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Anchoring Energy or Surface Melting in Nematic Liquid Crystals?

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The influence of the spatial variation of the scalar order parameter on the anchoring energy is analysed. It is shown that if the nematic average orientation is forced to change too rapidly, a surface melting is expected. Furthermore, by considering two typical situations in which the surface scalar parameter is different or equal to the bulk one, the renormalization of the anchoring energy is estimated.

1. INTRODUCTION

The influence of the spatial variation of the scalar order parameter[1] on the effective anchoring energy[2] has been discussed recently by many authors.[3–6] Despite the importance of the problem, only very rough solutions are reported. In this paper, by means of a perturbative expansion, the effective anchoring energy is connected to the spatial variation of the order parameter. Two typical cases are considered. In the first case the surface is supposed to impose a surface order parameter different from the bulk one. In the second case the surface order parameter and the bulk one are the same in the absence of distortions. As we will show, in both cases the scalar order parameter is not constant across the sample. From this non-uniformity it follows that even in the case of strong anchoring on the nematic average orientation, the effective anchoring energy is found finite.

2. BASIC EQUATIONS

Let us consider a nematic sample of thickness d, limited by two solid surfaces. Let $\vec{n}$ be the average nematic orientation (the "director") and S the scalar order

parameter.[1] The tensor order parameter for the nematic phase is[1]

$$Q_{ij} = S\left(n_i n_j - \frac{1}{3}\delta_{ij}\right). \tag{1}$$

In the case where Q_{ij} is position dependent, the "elastic" free energy density is

$$f = g + \frac{1}{2} L_1 Q_{ij,k} Q_{ij,k} + \frac{1}{2} L_2 Q_{ij,i} Q_{kj,k} \tag{2}$$

where g is the free energy density for a nematic characterized by Q_{ij} = constant and $Q_{ij,k} = \partial Q_{ij}/\partial x_k$.

As well known, g can be written in the form

$$g = g(0) + \frac{1}{2} A Q_{ij} Q_{ij} + \frac{1}{3} B Q_{ij} Q_{jk} Q_{ki} + \frac{1}{4} C_1 (Q_{ij} Q_{ij})^2 + \frac{1}{4} C_2 Q_{ij} Q_{jk} Q_{ke} Q_{ei}, \tag{3}$$

where $g(0)$ is the free energy density of the isotropic phase, and A, B and C_1, C_2 the usual Landau coefficients. The coefficient A is usually written as $A = a(T - T^*)$, where a is a positive constant and T^* a temperature slightly below the critical temperature T_c.

For the sake of simplicity, we limit ourselves to consider a planar problem, in which every physical quantity depends only on one coordinate indicating the distances of the considered point from the surfaces. We use a cartesian reference frame having the *z-axis* normal to the bounding walls, supposedly parallel to the (x, y) plane and placed at $z = \pm d/2$. Furthermore, we suppose $\vec{n}$ to be parallel everywhere to the (x, z) plane, i.e.

$$\vec{n} = (\cos\theta, 0, \sin\theta), \tag{4}$$

where $\theta = \cos^{-1}(\vec{n}.\vec{x})$ is the tilt angle. With the above assumptions f, given by Equation (2), is found to be

$$f = \frac{1}{2} L_1 \dot{S}^2 + \frac{1}{2} L_1 S^2 \dot{\theta}^2 + g(S), \tag{5}$$

where it is supposed $L_2 = 0$,[7] and the dot means d/dz. As follows from Equation (3) $g(S)$ is given by

$$g(S) = g(0) + \frac{1}{2} a(T - T^*) S^2 + \frac{1}{3} B S^3 + \frac{1}{4} C S^4, \tag{6}$$

where $C = C_1 + \frac{1}{2} C_2$. From Equations (5) and (6) one deduces immediately that

a spatial variation of θ is equivalent to a temperature variation of the nematic phase. In fact, by substituting Equation (6) into Equation (5) one obtains

