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Effect of surface electric field on the anchoring of nematic liquid crystals

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Abstract. We analyse the influence of adsorbed ions and the resulting surface electric field and its gradient on the anchoring properties of nematics with ionic conductivity. We take into account two physical mechanisms for the coupling of the nematic director with the surface electric field: (i) the dielectric anisotropy and (ii) the coupling of the quadrupolar component of the flexoelectric coefficient with the field gradient. It is shown that for sufficiently large fields near saturated coverage of the adsorbed ions, there can be a spontaneous curvature distortion in the cell even when the anchoring energy is infinitely strong. We also discuss the director distortion when the anchoring energy of the surface is finite.

PACS. 61.30.-v Liquid crystals – 61.30.Gd Orientational order of liquid crystals; electric and magnetic field effects on order – 61.30.Cz Theory and models of liquid crystal structure

1 Introduction

As in any other sample of condensed matter, the surface and interfacial properties of nematic liquid crystals (NLC) are rather complex. In many physical studies as well as practical devices like displays, it is necessary to anchor the orientation of the nematic director at appropriate surfaces in specific directions. Several techniques have been invented for this purpose [1,2]. In practice the anchoring has to be characterized by an angle dependent energy density and the simplest form proposed by Rapini and Papoular [3] consistent with the symmetry of the NLC has the following form:

$$F_{\rm s} = -\frac{1}{2}w(\mathbf{n}\cdot\mathbf{n}_0)^2, \qquad (1)$$

where $\mathbf{n}_0$ is the 'easy' axis along which $\mathbf{n}$ would align on the surface in the absence of all other external fields, and w is the anchoring energy.

Both w and $\mathbf{n}_0$ are assumed to arise from microscopic interactions between the anisotropic molecules of the liquid crystal and those forming the surface [4,5]. Any such surface however breaks the macroscopic symmetry of the nematic leading to the complexities referred to earlier, and discussed extensively in the literature [6]. Over the last few years another aspect of the problem, owing its origin to the "liquid" nature of the medium has been recognized; *viz.*, that it is a weak electrolyte [7]. In display devices the power consumption has to be reduced to the absolute minimum, and special care is taken to purify the sample. On the other hand, the effect of finite conductivity has very interesting consequences, for example leading to a 'nonlocal' character of the anchoring energy itself. Indeed there have been experimental studies [8,9] which have clearly demonstrated the necessity to take into account the influence of adsorbed charges on the surfaces in understanding the anchoring properties as functions of thickness and conductivity. In the first theoretical models the attention was confined to the coupling of the surface electric field produced by the adsorbed charges with the dielectric anisotropy of the medium.

All liquid crystals have flexoelectric properties, and in particular the nonzero quadrupole density arising out of the orientational order in the medium [10] couples with electric field gradients which can be quite large near the surfaces. In the present paper we will discuss the general electrostatic problem near surfaces which incorporates both the dielectric and flexoelectric properties of the medium. The previous treatments of the problem [11–15] were based on the naive assumption that the dielectric and the flexoelectric torques are reduced to only surface contributions, ignoring the elastic torque completely. These were balanced by the torque due to the anchoring energy at the surface. In turn the problem was simply treated as a renormalization in the effective anchoring energy. This approach implies that in the case of strong anchoring there cannot be an instability due to the surface electric field. In this paper we present a more general analysis

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of this problem and show that even in the case of $w \to \infty$, a curvature instability can indeed occur above a threshold double layer potential. If w is finite, the threshold potential naturally gets reduced.

