Flexoelectric polarization of a ferroelectric smectic liquid crystal

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The ratio of flexopolarization to piezopolarization in a chiral ferroelectric smectic C^* liquid crystal is determined both from the dependence of the polarization on the applied electric field and from the change of the amplitude of the electro-optical modulation. It is shown that the flexoelectric effect plays an important role in the deformation of the C^* helicoid.

1. INTRODUCTION

Most investigations[1–4] of the behavior of ferroelectric smectic C^* liquid crystals in an electric field are based on the Pikin–Indenbom theoretical model[1] in which the classical Landau–de Gennes approach is extended to the case of the specific symmetry of the C^* phase (C_2 group). The macroscopic polarization of C^*, which is usually determined from the general expression for the free energy, turns out to depend on a number of phenomenological coefficients that are material parameters of the particular liquid crystal (LC). In particular, a distinction is made between the piezoelectric contribution P_p due to the spontaneous tilt of the LC molecules in the layers, and the flexoelectric contribution P_f (the flexoelectric polarization) which appears only in the presence of nonzero gradients of the director-orientation angles. The first attempt to determine the relative value of the flexo- and piezoelectric contributions to the polarization was undertaken in Ref. 2, where the estimate obtained, $P_f/P_p \sim 1$–5, seemed quite crude, since the C^* viscoelastic constant used in these calculations were physically unrealistic.

It is stated in later papers[3,4] that the flexoelectric polarization of the C^* material *p*-*n*-decyloxbenzylidene-*p*-amino-2-methylbutylcinnamate (DOBAMBC) is negligibly small compared with the piezopolarization $P_f < 0.1$–$0.2P_p$; the arguments in these papers, however, cannot be regarded as convincing. The reasoning there was based on seemingly obvious qualitative considerations that more rigorous quantitative estimates proved to be wrong in principle. In Refs. 3, for example, it was not taken into account that the main contribution to the C^* flexoelectric polarization is made not by spontaneous helical twisting of the C^* director around the normal to the smectic layer, but mainly by the field-induced change of the C^* molecules relative to this normal. The pronounced minimum of the electro-optical modulation in

DOBAMBC mixed with the related ferroelectric liquid crystal *p*-*n*-hexyloxybnezylidine-*p*-amino-2-chlorpropylcinnamate (HOBAPC) is incorrectly interpreted in Ref. 4, where the authors have mistakenly assumed that this minimum corresponds to vanishing of the C^*-mixture total polarization, equal to the sum of the corresponding piezo- and flexoelectric contributions.

By a theoretical analysis of two types of independent experimental data, we show in this paper, within the framework of the theory of Refs. 1 and 5, that the contribution made to the polarization by the flexoelectric effect is of the same order as the contribution, heretofore assumed principal, made to the polarization by the spontaneous tilting of the molecules relative to the smectic planes. We have measured therefore, on the one hand, the macroscopic polarization $\langle P \rangle / P_c$ of a layer of C^* (DOBAMBC) as a function of the dc electric-field amplitude E right up to the total untwisting field E_c of the helicoid, $E_c : \langle P \rangle (E = E_c) = P_c$. We show that the plot of $\langle P \rangle / P_c$ vs (E/E_c) differs substantially from the universal $\langle \cos \varphi \rangle$ dependence[2] that would be obtained if the flexoelectric contribution to the polarization were negligibly small. On the other hand, we determine the amplitude ΔJ of the electro-optic modulation, in a low-amplitude ac field, of a mixture of two ferroelectric crystals (DOBAMBC + HOBACPC) and show that the observed strongly pronounced minimum of this amplitude (the compensation point) can be attributed only to the fact that the flexo- and piezoelectric contributions to the C^* polarization are comparable. We assess also the role of the various phenomenological coefficients that set the values of the spontaneous and induced polarizations of C^* in an electric field, and obtain the temperature dependence of the relative contribution of the flexoelectric polarization to the total polarization of the ferroelectric C^* LC DOBAMBC. The viscoelastic parameters of the LC, which enter in the expression for the C^* polarization, are determined from independent measurements. The paper consists of three parts: theoretical calculation of the flexoelectric polarization and of the electro-optical modulation amplitude in C^*, description of the corresponding experiment, and discussion of the results.

