

# Surface-Induced Flexoelectric Domains in Asymmetrically Strong-Weak Anchored MBBA Films

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The longitudinal surface-induced flexoelectric domains, previously obtained in symmetrically weak-anchored nematic layers have been further investigated in asymmetrically strong-weak anchored MBBA layers. The threshold voltages indicating the onset of the flexoelectric instabilities are high for thin cells ( $d = 9\ \mu\text{m}$ ,  $U_{\text{th}} = 10\ \text{V}$ ) and small for thick cells ( $d = 150\ \mu\text{m}$ ,  $U_{\text{th}} = 1.5\ \text{V}$ ). Pronounced minima in the curves  $p$  vs.  $d$  and  $U_{\text{th}}$  vs.  $d$ , where  $p$  is the domain period and  $d$  is the cell thickness, were obtained.

The flexoelectric domains were replaced by electrohydrodynamic instabilities only in the case of strong azimuthal anchoring of the liquid crystal layer.

The role of the sign and the value of the dielectric anisotropy, the homogeneity of the electric field as well as the asymmetry in the surface anchoring are discussed in detail.

## 1. Introduction

Flexoelectric domains in nematics have been proposed for the first time by Meyer [1]. A critical value of the dielectric anisotropy beyond which the domain structure cannot exist has been found independently by Meyer [2] and Fan [3].

Longitudinal electric field gradient stimulated domains in MBBA, PAA and APAPA have been experimentally observed and investigated by Markovski and Petrov [4] and later by Derzhanski and Hinov [5], who related their existence to the presence of flexoelectric properties.

The threshold voltage and the period of a new kind of flexoelectric two-dimensional domains arising from strongly-anchored planar nematics have been calculated by Bobylev and Pikin [6], [7]. A variable grating mode, discovered by Vistin' [8] has been interpreted by Barnik et al. [9], [10] in terms of the two-dimensional flexoelectric Bobylev-Pikin theory.

Longitudinal surface-induced flexoelectric domains created in weakly-anchored nematics with small dielectric anisotropy were recently investigated by us [11], [12]. The weak surface anchoring of the liquid crystal (LC) molecules was crucial for the formation of these domains. The flexoelectric deformations including variations in the

magnitude of the polar and azimuthal angles  $\theta$  and  $\varphi$  were very complex. The experimental results revealed a wall structure leading to a distinct separation of adjacent domains.

The shape of the flexoelectric domains was very different depending on the type of the boundary conditions, the amount of tilting of the molecules in the fieldless state, etc.

The aim of this paper is to propose a simple way for a reproducible and easy creation of flexoelectric domains in nematics under asymmetrical strong-weak anchoring conditions. The threshold voltages and the domain periods are measured for various thicknesses of the nematic layers. The crucial role of the azimuthal  $\varphi$ -anchoring of the layer is experimentally demonstrated. The possible contributions of the isotropic [13], electrohydrodynamic [14], [15], [16] and flexoelectric mechanisms are discussed in detail.

## 2. Experimental Setup and Results

The confining glasses of the LC cells coated with transparent conductive layers of  $\text{SnO}_2$  were cleaned by standard methods. The LC orientation was determined after rubbing with diamond paste (grain size  $0.25\ \mu\text{m}$  and  $0.5\ \mu\text{m}$ ) or by vacuum evaporation of  $\text{SiO}$  thin films. The surfactants utilized in the experiment for obtaining the weak anchoring were Na salts of fatty acids (common soap) and commercially available aliphatic monoamine  $\text{C}_8\text{H}_{17}\text{NH}_2$ . The way of the soap deposition was

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different from that described in [12]. We noticed that the efficiency of the soap deposition was higher when rubbing on with a cloth. The soap film was very thin and not observable with the naked eye. The weights of the glasses before and after treatment were equal within  $10^{-5}$  g. The coupling of the LC with the surfaces treated in this way was weak and the molecules were tilted without any degeneration.  $C_8H_{17}NH_2$  was deposited after adsorption of a monomolecular film [17], [18]. The cell gap was determined by mylar spacers, 9 to 205  $\mu\text{m}$  thick. The observations were performed under a Leitz type microscope in transmitted polarized monochromatic or white light. The nicols were normally crossed with the polarizer perpendicular to the direction of rubbing or rotated by an angle  $(\pi/2) - \varphi$  when  $\varphi$  deformations were being investigated. All the investigations were performed at room temperature.

