External electric-field effect on nematic anchoring energy

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The influence of an external field on the effective anchoring energy of a nematic liquid crystal in contact with a substrate is theoretically analyzed. Our analysis is performed on the hypothesis that the electrodes are perfectly blocking and that there is no selective ion adsorption. The proposed theory predicts an effective anchoring energy dependent on the applied dc voltage. According to the sign of the dielectric anisotropy and of the flexoelectric coefficient the dependence of the anchoring energy strength with the bias can be monotonic or not. For large bias voltage the effective anchoring energy strength tends to a constant value. Our theory is in qualitative agreement with published data investigating the influence of the bias on the saturation voltage.

I. INTRODUCTION

The influence of the selective ionic adsorption on the anisotropic part of the anchoring energy strength has been discussed by several authors in the last years [1–15]. According to the proposed models [2,3], the adsorption phenomenon is responsible for an ionic separation inside the liquid. To this charge separation is connected an electric-field distribution across the sample. The coupling of this field with the dielectric [16] and flexoelectric [17] properties of the liquid crystal gives rise to a dielectric energy density, localized near to the limiting surfaces, on mesoscopic thicknesses. This energy can be considered as a surface energy, which renormalizes the anisotropic part of the interfacial energy characterizing the interface nematic liquid-crystal substrate. The distribution of the field across the sample and its connection with the adsorption energy has been recently discussed in [6,18,19]. In the case in which the phenomenon of selective adsorption is absent because the adsorption energy for positive and negative ions is the same, the effect discussed above is absent.

In this paper, we analyze the influence of an external electric field on the nematic anchoring energy when the selective ion adsorption can be neglected. We show that the charge separation induced by the external field gives rise to an effect similar to the one connected to the selective ion adsorption. It is responsible for a bias-voltage dependence of the effective surface energy [14]. Our paper is organized as follows. In Sec. II the description of a liquid containing ions submitted to a dc electric field is presented. The analysis is performed in the framework of Poisson-Boltzmann theory. The peculiarities of the field distribution across the cell and its non-Debye's screening are discussed in the same section. The particular cases relevant to small and large bias voltage are presented in Sec. III. In Sec. IV, we evaluate the dielectric contribution to the surface energy and its dependence on the bias voltage. Section V is devoted to the conclusions, where our results are critically analyzed.

II. POISSON-BOLTZMANN THEORY FOR A LIQUID SUBMITTED TO AN EXTERNAL FIELD

Let us consider an isotropic liquid limited by two nonadsorbing surfaces, at a distance d apart. The z axis is normal to the bounding surfaces, with the origin in the middle of the sample. The liquid is supposed to contain ions. In the absence of external electric field the liquid is globally and locally neutral, i.e., $n_+(z)=n_-(z)=n_0$. If U is the difference of potential across the sample due to the external power supply we have, according to Poisson-Boltzmann theory

$$n_\pm(z)=ne^{\mp\psi(z)}, \tag{1}$$

where

$$\psi(z)=\frac{qV(z)}{k_BT} \tag{2}$$

is the electric potential in k_BT/q units and n is the density of ions where $\psi=0$. Equation (1) has been obtained by assuming $V(\pm d/2)=\pm U/2$, i.e., $V(z)=-V(-z)$. In Eq. (1) $n=n_\pm(0)$ has to be determined by imposing the conservation of the number of ions. From Eq. (1) the bulk density of electric charge is

$$\rho(z)=q[n_+(z)-n_-(z)]=-2nq\sinh\psi(z). \tag{3}$$

A simple calculation shows that

$$\int_{-d/2}^{d/2}\rho(z)dz=-2nq\int_{-d/2}^{d/2}\sinh\psi(z)dz=0, \tag{4}$$

as required.