$$f(S, \dot{S}, \dot{\theta}) = \frac{1}{2} L_1 \dot{S}^2 + \frac{1}{2} a(T - T^{*\prime})S^2 + \frac{1}{3} BS^3 + \frac{1}{4} CS^4, \tag{7}$$

where

$$T^{*\prime} = T^*(1 - \xi_o^2 \dot{\theta}^2), \tag{8}$$

in which

$$\xi_o^2 = L_1/aT^*, \tag{9}$$

is the square of the bare correlation length. If we substitute typical values[1] for L_1, a and T_c^* in Equation (9) we find $\xi_o \sim 10^{-7}$ cm.

Equation (7) shows that if $T - T^{*\prime} \sim 1°K$, i.e.

$$|\xi_o \dot{\theta}| \sim \sqrt{\frac{1 + (T^* - T)}{T^*}} \tag{10}$$

a phase transition is expected. This fact suggests that spatial variations of θ which are too large can induce a phase transition. When $\dot{\theta}$ is smaller than the value given by Equation (10), only a decrease of S is expected.

Recent experimental papers[8,9] investigate the surface properties of nematic liquid crystals by measuring the saturation field for the Freedericksz transition. In this situation, near the surface the θ-*gradient* is very large and given approximately by

$$\dot{\theta} \sim \frac{\pi}{2\xi_H}, \tag{11}$$

where ξ_H is the electric (or the magnetic) coherence length.[1] By using Equations (11) and (10) we can conclude that if

$$\xi_o/\xi_H \sim \frac{2}{\pi} \sqrt{\frac{1 + (T^* - T)}{T^*}} \tag{12}$$

the observed "saturation transition" refers to a surface melting and not to a surface reorientation. Let us consider now the experimental data of Blinov and coworkers.[9] The measured saturation field is of the order of $E_{sat} \sim 3 \times 10^5$ V/cm[9]; therefore, the electric coherence length is found to be $\xi_H \sim 10^{-6}$ cm, giving $\sqrt{T^*}\xi_o/\xi_H \sim 1$, i.e. Equation (12) is satisfied. This implies that probably the experimental data of

Blinov and coworkers refer to a surface melting. It would then be useful to reinterpret them in light of this observation.

Up to now we have considered situations in which $\dot{\theta}$ is large enough to satisfy Equation (10). However, even in the case of small $\dot{\theta}$ an S variation is expected. In order to analyse the connection between $S(z)$ and the effective anchoring energy, we will consider a nematic sample in which the $\theta(z)$ variation is due to the surface treatment. In this case, the total free energy of the nematic sample is given by

$$F = \int_{-d/2}^{d/2} f\, dz + F_s^{(-)}(S, \theta) + F_s^{(+)}(S, \theta), \tag{13}$$

where $F_s^{(\pm)}$ refers to the nematic-substrate interaction.[2]

By minimizing F and taking into account Equation (5), the following differential equations

$$2S\dot{S}\dot{\theta} + S^2\ddot{\theta} = 0 \tag{14}$$

$$\ddot{S} - [S\dot{\theta}^2 + R(S)/L_1] = 0 \tag{15}$$

where $R(S) = dg/dS$, for $\theta(z)$ and $S(z)$ are obtained.[10] Equations (14) and (15) must be solved with the boundary conditions

$$L_1 S^2\dot{\theta} + \frac{\partial F_s^{(\pm)}}{\partial \theta} = 0 \quad z = \pm d/2 \tag{16}$$

$$L_1\dot{S} + \frac{\partial F_s^{(\pm)}}{\partial S} = 0 \quad z = \pm d/2, \tag{17}$$

obtained by Equation (13). In the following, Equations (14) and (15) will be solved in different physical situations in order to evaluate an equivalent anchoring energy. For the sake of simplicity, we limit ourselves to consider a sample whose easy axes are characterized by two opposite tilt angles, $\pm\Theta$, and the substrates, at $z = \pm d/2$, having the same physical properties, giving $F_s^{(+)} = F_s^{(-)}$. In this antisymmetrical situation, $\theta(z)$ and $S(z)$ are odd and even functions, respectively: $\theta(z) = -\theta(-z)$, $S(z) = S(-z)$.

