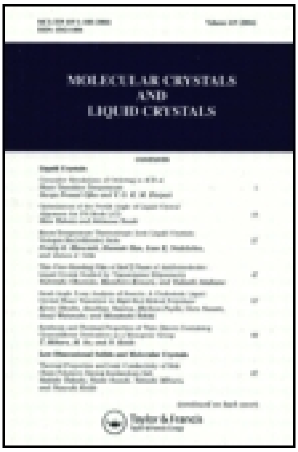

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Numerical Analysis of Deformations Induced by DC Electric Field in Homeotropic Nematic Layers with Giant Flexoelectricity

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Deformations of nematic layers induced by electric field were studied numerically. The role of very strong flexoelectric properties was considered. The threshold voltages for deformations were calculated for nematics with very low, moderate and high ion contents and for both signs of dielectric anisotropy. The director distributions were also determined. When the sum of flexoelectric coefficients reached large values, the threshold voltage decreased strongly and was independent on the sign of the dielectric anisotropy. The strong flexoelectric properties caused that even for extremely pure materials the threshold was lower than the theoretical value calculated for perfectly insulating material.

Keywords Director field deformations; flexoelectric properties; giant flexoelectricity

1. Introduction

Deformations of nematic liquid crystal layers induced by external electric field are due to torques acting on director which may have dielectric and flexoelectric nature. When the electric field is applied perpendicular to the layer plane, the deformations arise above some threshold voltage. The threshold value depends on dielectric anisotropy $\Delta\varepsilon$, sum of the flexoelectric coefficients $e_{11} + e_{33}$, anchoring energy W, ion mobilities $\mu^{\pm}$ and ion concentrations $N^{\pm}$. In previous papers, some of these relationships were studied numerically [1–5]. Some time ago, the bent-core nematic materials exhibiting giant flexoelectricity were reported [6]. The flexoelectric coefficients characterizing such materials reach the values which are three orders of magnitude larger than those of typical nematics. In this communication, the role of very strong flexoelectric properties in the electric field induced deformations is studied numerically.

Four kinds of homeotropic layers containing flexoelectric nematic liquid crystal, described by both signs of dielectric anisotropy and both signs of the sum of flexoelectric coefficients, were taken into account. The threshold voltage for deformations were calculated for nematic materials of various purity level, characterized by very

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low, moderate and high ion contents. The director distributions in the deformed states were also determined.

The main results are as follows: (i) Giant flexoelectric properties lead to drastic decrease of the threshold voltage. (ii) Even for extremely pure materials containing as low ion concentration as $10^{17}\,\mathrm{m}^{-3}$, the threshold is lower than the theoretical value calculated for perfectly insulating material. (iii) The influence of flexoelectricity on the behaviour of the nematic layer strongly depends on the ion content. (iv) In the case of high ion content, the form of director distributions is not sensitive to the flexoelectric properties.

2. Assumptions, Geometry and Parameters

A nematic liquid crystal layer of thickness $d = 20\,\mu\mathrm{m}$ was held between two infinite plates parallel to the xy plane of the Cartesian co-ordinate system. They were positioned at $z = \pm d/2$, and acted as electrodes. A voltage U was applied between them; the lower electrode ($z = -d/2$) was earthed. Homeotropic alignment, identical on both boundary plates, was assumed. The anchoring strength W was equal to $2 \times 10^{-5}\,\mathrm{Jm}^{-2}$. The director $\mathbf{n}$ was parallel to the xz plane, and its orientation was described by the angle $\theta(z)$, measured between $\mathbf{n}$ and the z-axis. The model substance was characterised by the elastic constants $k_{11} = 6.2 \times 10^{-12}\,\mathrm{N}$ and $k_{33} = 8.6 \times 10^{-12}\,\mathrm{N}$. Two values of dielectric anisotropy, $\Delta\varepsilon = \pm 2$, were adopted whereas $\Delta\varepsilon_{\perp} = 5.4$. The flexoelectric properties were expressed as the sum of the flexoelectric coefficients $e_{11} + e_{33}$, which ranged from -500 to $500\,\mathrm{pCm}^{-1}$ (the individual values of e_{11} and e_{33} are not relevant within the geometry involved [7]).

