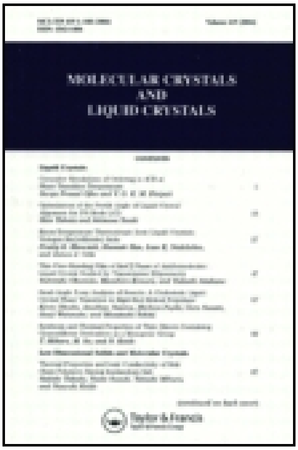

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Remarks on Critical Thickness of a Hybrid Aligned Nematic Cell with Large Elastic Anisotropy[†]

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We analyze the critical thickness of a hybrid aligned cell, below which the nematic liquid crystal assumes an undistorted configuration. We show that: (*a*) if $k_{11} \simeq k_{33}$, the critical thickness exists only in the case of different anchoring energies, between the walls; (*b*) if $k_{11} \ll k_{33}$, the critical thickness exists only if the anchoring energy for homeotropic orientation is greater than that for the planar orientation. Conversely, if $k_{11} \gg k_{33}$, the anchoring energy for the planar orientation should be greater than that for the homeotropic orientation.

INTRODUCTION

It is known that nematic liquid crystal films subjected to competitive boundary conditions are distorted,[1] and give the so-called hybrid alignment. However, Terminassian-Saraga, *et al.*,[2] have shown experimentally that there exists a critical thickness of the sample below which the director field becomes uniform along the easy axis with the bigger anchoring energy.[3] A theoretical analysis[4] shows that, with no external field, the critical thickness depends both on elastic constants and anchoring energies. The calculations performed in Ref. 4 are valid if $k_{11}/k_{33} < 1$, where k_{11} and k_{33} are the Frank elastic constants for splay and bend deformation, respectively.

If we suppose that the intermolecular force is the sum of hard rod repulsion and Maier-Saupe-type attraction, it is possible to calculate the

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Frank elastic constants k_{ii} in the mean field approximation. On this hypothesis, Priest[5] and Kimura, *et al.*,[6] have shown on expanding the total free energy density up to the second order in the scalar order parameters (that is a measure of the degree of alignment of the molecules along the director **n**) that it is possible to demonstrate that $k_{11} = k_{33}$ and all three elastic constants vary with temperature as S^2.

Furthermore, they have shown that deviations from the equality $k_{11} = k_{33}$ are related to the ratio $\langle P_4 \rangle / S$, in the following way:

$$k_{11} = \alpha S^2 \, (1 + \beta - 3\gamma \, \langle P_4 \rangle / S)$$

$$k_{33} = \alpha S^2 \, (1 + \beta + 4\gamma \, \langle P_4 \rangle / S)$$

where $\langle P_4 \rangle$ is the average value of the fourth Legendre polynomial, and α, β and γ are positive constants related to molecular properties. Therefore, in the hard-rod model, the inequality $k_{33} > k_{11}$ holds, and k_{33}/k_{11} decreases with temperature.

The provision $k_{33} > k_{11}$ is confirmed for many nematic liquid crystals.[7-13] However, as shown by Scudieri,[14,15] the elastic ratio can fall within a large range. Hence, it seems interesting to analyze the critical thickness in the case of a large elastic ratio.

DISCUSSION

De Jeu and Claassen[16] have found in certain nematic homologous series that the ratio k_{11}/k_{33} increases with the alkyl chain length and $k_{11}/k_{33} > 1$ for the longest chains. For these molecules, the elastic ratio increases with temperature, in contradiction of the theories of Priest and Kimura, *et al.* It is known that these deviations from theory, and other effects on the mechanical properties[17] are related to the flexibility of the molecules, implying that the hard rod model is not sufficient (for the molecular aspect of this problem see Ref. 18). In this case, it would be necessary to take into account the effect of molecular deformation in the calculation of the free energy, and consider, for example, the partial order parameters S_i relative to each molecular fragment, and some averages[19] of S_i as order parameters.