3. PERTURBATIONAL METHOD

Let us suppose that $\Theta \ll 1$. In this case it is possible to expand $\theta(z)$ and $S(z)$ in power series of Θ as follows[10]

$$\theta(z) = u_1\Theta + u_3\Theta^3 + \ldots \tag{18}$$

and

$$S(z) = S_o + S_2\Theta^2 + \ldots \tag{19}$$

where $u_i = u_i(z)$ and $S_i = S_i(z)$ $(i = 0, 1, \ldots)$.

By substituting expansions (18) and (19) into Equations (14) and (15) one obtains:

1) at the first order in Θ, u_1 is a solution of the differential equation

$$S_o^2(z)\dot{u}_1(z) = \alpha_1, \tag{20}$$

where α_1 is a constant; and

2) at the second order in Θ, $S(z)$ is obtained by solving the differential equations

$$\ddot{S}_o - (1/L_1)R(S_o) = 0 \tag{21}$$

and

$$\ddot{S}_2 - [S_o\dot{u}_1^2 + (1/L_1)(dR/dS)_{s_o} S_2] = 0 \tag{22}$$

At the lowest order in Θ, it is sufficient to solve Equation (21) for S_o. Subsequently, by means of Equation (20), $u_1(z)$ can be determined, and the equivalent surface energy estimated.

4. SURFACE SCALAR ORDER PARAMETER DIFFERENT FROM THE BULK ONE

If the surface treatment imposes a scalar parameter S_s different from the bulk one, S_b, $S_o(z)$ is not constant across the sample. In this framework, let us suppose that

$$S_o(z) = S_s + s_o(z), \quad \text{with} \quad s_o(z) \ll S_s. \tag{23}$$

In this case, Equation (21) can be rewritten as

$$\ddot{s}_o - (1/L_1)R(S_s + s_o) = 0, \tag{24}$$

which gives, by expanding $R(S_s + s_o)$ in a power series of s_o,

$$\ddot{s}_o - (1/\lambda^2)\, s_o = (1/L_1)R(S_s), \tag{25}$$

where

$$\lambda^{-2} = (1/L_1)(dR/dS)_{S_s} = (1/L_1)(d^2g/dS^2)_{S_s},$$

is the correlation length in the nematic phase[1] evaluated for $S = S_s$. The solution of Equation (25) is

$$s_o(z) = A_o ch(z/\lambda) - (\lambda^2/L_1)R(S_s), \tag{26}$$

where we have taken into account that in our situation $S(z) = S(-z)$ and hence $s(z) = s(-z)$.

In the hypothesis that $S_o(z = d/2) = S_s$, i.e. that the "anchoring" over S is strong at the surface, $s_o(d/2) = 0$; consequently, from Equation (26) we deduce for the integration constant A_o

$$A_o = \frac{\lambda^2}{L_1} R(S_s) \cdot \frac{1}{ch(d/2\lambda)}. \tag{27}$$

It follows that, in the case of the fixed surface order parameter, $s_o(z)$ is

$$s_o(z) = \frac{\lambda^2}{L_1} R(S_s) \left\{ \frac{ch(z/\lambda)}{ch(d/2\lambda)} - 1 \right\}; \tag{28}$$

therefore, at the zero $-$th order in Θ, the scalar order parameter is given by

$$S_o(z) = S_s + \frac{\lambda^2}{L_1} R(S_s) \left\{ \frac{ch(z/\lambda)}{ch(d/2\lambda)} - 1 \right\}. \tag{29}$$

Note that $s_o(0) \sim (\lambda^2/L_1)\, R(S_s)$, because, in general, $\lambda << d$. Since $S(0) = S_b = S_s + s_o(0)$ we find

$$(\Delta S)_o = S_s - S_b = (\lambda^2/L_1) R(S_s). \tag{30}$$

In the event that S_s is not fixed by the surface, the constant A_o appearing in Equation (26) must be determined by using the boundary condition (17), but this case will not be discussed here, since it is not very interesting.