2 Theoretical model

We consider the specific case of an NLC confined between two glass plates treated for homeotropic alignment, *i.e.*, the easy axis $\mathbf{n}_0$ is along $\mathbf{z}$, the normal to the surface. $\theta(z)$ is the polar angle made by the director with respect to the z-axis. The problem is considered to be one-dimensional, *i.e.*, we assume that the surface has uniform properties in the xy-plane. As we mentioned in Section 1, the medium is assumed to contain ionic impurities, and the surface selectively adsorbs one type of ions (usually positively charged) with an adsorption energy $\mathcal{E}$. As is well known in the electrolyte theory, such an adsorption produces a counterion cloud over a depth L_d, called the Debye screening length [16–18]. In turn, there is an electric field which is very strong near the surface ($= E$) and decays as we move away from it. As such, there is a fairly strong field gradient near the surface. In usual liquid crystals, $L_\mathrm{d} \ll d$, where d is the thickness of the sample. Hence, it is sufficient to treat the case of a semi-infinite sample bounded at $z = 0$. The free energy density of the bulk NLC has the following contributions:

(i) the elastic part which is given by

$$f^{\mathrm{el}} = \frac{1}{2}K_{11}(\boldsymbol{\nabla}\cdot\mathbf{n})^2 + \frac{1}{2}K_{33}(\mathbf{n}\times\boldsymbol{\nabla}\times\mathbf{n})^2; \tag{2}$$

(ii) dielectric coupling with the electric field given by

$$f^{\mathrm{diel}} = -\frac{\epsilon_\mathrm{a}}{8\pi}(\mathbf{n}\cdot\mathbf{E})^2, \tag{3}$$

where we have used the cgs system of units, and
(iii) the flexoelectric coupling with the field given by

$$f^{\mathrm{flexo}} = -\mathbf{P}_{\mathrm{fl}}\cdot\mathbf{E} = -[e_1\mathbf{n}(\boldsymbol{\nabla}\cdot\mathbf{n}) + e_3(\boldsymbol{\nabla}\times\mathbf{n}\times\mathbf{n})]\cdot\mathbf{E}. \tag{4}$$

For the sake of simplicity we use the one elastic constant approximation $K_{11} = K_{33} = K$, and in any case near an instability threshold in the homeotropic geometry, only K_{33} is relevant. Using the polar angle θ the total free energy density which is the sum of the above three terms takes the form

$$f = \frac{1}{2}K\theta'^2(z) - \frac{\epsilon_\mathrm{a}}{8\pi}E^2(z)\cos^2\theta(z) - \frac{e}{2}\sin[2\theta(z)]\theta'(z)E(z), \tag{5}$$

where $\theta' = \mathrm{d}\theta/\mathrm{d}z$ and $e = (e_1 + e_3)$ is the sum of the two flexoelectric coefficients defined in equation (4).

The surface energy which is generally assumed to be of the Rapini-Papoular form given by equation (1) can have another contribution if the molecules are polar and the anchoring is homeotropic [19]. It is now experimentally established that there is a surface polarization $\mathcal{P}_\mathrm{s}$ in such a case [20–22]. The angle dependent part of the total surface energy density now becomes [23,24]

$$f_\mathrm{s} = -\frac{1}{2}w(\mathbf{n}\cdot\mathbf{k})^2 - \boldsymbol{\mathcal{P}}_\mathrm{s}\cdot\mathbf{E}_\mathrm{s} = -\frac{1}{2}w\cos^2\theta_0 - \mathcal{P}_\mathrm{s}E_0\cos\theta_0, \tag{6}$$

where θ_0 and E_0 are the values of the polar tilt angle and the electric field at the surface ($z = 0$). Equation (6) was used to describe planar to homeotropic transitions at the nematic surface [25].

The equilibrium configuration in the bulk medium is given as usual by the Euler-Lagrange equation which yields

$$K\theta''(z) - \frac{\epsilon_\mathrm{a}}{8\pi}E^2(z)\sin[2\theta(z)] - \frac{e}{2}E'(z)\sin[2\theta(z)] = 0, \tag{7}$$

which has to be solved with the boundary conditions

$$-K\theta' + \frac{1}{2}(eE_0 + w)\sin(2\theta_0) + \mathcal{P}_\mathrm{s}E_0\sin\theta_0 = 0, \text{ at } z = 0, \tag{8}$$