The conservation of the number of ions implies that

$$n_0d=\int_{-d/2}^{d/2}n_+(z)dz. \tag{5}$$

The distribution of $\psi(z)$ across the sample is obtained by solving the Poisson equation $d^2V/dz^2=-(\rho/\epsilon)$, that by taking into account Eq. (3) reads

$$\frac{d^2\psi}{dz^2}=\frac{1}{\lambda^2}\sinh\psi(z), \tag{6}$$

with the boundary conditions

$$\psi(\pm d/2) = \pm u = \pm \frac{1}{2}\frac{qU}{k_B T}. \tag{7}$$

In Eq. (6), λ is the effective Debye screening length of the liquid when the bias voltage is U. It is given by

$$\lambda^2 = \frac{\epsilon k_B T}{2q^2 n} = \frac{n_0}{n}\lambda_0^2, \tag{8}$$

where $\lambda_0^2 = \epsilon k_B T/(2q^2 n_0)$ is the usual Debye screening length [20]. In the present problem λ depends on the applied voltage.

From Eq. (6) we easily obtain

$$\frac{d\psi}{dz} = \frac{\sqrt{2}}{\lambda}\sqrt{\cosh\psi + k}, \tag{9}$$

where k is an integration constant, whose value is directly connected with the electric field at $z=0$ by

$$k = \frac{\lambda^2}{2}\left(\frac{d\psi}{dz}\right)_0^2 - 1. \tag{10}$$

In particular, if $(d\psi/dz)_0 = 0$, $k = -1$ and Eq. (9) can be easily integrated. This case will be considered in the next section. By means of Eqs. (9) and (8), we can rewrite Eqs. (5) and (7) in the form

$$J(k,u) = \sqrt{2}\frac{d\lambda}{\lambda_0^2} \tag{11}$$

and

$$I(k,u) = \sqrt{2}\frac{d}{\lambda}, \tag{12}$$

respectively, where

$$J(k,u) = \int_{-u}^{u}\frac{e^{-\psi}}{\sqrt{\cosh\psi + k}}d\psi$$

and

$$I(k,u) = \int_{-u}^{u}\frac{1}{\sqrt{\cosh\psi + k}}d\psi. \tag{13}$$

From Eqs. (11) and (12) we obtain

$$I(k,u)J(k,u) = 2\left(\frac{d}{\lambda_0}\right)^2, \tag{14}$$

which determines $k = k(U)$. When this quantity is known, the effective Debye screening length is

$$\lambda = \sqrt{2}\frac{d}{I(k,u)}. \tag{15}$$

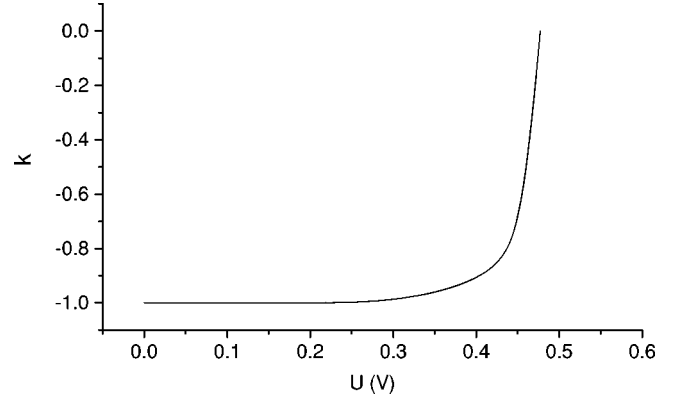In Fig. 1, $k = k(U)$ is reported for $d = 10$ μm and λ_0

FIG. 1. Integration constant k versus the applied voltage U. For small U, $k \to -1$. In this situation the electric field at $z=0$ vanishes. For $d = 10$ μm and $\lambda_0 = 0.36$ μm, this happens for $U < 0.2$ V.

$= 0.36$ μm, typical for a nematic cell made with commercial liquid crystal [21,22]. As it follows from this figure, for bias voltage in the range $0 \leq U \leq 0.2$ V, $(d\psi/dz)_0 \approx 0$, which implies that the electric field for $z=0$ vanishes. This means that for small bias voltage, the ionic charges screen completely the field due to the external power supply. The liquid behaves as a conductor: the ions move until the electric field inside it vanishes. In this case the electric field is localized close to the bounding surfaces over a thickness of the order of $\lambda_0 = \lambda(0)$.

