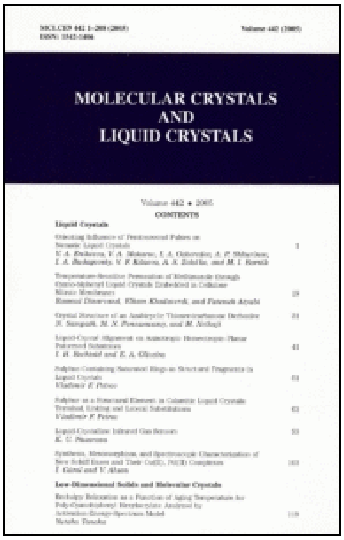
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On the Coexistence of the Flexo-Dielectric Walls-Flexoelectric Domains for the Nematic MBBA—A New Estimation of the Modulus of the Difference between the Flexoelectric Coefficients of Splay and Bend $|e_{1z} - e_{3x}|$

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On the Coexistence of the Flexo-Dielectric Walls–Flexoelectric Domains for the Nematic MBBA—A New Estimation of the Modulus of the Difference between the Flexoelectric Coefficients of Splay and Bend $|e_{1z} - e_{3x}|$

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The coexistence of the flexo-dielectric walls and the flexoelectric domains of Vistin'-Pikin-Bobylev has been again observed in the nematic methoxybenzylidene-parabutylaniline (MBBA). On the basis of the first observation of such coexistence by Vistin' et al. *and the new theory developed by us, the modulus of the difference between the flexoelectric coefficients of splay e_{1z} and bend e_{3x}, $|e_{1z} - e_{3x}|$, has been estimated to be around 2×10^{-4} statC/cm. It was experimentally found that the increase of the conductivity σ of the nematic MBBA from 10^{-9} to 10^{-8} $(\Omega \cdot cm)^{-1}$ replaced the flexoelectric domains by electrohydrodynamic movement of the fluid. The details of this movement are illustrated for the first time.*

Keywords Coexistence; estimation of flexoelectric coefficient $|e_{1z} - e_{3x}|$; flexo-dielectric walls; flexoelectric domains; MBBA

Introduction

It is well known that a direct current (DC) electric field applied across a thin nematic layer confined between two glass plates, at least one treated with a surfactant layer (soap or lecithin), can induce flexo-dielectric walls oriented along the initial alignment of the nematic [1–11]. On the other hand, Vistin' [12] and Greubel and Wolff [13] have discovered new DC voltage-induced static domains with an electrically controlled period oriented along the initial alignment of the nematic director **n**. These authors have also observed that the period of the domains decreases with the increase of the voltage. Bobylev and Pikin [14] were the first who proved the flexoelectric nature of these domains by developing a theory for the case of isotropic elasticity. They have obtained simple formulae for the threshold voltage U_c and the wave number q_c connecting the value of the electric field E with some of the material

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parameters of the nematic, such as the dielectric anisotropy $\Delta\varepsilon$, the mean elastic constant *K*, and the difference between the flexoelectric coefficients of splay e_{1z} and bend e_{3x}, $(e_{1z} - e_{3x})$, originally introduced by Meyer [15]. These important theoretical results stimulated further experimental study of such flexoelectric domains. Barnik *et al.* [16,17] have performed a very precise experiment, studying the flexoelectric domains very carefully. The Bobylev-Pikin theory was elaborated for the anisotropic elasticity by Bobylev *et al.* [18] and Schiller *et al.* [19]. Marinov and Hinov considered the behavior of the flexoelectric domains under the simultaneous action of a flexoelectrically deforming DC voltage and a high-frequency dielectrically orienting nematic voltage and found a solution based on matrix calculations [20]. The appearance of the flexoelectric domains under the influence of only a DC voltage was also studied [21]. We would like to stress here that the observation of the flexoelectric domains of Vistin'-Pikin-Bobylev in real MBBA nematic cells is very difficult due to the presence of many ions in this widely studied nematic [22,23]. Furthermore, according to our knowledge, there are only two measurements of the modulus of the difference between the flexoelectric coefficiens of splay e_{1z} and bend e_{3x}, $|e_{1z} - e_{3x}|$, for the case of the nematic MBBA [24,25] at room temperature as follows: Dozov *et al.* [24] have obtained the following value of $|e_{1z} - e_{3x}|$ for the case of MBBA:

$$|e_{1z} - e_{3x}| \cong (1.0 \pm 0.2) \times 10^{-4}\,\text{statC/cm} \qquad (1)$$

Takahashi *et al.* [25] have obtained a bigger value of this difference:

$$|e_{1z} - e_{3x}| \cong (4.2 \pm 0.3) \times 10^{-4}\,\text{statC/cm} \qquad (2)$$

Further, the flexoelectric domains of Vistin'-Pikin-Bobylev can appear in coexistence with other instabilities [26,27] (see Fig. 1).

