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► **To cite this version:**

Ph. Martinot-Lagarde, Geoffroy Durand. Flexoelectric effects in ferroelectric liquid crystals?. Journal de Physique Lettres, 1980, 41 (2), pp.43-45. 10.1051/jphyslet:0198000410204300 . jpa-00231717

HAL Id: jpa-00231717

<https://hal.science/jpa-00231717>

Submitted on 4 Feb 2008

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Classification
 Physics Abstracts
 61.30; 77.80; 77.20

Flexoelectric effects in ferroelectric liquid crystals ?

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(Reçu le 15 octobre 1979, révisé et accepté le 30 novembre 1979)

Résumé. — Nous montrons que la contribution flexoélectrique à la polarisation spontanée électrique des cristaux liquides smectique C* ne joue aucun rôle dans la constante diélectrique de la phase ferroélectrique. Les expériences récentes qui concluaient à une forte contribution flexoélectrique à partir de l'analyse de la constante diélectrique doivent être réinterprétées.

Abstract. — We show that the flexoelectric part of the spontaneous polarization in chiral smectic C* liquid crystal does not contribute at all to the static dielectric constant in the ferroelectric phase. Recent experimental data which concluded to a large flexoelectric contribution from dielectric constant analysis must be reinterpreted.

Meyer [1] has shown that chiral smectic C* liquid crystals are in general ferroelectrics. In this C* phase, the molecules are arranged in layers. They are tilted inside these layers, which pile on each other to built an helical texture. The spontaneous polarization \mathbf{P} , by symmetry, is inside the layers and perpendicular to the molecules. P is the sum of two contributions, one proportional to the molecular tilt (the *piezo* contribution) and one from a possible *flexoelectric* effect, analogous to the one described in nematics, because the molecules spiral around the helical axis along lines of constant bend. It is a problem to balance the flexo and piezo contributions to P . From dielectric measurements, the Russian group [2], using a simple model [3], concluded to the predominance of the flexoelectric contribution to P with a very large flexoelectric effect. A recent calculation from the Yugoslav group [4],

completed by a computer estimate [5], predicts a temperature dependence of the dielectric constant characteristic of the flexoelectric contribution. Recent measurements from this group [6] seem to exhibit this temperature dependence. In this letter, summing up the same model than ref. [4], we show analytically that the flexoelectric contribution to the dielectric constant is in fact exactly zero in the ferroelectric chiral smectic C* phase.

Let us compute the dielectric constant of a C*, when the temperature T is close to the smectic A \rightarrow C* phase transition temperature T_c . We apply a small DC electric field along OX , normal to the helical axis OZ . Taking the notations of ref. [4] for the coefficients, we write the increase of free energy density due to the molecular tilt θ and the polarization P in a convenient complex form, as :

$$\Delta g = \frac{1}{2} a \theta \theta^* + \frac{1}{4} b (\theta \theta^*)^2 + j \frac{A}{2} \left(\theta \frac{\partial \theta^*}{\partial z} - \theta^* \frac{\partial \theta}{\partial z} \right) + \frac{K_{33}}{2} \frac{\partial \theta}{\partial z} \frac{\partial \theta^*}{\partial z} + \frac{1}{2\chi} P P^* - \frac{\mu}{2} \left(P \frac{\partial \theta^*}{\partial z} + P^* \frac{\partial \theta}{\partial z} \right) + j \frac{C}{2} (P \theta^* - P^* \theta) - \frac{1}{2} (P + P^*) E. \quad (1)$$

Here, the complex θ is equal to the combination $\xi_1 + j\xi_2$ of ref. [4], i.e. $\theta = |\theta| e^{i\varphi}$, where $|\theta|$ is the (assumed small) projection of a molecular *director*

on the layer plane, and φ is the azimuthal angle of the molecule around OZ . In the same way, P is

$$P_x + iP_y \cdot a = \alpha(T - T^*)$$

is the usual tilt elastic constant which drives the ferroelastic A \rightarrow C phase transition. b is the associated

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saturation term in a Landau type expansion. A is the chirality source, K_{33} is a bend nematic curvature elastic constant. χ corresponds to ε of ref. [4], μ is the flexoelectric term. C characterizes the coupling between polarization and tilt. Minimizing Δg versus P^* , we obtain :

$$P = \chi \left[\mu \frac{\partial \theta}{\partial z} + jC\theta + E \right]. \quad (2)$$

Further minimization versus θ^* results in :

$$\frac{\partial^2 \theta}{\partial z^2} [K_{33} - \chi\mu^2] - 2j[\chi\mu C - A] \times \frac{\partial \theta}{\partial z} + [\chi C^2 - a - b\theta\theta^*] \theta = jC\chi E. \quad (3)$$

As previously described, we resume in absence of field the renormalization of curvature elastic constant $K = K_{33} - \chi\mu^2$ due to the flexo effect, and the existence of a constant helical twist $q = \left(\frac{\chi\mu C - A}{K_{33} - \chi\mu^2} \right)$ for which the transition temperature

$$T_c [T_c = T^* + \alpha^{-1}(Kq^2 + \chi C^2)]$$

is maximum. For a given temperature, θ is given by

$$\theta = \theta_q = \left(\frac{\alpha(T_c - T)}{b} \right)^{1/2} e^{i(qz + \varphi_0)},$$

φ_0 being arbitrary. In absence of field E , equation (2) gives $P = P_q = j\chi(\mu q + C) \cdot \theta_q$, showing clearly the two flexo and piezo contributions. E being uniform we are just interested in the uniform P_0 response to the field E . Equation (2) being linear in E , θ and P , P_0 can only be related to the uniform texture distortion θ_0 .

