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## SHEAR ELECTRICITY IN CHOLESTERIC

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**Résumé.** — Des considérations de symétrie montrent qu'un cristal liquide cholestérique soumis à l'action d'un gradient de vitesse, doit être le siège d'une polarisation électrique. La flexoélectricité donne une explication à ce phénomène.

**Abstract.** — From symmetry considerations it is possible to deduce that velocity gradients ought to induce electric polarizations in cholesteric liquid crystals. We show that flexoelectricity can be responsible for such a phenomenon.

**1. Introduction.** — In the hydrodynamic regime, cholesteric liquid crystals exhibit the same features as smectics A (composed of chiral molecules), that is the translational symmetry is broken in one direction of space, and the local point group relevant to the problem is  $C_\infty$  [1, 2]. One novel consequence of these considerations is that by shearing a cholesteric liquid crystal in certain ways, it is possible to induce an electric polarization in definite directions of space. To see this, one wants to write a relation such as :

$$P_i = \lambda_{ijk} \partial_k V_j \quad (1.1)$$

where  $P_i$  is the polarization density,  $V_j$  the barycentric velocity of the fluid, and  $\lambda_{ijk}$  is a third rank tensor. In a cartesian reference frame, and if  $z$  is chosen to be parallel to the helix axis, the non zero components of the tensor are :

$$\begin{cases} \lambda_{xyz} = -\lambda_{yxz} , \\ \lambda_{xzy} = -\lambda_{yzx} , \\ \lambda_{zxy} = -\lambda_{zyx} . \end{cases}$$

This coupling differs from piezoelectricity in the fact that it is not an equilibrium process : the fluid nature of cholesterics and smectics requires the use of velocity gradients rather than strains.

The polarization density resulting from a shear has already been observed in smectics A built up of chiral molecules (3). The phenomenon was interpreted [3, 4] as a pretransitional effect within the framework of a Landau theory of the  $S_A$  to the  $S_C$  (chiral) transition. More precisely the velocity gradient induces a tilt of the molecules within each layer, and the polarization results from this tilt.

In cholesteric phases, the situation is different and we will show in the second section that flexoelectricity may be used to understand the occurrence of such a phenomenon. In the third section, we give the detailed calculation of the hydrodynamic motions linked to the shear and justify the equations used in section 2. We conclude by giving order of magnitude considerations which suggests that this effect should be observable experimentally. A shear induced polarization has indeed been previously reported [5] but the geometry used in this experiment is in principle inconsistent with the symmetry considerations developed above.

**2. The link with flexoelectricity.** — In the first section, we considered the properties of a cholesteric in the hydrodynamic limit (that is on a large scale compared to the periodicity of the structure). We propose here a *microscopic* calculation of  $\lambda_{ijk}$ , considering that a cholesteric behaves locally like a twisted nematic (6, 2). It is known from nematic dynamics [2] that velocity gradients exert a torque on the director, which depends strongly on the very orientation of this director with respect to the shear. A homogeneous shear should then induce a tilt of the molecules (out of the cholesteric planes) modulated with the periodicity of the structure (Fig. 1). This tilt involves both a splay and a bend of the structure, but since these two vectors are even functions of the director field, they have non-zero macroscopic (that is averaged over a large number of periods) values. In turn, a mean electric polarization results from the flexoelectric phenomenon.