In Fig. 2, we show $\lambda = \lambda(U)$. As it is expected, for $U \to 0$, $\lambda \to \lambda_0$. On the contrary for large U, $\lambda \to \infty$ because all the ions are pushed at the surfaces, and there are no more ions in the liquid, which becomes a true insulating material.

III. LIMITING CASES OF SMALL AND LARGE BIAS VOLTAGE

The reduced voltage across the sample $\psi(z)$ for arbitrary bias voltages has been obtained in the previous section. Now we consider the particular cases of small and large bias volt-
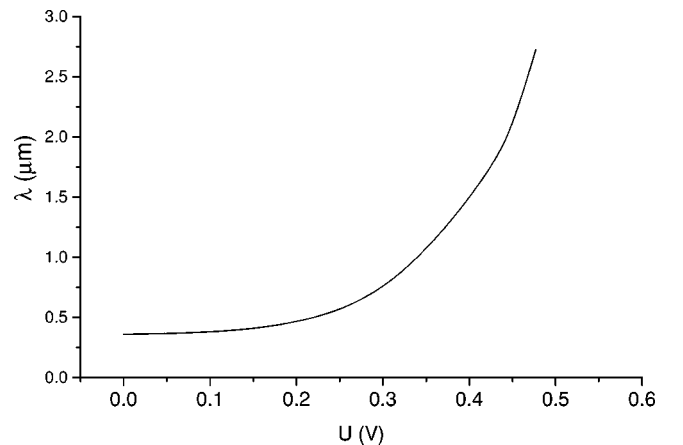

FIG. 2. Effective Debye's screening length λ versus the applied voltage U. For $U \to 0$, $\lambda \to \lambda_0$. For large U all the ions are pushed at the surfaces, and the liquid behaves as a perfect insulator, for which $\lambda \to \infty$. The curve refers to $d = 10$ μm and $\lambda_0 = 0.36$ μm.

age. In the case of a small applied voltage ($u=qU/2k_BT \ll 1$), $|\psi(z)| \le u \ll 1$. Consequently, the solution of Eq. (6) with the boundary conditions (7) is

$$\psi(z)=u\frac{\sinh(z/\lambda)}{\sinh(d/2\lambda)}. \qquad (16)$$

In this case

$$e^{-\psi(z)} \approx 1-\psi(z)=1-u\frac{\sinh(z/\lambda)}{\sinh(d/2\lambda)} \qquad (17)$$

and Eq. (5) gives $n \approx n_0$, which implies

$$\lambda=\lambda_0+\mathcal{O}(u^2). \qquad (18)$$

It follows that in the considered limit, Eq. (16) reads

$$\psi(z)=u\frac{\sinh(z/\lambda_0)}{\sinh(d/2\lambda_0)}. \qquad (19)$$

The charge distributions are then obtained by means of the relations (1), and in the present case are found to be

$$n_\pm(z)=n_0\left[1\mp u\frac{\sinh(z/\lambda_0)}{\sinh(d/2\lambda_0)}\right]. \qquad (20)$$

As expected, the positive ions are collected near the negative electrode, and the negative ions near to the positive one, in surface layers of thickness of the order of λ_0. Another important particular case is the one in which k is very close to -1. As underlined above, in this situation the electric field in the middle of the sample vanishes. By assuming $k=-1$, the solution of Eq. (6), with the boundary conditions (7) is

$$\psi(z)=\psi_-(z)+\psi_+(z), \qquad (21)$$

where

$$\psi_\pm=2\ln\left[\frac{1\pm\gamma e^{(z\mp d/2)/\lambda_0}}{1\mp\gamma e^{(z\mp d/2)/\lambda_0}}\right], \qquad (22)$$

where $\gamma=\tanh(u/4)$. In this limit the total electric potential $\psi(z)$ is obtained by adding the potential $\psi_-(z)$ and $\psi_+(z)$ created by the two surfaces separately. This means that the sample is considered as formed by two half spaces. This approximation works well if $d/2\lambda_0$ is very large.

