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Modulated Structures in Dielectric Liquid Crystals†

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Review of theoretical and experimental results concerned to formation of space-modulated orientational structures in liquid crystals is given. Properties of ideal liquid crystal dielectrics are considered. Origion conditions and properties of flexoelectric and flexoelectromagnetic structures in nematics, dielectric instabilities in cholesterics and smectics, ferroelectric structures in chiral smectics are discussed.

It is well known that an electric field induces a modulation of the orientational structure of liquid crystals. There are two types of such modulated structures (MS) having different physical natures. The dissipative one is connected with the transport of electric charges and mass of the substance. The thermodynamic MS are not accompanied by the irreversible transport processes. Such MS can exist in ideal liquid crystal dielectrics. In fact there are always some ion impurities and as a result two types of MS are possible in liquid crystals. However it is possible to change the thresholds of these orientational instabilities in dependence on the concentration of impurities, anisotropy of transport parameters, boundary conditions and frequency of an external field. Thus one can exclude or induce the effects of first and second types. Below we shall consider the thermodynamic orientational transformations and conditions of their observation.

1 MODULATED STRUCTURE PERTURBATIONS IN SMECTICS AND CHOLESTERICS

For generality it is useful to mention the mechanical instability because there is determinate similarity between the MS caused by the electromagnetic and mechanical actions. The first observed structure of this type was the instability of the smectic *A* under the action of a tension along the crystal axis.[1–4] The same effect can be induced by a compression in the smectic plane for example after the removal of the laser beam action. The critical values of the homogeneous deformations $u'_{\parallel}$ (tension) and $u'_{\perp}$ (compression)

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are

$$u'_{\parallel} = \frac{2\pi}{d}\left(\frac{K_1 C_{33}}{C_{13}^2}\right)^{1/2}, \qquad u'_{\perp} = -\frac{2\pi}{d}\left(\frac{K_1 C_{33}}{C_{11}^2}\right)^{1/2}, \tag{1}$$

the period of a square lattice of arising perturbations is

$$s = 2(2\pi d)^{1/2}\left(\frac{K_1}{C_{33}}\right)^{1/4}. \tag{2}$$

Here K_j are the Frank moduli, C_{ij} are the elastic moduli of the smectic A, d is the layer thickness.

The analogous MS can arise in cholesterics which are the layer systems too. The critical values u' and s are determined by the equations (1) and (2) but here the moduli K_1 and C_{ij} are some effective constants K_1 and C_{ij}:[5–7]

$$K_1 = \left(\frac{1}{8}\right)(3K_3 + K_2) \approx \left(\frac{3}{8}\right)K_3, \qquad C_{33} = K_2 q_0^2 = K_2\left(\frac{2\pi}{h_0}\right)^2$$

where h_0 is the pitch of a helix. Since $(K_j/C_{ik})^{1/2} \sim l$, where l is the molecular length, one can estimate the values

$$u' \sim l/d \sim 10^{-4} \quad \text{for smectics,}$$
$$u' \sim h_0/d \sim 10^{-1} \quad \text{for cholesterics,}$$

if $l \sim 10^{-7}$ cm, $h_0 \sim 10^{-4}$ cm, $d \sim 10^{-3}$ cm, $C_{ij} \sim K_2 h_0^2$. The Eqs. (1) and (2) differ from usually used ones by more accurate accounting of elastic positional and orientational properties of smectics and cholesterics.

2 PECULIAR BEHAVIOUR OF CHOLESTERICS

The same effect is possible under the action of electric and magnetic fields. It was described by Helfrich[5] in the framework of the coarse-grained model for cholesterics. In cholesterics with a small dielectric anisotropy ε_a the square lattice of perturbations arises if the value of electric field E is larger than the critical one, i.e., $E > E_c$. The values E_c and s are derived from the equations (1) and (2) by the replacement

$$K_1 \to \left(\frac{1}{8}\right)(3K_3 + K_2), \qquad C_{33} \to K_2 q_0^2, \qquad C_{13}u' \to \left(\frac{\varepsilon_a}{8\pi}\right)E^2.$$

Thus one has

$$E_c = \frac{(2\pi)^{3/2}(2K_2(3K_3 + K_2))^{1/4}}{(\varepsilon_a h_0 d)^{1/2}}, \qquad s = \left(\frac{3K_3 + K_2}{2K_2}\right)^{1/4}(2h_0 d)^{1/2}. \tag{3}$$