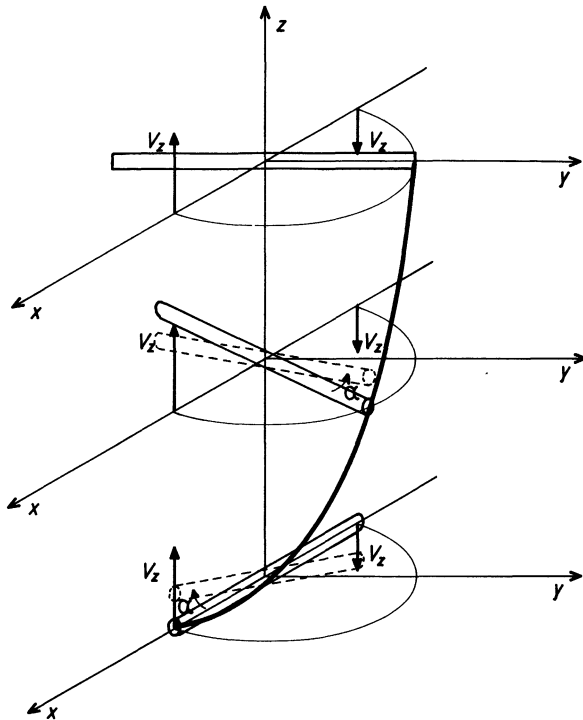


FIG. 1. — Solid batonnets : unperturbed configuration of the optical axis. Dashed batonnets : perturbed orientation due to the hydrodynamic torque exerted by  $\partial_x V_z^e$ . Note that the tilt angle  $\alpha$  is maximum when the optical axis lies along  $x$  and zero when along  $y$  : it is modulated at the spatial frequency of the structure. The resulting splay  $d^0 \frac{\partial \alpha}{\partial z}$  and bend  $\alpha \frac{\partial d^0}{\partial z}$  exhibit a constant component along  $y$ , which is the source of the induced polarization *via* the flexoelectric coupling.

Let us describe the cholesteric with :

$$\begin{aligned} \text{unperturbed helix} & \begin{cases} d_x^0 = \cos q_0 z, \\ d_y^0 = \sin q_0 z, \\ d_z^0 = 0, \end{cases} \\ \text{perturbed helix} & \begin{cases} d_x = \cos(q_0 z + \psi), \\ d_y = \sin(q_0 z + \psi), \\ d_z = \alpha. \end{cases} \end{aligned}$$

The deformations from the ground state are expressed by the angles  $\Psi$  and  $\alpha$ ;  $\Psi$ , although entering the problem is not very interesting since it contributes only to the twist deformation. The tilt out of the planes  $\alpha$ , involves both splay and bend deformations.

In the hydrodynamic limit, the dependences on  $x$  and  $y$  due to boundary conditions may be neglected, and the only important variable is  $z$  ( $\partial_x = \partial_y = 0$ ).

If a shear flow  $\partial_x V_z^e$  is imposed on the liquid crystal, a tilt is induced according to the relation :

$$\gamma_1^s \frac{\partial \alpha}{\partial t} + K_{33} q_0^2 \alpha - K_{11} \partial_{zz}^2 \alpha = \tilde{\gamma} \cos(q_0 z) \partial_x V_z^e. \quad (2.2)$$

The detailed derivation of this equation and the detailed expression of  $\gamma_1^s$  and  $\tilde{\gamma}$  will be given in the

next section. The left hand side expresses the response of the director, and the right hand side the torque exerted by the shear flow. The latter is obviously periodic in space, as previously claimed and illustrated in figure 1.

Furthermore, the shear is obtained by moving the glass plates defining the sample boundaries and it is convenient to impose a motion periodic in time, so that :

$$\partial_x V_z^e(t) = (\partial_x V_z^e) e^{j\omega t} \quad (2.3)$$

which leads to :

$$\alpha = \chi (\partial_x V_z^e) \cos(q_0 z) \quad (2.4)$$

$$\left| \chi = - \frac{\alpha_2}{(K_{33} + K_{11}) q_0^2 + q \gamma_1 \omega} \right. \quad \left. \text{if one assumes } \alpha_3 = 0 \right. \quad (2.5)$$

The resulting splay and bend vectors are :

$$\mathbf{S} \begin{cases} S_x = d_x \partial_z d_z \\ S_y = d_y \partial_z d_z \\ S_z = d_z \partial_z d_z \end{cases} \quad \mathbf{b} \begin{cases} b_x = d_z \partial_z d_x \\ b_y = d_z \partial_z d_y \\ b_z = d_z \partial_z d_z \end{cases} \quad (2.6)$$