A weak electrolyte model was adopted for the description of electrical phenomena in the layer [8]. The ion concentrations were determined by the dissociation and recombination constants. The dissociation constant β depended on the electric field strength E: $\beta = \beta_0\left[1 + |E|q^3/\left(8\pi\varepsilon\bar{\varepsilon}k_B^2T^2\right)\right]$, where $\bar{\varepsilon} = (2\varepsilon_{\perp} + \varepsilon_{\parallel})/3$ [9], and its zero field value β_0 was varied from 10^{16} to $10^{22}\,\mathrm{m}^{-3}\mathrm{s}^{-1}$. The transport of ions in the layer was described by typical values of mobility coefficients and diffusion coefficients. It was assumed that the mobility of anions was larger than that of cations: $\mu_{\parallel}^{-} = 1.5 \times 10^{-9}\,\mathrm{m}^2\mathrm{V}^{-1}\mathrm{s}^{-1}$, $\mu_{\perp}^{-} = 1 \times 10^{-9}\,\mathrm{m}^2\mathrm{V}^{-1}\mathrm{s}^{-1}$, $\mu_{\parallel}^{+} = 1.5 \times 10^{-10}\,\mathrm{m}^2\mathrm{V}^{-1}\mathrm{s}^{-1}$ and $\mu_{\perp}^{+} = 1 \times 10^{-10}\,\mathrm{m}^2\mathrm{V}^{-1}\mathrm{s}^{-1}$, which means that $\mu_{\parallel}^{\pm}/\mu_{\perp}^{\pm} = 1.5$. The Einstein relation was assumed for the diffusion constants: $D_{\parallel,\perp}^{\pm} = (k_BT/q)\mu_{\parallel,\perp}^{\pm}$ where q denotes the absolute value of the ionic charge, k_B is Boltzmann constant and T – absolute temperature. The recombination constant α was calculated from the formula $\alpha = 2q\bar{\mu}/(\varepsilon_0\bar{\varepsilon})$, where $\bar{\mu} = [(2\mu_{\perp}^{+} + \mu_{\parallel}^{+})/3 + (2\mu_{\perp}^{-} + \mu_{\parallel}^{-})/3]/2$ [9]. When $\Delta\varepsilon = 2$, this was equal to $3.8 \times 10^{-18}\,\mathrm{m}^3\mathrm{s}^{-1}$ and $4.9 \times 10^{-18}\,\mathrm{m}^3\mathrm{s}^{-1}$ when $\Delta\varepsilon = -2$. The resulting ion concentrations $N_0 = \sqrt{\beta_0/\alpha}$ represented the low, moderate and high values of ion content and were of the order $10^{17} \div 10^{20}\,\mathrm{m}^{-3}$. The conducting properties of the layer was characterized by the rate of the electrode processes (see Appendix). The rate of neutralization of ions as well as the rate of their generation at the electrodes were determined by a single parameter K_r. Its value, $K_r = 10^{-7}\,\mathrm{ms}^{-1}$, which represented the quasi-blocking character of the electrode contacts, reflected the fact that the electrodes are normally coated with insulating aligning films.

3. Method

The problem was considered to be one-dimensional. The reduced co-ordinate $\zeta = z/d$, is used in the following. The functions $\theta(\zeta)$, $V(\zeta)$ and $N^{\pm}(\zeta)$, which describe the director orientation, the potential and ions distribution within the layer, respectively, were determined by resolving of the set of ten equations which consisted of equation of balance of elastic, dielectric and flexoelectric torques for the bulk, two equations of balance of elastic, flexoelectric and anchoring torques for the boundaries, the Poisson equation, two continuity equations for the ion fluxes, four equations for ion concentrations on the boundaries [1]. Their detailed form as well as the assumptions of the model used for description of the electrical properties of the system, [1,10], are given in the Appendix.