It must be noted that the Eqs. (3) are correct for thick layers of cholesterics ($d \gg h_0$) where the coarse-grained model has sense. Here the perturbation is a local bend at a constant number of cholesteric planes in the layer with a fixed thickness d. At $E - E_c \ll E_c$ the perturbations are small and the square lattice is observed.[8] At $E - E_c > E_c$ the linear theory is not applied and the observed cholesteric texture is characterized by local deviations of the helix axis on the angle $\pi/2$. In this case defects of the cholesteric structure promote the memory effect.[9]

The most full description of the linear threshold effect in cholesterics with real boundary conditions at any relations between the thickness d and pitch h_0 is given by Chigrinov *et al.*[10] The numerical computation results in the next conclusions which agree with the experimental data (see Figure 1). In absence of an electric field in the cell with hard boundary conditions the pitch $h_0(d) = 2d/m$ where m is the zone number. At planar boundary conditions (see Figure 1a) there is the interval of thicknesses $d < h_0/4$ where a helix is totally unwinded. Here an external field **E** induces the Frederickz transition only as well as in the case of twist-orientations at $m = 1$ and $d < h_0/4$ (see Figure 1b). The Frederickz transition is not accompanied by a modulation of the orientational structure in the plane xy where the x axis is the director orientation at $z = 0$ (**E** is parallel to z axis).

At $d > h_0/4$ and $m \gtrsim 1$ an electric field ($E \geqslant E_c$) induces the domain

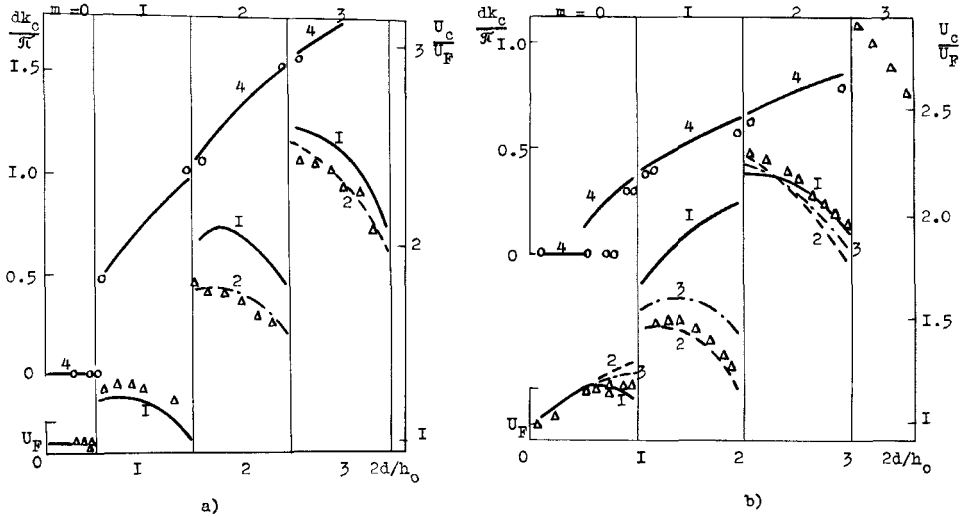

FIGURE 1 Dependence of threshold voltage (curves 1, 2, 3) and wave number (curve 4) upon cholesteric layer thickness for planar (a) and twist (b) orientations. Calculations correspond to the following values of orientation angles α: a) $\alpha = 0$—longitudinal domains (curve 1), $\alpha = \pi/2$ —transverse domains (curve 2), b) $\alpha = \pi/4$ (curve 1), $\alpha = 3\pi/4$ (curve 2), $\alpha = 0$ and $\alpha = \pi/2$ (curve 3). Experimental data are shown.[10]

picture with the wave vector $\mathbf{k}$ which has the components $\mathbf{k}_x = \mathbf{k} \cdot \sin\alpha$ and $\mathbf{k}_y = \mathbf{k} \cdot \cos\alpha$. The calculation of threshold characteristics of the polar and azimuth perturbations for the planar case gives the results (see Figure 1a):

at $m = 1$ the lowest threshold value $U_c = E_c d(k_c = 2\pi/s \neq 0)$ corresponds to the angle $\alpha = 0$,

at $m = 2$ and $m = 3$ the lowest value U_c corresponds to the angle $\alpha = \pi/2$.