Flexoelectric domains can be obtained more easily in asymmetrically-anchored nematic layers with strong anchoring of the LC molecules at one of the boundaries and weak anchoring at the other. The strong-weak anchoring of the LC molecules was achieved by means of rubbing of the two glass plates with diamond paste (or by  $\text{SiO}$  thin film deposition) followed by soap deposition on one of them.

The microscopic observations carried out on strong-weak anchored MBBA nematic films clearly show the crucial role of the strong anchoring for a realization of a relatively homogeneous LC orientation established after the relaxation of the metastable regions with different tilt (Figure 1). The strong-weak anchoring of the LC layer made possible the creation of surface-induced domains for thicknesses up to 210  $\mu\text{m}$ , which was not achievable with weakly-anchored MBBA layers [12]. The ratio  $p/d$ , where  $p$  is the domain period and  $d$  the thickness of the film, decreases with increasing  $d$  up to 150  $\mu\text{m}$  in accordance with the experimental results obtained for weakly-anchored MBBA layers [12] while the threshold voltages, higher for thinner cells and lower for thicker cells, were very different from those measured for weakly-anchored MBBA layers (Figs. 2 and 3) [12]. Of special interest are the surprisingly-small threshold voltages: 1.5–2 V obtained with thick MBBA cells. Up to now these threshold voltages for domain formation are the smallest ones ever obtained. The minima in the experimental curves are due to the limit of the

penetration depth of the strong surface forces, which was measured to be around 150  $\mu\text{m}$  [19], [20]. Above this thickness the LC orientation at the soap-treated wall was determined by local causes such as hydrodynamic movement during the filling of the cells [21], [22], thermal fluctuations etc. The domain formation in these cases was chaotic and slow (minutes after the voltage application).

The domain appearance in thin (Fig. 4a) and thick (Fig. 5a) LC cells as well as the development of the  $\theta$ ,  $\varphi$  LC flexoelectric deformations (Figs. 4b and 5b) were very similar to those reported in [12]. The optical textures of the flexoelectric domains in strong-weak anchored MBBA films confirmed again the wall structure of this special flexoelectric deformation, i.e. the novel anchoring did not change the type of the flexoelectric domains. As far as the detailed  $Y$ ,  $Z$  deformations are concerned we may say that the asymmetrical case is characterized by strong realignment of the nematic director along the  $Z$  direction, i.e. normal to the glass plates. In addition, the strength of the thermal fluctuations resulting in a flickering effect was much smaller due to the strong anchoring of the molecules at one of the boundaries (see the dark regions of the domains shown in Figure 5b). The LC deformations appeared to be finite at the threshold voltage (first-order transition) and increased with the voltage across the nematic layer up to  $90^\circ$  ( $45^\circ$  rotation of the polarizer).

The experimental results have shown that the surface-induced domains arise in the cathode region (after the soap-treatment of the cathode), in agreement with the previous experimental results [12]. Longitudinal surface-induced domains can arise as well in the anode region (after the soap-treatment of the anode) at higher voltages. However, it is difficult to understand the nature of these domains. They can be flexoelectric or electrohydrodynamic of Pikin's type. The novel anode-created domains were observed only after the application of an additional stabilizing a.c. electric field erasing the Williams' mode.

Asymmetrically-anchored MBBA layers were prepared as well with one aliphatic monoamine  $C_8H_{17}NH_2$  layer deposited by means of the well-known drawing technique allowing the adsorption of one or several monomolecular films [23], [24], [25]. In cells prepared in such a way we observed only  $\theta$ -hydrodynamic instabilities without any

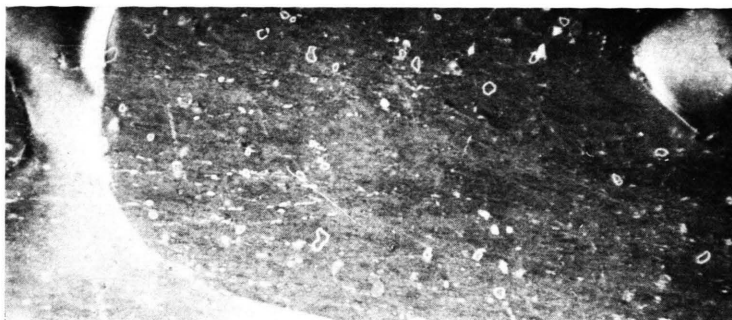


Fig. 1. An asymmetrical strong-weak anchored MBBA nematic layer, 42  $\mu\text{m}$  thick, after the I-N phase transition. The strong-weak anchoring of the liquid crystal molecules was achieved by means of a thin evaporated SiO film on one of the glass plates and soap rubbing of the other plate previously also treated with thin SiO film. The short side of the photo corresponds to 1350  $\mu\text{m}$ , crossed nicols.