$$\text{and } \lim_{z\to\infty}\theta'(z) = 0. \tag{9}$$

Note that in the bulk the flexoelectric term couples only with the field gradient as the purely field dependent part is integrated to give only a surface term. Near the threshold we can linearize equations (7, 8) to get

$$K\theta''(z) - \left[\frac{\epsilon_\mathrm{a}}{4\pi}E^2(z) + eE'(z)\right]\theta = 0, \tag{10}$$

for the Euler-Lagrange equation, and

$$-K\theta' + (eE_0 + w + \mathcal{P}_\mathrm{s}E_0)\theta_0 = 0, \tag{11}$$

for the boundary condition at $z = 0$.

We recall that the electric field $E(z)$ is generated in the present problem because of the adsorbed ions on the surfaces and the counterion cloud forming the diffuse double layer in the liquid crystal. The field distribution in this case is well known and it can be written to a very good approximation as [16]

$$E(z) = E_0\exp(-z/L_\mathrm{d}). \tag{12}$$

The total energy for unit surface area is given by

$$\begin{aligned} F &= \int_0^\infty f\,\mathrm{d}z + f_\mathrm{s} \\ &= \int_0^\infty \left[\frac{1}{2}K\theta'^2(z) + \frac{\epsilon_\mathrm{a}}{8\pi}E^2(z)\theta^2(z) - eE(z)\theta(z)\theta'(z)\right]\mathrm{d}z \\ &\quad + \frac{1}{2}(w + E_0\mathcal{P}_\mathrm{s})\theta_0^2, \end{aligned} \tag{13}$$

retaining only terms up to second order in θ.

3 Analysis

The field which acts as the source term for the instability (see Eq. (10)) is localized close to the limiting surface at $z = 0$. Thus it is appropriate to consider an approximate solution of the form

$$\theta(z) = \theta_0 + \Delta\theta[1 - \exp(-z/L_\mathrm{d})] = \theta_\mathrm{b} - \Delta\theta \exp(-z/L_\mathrm{d}), \tag{14}$$

where θ_b is the value of θ in the bulk (*i.e.* at $z \gg L_\mathrm{d}$).

The bulk energy density now takes the simple form

$$f = \frac{1}{L_\mathrm{d}}\left[A\mathrm{e}^{-2z/L_\mathrm{d}} + B\mathrm{e}^{-3z/L_\mathrm{d}} + C\mathrm{e}^{-4z/L_\mathrm{d}}\right], \tag{15}$$

where

$$A = \frac{K(\Delta\theta)^2}{2L_\mathrm{d}} + \frac{\epsilon_\mathrm{a}}{8\pi}E_0^2 L_\mathrm{d}\theta_\mathrm{b}^2 - eE_0\theta_\mathrm{b}\Delta\theta, \tag{16}$$

$$B = -\frac{\epsilon_\mathrm{a}}{4\pi}E_0^2 L_\mathrm{d}\theta_\mathrm{b}\Delta\theta + eE_0(\Delta\theta)^2, \tag{17}$$

$$\text{and} \quad C = \frac{\epsilon_\mathrm{a}}{8\pi}E_0^2 L_\mathrm{d}(\Delta\theta)^2. \tag{18}$$

The total energy per unit surface area in the harmonic approximation given by equation (13) is found to be

$$F = \alpha(\Delta\theta)^2 - \beta\theta_\mathrm{b}\Delta\theta + \gamma\theta_\mathrm{b}^2, \tag{19}$$

where

$$\alpha = \frac{K}{4L_\mathrm{d}} + \frac{eE_0}{3} + \frac{\epsilon_\mathrm{a}}{32\pi}E_0^2 L_\mathrm{d} + \frac{w + E_0\mathcal{P}_\mathrm{s}}{2}, \tag{20}$$

$$\beta = \frac{eE_0}{2} + \frac{\epsilon_\mathrm{a}}{24\pi}E_0^2 L_\mathrm{d} + w + \mathcal{P}_\mathrm{s}E_0, \tag{21}$$

$$\text{and} \quad \gamma = \frac{\epsilon_\mathrm{a}}{16\pi}E_0^2 L_\mathrm{d} + \frac{w + \mathcal{P}_\mathrm{s}E_0}{2}\,. \tag{22}$$