For the twist-orientation one has (see Figure 1b):

at $m = 1$ the value $U = U_c$ corresponds to $\alpha = \pi/4$,

at $m = 2$ the value $\alpha = 3\pi/4$ is favourable,

at $m =$ the values $\alpha = \pi/4$ in the lower half and $\alpha = 3\pi/4$ in the upper half of this zone are favourable. These conclusions are in accordance with the experiment. At $m \geqslant 4$ the calculated threshold characteristics of the square lattice of perturbations describe the experimental dependence $U_c(d)$ (see Figure 2).

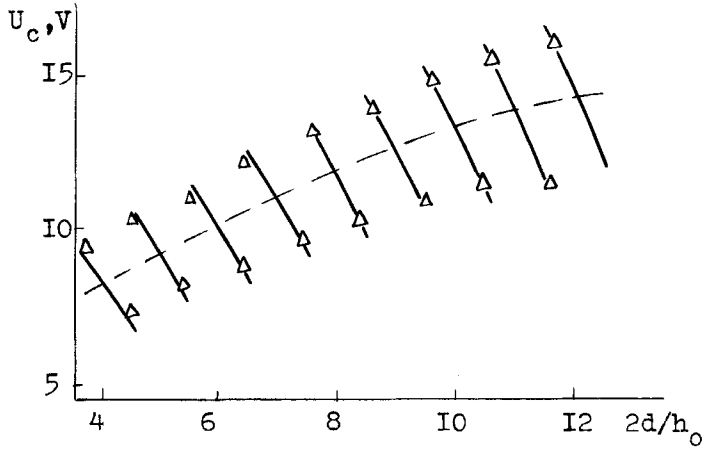

FIGURE 2 Theoretical and experimental dependences of threshold voltage upon cholesteric layer thickness for zone numbers $m > 4$.[10]

3 INSTABILITIES OF SMECTICS IN ELECTRIC FIELDS

In smectics the described MS have the threshold characteristics

$$U_c^2 = \frac{8\pi^2}{\varepsilon_a} d(K_1 C_{33})^{1/2}, \qquad s^2 = 8\pi d\left(\frac{K_1}{C_{33}}\right)^{1/2}. \tag{4}$$

One can see from (3) and (4) that in smectics the value U_c in $(h_0/1)^{1/2}$ times larger and the value s in $(h_0/l)^{1/2}$ times smaller than in cholesterics. It must

be noted that in smectics above the threshold an increase of perturbations must induce defects or systems of defects for example dislocations.

Parodi[11] noted that in smectics the phase transition of the first order can take place under the action of an electric field. In this case the favourable orientation of smectic layers arises by a jump. Here the volume energy gain compensates the surface energy loss which increases by a jump because of arising of dislocations. These dislocations conjugate the smectic layers near the solid surface with ones in the sample volume. The condition of such energy compensation determines the transition point E_c:

$$E_c^2 \sim \frac{8\pi W}{\varepsilon_a dl} \tag{5}$$

where W is the energy of a dislocation core, l is the core diameter. One has from the dimensionality consideration that $W \sim C_{33} l^2$ and consequently the expressions (4) and (5) give the same order of value $U_c \sim d^{1/2}$. The space periodic distribution of defects near the surface has similarity with the heterogeneity (4), i.e., $s \sim (dl)^{1/2}$. The value U_c must decrease with a temperature increasing because $C_{33} \sim |\psi|^2$ where ψ is the smectic order parameter. Since $U_c \sim |\psi|^{1/2}$ according to (4) and $U_c \sim |\psi|$ according to (5) the process of the generation of defects is more favourable near the smectic–nematic transition point. The experimental data[12] support these qualitative presentations.

