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Penetration Depth of Surface Forces Into Nematic Layers†

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The minima in the curves $U_{th}(d)$ and $p/d(d)/U_{th}$ is the threshold voltage, p is the period of the domains, d is the thickness of the cells of the flexoelectric domains in asymmetric anchored MBBA layers permit the determination of the penetration depth of the surface forces generated from the glass plates treated with SiO under vacuum evaporation. The value obtained, about 150 μm is in agreement with the results of other authors. The attenuation of these domains near the spacers on the other hand allows the determination of the penetration depth of the surface forces generated by the utilized polymer spacers which is about 350 μm. This simple method can be used for all nematics with positive or negative dielectric anisotropy displaying flexoelectric domains in asymmetric anchored layers.

The one-dimensional solutions of elastic problems for weakly anchored nematic layers demonstrate the importance of the value of the coherence length $b = K/W_s$ (W_s is a stiffness constant describing the boundary coupling strength which tries to keep the molecular orientation at the boundaries, K is the mean elastic coefficient) introduced by de Gennes.[1] It is seen that when W_s tends to infinity b becomes zero i.e. the penetration depth of the surface forces also tends to infinity. In effect the thermal fluctuation disturb the homogeneity of the molecular alignment. This fact was considered for the first time by de Gennes.[1] He obtained theoretically two basic types of the thermal fluctuations: the uncoupled modes of bend-twist and bend-splay deformations.

It has been known for years that surface forces act only on thin (about several hundred microns) liquid crystal layers. In thick cells the liquid crystal orientation is not affected by the limiting boundaries and the liquid crystal

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behavior is dictated chiefly by the thermal fluctuations of the liquid crystal molecules. This fact was established for the first time experimentally by Tsvetkoff.[2] He pointed out that the penetration depth of surface forces is several hundred microns. Kuhn and Finlayson[3] calculated the average liquid crystal orientation when only surface effects were considered i.e. in the absence of magnetic, electric and flow fields, for a homeotropic orientation of the liquid crystal on all surfaces. Their theoretical results were in agreement with the very well known experimental data about the influence of the side walls on liquid crystal orientation in one thin (along z) and long (along x and y) liquid crystal layer (see Figure 1 in the Ref. 3).

There is, however, another very simple method[4] for the determination of the penetration depth of surface forces for the one-dimensional case based on the behavior of surface induced domains[5] particularly in asymmetrically anchored nematic layers with SiO under vacuum evaporation and soap rubing of the glass plates respectively.[6] The penetration depth of the surface forces in this case corresponds to the minima of the curves $U_{th}(d)$ and $p/d(d)$ where p is the period of the domains, d—the thickness of the liquid crystal cell and U_{th}—the threshold voltage.

Let us consider briefly the experimental results obtained and the flexoelectric domain theory which clearly indicate the thickness, above which the influence of the wall treated with SiO under vacuum evaporation (strong anchoring) weakens.

FIGURE 1 MBBA liquid crystal layer 30 μm thick with asymmetric anchoring: strong and weak realized with SiO under vacuum evaporation and soap rubbing respectively.

EXPERIMENTAL RESULTS

1.1 Preparation of the cells

The liquid crystal cells under consideration were made of two glass plates coated with transparent conductive SnO_2 layers. One of the glass plates was treated with SiO under vacuum evaporation ensuring strong anchoring of the liquid crystal molecules. The other glass plate was treated with a surfactant: Na salt of the fatty acids (common soap from several hundreds to thousands Å thick) leading to a φ degeneration of the liquid crystal orientation in the glass plane and to θ weak anchoring: $W_{s\theta}d \sim K$. The cell gap was determined by polymer spacers 10 to 200 μm thick. The prepared cells were placed between two metallic holders. The degree of compression between the glasses and the polymer spacers was regulated with four screws.

The nematic liquid crystal with a negative dielectric anisotropy MBBA under investigation was with a critical frequency of the space relaxation in the range of 165 Hz.

The liquid crystal orientation in cells with thicknesses between 10 μm and 150 μm was slightly inclined in the plane determined from the direction of the strong anchoring (along x) and the normal to the glass plates (along z). The liquid crystal layer obtained was very homogeneous (Figure 1) with a strong asymmetry in the anchoring at the two boundaries. The orientation of the liquid crystal molecules in cells with greater thicknesses (> 150 μm) was determined by the simultaneous action of the rubbing, soap deposition and the direction of the liquid crystal filling flow coincident with that of the rubbing.

The thickness of the cells was measured as a difference between the top and bottom, focusing on the polymer edges with empty cells and on the liquid crystal edge with filled liquid crystal cells. The measurements were performed with the aid of a Leitz microscope with an accuracy of ± 1 μm for thin cells and ± 2 μm for thick cells.