Let us consider, finally, the case of large applied voltage ($u=qU/2k_BT \gg 1$). In this situation practically all the ions are collected at the surfaces, and in the bulk the liquid can be considered as a perfect insulator. In a first approximation the electric potential across the sample is given by

$$\psi_0(z) \approx 2u\frac{z}{d}. \qquad (23)$$

By substituting Eq. (23) into Eq. (5) we obtain

$$n=2un_0e^{-u}, \qquad (24)$$

and, using Eq. (8) the effective Debye screening length is

$$\lambda^2=\lambda_0^2\frac{e^u}{u}, \qquad (25)$$

showing that for $u \to \infty$, $\lambda \to \infty$ too. By means of Eq. (24) the charge distributions across the sample are found to be

$$n_\pm(z)=2un_0e^{[-u(1\pm 2z/d)]}. \qquad (26)$$

In particular, $n_+(-d/2)=n_-(d/2)=2n_0u$. Equation (26) shows that in the considered limit of large bias voltage, the ionic charges are confined in a surface layer whose thickness is of the order of $d/2u$. Hence, for $d/2u \approx \lambda_0$, i.e., $u \approx d/2\lambda_0$ the thickness of the surface layer is comparable with the Debye screening length.

By putting $\psi(z)=\psi_0(z)+\psi_1(z)$ into Eq. (6) and taking into account Eq. (25) and the boundary conditions (7) we obtain

$$\psi_1(z)=2\frac{e^{-u}}{u}\left(\frac{d}{2\lambda_0}\right)^2\sinh u\left[-2\frac{z}{d}+\frac{\sinh(2uz/d)}{\sinh u}\right]. \qquad (27)$$

Consequently, since in the present limit $u \gg 1$ and, hence, $\sinh u \approx e^u/2$, we have for $\psi(z)$ the expression

$$\psi(z)=2u\frac{z}{d}\left[1-\frac{1}{u^2}\left(\frac{d}{2\lambda_0}\right)^2\right]+2\frac{e^{-u}}{u}\left(\frac{d}{2\lambda_0}\right)^2\sinh\left(2u\frac{z}{d}\right). \qquad (28)$$

From this equation we derive that, in the considered case, in the bulk, the electrical potential is approximately given by

$$\psi_B(z)=2u\frac{z}{d}\left[1-\frac{1}{u^2}\left(\frac{d}{2\lambda_0}\right)^2\right]. \qquad (29)$$

Equation (29) shows that the presence of the ionic charges reduce the effective potential. The trend of $\psi(z)$ differs from $\psi_B(z)$ mainly close to the surfaces at $z=\pm d/2$, for a quantity

$$\Delta\psi(z)=2\frac{e^{-u}}{u}\left(\frac{d}{2\lambda_0}\right)^2\sinh\left(2u\frac{z}{d}\right). \qquad (30)$$

As it follows from Eq. (28) the analysis presented above holds for $(d/2\lambda_0 u)^2 \ll 1$, which implies $u \gg d/2\lambda_0$, or if $d \approx 10$ μm and $\lambda_0 \approx 0.36$ μm we have $U \gg 0.8$ V.