4. Results

Numerical resolving of the set of equations mentioned above yield the distributions of the angle $\theta(\zeta)$, electric potential $V(\zeta)$ and ion concentrations $N^{\pm}(\zeta)$. They allowed to determine the director field in the deformed layers and the threshold voltages for the deformations.

4.1. *Threshold Voltage*

4.1.1. *Negative Dielectric Anisotropy.* The threshold voltages for deformations were calculated for various values of the sum of flexoelectric coefficients $e_{11}+e_{33}$ and for four ion concentrations N_0. The results are shown in Figure 1. In the case of the lowest ion content, $N_0=10^{17}\,\mathrm{m}^{-3}$, and for sufficiently low $|e_{11}+e_{33}|$ value, the calculated threshold coincides with the theoretical value corresponding to the insulating nematic. For significantly higher values of $|e_{11}+e_{33}|$, there are ranges for which the deformations appear in two voltage regimes. Similar effect was found also in earlier works [3,4] concerning relatively low value of $e_{11}+e_{33}$, equal to $40\,\mathrm{pCm}^{-1}$, however it appeared at moderate ion concentration of the order $10^{19}\,\mathrm{m}^{-3}$. In the lower voltage regime, the deformations are extremely weak ($\theta(\zeta) < 2$ degree).

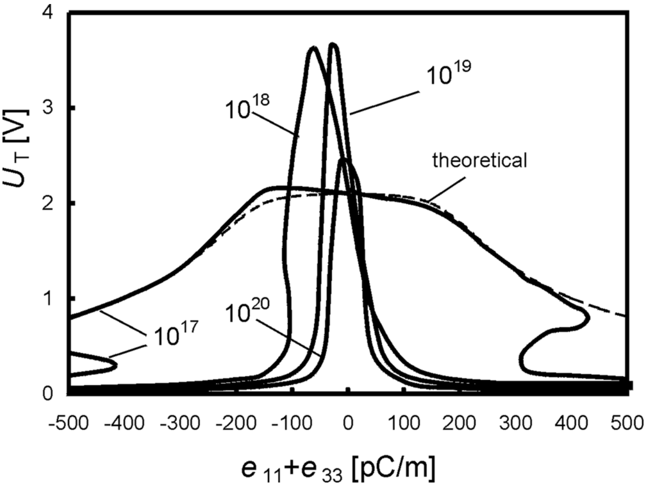

Figure 1. The threshold voltage as a function of the sum of flexoelectric coefficients calculated for four ion concentrations indicated in m^{-3} at the curves; $\Delta\varepsilon=-2$. Dashed line denotes the theoretical values corresponding to insulating nematic.

In the case of moderate and high ion content and for sufficiently low absolute value of negative sum of flexoelectric coefficients, the threshold exceeds remarkably the theoretical value corresponding to the insulating nematic. For every ion content, the giant flexoelectric properties lead to drastic decrease of the threshold voltage down to values below 0.1 V. The threshold voltages for negative sum of flexoelectric coefficients are slightly higher than those for the positive sum with the same $|e_{11}+e_{33}|$.

4.1.2. *Positive Dielectric Anisotropy.* In the case of positive dielectric anisotropy, the torques of dielectric nature stabilize the director distribution. The deformations arise if the absolute value of the sum of flexoelectric coefficients exceeds a critical value determined by the formula $(e_{11}+e_{33})^2 > k_{33}\varepsilon_0\Delta\varepsilon$ [7]. The corresponding limiting values are shown in Figure 2 by two vertical asymptotes. When the sum of flexoelectric coefficients tends to these limiting values, the threshold tends to infinity. For moderate ion content, 10^{18}–$10^{19}\,\mathrm{m}^{-3}$, there are narrow regimes of $e_{11}+e_{33}$ for which the deformations arise in two voltage regions. In the case of the lowest ion content, $N_0 = 10^{17}\,\mathrm{m}^{-3}$, the calculated threshold coincides with the theoretical value corresponding to the insulating nematic. For giant flexoelectric properties the threshold voltage decreases drastically down to values below 0.1 V. This effect is particularly evident for high ion content.