1.2 Remarks

1) The thickness deviation in the plane of the electrodes is of no great importance for the creation of the domains investigated. They exist also in wedge-shaped cells.

2) The soap was rubbed on with a cloth. This technique gave the possibility for a very homogeneous deposition. The thickness of the soap coating had to be larger than the amplitudes of the surface grooves created by the SiO vacuum evaporation, or by mechanical rubbing.

1.3 Experimental procedure

A d.c. voltage was applied to the liquid crystal cell thus prepared. In some cases a high frequency orienting electric field was applied as well. Both vol-

tages were separated by an RC circuit. The observations were performed with a Leitz polarization microscope in transmitted polarized white light. The nicols were normally crossed with an analyser oriented along the easy direction or rotated at an angle φ- when φ-deformations were investigated. When studying deformations along θ the easy axis of the molecules was placed at an angle 45° between the crossed polarizer and the analyser. The investigations were performed at room temperature.

When a d.c. voltage of the order of 1.5–5 V (depending very strongly on the cell thicknesses) was applied alternating dark and grey bands were formed, oriented in the easy direction of the molecules OX (Figure 2) when the layer was tilted (the analyser was partially rotated at an angle $+\varphi$; if the analyser was rotated at the angle $-\varphi$ the bands changed places). These bands are due to the alternating twist-splay-band deformation (or walls) of the director in the ZOY plane. The twist rotation of the molecules in the adjacent domains is in two opposite directions. The simultaneous action of the strong anchoring of the lower glass plate and the flexoelectric bend-splay deformations at the upper glass plate generate flexoelectric domains or walls. The finite liquid crystal deformations arising as a first order transition are very complex with simultaneous variation of the liquid crystal orientation along θ and φ (Figure 3). Having this in mind we want to show the complication of the flexoelectric deforma-

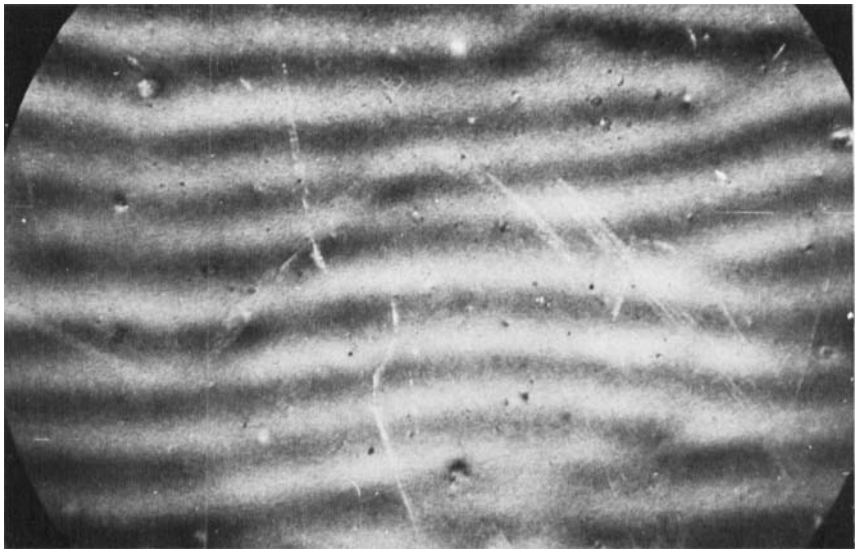

FIGURE 2 A threshold ($U_{th} = 2$ V) of parallel domains in a MBBA asymmetric layer 105 μm thick. Initially crossed nicols: $A \parallel \mathbf{n}_0 \perp P$; the analyser is rotated at an angle $\varphi \simeq 20°$; magnification ×60.

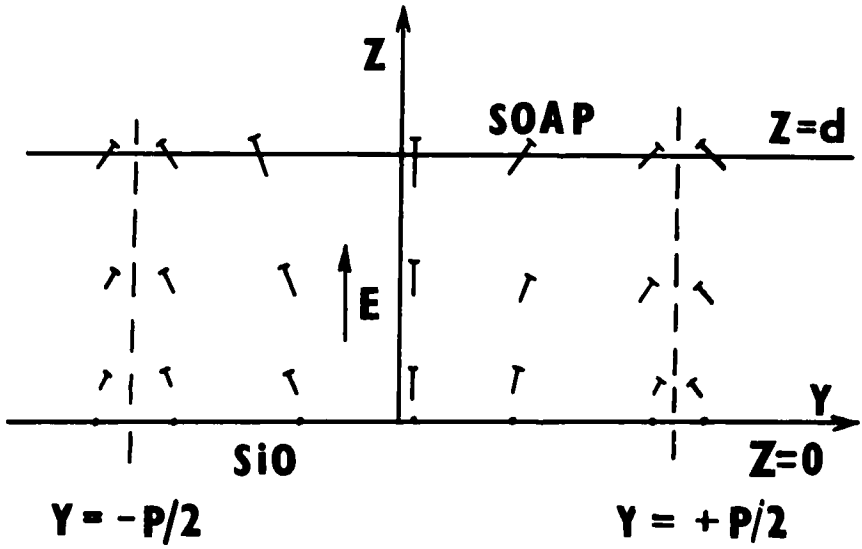

FIGURE 3 A director distribution in the flexoelectric domains.