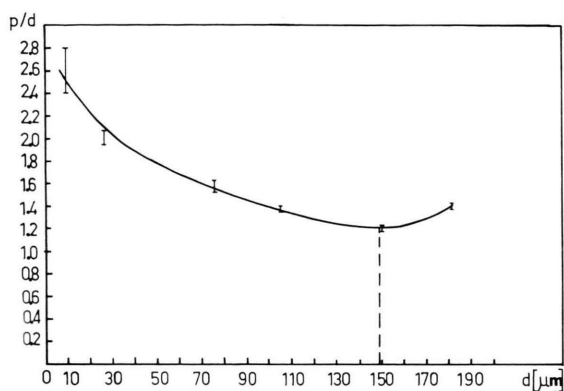


Fig. 2. The ratio  $p/d$  ( $p$  is the period of the flexoelectric domains) in strong-weak anchored MBBA nematic films versus cell thickness  $d$ . The minimum in the curve shows the penetration depth of the strong surface forces.

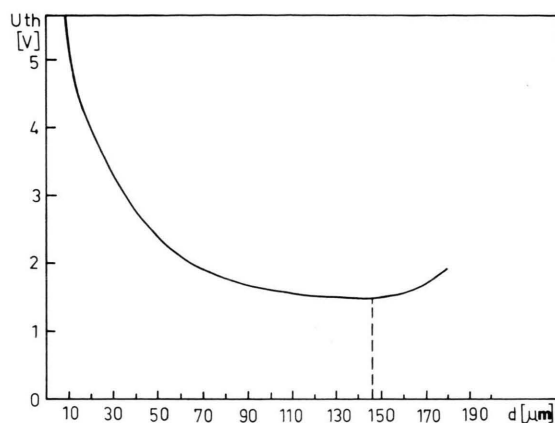
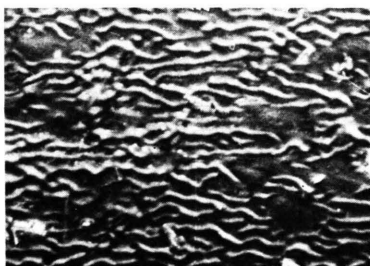


Fig. 3. The voltage threshold  $U_{\text{th}}$  indicating the onset of domain formation in strong-weak anchored MBBA nematic films vs. cell thickness  $d$ . The minimum in the curve shows the penetration depth of the strong surface forces.

a)



b)

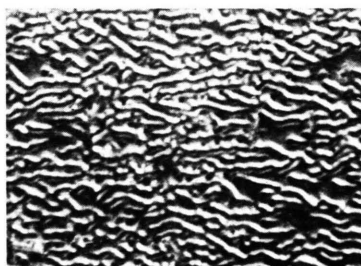


Fig. 4a. Onset of domain formation in a thin (9  $\mu\text{m}$ ) strong-weak anchored MBBA nematic layer at threshold voltage. Initially crossed nicols:  $n_0 \parallel A \perp \vec{P}$ ; The polarizer was rotated by an angle of approximately  $5-6^\circ$ . The applied d.c. voltage across the layer was 10 V. The short side of the photo corresponds to 1350  $\mu\text{m}$ .

Fig. 4b. The same MBBA nematic layer in the presence of d.c. voltage of 14 V.

