

On the “Gradient Flexoelectric Effect” in Nematics Thirty Years Later*

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Abstract. The “gradient flexoelectric effect” in nematics discovered in 1974 by Derzhanski *et al.* [8] is considered thirty years later. The following topic questions are discussed: the kind of the non-homogeneity of the electric field in the presence of ions or in non-planar geometry, or in planar cells with interdigital, quadrupolar, etc. position of the electrodes, the boundary conditions and their dependence on the ions and the flexoeffect, the sign and the value of some flexoelectric coefficients. The experimentally observed in 1974 flexodeformations are discussed on the basis of the type of the electric field, calculated for a simpler electrochemical liquid cell [49].

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1 Introduction

The piezoelectric effects in nematics were proposed for the first time by Meyer in 1969 [1]. This theory was enunciated by analogy with the theory of piezoelectricity in solids. The basic idea of Meyer was that the macroscopic splay ($\text{div } \mathbf{n}$) and the macroscopic bend ($\mathbf{n} \times \text{rot } \mathbf{n}$), where \mathbf{n} is the unit vector showing the mean direction of the liquid crystal molecules along one preferred axis, called by Frank [2] director, can lead to macroscopic polarization (Figure 1). From this picture it is clear that the initial non-deformed state of the nematic is not polarized since the permanent electric dipoles of the molecules are locally compensated, which can be removed in two ways:

- (a) The confinement of the liquid crystal in a non-planar geometry with imposed homogeneous boundary conditions leads to macroscopic polarization of the liquid crystal by the so-called “direct piezoeffect”, and

*This work is dedicated to Professor Alexander Derzhanski, DSci., Corresponding Member of the Bulgarian Academy of Sciences, on the occasion of his 70th anniversary.

- (b) The exposition of the liquid crystal under the action of a DC voltage or AC voltage, when the frequency is sufficiently low, leads at appropriate conditions to the observation of piezodeformations caused by the so-called “converse piezoeffect”.

This piezoelectric effect for both examples has been called by Meyer a “dipolar piezoelectric effect”. There are similarities and differences between the piezoelectricity in the crystals [3] and the piezoelectricity in the liquid crystals. For instance, one similarity is that the **splay** or **bend** of either the liquid crystal or the piezoelectric crystal should lead to a macropolarization. A second similarity is that “converse piezoeffect” can exist in both: crystals and liquid crystals. There is one important difference, which should be mentioned. The existence of macro-dislocations in the piezoelectric crystals should decrease the net polarization, since the piezoeffect in the crystals is connected with the crystal lattice, while around the disclinations in an insulating nematic, there should exist a very strong polarization [4] (the situation is quite different, however, in a conductive liquid crystal due to the screening effect of the impurity ions [5]). De Gennes suggested the term “piezoelectricity” to be replaced by “flexoelectricity” [6] and it is now accepted in the terminology of the liquid crystals (the term “flexoelectricity” can be found in the crystal terminology as well [7]).

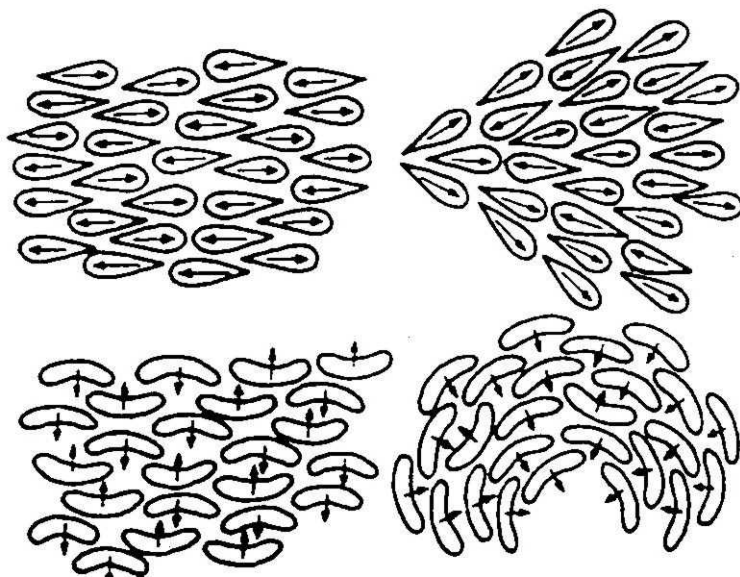


Figure 1. The dipolar model of R. B. Meyer [1] for unstrained nematic structures containing polar molecules with wedge-shaped and banana-shaped molecules (left) and the splayed structure and bent structure (right) coupled either with the non-planar geometry (direct flexoeffect) or with an applied external electric field (converse flexoeffect).