tion in asymmetrical cells in comparison with the flexoelectric threshold behavior in symmetrical weak anchored MBBA films.[5]

The further increase of the voltage brings about the establishment of parallel domains of long term stability, oriented everywhere in the easy direction (Figures 4 and 5). When the thickness of the liquid crystal cells investigated was small (around 10μm) the domains formed were observed after irradiation with white polarizing light in the field of a microscope as alternating red and green bands almost equal in width. These observations as well as those of thicker cells demonstrated that these domains have a double period. The simultaneous applications of a deforming d.c. electric field and an orienting high a.c. electric field with a frequency above the space charge relaxation clearly confirmed the existence of coupled θ, φ deformations in plane YOZ (Figures 6 and 7).

An experimental measurement of the dependence of the period and the voltage threshold of these domains on the thickness of the liquid crystal cell investigated for the same boundary conditions (the same glasses) was carried out for MBBA liquid crystal. The results are given in Figures 8 and 9. A comparison is made with the experimental curves obtained for symmetrical MBBA layers with weak anchoring.

The curves $U_{th}(d)$ and $p/d(d)$ show minima around 150 μm. We anticipate that this thickness corresponds to the penetration depth of the surface forces generated from the SiO deposition under vacuum evaporation of the lower

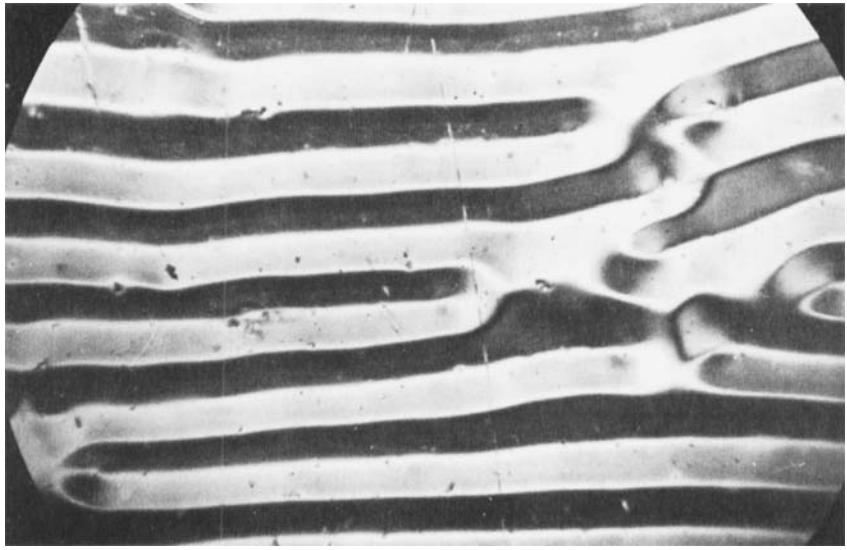

FIGURE 4 Parallel domains in a MBBA asymmetric layer 105 μm thick at applied d.c. voltage $U = 4$ V, $\mathbf{n}_0 \| A \perp P$; magnification ×60.