2. THEORY

A. Polarization of deformed C^* helicoid in an external field.

If the electric field E is parallel to the y axis of the smectic layer, the director of the ferroelectric smectic C^* liquid crystal undergoes a deformation characterized by two degrees of freedom[5]: the polar angle $\theta = \theta(z) = \theta_0 + \theta_1(z)$, where $|\theta_0| \ll 1$ and $|\theta_1| \ll |\theta_0|$, and the azimuthal angle $\varphi(z)$, where the coordinate z is perpendicular to the plane of the smectic C^* layers (Fig. 1). The total polarization $P(z)$ of the smectic C^* layer is the sum of the components $\mathbf{P}_{\parallel}(z)$ and $\mathbf{P}_{\perp}(z)$ parallel and perpendicular to the C_2 axis in the C^* layer, so that the spontaneous polarization $\mathbf{P}_0(z)$, which is directed along the C_2 axis, contributes only to $\mathbf{P}_{\parallel}$. The nonzero values of the polarization components $\mathbf{P}_{\perp}(z)$ and

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$\mathbf{P}_\parallel(z) - \mathbf{P}_0(z)$ are due to the mutual influence of the layers on the smectic C^* or to the flexoelectric effect.

According to the Pikin–Indenbom phenomenological model that describes the ferroelectricity in C^*, the free energy takes in this case the form[5]

$$\mathscr{F}_0 = a\theta^2 + b\theta^4 + \frac{1}{2}g'\left(\frac{\partial\theta}{\partial z}\right)^2 + \frac{1}{2}k'\theta^2\left(\frac{\partial\varphi}{\partial z} - q_0\right)^2 + \chi_\perp\theta E\cos\varphi\left(\mu_p - \mu_f\frac{\partial\varphi}{\partial z}\right) - \chi_\perp\mu_f' E\sin\varphi\frac{\partial\theta}{\partial z},$$
$$a = a_0'(T - T_c) - \frac{1}{2}\chi_\perp\mu_p^2 - \frac{1}{2}k' q_0^2,$$
$$q_0 = -\frac{1}{k'}(\lambda + \mu_p\mu_f\chi_\perp). \quad (1)$$

The parameters T_c, $a_0' > 0$, b, g', k', $\chi_\perp$, μ_p, μ_f, μ_f', λ in (1) are the LC material parameters. The quantities a_0, b, g', and $k'\theta^2$ are the elasticity coefficients for the θ and φ deformations of the director, $-\lambda/k$ characterizes an initial twist of the "cholesteric type," $\chi_\perp$ is the dielectric susceptibility, μ_p is the piezoelectric modulus, μ_f and μ_f' are the flexomoduli that characterize the flexodeformation along the spontaneous polarization axis C_2 of the C^* layer and the axis perpendicular to it. The components $\mathbf{P}_\parallel(z)$ and $\mathbf{P}_\perp(z)$ of the C^* polarization take in this case the form (Fig. 1)

$$\mathbf{P}_\parallel(z) = \{P_x, P_y\} = \{P_\parallel\sin\varphi, -P_\parallel\cos\varphi\} = \left\{\chi_\perp\left(\mu_p - \mu_f\frac{\partial\varphi}{\partial z}\right)\theta\sin\varphi, -\left(\mu_p - \mu_f\frac{\partial\varphi}{\partial z}\right)\theta\cos\varphi\right\},$$
$$\mathbf{P}_\perp(z) = \{P_x', P_y'\} = \{P_\perp\cos\varphi, P_\perp\sin\varphi\} = \left\{\chi_\perp\mu_f'\frac{\partial\theta}{\partial z}\cos\varphi, \chi_\perp\mu_f'\frac{\partial\theta}{\partial z}\sin\varphi\right\}. \quad (2)$$