Such instabilities of dielectric cholesterics and smectics have their electric current analogies. The thresholds of the formation of MS connected with a non-homogeneous distribution of an electric current are derived from (3) and (4) by the substitution

$$1/\varepsilon_a \to \sigma_{\parallel z}/\varepsilon_{\parallel z}(\sigma_{\perp z} - \sigma_{\parallel z})$$

where the designations $\perp z$ and $\parallel z$ correspond to the directions which are perpendicular and parallel to the crystal axis z, $\sigma_{\perp z}$ and $\sigma_{\parallel z}$ are the electroconductivity values. Thus the dissipative MS are less energetically favourable if $\varepsilon_a \sigma_{\perp z} \gg \varepsilon_{\parallel z}(\sigma_{\perp z} - \sigma_{\parallel z})$.

4 FLEXOELECTRIC INSTABILITY IN NEMATICS

Another physical phenomenon accompanied by the formation of a modulated orientational structure is the flexoelectric (FE) instability in nematics.[13] It is well known that there are possible the one-dimensional FE deformation connected with the surface effects[14] and two-dimensional FE deformation which has the volume character. The last one can be conditioned by a heterogeneity of an electric field caused by the presence of a volume electric charge[15]

or special disposition of electrodes.[16] However the volume FE deformation induced by a homogeneous electric field is of principal interest.

Meyer[13] have shown that in an infinite nematic or in the case of free boundary conditions the director distribution $\mathbf{n}(\mathbf{r})$ is described by the law $\theta = (f/K)\mathrm{Ex}$. Here the field is parallel to the z-axis, θ is the angle between the director $\mathbf{n}$ and x-axis in the plane xz, $f = f_2 = -f_1$ is the FE coefficient, $K = K_3 = K_1$ is the Frank constant, $\varepsilon_a = 0$. Thus the Meyer structure has the period $X = 2\pi K/fE$. At hard boundary conditions such structure was not observed even in strong electric fields.

Bobylev and Pikin[17] have shown that another FE structure being two-dimensional can take place even at hard boundary conditions (see Figure 3).

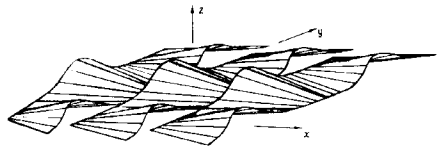

FIGURE 3 Two-dimensional flexoelectric structure.

This structure being periodical along the y-axis with the period Y has two angular director deviations from the x-axis: $\theta(y, z) \sim \sin(qy)$ and $\varphi(y, z) \sim \cos(qy)$. The threshold characteristics of this structure are

$$U_f = \frac{2\pi K}{|f_1 - f_2|(1 + a)}, \qquad Y_f = \frac{2\pi}{q_f} = 2d\left(\frac{1 + a}{1 - a}\right)^{1/2} \tag{6}$$

where $a = \varepsilon_a K/4\pi(f_1 - f_2)^2$, $|a| < 1$. One can make the next conclusions from (6) if the difference $|f_1 - f_2|$ is large or the values $|\varepsilon_a|$ and K are small: $U_f \to \infty$ if $a \to -1$, i.e., the system is stabilized, $Y_f \to \infty$ if $a \to 1$, i.e., the Frederickz-transition is induced. These qualitative conclusions are preserved at $K_1 \neq K_3$ in real nematics[18] and are confirmed by the experiment[19] (see Figure 4). The expressions (6) allow a generalization with due regard for the heterogeneity of an electric field and corresponding contribution to the free energy.[20] It must be noted that described structure can arise without a threshold if the boundary conditions are weak. At hard boundary conditions and initial tilt orientations the threshold U_f depends on the tilt angle Θ: $U_f \sim \cos^{-1}\Theta$, i.e., the threshold is infinite in the homeotropic situation.

Above the threshold at $E \gg E_f$ the stationary FE modulated structure with large wave numbers $q \gg q_f$ is energetically favourable. Thus it essentially differs from dissipative MS which have several instability modes slightly above the first threshold. This peculiarity of the flexoeffect is connected with the thermodynamic character of the phenomenon: the MS correspond to the minimal free energy of a nematic and preserve the number of freedom