Consequently, in order to determine the critical thickness in a general way, we must consider either $k_{33} > k_{11}$ or $k_{33} < k_{11}$. The main purpose of the present paper is to recalculate d_c in the case of an arbitrary elastic ratio.

We suppose that:

(1) the cell is an infinite slab (i.e., the sample thickness is much smaller than the linear dimensions of the cell plates);

(2) the z-axis is normal to the walls limiting the sample and the walls lie in the x-y plane;

(3) the director lies in the plane (x, y): in this way, only one polar angle is necessary to describe the nematic liquid crystal deformation (the twist deformation is absent);

(4) ϕ is the tilt angle between the director $\mathbf{n}$ and the z-axis;

(5) the boundary conditions imposed by the walls at $z = \pm d/2$ are constant, i.e. the tilt angle is invariant with respect to translation parallel to the x-axis (ϕ depends only on z: $\phi = \phi(z)$);

(6) the boundary walls are treated to give homeotropic orientation at $z = -d/2$ and planar orientation at $z = +d/2$;

(7) the surface interaction is $(w/2)\sin^2(\phi-\Phi)$,[20] indicating the easy direction by Φ and the anchoring strength coefficient by w. Labelling with 1 and 2 the lower and upper wall respectively, we have $\Phi_1 = 0$ and $\Phi_2 = \pi/2$; hence, the surface energy results as: $(w_1/2)\sin^2\phi_1 + (w_2/2)\cos^2\phi_2$, where $\phi_1 = \phi(-d/2)$ and $\phi_2 = \phi(d/2)$.

The case $k_{33} \geq k_{11}$

In the frame of the continuum theory, the total free energy per unit wall area of the hybrid aligned cell is given by

$$F(\phi) = (k_{33}/2)\left\{\int_{d/2}^{d/2} C^2(k,\phi)(d\phi/dz)^2\,dz + L_1^{-1}\sin^2\phi_1 + L_2^{-1}\cos^2\phi_2\right\} \tag{1}$$

where $C^2(k,\phi) = 1-k\sin^2\phi$, $k = 1-(k_{11}/k_{33})$, $L_i = k_{33}/w_i$. In Eq. 1, there are a volume term and a surface term. Cognard[20] has shown the existence of a thin "smectic-like" interfacial layer where the anchoring energy is very strong (>1 erg/cm^2) and where the order parameter is greater than the value in the bulk. The orientation in this layer cannot be modified by any elastic bulk distortion. However, the experimental values for the anchoring energy are much smaller ($\sim 10^{-3}$ erg/cm^2), showing that besides the "smectic-like" layer, the anchoring energies are of the same order of magnitude as the volume terms on the shear-plane (where the order parameter reaches the bulk value). Then, in order to know the director deformation inside the cell, we work out the minimization of Eq. 1 including both the terms.[21]

In this case, since $k_{11} < k_{33}$, $C(k,\phi)$ is real for any k. It is well known that the functional $F = F(\phi)$ has a stationary value if the function ϕ is a solution of the Euler-Lagrange Eq. 2.

$$C^2(k,\phi)(d\phi/dz)^2 = p^2 \tag{2}$$

for any z falling in the range $(-d/2, d/2)$, and satisfies the boundary conditions imposed by the finite anchoring energies, i.e.:[22]

$$2L_iC(k, \phi_i)\,(d\phi/dz)_i = \sin 2\phi_i, \qquad \text{for} \quad z = \pm d/2 \tag{3}$$

In Eq. 2, p is an integration constant, fixed by the boundary conditions. From Eqs. 2 and 3 we obtain

$$p = (1/d)\, I(\phi_1, \phi_2; k) \tag{4}$$

$$2(L_i/d)\, C(k, \phi_i)\, I(\phi_1, \phi_2; k) = \sin 2\phi_i \tag{5}$$

where

$$I(\phi_1, \phi_2; k) = \int_{\phi_1}^{\phi_2} C(k, \mu)\, d\mu\,.$$

Using Eqs. 4 and 5, it is possible to calculate ϕ_1, ϕ_2 and p and hence from Eq. 2, the function ϕ. In this way, we obtain the tilt angle function ϕ for which $F(\phi)$ has an extremum.