By using Equation (29), Equation (20) gives

$$\dot{u}_1 = \frac{\alpha_1}{S_o^2(z)} \sim \frac{\alpha_1}{S_s^2} \left[1 - 2 \frac{s_o(z)}{S_s} \right], \tag{31}$$

from which we deduce

$$u_1(z) = \frac{\alpha_1}{S_s^2} \left\{ z - \frac{2}{S_s} \Delta_o(z) \right\}, \tag{32}$$

where

$$\Delta_o(z) = \int_o^z s_o(z')dz'. \tag{33}$$

Equation (32) is valid in general. To obtain Equation (32) we have taken into account that in our case $\theta(z) = -\theta(-z)$, and hence $u_i(z) = -u_i(-z)$.

4.1 Strong Anchoring on θ

In the event that the anchoring energy on θ is strong

$$\theta(d/2) = \Theta, \tag{34}$$

for every bulk distortion; consequently,

$$u_1(d/2) = 1 \quad \text{and} \quad u_i(d/2) = 0, \, i = 2, 3 \ldots \tag{35}$$

In this case Equation (32) gives

$$\alpha_1 = \frac{2}{d} S_s^2 \frac{1}{1 - (4/d)[\Delta_o(d/2)]/S_s}, \tag{36}$$

and hence

$$u_1(z) = \frac{2}{d} \frac{z - (2/S_s)\Delta_o(z)}{1 - (4/d)[\Delta_o(d/2)]/S_s}. \tag{37}$$

By substituting Equation (28) in Equation (33), Equation (37) gives

$$u_1(z) = \frac{2}{d} z \frac{1 - 2\dfrac{(\Delta S)_o}{S_s}\left\{\dfrac{\lambda}{z}\dfrac{sh(z/\lambda)}{ch(d/2\lambda)} - 1\right\}}{1 - 2\dfrac{(\Delta S)_o}{S_s}\left\{2\dfrac{\lambda}{d}\dfrac{sh(d/2\lambda}{ch(d/2\lambda)} - 1\right\}}. \tag{38}$$

It follows that, at the first order in Θ, θ(z) is found to be

$$\theta(z) = u_1(z)\Theta, \tag{39}$$

where $u_1(z)$ is given by Equation (38). From Equation (39) we obtain for the bulk θ − *gradient*

$$\dot{\theta}(0) = \dot{u}_1(0)\Theta \sim \frac{2}{d}\Theta \frac{1 + 2\dfrac{(\Delta S)_o}{S_s}}{1 + 2\dfrac{(\Delta S)_o}{S_s}\left(1 - 2\dfrac{\lambda}{d}\right)}, \tag{40}$$

giving for the extrapolated surface tilt angle the expression

$$\theta_s = \dot{\theta}(0)\frac{d}{2} \sim \Theta\left\{1 + 4\frac{(\Delta S)_o \lambda}{S_s d}\right\}. \tag{41}$$

In the hypothesis of $S(z) = S_b = \text{const}$ and weak anchoring energy, θ_s is given by

$$\theta_s = \frac{\Theta}{1 + 2(L/d)}, \tag{42}$$

where $L = L_1 S_s^2/W$ is the surface extrapolation length.[2]

From Equations (41) and (42) we deduce that the equivalent extrapolation length is

$$L_{eq} = -2\frac{(\Delta S)_o}{S_s}\lambda. \tag{43}$$

In the event that $(\Delta S)_o = 0$, i.e. $S_o = \text{const}$, Equation (43) gives $L_{eq} = 0$ at the first order in Θ. In this case, the analysis must be performed by also considering the third order in Θ as we will show in Section 5.

