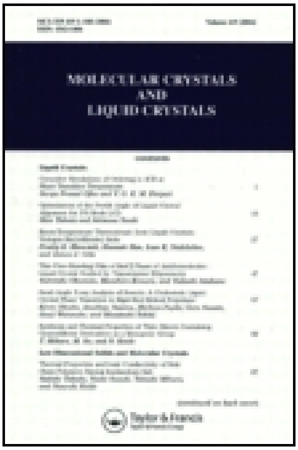

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Flexoelectricity in Nematics : Continuous Distortions and Instabilities

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FLEXOELECTRICITY IN NEMATICS : CONTINUOUS DISTORTIONS AND INSTABILITIES

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Abstract We review recent experiments mostly performed by the Orsay Group, which allow the independent measurement of the bulk flexoelectric and electric quadrupolar constants for nematic liquid crystals. Most of them use the hybrid align geometry, where a splay bend distorted nematic behaves as a liquid ferroelectric domain.

In this review, we give a survey of recent work done essentially in Orsay in the field of flexoelectricity of nematic liquid crystals. For the sake of completeness, we first recall a few elementary points about flexoelectricity; we then present results on continuous texture distortions, and our more recent unpublished work on flexoelectrically induced texture instabilities in nematics.

A - WHAT IS FLEXOELECTRICITY IN NEMATICS ?

It is known for a long time that an uniformly oriented nematic texture possesses inversion symmetry, i.e. is not ferroelectric. As shown by Meyer[1], one can break this inversion symmetry by imposing to the mean molecular orientation (the unitary director $\vec{n}$) a splay or bend curvature distortion. Per unit volume, one expects an electric pola-

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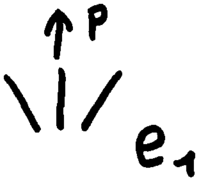
rization $\vec{P}$ to occur, equal to : $\vec{P} = e_1\ \vec{n}(\mathrm{div}\ \vec{n}) + e_3\ \mathrm{curl}\ \vec{n} \times \vec{n}$. The two coefficients, e_1 and e_3, of the order of a molecular polarization (10^{-18} cgs) divided by m^2 (molecular dimension) are of the order of $10^{-18}/10^{-14} \sim 10^{-4}$ cgs. The flexopolarization is the nematic equivalent of the piezoelectric polarization observed in strained solids.



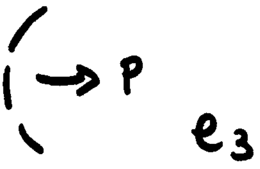


Fig. 1

The interest of flexoelectricity is to provide a linear coupling with an external electric field E in the form $-\vec{E}.\vec{P}$, where P can be proportional to some imposed distortion. By comparison, when $\vec{P} = \chi\vec{E}$ is induced by $\vec{E}$, one obtains a quadratic coupling $- \chi E^2$. The first experiment, discussed by Helfrich^2 and many others, to demonstrate the inverse flexo effect, consisted to induce (Fig.2) a bend distortion in an homeotropic sample. Balancing electric an mechanic terms in the free energy, results in $EP \sim \frac{K}{d^2}\ \theta^2$, where K is the curvature elastic constant, d the sample thickness, θ the surface angle. This results in $\theta \sim E\ \frac{ed}{K}$. In this description, we have neglected the surface anchoring energy, assuming the anchoring infinitely weak. When the anchoring is strong (molecular orientation fixed at the surface), there is an exact compensation in the electric energy $-\overline{P}.\overline{E}$ between the two differently curved parts of molecular lines