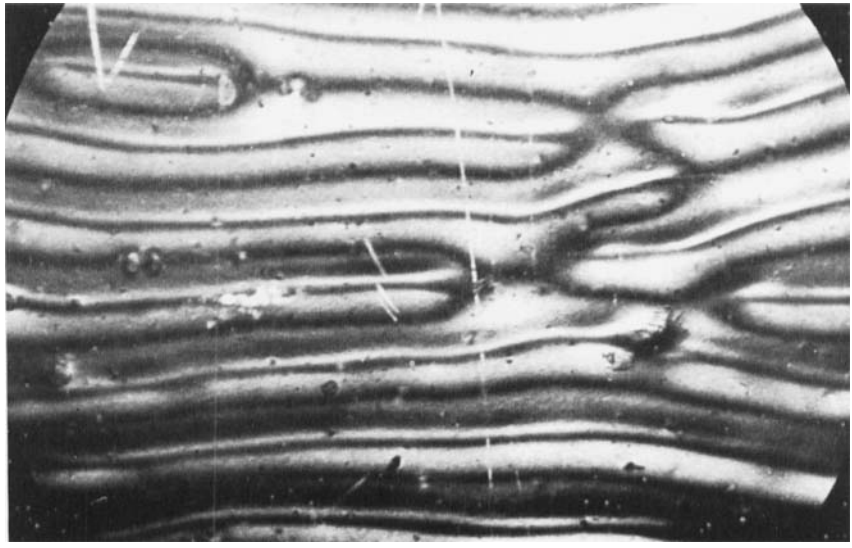

FIGURE 5 Parallel domains in a MBBA asymmetric layer 105 μm thick at applied d.c. voltage, $U = 7$ V, initially crossed nicols: $A \| \mathbf{n}_0 \perp P$; analyser rotation angle $\varphi \simeq 35°$; magnification ×60.

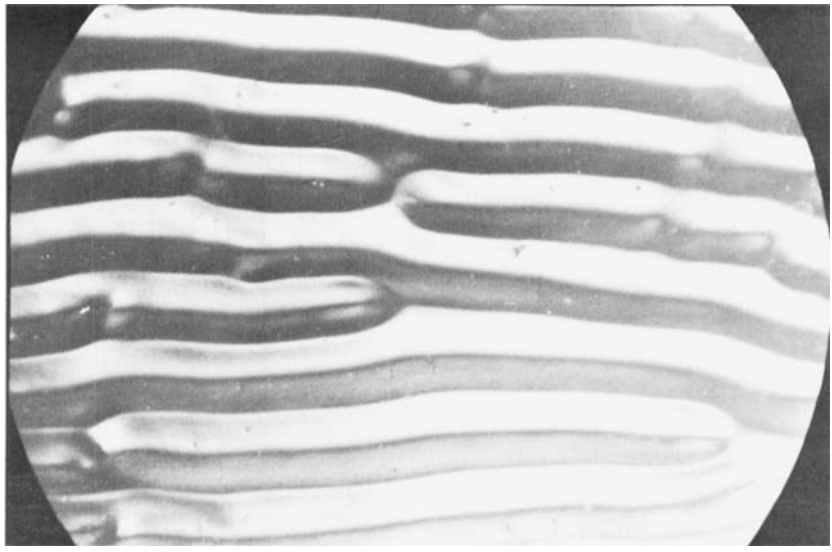

FIGURE 6 Parallel domains in a MBBA asymmetric layer 30 μm thick at an applied d.c. voltage 6 V, initially crossed nicols: $A \parallel \mathbf{n}_0 \perp P$; analyser rotation angle $\varphi \simeq 30°$; magnification ×60.

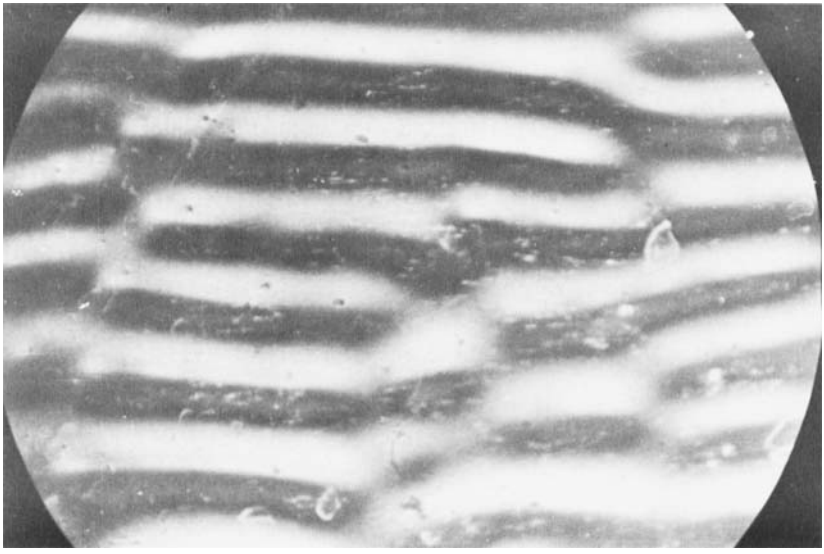

FIGURE 7 Parallel domains in a MBBA asymmetric layer 30 μm thick at simultaneously applied d.c. voltage $U = 6$ V and a.c. voltage $U = 20$ V 10 kHz, initially crossed nicols: $A \parallel \mathbf{n}_0 \perp P$, analyser rotation angle $\varphi \simeq 30°$; magnification ×60.