As usual the equilibrium conditions are

$$\frac{\partial F}{\partial(\Delta\theta)} = 2\alpha\Delta\theta - \beta\theta_\mathrm{b} = 0, \tag{23}$$

$$\text{and} \quad \frac{\partial F}{\partial\theta_\mathrm{b}} = -\beta\Delta\theta + 2\gamma\theta_\mathrm{b} = 0. \tag{24}$$

The undistorted state is stable if

$$\mathcal{R} = \frac{\partial^2 F}{\partial(\Delta\theta)^2} = 2\alpha > 0, \tag{25}$$

and the Hessian determinant

$$\mathcal{H} = \frac{\partial^2 F}{\partial(\Delta\theta)^2}\frac{\partial^2 F}{\partial\theta_\mathrm{b}^2} - \left(\frac{\partial^2 F}{\partial(\Delta\theta)\partial\theta_\mathrm{b}}\right) = 4\alpha\gamma - \beta^2 > 0. \tag{26}$$

By taking into account equations (20, 21, 22), relations (25, 26) can be rewritten as

$$\frac{\epsilon_\mathrm{a}}{16\pi}E_0^2 L_\mathrm{d} + \left(\frac{2e}{3} - \mathcal{P}_\mathrm{s}\right)E_0 + \left(\frac{K}{2L_\mathrm{d}} + w\right) > 0, \tag{27}$$

$$\begin{aligned}\text{and} \quad &\left(\frac{K}{4L_\mathrm{d}} + \frac{eE_0}{3} + \frac{\epsilon_\mathrm{a}}{32\pi}E_0^2 L_\mathrm{d}\right)\frac{\epsilon_\mathrm{a}}{4\pi}E_0^2 L_\mathrm{d} \\ &+ \left(\frac{K}{2L_\mathrm{d}} - \frac{eE_0}{3} + \frac{\epsilon_\mathrm{a}}{48\pi}E_0^2 L_\mathrm{d}\right)(w + \mathcal{P}_\mathrm{s}E_0) \\ &- \left(\frac{eE_0}{2} + \frac{\epsilon_\mathrm{a}}{12\pi}E_0^2 L_\mathrm{d}\right)^2 > 0.\end{aligned} \tag{28}$$

Before discussing the general case of arbitrary values of the anchoring energy w, it is instructive to consider the specific case of strong anchoring, *i.e.* $w \to \infty$. It may be noted that this case has not been discussed in the earlier treatments of the problem [11–15], as only the inequality (27) was treated in those papers. As mentioned in the Introduction, this approach is equivalent to reducing the dielectric and flexoelectric contributions to surface terms.

4 Instability thresholds for strong anchoring

When $w \to \infty$, the inequality (27) is always satisfied and it is sufficient to discuss different possibilities using relation (28). It is clear that the instability thresholds correspond to the zero crossings of the coefficient of the $(w + \mathcal{P}_\mathrm{s}E_0)$ term which dominates in the strong anchoring limit. We denote this coefficient by μ. The extremum of this coefficient with respect to E_0, $(\mathrm{d}\mu/\mathrm{d}E_0 = 0)$, corresponds to $E_0 = (8\pi e)/(\epsilon_\mathrm{a}L_\mathrm{d})$, which is a minimum for $\epsilon_\mathrm{a} > 0$ and a maximum for $\epsilon_\mathrm{a} < 0$. The extremum value of μ is given by

$$\mu_\mathrm{e} = \frac{K}{2L_\mathrm{d}} - \frac{4\pi}{3}\frac{\mathrm{e}^2}{\epsilon_\mathrm{a}L_\mathrm{d}} \tag{29}$$

and the fields corresponding to zero crossings of μ are given by [26]

$$E_0 = \frac{8\pi e}{\epsilon_\mathrm{a}L_\mathrm{d}} \pm \sqrt{\left(\frac{8\pi e}{\epsilon_\mathrm{a}L_\mathrm{d}}\right)^2 - \frac{24\pi K}{\epsilon_\mathrm{a}L_\mathrm{d}^2}}. \tag{30}$$

We can now discuss different possibilities.