Solving the Euler equations for the functional (1) we can find an approximate expression for the C^* director orientation in a low-amplitude electric field $E \ll E_c$, where

$$E_c = (\pi^2/16)k' q_0^2\theta_0/\chi_\perp\mu_p \quad (3)$$

is the threshold untwisting field for the C^* helicoid

$$\tilde{\theta}_1 = \frac{\theta_1}{\theta_0} = \frac{\alpha(1-\delta)}{\beta - \gamma}\cos q_0 z, \quad \varphi = q_0 z + \alpha\sin q_0 z, \quad (4)$$

θ_0 is the equilibrium tilt angle of C^* in the absence of a field. Here $\alpha = (\pi^2/16)(E/E_c) \ll 1$ is the relative amplitude of the field, $\beta = 4a/k'q_0^2$, $\gamma = q'/k'$ the relative values of the elasticity coefficients, and $\delta = (\mu_f - \mu_f')q_0/\mu_p$ the normalized flexoelectric coefficient of C^*.

When $E \sim E_c$ the system of Euler's equations for the

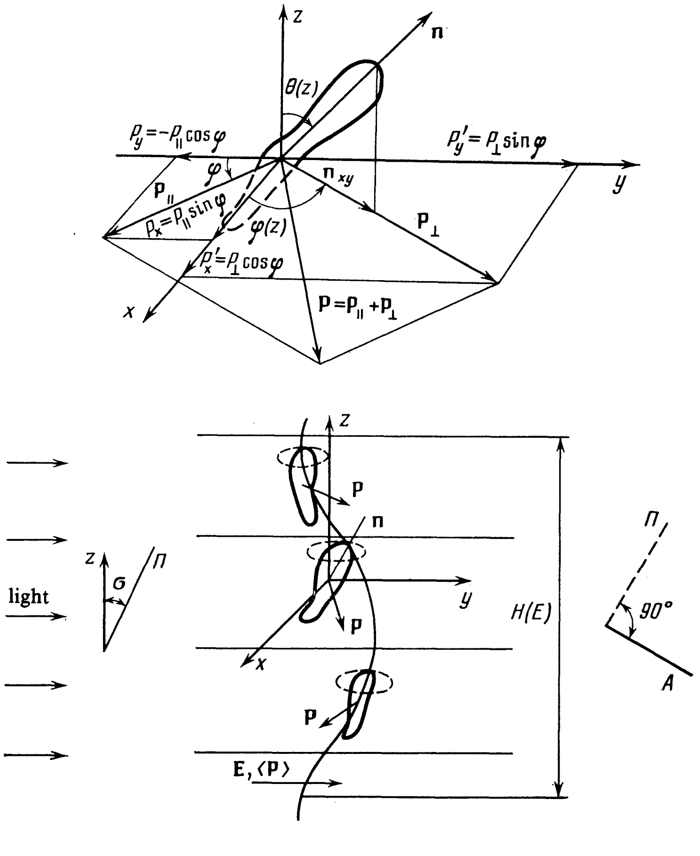

FIG. 1. Top: polarization **P** of ferroelectric smectic C^* layer of the LC; the component $\mathbf{P}_\parallel$ and $\mathbf{P}_\perp$ are respectively parallel and perpendicular to the smectic C_2 axis in the layer, $\theta(z)$ and $\varphi(z)$ are the orientation angles of the director **n** relative to the normal to the smectic layers (the z axis). Bottom: the C^* helicoid in an electric field $\mathbf{E}\parallel y$, $H(E)$ is the helicoid pitch, $\pi \perp A$ are the polarizer and analyzer between which the C^* layer is placed, σ is the polarizer orientation angle relative to the z axis, and $\langle P\rangle$ is the polarization of the C^* layer averaged along the z axis.