Actually, the basic principle involved in the application of the fundamental equation of the continuum theory to the solution of this problem is that the equilibrium state of the director field is always given by the director configuration that minimizes the total free energy of the system with specified boundary conditions.

Then in order to know the director deformation inside the cell, we must consider the minimum of the function

$$\psi(\phi_1, \phi_2; k) = (1/d) I^2(\phi_1, \phi_2; k) + \psi_s(\phi_1, \phi_2) \tag{6}$$

obtained by substituting Eqs. 4 and 2 in 1. In Eq. 6 we put $\psi = 2F/k_{33}$ and $\psi_s = L_1^{-1} \sin^2 \phi_1 + L_2^{-1} \cos^2 \phi_2$. Hence it is necessary to study the following partial derivatives of $\psi(\phi_1, \phi_2; k)$:

$$\begin{aligned} g_i(\phi_1, \phi_2; k) &= \partial_i \psi(\phi_1, \phi_2; k) \\ f_{ij}(\phi_1, \phi_2; k) &= \partial_{ij} \psi(\phi_1, \phi_2; k) \\ H(\phi_1, \phi_2; k) &= \det[f_{ij}(\phi_1, \phi_2; k)] \end{aligned} \tag{7}$$

where $\partial_i = \partial/\partial\phi_i$ and so on, and the minimum is found setting[23] $g_i(\phi_1, \phi_2; k) = 0$, $f_{ii}(\phi_1, \phi_2; k) > 0$ $H(\phi_1, \phi_2; k) > 0$. Using well known theorems[24] in order to calculate the derivative of the function $I(\phi_1, \phi_2; k)$ we get:

$$\begin{aligned} \partial_i \psi_s(\phi_1, \phi_2) &= (-1)^{i+1} \sin 2\phi_i / L_i \\ \partial_{ij} \psi_s(\phi_1, \phi_2) &= 2(-1)^{i+1} \delta_{ij} \cos 2\phi_i / L_i \\ \partial_i I(\phi_1, \phi_2; k) &= (-1)^i C(k, \phi_i) \\ \partial_{ij} I(\phi_1, \phi_2; k) &= (-1)^{i+1} \delta_{ij} S(k, \phi_i) \end{aligned} \tag{8}$$

where $S(k, \phi_i) = k \sin 2\phi_i/2C(k, \phi_i)$. In the previous relations we consider linearly independents ϕ_i so that $\partial_j\phi_i = \delta_{ij}$. If we take into account Eqs. 8, after trivial calculations, the relations, Eq. 7 become

$$g_i(\phi_1, \phi_2; k) = (-1)^i\{(2/d)I(\phi_1, \phi_2; k)C(k, \phi_i) - (1/L_i) \sin 2\phi_i\}$$

$$f_{11}(\phi_1, \phi_2; k) = 2\{(1/d)\,[C^2(k, \phi_1) + I(\phi_1, \phi_2; k)S(k, \phi_1)] + (1/L_1) \cos 2\phi_1\}$$

$$\begin{aligned} H(\phi_1, \phi_2; k) = {} & (4/d)\{[G_-(k, \phi_2)S(k, \phi_1) - G_+(k, \phi_1)S(k, \phi_2) \\ & - (1/d)S(k, \phi_1)S(k, \phi_2)I(\phi_1, \phi_2; k)]I(\phi_1, \phi_2; k) \\ & + (1/L_1L_2)\,[L_2C^2(k, \phi_2) \cos 2\phi_1 - L_1C^2(k, \phi_1) \cos 2\phi_2 \\ & - d \cos 2\phi_1 \cos 2\phi_2]\} \end{aligned} \qquad (9)$$

where

$$G_\pm(k, \phi_i) = (1/d)C^2(k, \phi_i) \pm (1/L_i) \cos 2\phi_i\,.$$

We observe that $g_i(\phi_1, \phi_2; k) = 0$ for the boundary conditions (see Eq. 5).