4.1 Nematic with negative dielectric anisotropy, $\epsilon_\mathrm{a} < 0$

In this case the term in the square root of equation (30) is always positive and larger in magnitude than the first term. Hence in general there are two values of the surface field, one negative and another positive, corresponding to different species of adsorbed charges, between which the homeotropic anchoring is *stable*, and beyond which it gets destabilized. If the flexoelectric coefficient e is positive, the negative threshold field is much larger than the positive

field, and *vice versa* for a negative e. The physical meaning of these results is obvious: while a negative ϵ_a leads to an instability of the director if the electric field is large enough and has either sign, the flexoelectric term stabilizes the homeotropic alignment for one of the signs of the field gradient depending on its own sign.

4.2 Nematic with positive dielectric anisotropy, $\epsilon_a > 0$

In this case two possibilities have to be considered according to the value of ϵ_a.

$$\text{If} \quad \epsilon_a < \frac{8\pi}{3}\frac{e^2}{K} \tag{31}$$

the term in the square root is still positive, but smaller in magnitude than the first term in equation (30). The instability occurs for some value of the field, but the homeotropic alignment gets restabilized at a second higher threshold field in view of the quadratic dependence of the stabilizing dielectric torque on the field. The sign of e decides the sign of the field for which the destabilization occurs: for positive e, E_0 also should be positive, *i.e.* the field gradient should be negative, and *vice versa.*

$$\text{If} \quad \epsilon_a > \frac{8\pi}{3}\frac{e^2}{K} \tag{32}$$

the term under the square root becomes negative and there cannot be any destabilization of the homeotropic alignment.

The case when $\epsilon_a = 0$ will be discussed separately.

5 Threshold values for finite anchoring energy

When the anchoring energy is finite, the zero crossings in relations (27, 28) have to be numerically evaluated for given material parameters and the value of w. This has been done, and as can be expected, as w gets smaller, the threshold field needed for instability becomes lower. For example, if $w = 10^{-2}$ erg/cm^2, which corresponds to an extrapolation length of $\sim 0.5\ \mu$m, which can be attained in the laboratory [8], and $\epsilon_a = -1$, $\mathcal{P}_s = -10^{-3}$ esu [23,24], $L_d = 0.1\ \mu$m [17,18], $K = 10^{-7}$ dyn [27], the threshold double layer potential is about 22 mV for $e = +5 \times 10^{-4}$ esu and ~ 30 mV for $e = -5 \times 10^{-4}$ esu [28]. These values are easily attained in conducting nematic liquid crystals [17,18]. We consider now two simple limiting cases.

5.1 Threshold for a dielectrically isotropic medium

For simplicity we assume that $\mathcal{P}_s = 0$ in further analysis. In this case, when $\epsilon_a = 0$, the stability conditions equations (27, 28) read as

$$\frac{2e}{3}E_0 + \left(\frac{K}{2L_d} + w\right) > 0, \tag{33}$$

$$\text{and} \quad \left(\frac{K}{2L_d} - \frac{eE_0}{3}\right)w - \left(\frac{eE_0}{2}\right)^2 > 0. \tag{34}$$