2 On the “gradient flexoelectric effect”: a history and topic questions

In this paper the “gradient flexoelectric effect” in nematics, theoretically proposed and experimentally observed by Derzhanski *et al.* in 1974 [8], is considered thirty years later. The basic idea of Derzhanski was first, that the electric field in conventional nematic cell of a sandwich type under the influence of a DC or low-frequency electric field should be non-homogeneous due to the space charges accumulated near the electrodes, and second, that the coupling of this non-homogeneous electric field with the flexoelectric properties of the nematic should lead to the creation of a new flexoelectric effect, called by him “gradient flexoelectric effect”. Indeed, such an effect was experimentally observed by the Sofia Liquid Crystal Group during 1972-1974 after the application of a lower DC voltage across thin nematic cells of MBBA, PAA, APAPA, *etc.* with a thickness between 10 and 100 micrometers. At these conditions, the integral intensity of the transmitted through the liquid crystal polarized light increases due to the flexodeformations (Figure 2), while the application of a high-frequency electric

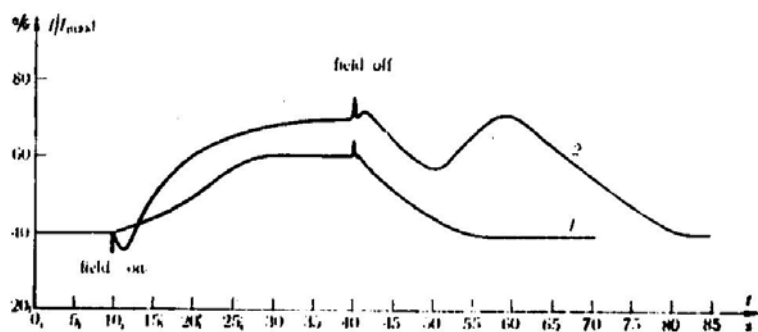


Figure 2. The registered by A. Derzhanski *et al.* gradient flexoelectric effect [8]: the intensity of the transmitted light as a function of the applied DC voltage: curve 1 — $U = 1$ V, curve 2 — $U = 2$ V.

field with a frequency above the cut-off for the relaxation of the ions, decreases the intensity. The flexodeformations could be registered at a very low voltage with a magnitude of millivolts. During the experimental research of the gradient flexodeformations it was understood that additionally there are two other important requirements for the observation of the gradient flexoeffect: weak anchoring of the liquid crystal and existence of initial deformations of the director \mathbf{n} . In the beginning, after the report on the “gradient flexoelectric effect” in 1974, it did not receive much attention among the researchers working in the field of the liquid crystals. In our opinion, the reason was the common view that the flexodeformations are “ghost” deformations. For instance, in the book of De Gennes [6] it was noted that the experimental observation of the flexoeffect in nematics

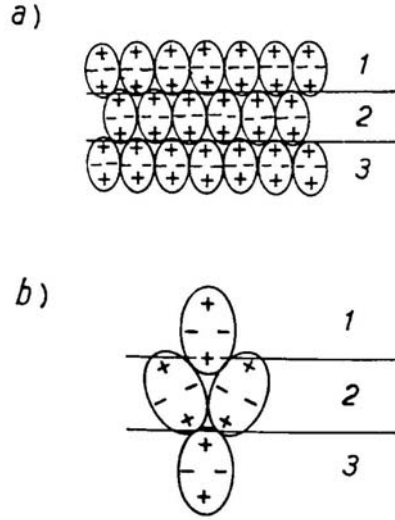


Figure 3. Quadrupoles and quadrupolar flexoeffect [9] and the “gradient flexoeffect”: a) there is no bulk polarization, b) quadrupoles with a splay imposed within the central “layer”, plus charges from above are allowed to enter, while plus charges below are expelled, leading to a net polarization upwards.