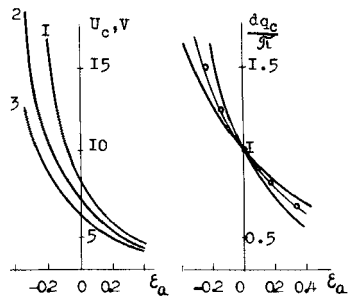

FIGURE 4 Dependence of flexoelectric threshold characteristics upon dielectric anisotropy. Calculations correspond to the following values of difference $|f_1 - f_2|$: $1.5 \cdot 10^{-4}$ (curve 1), $1.8 \cdot 10^{-4}$ (curve 2), $2 \cdot 10^{-4}$ (curve 3) CGS units.[17–19]

degrees at any values $E > E_f$. The solution of the nonlinear problem in the approximation $K_1 = K_3 = K$, $f_1 = -f_2 = f$, $\varepsilon_a = 0$ and $q \gg 2\pi/d$ shows that such structure is described by the functions

$$\theta(y) = \theta_0 \sin(qy) + \theta_1 \sin(3qy) + \ldots,$$
$$\varphi(y) = \varphi_0 \cos(qy) + \varphi_1 \cos(3qy) + \ldots,$$

where $\theta_0 \approx 1.1$, $\varphi_0 \approx -1.0$, $\varphi_1 \approx -\theta_1 \approx \theta_0^3/8$, $q \approx 1.8fE/K$, i.e., $Y = 2\pi/q \approx 1.1K/fE$. In this case the free energy density is $F(Y) \approx -1.1(fE)^2/K$ just as the Meyer-structure is characterized by the value $F(X) \approx 0.5F(Y)$, i.e., the considered FE structure is more energetically favourable in strong fields. The dependence $Y \sim dU_f/U$ is confirmed by the experiment.[19]

At $E > E_f$ the induced polarized state in nematics is described by the macroscopic polarization

$$P = (\chi_e + \delta\chi_e)E, \qquad \delta\chi_e \sim \frac{(f_1 - f_2)^2}{4K}.$$

Usually this phenomenon is observed in constant and low-frequency alternating electric fields. At frequencies $\omega > 2\pi/\tau_0 \sim 10\,\mathrm{sec}^{-1}$ where τ_0 is the orientational relaxation time the hysteresis phenomena must take place. The observed hysteresis loops can be caused also by other reasons for example by the volume electric charge attached to electrode region. One can see also that at $\omega \gg 2\pi/\tau_0$ and $\varepsilon_a < 0$ the value U_f has the frequency dependence[17]

$$U_f \sim 4d(\gamma_1\omega/|\varepsilon_a|(1 - |a|))^{1/2}$$

where γ_1 is the viscosity coefficient, just as the wave number q_f depends on ω weakly. At $\omega \gg 2\pi/\tau_0$ and $\varepsilon_a > 0$ the Frederickz-transition can realize

only. Thus in nematics with $\varepsilon_a < 0$ at comparatively low frequencies $\omega/2\pi \gtrsim 10$ Hz the dissipative instabilities can precede the flexoeffect. In principle the high-frequency FE effect can be observed in nematics near the temperatures of phase transitions to smectic phases where ordinary dissipative MS can not arise.

5 FLEXOELECTROMAGNETIC INSTABILITY IN NEMATICS

The FE effect in nematics has not the pure magnetic analogy because the last one is forbidden by the symmetry consideration. In principle paramagnetic nematics can exist. Non-chiral molecules of such substances must have the symmetry plane and permanent magnetic moment **m** which is perpendicular to this plane (see Figure 5). If the nonchiral paramagnetic molecule has the permanent electric dipole moment **p** one can make a conclusion about simultaneous electric and magnetic dipole ordering which is accompanied by the orientational deformation of such nematics.[21] It is obvious that this deformation must arise at the simultaneous action of electric (**E**) and magnetic (**H**) fields. Thus the considered effect is linear in the EH value just as the flexoelectric effect is linear in the **E** value. Therefore the flexoelectromagnetic (FEM) effect can precede the FE effect if the threshold U_f is sufficiently high. Such possibility arises for high ε_a values or at the equality $f_1 = f_2$ which is connected with a special form of anisotropic molecules (see Figure 5a). However owing to the interaction with magnetic moments the sufficiently strong magnetic field orders the plane orientation of molecules and promotes the modulation of the director distribution $n(r)$ under the action of electric fields (see Figure 5b).