functional (1) admits of a solution $\varphi(z)$ that satisfies the condition[2]

$$\frac{z}{H}=x=\int_0^{\varphi/2}\frac{du}{G_1(k)\,(1-k^2\sin^2 u)^{1/2}},\qquad \tilde{\theta}_1(x)\approx\frac{F(x)}{\beta},\ (5)$$

where $H(E)$ is the C^* helix pitch in an external field satisfying the relation

$$H/H_0=\pi^{-2}G_1(k)\,G_2(k),$$

where

$$G_1=\int_0^{\pi}(1-k^2\sin^2 u)^{-1/2}\,du,\qquad G_2=\int_0^{\pi}(1-k^2\sin^2 u)^{1/2}\,du,$$

are elliptic integrals of the first and second kind, $H_0=H|_{E=0}$, and the parameter $0\leqslant k\leqslant 1$ is obtained from the relation $4k^2/G_2(k)=E/E_c$. The function contained in the second expression of (5) is equal to

$$F(x)=(\xi-1)^2+\alpha(1-\delta\xi)\cos\varphi,$$

$$\xi=q_0^{-1}\frac{d\varphi}{dz}=\frac{\pi}{G_2(k)}(1-k^2\sin^2\varphi/2)^{1/2}. \qquad (6)$$

the solutions (5) were obtained with account taken of the following assumptions:

$$\delta\ll\beta^2/\gamma,\quad \delta^2\leqslant\beta,\quad \gamma\ll\beta. \qquad (7)$$

An electric field E applied along the y axis that lies in the plane of the smectic layer deforms the helix of C^* and this in turn causes macroscopic polarization that is inhomogeneous along z.[6] The polarization $\langle P\rangle$ averaged over z, which is the quantity usually measured in experiments,[1)] is equal to

$$\langle P\rangle=\langle P_y+P_y'\rangle=-\chi_\perp\left\langle\left(\mu_p-\mu_f\frac{d\varphi}{dz}\right)\theta\cos\varphi-\mu_f'\frac{d\theta}{dz}\sin\varphi\right\rangle, \qquad (8)$$

where we have omitted terms that are of no importance in this case and are independent of the deformation. Recognizing that

$$\theta=\theta_0(1+\tilde{\theta}_1(z))\ \text{ and }\ \langle(d\varphi/dz)\cos\varphi\rangle=0,$$

we obtain from (8) for the polarization $\mathscr{P}$ of a ferrelectric C^* liquid crystal:

$$\mathscr{P}=\frac{\langle P\rangle}{\chi_\perp\mu_p\theta_0}=-\langle\cos\varphi\rangle-\left\langle\tilde{\theta}_1(z)\left(1-\delta_1 q_0^{-1}\frac{d\varphi}{dz}\right)\cos\varphi\right\rangle$$
$$+\delta_2\left\langle q_0^{-1}\frac{d\tilde{\theta}_1}{dz}\sin\varphi\right\rangle, \qquad (9)$$

$$\delta_1=\mu_f q_0\mu_p^{-1},\quad \delta_2=\mu_f' q_0\mu_p^{-1},\quad \delta=\delta_1-\delta_2.$$

Two methods can be proposed to find the polarization $\mathscr{P}$ as a function of the relative external field E/E_c. They are based either on an approximate calculation at [$E/E_c\ll 1$, Eq. (4)] or on an exact calculation [Eq. (5)] of the C^* director deformation in the external field. Thus, At $E/E_c\ll 1$, substituting (4) in (9), we get[2,5]

$$\mathscr{P}={}^1/_2\alpha[1-(1-\delta)^2/\beta]. \qquad (10)$$

In the general case when $E/E_c\sim 1$ we obtain according to (5) the expression

$$\mathscr{P}=-\int_0^{2\pi}\frac{d\varphi}{2G_1(1-k^2\sin^2\varphi/2)^{1/2}}$$
$$\times\left\{\cos\varphi+\frac{1}{\beta}\left[(\xi-1)^2(1-\delta_1\xi)\cos\varphi\right.\right.$$
$$\left.-\delta_2(2-2\xi+\alpha\delta\cos\varphi)\frac{k^2\pi^2\sin^2\varphi}{4G_2^2}\right]$$
$$\left.+\frac{\alpha(1-\delta\xi)}{\beta}[\cos^2\varphi(1-\delta_1\xi-\delta_2\xi)+\delta_2\xi]\right\}, \qquad (11)$$

in which the coefficients δ_1 and δ_2 enter separately.