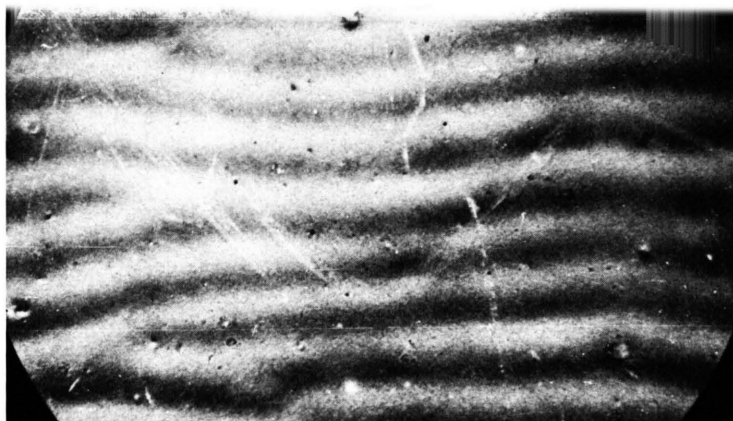


Fig. 5a. Onset of domain formation in a thick ( $105\text{ }\mu\text{m}$ ) strong-weak anchored MBBA nematic layer at threshold voltage. Initially crossed nicols;  $n_0 \parallel A \perp P$ . The polarizer was rotated by an angle of approximately  $20^\circ$ . The applied d.c. voltage across the layer was 2 V. The short side of the photo corresponds to  $1350\text{ }\mu\text{m}$ .

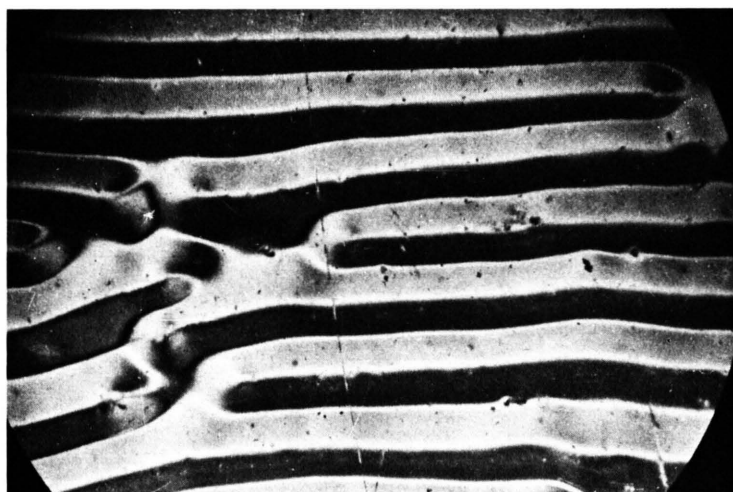


Fig. 5b. The same MBBA nematic layer when the magnitude of the applied d.c. voltages was 7 V. Initially crossed nicols;  $A \parallel n_0 \perp P$ . The polarizer was rotated by an angle of approximately  $35^\circ$ .



Fig. 6. The flexoelectric static domains were totally replaced by hydrodynamical ones in an MBBA layer,  $26\text{ }\mu\text{m}$  thick, when the glasses were treated with SiO thin films followed by one layer of aliphatic monoamine  $\text{C}_8\text{H}_{17}\text{NH}_2$  deposition on one of them. The magnitude of the applied voltage was about 3.5 V,  $n_0 \parallel P \parallel A$ . The short side of the photo corresponds to  $1350\text{ }\mu\text{m}$ .

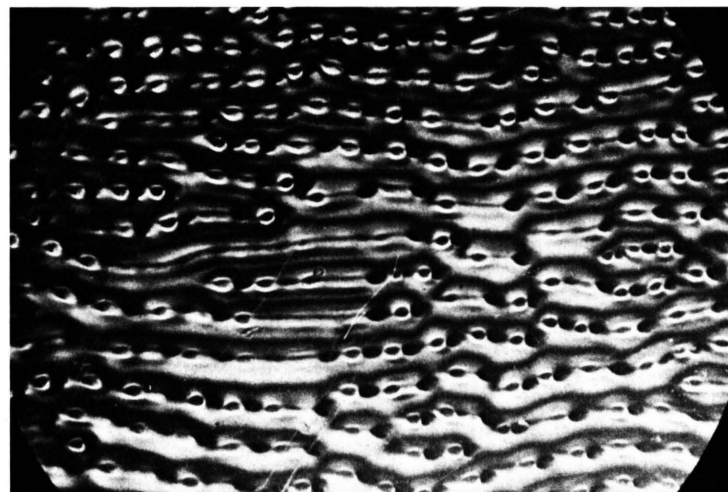


Fig. 8. Novel hydrodynamics in the form of isolated loops generated from the surface-induced flexoelectric domains in a strong-weak anchored MBBA layer,  $60\text{ }\mu\text{m}$  thick. A  $15^\circ$ ,  $n_0 \perp P$ . The applied voltage across the layer was 5 V. This kind of hydrodynamics is characteristic for thick soap deposition and liquid crystal thickness below  $100\text{ }\mu\text{m}$ .



formation of static surface-induced domains (Figure 6). This electrodynamic mode was easily erased with the aid of an additionally applied a.c. electric field. The further increase of the d.c. voltage led to movement of the fluid chiefly along the walls.