First, we show our observation of the coexistence of flexo-dielectric walls and of Visin'-Pikin-Bobylev flexoelectric domains under various circumstances: the decrease of their period under the application of a higher DC voltage and their disappearance under the simultaneous application of DC voltage, forming the flexoelectric domains, and a high-frequency alternating current (AC) dielectrically orienting the nematic voltage, removing them. Second, we have calculated the modulus of $|e_{1z} - e_{3x}|$ for the case of nematic MBBA, based on the flexoelectric domains that are seen on the lower part of Fig. 1. As far as we know, this is a third assessment of the difference between the two flexoelectric coefficients of splay and bend for the case of MBBA. Third, we show that the purity of the nematic MBBA is of crucial significance for the existence of the flexoelectric domains and demonstrate that for the case of more conductive MBBA, a very complex electrohydrodynamic movement of the fluid replaced the flexoelectric domains. Such an electrohydrodynamic picture in the initial phase of development has been observed by us [1,2] and by Delev *et al.* [28].

Observation of Coexistence of the Flexo-Dielectric Walls and of the Flexoelectric Domains of Vistin'-Pikin-Bobylev

We have again observed the coexistence of the flexo-dielectric walls and the flexoelectric domains of Vistin'-Pikin-Bobylev in the nematic MBBA. Let us stress that

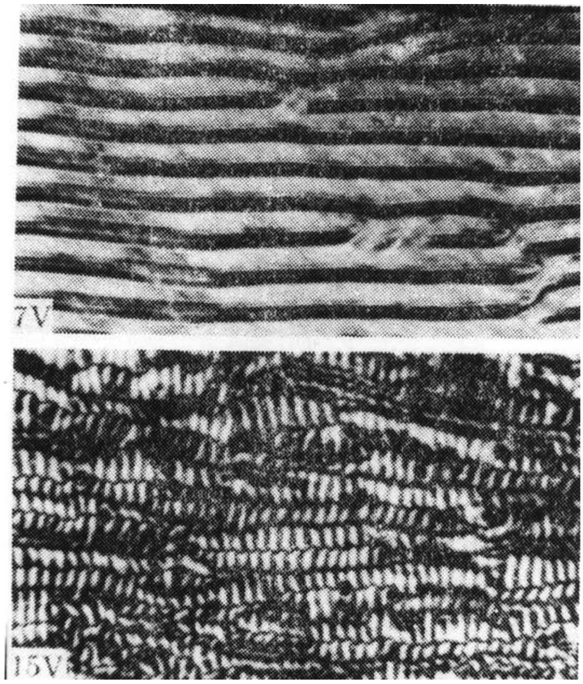

Figure 1. A microphotograph from Vistin' *et al.* [26] showing the simultaneous existence of longitudinal domains aligned along the long side of the picture and the flexoelectric domains of Vistin' *et al.* [26], aligned in the perpendicular direction. The liquid crystal studied was MBBA with a thickness of 20 μm and a conductivity $\sigma \sim 10^{-10}\ (\Omega \cdot \mathrm{cm})^{-1}$. The applied voltage is shown in the image. The sizes of the two pictures are 650 μm × 380 μm, respectively.

it is not easy to observe such a coexistence and to our knowledge there are only two such observations. Our observation is shown in Fig. 2 for a nematic layer with a thickness of 100 μm under a DC voltage excitation. The voltage threshold for the appearance of the flexo-dielectric walls was about 2 V and the voltage threshold for the appearance of the flexoelectric domains was about 6 V. The period of the flexo-dielectric walls is about 200 μm (magnification ×60) and the period of the flexoelectric domains is about 33 μm. Further, the flexoelectric domains were formed in the regions of the cells where the free ions are depleted. Inversely, in the regions with ions, the flexoelectric domains could not be formed. The application of additional, dielectrically orienting the nematic AC voltage with an effective value of 14 V_{rms}, 20 kHz, removed a great part of the flexoelectric deformations. This is shown in Fig. 3. The stronger flexoelectric deformations, however, cannot be removed with such a value of the AC voltage. From theory and experiments it is well known that the period of the flexoelectric domains decreases with the increase of magnitude of the DC voltage. This was confirmed by our observations. The increase of the DC voltage from 6 to 8 V led to the apparent decrease of the period of the flexoelectric domains. This process is illustrated in Fig. 4. The mechanism of decreasing the period of the flexoelectric domains in the conventional case is well known. It consists of a movement of charged dislocations along the domains [16,17]. However, the application of simultaneous DC and AC voltages can continuously decrease the