B. Electro-optical modulation of planar C layer.*

If a cell with a planarly oriented (the smectic layers are perpendicular to the substrates) ferroelectric C^* liquid crystal is placed between crossed polarizers $\pi\perp A$ (Fig. 1), the calculation of the the relative intensity $J=I/I_0$ of the light passing through such a system is[7]

$$J=I/I_0=\langle(\sin^2 2\sigma+2\theta\sin 4\sigma\cos\varphi$$
$$+4\theta^2\cos 4\sigma\cos^2\varphi-{}^{16}/_3\theta^3\sin 4\sigma\cos^3\varphi)\sin^2\Delta\Phi/2\rangle. \qquad (12)$$

As before, the z axis is perpendicular here to the substrates of the C^* cell (of thickness L, $0\leqslant z\leqslant L$), σ is the angle between the optic axis of one of the polarizers and the z axis, $\Delta\Phi(z)=(2\pi L/\lambda)\Delta n$ is the phase delay, $\Delta n=n_e(z)-n_0$ is the birefringence, and n_e and n_o are the refractive indices for the extraordinary (e) and ordinary (o) monochromatic waves of length λ. When the sign of the external electric field applied to the C^* cell is reversed, $E\to -E$, the relative intensity of the transmitted light is correspondingly altered (electro-optical modulation[4)]):

$$\Delta J=J_E-J_{-E}. \qquad (13)$$

Let us obtain and expression for ΔJ for a weak deformation of the C^* helicoid, when $E/E_c\ll 1$. The deformation angles $\theta=\theta_0(1+\tilde{\theta}_1(z))$ and $\varphi(z)$ are defined here in accordance with (4) as

$$\tilde{\theta}_1|_{\pm E}=\pm\frac{\alpha(1-\delta)}{\beta}\cos q_0 z,\qquad \varphi|_{\pm E}=q_0 z\pm\alpha\sin q_0 z. \qquad (14)$$

By substituting (14) in (12) we can show that, accurate to small α^2 and θ_0^5, the phase factor $\sin^2(\Delta\Phi/2)$ in (12) can be regarded as constant and therefore excluded from the averaging in (12). Substituting next (14) in (12) and carrying out the appropriate averaging, we obtain

$$\Delta J\approx 2\theta_0\sin 4\sigma\sin^2\frac{\Delta\Phi}{2}\{[\langle(1+\tilde{\theta}_1)\cos\varphi\rangle_E-\langle(1+\tilde{\theta}_1)\cos\varphi\rangle_{-E}]$$
$$-{}^8/_3\theta_0^2(\langle\cos^3\varphi\rangle_E-\langle\cos^3\varphi\rangle_{-E})\}$$
$$+4\theta_0^2\cos 4\sigma\sin^2\frac{\Delta\Phi}{2}(\langle\cos^2\varphi\rangle_E-\langle\cos^2\varphi\rangle_{-E})$$
$$\approx 2\theta_0\alpha\sin 4\sigma\sin^2\frac{\Delta\Phi}{2}\left[-1+\frac{1-\delta}{\beta}+2\theta_0^2\right]. \qquad (15)$$

It follows from (15) that the amplitude of the electro-optical modulation of a planar C^* layer in a weak field can vanish at the compensation point of the following condition is satisfied:

$$1-2\theta_0^2 \sim -(\delta-1)/\beta. \tag{16}$$

3. EXPERIMENT

The dependence of the macroscopic polarization in DOBAMBC on the amplitude E of the dc electric field was determined by a pyroelectric technique[6,8] at temperatures T corresponding to different deviations of $T_c - T$ from the phase-transition temperature T_c. We measure directly the coefficient $\gamma = d\langle P\rangle/dT$, from which the polarization $\langle P\rangle$ was determined by graphic integration. This method of measuring $\langle P\rangle$ at a given temperature T is correct, since the duration of the measuring thermal pulse exceeded by two orders the relaxation times of a C^* sample weakly field-deformed by pulsed heating. We determined in the experiment the untwisting field E_c of the C^* helicoid, as well as the polarization $P_c = \chi_\perp \mu_p \theta_0$, of a completely untwisted helicoid. The corresponding plots of $\langle P\rangle/Pc$ vs E/E_c and of $\langle P\rangle/\chi_\perp\mu_p\theta_0$ vs E are shown in Fig. 2.