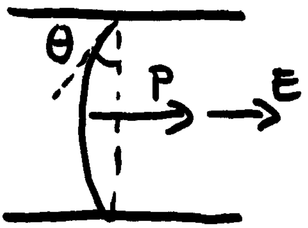


Fig. 2

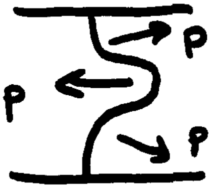


Fig.3

(Fig.3) and the solution is $\theta = 0$. One sees easely why :
a) assume $e_1 = e_3 = e$. $\vec{P}$ can be written as $\vec{P} = e\,\mathrm{div}(\bar{n}\,\bar{n})$, i.e. the divergence of the electric quadrupolar momentum density $\overline{\overline{Q}} = -e\,(\bar{n}\,\bar{n})$. The electric energy density can be written as : $-\bar{E}.\bar{P} = -[\nabla.\bar{E}(\bar{n}\,\bar{n}) - e\,\bar{n}\,\bar{n} : \bar{\nabla}\,\bar{E}]$.
When integrated in the volume, the first term gives a surface term which is zero for strong anchoring, and one is left with a pure quadrupolar effect, coupling of $\overline{\overline{Q}}$ with the field gradient, as shown by Prost and Marcerou[3]. For uniform $\vec{E}$ field this term vanishes also and there is no volume flexoelectric effect.
b) assume $e_1 \neq e_3$. For small distortions, $\vec{P}$ remains a divergence and can always be integrated out[4]. The quadrupolar effect is now proportionnal to $e_1 + e_3$. For large distortions only, one expects to observe with strong anchoring a pure volume flexoelectric effect, proportionnal to $e^* = e_1 - e_3$.
To conclude, with strong anchoring, the "flexoelectric" polarization gives rise to two kinds of effects :

- a pure volume flexoelectric effect, with an effective polarization $\vec{P}_{eff} = e^*\vec{n}(\mathrm{div}\,\vec{n})$, $(e^* = e_1 - e_3)$ which couples with $\vec{E}$.

- a pure volume quadrupolar effect, where the electric quadrupole density $\overline{\overline{Q}} = -(e_1 + e_3)\,(\bar{n}\,\bar{n})$ couples with the field gradient $\bar{\nabla}\,\bar{E}$.

B - CONTINUOUS DISTORTIONS

One can measure independently e_1+e_3 and e_1+e_3 , by observing the texture distortions induced in a nematic liquid crystal by an electric field gradient, or a field alone.

I) Quadrupolar effect

Prost and Marcerou[5] have observed by light scattering the distortion induced by the coupling of a E field gradient in an uniform homeotropic aligned sample. This geometry has been recently resumed by Dozov et al[6] , who wanted to measure the quadrupolar coefficient $e_1 + e_3$ and its sign in MBBA. The cell geometry is sketched on Fig.4. The electric torque density, on one central molecule, is estimated as $q\,\frac{E\ m}{m^3}$, where q is the assumed end charge giving rise to $\overline{\overline{Q}}$, and m is a molecular dimension. E itself is a pure gradient, and can be written as $E = \nabla E.\ m$. The resulting torque density is now $\nabla E\ \frac{qm^2}{m^3} = \nabla E\ Q$, to be compensated by the elastic torque density $K\theta/d^2$ where θ is the small induced tilt. ∇E is of the order of $V/d\ell$, where V is the voltage difference between the electrodes, d the sample thickness and ℓ the electrode separation.The resulting tilt is : $\theta = \frac{Q\ V}{K}\ \frac{d}{\ell}$. For MBBA, the conoscopic observation of θ gives : $e_1 + e_3 = -1\ 10^{-4}$ cgs, a very strange result, for its sign, opposed to what is known[4] in BBMBA and to the previous estimate of Derjhanski[7] et al, and for its absolute value, 7 times smaller than measured[3] by Prost and Marcerou. Screening effect may be invoqued to explain this discrepancy.

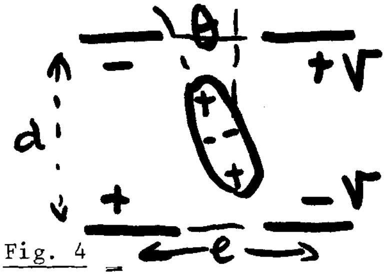


Fig. 4

II) Flexoelectric effect

The existence of $\vec{P}_{eff}$ has been demonstrated by Dozov et al[8] using a hybrid aligned cell (HAC) to obtain the large distortion (rotation of $\pi/2$, see Fig.5) necessary to the existence of the pure volume flexoelectric effect. A DC field E is applied perpendicular to the splay bend texture. The resulting torque density $\vec{P}_{eff} \times \vec{E}$ induces a twist ϕ of the splay bend molecular lines. Writting $P_{eff} \sim e^*/d$, with the same elastic torque density $\frac{K\phi}{d^2}$, one expects : $\phi \sim \frac{e^*}{K}\, dE$. This is well verified experimentally. The two splay bend domains rotate in opposite direction, as expected from the sign of P_{eff}. ϕ is found $\sim$ E, for d = 100 µm, which results in $e^* \sim 10^{-4}$ cgs for MBBA. The twist ϕ is optically observed by the rotation of the light polarization propagating across the HAC. This method, applied to other materials, has given interesting results. For instance, in 8CB, e^* is found + 1.10^{-4} cgs, although in 8OCB which differs only by the existence of an oxygen, e^* is $-6\ 10^{-4}$ cgs. This change has been interpreta-