4.2 Weak Anchoring on θ

In the event that the anchoring energy on θ is finite, the constant α_1, appearing in Equation (42), must be determined by using the boundary conditions in Equations (16) and (17). By supposing that

$$F_s^{(\pm)} = \frac{1}{2} W[\theta(\pm d/2) \mp \Theta]^2, \tag{44}$$

Equation (16) is written

$$L_1 S_s^2 \dot{\theta} + W[\theta(d/2) - \Theta] = 0. \tag{45}$$

At the first order in Θ, Equation (45) gives

$$L_1 S_s^2 \dot{u}_1(d/2) + W[u_1(d/2) - 1] = 0. \tag{46}$$

By using Equations (46) and (42) we obtain

$$\alpha_1 = \frac{2}{d}\frac{S_s^2}{2(L/d) + 1 - (2/S_s)\dfrac{\Delta_o(d/2)}{d/2)}}, \tag{47}$$

instead of Equation (36). It follows that in the case of weak anchoring $u_1(z)$ is given by

$$u_1(z) = \frac{2}{d}\cdot z \cdot \frac{1 - (2/S_s)\dfrac{\Delta_o(z)}{z}}{1 - (2/S_s)\dfrac{\Delta_o(d/2)}{d/2} + 2\dfrac{L}{d}}. \tag{48}$$

By following the previous step-by-step procedure, we have for the bulk θ-*gradient*

$$\dot{\theta}(0) = \Theta \dot{u}_1(0) = \frac{2}{d}\Theta \frac{1 - \frac{2}{S_s} s_o(0)}{1 + \frac{2}{d}\left[L - \frac{2}{S_s}\Delta_o(d/2)\right]}, \tag{49}$$

and hence the extrapolated tilt angle is found to be

$$\theta_s = \dot{\theta}(0)\frac{d}{2} = \Theta \frac{1 - \frac{2}{S_s} s_o(0)}{1 + \frac{2}{d}\left[L - \frac{2}{S_s}\Delta_o(d/2)\right]}. \tag{50}$$

By comparing Equation (50) with Equation (42), we deduce that an S variation can be considered equivalent to:

i) a variation of the easy axis:

$$\Theta \to \Theta_{eff} = \Theta\left[1 - \frac{2}{S_s} s_o(0)\right] = \Theta\left[1 + 2\frac{(\Delta S)_o}{S_s}\right], \text{ and}$$

(ii) a variation of the extrapolation length

$$L \to L_{eff} = L - \frac{2}{S_s}\Delta_o(d/2) = L - 2\frac{(\Delta S)_o}{S_s}[\lambda - d/2], \tag{52}$$

as follows from Equations (28), (30) and (33). We can interpret Equation (50) also in another way, taking into account that it can be rewritten as

$$\theta_s = \Theta\left[1 - 2\frac{s_o(0)}{S_s}\right] \Big/ \left[1 - \frac{2}{S_s}\frac{\Delta_o(d/2)}{d/2} + 2\frac{L}{d}\right], \tag{50'}$$

and that, from Equations (29) and (30) and (33)

$$1 - 2\frac{s_o(0)}{S_s} = 1 + 2\frac{(\Delta S)_o}{S_s}, \frac{\Delta_o(d/2)}{d/2} = -(\Delta S)_o\left(1 - 2\frac{\lambda}{d}\right),$$

we obtain

$$\theta_s = \Theta \frac{1 + 2\frac{(\Delta S)_o}{S_s}}{1 + 2\frac{(\Delta S)_o}{S_s} + \frac{2}{d}\left[L - 2\lambda\frac{(\Delta S)_o}{S_s}\right]} = \Theta \cdot \frac{1}{1 + \frac{2}{d}\frac{L - 2\lambda\frac{(\Delta S)_o}{S_s}}{1 + 2\frac{(\Delta S)_o}{S_s}}}. \tag{50''}$$

