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# ONE-DIMENSIONAL DIELECTRIC-FLEXOELECTRIC DEFORMATIONS IN NEMATIC LAYERS

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**Résumé.** — On a développé une théorie générale pour la description unitaire de différents effets flexoélectriques dans des champs électriques homogènes constants. Sur cette base on a étudié théoriquement quatre types de géométrie :

*a*) couche homéotrope sous l'action d'un champ électrique orienté normalement à l'axe optique ; *b*) couche homéotrope et champ électrique orienté parallèlement ; *c*) couche planaire et champ électrique orienté parallèlement ; *d*) couche planaire et champ électrique orienté normalement. En plus on a étudié les cas d'une anisotropie diélectrique positive et négative aussi bien que les cas d'égales et de non égales énergies d'ancrage. C'est-à-dire que 16 cas différents sont l'objet de cet article. Dans les cas homéotropes on a tenu compte de la polarisation de surface. Dans les cas *a*) et *c*) il n'existe pas de champ critique et les déformations commencent depuis le zéro. Le champ critique dans les cas *b*) et *d*) peut dépendre de la polarité. Cette polarité a déjà été observée expérimentalement dans une couche planaire de MBBA.

**Abstract.** — A general theoretical framework for a unified description of various flexoelectric effects in uniform d.c. electric fields is developed. With this framework four basic geometries are studied theoretically as follows :

*a*) homeotropic layer under the action of a d.c. electric field normal to the optic axis ; *b*) homeotropic layer and parallel electric field ; *c*) planar layer and parallel electric field ; *d*) planar layer and normal electric field. Moreover the cases of positive and negative dielectric anisotropy are considered as well as the cases of equal and unequal anchoring energies. This means that a total number of 16 different cases are the subject of this report. For the homeotropic cases the surface polarization is taken into account as well. In the cases *a*) and *c*) the effects are nonthreshold. The threshold for cases *b*) and *d*) can be polarity dependent. This polarity has already been observed experimentally in planar MBBA layers.

1. **Introduction.** — A theoretical study of static deformations in planar and homeotropic nematic layers, arising in weak d.c. electric fields (EF) is presented in this paper. It is well known that one of the basic mechanisms of orientational action of the EF on liquid crystals (LC) is the volume dielectric torque caused by the dielectric anisotropy of the nematic. If the initial orientation of the director with respect to the EF is dielectrically unstable, this torque leads to the familiar electric analogue of the Freedericksz transition [1]. Details of the theoretical description of the above are given in ref. [2, 3].

The dielectric torque acts for a.c. as well as d.c. EF. For d.c. fields however another orientational mechanism arises — the mechanism of flexoelectric deformation as pointed out for the first time by R. Meyer [4].

It is worth mentioning now that flexoelectric deformations of nematics possess two characteristic features, distinguishing them from any other electrostructural effects :

1. Flexoelectric effects can be observed in uniform and nonuniform d.c. electric fields. First the effects in uniform EF were described in [4] and observed in [22]. In nonuniform EF flexoelectric deformations were observed and theoretically interpreted for the first time by us in 1973 [20] and later on by Prost and Pershan in 1976 [7]. We will consider here only uniform d.c. EF and planar deformations (when the director is restricted to a plane containing the EF). In this case the flexoeffect does not create volume torques and flexoelectric terms do not contribute to the torque-balance equation. The flexoelectric deforming action is concentrated on the surfaces alone and leads only to surface torques on the glass-liquid crystal interfaces, as pointed out first by one of us in 1974 [5]. This can be easily understood

remembering that the torque is expressed as the derivative of the stress, which in uniform EF is uniform throughout the volume e.g. for flexoelectric bending of homeotropic layer [11] according to [4] $t_3 = e_{3x} E_x$ so $\Gamma_y = \partial t_3/\partial z = 0$ and jumps to zero at the interface (when instead of the derivative we can take the difference :

$$(\Gamma_{\text{surf}})_y = e_{3x} E_x - 0 = e_{3x} E_x).$$

2. As a result of this planar flexoelectric deformations in uniform d.c. EF can be realized only in the case of *soft* boundary conditions permitting substantial changes in the director orientation on the interfaces. Some of these deformations have already been described by the authors using the boundary condition derived in ref. [5] and [6] and expressing the surface torque balance; in these boundary conditions terms depending on the EF and flexoelectric moduli play a part as well. In this manner solutions for electrostructural deformation problems with boundary conditions depending on the electric field were obtained for the first time.

On the other hand however it has been pointed out recently by Prost and Pershan [7] that the flexoeffect is not the only factor giving rise to surface torques depending on the EF. Another possible effect is the presence of surface polarization which couples directly with the EF. These two effects can hardly be separated and substantial errors may be caused when determining flexoelectric moduli from this kind of experiment. This question will be discussed in detail later.

Due to these two features the exact solution of the deformation problem requires a detailed account of the orientational interaction between liquid crystal and substrate as well as all elastic terms which do not give volume contribution. One of these terms is related to the second-order elasticity of Nehring and Saupe [8] :

$$K_{13} \operatorname{div} (n.\operatorname{div} n)$$

Its influence is given for some cases in three separate papers [9, 10, 29].

It is our aim in this paper to develop first a general framework in vector notation for a unified description of various flexoelectric effects in uniform fields. As sketched above we derive an elastic-dielectric-flexoelectric torque balance equation and flexoelectrically dependent boundary conditions. Then on these grounds we will obtain solutions of some special planar one-dimensional cases and will discuss their importance. We will consider the four basic geometries presented on figure 1 :

*a*) a homeotropic layer and horizontal EF

*b*) a homeotropic layer and vertical EF

*c*) a planar layer and horizontal EF, parallel to the easy axis

*d*) a planar layer and vertical EF.

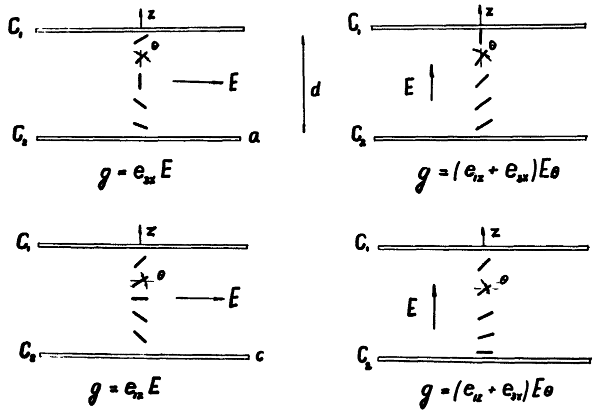


FIG. 1. — Schematical representation of the basic geometries.

The subcases of positive and negative dielectric anisotropy as well as equal and unequal surface energies have been considered for each of these basic geometries, making a total of 16 cases presented in this paper. Two of the cases have been considered previously in a different manner by Helfrich [11, 12]. A unified description based on a general theoretical approach however has not been published so far.

2. **Basic equations.** — The basis of our theoretical description is given by the *electric enthalpy* expression of a flexoelectric nematic [4] :

$$\begin{aligned} H_v = & \\ = 1/2 \{ & K_{11}(\operatorname{div} n)^2 + K_{22}(n.\operatorname{rot} n)^2 + K_{33}[(\operatorname{rot} n) \times n]^2 \} \\ & - e_{1z} E.(n.\operatorname{div} n) - e_{3x} E.[(\operatorname{rot} n) \times n] \\ & - 1/2[\chi_{\parallel}(E.n)^2 + \chi_{\perp}(E \times n)^2] \end{aligned} \quad (1)$$

where $n$ is the nematic director, $K_{ii}$ : the elastic moduli, $e_{1z}$ and $e_{3x}$ : the flexoelectric coefficients of splay and bend respectively, $\chi_{\parallel}$ and $\chi_{\perp}$ : the dielectric susceptibilities.

In case of *soft* boundary conditions (i.e. in case of weak anchoring) the total enthalpy of the sample is a sum of two terms, a volume and a surface one :

$$\mathcal{H} = \mathcal{H}_v + \mathcal{H}_s . \quad (2)$$

The volume term is given by an integral over the volume of the expression (1)

$$\mathcal{H}_v = \int_v H_v \, dv .$$

The surface energy is determined as we have already noted by the orientational interaction between substrates and LC. Following Rapini and Papoular [13], we can write :

$$\mathcal{H}_{\text{surf}} = 1/2 \int_s C_s(v \times n)^2 \, ds \quad (3)$$

in the case of homeotropically acting surfaces (a and b) and

$$\mathcal{H}_{surf} = 1/2 \int_s C_s(\nu . n)^2 \, ds \qquad (4)$$

for homogeneously acting surfaces (c and d).