In order to get the critical thickness below which the sample is undistorted, we consider only the situation $d\psi/dz = 0$, for any z in the range $(-d/2, d/2)$ and hence, from Eq. 2, $p = 0$. The latter condition, for Eq. 4, implies $I(\phi_1, \phi_2; k) = 0$, i.e., $\phi_1 = \phi_2$, since $C(k, \mu) > 0$, for any μ. In this case, setting $\phi^* = \phi_1 = \phi_2$, the previous Eqs. 9 assume the simple form:

$$\begin{aligned} g_i(\phi^*, \phi^*; k) &= (-1)^{i+1} \sin 2\phi^*/L_i = 0 \text{ and hence } \phi^* = 0 \text{ or } \pi/2 \\ f_{11}(\phi^*, \phi^*; k) &= 2\{(1/d)C^2(k, \phi^*) + (1/L_1) \cos 2\phi^*\} \\ H(\phi^*, \phi^*; k) &= (4/dL_1L_2)\{(L_2 - L_1)C^2(k, \phi^*) - d \cos 2\phi^*\} \cos 2\phi^* \end{aligned} \qquad (10)$$

The solution $\phi^* = 0$ means that the molecules are everywhere normal to the walls (homeotropic configuration); the other solution $\phi^* = \pi/2$ is the case with the molecules parallel to the walls (planar configuration). In both cases, this means that the sample undergoes an order transition.

To compute the critical parameter (sample thickness) governing this transition, we will analyze separately different situations.

(a) $k = 0$ (elastic isotropy).[25] Since $C(0, \phi) = 1$, from Eqs. 10, we get:

$$f_{11}(0, 0; 0) = 2(L_1 + d)/dL_1 > 0$$

$$H(0, 0; 0) = (4/dL_1L_2)(L_2 - L_1 - d) > 0$$

$$\text{if} \quad d < d_c^{(H)} = L_2 - L_1 \qquad (10\text{-}1)$$

and

$$f_{11}(\pi/2,\pi/2;0) = 2(L_1 - d)/dL_1 > 0 \qquad \text{if} \qquad d < L_1$$
$$H(\pi/2,\pi/2;0) = (4/dL_1L_2)(L_1 - L_2 - d) > 0$$
$$\text{if} \qquad d < d_c^{(P)} = L_1 - L_2. \qquad (10\text{-}2)$$

We see that in both cases a critical thickness for the order transition exists depending on the difference of the anchoring energies: for thicknesses $d > d_c$ the sample stays in the distorted configuration, while the configuration $\phi = 0$, for any z, is stable if $d < d_c^{(H)}$ and $\phi = \pi/2$, for any z, is stable if $d < d_c^{(P)}$.

Hence, we conclude that if $k \simeq 0$ and $w_1 \neq w_2$, a critical thickness always exists.

(b) $k = 1$ (i.e., $k_{11} \ll k_{33}$). This case can be found when a nematic-smectic A phase transitions is approached;[14] it implies that the bend deformation is impeached. Since $C(1,\phi) = \cos\phi$, Eqs. 10 give

$$f_{11}(0,0;1) = 2(L_1 + d)/dL_1 > 0$$
$$H(0,0;1) = (4/dL_1L_2)(L_2 - L_1 - d) > 0$$
$$\text{if} \qquad d < d_c^{(H)} = L_2 - L_1 \qquad (10\text{-}3)$$

and

$$f_{11}(\pi/2,\pi/2;1) = -2/L_i < 0$$
$$H(\pi/2,\pi/2;1) = -4/L_1L_2 < 0 \qquad (10\text{-}4)$$

We see that now only the homeotropic configuration can be reached by varying the cell thickness, whereas the planar configuration is forbidden, because the free energy is not minimized in this case.