4.2. *Director Distributions*

In order to determine qualitative character of the deformations arising for giant values of flexoelectric coefficients, the director distribution were calculated for voltages exceeding the thresholds by 10%. They are exemplified in Figure 3 for $\Delta\varepsilon = 2$ and $e_{11}+e_{33} = -400\,\mathrm{pCm}^{-1}$. Their form can be interpreted by means of torques which are determined by the electric field distributions plotted in Figure 4.

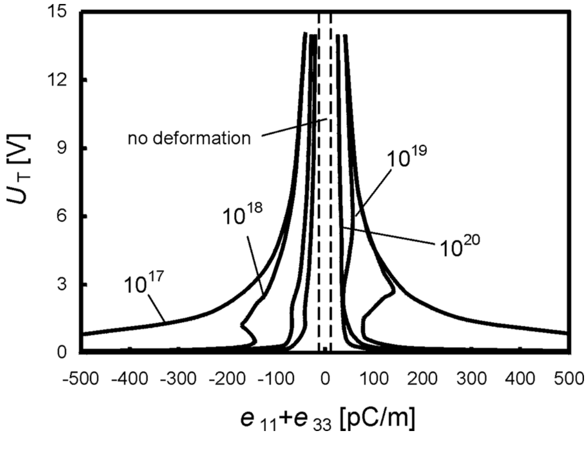

Figure 2. The threshold voltage as a function of the sum of flexoelectric coefficients calculated for four ion concentrations indicated in m^{-3} at the curves; $\Delta\varepsilon = 2$. The theoretical values corresponding to insulating nematic coincide with the data for $N_0 = 10^{17}\,\mathrm{m}^{-3}$. Vertical dashed lines limit the range of $e_{11}+e_{33}$ in which the deformations are absent.

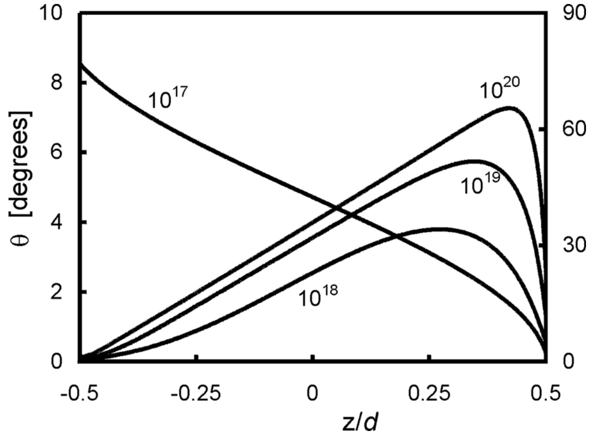

Figure 3. The director distribution as a function of the reduced co-ordinate for four ion concentrations indicated in m^{-3} at the curves; $e_{11}+e_{33}=-400\,\mathrm{pCm}^{-1}$, $\Delta\varepsilon=2$. Curve for $N_0=10^{17}\,\mathrm{m}^{-3}$ corresponds to the right scale, curves for $N_0=10^{18}\div10^{20}\,\mathrm{m}^{-3}$ correspond to the left scale. The bias voltages (in volts) are as follows: 10^{17} – 1.155, 10^{18} – 0.1045, 10^{19} – 0.0605, 10^{20} – 0.0418.