We measured also the electro-optical modulation amplitude in the ferroelectric smectic C^* liquid crystal in a low-amplitude ac field $E/E_c \ll 1$. In this case E^* was a mixture of DOBAMBC and HOBACPC. The experimental procedure was described in detail earlier.[4] The depth ΔJ of the electro-optical modulation was measured in the rather large temperature interval $0 < T_c - T < 5\,^\circ\mathrm{C}$ and at different HOBACPC concentrations. We have observed that at a HOBACPC concentration c equal to 23.5 wt. % there is practically no modulation of the optical signal by the electric field in the entire observable temperature region and in a wide range of external-field frequencies f. Figure 3 shows a plot of the temperature-averaged amplitude of the electro-optical modulation of a planar C^* layer at two frequencies, $f = 40$ Hz and $f = 10$ kHz, as a function of the HOBACPC concentration. It can be seen that the corresponding compensation points practically coincide.

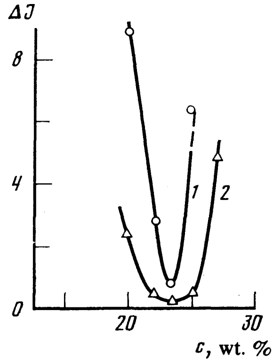

FIG. 3. Temperature-averaged amplitude ΔJ of the electro-optical polarization of a planar C^* layer at $E/E_c \ll 1$ for two ac frequencies, $f = 40$ Hz, $D = 250$ V/cm (curve 1) and $f = 10$ kHz, $E = 1500$ V/cm (curve 2), as a function of the HOBACPC concentration.

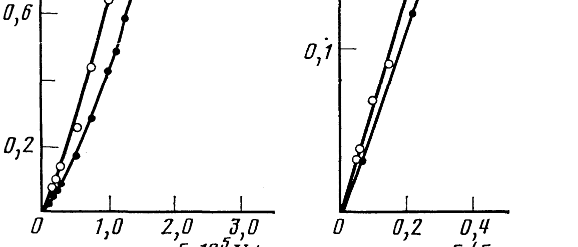

FIG. 2. a) Dependence of the relative polarization $\langle P\rangle/\chi_\perp\mu_p\theta_0$ on the E field for DOBAMC at various temperatures: curve 1—$T_c - T = 0.7\,^\circ$C, 2—2.7 °C, 3—4 °C. An estimate of the relative contribution to the flexopolarization (10) yields $\delta = 30$, 48, and 63 respectively for curves 1, 2, and 3. According to (18), $\beta(T - T_c) \sim 10^3$ [deg − 1]. b) Linear sections of the corresponding plots of $\langle P\rangle/P_c$ vs E/E_c in enlarged scale.

4. RESULTS AND DISCUSSION

Our experiments lead to estimates of the relative C^* flexoelectric coefficients $\delta_1 = \mu_f q_0/\mu_p$ and $\delta_2 = \mu_f' q_0/\mu_p$ (including their differences $\delta = \delta_1 - \delta_2$) that show the ratio of the flexo- and piezopolarization in a ferroelectric smectic C^* liquid crystal. The estimates call for knowledge of the parameter

$$\beta = \frac{4a}{k' q_0^2} = \frac{4(a_0 - {}^1/_2\chi_\perp\mu_p^2 - {}^1/_2 k' q_0^2)}{k' q_0^2} \sim \frac{4a_0}{k' q_0^2}, \tag{17}$$

$$a_0 = a_0'(T - T_c).$$

Estimates of the material DOBAMBC parameters $\chi_\perp, \mu_p, k', q_0$ are given in Refs. 2 and 5 and agree with those obtained by us in Ref. 9: $k' \sim 10^{-5}$ dyn, $\chi_\perp \sim 0.2$–0.3, $\mu_p \sim 79$–90 cgs esu, and $q_0^2 \sim 10^8$ cm^{-2}. The value of a_0' measured in Ref. 9, however, differs from those in Refs. 2 and 5 by three orders: $a_0' = 2.5\ 10^5$ cgs esu/deg. The correctness of the latter estimate of a_0' for the phase transition in DOBAMBC, as well as the causes of the erroneous estimates in Refs. 2 and 5, were considered in detail in Ref. 9 and will not be repeated here. We note only that the value of a_0' presented here agree well with the corresponding estimates of other studies, viz., $\gtrsim 2\cdot 10^4$, $\sim 10^5$, and $\sim 2.5\ 10^5$ cgs esu/deg in Refs. 10, 11, and 12, respectively. According to our data we have