Equation (31) reveals that Θ (the easy axis) can be considered unchanged, and the equivalent anchoring extrapolation length is

$$L_{eq} = \frac{L - 2\lambda \dfrac{(\Delta S)_o}{S_s}}{1 + 2\dfrac{(\Delta S)_o}{S_s}}, \tag{51}$$

which is a generalization of a known formula.[5,6]

5. SURFACE SCALAR ORDER PARAMETER EQUAL TO THE BULK ONE

In the event that $(\Delta S)_o = 0$, i.e. $S_s = S_b$, the previous analysis must be performed by taking into account that at the zero order in Θ, S_o is constant across the sample. In this case, by following the previous step-by-step procedure we obtain from Equations (21) and (22)

$$s_o(z) = 0 \tag{52}$$

$$s_2(z) = \left(2\frac{\lambda}{d}\right)^2 S_o\left\{\frac{ch(z/\lambda)}{ch(d/2\lambda)} - 1\right\} \tag{53}$$

and

$$u_1(z) = (2/d)z \tag{54}$$

$$u_2(z) = 2\left(2\frac{\lambda}{d}\right)^3 \frac{ch(d/2\lambda)}{ch(d/2\lambda)}\left\{2\frac{z}{d} - \frac{sh(z/\lambda)}{sh(d/2\lambda)}\right\}, \tag{55}$$

in the strong anchoring case for $\theta(z)$ and $S(z)$.

Since $d >> \lambda$, $sh(d/2\lambda)/ch(d/2\lambda) \sim 1$, and $sh(z/\lambda)/sh(d/2\lambda)$ is different from zero only in a surface layer of thickness λ. The tilt angle, up to the third order in Θ, is given by

$$\theta(z) \sim 2\frac{z}{d}\Theta + 2\left(2\frac{\lambda}{d}\right)^3\left\{2\frac{z}{d} - \frac{sh(z/\lambda)}{sh(d/2\lambda)}\right\} \tag{56}$$

instead of (38) and (39). Note that in the considered situation the scalar order parameter is given by

$$S(z) = S_o\left\{1 + \left(2\frac{\lambda}{d}\right)^2\left[\frac{ch(z/\lambda)}{ch(c/2\lambda)} - 1\right]\right\}. \tag{57}$$

Consequently,

$$S_b = S(0) = \left[1 - \left(2\frac{\lambda}{d}\right)^2 \Theta^2\right] S_o < S_o. \tag{58}$$

as expected, the bulk order parameter is smaller than the surface one.

Now the extrapolated surface tilt angle is given by

$$\theta_s \sim \Theta \left\{1 + 2\left(2\frac{\lambda}{d}\right)^3 \Theta^2\right\} \tag{59}$$

and the equivalent anchoring energy is then

$$L_{eq} = \left(2\frac{\lambda}{d}\right)^2 \Theta^2 \cdot 2\lambda = -2\frac{\Delta s}{s_o}\lambda, \tag{60}$$

where, according to Equation (38) we have put

$$\Delta S = S_o - S_b = \left(\frac{2\lambda}{d}\right)^2 \Theta^2, \tag{61}$$

Equation (60) coincides with Equation (43) reported above.

CONCLUSIONS

The influence of a spatial variation of the scalar order parameter on the macroscopic anchoring energy has been considered. Our analysis shows that if:

1) the nematic average orientation θ is forced to change in such a way that $T^*\xi_o^2\dot{\theta}^2 \sim 1$, for the liquid crystal it is better to melt than to orient itself in elastic manner;
2) the surface treatment imposes a surface order parameter different from the bulk one (depending only on the temperature), the spatial variation of S is equivalent to a finite anchoring energy, that renormalizes the total anchoring energy, and
3) the surface treatment imposes a surface order parameter equal to the bulk one (at a given temperature), the spatial variation of the average orientation produces a spatial variation of the scalar order parameter. This last effect is equivalent to a finite anchoring energy. This fact implies that, in general, the equivalent anchoring energy depends, nonlinearly, on the imposed deformations.

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