Here $\nu$ is the interface normal and $C_s$ is a surface energy constant which in general could be a function of the position.

To this term we will also add, following Prost and Pershan [7], the energy of the polarized surface layer in an EF

$$\mathcal{H}_{pol} = - \int_s (m_p . E) \, ds \qquad (5)$$

where $m_p$ is the surface dipole density which might also depend on the position. The surface polarization plays an essential part in the homeotropic case (a and b). The reason it arises may be the specificity in the interaction of the substrate with the two ends of the molecule if they are different. As a result, a preferable orientation of the longitudinal molecular dipole will take place in the surface layer and an uncompensated dipole moment will exist. In such a case this moment will be parallel to the director and (5) becomes :

$$\mathcal{H}_{pol} = - \int_s m_p(n . E) \, ds \, . \qquad (6)$$

An estimate for $m_p$ for this case is given elsewhere [14]. The result for MBBA is :

$$m_p \sim 4 \times 10^{-4} \text{ statcoul.cm}^{-1} \, .$$

Molecules with equal end substituents probably give small $m_p$. There will also be a small $m_p$ in homogeneously oriented layers (c and d) because specific interactions orienting the normal molecular dipole are probably weaker and reorientation around the long axis is fast.

We will now proceed with the general form of the volume torque and the surface torque equations in a vector notation. Let us make a variation of the director $n \to n + \delta n$ with the condition

$$n^2 = 1 \text{ i.e. } n . \delta n = 0 \qquad (7)$$

as a constraint on $\delta n$ which means that $\delta n \perp n$. The variation $\delta \mathcal{H}$ will then generally have the form :

$$\begin{aligned} \delta \mathcal{H} &= \mathcal{H}(n + \delta n) - \mathcal{H}(n) = \delta \mathcal{H}_v + \delta \mathcal{H}_g \\ &= - \int_v (h . \delta n) \, dv + \int_s (g . \delta n) \, ds \, . \end{aligned} \qquad (8)$$

In the analogy with magnetism $h$ was called in ref. [15] a *molecular field*. In our case we will distinguish between :

$h$ : volume molecular field and

$g$ : surface molecular field .

With the constraint $n . \delta n = 0$ in mind, it is obvious that the condition for a minimum value of the functional $\delta \mathcal{H} = 0$ corresponding to the equilibrium state of the system is fulfilled if $h$ and $g$ are colinear with $n$. If this is not the case, torques arise, tending to orient $n$ parallel to $h$ in the volume and parallel to $g$ on the interfaces. The equilibrium equations will then read :

$$n \times h = 0 \text{ in } V \qquad (9)$$

$$n \times g = 0 \text{ on } S \qquad (10)$$

As will be seen, (10) plays the role of a boundary condition for the eq. (9).

The exact expressions of $h$ and $g$ are calculated by means of representing $\delta \mathcal{H}$ in the form (8). All divergence terms from $H_v$ take part in $g$ as well. The result for $h$ is as follows :

$$h = h_{elast} + h_{flexo} + h_{diel} \qquad (11)$$

where $h_{elast}$, $h_{flexo}$ and $h_{diel}$ are the elastic, flexoelectric and dielectric part of the volume molecular field. $h_{elast}$ was given by the Orsay group [15] :

$$\begin{aligned} h_{elast} = K_{11} \text{ grad div } n - K_{22}[T \text{ rot } n + \text{rot } (Tn)] + \\ + K_{33}[(\text{rot } n) \times B + \text{rot } (B \times n)] \end{aligned} \qquad (12)$$

where $T = n . \text{rot } n$ is the twist pseudoscalar

$B = (\text{rot } n) \times n$ is the bend vector

$h_{flexo}$ was obtained by Fan [16] :

$$\begin{aligned} h_{flexo} = e_{1z}[E \text{ div } n - \text{grad } (E . n)] + \\ + e_{3x}[E \times \text{rot } n - \text{rot } (E \times n)] \end{aligned} \qquad (13)$$

and $h_{diel}$ is analogous to the magnetic case [15] :

$$h_{diel} = \Delta \chi (E . n) \, E \, , \qquad \left( \Delta \chi = \frac{\Delta \varepsilon}{4 \pi} \right) . \qquad (14)$$

We will assume here that $\Delta \chi = \chi_{\parallel} - \chi_{\perp}$ is small enough so that $E$ does not depend substantially on the position, in contrast to ref. [2].

The surface molecular field has four components as follows :

$$g = g_{elast} + g_{flexo} + g_{surf} + g_{pol} \, . \qquad (15)$$

It must be noted that in fact only $g_{surf}$ and $g_{pol}$ really arise from surface effects while $g_{elast}$ and $g_{flexo}$ are just mathematical expressions of bulk contributions. Thus we obtain :

$$\begin{aligned} g_{elast} = K_{11} \, \nu \text{ div } n + K_{22}(Tn \times \nu) + \\ + K_{33} \, \nu \times (B \times n) \, . \end{aligned} \qquad (16)$$

An additional contribution to $g_{elast}$ also gives the term which describes the *saddle splay* in the form given by Frank [17] :

$$f_{\text{saddle}} = -(K_{22} + K_{24}) \sum_{j,i<j} (n_{i,i}.n_{j,j} - n_{i,j}.n_{j,i}) . \qquad (17)$$

It is well known that this term does not contribute to the volume torque because of its divergence character [18]. For its contribution to the surface molecular field we obtain :

$$(g_{\text{saddle}})_i = (K_{22} + K_{24}) \left( v_i \sum_{j \neq i} n_{j,j} - \sum_{j \neq i} v_j n_{j,i} \right) . \qquad (18)$$

This term will be of importance if the deformation is nonplanar and two components of the splay and twist exist. Boundary conditions paying attention to this term were derived in some other papers [31, 32]. The flexoelectric component of $g$ is :

$$g_{\text{flexo}} = -e_{1z}(E.n)\, v - e_{3x}[v \times (n \times E)] . \qquad (19)$$

For $g_{\text{surf}}$ we obtain from (3) and (4) respectively :

$$g_{\text{surf}} = -C_s(v.n)\, v \qquad (20')$$

for the homeotropic case and

$$g_{\text{surf}} = C_s(v.n)\, v \qquad (20'')$$

for the homogeneous case. Finally

$$g_{\text{pol}} = -m_p E . \qquad (21)$$

As we have noticed already this term is of importance only in the homeotropic case.

Now we have to specify these general results for the problem formulated above.

The planar character of our problem means that only deviations of the director in a plane normal to the substrate also containing the electric field vector (the $xOz$ plane, Fig. 1) are taken into account. Since in case *b*) this plane is not defined uniquely, we shall assume that the $Ox$ direction is specified by rubbing of the plates. The deviations of the director are described by the angle $\theta$ between the initial and the final orientation. It is also assumed that $\theta$ depends only on the distance to the glass surfaces — the $z$ coordinate. This makes the problem one-dimensional. A two-dimensional non-planar problem for the case *d*) where azimuthal deviations are considered as well — $\theta(y, z)$ and $\varphi(y, z)$ was solved recently by Bobilev and Pikin [19] assumming strong $\theta$ and $\varphi$ anchoring on the walls and a threshold for domain formation was found. In that case a volume flexoelectric torque is acting. The authors also have in mind strong $\varphi$ anchoring, but weak $\theta$ anchoring. In this case :

*a*) $n(\sin\theta(z), 0, \cos\theta(z))$ $E(E, 0, 0)$
*b*) $n(\sin\theta(z), 0, \cos\theta(z))$ $E(0, 0, E)$ $v(0, 0, 1)$
*c*) $n(\cos\theta(z), 0, \sin\theta(z))$ $E(E, 0, 0)$
*d*) $n(\cos\theta(z), 0, \sin\theta(z))$ $E(0, 0, E)$ .

It is easily seen that in all these cases the flexoelectric contribution to the molecular field is parallel to the director so that

$$n \times h_{\text{flexo}} = 0$$

i.e. a volume flexoelectric torque does not act as already pointed out by the authors in the introduction. The same is valid for the saddle-splay component of the surface field which in the planar case is identically equal to zero.