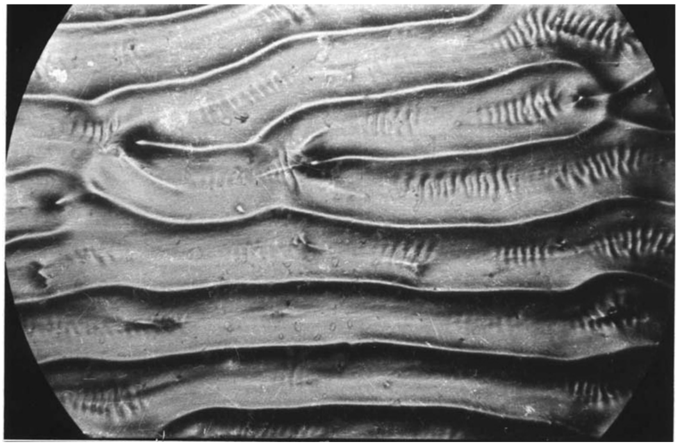

Figure 2. Flexo-dielectric walls and the flexoelectric domains of Vistin'-Pikin-Bobylev in a nematic MBBA layer with a thickness of 100 μm and a conductivity $\sigma \sim 10^{-9}$ $(\Omega \cdot \mathrm{cm})^{-1}$. The liquid crystal is excited by a DC voltage of 6 V; only analyzer is used; room temperature; magnification ×60. The size of the picture is 1350 μm × 879 μm.

period of the flexoelectric domains, as was shown by us [20]. In our case, however, the decrease of the period of the flexoelectric domains is more complex. On the other hand, the image of the domains of Fig. 4 shows undoubtedly that the flexoelectric domains cross the flexoelectric walls, thus generating point singularities that are visible as a series of bright spots. The second important note concerns the purity of the nematic phase of MBBA. The cutoff frequency showing the appearance of the chevrons has been measured to be around 165 Hz, which corresponds to conductivity σ in

Figure 3. Flexo-dielectric walls and the flexoelectric domains of Vistin'-Pikin-Bobylev in the same nematic MBBA layer with a thickness of 100 μm and a conductivity $\sigma \sim 10^{-9}$ $(\Omega \cdot \mathrm{cm})^{-1}$. The liquid crystal is excited by a DC voltage of 6 V and a high-frequency orienting voltage of 14 V_{rms} and a frequency $f = 20$ kHz; only analyzer is used; room temperature; magnification ×60. The size of the picture is 1350 μm × 879 μm.

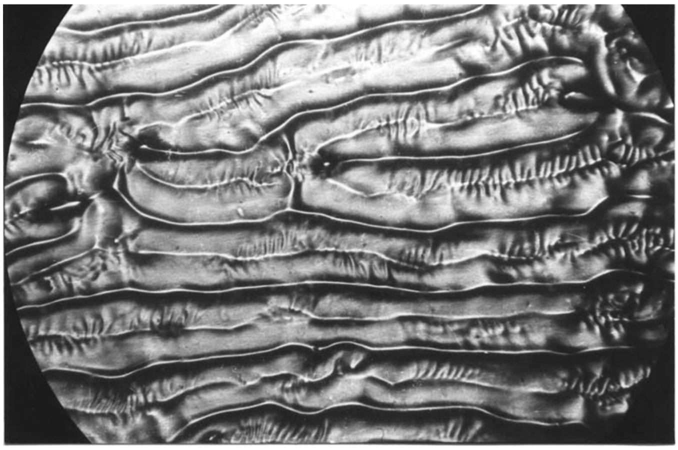

Figure 4. Flexo-dielectric walls and the flexoelectric domains of Vistin'-Pikin-Bobylev in the same nematic MBBA layer with a thickness of 100 μm and a conductivity $\sigma \sim 10^{-9}$ $(\Omega \cdot \mathrm{cm})^{-1}$. The liquid crystal is excited by a DC voltage of 8 V; only analyzer is used; room temperature; magnification ×60. The size of the picture is 1350 μm × 879 μm.