$$|\beta| \sim \frac{4a_0'}{k' q_0^2}(T_c - T) \sim (T_c - T)\cdot 10^3\ [\deg^{-1}]. \tag{18}$$

The relative difference of the flexoelectric moduli can be estimated from our experiments in two ways: from the slope of the field dependence of the polarization (10), using the linear section of the plot $\langle P\rangle/P_c$ vs E/E_c (Fig. 2), and

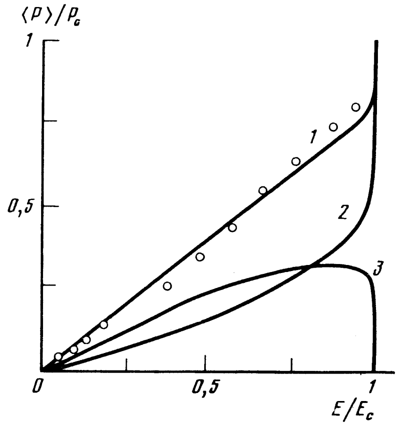

FIG. 4. Least-squares approximation of the experimental $\langle P\rangle/P_c$ dependence on E/E_c by the theoretical relation (11) for $T_c - T = 0.7$ °C and $\beta = 0.7\cdot 10^3$. O experiment, solid curve 1—calculated data. The relative deviation of the theoretical values from experiment does not exceed 20%. Curves 2 and 3 show the piezoelectric and flexoelectric contributions to the polarizations, respectively, as calculated from Eq. (11).

from the condition that the electro-optical modulation (16) of the planar C^* layer have zero amplitude. It follows thus from (16) that $\delta > 1$. Using the first of the proposed methods we obtain

$$\delta=\left|\left(2\frac{d\mathscr{P}}{d\alpha}\bigg|_{\alpha=0}-1\right)\beta\right|^{1/2}+1,\qquad \alpha=\frac{\pi^2}{16}\frac{E}{E_c},\quad \mathscr{P}=\frac{\langle P\rangle}{P_c}. \tag{19}$$

It follows from Fig. 2 that at $0.7 < T_c - T < 4$ °C the value of δ increases from 30 to 62.

Before we use the second method of estimating (16), we must note that our experiment was on a mixture of DOBAMBC and HOBACPC with $\theta_0 \sim \pi/6$ (Ref. 4), so that the corresponding values of δ and β differ from those obtained above. In particular, according to our estimates the threshold helicoid-untwisting field $E_c \sim k'q_0^2$ in the mixtures increases strongly compared with "pure" DOBAMBC corresponding to $\beta \sim (T - T_c)\cdot 10^2$ [deg^{-1}]. An estimate using (16) yields in this case $\delta \sim 0.5|\beta| + 1 \approx 50$, i.e., in the same range as the previously obtained δ.

If we use for the polarization the general formula (11) that is valid also for the case $E \sim E_c$, we can estimate the two normalized flexoelectric coefficients δ_1 and δ_2 (9) directly from the experimental dependence of $\mathscr{P} = \langle P\rangle/P_c$ on E/E_c, and calculate the relative values of the flexoelectric coefficients at $0 \leqslant E/E_c \leqslant 1$ (Fig. 4). A computer program was compiled for this purpose to obtain a least-squares fit of the experimental and theoretical curves (Fig. 4). The values of the parameter β for different $\Delta T = T_c - T$ were taken in this case from Eq. (18). The calculation yielded the relative values of the flexoelectric coefficients listed in the table. It can be seen that $|\delta_2| \gg |\delta_1|$ in all three cases, and the estimate of δ agrees with the data obtained by analyzing the linear section of the $\mathscr{P}(E/E_c)$ plot. We note that the values of δ in the table do not contradict the experimental conditions.[3,5]

TABLE I.