Finally it is important to stress that if the electric field is nonhomogeneous, a volume flexoelectric torque will arise even in the planar case. This effect was called by the authors *gradient flexoelectric effect* (1974) because the volume torque is dependent on the gradient of the electric field and the sum of the flexocoefficients ($e_{1z} + e_{3x}$) [20]. We have studied this effect in an inhomogeneous EF, the inhomogeneity being produced by a volume space charge. The importance of the volume flexoeffect was recognized later by Prost and Pershan and an excellent way for producing EF inhomogeneity by means of interdigital electrodes was used [7]. The analogy between gradient torque and the induced birefringence in gases in EF gradient leads Prost and Marcerou to important ideas for quadrupolar flexoelectricity as well [21].

3. **General classification.** — The general features of the different cases are summarized in Table 1. Basically this classification makes use of the fact that in the cases a and c the flexoelectric surface torques (19) are equal to (Fig. 1)

$$\Gamma_{\text{flexo}} = e_{3x} E, \text{ resp. } \Gamma_{\text{flexo}} = e_{1z} E \qquad (22)$$

so that they are different from zero even in an undeformed state. On the other hand in the cases b and d the surface torques are (again Fig. 1)

$$\Gamma_{\text{flexo}} = (e_{1z} + e_{3x}) E\theta \qquad (23)$$

so that in the case of zero $\theta$ — undeformed state, they are also zero. It will be seen that for the first two cases, the theory predicts nonthreshold deformation of the Haas *et al.* type [22] and for the second two cases we have a typical instability problem solved for the first time in a special subcase of *b*): $\Delta\chi = 0$, $C_1 = 0$, $C_2 \to \infty$ by Helfrich [12]. This polar transition whose polarity is connected with the surface energy asymmetry is called the *Helfrich transition* by the authors. In this case the theory predicts that at a given value of the voltage for one polarity, deformation will be obtained, but for the opposite polarity the flexoeffect will tend to improve the initial orientation if it is not ideal. Such an effect has already been observed by one of the authors in 1974 [23] using a planar MBBA layer (Fig. 2) and an interference technique.

The effects of dielectric anisotropy on flexoelectric deformations are also summarized in table I. The cases

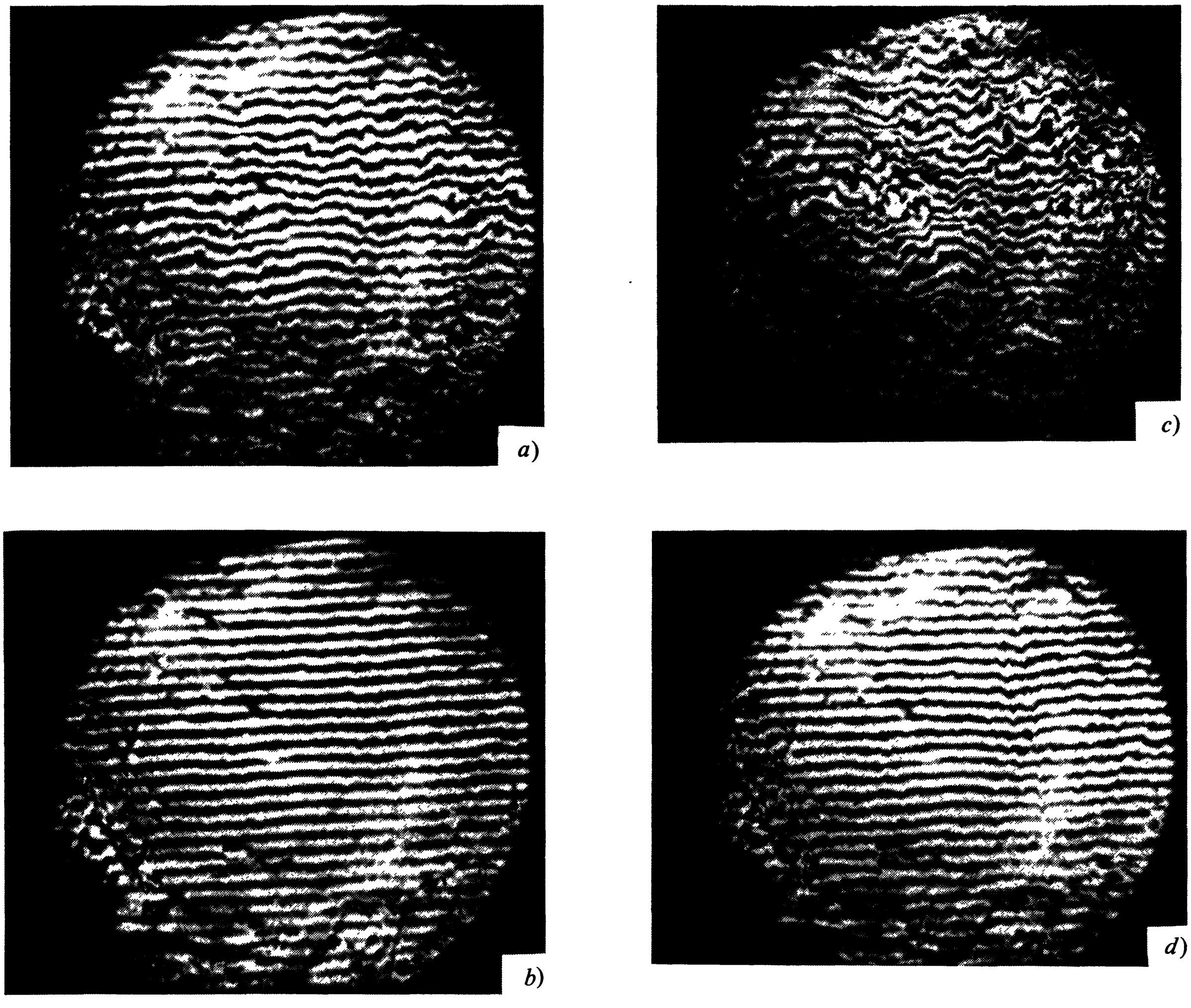

FIG. 2. — Interference-polarizing pictures of planar MBBA layer. Microscope — MPI-5, green light, layer thickness — 30 μm. With homogeneous orientation the lines are parallel. *a*) Zero voltage — the initial orientation is not ideal. *b*) High frequency a.c. voltage (10 V, 20 kHz) — the dielectric torque improves the planar orientation. *c*) d.c. voltage (1V) — deforming action. The deformation pattern is somewhat irregular. In the upper left corner a region with strong anchoring can be seen where there is no deformation. *d*) d.c. voltage reversed (−1V) — the reversed polarity tends to improve the planar orientation, similar to a.c. voltage. The planar orientation is accomplished by the rubbing method. Low anchoring conditions are obtained by keeping the sample for a period of about two weeks (in a desiccator). During this time a decrease of the surface energy *C* was observed, probably due to the slow building up of adsorption layers of impurities and liquid crystal molecules on the surfaces.

are divided into stable and unstable on the basis of their dielectric stability. When they are unstable, they will be treated in terms of a flexoelectrically influenced Freedericksz transition.

The cases a and c are formally equivalent. The solution of *c*) is obtainable from *a*) by replacing $e_{3x}$ by $e_{1z}$, $K_{33}$ by $K_{11}$ and $\Delta\chi > 0$ by $\Delta\chi < 0$ or vice versa. In this manner case *c*) can be used to measure the splay flexocoefficient $e_{1z}$ only, as already proposed by the authors [24]. In the same manner solutions of *b*) and *d*) become equivalent with the substitution of $\Delta\chi > 0$ by $\Delta\chi < 0$ or *vice versa*. For this reason we will consider in more detail only cases *a*) and *b*).

Some words about the general idea of the calculations are necessary. Let us look at the nonthreshold cases *a*) and *c*) first. The most simple situation is that of a zero $\Delta\chi$. The solution is an odd function of $z$, taking maximal values at the boundaries where flexoelectric torques are concentrated. In the small angle approximation the amplitude of the solution is linear in $e_{in} E$. So in fact we calculate the flexoelectric linear response function for the orientation.