Formally, from the symmetry consideration, one can write the contribution δF to the free energy density as

$$\delta F = -e_1([\mathbf{EH}]\mathbf{n})\mathrm{div}\mathbf{n} - e_2(\mathbf{H}\mathrm{rot}\mathbf{n})(\mathbf{En}) - e_3(\mathbf{E}\mathrm{rot}\mathbf{n})(\mathbf{Hn}) - e_4(\mathbf{n}\mathrm{rot}\mathbf{n})(\mathbf{EH}) \tag{7}$$

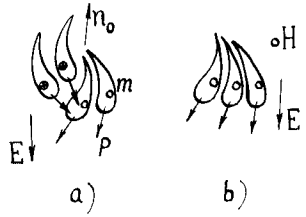

FIGURE 5 Flexoelectromagnetic effect: a) action of electric field, b) simultaneous action of magnetic and electric fields.

where e_j are FEM coefficients. At $e_4 \neq 0$ and $\mathbf{E} \| \mathbf{H} \perp \mathbf{n}_0$, where $\mathbf{n}_0$ is an initial director orientation, the paramagnetic nematic must gain the finite space twisting with the pitch $h = \pi K_2/e_4 EH$ (see Figure 6a). This effect is possible if the influence of boundaries is absent and the anisotropies $\varepsilon_a = \chi_a = 0$ where χ_a is the diamagnetic anisotropy.

In nematic layers with a finite thickness d along the z-axis and hard boundary conditions the next threshold FEM effects are possible:[21]

1) At $\vec{\mathbf{E}} \perp \vec{\mathbf{H}}$, $\vec{\mathbf{H}} \| \vec{\mathbf{n}}_0$ and $\vec{\mathbf{E}} \perp z$-axis (see Figure 6b) the arising domaine structure and threshold characteristics are similar to the FE ones, i.e.,

$$E_c = \frac{2\pi K}{|e_1 + e_2|(1 + b_1)dH}, \quad Y_c = 2d\left(\frac{1 + b_1}{1 - b_1}\right)^{1/2}, \quad b_1 = \frac{\varepsilon_a K}{4\pi(e_1 + e_2)^2 H^2},$$

$$|b_1| < 1.$$

2) At $\vec{\mathbf{E}} \perp \vec{\mathbf{H}}$, $\vec{\mathbf{E}} \| \vec{\mathbf{n}}_0$ and $\vec{\mathbf{H}} \perp z$-axis (see Figure 6c) there is the FE analogy too, i.e.,

$$H_c = \frac{2\pi K}{|e_1 - e_3|(1 + b_2)dE}, \quad Y_c = 2d\left(\frac{1 + b_2}{1 - b_2}\right)^{1/2}, \quad b_2 = \frac{\chi_a K}{4\pi(e_1 - e_3)^2 E^2},$$

$$|b_2| < 1.$$

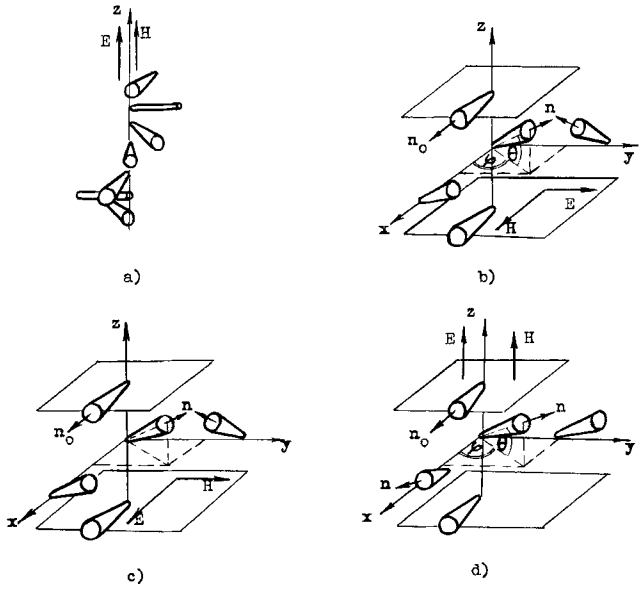

FIGURE 6 Flexoelectromagnetic modulated structures: a) helix, b) and c) longitudinal domains, d) transverse domains.