	$\Delta T = T_c - T$, °C		
	0,7	2,7	4
δ_1	5	4	5
δ_2	−30	−60	−77
$\delta=\delta_1-\delta_2$	35	64	82

Indeed, according to (1), when the real DOBAMBC parameters are substituted in the equation for the pitch q_0 we have

$$\delta_1=\frac{\mu_f q_0}{\mu_p}\sim\frac{k'q_0^2}{\chi_\perp \mu_p^2}\sim 1. \tag{20}$$

It follows from the table that the quantity $\delta_1 \sim \mu_f$ varies little with temperature, and practically the entire contribution to the temperature dependence of $\delta(\Delta T)$ is made by the coefficient $\delta_2 \sim \mu_f'(-\delta_2 \gg \delta_1)$, which increases strongly in absolute value with increasing distance from the point T_c of the transition into the C^* phase, i.e., with increasing ΔT. It follows from the experiment that the normalized polarization $\mathscr{P} = \langle P\rangle/\chi_\perp \mu_p \theta_0$ remains practically unchanged as a function of (E/E_c) when the temperature T is raised: $\partial\mathscr{P}/\partial(E/E_c) \sim$ const (Fig. 3). This means according to (10) that the dielectric susceptibility

$$\delta\chi_\perp=\frac{\partial\langle P\rangle}{\partial E}\sim\frac{\chi_\perp\mu_p\theta_0}{E_c}\frac{\partial\mathscr{P}}{\partial(E/E_c)}\sim\frac{\chi_\perp^2\mu_p^2}{k'q_0^2}\frac{\partial\mathscr{P}}{\partial(E/E_c)} \tag{21}$$

is a smooth function of temperature. If we assume that $(\chi_\perp\mu_p^2/k')[\partial\mathscr{P}/\partial(E/E_c)]$ is practically temperature-independent, then $\delta\chi_\perp(\Delta T) \sim q_0^2 \sim H_0^2(\Delta T)$ increases with increasing ΔT. An increase of $\delta\chi_\perp$ as a function of ΔT was observed also in experiment.[13,14] We note that this contradicts the data of Ref. 2, which are cited also in Ref. 5, where the relative flexoelectric coefficient δ is regarded as independent of temperature.

We point out in conclusion that our results on the ratios of flexopolarization and piezopolarization in a ferroelectric C^* liquid crystal (DOBAMBC or the mixture DOBAMBC + HOBACPC) allow us to conclude that the flexoelectric effect plays an important role in the deformation of the C^* helicoid in an external electric field. Of greatest importance here are the inhomogeneous polar deformations $d\theta/dz$ of the helicoid, which make an appreciable contribution to the C^* polarization. The ratio of this flexocontribution to the piezocontribution is in our opinion best described not by the dimensionless coefficient $\delta = (\mu_f - \mu_f')q_0/\mu_p$ or $\delta_1 = \mu_f q_0/\mu_p$, as was done up to now,[1–6] but by the ratio of the flexopolarization P_f of the C^* helicoid to the corresponding piezopolarization P_p, given according to (10) and the table ($E/E_c \ll 1$) by

$$\frac{P_f}{P_p}=\frac{(1-\delta)^2}{|\beta|}\sim\left(\frac{\mu_f' q_0}{\mu_p}\right)^2\frac{k'q_0^2}{4a}\sim 1.4\text{–}1.7 \tag{22}$$

and amounting to $P_f/P_p \sim 1$ at $0 \leqslant E \leqslant 0.95\,E_c$ (Fig. 4).

A more accurate quantitative estimate of the relative value of the flexoelectric polarization of the ferroelectric smectic C^* liquid crystal can be obtained only by taking into account the orienting action of the substrates on the distribution of the C^* director, an action that becomes particularly substantial in thin C^* samples.

[1)] The usual smectic-planes orientation perpendicular to the LC cell substrate is assumed, and the boundary conditions on the substrates are neglected (Fig. 1).

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