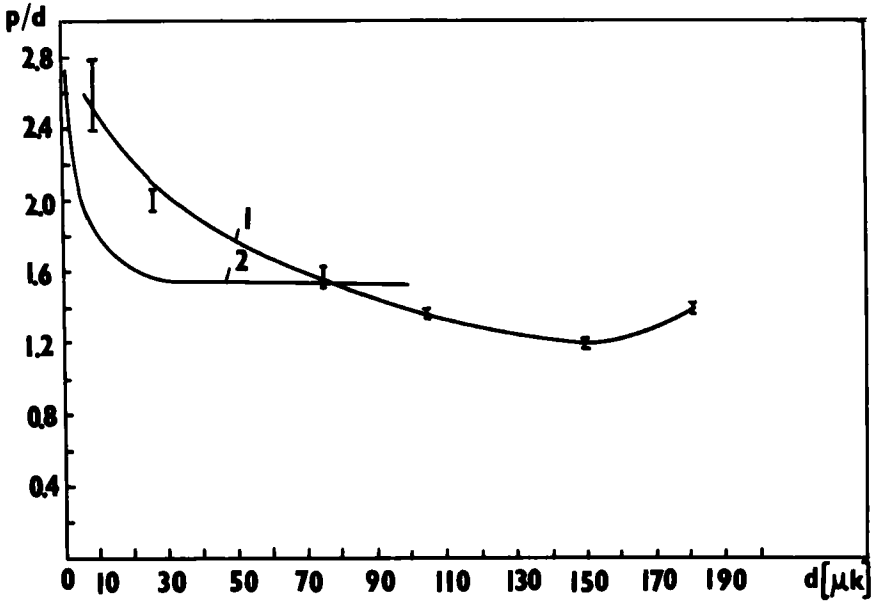

FIGURE 8 The ratio p/d (p is the domain period, d is the liquid crystal layer thickness) in asymmetric MBBA films (curve 1) and in symmetric MBBA films (curve 2) with weak anchoring as a function of the cell thickness under investigation. The minimum of the curve 1 is around 150 μm.

glass plate. Above this thickness the strength of the coupling of the liquid crystal layer with the lower glass plate displaying the strong anchoring decreases and the domains are formed in liquid crystal layers with weak anchoring.[5] Consequently this leads to a larger voltage threshold and to a higher ratio p/d.

In cells formed of two glass plates treated with SiO under vacuum evaporation the penetration depth of the surface forces would be approximately 300–400 μm which is a fairly good agreement with the experimental results of Tsvetkoff[2] obtained by other more complex techniques. We want to show also by theoretical calculations the validity of our assumption on the penetration depth of the surface forces.

THEORY

The flexoelectric instability arising at a certain threshold voltage is characterized by small director deviations from X axis in two planes: at angle θ in XY plane and at angle φ in XY plane (Figure 3).

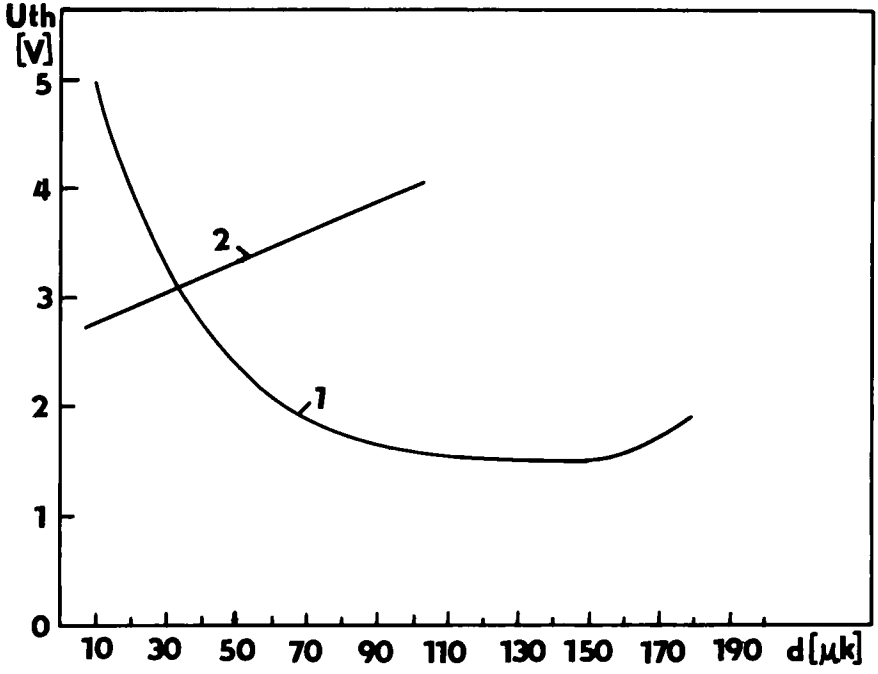

FIGURE 9 The threshold voltages in asymmetric MBBA films (curve 1) and in symmetric MBBA films (curve 2) with weak anchoring as a function of the cell thickness under investigation. The minimum of the curve 1 is around 150 μm.