### 3. Discussion

Before giving evidence of the flexoelectric character of the surface-induced static domains we shall discuss possible contributions due to Felici's and Pikin's mechanisms.

#### a) Felici's Mechanism

One notes a great similarity in the behaviour of the flexoelectric and isotropic instabilities revealed in their strong dependence on the amount, sign and distribution of the space charges closely related with the nature and treatment of the electrodes. For instance, the time necessary for the flexoelectric domains appearance in asymmetrically strong-weak anchored MBBA layers is either milliseconds for thin LC cells with a thickness below  $10\text{ }\mu\text{m}$  or several seconds for  $150\text{ }\mu\text{m}$  thick cells. In addition, the flexoelectric domain formation depends strongly on the current prehistory of the cells being under voltage excitation including the number and duration of the voltage applications [26], the amount of the impurity charges [27], etc.

It appears that the flexoelectric domains are created after the transient space charge separation leading to the saturation of the two double electric layers being formed at the electrodes and to the formation of a bulk charge-depletion layer. In general, this is an unfavourable case for the isotropic instability creation, requiring first of all a high purity of the LC accompanied with a strong single or double injection from the electrodes [28], [29]. Furthermore, the threshold voltages indicating the onset of the isotropic instabilities being in the range of  $4\text{--}5\text{ V}$  [30–34] were much above those showing the surface-induced domain appearance in thick cells (Figure 3).

All these considerations clearly show the incapability of Felici's mechanism to explain the surface-induced domain formation.

#### b) Pikin's Mechanism

Some of the features characterizing Pikin's domains and the flexoelectric ones are very similar:

first, the long axis of each of these two types of domains follows always the initial orientation of the LC molecules and second, the realignment in the LC orientation includes not only  $\theta$ -polar deformations, typical for the classical Williams instability [35], but also azimuthal deformations.

During the experimental investigations, however, a number of differences were noted as well:

1. The increase in the initial tilt of the LC layer in accordance with our experimental results leads to a considerable decrease in the threshold voltage indicating the formation of the flexoelectric domains while Pikin's theory predicts a significant rise in the threshold voltage, necessary for the creation of azimuthal electrohydrodynamic domains.

2. The flexoelectric domains slightly depend on the influence of the dielectric torques while  $Z$ -variations in the  $\theta$ -deformations should lead to a considerable rise of the threshold voltage, necessary for the creation of Pikin's domains (the theory describing these domains has been developed under the essential suggestion for equality of the  $\theta$ -tilt over the entire LC layer [6], [7]).

3. The immobility of the flexoelectric domains around and slightly above the threshold voltage together with the flickering effect due to the strong thermal fluctuations of the molecules at the weak-anchoring wall clearly show their static character.

### 4. Flexoelectric Mechanism

The imperfections peculiar for the LC orientation of weakly-anchored MBBA layers [12] can be easily removed by the strong anchoring of the LC at one of the boundaries (Figure 1). In addition, the weak anchoring of the LC at the other boundary favours the easy and reproducible creation of various flexoelectric effects.

Note the crucial role of several parameters for the flexoelectric domains formation.

#### 1. Influence of the Dielectric Anisotropy

The various flexoelectric theories [1], [3], [6], [7] unambiguously point out a critical value of the dielectric anisotropy beyond which the domain structure cannot exist. However, this important condition has been obtained either without taking into account the real boundary conditions [1], [3] or

after an essential suggestion for strong anchoring of the LC layer [6], [7]. We shall see below that flexoelectric walls can be created even at higher values of the dielectric anisotropy.

The flexoelectric instability arising at a certain threshold voltage is characterized by a director deviation from the X axis in two planes: at an angle  $\varphi$  in the XOY plane (the plane of the electrodes) and at angle  $\theta$  in the XOZ plane (this plane is determined by the easy axis of the molecules OX and by the direction of the electric field applied across the LC layer).