The situation is more complicated when the dielectric anisotropy comes into play. Now the solution is a linear combination of an even function of $z : f_1(z)$, describing the dielectric response, with a maximum in the middle where the layer is most free from the boundary constraints, and an odd function $f_2(z)$ with a minimum in the middle describing flexoelectric response :

$$\theta(z) = \theta_1 f_1(z) + \theta_2 f_2(z) .$$

TABLE I

*General characteristics of the basic subcases*

| Stable | Unstable | Stable | Unstable |
|---|---|---|---|
| *a*) $\Delta\chi < 0, C_1 = C_2$ Nonthreshold flexodeformations, dielectrically depressed at higher voltages. Zero deformation in the middle of the layer. | $\Delta\chi > 0, C_1 = C_2$ Nonthreshold flexodeformations, followed by Freedericksz transition. | *b*) $\Delta\chi > 0, C_1 = C_2$ Polarity independent, dielectrically depressed Helfrich transition. | $\Delta\chi < 0, C_1 = C_2$ Polarity independent, flexoelectrically enhanced Freedericksz transition. |
| $\Delta\chi < 0, C_1 \neq C_2$ Nonthreshold flexodeformation, dielectrically depressed at higher voltages. The point of zero deformation moves with the voltage. | $\Delta\chi > 0, C_1 \neq C_2$ Flexoelectrically degenerated Freedericksz transitions. | $\Delta\chi > 0, C_1 \neq C_2$ Polarity dependent, dielectrically depressed Helfrich transition. | $\Delta\chi < 0, C_1 \neq C_2$ Polarity dependent, flexoelectrically enhanced Freedericksz transition |
| *c*) $\Delta\chi > 0, C_1 = C_2$ Nonthreshold flexodeformations, dielectrically depressed at higher voltages. Zero deformation in the middle of the layer. | $\Delta\chi < 0, C_1 = C_2$ Nonthreshold deformations, followed by Freedericksz transition. | *d*) $\Delta\chi < 0, C_1 = C_2$ Polarity independent, dielectrically depressed Helfrich transition. | $\Delta\chi > 0, C_1 = C_2$ Polarity independent, flexoelectrically enhanced Freedericksz transition. |
| $\Delta\chi > 0, C_1 \neq C_2$ Nonthreshold flexodeformation, dielectrically depressed at higher voltages. The point of zero deformation moves with the voltage. | $\Delta\chi < 0, C_1 \neq C_2$ Flexoelectrically degenerated Freedericksz transition. | $\Delta\chi < 0, C_1 \neq C_2$ Polarity dependent, dielectrically depressed Helfrich transition. | $\Delta\chi > 0, C_1 \neq C_2$ Polarity dependent, flexoelectrically enhanced Freedericksz transition. |

In the symmetrical dielectrically stable situations (Table I) there is no dielectric response of the system at all. The dielectric torque only leads to a depression of the flexoelectric response. In the symmetrical unstable situations there is also no dielectric response of the system — $\theta_1 = 0$ for electric fields up to the threshold for Freedericksz transition $E_{tr}$ — due to the different symmetry of the deformation pattern the flexoelectric and dielectric response are not coupled. In the unsymmetrical stable situations there is flexoelectric as well as dielectric response but both of them are dielectrically depressed. And finally in the unsymmetrical unstable situations both the dielectric and flexoelectric response tend to infinity in the vicinity of a threshold field. This resembles the behaviour of the response function for the electric polarization $\chi$ of a pyroelectric near the Curie temperature for the phase transition pyroelectric-ferroelectric (e.g. [33]).

The somewhat unusual character of our problem consists in that : it is the same electric field which *via* the boundary conditions determines the flexoelectric response and *via* the volume action leads to the instability in the system. Strictly speaking in that case the net tilt of the layer, produced by the flexoeffect, causes a degeneration of the Freedericksz transition — the coupling between the flexoelectric deformation and the dielectric torque tends to amplify the flexoelectric tilt even at small fields. The increase of the deformation is continuous and no threshold field can be consistently defined — as we say the transition is degenerated.

In the cases *b*) and *d*) this problem does not exist. Either flexoelectric or dielectric torque or both of them lead to instability behaviour — below a definite value of the electric field, depending both on dielectric and flexoelectric parameters of the material the deformation is identically zero. There is no linear response of the system. The small angle approximation permits only the threshold field determination. The field dependence of the deformation above the threshold necessitates keeping third powers of $\theta$ in the equation and boundary conditions (equivalent to the truncation of the total enthalpy expression (2) to fourth order). This is done by us in two cases only — formulae (64) and (68). Due to the fact that there is neither first nor third powers of $\theta$ in (2) the combined dielectric-flexoelectric transition in these cases is truly second order.

Another point deserves mention as well. In our calculations we do not take into account the reverse action of the depolarizing field due to the flexoelectric polarization of the deformed structure itself as was done in the papers of Deuling [34, 27]. This correction field is important in very pure materials, because the flexoelectric polarization once created is screened by redistribution of the space charges within a time $\varepsilon/4\pi\sigma$, where $\sigma$-conductivity. It also becomes important at low temperatures, where $\sigma$ decreases several orders of magnitude and the space charge is *frozen* [30].

4. **Solutions.** — 4.1 THE HOMEOTROPIC LAYER UNDER THE INFLUENCE OF A HORIZONTAL ELECTRIC FIELD. — In this case the torque balance equation for a small angle approximation takes the form (following (9), (11), (13) and (14)) :

$$K_{33}\frac{d^2\theta}{dz^2} + \Delta\chi E^2\,\theta(z) = 0\,. \qquad (24)$$

Boundary conditions in the same approximation are given by (following (10), (15), (16), (19), (20′) and (21)) :

$$C_1\,\theta + K_{33}\frac{d\theta}{dz} = (e_{3x} + m_{p1})\,E\,, \quad z = \frac{d}{2}$$

$$C_2\,\theta - K_{33}\frac{d\theta}{dz} = -\,(e_{3x} - m_{p2})\,E\,, \quad z = -\frac{d}{2}\,.$$

Different cases of surface polarization (hydrophilic and hydrophobic surfaces) are discussed in detail elsewhere [14]. If the surfaces are different in general $m_{p1} + m_{p2} \neq 0$. But if they are identical, the surface polarization moments will be equal and in the opposite direction, i.e. :

$$m_{p1} = -\,m_{p2} = m_p\,. \qquad (26)$$

The field dependent surface torque will then take the same form at the two boundaries : $(e_{3x} + m_p)\,E$. This means that the surface polarization acts together with the flexoeffect increasing or decreasing its action depending on its direction (see ref. [14]). For bigger deformations however these two terms become different due to their different dependence on $\theta$ : $e_{3x}\,E\cos^2\theta$ versus $m_p\,E\cos\theta$.

It is convenient to discuss the solutions in terms of the following parameters :

— dielectric coherence length :

$$\xi(E) = \frac{1}{E}\sqrt{\frac{K_{33}}{|\Delta\chi|}} = \frac{U_F}{\pi E} \qquad (27)$$

— flexoelectric coherence lengths :

$$\eta_{1,2}(E) = \frac{1}{E}\cdot\frac{K_{33}}{e_{3x} \pm m_{p1,2}}\,, \quad +\text{ for } m_{p1}\,, \quad -\text{ for } m_{p2} \qquad (28)$$

— extrapolation lengths :

$$b_{1,2} = \frac{K_{33}}{C_{1,2}} \qquad (29)$$

— threshold voltage for Freedericksz transition :

$$U_F = \pi\sqrt{\frac{K_{33}}{|\Delta\chi|}} = E_F . d \qquad (30)$$

— threshold voltages for Helfrich transition

$$U_{H1,2} = \frac{K_{33}}{e_{1z} + e_{3x} \pm m_{p1,2}}\,, \quad +\text{for } m_{p1}\,, \quad -\text{for } m_{p2}\,. \qquad (31)$$

4.1.1 *Dielectrically compensated* LC ($\Delta\chi = 0$). — In this special subcase (which can be realized for instance with mixtures) the solution of (24) can be expressed in the form :

$$\theta(z) = \theta_0\,\frac{z - z_0}{d} \qquad (32)$$

where

$$\theta_0 = \left(\frac{b_1}{\eta_1} + \frac{b_2}{\eta_2}\right)\Big/(b_1 + b_2 + d) \qquad (33)$$

and

$$z_0 = \frac{d}{2}\left[\left(b_2\frac{\eta_1}{\eta_2} - b_1\right) + b_1\,b_2\left(1 - \frac{\eta_2}{\eta_1}\right)\right] \times \left(b_1 + b_2\frac{\eta_1}{\eta_2}\right)^{-1} \qquad (34)$$

where the ratio $\eta_1/\eta_2 = (e_{3x} - m_{p2})/(e_{3x} + m_{p1})$ is not dependent on $E$.