The electric enthalpy for the nematic under consideration can be written in the following form:[7]

$$H_E = \left(\frac{1}{2}\right)\iiint\left\{K\left(\frac{\partial\varphi}{\partial y}+\frac{\partial\theta}{\partial z}\right)^2 + K\left(\frac{\partial\theta}{\partial y}-\frac{\partial\varphi}{\partial z}\right)^2 + \frac{|\epsilon_a|}{4\Pi}E^2\theta^2 - 2e_{1z}E\theta\frac{\partial\varphi}{\partial y} - 2e_{3x}E\varphi\frac{\partial\theta}{\partial y} - 2(e_{1z}+e_{3x})\theta\frac{\partial\theta}{\partial z}\right\}dx\,dy\,dz + \iint\left\{W_{s\theta}\theta^2 + W_{s\varphi}\varphi^2 + 2\bar{\omega}\theta\varphi\right\}dx\,dy \quad (1)$$

for small deformations $|\varphi|, |\theta| \ll 1$, equal elastic moduli $K_{11} = K_{22} = K_{33} = K$ and the following director components: $n_x = 1$, $n_y = \varphi$, $n_z = \theta$.

In (1) e_{1z} and e_{3x} are the splay and bend flexoelectric coefficients respectively, $W_{s\theta}$ is the θ stiffness surface constant, $W_{s\varphi}$ is the φ stiffness surface con-

stant. The constant introduced by Klèman and Pikin[8] shows the coupling between the θ and φ anchoring.

Minimizing the electric enthalpy H_E with respect to the deformation angles θ and φ, one can obtain the following Lagrange equations:

$$K\left(\frac{\partial^2\theta}{\partial y^2}+\frac{\partial^2\theta}{\partial z^2}\right)+(e_{1z}-e_{3x})E\frac{\partial\varphi}{\partial y}-\frac{|\epsilon_a|E^2}{4\Pi}\theta=0$$
$$K\left(\frac{\partial^2\varphi}{\partial y^2}+\frac{\partial^2\varphi}{\partial z^2}\right)-(e_{1z}-e_{3x})E\frac{\partial\theta}{\partial y}=0 \tag{2}$$

and the following boundary conditions at the soap treated wall:

$$K\left(\frac{\partial\varphi}{\partial y}+\frac{\partial\theta}{\partial z}\right)-E(e_{1z}+e_{3x})\theta+W_{s\theta}\theta+2\bar{\omega}\varphi=0$$
$$K\left(\frac{\partial\theta}{\partial y}-\frac{\partial\varphi}{\partial z}\right)-W_{s\varphi}\varphi-2\bar{\omega}\theta=0 \quad z=d \tag{3}$$

The liquid crystal orientation at the wall treated with SiO under vacuum evaporation will be assumed fixed: $\theta(0)=0, \varphi(0)=0$ by the strong anchoring.

The liquid crystal is much longer in the direction of y. Consequently the liquid crystal behavior will be determined by the two glass plates forming the cells.

The boundary conditions along y:

$$K\left(\frac{\partial\varphi}{\partial y}+\frac{\partial\theta}{\partial z}\right)\mp e_{1z}E\theta=0$$
$$K\left(\frac{\partial\theta}{\partial y}-\frac{\partial\varphi}{\partial z}\right)\mp e_{3x}E\varphi=0 \quad y=\pm p/2 \tag{4a}$$

are from the boundaries along Y would be important (e.g. for a determination of the voltage threshold or the period of the domains) for the behavior of a liquid crystal free in all directions.[5]

The boundary conditions near the spacers are much complex and include not only the flexoelectric energy [see the relations (4a)] but also the interaction elastic energy between the liquid crystal molecules and the spacer boundaries (Figure 10).

The coupling along φ, as was pointed out in the first part of the paper, becomes zero with thick soap deposition: $W_{s\varphi}\to 0$. The coupling along θ however is finite, but small.