To first order in  $\partial_x V_z^e$

$$\left. \begin{aligned} S_x &= \frac{1}{2} q_0 \chi \sin(2 q_0 z) \partial_x V_z^e \\ S_y &= -q_0 \chi \sin^2(q_0 z) \partial_x V_z^e \\ S_z &= 0 \\ b_x &= \frac{1}{2} q_0 \chi \sin(2 q_0 z) \partial_x V_z^e \\ b_y &= +q_0 \chi \cos^2(q_0 z) \partial_x V_z^e \\ b_z &= 0. \end{aligned} \right\} \quad (2.7)$$

And eventually :

$$\left. \begin{aligned} \langle S_x \rangle &= \langle b_x \rangle = \langle S_z \rangle = \langle b_z \rangle = 0 \\ \langle -S_y \rangle &= \langle b_y \rangle = \frac{q_0 \chi}{2} \partial_x V_z^e \end{aligned} \right\} \quad (2.8)$$

The flexoelectric polarization is given by [7] :

$$\mathbf{P} = f_{\parallel} \mathbf{d} \text{Div } \mathbf{d} + f_{\perp} (\mathbf{d} \cdot \nabla) \mathbf{d} \quad (2.9)$$

(we use notations defined in [8]) which gives :

$$\begin{aligned} P_x &= P_z = 0 \\ P_y &= q_0 \chi \frac{f_{\perp} - f_{\parallel}}{2} \partial_x V_z^e. \end{aligned} \quad (2.10)$$

Thus we find :

$$\lambda_{zxx} = \lambda_{zzx} = 0; \quad \lambda_{yzx} = q_0 \chi \frac{f_{\perp} - f_{\parallel}}{2}. \quad (2.11)$$

This polarization is fundamentally different from the one observed with chiral smectics C (3) in similar

experiments : this latter one results from the modulation of the  $\psi$  angle, and exists only to the extent that there is a macroscopic polarization in the ground state of those materials. In particular, it vanishes when the director is perpendicular to the helical axis, which is the definition of cholesterics.

The next point is the occurrence of the difference ( $f_{\perp} - f_{\parallel}$ ) in (2.22); it has been argued in [9], that there was only one flexoelectric coefficient ( $f_{\perp} + f_{\parallel}$ ) in nematics and smectics A, ( $f_{\perp} - f_{\parallel}$ ) being indistinguishable from surface polarization terms. However, the phenomenon we describe here is typically *bulk*, and cannot be confused with surface polarization effects (see appendix I). The contradiction between these two statements is only apparent : the proof proposed in [8] is valid to the extent that we deal with small deviations of the director with respect to a uniform equilibrium distribution. The cholesteric ground state does not obey the uniformity requirement, and the argument developed for nematics and smectics A is not valid here. This is interesting since it allows us to measure  $f_{\perp}$  and  $f_{\parallel}$  independently (for instance, starting from a nematic, one could measure ( $f_{\perp} + f_{\parallel}$ ) with the interdigital technique [9] and ( $f_{\perp} - f_{\parallel}$ ) using the technique we describe here with the same compound slightly doped with an optically active molecule).

The last remark is that whereas one expects and finds an S dependence for ( $f_{\perp} + f_{\parallel}$ ), an S<sup>2</sup> law is predicted for  $f_{\perp} - f_{\parallel}$  from equations (2.9) and (4.5) of reference [8] since it is essentially given by the Meyer dipolar contribution, and the anisotropic part of the quadrupolar contribution <sup>(1)</sup>.

**3. The hydrodynamic equations.** — A similar problem has already been investigated by Leslie [6], who was mainly interested in studying a (non linear) steady state solution ( $\partial \mathbf{d} / \partial t = 0$ ). We are here concerned with a linearized problem, in which the perturbation is a periodic function of time ( $\partial / \partial t = j\omega$ ); this allows us to find a closed solution to our problem, whereas in the general case this is not possible.