is questionable due to its subtle character. The situation changed considerably after the work of Prost and Marcerou on the quadrupolar flexoeffect in nematics [9]. These authors, regarded for the first time the question about the existence of quadrupolar flexoelectricity. For the case of a pure splay this effect is illustrated in Figure 3, which clearly shows that it is in effect a gradient flexoeffect (the theory of Prost and Marcerou has been reconsidered by Goossens [10] in relation with the “order electricity”). We shall not deal with the basic torques, related to the flexoelectricity, explained in detail in several papers of Derzhanski et al [11-14]. Important questions about the quadrupolar flexoeffect in nematics have been considered by Durand [15]. Nevertheless, we shall rewrite the original differential equation obtained for the first time in [8], which includes the gradient flexoelectric term:

$$\nabla^2\theta - ((e_{1z} + e_{3x}/K)(dE/dz) - (\Delta\varepsilon/4\pi K)E^2(z)) \sin\theta \cos\theta = 0 \quad (1)$$

where θ is the deformation angle (see Figure 4), K is the mean elastic constant of the liquid crystal, $(e_{1z} + e_{3x})$ is the total flexoelectric coefficient, responsible for the bulk gradient flexoelectric torques and $\Delta\varepsilon$ is the dielectric anisotropy of the liquid crystal, which for the given case (MBBA) is negative. In [8], two types of boundary conditions have been considered: a strong planar anchoring, without initial deformation of the liquid crystal (case A), and a strong periodic anchoring of the liquid crystal (case B). According to our knowledge, this is the

On the “Gradient Flexoelectric Effect” in Nematics

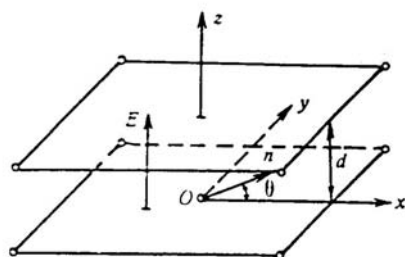


Figure 4. A schematic representation of a sandwich nematic layer. The directions of the axis z and the applied external DC voltage coincide, while the axis x is along the initial orientation of the liquid crystal, n is the director, θ is the deformation angle.

first periodic boundary condition included in the theory of liquid crystal deformations. Furthermore, the authors of [8] introduced two types of electric field: a linear field acceptable for thinner liquid crystal cells, and a hyperbolic electric field which is justified for thicker cells. The numerical solution of the problem for the case B clearly showed the existence of flexoelectric deformations, which for the chosen sign of the total flexocoefficient (positive) leads to bigger flexodeformations in the region of the anode for a linear electric field (see Figure 3 in [8]), while the calculated flexodeformations for a hyperbolic electric field have been in the region of the cathode (see Figure 5). The flexoeffect itself is shown in Figure 6, which clearly illustrates the flexoelectrical texturing of the nematic layer. It should be mentioned that the calculated flexodeformations in [8] were very small, in the range of several percent. There are two causes for this value. First, in the numerical procedure, a very small value of the total flexoelectric coefficient $e = 4 \times 10^{-5} \text{ dyne}^{1/2} ((4/3) \times 10^{-12} \text{ C/m})$ has been taken and, second, it was assumed, as noted, a strong anchoring of the liquid crystal. Let us stress

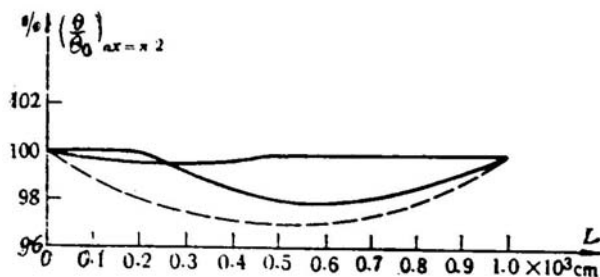


Figure 5. The initial deformations (the dashed line) and the flexoelectric deformations for the case of a linear electric field (left, upper solid line) and a hyperbolic electric field (left, lower solid line)(after [8]).