3) At $\vec{\mathbf{E}} \| \vec{\mathbf{H}} \perp \vec{\mathbf{n}}_0$ (see Figure 6d) the modulation arises along the x-axis. At $\chi_a = 0$ the threshold values are

$$E_c = \frac{2\pi K}{|e_1 + e_3 + 2e_4|(1 + b_3)dH}, \qquad X_c = 2d\left(\frac{1 + b_3}{1 - b_3}\right)^{1/2},$$

$$b_3 = \frac{\varepsilon_a K}{4\pi(e_1 + e_3 + 2e_4)^2 H^2}, \qquad |b_3| < 1.$$

Supposing the coefficients e_j being proportional to the moments p and m one can estimate $|e_j|$ from the dimensionality consideration: $|e_j| \sim mp/Kl^3 \sim 10^{-20}$ sec. Thus one has the estimation $H_c \sim 10^7$ ampere/meter at $U = dE \sim 10^2$ volts, i.e., the strong fields are necessary for an observation of these effects.

6 FLEXOEFFECTS IN CHIRAL SMECTICS

The induced polarization arising as a result of the FE effect is a quadratic function of the director perturbations θ and φ where θ is the polar angle, φ is the azimuth angle. The spontaneous polarization $\mathbf{P}_0$ in the chiral smectic $\tilde{C}$ can be partly caused by the spontaneous modulation $\mathbf{n}(\mathbf{r})$. The corresponding contribution to $\mathbf{P}_0$ is proportional to the FE coefficient f_0, wave number of a helix $q_0 = d\varphi_0/dz$ and polar angle θ_0 being the phase transition parameter:[22,23]

$$P_0 = (\mu - f_0 q_0)\theta_0$$

where μ is the piezoelectric coefficient. It must be noted that $\mathbf{P}_0$ is parallel to the second order symmetry axis in the monomolecular smectic layer.

The induced polarization $\mathbf{P}$ in the $\tilde{C}$ phase is not obviously parallel to the symmetry axis and has in general three independent contributions:

$$\mathbf{P} = \mathbf{P}_0 + \mathbf{P}_\| + \mathbf{P}_\perp$$

where $\mathbf{P}_\|$ is parallel to the crystal axis z, $\mathbf{P}_\perp$ is perpendicular to $\mathbf{P}_0$ and $\mathbf{P}_\|$. Correspondingly there are three independent FE coefficients $f_0, f_\|$ and $f_\perp$ in the $\tilde{C}$ phase. The coefficient $f_\perp$ relates $\mathbf{P}_\perp$ to $\partial\theta/\partial z$, the coefficient $f_\|$ relates $\mathbf{P}_\|$ to space derivatives of the angles θ and φ.

Thus under the action of an electric field $\mathbf{E} \perp z$-axis the orientational helix in the $\tilde{C}$ phase is perturbed in two ways. At weak fields the perturbations $\theta(z) - \theta_0$ and $\varphi(z) - \varphi_0(z)$ are small and independent. These modulations give the correction $\delta\chi_e$ to the dielectric susceptibility, i.e., to the linear response of the $\tilde{C}$ phase in weak fields:[22]

$$\langle P \rangle = (\chi_e + \delta\chi_e)E, \qquad \delta\chi_e = \frac{1}{2}\left[\frac{\mu^2}{Kq_0^2} + \frac{(\mu - (f_0 - f_\perp)q_0)^2}{gq_0^2 - 4A}\right]$$

where K and g are the elastic constants, $A \sim (-\theta_0^2) \sim (T - T_c)$, T_c is the phase transition temperature. One can see that the term $\delta\chi_e$ has both piezoelectric and FE contributions, the last one holding the difference of flexocoefficients $f_0 - f_\perp$ as in nematics. The experiment[24] shows that the f_0 and $f_\perp$ values are comparable with μ/q_0. At $T < T_c$ and $0 < E < E_c$ the finite difference $f_0 - f_\perp$ can cause the periodical perturbation of the polar angle, the period being equal to the pitch of a helix. At $|T - T_c| \ll T_c$ this perturbation must be small because $(f_0 - f_\perp)/f_0 \sim \theta_0^2 \ll 1$ at these temperatures. At $T < T_c$ and $E < E_c$ the non-monotonous dependence $\langle P \rangle(E)$ can take place, the E_c value corresponding to the total unwinding of a helix. The considered examples of MS in dielectric liquid crystals show the large variety of possible phenomena, conditions of their observation and their applications in diffraction lattices with managed period.

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