In Fig. 3, the potential across the sample, numerically evaluated by means of Eq. (9) with k and λ given by Eqs. (14) and (15), respectively, is shown for different bias voltages. The trend agrees with the one discussed above. In Fig. 4, the electric field in the bulk $E_B=E(0)$ for different λ_0 is reported. As expected, the presence of the surface charges of ionic origin reduces the bulk electric field. This reduction is, for commercial liquid crystals, rather important. In fact, for a bias voltage of the order of 1 V the actual bulk field is negligible with respect to the one in the absence of the ions. It follows that to identify the bulk electric field with the applied one is not correct.
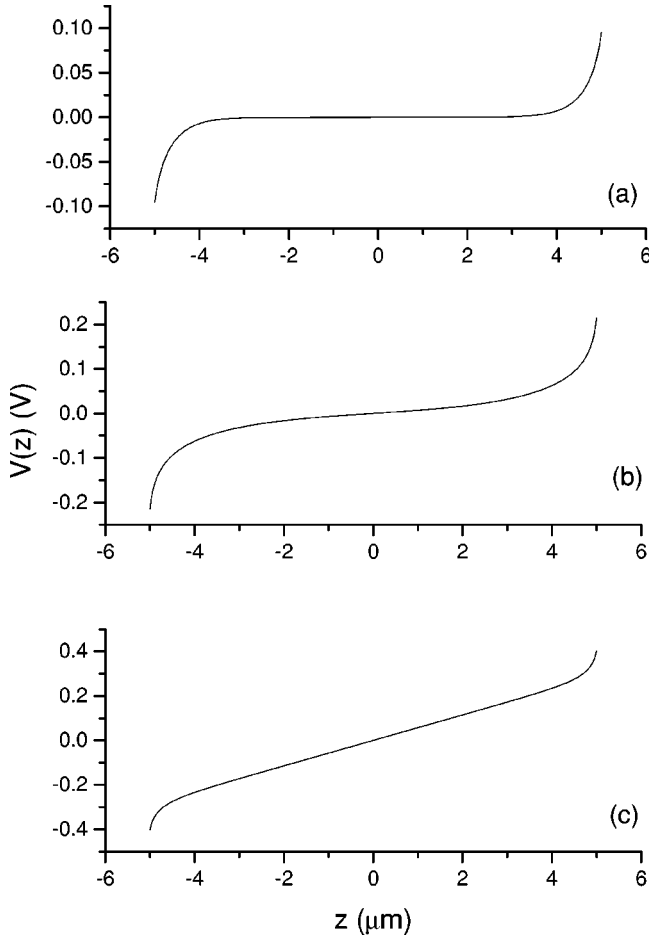
FIG. 4. Bulk electric field $E_B = E(0)$ versus the bias voltage U for $d = 10$ μm, $\lambda_0 \to \infty$ (solid), $\lambda_0 = 0.36$ μm (dotted), $\lambda_0 = 0.1$ μm (dash-dotted).

FIG. 3. Voltage $V(z)$ versus z, for $d = 10$ μm and $\lambda = 0.36$ μm. For small U, E vanishes at $z = 0$. (a) $U = 0.2$ V, (b) $U = 0.4$ V, and (c) $U = 0.8$ V.

IV. CONTRIBUTION TO THE SURFACE ENERGY OF DIELECTRIC ORIGIN

In previous sections, we have evaluated the influence of the ionic charge on the electric potential across a sample of an isotropic liquid. If the liquid is an anisotropic fluid, as a nematic liquid crystal, in a first approximation the electric potential is still given by the equations reported above. However, in this event the presence of the ionic charges gives rise to a surplus of surface energy characterizing the nematic liquid-crystal-substrate interface. To evaluate the dielectric contributions to the surface energy we have to take into account the coupling of the external field with the dielectric anisotropy, f_D, and with the flexoelectric properties of the liquid crystal, f_Q [2,3,16,17].

The quantities f_D and f_Q, which are bulk energy densities, are given by

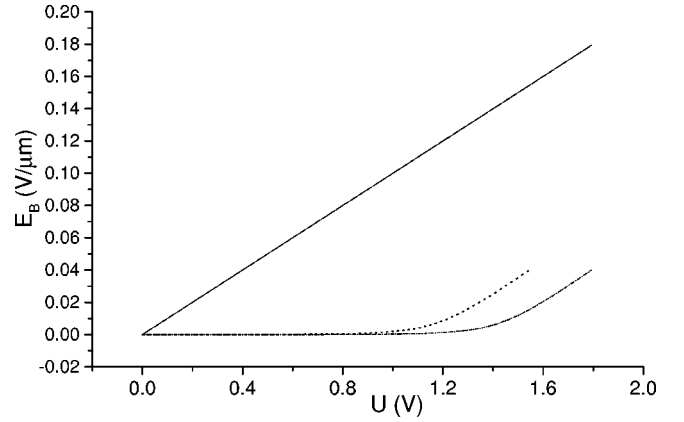$$f_D = -\frac{1}{2}\epsilon_a E(z)^2 \cos^2\theta \tag{31}$$

