considered dislocation cluster and is connected with the number of defects in the cluster (N) by the relation $N=\pi l\sigma/2\Psi(q^*_{jeff})b_{\rm eff}$ (see Ref. 6, part IV).

Let us estimate the equilibrium number of dislocations in the modulated flexostructure. Since the defects are uniformly distributed over the NLC volume, we shall assume in the estimates that the total number of dislocations in the entire volume is $N_D\sim N^2$. The critical stress necessary to tear a dislocation away from the source is defined as $\sigma_c=2\tau/db$, where τ is the linear tension in the dislocation. If the volume contains already N_D dislocations, this stress takes the form

$$\sigma_c=\frac{2\tau(\dot{q}_{eff})}{db_{eff}}\approx\frac{0.6fE_0}{d}\theta_1^{*2}\left(1+\frac{\pi N_DK}{0.6fE_0L}\right)\ln\frac{d}{a}. \qquad (17)$$

The stress acting on the source is in this case

$$\sigma_D=\sigma(\dot{q}_{eff})+\sigma_{06}(\dot{q}_{eff})\approx1.6\frac{f^2}{K}\theta_1^{*2}E\left(E-E_0-\frac{\pi N_DK}{0.6fL}\right)\frac{L-l}{2L-l}. \qquad (18)$$

Taking into account the connection between l and $N_D\sim N^2$, which now takes the form

$$l\approx NK\left(E_0+\frac{\pi N_DK}{0.6fL}\right)\left[0.6fE\left(E-E_0-\frac{\pi N_DK}{0.6fL}\right)\right]^{-1},$$

we arrive at a relation between the number of dislocations and the field, determined by the condition $\sigma_c\leqslant\sigma_D$. In the case $l\ll L$ (when the number of dislocations in each cluster, meaning also in the entire volume, is still small), this relation takes the form

$$N_D\sim N^2\leqslant\left[\frac{1.2fd}{K}E(E-E_0)-E_0\ln\frac{d}{a}\right]\times\left[\frac{\pi K}{0.6fL}\ln\frac{d}{a}+\frac{2\pi d}{L}E\right]^{-1}. \qquad (19)$$

Substituting in (19) for the initial field E_0 the threshold value E_c at which an ideal domain flexostructure is produced, we can estimate that critical value of the electric field at which dislocation production is possible at all, i.e., the change of the period of the modulated structure

$$E^*\geqslant\frac{\pi K}{2fd}\left[1+\left(1+\frac{1}{0.3\pi}\ln\frac{d}{a}\right)^{1/2}\right]. \qquad (20)$$

This is the field correspondant to the end point of the plateau on the plot of $b=b(1/E)$ obtained in experiment.[4]

At large values of the field $E\gg E^*$ the modualted structure contains many dislocations. Using the approximation $l\lesssim L$ we obtain the field dependence of the number of defects:

$$\frac{\pi K^2}{0.6fL}N^3+\left(\frac{\pi K^2}{2.4fd}\ln\frac{d}{a}+\pi KE\right)N^2+KE_0N+\frac{KLE_0}{4d}\ln\frac{d}{a}-0.6fLE(E-E_0)\leqslant0. \qquad (21)$$

We use now the condition $qL\gg qd\gg1$ corresponding to strong fields and obtain from (21) the qualitative estimate

$$N_D\sim N^2\leqslant0.6(fL/\pi K)(E-E_0). \qquad (22)$$

We emphasize that it is precisely this value of N_D which is needed to change the flexostructure period from $b(E_0)$ to $b(E)$. Thus, the production of dislocations, grouped in definite clusters, leads to a dependence, dictated by thermodynamic considerations, of the period of the modulated structure on the field (see Sec. 2). Namely, at $E^*>E>E_c$ the domain size changes very little, and at $E\gg E^*\sim E_c$ the wave number of the structure depends linearly on the field. We note in conclusion that foregoing analysis points to the inevitable formation of a system of defects in a controllable diffraction grating (flexoelectric domain structure) when the electric field is greatly strengthened.

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[1)]Allowance for terms of higher order leads to corrections $\sim10^{-3}$ in the final results.

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