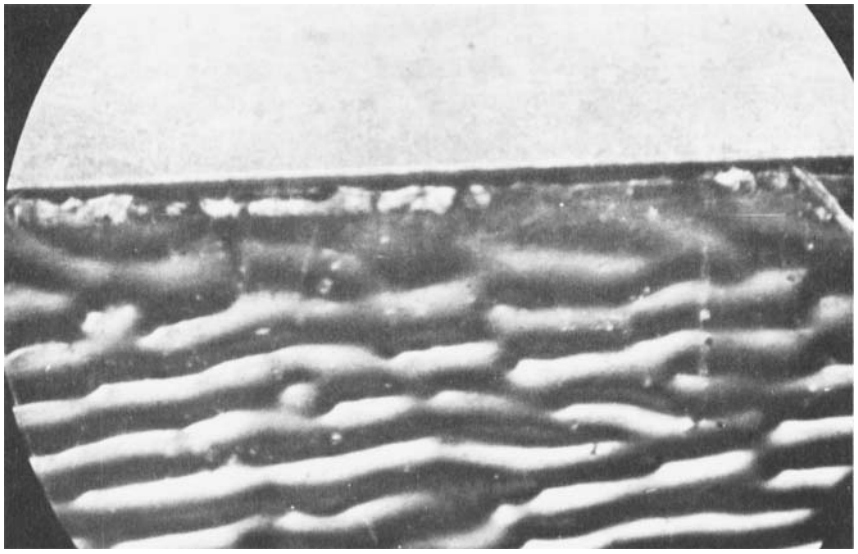

FIGURE 10 Attenuation of the parallel domains in a MBBA asymmetric layer 76 μm thick near the polymer spacer: the applied d.c. voltage is about 3,5 V, initially crossed nicols $A \parallel \mathbf{n}_0 \perp P$, the analyser rotation angle $\varphi \simeq 20°$; magnification ×60. The penetration depth of the surface forces at the spacers is about 350 μm.

With these simplifications the boundary conditions (3) can be written in the following form:

$$K\left(\frac{\partial \varphi}{\partial y} + \frac{\partial \theta}{\partial z}\right) - (E(e_{1z} + e_{3x}) - W_{s\theta})\theta = 0$$
$$K\left(\frac{\partial \theta}{\partial y} - \frac{\partial \varphi}{\partial z}\right) = 0 \quad z = d \tag{5}$$

Let us take the solution satisfying the boundary conditions (5) at the upper glass plate ($z = d$) and for vanishing values of the deformation angles θ and φ at the lower glass plate ($z = 0$) (Figure 3) such as:

$$\theta = \text{Azch}\,(qy), \quad \varphi = \text{Bzsh}\,(qy) \tag{6}$$

when the flexoelectric energy is greater than the surface coupling constant $W_{s\theta}$:

$$(e_{1z} + e_{3x})E > W_{s\theta} \tag{7}$$

Let us note that the dimension of the constants A and B in the solution (6) is cm^{-1}.

Inserting (6) into the differential Eq. (2) and the boundary conditions (5) and taking into account the condition of nontriviality of the solution: $\theta \neq 0$ $\varphi \neq 0$, e.g. $A \neq 0$ and $B \neq 0$, one can write the following two equations connecting the value of the electric field and the parameter q:

$$\frac{(e_{1z} - e_{3x})^2}{K^2} E^2 + q^2 = \frac{|\epsilon_a|}{4\Pi K} E^2 \tag{8}$$

$$Kq^2d^2 = E(e_{1z} + e_{3x})d - (K + W_{s\theta}d) \tag{9}$$

The elimination of the parameter q leads to the quadratic equation for the value of the electric field at the domain formation:

$$E^2\left(\frac{|\epsilon_a|}{4\Pi} - \frac{(e_{1z} - e_{3x})^2}{K}\right) - E\frac{(e_{1z} + e_{3x})}{d} + \frac{K + W_{s\theta}d}{d^2} = 0 \tag{10}$$

The solution of this equation gives the threshold voltage U_{th} as a function of all other parameters in the problem under consideration:

$$U_{th} = \frac{(e_{1z} + e_{3x}) + \left[(e_{1z} + e_{3x})^2 - 4(K + W_{s\theta}d)\left(\frac{|\epsilon_a|}{4\Pi} - \frac{(e_{1z} - e_{3x})^2}{K}\right)\right]^{1/2}}{2\left(\frac{|\epsilon_a|}{4\Pi} - \frac{(e_{1z} - e_{3x})^2}{K}\right)} \tag{11a}$$

The corresponding solution for the dimensionless parameter dq (d is the cell thickness) has the form:

$$dq = \frac{(e_{1z} + e_{3x}) + \left[(e_{1z} + e_{3x})^2 - 4(K + W_{s\theta}d)\left(\frac{|\epsilon_a|}{4\Pi} - \frac{(e_{1z} - e_{3x})^2}{K}\right)\right]^{1/2}}{2K^{1/2}\left(\frac{|\epsilon_a|}{4\Pi} - \frac{(e_{1z} - e_{3x})^2}{K}\right)^{1/2}} \tag{11b}$$