We have shown in section 1 that three independent geometries corresponding to the three externally

applied shear ( $\partial_x V_z^e, \partial_z V_x^e, \partial_x V_y^e$ ) have to be investigated. In the incompressible cholesteric, the variables are  $\psi, \alpha, V_x, V_y, V_z$  and  $P$  ( $\mathbf{V}$  is the barycentric velocity of the fluid,  $P$  the pressure and the coupling with the temperature is omitted). It must be noted again that boundary perturbations do not extend outside a region of linear dimensions of the order of ( $\Pi/q_0$ ) and hence in the hydrodynamic limit the only relevant parameter is  $z$ .

One needs to satisfy the momentum and mass conservation equations, and the director equation. The mass conservation (incompressible fluid) equation gives (3.1) :  $\partial_z V_z = 0$  (or  $V_z = 0$ ; except when the applied shear is  $\partial_x V_z^e$ , in which case  $\partial_z V_z = 0$  implies the absence of any counterflow in the  $z$  direction).

Furthermore, the  $z$  component of the momentum conservation equation provides the definition of pressure, so that we are left with four equations and four unknowns :

$$+ g'_z = k_{33} q_0^2 \alpha - k_{11} \partial_{zz}^2 \alpha \tag{3.2}$$

$$d_y^0 g'_x - d_x^0 g'_y = k_{22} \partial_{zz}^2 \psi \tag{3.3}$$

$$\partial_z \sigma'_{zx} = \partial_z \sigma'_{zy} = 0 \tag{3.4}$$

$g'_i$  is, in Leslie's notation, the dissipative part of the director body force. Its actual expression depends on the particular case under study. The last two equations (3.4), do not involve reactive terms which are second order (2) and inertial terms which are negligible. They may be integrated to yield :

$$\sigma'_{zx} = \frac{a}{2} \quad \sigma'_{zy} = \frac{b}{2} \tag{3.5}$$

$a$  and  $b$  are constant numbers independent of  $x, y$  and  $z$  (the factor  $\frac{1}{2}$  is included for convenience).

We now have a set of four coupled linear equations, and six unknown parameters.  $a$  and  $b$  have to be determined from boundary conditions. As we will see the relevant one in the hydrodynamic limit is that  $\mathbf{V}$  must remain finite as the size of the sample goes to infinity.

CASE *a.* —  $\partial_x V_z^e \neq 0$ .

The set of equations (3.2, 3, 5) exhibit the detailed form

$$\gamma_1 \frac{\partial \alpha}{\partial t} + K_{33} q_0^2 \alpha - k_{11} \partial_{zz}^2 \alpha = -\alpha_3 dx^0 \partial_z V_x - \alpha_3 d_y^0 \partial_z V_y - \alpha_2 d_x^0 \partial_x V_z^e \tag{3.6}$$

$$\gamma_1 \frac{\partial \psi}{\partial t} = k_{22} \partial_{zz}^2 \psi - \gamma_1 q_0 V_z^e \tag{3.7}$$

$$(\alpha_4 + (\alpha_3 + \alpha_6) d_x^{02}) \partial_z V_x + (\alpha_6 + \alpha_3) dx^0 dy^0 \partial_z V_y + (\alpha_4 + (\alpha_6 - \alpha_3) d_x^{02}) \partial_x V_z^e + 2 \alpha_3 d_x^0 \frac{\partial \alpha}{\partial t} = a \tag{3.8}$$

$$(\alpha_3 + \alpha_6) d_x^0 d_y^0 \partial_z V_x + (\alpha_4 + d_y^{02} (\alpha_6 + \alpha_3)) \partial_z V_y + (\alpha_6 - \alpha_3) d_x^0 d_y^0 \partial_x V_z^e + 2 \alpha_3 d_y^0 \frac{\partial \alpha}{\partial t} = b \tag{3.9}$$

(3.7) expresses the convection of  $\psi$  due to the velocity parallel to the helical axis.