the range of 10^{-9} $(\Omega \cdot \mathrm{cm})^{-1}$. Let us note that the picture shown in Fig. 1 has been obtained in pure nematic MBBA with $\sigma \sim 10^{-10}$ $(\Omega \cdot \mathrm{cm})^{-1}$ [26]. The decrease of the conductivity σ from 10^{-9} to 10^{-8} $(\Omega \cdot \mathrm{cm})^{-1}$ removes the flexoelectric domains of Vistin'-Pikin-Bobylev by complex electrohydrodynamic movement of the fluid shown in Figs. 5 and 6.

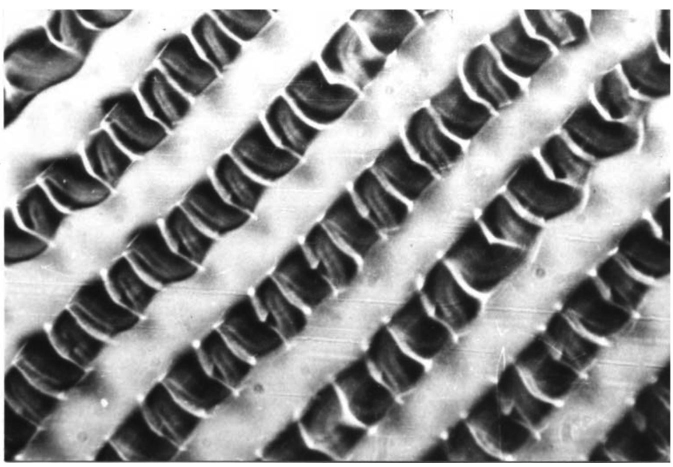

Figure 5. Flexo-dielectric walls and electrohydrodynamic domains in MBBA nematic layer with a thickness of 90 μm and a lower conductivity $\sigma \sim 10^{-8}$ $(\Omega \cdot \mathrm{cm})^{-1}$. $P \parallel n_0 \perp A$. The nematic layer is excited by a DC voltage of 10 V. The minus (–) is applied to the liquid-crystal cell from the side of the observer. The image shows the beginning of the electrohydrodynamics; room temperature; magnification 12.5 × 12.5. The size of the picture is 846 μm × 575 μm.

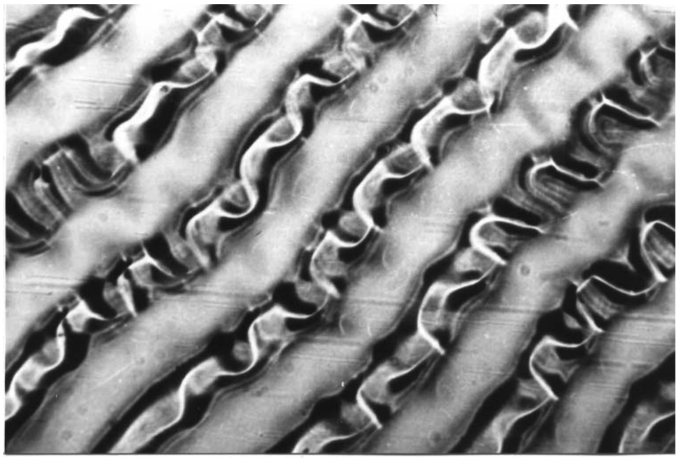

Figure 6. The same MBBA layer shown in Fig. 5 after 10 s.