Then only if $w_1 > w_2$ (homeotropic surface energy greater than the planar one) do we get a critical thickness, while in the opposite case the nematic liquid crystal is always distorted.

(c) In the general case ($k_{11}/k_{33} < 1$), the most common case,[7,13] we note that $C(k,0) = 1$ and $C(k,\pi/2) = 1 - k$, so Eqs. 10 for $\phi = 0$ give again Eq. (10-3), while for $\phi = \pi/2$ we obtain

$$f_{11}(\pi/2,\pi/2;k) = 2\{(1 - k)/d - 1/L_1\}$$
$$H(\pi/2,\pi/2;k) = -(4/dL_1L_2)\{(1 - k)(L_2 - L_1) + d\} \qquad (10\text{-}5)$$

Then the planar configuration is a stable configuration only if $d < d_c^{(P)} = \tilde{L}_1 - \tilde{L}_2$ where $\tilde{L}_i = k_{11}/w_i$.

We observe that, as may be expected, the critical thicknesses for homeotropic and planar configurations are related to only one elastic constant.

This result is analogous to the Fréedericsz transition in a homeotropic or planar nematic cell: the order transition from the uniform orientation to the distorted orientation depends upon just one elastic constant.[26] These results are in agreement with those reported in Ref. 3.

The case $k_{11} \geq k_{33}$

Now let us analyze the situation in which the splay elastic constant is bigger than the bend constant (this could be found with flexible molecules).[16] We set $h = 1 - (k_{33}/k_{11})$, $(0 \leq h \leq 1)$, and minimize the total free energy defined as

$$F(\phi) = (k_{11}/2)\left\{\int_{-d/2}^{d/2} C^2(h,\phi)(d\phi/dz)^2\,dz + \tilde{L}_1^{-1}\sin^2\phi_1 + \tilde{L}_2^{-1}\cos^2\phi_2\right\}$$

We then get

$$C^2(h,\phi)(d\phi/dz)^2 = q^2, \tag{11}$$

for any z falling in the range $(-d/2, d/2)$ and

$$2\tilde{L}_i C^2(h,\phi_i)(d\phi/dz)_i = \sin 2\phi_i, \qquad \text{for} \qquad z = \pm d/2, \tag{12}$$

where $C^2(h,\phi) = 1 - h\cos^2\phi > 0$ and q^2 is an integration constant.

We perform the calculations as in the previous case and obtain:

$$g_i(\phi^*,\phi^*;h) = (-1)^{i+1}\sin 2\phi^*/\tilde{L}_i = 0$$

$$\text{and hence} \quad \phi^* = 0 \quad \text{or} \quad \pi/2$$

$$f_{11}(\phi^*,\phi^*;h) = 2\{(1/d)C^2(h,\phi^*) + (1/\tilde{L}_1)\cos 2\phi^*\}$$

$$H(\phi^*,\phi^*;h) = (4/d\tilde{L}_1\tilde{L}_2)\{(\tilde{L}_2 - \tilde{L}_1)C^2(h,\phi^*) - d\cos 2\phi^*\}\cos 2\phi^* \tag{13}$$

The case $h = 0$ is the same as that in the previous section with $k = 0$. For $h = 1$ (i.e., $k_{11} \gg k_{33}$) $C(1,\phi) = \sin\phi$, and then,

$$\begin{aligned} f_{11}(0,0;1) &= 2/\tilde{L}_1 > 0 \\ H(0,0;1) &= -4/\tilde{L}_1\tilde{L}_2 < 0 \end{aligned} \tag{13-1}$$

and

$$f_{11}(\pi/2,\pi/2;1) = 2(\tilde{L}_1 - d)/d\tilde{L}_1 > 0 \qquad \text{if} \qquad d < \tilde{L}_1 \tag{13-2}$$