and

$$f_Q = e\left(\cos^2\theta - \frac{1}{3}\right)\frac{dE}{dz}, \tag{32}$$

where $\theta = \cos^{-1}(\vec{n}\cdot\vec{z})$ is the angle formed by the nematic director $\vec{n}$ with the z axis. Furthermore, $\epsilon_a = \epsilon_\parallel - \epsilon_\perp$ is the dielectric anisotropy ($\parallel$ and $\perp$ refer to $\vec{n}$), and $e = e_{11} + e_{33}$ the total flexoelectric coefficient [2,3,16,17].

Let us indicate by $E_B = E(0)$ and by $E_S = E(d/2)$ the values of the electric field in the middle and at the surface of the sample. The dielectric energy, per unit surface, is

$$F_E = \int_{-d/2}^{d/2}\left[-\frac{1}{2}\epsilon_a E(z)^2\cos^2\theta + e\left(\cos^2\theta - \frac{1}{3}\right)\frac{dE}{dz}\right]dz. \tag{33}$$

This quantity can be written as

$$F_E = \int_{-d/2}^{d/2}\left[-\frac{1}{2}\epsilon_a[E(z)^2 - E_B^2]\cos^2\theta + e\left(\cos^2\theta - \frac{1}{3}\right)\right.$$
$$\left.\times\left(\frac{dE}{dz} - \frac{dE_B}{dz}\right)\right]dz + \int_{-d/2}^{d/2}\left[-\frac{1}{2}\epsilon_a E_B(z)^2\right.$$
$$\times\cos^2\theta$$
$$\left.+ e\left(\cos^2\theta - \frac{1}{3}\right)\frac{dE_B}{dz}\right]dz. \tag{34}$$

Taking into account that $E(z)$ is changing very rapidly close to the limiting surfaces we can put Eq. (34) in the form

$$F_E = f_1 + f_2 + \int_{-d/2}^{d/2}\left[-\frac{1}{2}\epsilon_a E_B(z)^2\cos^2\theta\right.$$
$$\left.+ e\left(\cos^2\theta - \frac{1}{3}\right)\frac{dE_B}{dz}\right]dz, \tag{35}$$

where

$$f_1 = -\frac{1}{2}\epsilon_a \cos^2\theta_1 \int_{-d/2}^{0}[E^2(z)-E_B^2]dz - e\left(\cos^2\theta_1-\frac{1}{3}\right)$$
$$\times(E_S-E_B) \tag{36}$$

and

$$f_2 = -\frac{1}{2}\epsilon_a \cos^2\theta_2 \int_{0}^{d/2}[E^2(z)-E_B^2]dz + e\left(\cos^2\theta_2-\frac{1}{3}\right)$$
$$\times(E_S-E_B), \tag{37}$$

with $\theta_1 = \theta(-d/2)$ and $\theta_2 = \theta(d/2)$. f_1 and f_2 are the dielectric contributions, due to the ions, to the surface energy. The relevant anchoring energy strengths, coinciding with the coefficient of $\cos^2\theta_i$ ($i=1,2$), are then

$$W_D = -\frac{1}{2}\epsilon_a \int_{0}^{d/2}[E^2(z)-E_B^2]dz \tag{38}$$

and

$$W_Q = \pm e(E_S - E_B), \tag{39}$$

where $\pm$ refer to $z=\pm d/2$. Using now Eq. (9) and boundary conditions (7), W_D and W_Q can be written as

$$W_D = -\frac{1}{2}\epsilon_a\left(\frac{k_BT}{q}\right)^2\frac{\sqrt{2}}{\lambda}\int_{0}^{u}\frac{\cosh\psi}{\sqrt{\cosh\psi+k}}d\psi \tag{40}$$

and

$$W_Q = \pm e\frac{k_BT}{q}\frac{\sqrt{2}}{\lambda}(\sqrt{\cosh u+k}-\sqrt{1+k}). \tag{41}$$

In Fig. 5, we report the quantity $W_E = W_D + W_Q$ versus the bias voltage U for $\epsilon_a = 10$ and typical values of e.