This solution generalizes the Helfrich solution [11] obtained at equal anchoring energies ($b_1 = b_2$) and zero surface polarization ($m_p = 0$). It demonstrates that as a result of the surface asymmetry, the plane of zero deformation is no longer in the middle of the layer. For this reason with asymmetrical boundary conditions the influence of the anchoring energy is sufficient even for small values of layer thickness while in the symmetrical conditions it is of importance only at greater thicknesses of the order $(b_1 + b_2)$.

Making the assumption (26) which means that $\eta_1 = \eta_2$, we obtain :

$$\theta(z) = \frac{e_{3x} + m_p}{K_{33}}\,E\,\frac{z - z_0}{1 + d/(b_1 + b_2)} \qquad (35)$$

with

$$z_0 = \frac{d}{2}\,\frac{b_2 - b_1}{b_1 + b_2}\,.$$

The zero deformation plane is nearer to the boundary when $C$ is larger.

It is evident from this result that the homeotropic layer deformation proposed by Helfrich as a method of measuring the bend flexocoefficient $e_{3x}$ requires the elucidation of the problem for the values of two important corrections, which if not taken into account will lead to substantial errors :

*a*) boundary asymmetry correction ($b_1 \neq b_2$);
*b*) surface polarization correction ($m_p \neq 0$).

The boundary asymmetry correction can be best estimated by calculating the field induced birefringence which is the experimentally measured quantity in this type of experiment [25] :

$$\delta = \frac{1}{2} n_0 \left(1 - \frac{n_0^2}{n_e^2}\right) \int_{-d/2}^{d/2} \theta^2(z)\, dz = \\ = \frac{1}{24} n_0 \left(1 - \frac{n_0^2}{n_e^2}\right) \left(\frac{e_{3x} + m_p}{K_{33}}\right)^2 E^2 d^3 \times \\ \times \frac{1 + 3(b_1 - b_2)^2/(b_1 + b_2)^2}{[1 + d/(b_1 + b_2)]^2} \qquad (36)$$

where $n_0$ and $n_e$ are the ordinary and extraordinary refractive indices respectively.

E.g. if $b_1 = 2\, b_2$ the correction in the $\delta$ value is 33 % regardless of the layer thickness.

The dependence of $\delta$ on the layer thickness $d$ permits the evaluation of the total extrapolation length : for smaller $d$ from (36) we obtain a cubic growth

$$\delta \sim d^3 \qquad (37)$$

but for greater $d$, a linear growth with a slope $(b_1 + b_2)^2$ is established :

$$\delta \sim (b_1 + b_2)^2\, d\,. \qquad (38)$$

The surface polarization correction is estimated by the authors elsewhere [14]. It can easily be seen that depending on its direction, the surface polarization torque can act in the same or in the opposite direction as the flexoelectric one, thus increasing or decreasing the apparent value of $e_{3x}$ which was found in fact in two experiments with MBBA [25, 22] using different boundary conditions. From the two very different apparent values of $e_{3x}$ : $3.7 \times 10^{-5}$ dyn.$^{1/2}$ and $2.7 \times 10^{-4}$ dyn.$^{1/2}$ the true value of $e_{3x}$ was estimated to be

$$e_{3x} \approx 10^{-4} \text{ dyn.}^{1/2}\,.$$

This value is more realistic than the broadly accepted value $3.7 \times 10^{-5}$ dyn.$^{1/2}$. It resolves the apparent contradiction between different flexoelectric experiments with MBBA [25, 26].

4.1.2 *Dielectrically stable layer* ($\Delta\chi < 0$). — This is the most probable case for substances with large $e_{3x}$ because of the large normal component of the molecular dipole. In this case the volume dielectric torque acts against the surface flexoeffect with a tendency to depress the flexodeformation. Due to the fact however that the flexoeffect is linear, it prevails at small voltages.

Our torque balance equation (24) can be written as

$$\xi^2 \frac{d^2\theta}{dz^2} - \theta(z) = 0 \qquad (39)$$

with a general solution of the type

$$\theta(z) = \theta_1 \operatorname{ch} \frac{z}{\xi} + \theta_2 \operatorname{sh} \frac{z}{\xi}\,. \qquad (40)$$

The constants $\theta_1$ and $\theta_2$ are given by the solution of the boundary value problem :

$$\left(\operatorname{ch}\frac{d}{2\,\xi} + \frac{b_1}{\xi}\operatorname{sh}\frac{d}{2\,\xi}\right)\theta_1 + \\ + \left(\operatorname{sh}\frac{d}{2\,\xi} + \frac{b_1}{\xi}\operatorname{ch}\frac{d}{2\,\xi}\right)\theta_2 = \frac{b_1}{\eta_1} \\ \left(\operatorname{ch}\frac{d}{2\,\xi} + \frac{b_2}{\xi}\operatorname{sh}\frac{d}{2\,\xi}\right)\theta_1 - \\ - \left(\operatorname{sh}\frac{d}{2\,\xi} + \frac{b_2}{\xi}\operatorname{ch}\frac{d}{2\,\xi}\right)\theta_2 = -\frac{b_2}{\eta_2}\,. \qquad (41)$$

The determinant of the system

$$\Delta = -\left[\left(1 + \frac{b_1 b_2}{\xi^2}\right)\operatorname{sh}\frac{d}{\xi} + \frac{(b_1 + b_2)}{\xi}\operatorname{ch}\frac{d}{\xi}\right] \neq 0$$

is always different from zero. The system always has a solution. The deformation is not a threshold one.

If the assumption (26) is made, the solution can be written as :

$$\theta = \theta_0 \operatorname{sh}\frac{z - z_0}{\xi} \qquad (42)$$

where

$$\operatorname{th}\frac{z_0}{\xi} = (b_2 - b_1)\left[(b_1 + b_2)\operatorname{cth}\frac{d}{2\,\xi} + 2\frac{b_1 b_2}{\xi}\right]^{-1} \qquad (43)$$

$$\theta_0 = \frac{\xi}{\eta}\left[\frac{\xi}{b_1}\operatorname{sh}\frac{d - 2\,z_0}{2\,\xi} + \operatorname{ch}\frac{d - 2\,z_0}{2\,\xi}\right]^{-1}\,. \qquad (44)$$

It is evident from (43) that in this case the zero deformation plane $z = z_0$ moves with the voltage towards the middle of the layer. Only when a strong anchoring is imposed on one of the walls ($b_1$ or $b_2 = 0$) is the plane fixed, and it then coincides with this wall.

If now $b_1 = b_2 = b$ — the symmetrical surface case — $z_0 = 0$ and the solution (42) is :

$$\theta(z) = \frac{\operatorname{sh}\frac{z}{\xi}}{\frac{\xi}{b}\operatorname{sh}\frac{d}{2\,\xi} + \operatorname{ch}\frac{d}{2\,\xi}} \cdot \frac{\xi}{\eta}\,. \qquad (45)$$

It can immediately be seen that if $|\Delta\chi| \to 0$ or $E \ll \frac{2}{d}\sqrt{\frac{K_{33}}{|\Delta\chi|}}$ so that $1/\xi \to 0$ the solution (45) is reduced to solution (55) discussed above. At small voltages the dielectric effect is negligible but at higher voltages the maximum deformations on the walls tend to a saturation value (for $C = 0$) :

$$\theta\left(\frac{d}{2}\right) \xrightarrow[E\to\infty]{} \frac{e_{3x} + m_p}{\sqrt{|\Delta\chi| K_{33}}} \sim 0.5 \text{ rad for MBBA}$$

and the deformation penetrates into the layer only at distances of the order of $\xi$ — the dielectric coherence length. One can obtain for the induced phase difference :

$$\delta \sim \frac{(e_{3x} + m_p)^2}{|\Delta\chi| K_{33}} \cdot \frac{\xi \operatorname{sh}\frac{d}{\xi} - d}{\operatorname{ch}\frac{d}{\xi} + 1}. \tag{46}$$

For weak voltages $\left(\frac{e_{3x} + m_p}{K_{33}} E\right)^2 d^3$ i.e. the increase follows $E^2$ but for higher voltages there is a decrease following $\xi$, i.e. $E^{-1}$.

4.1.3 *Dielectrically unstable layer* ($\Delta\chi > 0$). — In this case layers with strong anchoring ($b_1 = b_2 = 0$) will display a Freedericksz transition at a critical field $E_F = \frac{\pi}{d}\sqrt{K_{33}/\Delta\chi}$. With weak anchoring this threshold field decreases as has already been calculated by Rapini and Papoular. In our case we have to take into account the surface field-dependent torques as well. The results can be described briefly as follows : if the problem is symmetrical ($b_1 = b_2$, $m_{p1} = - m_{p2}$) the flexodeformation established at small voltages is zero in the middle of the layer and is an odd function of $z$ — formula (35). On the other hand the Freedericksz deformation is characterized by an even function of $z$ with a maximum in the middle (e.g. ref. [2, 3]). For this reason in the symmetrical problem these two effects remain uncoupled as we have already noticed. On the other hand when $b_1 \neq b_2$ the zero deformation is not in the middle and the flexoeffect produces a net tilt of the layer which degenerates the Freedericksz transition because the volume dielectric torque immediately tends to increase this tilt.