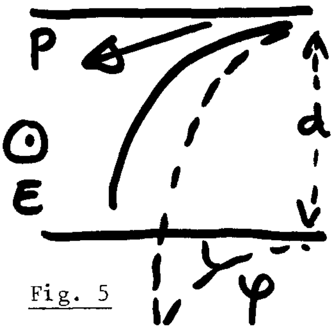


Fig. 5

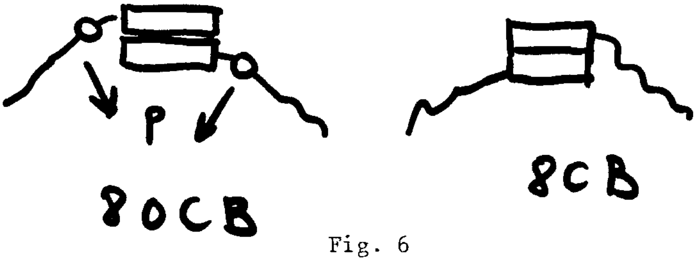


Fig. 6

ted as "conformational" flexoelectricity[9] . The strong dipoles of the CN groups seem to cancel each other, by some molecular pairing. In presence of a bend, such a molecular pair would see a prefered configuration of the end chains which would tend to align the oxygen dipoles toward the center of curvature (see Fig.6).

C - FLEXOELECTRICALLY INDUCED INSTABILITIES

We now report on two texture instabilities, using the quadrupolar or the flexoelectric coupling, discovered recently in Orsay.

I/ Uniform twist instability of the HAC (Flexoelectric coupling)

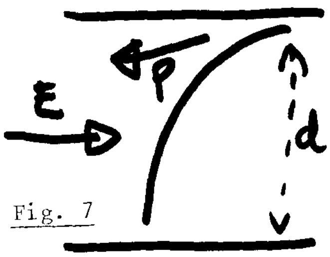


Fig. 7

We can demonstrate the existence of $\vec{P}_{eff}$ in a HAC, by applying a DC electric field $\vec{E}$ opposed to $\vec{P}_{eff}$. Above a certain threshold E_{th}, a twist ϕ becomes unstable, because $\vec{P}_{eff}$ tends to align along $\vec{E}$. E_{th} is given classically by the balance of torques equation:

$$E_{th}\ P\phi \sim \frac{K}{d^2}\ \phi, \text{ i.e. } \quad E_{th} \sim \frac{K}{e^* d}$$

We do observe[10] such an instability, for $d \sim 50$ µm, at $E_{th} \sim 1$ cgs, i.e. for the expected value calculated with e^* determined in the continuous texture twist experiment. The influence of the dielectric anisotropy must anyway be considered for a quantitative fit of the data. For larger fields, electrohydrodynamic instabilities can develop, as always, and limit the range of observable twist.

II/ Non uniform twist instability of the HAC (Quadrupolar coupling)

We resume the HAC geometry of Fig.5 where we have observed the continuous twist of the splay bend texture which behaves as a ferroelectric domain rotating under the action of a perpendicular field. We have observed[11] , superimposed to the average twist ϕ_0 a sharply localized twist ϕ which appears between crossed polarizers as a "wall" parallel to the electrodes, of lateral extension comparable to the thickness d. This wall propagates from the cathode to the anode, with a velocity linear in field and a resulting mobility in the 10^{-4} cm/Vs range, comparable to the one of ions.

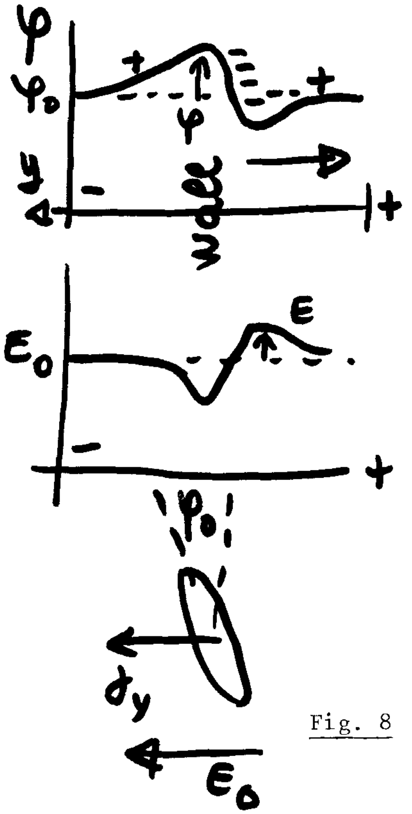


Fig. 8

With blocking electrodes, we check that these walls can only exist in presence of DC current. They stop if we turn the field off, they start moving again if we turn the field on. They vanish when reversing the field. We understand this wall as a localized charged twist of the texture, created by the following mechanism :