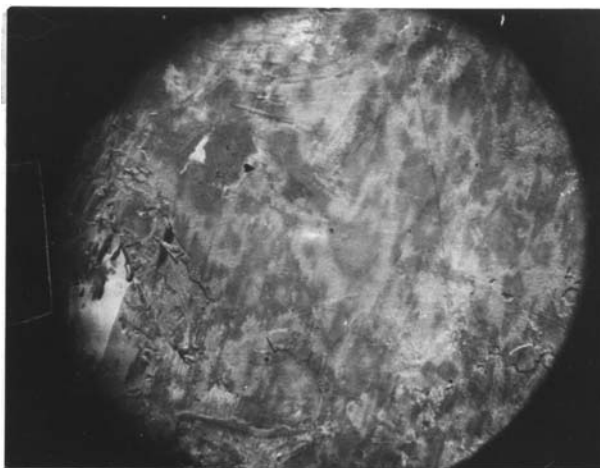


Figure 6. Flexoelectrically textured MBBA layer with weak anchoring at a DC voltage of 1 V (similar flexoelectric deformations were observed during the study of the gradient flexoeffect in [8]) (from Diploma work of Marin Mitov, Sofia University, 1974).

that by 1974 there was only one measurement of the flexoelectric coefficient of bend $e_{3,x}$ by Helfrich [16], which was based on the bending of a homeotropic nematic layer in a transversal electric field, first observed by Haas *et al.* [17]. The same flexocoefficient $e_{3,x}$ has been re-measured by Schmidt *et al.* [18]. The measured flexocoefficient $e_{3,x}$ has been very small, probably due to the surface polarization, first mentioned by Prost and Pershan [19] and further elaborated by Petrov and Derzhanski [20]. Now, it is well known that the value of the total flexoelectric coefficient for the case of MBBA is one order of magnitude bigger [21,22,14]. It should be pointed out, however, that there is no consensus about the magnitude and the sign of the total flexocoefficient of MBBA [14]. In addition, the recent measurement of this coefficient performed by Blinov *et al.* [23] by pyroelectric method yield again a small value: $(1.7 \pm 0.7) \times 10^{-12}$ C/m or $(5.1 \pm 2.1) \times 10^{-5}$ dyn^{1/2}. As far as the sign is concerned, both signs (+) [8,23] and (-) [22,24] have been pointed out by the authors (see also [14]). The second important cause for the calculation of so-small flexoelectric deformations was the acceptance of strong anchoring of the nematic irrespective of the initial state of the liquid crystal: planar or periodically deformed. The inclusion of a soft boundary condition with a definition of a relatively small surface energy of interaction between the walls confining the liquid crystal and the liquid crystal itself ($W_s = 10^{-3}-10^{-4}$ erg/cm²) ($(10^{-6}-10^{-7})$ N/m²) should considerably increase the magnitude of the flexoelectric deformations (it is sufficient to regard the flexoelectric deformations shown in Figure 6: let us stress that the initial state of the nematic cell is with different texturing, which can change even at a DC voltage of 1 V). The simplest, but still including the flexoeffect boundary

On the “Gradient Flexoelectric Effect” in Nematics

condition is of the form

$$K\theta' \pm ((e_{1z} + e_{3x})E - W_s) \sin \theta \cos \theta = 0 \quad (2)$$

where the mean elastic coefficient K is positive, while θ' and $(e_{1z} + e_{3x})E$ can be either positive or negative (the surface polarization is ignored). This boundary condition clearly shows that at an appropriate sign of the total flexocoefficient the flexoelectric effect itself can lead to surface deformed flexoelectric torques, which even at a homogeneous electric field cause flexoelectric deformations [11-18,22].

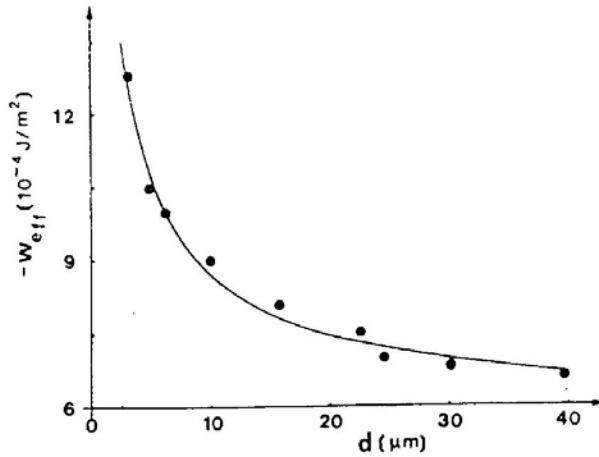


Figure 7. Experimental data given by Blinov and Kabaenkov [59] of the effective surface energy versus the thickness of the cell concerning one initially planar 5CB nematic (points) and the theoretical curve obtained by means of the proposed model by Alexe-Ionescu, Barbero and Petrov, taking into account dielectric and quadrupolar flexoelectric couplings at the boundary ([28]).