The torque balance equation (24) now reads as follows :

$$\xi^2 \frac{d^2\theta}{dz^2} + \theta(z) = 0 \tag{47}$$

and its solutions can be obtained from those of (39) by changing $\xi$ to $i\xi$ :

$$\theta(z) = \theta_1 \cos\frac{z}{\xi} + \theta_2 \sin\frac{z}{\xi}. \tag{48}$$

The boundary value problem now has a solution when $\Delta \neq 0$. The condition $\Delta = 0$ leads to the following equation for the Freedericksz threshold : in the absence of flexoelectric surface torques :

$$\operatorname{ctg}\frac{d}{\xi} = \left(\frac{b_1 b_2}{\xi^2} - 1\right)\frac{\xi}{b_1 + b_2} \tag{49}$$

which determines the threshold value of $\xi(E)$. Having in mind that

$$\xi(E) = \frac{d}{\pi}\sqrt{\frac{E_F}{E}} \tag{50}$$

expression (49) can be rewritten in terms of the threshold field $E_{tr}$ :

$$\operatorname{ctg}\pi\frac{E_{tr}}{E_F} = \left(b_1 b_2 \frac{\pi^2}{d^2}\frac{E_{tr}^2}{E_F^2} - 1\right)\frac{d}{\pi(b_1 + b_2)}\frac{E_F}{E_{tr}}. \tag{51}$$

This equation is a generalization of the Rapini-Papoular equation for the unsymmetrical boundary condition. It can easily be demonstrated using the identity

$$2 \operatorname{ctg}\alpha = \operatorname{ctg}\frac{\alpha}{2} - \operatorname{tg}\frac{\alpha}{2}$$

that when $b_1 = b_2$ eq. (51) contains the solution of the Rapini-Papoular equation :

$$\operatorname{ctg}\frac{\pi}{2}\frac{E_{sym}}{E_F} = \frac{\pi b}{d}\frac{E_{sym}}{E_F}. \tag{52}$$

If we make the assumption (26), the solution in the unsymmetrical case can be written in the form similar to (42) :

$$\theta(z) = \theta_0 \sin\frac{z - z_0}{\xi} \tag{53}$$

where

$$\operatorname{tg}\frac{z_0}{\xi} = (b_1 - b_2)\left[(b_1 + b_2)\operatorname{ctg}\frac{d}{2\xi} - 2\frac{b_1 b_2}{\xi_1}\right]^{-1} \tag{54}$$

$$\theta_0 = \frac{\xi}{\eta}\left[\frac{\xi}{b_1}\sin\frac{d - 2z_0}{2\xi} + \cos\frac{d - 2z_0}{2\xi}\right]^{-1}. \tag{55}$$

In the symmetrical case $b_1 = b_2$, eq. (54) shows that $z_0 = 0$ until the denominator becomes equal to zero. In view of (50) this condition is equivalent to eq. (52). So up to the threshold $E_{sym}$, the two effects are uncoupled, as we noticed at the beginning. The complete solution in the flexoelectric region is then, analogously to (45) :

$$\theta(z) = \frac{\xi}{\eta}\sin\frac{z}{\xi}\left(\frac{\xi}{b}\sin\frac{d}{2\xi} + \cos\frac{d}{2\xi}\right)^{-1}. \tag{56}$$

In the unsymmetrical case it is evident from (54) that $z_0$ increases with the voltage, the field tends to increase the flexoelectric tilt and the Freedericksz transition is degenerated.

The solution of eq. (51) in the case of weak asymmetry $\Delta b/b \ll 1$ can be expressed by the solution $E_{\text{sym}}$ of (52) with $b = 1/2(b_1 + b_2)$ in the following manner :

$$E_{\text{tr}} = E_{\text{sym}} + \Delta E \tag{57}$$

where

$$\frac{\Delta E}{E_F} = \frac{\pi b}{d} \frac{E_F}{E_{\text{sym}}} \left(\frac{\Delta b}{2 b}\right)^2 \times \\ \times \left[ \frac{\pi b}{d} + \frac{d}{\pi b} \frac{E_F^2}{E_{\text{sym}}^2} + \frac{2 \pi}{\sin^2 \dfrac{\pi E_{\text{sym}}}{E_F}} \right]^{-1} . \tag{58}$$

An analysis of eq. (57) shows that the boundary asymmetry increases the threshold in comparison to that corresponding to the mean value of the anchoring energy.

4.2 A HOMEOTROPIC LAYER UNDER THE ACTION OF A VERTICAL ELECTRIC FIELD. — In this case, in a small angle approximation, the torque balance equation takes the form :

$$K_{33} \frac{\mathrm{d}^2\theta}{\mathrm{d}z^2} - \Delta\chi E^2 \theta(z) = 0 . \tag{59}$$

The boundary conditions in the same approximation are given by :

$$[C_1 + (e_{1z} + e_{3x} + m_{p1}) E] \theta + K_{33} \frac{\mathrm{d}\theta}{\mathrm{d}z} = 0 \quad z = \frac{d}{2}$$

$$[C_2 - (e_{1z} + e_{3x} - m_{p2}) E] \theta - K_{33} \frac{\mathrm{d}\theta}{\mathrm{d}z} = 0 \quad z = -\frac{d}{2} . \tag{60}$$

As we have already noticed in this geometry the deformation is characterized by a definite threshold voltage.

4.2.1 *Dielectrically compensated* LC ($\Delta\chi = 0$). — In this special subcase the solution is a linear function of $z$ and it arises when the field exceeds a value which is given by the condition that the determinant of the system (60) becomes zero :

$$b_1 b_2 \frac{E_{\text{tr}}^2}{U_{H1} U_{H2}} - \left[ \frac{b_1}{U_{H1}} - \frac{b_2}{U_{H2}} + \\ + b_1 b_2 \left( \frac{1}{U_{H1}} - \frac{1}{U_{H2}} \right) \right] E_{\text{tr}} - \left( 1 + \frac{b_1 + b_2}{d} \right) = 0 . \tag{61}$$

Because of the potential jump at the surface polarized electrodes the field acting on the layer is given by $E_d = U + 4 \pi(m_{p1} + m_{p2})$. $U_{H1}$ and $U_{H2}$ in (61) are given by (31). Because of the term linear in $E_{\text{tr}}$ whose existence is due to the anchoring energy and surface polarization asymmetry, the effect is polar — the positive and negative thresholds are different (positive means that the electric field is in the positive O$z$ direction — from $C_2$ to $C_1$). Let us now assume surface polarization symmetry (26). Then $U_{H1} = U_{H2} = U_H$ and $E . d = U$. The solution of (61) is given by :

$$2\left(\frac{U_{\text{tr}}}{U_H}\right) = -\left(\frac{d}{b_1} - \frac{d}{b_2}\right) \pm \\ \pm \sqrt{\left(\frac{d}{b_1} - \frac{d}{b_2}\right)^2 + 4\left(\frac{d}{b_1} + \frac{d}{b_2} + \frac{d^2}{b_1 b_2}\right)} . \tag{62}$$

If $C_1 > C_2$ ($b_1 < b_2$) and $e_{1z} + e_{3x} + m_p > 0$ ($U_H > 0$) then the positive threshold is lower than the negative, so a lower threshold is obtained when the electrode with a lower anchoring energy is an anode.