a) a localized fluctuation ϕ of the twist ϕ_0, because of the anisotropy of conductivity σ_a, gives rise to a localized space charge density ρ. Calling y the $\vec{E}_0$ field orientation (Fig.8), the current density writes as : $j_y = \sigma E_o - \sigma_a \frac{\cos 2\phi_o}{2} E_o$ which results in a stationnary space charge density

$$\rho = -\frac{\varepsilon}{4\pi\sigma}\ \sigma_a \sin 2\phi_o \frac{d\phi}{dy} E_o$$ (ε is the assumed isotropic dielectric constant, σ the isotropic conductivity)

b) these localized space charge ρ gives rise to an additionnal field E and an additional field gradient ∇E. The additionnal E is calculated by : $\frac{dE}{dy} = 4\pi\rho$ which gives :

$E = -E_o \frac{\varepsilon}{\sigma} \sigma_a \sin 2\phi_o$; because σ_a is positive for most nematic materials, E is opposed to ϕ. The additionnal field has a stabilizing effect, through the flexoelectric coupling. The resulting torque $\vec{P}_{eff} \times \vec{E}$ is smaller were ϕ is larger, and vice versa. On the other hand, the additionnal field gradient ∇E may be destabilizing, through the quadrupolar coupling, if $e_1 + e_3 > 0$, (see Fig. 9) and an instability occurs. The threshold $E_{o_{th}}$ for the wall formation can be estimated by the following argument : just above threshold, the elastic and quadrupolar torques are almost balanced :

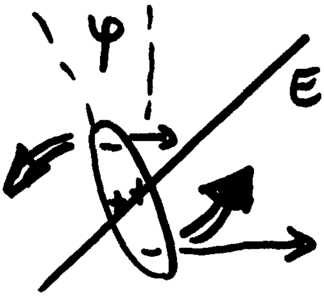


Fig. 9

$$\frac{K\phi}{d^2} + (e_1 + e_3) \sin 2\phi_o \frac{E}{d} = 0$$

where E/d stands for ∇E. This relationship between ϕ and E must be compatible with the field equation

$$\varepsilon \frac{\sigma_a}{\sigma} \sin 2\phi_o\ E_o\phi + E = 0$$

This is possible for $E_{o_{th}} \sim \dfrac{E_1}{\sin^2 2\phi_o}$

with $E_1 \sim \dfrac{K\,\varepsilon}{(e_1 + e_3)d}\,\dfrac{\sigma}{\sigma_a} \sim 0.5$ cgs.

Remember now that the initial twist ϕ_o and the field E_o are already related by the equation :

$$E_o = \frac{\phi_o}{\cos\phi_o}\,E' \quad \text{where} \quad E' = \frac{K}{e^* d}$$

as seen previously for the flexoelectric induced continuous twist experiment. As shown on Fig.10, the wall can only appear in a range of finite ϕ_o , well above zero. The "wall" can be considered as a negatively charged twisted "soliton". Because of charge injection, the threshold is more probably easely reached close to the cathode, as observed. The only disturbing point is the necessary positive sign for $e_1 + e_3$ in MBBA. Further critical experiments are necessary to be sure of this sign.

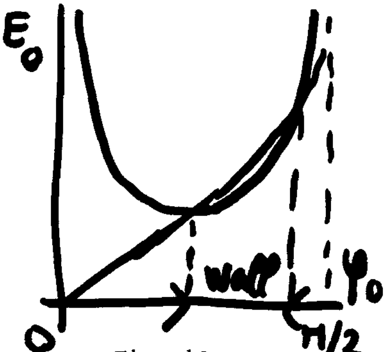


Fig. 10

D - CONCLUSION

Assuming strong anchoring to avoid surface effects, we have demonstrated simple experiments which demonstrate independently the volume flexoelectric effect and the electric quadrupolar effect. These experiments allow the independent measurement of the two associated constants $e^* = e_1 - e_3$ and $e_1 + e_3$, for the same material. In principle the best measurements are obtained with the continuous texture distortion experiments, but screening effects may perturb the results, specially when creating a field gradient. The new soliton-like charged twist wall is in itself a interesting subject of investigation. The hybrid aligned cell appears to be an interesting system to study nematic liquid crystals.

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