The threshold voltage U_{th} in accordance with (11a) and with part of the experimental results (Figure 9) decreases with the thickness d until the following relation:

$$(e_{1z} + e_{3x})^2 - 4(K + W_{s\theta}d)\left(\frac{|\epsilon_a|}{4\Pi} - \frac{(e_{1z} - e_{3x})^2}{K}\right) \tag{12}$$

which determines the minimal threshold voltage:

$$U_{th\,min} = \frac{(e_{1z} + e_{3x})}{2\left(\frac{|\epsilon_a|}{4\Pi} - \frac{(e_{1z} - e_{3x})^2}{K}\right)} \tag{13}$$

and the minimal value of the parameter *dq:*

$$dq_{\min} = \frac{(e_{1z} + e_{3x})}{2\left(\frac{|\epsilon_a|}{4\Pi} - \frac{(e_{1z} - e_{3x})^2}{K}\right)} \tag{14}$$

The real period p of the domains can be obtained only after minimization of the electric enthalpy including the solutions of the problem and the elastic energy of the walls and the surface disclinations adjusted the flexoelectric deformations in the adjacent domains. This is not possible at the moment. It is interesting to note however, that the difference between the value of the dimensionless parameter dq calculated from the relations (13) and (14) and the value of p/d experimentally obtained (Figure 8) is not very high and is of the order of a unit.

After the critical thickness of the cells under investigation determined from the relation (12) in the liquid crystal is impossible the formation of the flexoelectric domains. Here the theoretical results are in contradiction with the experimental data about the formation of these domains. The penetration of the surface forces with the other thicknesses became weak and the object of theoretical description should be another liquid crystal layer with different boundary conditions.

Let us stress the following important features of the problem under consideration:

(a) the flexoelectric domains appear in asymmetrical MBBA films in the form of walls when the well known flexoelectric domains of Bobylev *et al.*[9,10] are impossible:

$$\frac{|\epsilon_a|}{4\Pi} > \frac{(e_{1z} - e_{3x})^2}{K} \tag{15}$$

Their existence is due only to the weak anchoring at the soap treated wall.

(b) the lowest value of the threshold voltage gives an important relation between the sum of the flexoelectric coefficients: $(e_{1z} + e_{3x})$, their difference: $(e_{1z} - e_{3x})$ and the values of the dielectric anisotropy and the mean elastic coefficient, respectively: (13) Our experimental results (Figure 9) show the range of the total flexoelectric coefficient $(e_{1z} + e_{3x}) \sim 4.10^{-4}$ c.g.s. for $U_{th} = 1{,}5$ V and $|\epsilon_a|/4\Pi > (e_{1z} - e_{3x})^2/K \sim 4.10^{-2}$ which is in excellent agreement with the value obtained by Prost and Pershan[11] and by Derzhanski and Mitov.[12]

(c) the surface polarization[11] in accordance with the results of Petrov and Derzhanski[13] is small for slightly tilted layers. This shows that the observed domains are purely flexoelectric.

(d) the minima in the curves $U_{th}(d)$ and $p/d(d)$ clearly indicate the penetration depth of the surface forces. This was also confirmed by the liquid crystal behavior without electric field. Above this thickness the liquid crystal orientation at the upper soap treated plate was very unstable and was strongly dependent on flow and thermal fluctuations etc. On the other hand the surface forces due to the glass plate treated with SiO under vacuum evaporation are strong and impose the orientation in the whole liquid crystal layer especially in thin cells. This influence is very big in cells with thicknesses below 10 μm preventing the formation of the flexoelectric domains which exist only in tilted layers.

(e) the attenuation of the parallel domains in MBBA asymmetric anchored layers near the spacers permits the simple determination of the penetration depth of the surface forces from the boundaries along Y. The value is estimated about 350 μm (Figure 10).

(f) the anisotropic case for the values of the elastic coefficients $K_{11}\, K_{22}\, K_{33}$ changing the type of the possible solution (6) as well as the inclusion of finite $W_{s\varphi}$ energy has been considered in a forthcoming paper.[6]

CONCLUSIONS

The minima in the curves $U_{th}(d)$ and $p/d(d)$, where U_{th} is the threshold voltage for the formation of flexoelectric domains (or flexoelectric walls) in asymmetric anchored MBBA layers, p is the domain period and d is the thicknesses of the investigated liquid crystal layers, allow the determination of the penetration depth of the surface forces due to SiO treatment under vacuum evaporation. The results obtained—300–400 μm are in good agreement with other results. This method is very simple and can be applied for other treatments of the glass plates giving a strong anchoring and for other liquid crystals with positive or negative dielectric anisotropy. For liquid crystals which do not interact with the soap deposition one can use very well polished glasses giving a degeneration of the liquid crystal orientation along φ. In this way Rivière[14] obtained flexoelectric domains in 6CB possessing high positive dielectric anisotropy.

The attenuation of the flexoelectric domains near the spacers permits the determination for the first time of the penetration depth of the surface forces generated. The result obtained: around 350 μm in our opinion is not surprising. This method can be used also at the free boundaries or around holes, etc.

The estimation of the penetration depth of the surface forces is of great importance for all displays requiring strong anchoring.

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