For very low ion content, the angle θ describing the deformations reaches high values on one of the electrodes. The director distributions resemble the deformations predicted theoretically for the insulating nematics. This is due to relatively high electric field ensuring high destabilizing flexoelectric torque acting on the negative electrode. Stabilizing torque on the positive electrode quenches the deformation at $\zeta=1/2$. The bulk flexoelectric torque in the left half of the layer is weak and therefore negligible. For higher ion concentrations, the deformations are weaker and have different character. Their form is mainly due to the action of flexoelectric

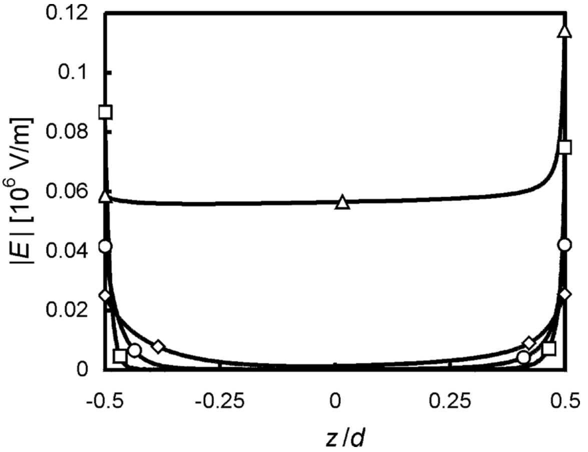

Figure 4. The electric field distributions as functions of the reduced co-ordinate for $e_{11}+e_{33}=-400\,\mathrm{pCm}^{-1}$, $\Delta\varepsilon=2$ corresponding to the director profiles shown in Figure 3. The ion concentrations N_0 expressed in m^{-3} are as follows: 10^{17} – triangles, 10^{18} – diamonds, 10^{19} – circles, 10^{20} – squares.

torques caused by strong electric field gradient induced by space charge of separated ions. The deformation is governed by bulk flexoelectric torques destabilizing in the vicinity of the positive electrode and stabilizing in the neighbourhood of the negative electrode. These features are the more evident, the higher is the ion concentration. In the case of $N_0 = 10^{20}\,\mathrm{m}^{-3}$, the electric field in the bulk of the layer is practically negligible. The deformation is caused only by very high subsurface fields and gradients. The angle describing the director orientation varies almost linearly with the z co-ordinate.

The director profiles for $\Delta\varepsilon = -2$ are qualitatively the same which shows that the sign of dielectric anisotropy is inessential. The director distributions obtained for $e_{11} + e_{33} = 400\,\mathrm{pCm}^{-1}$ and $\Delta\varepsilon = \pm 2$ have also the same form but are reversed with respect to $z = 0$.

5. Summary

The calculations showed that the influence of flexoelectricity on the behaviour of the nematic layer strongly depends on the ion content. In the case of large values of $e_{11} + e_{33}$, and for $N_0 > 10^{18}\,\mathrm{m}^{-3}$, the form of director distributions is not sensitive to ion concentration. In the case of giant flexoelectric properties, the dielectric anisotropy is meaningless. There are some minor quantitative differences in director distributions observed for positive and negative sum of flexoelectric coefficients which are due to difference between mobilities of anions and cations.

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Appendix

The functions $\theta(\zeta)$ and $V(\zeta)$ which describe the director orientation and the potential distribution within the layer are determined by the torque equation:

$$\frac{1}{2}(k_b - 1)\sin 2\theta\left(\frac{\mathrm{d}\theta}{\mathrm{d}\zeta}\right)^2 - \left(\sin^2\theta + k_b\cos^2\theta\right)\frac{\mathrm{d}^2\theta}{\mathrm{d}\zeta^2} + \frac{1}{2}\frac{\varepsilon_0\Delta\varepsilon}{k_{11}}\sin 2\theta\left(\frac{\mathrm{d}V}{\mathrm{d}\zeta}\right)^2 + \frac{1}{2}\frac{e_{11} + e_{33}}{k_{11}}\sin 2\theta\left(\frac{\mathrm{d}^2 V}{\mathrm{d}\zeta^2}\right) = 0 \tag{A1}$$

and the electrostatic equation:

$$\rho(\zeta)d^2 + \varepsilon_0\left(\varepsilon_\perp + \Delta\varepsilon\cos^2\theta\right)\frac{\mathrm{d}^2 V}{\mathrm{d}\zeta^2} - \varepsilon_0\Delta\varepsilon\sin 2\theta\frac{\mathrm{d}V}{\mathrm{d}\zeta}\frac{\mathrm{d}\theta}{\mathrm{d}\zeta} + (e_{11} + e_{33})\cos 2\theta\left(\frac{\mathrm{d}\theta}{\mathrm{d}\zeta}\right)^2 + \frac{1}{2}(e_{11} + e_{33})\sin 2\theta\frac{\mathrm{d}^2\theta}{\mathrm{d}\zeta^2} = 0 \tag{A2}$$

where $k_b = k_{33}/k_{11}$ and $\rho(\zeta) = q[N^+(\zeta) - N^-(\zeta)]$ is the space charge density. The boundary conditions for $\theta(\zeta)$ are determined by the equations describing the surface torques equilibrium

$$\pm\left[\frac{1}{2}\frac{e_{11}+e_{33}}{k_{11}}\sin 2\theta(\pm 1/2)\frac{\mathrm{d}V}{\mathrm{d}\zeta}\bigg|_{\pm 1/2} - \left(\sin^2\theta(\pm 1/2) + k_b\cos^2\theta(\pm 1/2)\right)\frac{\mathrm{d}\theta}{\mathrm{d}\zeta}\bigg|_{\pm 1/2}\right] - \frac{1}{2}\gamma\sin 2\theta(\pm 1/2) = 0 \tag{A3}$$

for $\zeta = \pm 1/2$, where $\gamma = Wd/k_{11}$. The boundary conditions for the potential are $V(-1/2) = 0$ and $V(1/2) = U$.

The transport of ions in the bulk is governed by two equations of continuity of the ion fluxes

$$d(\beta - \alpha N^+ N^-) = \frac{\mathrm{d}J_z^\pm}{\mathrm{d}\zeta}, \tag{A4}$$

where $J_z^\pm = \mp\frac{1}{d}\left(\mu_{zz}^\pm N^\pm \frac{\mathrm{d}V}{\mathrm{d}\zeta} - D_{zz}^\pm \frac{\mathrm{d}N^\pm}{\mathrm{d}\zeta}\right)$ denote the flux of ions of given sign, i.e., the numbers of ions which pass through a surface $\zeta = \mathrm{const}$, counted per unit area and per unit time. The z-components of the mobility and diffusion coefficients are given by $\mu_{zz}^\pm = \mu_\perp^\pm + \Delta\mu^\pm\cos^2\theta$ and $D_{zz}^\pm = D_\perp^\pm + \Delta D^\pm\cos^2\theta$, respectively, where $\Delta\mu^\pm = \mu_\parallel^\pm - \mu_\perp^\pm$ and $\Delta D^\pm = D_\parallel^\pm - D_\perp^\pm$ denote the anisotropies of the two quantities.

In order to establish the boundary conditions for the ion concentrations, a model of phenomena taking place in the bulk and at the electrode-liquid interfaces was used in which the following processes were taken into account:

1. dissociation of dissociable neutral molecules D and recombination of resulting ions: $D \underset{\alpha}{\overset{\beta}{\leftrightarrows}} A^+ + B^-$
2. neutralization of ions at the electrodes: $A^+ + e^- \xrightarrow{K_r} A$ and $B^- \xrightarrow{K_r} e^- + B$,
3. generation of ions at the electrodes

$$\begin{aligned} &D + e^- \xrightarrow{K_g} D^-, \quad D \xrightarrow{K_g} e^- + D^+ \quad \text{and} \\ &B + e^- \xrightarrow{K_g} B^-, \quad A \xrightarrow{K_g} e^- + A^+, \end{aligned} \tag{A5}$$

4. restoring the dissociable molecules

$$A + B = D. \tag{A6}$$

In the equations above, K_r and K_g are reaction rates defined below, common for all the neutralization and generation processes respectively. In order to limit the number of variables and parameters the assumption that the concentrations of A, B, A^+ and B^- are small was used: $N_A \ll N_D$ and $N_B \ll N_D$, as well as $N^+ \ll N_D$ and $N^- \ll N_D$. In this way one may neglect reactions (A5) and (A6). Equivalently, neutral molecules of every kind, A and B (resulting from neutralization of ions) as well as dissociable molecules D, may accept electrons at negative electrode (or donate them at the positive electrode) with identical rate. The total concentration of neutral molecules at the electrode, $N_A + N_B + N_D$, is approximately equal to the equilibrium concentration of dissociable molecules in the absence of electric field, N_d.