Another important problem connected with the gradient flexoeffect concerns the real value of the surface energy in the presence of ions in the liquid crystal, when the electrodes adsorb these ions [25-27]. This problem has been considered by Alexe-Ionescu *et al.* [28] (see Figure 7) and by Barbero *et al.* [29] for the case of weak adsorption of the ions and by Barbero and Olivero [30] for the case of strong adsorption of the ions. This question is of importance also for the surface polarization [31] and for the liquid crystal displays [32]. The presence of impurity ions (inherent or doped) in the liquid crystal leads to the formation of double electric layers which are saturated at a DC voltage of 1 V while the inverse voltage, concentrated within the double electric layers is of the order of 0.4 V [33-35]. Another challenge is the solution of the Gouy-Chapman equation. The exact solution of this equation is expressed in elementary logarithmic and inverse trigonometric functions [30,36,37]. A very good approximation for the

electric field near the electrodes is the exponential variation with the distance [30,31] for the case of weak adsorption of the ions whereas for the case of strong adsorption the kind of the electric field is more complex (see Eq.(6) in [36])(see also the calculations in [30]). There are also examples of a hyperbolic kind of the electric field near the electrodes at certain assumptions [38].

3 The electrochemical behaviour of the liquid crystal and the “gradient flexoeffect”: an example

The non-homogeneity of the electric field inside a liquid crystal cell is determined not only by the presence of impurity ions, but also by the electrochemical behaviour of the given liquid crystal. This is especially important for the case of the liquid crystal MBBA (methoxy-benzylidene-para-butyl-aniline), which is not a stable chemical substance [39-43] (we refer only to the first papers devoted to this problem). The electrochemical behaviour of every liquid or every liquid crystal is revealed by the current-voltage or by transition current curves (see, for instance, the results presented in [44] and [45]). Detailed calculations of the electric field can be found, for instance, in [46] and [47]. The possible degradation of the liquid crystal under the action of a DC or low-frequency electric field can be considerably or totally removed by adding a small amount (several wt%) of a substance, which shows reversible electrochemical reactions at a very low redox or oxide potential [48]. Nevertheless, ions generated by the electrochemical behaviour of the liquid crystal are evidently connected with the observation of the “gradient flexoeffect” in nematics. We would like to illustrate this (among the many papers devoted to this question) by the electrochemical behaviour of one electrolyte-free liquid used for the observation of electrochemiluminescence [49], [50]. There are great similarities between the flexoeffect observed in one MBBA cell and the electrochemiluminescence observed in such cells. The electrochemical current as a function of the applied voltage V , measured for such a cell with a different electrode spacing d is shown in Figure 8. The calculated real value of the electric field inside the cell for a different value of the applied

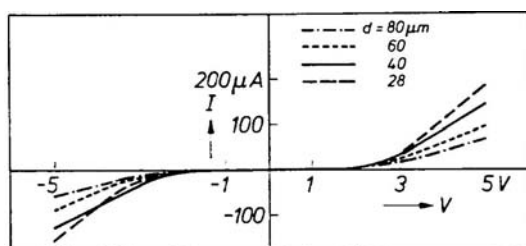


Figure 8. The current-voltage curves for the electrochemical cell of Schaper, Köstlin and Schnedler (see [49]) with a different electrode spacing d .