Equation (33) leads to a threshold condition only if $e < 0$, and in this case it is given by

$$E_1 = \frac{3}{2|e|}\left(\frac{K}{2L_d} + w\right). \tag{35}$$

Equation (34) yields

$$E_2 = -\frac{2w}{3e} - \sqrt{\left(\frac{2w}{3e}\right)^2 + \frac{2Kw}{L_d e^2}}, \tag{36}$$

$$\text{and} \quad E_3 = -\frac{2w}{3e} + \sqrt{\left(\frac{2w}{3e}\right)^2 + \frac{2Kw}{L_d e^2}}\,. \tag{37}$$

In the strong anchoring case $w \to \infty$, both E_1 and E_2 tend to infinity. In this case we get a destabilization of the director field at the threshold given by

$$\lim_{w\to\infty} E_3 = \frac{3K}{eL_d}. \tag{38}$$

E_3 is of course positive for positive e. This means that the threshold occurs for a double layer potential [27,28]

$$V_{th} \sim \frac{K}{|e|} \sim \frac{5\times 10^{-7}}{5\times 10^{-4}} \sim 10^{-3}\ \text{stat V} \sim 0.3\ \text{V}. \tag{39}$$

Indeed such voltages are possible across double layers [29].

5.2 Threshold for a non flexoelectric medium

In this case $e = 0$ and as before we assume $\mathcal{P}_s = 0$. Now the instability threshold, which can occur only if $\epsilon_a < 0$, is given by

$$\frac{|\epsilon_a|}{16\pi}E_0^2 L_d - \left(\frac{K}{2L_d} + w\right) < 0, \tag{40}$$

and

$$-\frac{\epsilon_a^2}{1152\pi^2}L_d^2 E_0^4 + \frac{|\epsilon_a|}{4\pi}\left(\frac{K}{4} + \frac{L_d w}{12}\right)E_0^2 - \frac{Kw}{2L_d} < 0. \tag{41}$$

As before in the strong anchoring limit the threshold condition becomes

$$E_{th} = \frac{1}{L_d}\sqrt{\frac{24\pi K}{|\epsilon_a|}}, \tag{42}$$

whose form is reminiscent of the condition for Freedericksz transition. Again we get a double layer threshold voltage for $\epsilon_a = -4$ to be $V_{th} \sim 1$ V; of the same order as in the previous case.

The above analysis shows that *even when* the anchoring energy w is considered to be infinite, a sufficiently

strong surface electric field generated by adsorbed ions can lead to a destabilization of the homeotropic alignment. If the dielectric anisotropy is negative and the flexoelectric coefficient e is positive, and the anchoring energy is moderate, the destabilizing double layer voltage can be quite low, of the order of 0.1 V, which can be easily attained in practical cases.

Similar considerations are valid for planar alignment. In this case it is easier to destabilize nematic liquid crystals with positive ϵ_a and for a positive surface field, materials with negative e.

6 Conclusions

We have reexamined the influence of adsorbed ions on the orientation of nematic liquid crystals doped with ionic impurities. The earlier approaches treated the problem only for the case of a weak surface anchoring, and treated the effect of the double layer potential as a purely surface effect, ignoring the elastic distortion in the bulk. In this approximation, the surface electric field just renormalizes the finite anchoring energy at the surface and hence it does not influence the director profile if the anchoring is strong. We have removed this limitation in our analysis and shown that the surface electric field can affect the bulk orientation of the director by distorting the director profile near the surface. Indeed such a distortion is found even when $w \to \infty$. This destabilization has two origins, due to both the dielectric anisotropy coupling with E_0^2, and to the flexoelectric coefficient $e = e_1 + e_3$ coupling with the strong field gradient near the surface. For homeotropic anchoring, a positive sign of e leads to a destabilization of the director for a negative field gradient and hence a positive surface field, and is the origin of destabilization in materials with positive ϵ_a. In this case at a second larger threshold the director profile gets restabilized in view of the E^2-dependence of the dielectric coupling. In the case of negative dielectric anisotropy materials, such a restabilization is not possible. In fact it is often found that it is rather difficult to get a homeotropic alignment of materials with negative dielectric anisotropy [29]. We feel that the phenomena discussed in this paper can account for these experimental results.

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