$$H(\pi/2,\pi/2;1) = (4/d\tilde{L}_1\tilde{L}_2)(\tilde{L}_1 - \tilde{L}_2 - d) > 0$$

$$\text{if} \qquad d < d_c^{(P)} = \tilde{L}_1 - \tilde{L}_2 \tag{13-2}$$

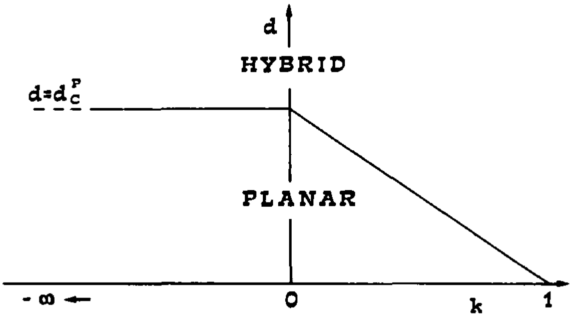

FIGURE 1 Sketch of the stability of homeotropic configurations, in the d-k plane, where d is the sample thickness and k the elastic anisotropy ($k = 1 - k_{11}/k_{33}$) in the case $w_1 > w_2$. The full line represents the critical thickness below which the sample assumes the undistorted configuration.

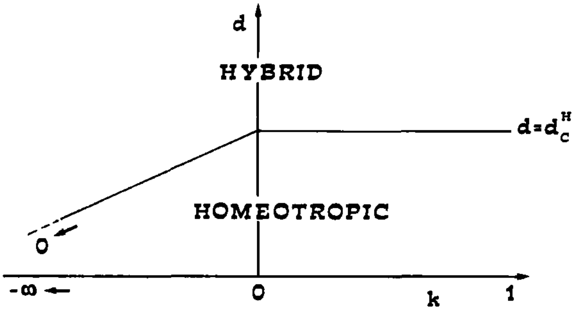

FIGURE 2 The same as Figure 1, but here is $w_1 < w_2$.

We can consider this case as the symmetric case of $k = 1$: if $h \rightarrow 1$ and $\bar{L}_1 > \bar{L}_2$ there exists a critical thickness below which the sample is planar, while if $\bar{L}_1 < \bar{L}_2$, d_c does not exist. Here also the transition depends just upon one elastic constant: now the homeotropic configuration is never attained, because the splay deformation is forbidden.

CONCLUSION

We have shown that:

(a) if $k_{11} \simeq k_{33}$ and $L_1 \neq L_2$, a critical thickness d_c always exists such that for $d < d_c$ the sample is undistorted. In the case $L_1 < L_2$ and $d < d_c^{(H)}$ the liquid crystal is entirely homeotropic, while if $L_1 > L_2$ and $d < d_c^{(P)}$ it is planar;

(b) if $k_{11} \leqslant k_{33}$ and $L_1 \neq L_2$ the critical thickness exists only when $L_2 > L_1$. In this situation, for $d < d_c^{(H)}$ the liquid crystal assumes the homeotropic configuration. In the opposite case ($L_2 < L_1$) the sample is distorted for any cell thickness. Conversely if $k_{11} \geqslant k_{33}$ and $L_1 \neq L_2$ the critical thickness is defined if $\tilde{L}_2 < \tilde{L}_1$; for $d < d_c$ the sample is planar. If $\tilde{L}_2 > \tilde{L}_1$, then $d\phi/dz \neq 0$ for any sample thickness.

In conclusion, we point out what is the main result of our calculation. When a liquid crystal cell is prepared with a different orientation at the walls, different final configurations can be achieved depending upon the sample thickness and the elastic constant values. In a thickness-elastic anisotropy diagram, a critical thickness line can be defined over which the cell has a hybrid configuration, and under which only a uniform (planar or homeotropic) configuration is possible (see Figure 1 and 2).

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