<sup>(1)</sup> The anisotropy of the quadrupolar contribution, due to the Lorentz field corrections, has been pointed out to us by the Sofia group.

From (3.8) and (3.9) we find  $\partial_z V_x$  and  $\partial_z V_y$  as a function of  $\partial_x V_z^e$ ,  $\partial\alpha/\partial t$ ,  $a$  and  $b$ . Plugging them back into (3.6), we obtain :

$$\gamma_1^s \frac{\partial\alpha}{\partial t} + k_{33} q_0^2 \alpha - k_{11} \partial_{zz}^2 \alpha = \gamma d_x^0 \partial_x V_z^e - \frac{\alpha_3}{2\eta_b} (ad_x^0 + bd_y^0) \quad (3.10)$$

$$\gamma_1^s = \gamma_1 - \frac{2\alpha_3^2}{\alpha_3 + \alpha_4 + \alpha_6} = \gamma_1 - \frac{\alpha_3^2}{\eta_b} \text{ is the splay viscosity} \quad (3.11)$$

$$\gamma = -\alpha_2 + \frac{\alpha_3(\alpha_4 + \alpha_6 - \alpha_3)}{(\alpha_3 + \alpha_4 + \alpha_6)} = -\alpha_2 + \alpha_3 \frac{(\alpha_4 + \alpha_6 - \alpha_3)}{2\eta_b}. \quad (3.12)$$

$\eta_b$  is a Miesowicz viscosity (see for instance reference [2]).

Equation (3.10) shows that  $\alpha$  is a periodic function of period  $2\pi/q_0$ , and hence (with

$$(\partial_x V_z^e(t) = \partial_x V_z^e e^{j\omega t})$$

using this  $\alpha$  value in the expressions for  $\partial_z V_x$  and  $\partial_z V_y$ , one gets :

$$\begin{aligned} \partial_z V_x = (2\alpha_4 \eta_b)^{-1} & \left\{ -\partial_x V_z^e \alpha_4 \left( \alpha_4 + \alpha_6 + \alpha_3 (d_y^{02} - d_x^{02}) + \frac{2j\alpha_3 \gamma \omega d_x^{02}}{(K_{11} + K_{33}) q^2 + j\gamma_1^s \omega} \right) + \right. \\ & + a \left( \alpha_4 + (\alpha_3 + \alpha_6) d_y^{02} + j \frac{\alpha_4 \alpha_3^2 \omega d_x^{02}}{\eta_b [(K_{11} + K_{33}) q^2 + j\gamma_1^s \omega]} \right) \\ & \left. + b \left( \frac{j\alpha_3^2 \alpha_4 \omega}{\eta_b [(K_{11} + K_{33}) q^2 + j\gamma_1^s \omega]} - 1 \right) d_x^0 d_y^0 \right\} \quad (3.13) \end{aligned}$$

$$\begin{aligned} \partial_z V_y = (2\alpha_4 \eta_b)^{-1} & \left\{ 2\partial_x V_z^e d_x^0 d_y^0 \alpha_4 \alpha_3 \left( 1 - \frac{j\gamma\omega}{(K_{11} + K_{33}) q^2 + j\gamma_1^s \omega} \right) + \right. \\ & + ad_x^0 d_y^0 \left( \frac{j\alpha_4 \alpha_3^2 \omega}{\eta_b [(K_{11} + K_{33}) q^2 + j\gamma_1^s \omega]} - \alpha_6 - \alpha_3 \right) \\ & \left. + b \left( \alpha_4 + (\alpha_3 + \alpha_6) d_x^{02} + j \frac{\alpha_4 \alpha_3^2 \omega d_y^{02}}{\eta_b [(K_{11} + K_{33}) q^2 + j\gamma_1^s \omega]} \right) \right\} \quad (3.14) \end{aligned}$$

According to (3.13), the averaged value of  $\partial_z V_x$  and  $\partial_z V_y$  over a large number of periods is non zero ; this is an unphysical situation since a vanishingly small shear cannot induce an infinitely large velocity ( $\partial_z V_x = \text{Const.}$   $V_x = (\text{Const.}) z$ ).