Estimation of $|e_{1z} - e_{3x}|$ for the Case of MBBA

As mentioned earlier, as far as we know, there are only two measurements of the modulus of the difference between the flexoelectric coefficients of splay e_{1z} and bend e_{3x}, $|e_{1z} - e_{3x}|$, for the case of the nematic MBBA [24,25]. On the other hand, it is possible to estimate this modulus from the pictures shown in Figs. 1 to 4 by using the complete theory describing the appearance of the flexoelectric domains of Vistin'-Pikin-Bobylev, which was developed by us [20,21]. It is important to mention here that for the case of MBBA the flexo-dielectric walls and, consequently, the flexoelectric domains of Vistin'-Pikin-Bobylev appear near the cathode for the case of thicker cells with a thickness greater than 20 μm. In these cases we do not know the thickness of penetration of the flexoelectric domains. Consequently, we decided to use the picture of the flexoelectric domains shown in the lower part of Fig. 1, from which we can determine with higher precision the value of $|e_{1z} - e_{3x}|$ for the case of MBBA. The other physical parameters we must know are the value of the dielectric anisotropy $\Delta\varepsilon$, the value of the twist elastic constant K_{22}, the value of the ratio between the elastic constants of splay and twist, K_{11}/K_{22}; and, finally, the period of the flexoelectric domains λ_c ($\lambda_c = 2\pi/q_c$, where q_c is the wave number) [29–31]. With a high precision we take the following values at room temperature:

$$K_{22} = 4.0 \times 10^{-7}\,\text{dyne}, \quad K_{11}/K_{22} = 1.6\ [32\text{–}43], \quad \Delta\varepsilon = 0.7\ [44] \tag{3}$$

and $\lambda_c = 15\,\mu\text{m}$ [27] (Fig. 11) (the thickness of the liquid crystal cell has been 20 μm) [27].

The expressions for U_c and q_c have the following forms [20]:

$$U_c^2 = \frac{\pi^2 K_{11} K_{22}}{(e_{1z} - e_{3x})^2} \frac{\left[1 + b_1\left(\frac{\mu}{1-b_1\mu} + \frac{1}{1-b_1\mu} P_{SQR}\right)\right]\left[1 + a_1\left(\frac{\mu}{1-b_1\mu} + \frac{1}{1-b_1\mu} P_{SQR}\right)\right]}{P_{SQR}} \tag{4}$$

$$q_c^2 = \left(\frac{\pi}{d}\right)^2 \left(\frac{\mu}{1 - b_1\mu} + \frac{1}{1 - b_1\mu} P_{SQR}\right) \tag{5}$$

where

$$P_{SQR} = \sqrt{\frac{(1 - b_1\mu) + a_1\mu}{a_1 b_1}}, \quad a_1 = \frac{1}{2} + \frac{3}{4}\frac{K_{22}}{K_{11}} - \frac{1}{4}\frac{K_{11}}{K_{22}},$$
$$b_1 = \frac{1}{2} + \frac{3}{4}\frac{K_{11}}{K_{22}} - \frac{1}{4}\frac{K_{22}}{K_{11}}, \quad \mu = \frac{|\Delta\varepsilon| K_{22}}{4\pi(e_{1z} - e_{3x})^2} \tag{6}$$

The requirements:

$$a_1 > 0, b_1 > 0; (1 - b_1\mu) > 0 \tag{7}$$

lead to the following inequalities:

$$\frac{1}{3} < \frac{K_{22}}{K_{11}} < 3 \tag{8}$$

and to a restriction on the other parameter μ:

$$|\mu| < \left(\frac{1}{b_1}\right) \tag{9}$$

Based on the relations (4) and (5), we have calculated the threshold voltage U_c and the wavelength $\lambda_c = q_c/2\pi$ as a function of the difference between the two

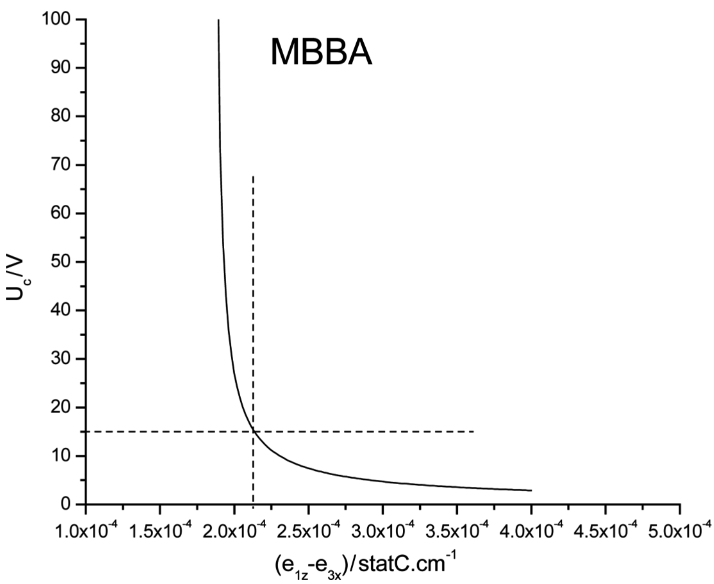

Figure 7. Determination of the value of $(e_{1z} - e_{3x})$ for the case of MBBA after comparison of the threshold voltage U_c of the flexoelectric domains (see Fig. 1) extracted from the experimental data obtained in Vistin' *et al.* [26] to the theoretical values represented by the solid curve.