The small angle approximation permits the derivation of the deformation threshold but the amplitude of the deformation remains undetermined. In order to calculate it for voltages greater than $U_{\text{tr}}$, cubic terms have to be included in the expansion of $\sin 2\theta$ in the boundary conditions. Let us now consider for the sake of simplicity the symmetrical case ($b_1 = b_2 = b$) when the thresholds in two directions are the same :

$$U_{\text{sym}} = \pm \frac{U_H}{2} \sqrt{\frac{d^2}{b^2} + \frac{2 d}{b}} . \tag{63}$$

With $K_{11} = K_{33}$, we obtain

$$\theta(z) = \frac{d}{b} \frac{\sqrt{1 + \dfrac{2 b}{d} \left( \dfrac{z}{d} - \dfrac{b}{2 d} \dfrac{U_{\text{sym}}}{U_H} \right)}}{\sqrt{1 + \dfrac{2 d}{3 b} - \dfrac{2}{3} \kappa \left(1 + \dfrac{d}{b}\right)\left(2 + \dfrac{d}{b}\right)}} \times \\ \times \sqrt{\frac{U - U_{\text{sym}}}{U_{\text{sym}}}} \tag{64}$$

where

$$\kappa = \frac{3}{4} m_p/(e_{1z} + e_{3x} + m_p) .$$

The case where $b \to \infty$ ($C = 0$) and the deformation is, according to (63), not a threshold one is of some theoretical interest. Analysing now the full boundary conditions, one finds that the boundary angles $\theta_1$ and $\theta_2$ are connected by the relation

$$\theta_1 + \theta_2 = \pi/2 \tag{65}$$

and that $\theta_1$ (at the wall $C_1$) for small voltages is given by

$$\theta_1 = \frac{\pi}{4} \Big/ \left(1 - \frac{1}{2} \frac{U}{U_H}\right) . \tag{66}$$

We got nonthreshold behaviour and as a result a linear response.

This result means that at a voltage just above zero the layer is inclined as a whole at an angle of 45° to the field and after that it starts deforming. When $U$ increases, $\theta_1 \to \pi/2$ and $\theta_2 \to 0$. This resembles the situation realizable in the case $C_1 \to \infty$ as will now be demonstrated. In the latter situation ($b_1 = 0$) the positive threshold is given by :

$$U_{\mathrm{tr}} = U_{\mathrm{H}}\left(1 + \frac{d}{b_2}\right) \tag{67}$$

while the negative is infinite. The Helfrich solution corresponds to the case $b_2 \to \infty$ ($U_{\mathrm{tr}} = U_{\mathrm{H}}$). The maximal deformation at the wall $C_1$ as a function of voltage not far from the threshold (67) is given by formula (68). As usual the response is non linear :

$$\theta_1^2 = \frac{3}{4}\,\frac{(U/U_{\mathrm{tr}}) - 1}{(U/U_{\mathrm{tr}}) - \left(1 + \dfrac{b^2}{d}\right)^{-1}}. \tag{68}$$

4.2.2 *Dielectrically stable layer* ($\Delta\chi > 0$). — The torque balance equation is

$$\xi^2 \frac{\mathrm{d}^2\theta}{\mathrm{d}z^2} - \theta(z) = 0 \tag{69}$$

with a solution

$$\theta(z) = \theta_1 \operatorname{ch}\frac{z}{\xi} + \theta_2 \operatorname{sh}\frac{z}{\xi} \tag{70}$$

$\theta_1$ and $\theta_2$ are given by the solution of the boundary value problem

$$\begin{aligned}
&\left[\left(1 + \frac{b_1}{\eta_1}\right)\operatorname{ch}\frac{d}{2\xi} + \frac{b_1}{\xi}\operatorname{sh}\frac{d}{2\xi}\right]\theta_1 + \\
&\quad + \left[\left(1 + \frac{b_1}{\eta_1}\right)\operatorname{sh}\frac{d}{2\xi} + \frac{b_1}{\xi}\operatorname{ch}\frac{d}{2\xi}\right]\theta_2 = 0 \\
&\left[\left(1 - \frac{b_2}{\eta_2}\right)\operatorname{ch}\frac{d}{2\xi} + \frac{b_2}{\xi}\operatorname{sh}\frac{d}{2\xi}\right]\theta_1 - \\
&\quad - \left[\left(1 - \frac{b_2}{\eta_2}\right)\operatorname{sh}\frac{d}{2\xi} + \frac{b_2}{\xi}\operatorname{ch}\frac{d}{2\xi}\right]\theta_2 = 0 .
\end{aligned} \tag{71}$$

The condition $\Delta = 0$ leads to the following equation for the threshold voltage $U_{\mathrm{tr}}$ (with $\eta_1 = \eta_2$, $U_{\mathrm{H1}} = U_{\mathrm{H2}} = U_{\mathrm{H}}$)

$$\begin{aligned}
\coth\frac{\pi U_{\mathrm{tr}}}{U_{\mathrm{F}}} = &\left[\left(\frac{U_{\mathrm{F}}^2}{\pi^2 U_{\mathrm{H}}^2} - 1\right)\frac{\pi^2 b_1 b_2}{d^2} \times \right.\\
&\left.\times \frac{U_{\mathrm{tr}}^2}{U_{\mathrm{F}}^2} - \frac{b_1 - b_2}{d}\,\frac{U_{\mathrm{tr}}}{U_{\mathrm{H}}} - 1\right]\frac{d}{\pi(b_1 + b_2)}\,\frac{U_{\mathrm{F}}}{U_{\mathrm{tr}}}.
\end{aligned} \tag{72}$$

This equation has a solution if $U_{\mathrm{F}}^2/\pi^2 U_{\mathrm{H}}^2 < 1$. This means that a limiting value of the dielectric anisotropy in such a geometry there exists above which the layer is absolutely dielectrically stable — the flexoeffect is not capable of creating deformations at all. The same conclusion was reached by Fan [16] when considering the influence of the dielectric anisotropy on Meyer's domain formations. In our case when

$$\Delta\chi \to \Delta\chi_{\mathrm{lim}} \qquad U_{\mathrm{tr}} \to \infty .$$

The limiting anisotropy is :

$$\Delta\chi_{\mathrm{lim}} = \frac{(e_{1z} + e_{3x} + m_p)^2}{K_{33}}. \tag{73}$$

If the previously assumed value is taken for the total flexocoefficient, namely

$$e_{1z} + e_{3x} \sim 5 \times 10^{-5}\ \mathrm{dyn.}^{1/2}$$

then $\Delta\chi_{\mathrm{lim}} \sim 2 \times 10^{-3}$ which is an extremely small value and the appearance of the effect will require very careful compensation of $\Delta\chi$. But if

$$e_{1z} + e_{3x} \sim 5 \times 10^{-4}\ \mathrm{dyn.}^{1/2}$$

as already discussed in case 4.1.1, then $\Delta\chi \sim 2 \times 10^{-1}$. This means that the flexoelectric effect will be important for the usual nematic materials with small dielectric anisotropy, provided that the surface polarization does not act in the opposite direction. On the other hand in this geometry it is possible to display such an effect in nonflexoelectric materials, using surface polarization only.

Equation (72), due to the presence of the linear term $\dfrac{(b_1 - b_2)\,U_{\mathrm{tr}}}{dU_{\mathrm{H}}}$, has two different roots for positive and negative direction of $E$ — the effect remains polar. If the symmetrical case is considered, the following equation will be obtained for $U_{\mathrm{sym}}$ :

$$2\coth\frac{\pi U_{\mathrm{sym}}}{U_{\mathrm{F}}} = \left[\left(\frac{U_{\mathrm{F}}^2}{\pi^2 U_{\mathrm{H}}^2} - 1\right)\frac{\pi^2 b^2}{d^2}\,\frac{U_{\mathrm{sym}}^2}{U_{\mathrm{F}}^2} - 1\right] \times \frac{d}{\pi b}\cdot\frac{U_{\mathrm{F}}}{U_{\mathrm{sym}}}. \tag{74}$$

When $\dfrac{\Delta b}{b} \ll 1$ we can write for $U_{\mathrm{tr}}$

$U_{\mathrm{tr}} = U_{\mathrm{sym}} + \Delta U$, where $U_{\mathrm{sym}}$ is a root of (74) with $b = 1/2(b_1 + b_2)$ and $\Delta U$ is given by the equation

$$\Delta U = \frac{\Delta b}{2b}\left[\frac{\pi^2 U_{\mathrm{H}}}{U_{\mathrm{F}}^2 \operatorname{sh}^2\dfrac{\pi U_{\mathrm{sym}}}{U_{\mathrm{F}}}} + \frac{\pi^2 b}{2d}\left(\frac{U_{\mathrm{F}}^2}{\pi^2 U_{\mathrm{H}}^2} - 1\right)\frac{U_{\mathrm{H}}}{U_{\mathrm{F}}^2} + d/2b\,\frac{U_{\mathrm{H}}}{U_{\mathrm{sym}}^2}\right]^{-1}. \tag{75}$$

Now $\Delta U \sim \Delta b$ in contrast to case 4, where $\Delta E \sim (\Delta b)^2$ — eq. (58). Eq. (75) demonstrates explicitly how anchoring energy asymmetry produces polarity of the effect. Where $C_1 > C_2$ the positive threshold is lower than the negative in accordance with eq. (62) for $\Delta\chi = 0$.