The speed of neutralisation of the ions, $n_r^{\pm}$, is proportional to their concentration: $n_r^{\pm} = K_r^{\pm} N^{\pm}$ and the speed of generation, $n_g^{\pm}$, is proportional to the concentration N_d of the neutral dissociable molecules: $n_g^{\pm} = K_g^{\pm} N_d$, where $K_r^{\pm}$ and $K_g^{\pm}$ are suitable constants of proportionality. In the case of zero applied voltage, the equilibrium state realizes in which $N^{\pm} = N_0 = \sqrt{\beta_0/\alpha}$. Therefore $K_r^{\pm} N_0 = K_g^{\pm} N_d$. Since $N^{\pm} \ll N_d$, one may neglect the dependence of N_d on z and ignore the flux of the dissociable molecules. This enables to determine $K_g^{\pm}$ if $K_r^{\pm}$ and N_d are estimated. In general, $K_g^{\pm}$ and $K_r^{\pm}$ can be different for each electrode process. For simplicity however, the calculations were performed using common value of K_r for all neutralization processes as well as common value of $K_g = K_r N_0 / N_d$ for all generation processes at the electrodes.

The processes of charge transfer between the molecules or ions in the liquid crystal and the electrodes are limited by energy barriers arising between the liquid and electrodes. For instance, the transfer of the electron from cathode to a neutral molecule depends on the work function of the electrode as well as on the affinity of the electron to the molecule. Similarly, the transfer of the electron from molecule to anode depends on the ionisation energy of the molecule and also on the work function of the electrode. The rates of generation and neutralisation of the ions at the electrodes can be interpreted in terms of a model in which they are determined by the activation energies ϕ of corresponding electrochemical reactions. For example, the rate of neutralization of a negative ion occurring by the transfer of an electron from the ion to the electrode is equal to $K_r = k_r \exp(-\varphi/k_B T)$, where k_r is a constant. (Absolute values of the parameters φ and k_r are inessential, only the resulting K_r value is important.) Similar formula can be used for the generation constant of positive ions occurring by the transfer of an electron from a neutral molecule to the electrode. (In general, the energy barriers φ can be of different height for every electrode process.) The energy barrier is affected by the electric field existing at the electrode, i.e., increased or decreased by $\Delta\varphi(\pm 1/2) = |E(\pm 1/2)| qL$, where L is the thickness of the subelectrode region, of the order of several molecular lengths. In our calculations $L = 10\,\text{nm}$ was assumed.

As a result of the above assumptions, the boundary conditions take the forms:

$$\mp \mu_{zz}^{\pm} N^{\pm} \frac{dV}{d\zeta} - D_{zz}^{\pm} \frac{dN^{\pm}}{d\zeta} = \left[-N^{\pm} K_r \exp(\pm\Delta\varphi/k_B T) + N_0 K_r \exp(\mp\Delta\varphi/k_B T)\right] d \quad \text{for } \zeta = -1/2 \tag{A7}$$

$$\mp \mu_{zz}^{\pm} N^{\pm} \frac{dV}{d\zeta} - D_{zz}^{\pm} \frac{dN^{\pm}}{d\zeta} = \left[-N^{\pm} K_r \exp(\mp\Delta\varphi/k_B T) + N_0 K_r \exp(\pm\Delta\varphi/k_B T)\right] d \quad \text{for } \zeta = 1/2 \tag{A8}$$

The left hand sides of these equations represent the fluxes of the ions at the electrodes. The first terms on the right hand sides denote the numbers of ions which are neutralised and the second terms – the numbers of ions generated at the electrodes in course of accepting or donating the electrons by the neutral molecules.

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