Then, the flexoelectric contribution $\mu \frac{\partial \theta_0}{\partial z}$ to P_0 is exactly zero. There is no flexo contribution to the dielectric constant.

At this point, it is simple to give the analytical solution to the dielectric constant calculation of ref. [4]. Equation (3) is more compactly written in a frame twisting around Oz at the spatial frequency q . We define $\psi_0 = \theta e^{-jqz}$. In this frame, any component θ_k corresponds to ψ_{k-q} i.e. the undisturbed texture appears uniform. Assuming the existence of two modes ψ_k and ψ_l , the saturation term generates additional frequencies to k and l . In the case of interest, with ψ_0 and a much smaller $\psi_{-q}(E)$, we can keep only the two coupled modes ψ_q and ψ_{-q} which follow the equations :

$$\begin{aligned} \psi_{-q}(Kq^2 + b\psi_0\psi_0^*) + b\psi_0\psi_0^*\psi_q^* &= -jC\chi E_{-q} \\ \psi_q^*(Kq^2 + b\psi_0\psi_0^*) + b\psi_0\psi_0^*\psi_{-q} &= 0. \end{aligned}$$

The two eigen modes $\psi_{\pm} \equiv \psi_{-q} \pm \psi_q^*$ are given by :

$$\begin{aligned} \psi_+ &= \frac{-jC\chi}{Kq^2 + 2b\psi_0\psi_0^*} E_{-q} \\ \psi_- &= \frac{-jC\chi}{Kq^2} E_{-q}. \end{aligned}$$

Reverting to the laboratory frame, we identify ψ_+ with the *soft mode* (distorsion of $|\theta|$), and ψ_- to the *Goldstone mode* (distorsion of φ). The induced polarization along Ox can be written as :

$$P = \chi E \left[1 + \frac{1}{2} \left(\frac{C^2\chi}{Kq^2} + \frac{C^2\chi}{Kq^2 + 2\alpha(T_c - T)} \right) \right].$$

The additional contribution to the transverse dielectric constant in the smectic C* phase is :

$$\Delta\varepsilon_{C^*} = 2\pi \left(\frac{C^2\chi^2}{Kq^2} + \frac{C^2\chi^2}{Kq^2 + 2\alpha(T_c - T)} \right).$$

For $T = T_c$, the two contributions from the soft mode and the Goldstone mode are equals. In the A phase, ψ_0 is zero, there is only one mode ψ coupled to the field as :

$$\psi_{-q}[Kq^2 + \alpha(T - T_c)] = -jC\chi E_{-q}$$

where we neglect the high order terms. ψ_q being zero, the two soft and Goldstone modes ψ_{\pm} degenerate into a two dimensional soft mode which contributes to the dielectric constant as :

$$\Delta\varepsilon_A = 4\pi \frac{C^2\chi^2}{Kq^2 + \alpha(T - T_c)}.$$

As expected, $\Delta\varepsilon_A$ equals $\Delta\varepsilon_{C^*}$ at $T = T_c$. As previously predicted by the Israeli group [7], there is no critical divergence of $\Delta\varepsilon$.

It is not too complicated to extend this calculation to the case of a time dependent electric field excitation at frequency ω . We need just to introduce a dissipation function, the more general form of which compatible with the symmetry of our system is :

$$\frac{1}{2\Gamma_1} \dot{\theta}\dot{\theta}^* + \frac{1}{2\Gamma_2} \dot{P}\dot{P}^* + j\frac{1}{2\Gamma_3} (\dot{P}\dot{\theta}^* - \dot{P}^*\dot{\theta})$$

where Γ_1 and Γ_2 and Γ_3 are usual kinetic coefficients. Note that in ref. [4], the last term Γ_3 was not considered. We see the appearance of three characteristic frequencies : a Γ_1 , Γ_2/χ and $C\Gamma_3$, describing respectively, the relaxation of the tilt, of the electric polarization and of the piezo (or electroclinic) effect. Each mode has a complicated frequency dependence, superposition of the three mentioned Lorentzians relaxations, which will be discussed in a forthcoming paper.

We have just to reinterpret the existing experimental data, which seem to demonstrate a large

flexoelectric effect in $\Delta\epsilon$. The analysis of the Russian group was based on an over-simplified expression [3] for $\Delta\epsilon$. Taking our expression for $\Delta\epsilon_{C^*}$, we can explain their observed temperature dependence by the temperature dependence of the damping frequency of the helix (through $q(T)$). The computer analysis of ref. [5] is probably incorrect if it has been done at constant E field. The very large assumed flexo coefficient

$$(\epsilon\mu^2 = 0.8 K_{33})$$

implies a factor 5 decrease for the renormalized curvature elastic constant K . Close to T_c , the field E could be large enough to start unwinding the helix. Note that even for $\mu = 0$, we do not agree with the calculation of ref. [5], where one does not find equal amplitudes for the Goldstone and the soft mode contributions at T_c . On the other hand, our expression explains correctly the widely observed [8, 9, 10] temperature variation of $\Delta\epsilon$ in the C^* phase, by just

the temperature dependence of the predominant Goldstone mode (through $q(T)$).

In conclusion, we have derived simple expressions for the transverse dielectric constant of a chiral smectic C^* close to a smectic A transition. We have shown that the flexoelectric contribution to the spontaneous polarization, gives no contribution at all to the dielectric constant of the ferroelectric C^* phase. The flexoelectric contribution, if any, should be looked upon with experiments measuring P directly, while keeping the twisted helical texture. Experimental observations [11] seem to indicate that the flexo contribution may be of the order of (or less than) 10% of the piezo contribution, at least for the most usual compounds, in agreement with the usual values of the flexoelectric coupling in nematics

$$(\mu \sim 10^{-5} \text{ cgs}).$$

Further experiments would be useful to clarify this point.

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