4.2.3 *Dielectrically unstable layer* ($\Delta\chi < 0$). — If there are no surface torques in this case, a pure Freedericksz transition will take place at a threshold voltage given by an equation similar to eq. (51)

$$\operatorname{ctg} \frac{\pi U_{tr}}{U_F} = \left( \frac{\pi^2 b_1 b_2}{d^2} \frac{U_{tr}^2}{U_F^2} - 1 \right) \frac{d}{\pi(b_1 + b_2)} \frac{U_F}{U_{tr}} \quad (76)$$

$U_{tr}$ is independent of polarity.

The influence of flexoelectricity will express itself in that it will cause a decrease in the threshold for one polarity and an increase for the opposite polarity, provided that there exists a surface asymmetry. This feature has already been mentioned by Helfrich [12].

The equation for $U_{tr}$ derived in analogy to (72) by changing $U_F$ in $iU_F$ now reads :

$$\operatorname{ctg} \frac{\pi U_{tr}}{U_F} = \left[ \left( \frac{U_F^2}{\pi^2 U_H^2} + 1 \right) \frac{\pi^2 b_1 b_2}{d^2} \frac{U_{tr}^2}{U_F^2} - \frac{b_1 - b_2}{d} \frac{U_{tr}}{U_H} - 1 \right] \frac{d}{\pi(b_1 + b_2)} \frac{U_F}{U_{tr}} . \quad (77)$$

It reduces to eq. (76) if $U_H \to \infty$.

The equation for the symmetrical case is written to estimate the effect of the surface asymmetry on the polarity :

$$2 \operatorname{ctg} \frac{\pi U_{sym}}{U_F} = \left[ \left( \frac{U_F^2}{\pi^2 U_H^2} + 1 \right) \frac{\pi^2 b^2}{d^2} \frac{U_{sym}^2}{U_F^2} - 1 \right] \times \frac{d}{\pi b} \frac{U_F}{U_{sym}} . \quad (78)$$

If $\frac{\Delta b}{b} \ll 1$, $U_{tr} = U_{sym} + \Delta U$, where $U_{sym}$ is a root of eq. (78) with $b = 1/2(b_1 + b_2)$ and

$$\frac{\Delta U}{U_H} = \frac{\Delta b}{2b} \left[ \frac{\pi^2}{\sin^2 \frac{\pi U_{sym}}{U_F}} \frac{U_H}{U_F^2} + \frac{\pi^2 b}{2d} \times \left( \frac{U_F^2}{\pi^2 U_H^2} + 1 \right) \frac{U_H}{U_F^2} - \frac{d}{2b} \frac{U_H^2}{U_{sym}^2} \right]^{-1} .$$

Here, just as in case 4.2, $C_1 - C_2 > 0$ leads to a decrease of the positive threshold and an increase of the negative with respect to $U_{sym}$.

Another aspect of eq. (78) is worth mentioning. The value of $U_0$, the Freedericksz threshold with soft boundary conditions, can be measured using a high frequency electric field and symmetrical boundary conditions because over several hundred Hz the flexo-effect is not active. If a constant electric field is now applied, the value of $U_{sym}$ will be measured. The difference between these two values can be used to determine $U_H$ and consequently $(e_{1z} + e_{3x} + m_p)$ because $U_0$ is given by a special form of (78) taking $U_H \to \infty$. If the difference $\Delta U = U_{sym} - U_0$ is small enough, it is given by

$$\frac{\Delta U}{U_0} = - \frac{(e_{1z} + e_{3x} + m_p)^2}{|\Delta\chi| K_{33}} \times \left[ 1 + \frac{d^2}{b^2} \frac{U_F^2}{\pi^2 U_0^2} + \frac{d}{b} \sin^2 \frac{\pi U_0}{U_F} \right]^{-1}$$

Such a method of determining $(e_{1z} + e_{3x} + m_p)$ can provide some advantages as compared to Deuling's method [27] because for the latter extremely pure materials are needed. For MBBA an estimate of this difference for the case b $\sim$ d, i.e. $U_0 \sim (1/2)\, U_F$ gives $\Delta U/U_0 \sim 1/3$. Such a difference could be registered quite unambiguously.

4.3 PLANAR LAYER UNDER THE ACTION OF A HORIZONTAL ELECTRIC FIELD PARALLEL TO THE EASY AXIS. — As already noted, this case is formally equivalent to case a. A simplification of the solutions will take place if in the planar case the surface polarization $m_p$ is small enough. Then $\eta_1 = \eta_2$ (28) where we have $e_{1z}$ instead of $e_{3x}$. Also instead of $K_{33}$, we should put in the formulae (27)-(31) the splay elasticity constant $K_{11}$.

The importance of this geometry is that it gives the possibility of experimental determination of $e_{1z}$ only [28]. The corresponding formula describing the flexoelectrically induced change of the phase difference will look like (36) :

$$\delta(0) - \delta(E) = \frac{1}{24} n_e \left( \frac{n_e^2}{n_0^2} - 1 \right) \times \left( \frac{e_{1z}}{K_{11}} \right)^2 E^2 d^3 \frac{1 + 3(b_1 - b_2)^2/(b_1 + b_2)^2}{[1 + d/(b_1 + b_2)]^2} .$$

The problem is that in planar layers the anchoring is much stronger and one can hardly find a surface with $b > 1$ μm, so that very thin cells are required. The Rychenkow-Kleman technique [28] for depositing amorphous coating seems to be promising but unfortunately in that case the LC layer becomes spontaneously homeotropic. The construction of an asymmetrical cell could possibly help to solve the problem-namely imposing a strong planar anchorage on one of the glass plates which for elastic reasons should be capable of orienting the whole layer. The flexoelectric surface torque will then act only on the opposite amorphous carbon surface.

4.4 PLANAR LAYER UNDER THE ACTION OF A VERTICAL ELECTRIC FIELD. — Here again in formulae (27)-(31) we should set $m_p = 0$ so that $U_{H1} = U_{H2}$ and replace $K_{33}$ by $K_{11}$. As in the previous case the subcases with the same dielectric stability are with opposite dielectric anisotropy (Table I).

An important difference is that surface asymmetry induced polarity is just the opposite to the case b : the lower threshold corresponds to an EF in the direction strong anchoring electrode-weak anchoring electrode. The reason for this is that in the boundary conditions the flexoelectric terms enter with a sign opposite to that in (60).

As we have already pointed out such an asymmetric effect was registered by one of the authors. It is shown on figure 2.

5. **Conclusion.** — At least 16 cases of electrostructural deformations in a d.c. electric field are considered by the authors using a general theoretical approach with regard to different flexoelectric, dielectric, anchoring and surface polarization parameters of the nematic. In all the geometries considered the volume flexoelectric torque is zero and the flexoeffect comes into play with surface torques only which definitely requires weak anchoring for its manifestation.

The importance of these experimental geometries can be summarized in two points. First, some of them, e.g. the polar flexoeffects, could find interesting display applications. For this reason it is important to know the influence of the different dielectric and surface parameters on their performance. Such a polar effect has already been observed by the authors. Second, others have been proposed as methods for measuring the flexoelectric moduli of the material. The authors demonstrate here that such experiments could not give a direct means since in the total effect, the flexoeffect is related to the surface properties, often in a complicated manner. The evaluation of the moduli therefore from a single type of experiment is often not possible. The necessity of complementary experiments is obvious. The importance of the surface polarization can for instance be estimated only after a careful comparison of the results with hydrophilic and hydrophobic substrates.

On the other hand the exact determination of the flexoelectric parameters generates exact information about the boundary conditions as well. In this manner the results of the authors could be useful in studying surface interactions (e.g. the thickness dependence of the electrooptical effects). The importance of the exact knowledge about the surface properties of liquid crystals need not be specifically stressed. The situation in this field now resembles solid state physics where volume properties are much better understood than the surface ones. The development of methods for weak and anisotropic anchoring will bring about a great advance. This will give the possibility of splay flexocoefficient measurements.

Interesting new information can be obtained from the dynamic behaviour of these surface driven effects, e.g. information can be obtained about the dissipation of energy on the surface. Some preliminary theoretical and experimental studies of flexoelectric oscillations have already been performed and reported elsewhere [5, 6].

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