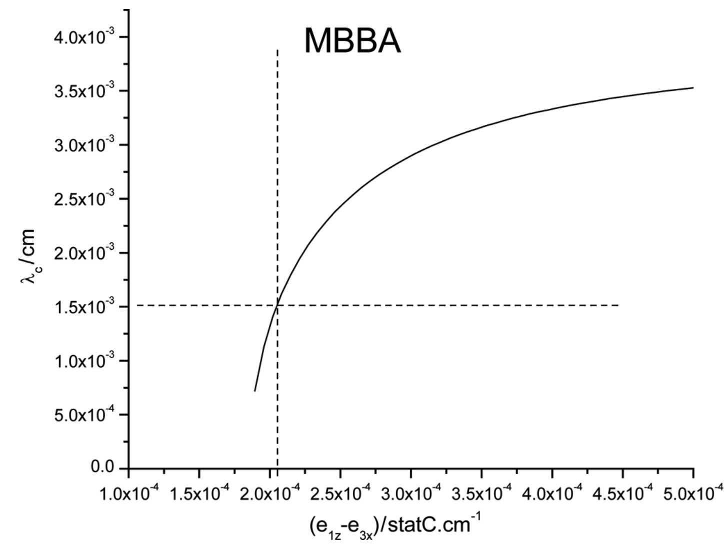

Figure 8. Determination of the value of ($e_{1z} - e_{3x}$) for the case of MBBA after comparison of λ_c of the flexoelectric domains for a thickness of the liquid crystal cell of 20 μm (see Fig. 1) extracted from the experimental data obtained in Vistin' *et al.* [26] with the theoretical threshold values represented by the solid curve.

flexoelectric coefficients ($e_{1z} - e_{3x}$) represented by the solid curves shown in Figs. 7 and 8. Finally we have obtained the following range of the modulus $|e_{1z} - e_{3x}|$:

$$2.06 \times 10^{-4}\,\text{statC/cm} \leq |e_{1z} - e_{3x}|_{\text{MBBA}} \leq 2.13 \times 10^{-4}\,\text{statC/cm} \tag{10}$$

for $K_{22} = 4.0 \times 10^{-7}$ dyne, $K_{11}/K_{22} = 1.6$, and $\Delta\varepsilon = 0.7$ and the experimental values of $U_c = 15$ V and $\lambda_c = 15$ μm obtained by Vainstein *et al.* [27].

Finally, we can estimate the penetration depth of the flexoelectric domains of Vistin'-Pikin-Bobylev for the domains observed by us. Because the period of the domains is smaller relative to the thickness of the cell for the case when the domains penetrate into the whole cell (see Fig. 1), in our case the penetration of the domains is about 40 μm. This value is less than the value of the cell thickness, which was 100 μm. Because the large flexo-dielectric walls are created in the cathode region, the flexoelectric domains of Vistin'-Pikin-Bobylev appeared in the same region as well.

Conclusion

In summary, we have observed the coexistence of flexo-dielectric walls and the flexoelectric domains of Vistin'-Pikin-Bobylev for the nematic MBBA with a conductivity $\sigma \sim 10^{-9}\ (\Omega \cdot \text{cm})^{-1}$ and details of the coexistence of the flexo-dielectric walls and the developed complex electrohydrodynamic at lower conductivity: $\sigma \sim 10^{-8}\ (\Omega \cdot \text{cm})^{-1}$. Such a coexistence can be rarely observed in nematics. Further, we have calculated the range of the values of the difference between the flexoelectric coefficients of splay e_{1z} and bend e_{3x}, $|e_{1z} - e_{3x}|$, for the case of MBBA on the basis of previous results of other authors concerning the coexistence of the flexo-dielectric walls and the flexoelectric domains of Vistin'-Pikin-Bobylev. Thus, we have obtained the value of $|e_{1z} - e_{3x}|$ for the case of MBBA. The depth of penetration of the flexoelectric domains of Vistin'-Pikin-Bobylev for our case (see Fig. 2) has been estimated as well.

Acknowledgments

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