The conditions  $\langle \partial_z V_x \rangle = \langle \partial_z V_y \rangle = 0$  allow us to calculate  $a$  and  $b$  :

$$\left. \begin{aligned} a &= \partial_x V_z^e \frac{\alpha_4 \left( \alpha_4 + \alpha_6 + \frac{j\alpha_3 \gamma \omega}{(K_{11} + K_{33}) q^2 + j\gamma_1^s \omega} \right)}{\alpha_4 + \frac{\alpha_3 + \alpha_6}{2} + \frac{j\alpha_4 \alpha_3^2 \omega}{2\eta_b [(K_{11} + K_{33}) q^2 + j\gamma_1^s \omega]}} \\ b &= 0 \end{aligned} \right\} \quad (3.15)$$

and eventually :

$$\alpha = \frac{\tilde{\gamma}_a d_x^0 \partial_x V_z^e}{(K_{11} + K_{33}) q_0^2 + j\gamma_1^s \omega} \quad (3.16)$$

with

$$\tilde{\gamma}_a = -\alpha_2 - \frac{\alpha_3}{2} \frac{\alpha_3 - \alpha_6 - j\omega \frac{\alpha_3 \alpha_4 \alpha_2}{\eta_b [(K_{11} + K_{33}) q_0^2 + j\gamma_1^s \omega]}}{\alpha_4 + \frac{\alpha_3 + \alpha_6}{2} + j \frac{\alpha_4 \alpha_3^2 \omega}{2\eta_b [(K_{11} + K_{33}) q_0^2 + j\gamma_1^s \omega]}}. \quad (3.17)$$

This is precisely the proof of equation (2.4).

CASE *b*. —  $\partial_z V_x^e \neq 0$ .

In this case  $V_z = 0$ ; on the other hand, a reaction velocity  $\delta V_x(z)$ ,  $V_y(z)$  modulated at the spatial frequency of the structure is still allowed. The problem then is formally identical to the former. One finds :

—  $\psi$  uncoupled to the shear.

$$\alpha = \frac{\tilde{\gamma}_b d_x^0 \partial_z V_x^e}{(K_{11} + K_{33}) q_0^2 + j\gamma_1^s \omega} \quad (3.18)$$

$$\tilde{\gamma}_b = \frac{\alpha_3 \alpha_4}{\alpha_4 + \frac{\alpha_6 + \alpha_3}{2} + \frac{j\omega\alpha_3^2}{2\eta_b[(K_{11} + K_{33}) q^2 + j\gamma_1^s]}} \quad (3.19)$$

CASE *c*. —  $\partial_x V_y^e \neq 0$ .

$\psi$  is coupled to the shear but not  $\alpha$  and no polarization effect is to be expected.

Using the results of section 2, one can summarize the results of section 3 in the following way :

$$\begin{aligned} \lambda_{yzx} = -\lambda_{xzy} &= q_0 \frac{f_{\perp} - f_{\parallel}}{2} \frac{\tilde{\gamma}_a}{(K_{11} + K_{33}) q_0^2 + j\gamma_1^s \omega} \\ \lambda_{yxz} = -\lambda_{xyx} &= q_0 \frac{f_{\perp} - f_{\parallel}}{2} \frac{\gamma_b}{(K_{11} + K_{33}) q_0^2 + j\gamma_1^s \omega} \\ \lambda_{zyx} = -\lambda_{xzy} &= 0. \end{aligned} \quad (3.20)$$

**Conclusion.** — This shear induced polarization effect will be of practical importance only to the extent that it is measurable. An experiment well suited to seek it has been developed by Pieranski, Guyon and Keller, to investigate the chiral smectic C phase (3). The geometry could correspond either to case a, or b (figure 2). The induced polarization gives rise