For small deformations  $|\varphi| \ll 1$ ,  $|\theta| \ll 1$ , equal elastic coefficients  $K_{11} = K_{22} = K_{33} = K$  and the director components  $n_x \cong 1$ ,  $n_y \cong \varphi$ ,  $n_z \cong \theta$  one obtains two coupled differential equations:

$$\begin{aligned} \frac{\partial^2 \theta}{\partial y^2} + \frac{\partial^2 \theta}{\partial z^2} + E(e_{1z} - e_{3x})/K \frac{\partial \varphi}{\partial y} \\ - \frac{|\Delta \varepsilon|}{4\pi K} E^2 \theta = 0, \\ \frac{\partial^2 \varphi}{\partial y^2} + \frac{\partial^2 \varphi}{\partial z^2} - E(e_{1z} - e_{3x})/K \frac{\partial \theta}{\partial y} = 0, \quad (1) \end{aligned}$$

describing complex  $\theta$ ,  $\varphi$  flexoelectric reorientations of the nematic director [6], [7]. This system can be further transformed into the convenient equation

$$\begin{aligned} \frac{\partial^4 u}{\partial y^4} + 2 \frac{\partial^4 u}{\partial y^2 \partial z^2} + \frac{\partial^4 u}{\partial z^4} \\ + \left[ \frac{(e_{1z} - e_{3x})^2 E^2}{K^2} - \frac{|\Delta \varepsilon| E^2}{4\pi K} \right] \frac{\partial^2 u}{\partial y^2} \\ - \frac{|\Delta \varepsilon|}{4\pi K} E^2 \frac{\partial^2 u}{\partial z^2} = 0 \quad (2) \end{aligned}$$

describing either  $\varphi$ -azimuthal or  $\theta$ -polar flexoelectric deformations:  $u = \theta$  or  $\varphi$  (note the homogeneity of the electric field).

Hence in the case of a linear Z-solution (i.e. strong-weak anchoring) the flexoelectric deformations along Y can be either periodic:

$$\frac{(e_{1z} - e_{3x})^2}{K} > \frac{|\Delta \varepsilon|}{4\pi} \quad (3)$$

or should be with a wall structure [20]:

$$\frac{(e_{1z} - e_{3x})^2}{K} < \frac{|\Delta \varepsilon|}{4\pi}. \quad (4)$$

(The validity of the latter of these two inequalities was verified for the case of MBBA in two separate papers [36], [37]).

Flexoelectric domains can be created in nematics with positive dielectric anisotropy as well [12], [20]. It seems that static flexoelectric domains can exist in every nematic whatever the sign and the magnitude of the dielectric anisotropy.

## 2. Influence of the Homogeneity of the Electric Field

The space charges, as noted, are accumulated near to the electrodes forming in this way two double electric layers with typical thickness of several hundred angstroms [38], [40]. The formation of the space charge-depletion layer was facilitated by the asymmetry in the electrode treatment as well [39]. Moreover, part of the space charges are accumulated by the surface disclinations including nematic reorientations up to the strong-anchoring wall [41], [42] (Figure 7). The experimental results have clearly shown that the flexoelectric domains are formed after the saturation of the electric double layers and the establishment of a relatively homogeneous electric field (an ideal homogeneous electric field is not possible to achieve owing to the anisotropy in the conducting and dielectric tensor elements [43], [44]).

## 3. Influence of the $\varphi$ -azimuthal Anchoring of the LC Layer

Asymmetrically strong-weak  $\theta$ -anchored MBBA layers were first prepared after a monomolecular aliphatic monoamine  $C_8H_{17}NH_2$  deposition [17] on one of the two conductive glass plates previously

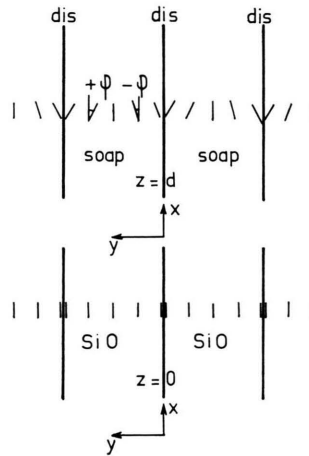


Fig. 7. Schematic representation of the  $\varphi$  deformations in the electrode planes.