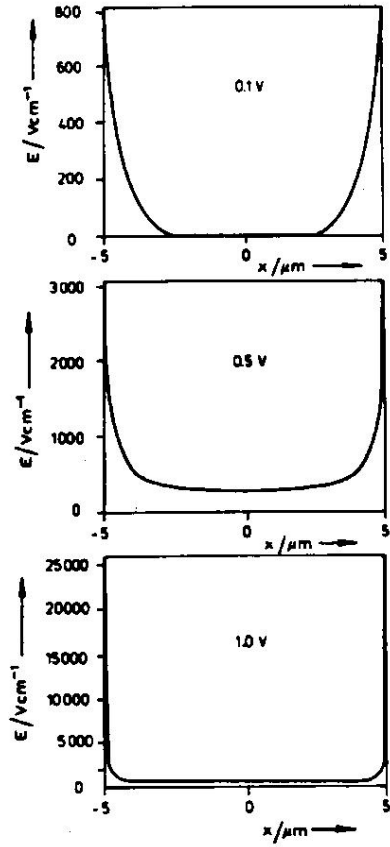


Figure 9. The calculated real value of the electric field inside the cell of Schaper *et al.* (see [9]) for three different values of the applied DC voltage: 0.1 V, 0.5 V, and 1 V (the thickness of the cell is 5 μm after [50]).

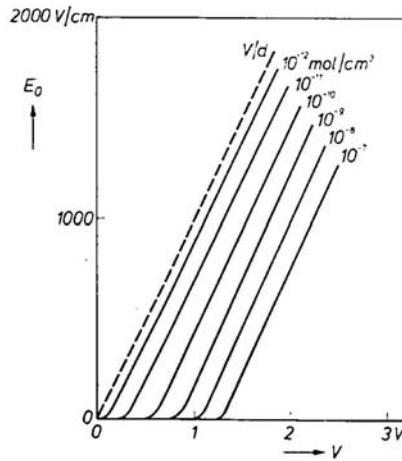


Figure 10. The calculated mid-plane electric field-strength E_0 versus applied voltage V at different ion concentrations (the thickness of the cell is 10 μm) for the cell of Schaper *et al.* (after [50]).

voltage (0.1–1 V) is shown in Figure 9. The calculated mid-plane electric field-strength E_0 as a function of the applied voltage V , for the electrochemical cell with electrode spacing d of 10 μm , at different ion concentrations is shown in Figure 10. The calculated electric field-strength E_s near the electrodes as a function of the applied voltage V for the same cell and different ion concentrations is shown in Figure 11. E_s increases strongly while E_0 decreases with increased ion concentration (Figure 10). Since the gradient of the electric field is stronger near the electrodes (Figure 11) one expects, the stronger flexoeffect to be namely in these regions. The most interesting curves are shown in Figure 12. They rep-

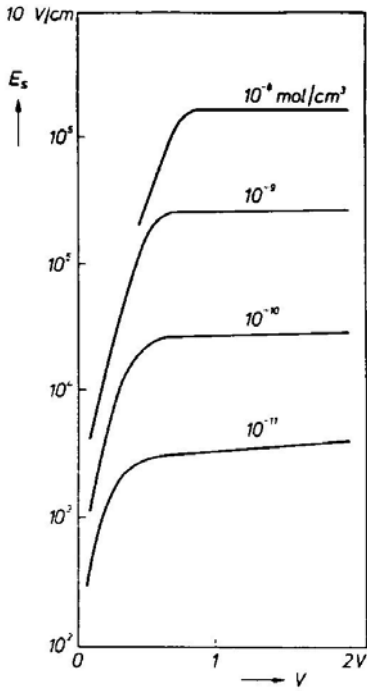


Figure 11. The calculated electric field-strength E_s near the electrodes versus the applied voltage V for the same cell of Schaper *et al.* at different ion concentrations (after [49]).

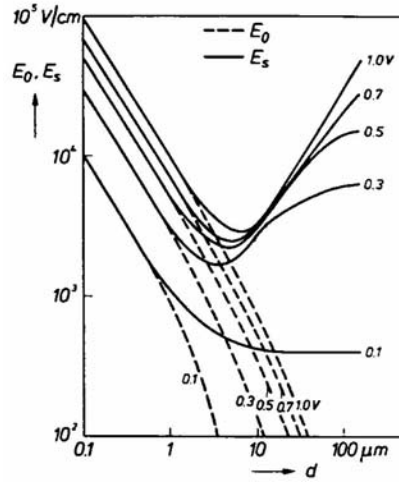


Figure 12. The calculated mid-plane field-strength E_0 (the dashed curves) and the calculated electric field-strength E_s near the electrodes (the solid curves) versus the thickness of the cell and the applied voltage.