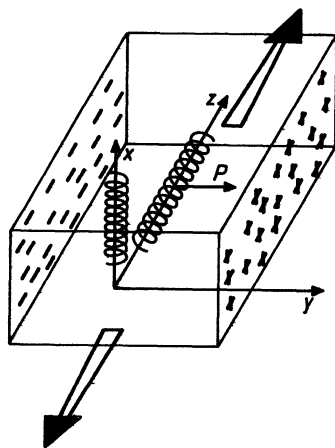


FIG. 2. — Parallelepiped cholesteric sample subjected to shear (the arrows represent the velocity of the sample holders). The two helices show the two directions of the cholesteric axis leading to a non-zero polarization. This induced volume polarization creates a surface charge at the sample boundary which is the measurable quantity.

to surface charges, which are directly measurable with non contact techniques (3).

$$Q = \int_s P_y dS = S \lambda_{yzz} \partial_x V_z$$

( $S$  = surface of the sample defined by  $y = P/2$ ) or for order of magnitude purposes ( $\alpha_3 \simeq 0$ ) :

$$Q \simeq \frac{q_0 \alpha_2 \partial_x V_z^e}{(K_{33} + K_{11}) q_0^2 + j\gamma_1 \omega} \frac{(f_{\perp} - f_{\parallel})}{2} S.$$

In the high frequency limit ( $\gamma_1 \omega \gg (K_{33} + K_{11}) q_0^2$  which seems appropriate to get large values of  $\partial_x V_z^e$ , and to avoid the  $1/f$  noise in detection systems) : with :

$$q_0 = 3 \times 10^4 \text{ cm}^{-1} \quad (\text{one micron pitch})$$

$$\frac{\partial_x V_z^e}{\omega} \simeq 10^{-2}$$

(one micron excursion of the holding plates, for a sample thickness of  $10^{-2}$  cm, as in reference [5])

$$\frac{f_{\perp} - f_{\parallel}}{2} \simeq 10^{-4} \text{ e s u cm}^{-1}$$

$$S \simeq 10^{-2} \text{ cm}^2$$

one gets :

$$Q \simeq 3 \times 10^{-3} \text{ e s u}.$$

Such a charge should be measurable since it corresponds to a 1 mV voltage on a one hundred picofarad capacitance, typical for this type of experimental arrangement.

Experiments in well controlled geometries are currently underway (E. Guyon, P. Pieranski and L. Petit private communication) and will provide the first evidence of the direct flexoelectric effect in conventional liquid crystals.

**Acknowledgments.** — It is a pleasure to acknowledge stimulating conversations with E. Guyon and to thank H. Gasparoux for his constant encouragement.

**Appendix I.** — The experiment described in this paper consists of detection of the surface charges resulting from the existence of the bulk polarization :

$$d\sigma^s = P_i \cdot dS_i \quad (A.1)$$

( $dS_i$  surface element pointing out of the sample).

The question is to know whether or not it is possible to obtain surface charges from surface polarizations.

Symmetry considerations allow us to write :

$$dP_i^s = \lambda_{ij} dS_j \quad (A.2)$$

$dP_i^s$  is the surface polarization,  $\lambda_{ij}$  has the symmetry properties of the bulk. For instance, quadrupolar densities  $\theta$  lead to such a relation with  $\lambda_{ij} = \frac{1}{3} \textcircled{H}_{ij}$ .

The corresponding surface charge, will arise from but  $\lambda_{zy} = \lambda_a d_z d_y$  and the divergence of (A.2) :

$$d\sigma_{ps}^s = - \partial_i(\lambda_{ij} dS_j) . \quad (\text{A.3})$$

$$d\sigma^s = - \lambda_a \chi q_0 |d_x^{02} - d_y^{02}| dS_y \quad (\text{A.5})$$

$$\langle d\sigma_{ps}^s \rangle = 0$$

More precisely in the hydrodynamic limit :

$$d\sigma_{ps}^s = - \partial_z | \lambda_{zy} | dS_y \quad (\text{A.4}) \quad \text{which is the point stated in section 2.}$$

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