rubbed or coated with thin SiO films to ensure the strong  $\varphi$ -azimuthal anchoring. The application of a d.c. voltage across such a cell led only to the formation of electrohydrodynamic domains already observed by Little et al. in hybrid-aligned nematic layers [45] (Figure 6). This result clearly demonstrates the crucial role of the weak azimuthal  $\varphi$ -anchoring of the LC at the soap-treated wall leading to the flexoelectric domain formation. It is reasonable to accept that the LC molecules can easily rotate at the weak-anchoring wall when the soap completely covers the surface grooves being made in different ways [46]–[52] on the confining glass plates.

Similarly, surface-induced flexoelectric domains cannot be created only by asymmetry in the  $\varphi$ -anchoring. For the present, however, this is not possible to be experimentally verified, since the weak  $\varphi$ -azimuthal anchoring when achieved by thick soap deposition is always accompanied by weak  $\theta$ -polar anchoring of the LC layer.

#### 4. Role of the Flexoelectric Deformations for the Creation of Novel Types of Electrohydrodynamic Instabilities

The insignificance of the flexoelectric phenomena in the conventional hydrodynamics has been correctly discussed by Fan [3]. The decisive role of the initial static flexoelectric deformations for the creation of electrohydrodynamic movement of the LC, however, has been touched on already in 1973 by Helfrich [53].

Indeed, the initial flexoelectric deformations just below the electrohydrodynamic threshold can determine various types of electrohydrodynamic movement which strongly depends on the strength of the surface coupling and the amount of the initial tilt closely related to the thickness of the LC cells being under study. For instance, at thin soap deposition the static  $\theta$ ,  $\varphi$  flexoelectric deformations of strong-weak anchored LC layers with a thickness below 100  $\mu\text{m}$  led to a complex three-dimensional flow of the fluid (Fig. 4b) already observed in weakly-anchored MBBA layers [12]. The increase of the soap thickness up to several microns led to a new electrohydrodynamics illustrated in Figure 8. It is evident that the flexoelectric two-dimensional deformations can create various electrohydrodynamic modes which await their further explanation.

#### Concluding Remarks

The longitudinal surface-induced flexoelectric domains arising in weakly-anchored MBBA layers [12] have been further investigated in asymmetrically strong-weak anchored MBBA layers. In summary, pronounced minima in the curves  $p$  vs.  $d$  and  $U_{th}$  vs.  $d$ , where  $p$  is the domain period,  $d$  is the cell thickness and  $U_{th}$  is the threshold voltage, are obtained. The threshold voltage indicating the onset of the flexoelectric instability is high for thin cells ( $d = 9 \mu\text{m}$ ,  $U_{th} = 10 \text{ V}$ ) and low for thick cells ( $d = 150 \mu\text{m}$ ,  $U_{th} = 1.5 \text{ V}$ ). Disappearance of these domains was not observed up to 210  $\mu\text{m}$  thickness of the layers being under study.

It appears that these domains are a two-dimensional analogue of the well-known flexoelectric bending of a homeotropic MBBA layer caused by a transversal and constant electric field, investigated by Helfrich et al. [54], [55] (strictly speaking the real two-dimensional analogue are the cross-like domains experimentally investigated already by us [12]).

The low threshold voltage, especially for thick cells, being in the range of 1.5–3 V permitted the clear separation of the flexoelectric phenomena from the possible electrohydrodynamic effects capable to disturb the flexoelectric deformations. The surface polarization, on the other hand, [56] in accordance with the recent results obtained by Petrov and Derzhanski [57] is negligible for the case of small tilting of the LC layers, i.e. the surface-induced domains observed in our experiment are purely flexoelectric.

The preparation of strong-weak anchored nematic layers facilitates very much the creation and the investigation of the flexoelectric domains. The new experimental results support the flexoelectric character of these domains and further reveal the insignificance of the mechanisms proposed by Felici [13] and Pikin [14], which are not capable to explain the formation of these domains.

The flexoelectric domains were only replaced by electrohydrodynamic instabilities in the case of strong  $\varphi$ -azimuthal anchoring of the LC molecules.

Our experimental results clearly demonstrate the significant role of the asymmetry in the surface anchoring of the nematic layers leading to an easy and reproducible creation of static surface-induced flexoelectric domains.

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