resent E_0 and E_s at an ion concentration of 10^{-11} mol/cm² as a function of the electrode spacing d at different voltage. It is seen that the gradient of the electric field is very complex at a DC voltage with a magnitude of 0.3 V. Consequently, the electrooptic response in one liquid crystal cell with a similar ionic behaviour would be complex, with a flexoelectric deformation at one of the two electrodes and flexoelectric and dielectric alignment at the other electrode. It is evident that the change of the sign of the total flexocoefficient would lead to the interchange of the behaviour of the liquid crystal at both electrodes.

4 On the “gradient flexoeffect” and some experimental and theoretical questions

The gradient flexoelectric effect can be observed in any non-homogeneous electric field, which could be realized under an appropriate form of the cell, confin-

ing the liquid crystal [51-53] or under appropriate disposition of the electrodes, for instance interdigital [19,21,54,55] or quadrupolar [24], *etc.* Evidently, such kinds of electric field can be used not only at a DC voltage but at an alternating electric field with a frequency below, for instance, 5 kHz since the flexoeffect can be observed in this frequency region.

Finally, we stress that it is possible to separate the flexoelectric deformations arising in a gradient electric field from the flexoelectric deformations arising in a homogeneous electric field and gradients in the director \mathbf{n} . Mathematically this calculation has been done for the first time by Derzhanski and Petrov [56] on the basis of theoretical results obtained by Fan [57], which concern the bulk distribution of the flexoelectricity:

$$\mathbf{h}_f = e_{1z}(\mathbf{E}\nabla \cdot \mathbf{n} - \nabla(\mathbf{E} \cdot \mathbf{n})) + e_{3x}(\mathbf{E} \times \nabla \times \mathbf{n} - \nabla \times (\mathbf{E} \times \mathbf{n})) \quad (3)$$

where \mathbf{h}_f is the bulk flexoelectric component of the molecular field [6,11,57]. This expression can be easily transformed into an equivalent form, which we prefer to write as

$$\mathbf{h}_f = (e_{1z} - e_{3x})\mathbf{h}_{f1} + (e_{1z} + e_{3x})\mathbf{h}_{f2} \quad (4)$$

The exact form of this expression [56] shows that the bulk flexodeformations can be observed when there is a gradient in the distribution of either the nematic director, or an external electric field, or both. Effects of the first kind are known as linear (with respect to the electric field) and they depend on the difference of splay and bend flexocoefficients. Those of the second kind are known as gradient effects, and they depend on the sum of both flexocoefficients. It can be easily shown that for planar problems, where the director is confined to a plane, the first term in q. (4) is identically zero. In such a case, with a homogeneous field, the only source of flexodeformation comes from the surface torque \mathbf{g}_f , the flexoelectric surface molecular field [11] defined by the following equation:

$$\mathbf{g}_f = -e_{1z}(\mathbf{E} \cdot \mathbf{n})\mathbf{s} + e_{3x}(\mathbf{s} \times (\mathbf{n} \times \mathbf{E})) \quad (5)$$

where \mathbf{s} is the unit surface normal.

Let us point out that Eq. (2) is a particular case of Eq. (5) (most of these considerations have been done by Petrov in [14]).

5 Conclusions

In this paper we have considered only the one-dimensional flexoelectric deformations induced by the “gradient flexoeffect”. The two-dimensional flexoelectric deformations are more complex and will not be considered here. The periodic flexo-deformations are also complex and deserve to be considered in a separate paper [58]. We finish this short review paper with a citation from the

Report of three famous scientists: P. G. de Gennes (with a Nobel Prize in physics for 1991), E. Guyon and J. Prost, written in 1979 for the synopsis of the Dr. Sciences Thesis of A. Derzhanski entitled "Flexoelectricity in Nematic Liquid Crystals": "... But a most original contributon of Derzhanski was to propose that the presence of inhomogeneous field within the cell (caused by existence of space charges near the electrodes) could also lead to the observation of flexoelectricity. Although not widely accepted at the start, the results of this "gradient flexoelectric effect" have proved to be essentially correct and have been confirmed by different techniques ...".

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