As it is evident from Fig. 5, W_E tends to a saturation voltage for large bias, as experimentally observed [14]. In the limit of small voltage, where $k \approx -1$, using Eq. (21) and Eq. (37), we obtain (for $z=d/2$)

$$W_E = -2\frac{k_BT}{q\lambda_0}\left[2\epsilon_a\frac{k_BT}{q}\sinh^2\left(\frac{u}{4}\right)+e\sinh\left(\frac{u}{2}\right)\right]. \tag{42}$$

This equation shows that according to the value of e/ϵ_a are possible nonmonotonic behaviors of $W_E(U)$. For large bias voltage, using Eq. (28) we obtain (for $z=d/2$)

$$W_E = -\frac{1}{2}\left(\epsilon_a\frac{k_BT}{q}+e\right)\frac{k_BT}{q}\frac{d}{\lambda_0^2}+\mathcal{O}(u^{-1}). \tag{43}$$
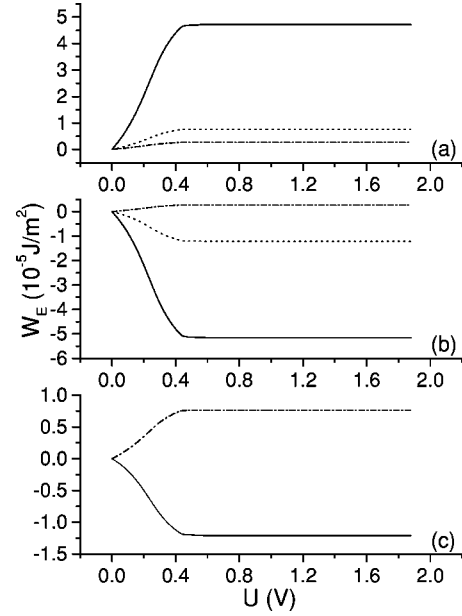

FIG. 5. Dielectric contribution to the surface energy W_E versus the applied voltage U for $\epsilon_a = 10\epsilon_0$, $d=10$ μm, $\lambda_0 = 0.36$ μm and (a) $e=5\times10^{-11}$ C/m (solid), $e=10^{-11}$ C/m (dotted), and $e=5\times10^{-12}$ C/m (dashed); (b) $e=-5\times10^{-12}$ C/m (solid), $e=-10^{-11}$ C/m (dotted); and $e=-5\times10^{-11}$ C/m (dashed), and (c) comparison of W_E versus U for $e=5\times10^{-11}$ C/m (dashed) and $e=-5\times10^{-11}$ C/m (solid).

From this equation we can estimate the order of magnitude of the renormalization of the bare anchoring energy due to the ions dissolved in the liquid crystal. If $\epsilon_a = 10\epsilon_0$, $e=10^{-11}$ C/m [23–25], $\lambda_0 = 0.36$ μm [21,22], and $d=10$ μm, the saturation value for W_E is of the order of 10^{-5} J/m^2. However, if $\lambda_0 = 0.1$ μm, W_E tends to 10^{-4} J/m^2. In other words, the saturation value of W_E strongly depends on λ_0.

V. CONCLUSIONS

We have analyzed the effect of a bias voltage on the ionic distribution in a liquid sample, in the shape of a slab. The case in which the liquid is a nematic liquid crystal has also been considered. In this case we have shown that the nonhomogeneous distribution of the electric field can be described by means of a surface energy of dielectric origin. It is due to the coupling of the electric field with the dielectric anisotropy and with the quadrupolar properties of the liquid crystal. The surface energy of dielectric origin is bias dependent and tends to a saturation value for large applied voltage. Its value ranges, for typical values of ϵ_a and e, from 5×10^{-5} to 10^{-4} J/m^2, for 0.1 μm$\leqslant\lambda_0\leqslant0.36$ μm. This means that in the presence of a dc voltage the renormalization of the anchoring energy due to the bias could be important. To neglect it can be the source of mistakes in the determination of the flexoelectric coefficients.

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