Surface induced nematic ordering and the localization of a twisted distortion in a nematic cell

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A Landau-de Gennes phenomenological model is used to analyze the coupling between the surface-induced scalar order parameter variation and the orientational ordering in a nematic slab, considering also non-planar distortions where the twist angle (ω) can vary. The results show that a weak variation $\delta\omega$ additional to that predicted by the Frank elastic theory for a pure twist deformation can appear and that it can be either globally spread over the whole sample or localized to a subsurface region of thickness comparable to the nematic correlation length. In the former case $\delta\omega$ results from a global tilt angle variation, while in the latter it is caused by a variation of the scalar order parameter (S) close to the surface. In contrast to the recently analysed S variation-induced subsurface deformation of the tilt angle [see, e.g., Phys. Rev. E **57**, 1780 (1998)], an intrinsic (chiral nematic) or confinement-induced global twist is needed to yield the localized $\delta\omega$ effect.

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In the bulk of a nematic liquid crystal sample the degree of molecular ordering is given by the scalar order parameter S and depends (beside on material constants of the liquid crystal) on temperature. If the nematic sample is confined, e.g., by a solid substrate, the value of S at the interface (S_0) can be different from its bulk value (S_b), and a variation of S occurs in a layer whose thickness is of the order of the nematic correlation length ξ [1]. In the framework of the phenomenological Landau-de Gennes theory such a variation of S can give rise to a subsurface deformation of the nematic director field if the splay and bend elastic constants are different from the twist one, as it has been considered by several authors [2–5]. In all these analyses only planar elastic distortions of the nematic have been dealt with, thereby neglecting the twist distortion, and the subsurface deformation was present in the tilt angle profile. The aim of this paper is to extend the analysis performed in Ref. [5] for the strong anchoring limit to non-planar distortions. In particular, we would like to examine whether a localized subsurface twist deformation similar to that reported for the tilt angle in, e.g., [5] can be observed, and, further, to explore also the global (delocalized) coupling between the tilt and the twist angle in a twisted nematic slab. Analyses allowing also non-planar distortions have been performed in Refs. [6,7], but were used to investigate substrate-induced orientational phase transitions. Indication for strong localized variation in molecular orientation that can be explained in terms of a variation of S [2,3], or both of S and biaxiality [8], has been found also experimentally. It should be mentioned that questions about subsurface director deformations were first raised within the second order elastic theory and that these deformations have been predicted later also by several pseudomolecular models (for a brief review see, e.g., Ref. [5] and the references therein), but no effects of the order variation were taken into account. Recently, an analysis allowing also nematic density variations has been performed [9].

Let us consider a nematic slab of thickness d where the z-axis is oriented along the surface normal and the surfaces are lying at $z = \pm d/2$, being parallel to the xy-plane. The nematic director can then be parametrized as $\vec{n} = \vec{n}(z) = [\sin\phi(z)\cos\omega(z), \sin\phi(z)\sin\omega(z), \cos\phi(z)]$, $\phi(z)$ being the angle between $\vec{n}$ and the z-axis (the tilt angle) and $\omega(z)$ the azimuthal angle defined with respect to the x-axis (the twist angle). In the framework of the Landau-de Gennes phenomenological theory the free energy density expansion up to the second order in first director component and scalar order parameter derivatives is given (neglecting biaxiality) by

$$f = f_0(S) + f_1(\phi, S') + f_2(\phi, \phi', \omega', S) + f_3(\phi, \phi', S, S'), \tag{1}$$

where the symbol $'$ denotes a derivative with respect to z. Here

$$f_0(S) = \frac{1}{2}a(T - T^*)S^2 - \frac{1}{3}BS^3 + \frac{1}{4}CS^4 \tag{2}$$

is the free energy density of the unperturbed uniform nematic ($a > 0$, $B > 0$, $C > 0$, and T^* are material constants) and

$$f_1(\phi, S') = \frac{3}{4}L_1\left\{1 + \frac{L_2}{2L_1}\left(\cos^2\phi + \frac{1}{3}\right)\right\}S'^2 \tag{3}$$

is the source of "polar" intrinsic anchoring [5], the constants $L_1 > 0$ and L_2 being related to the elastic constants; $L_2 = 0$ represents the one-constant approximation in which the splay and bend elastic constants are equal to the twist constant [1,5]. In fact L_2 is a sum of two phenomenological parameters [5] but since in a planar geometry only their sum appears in the free energy we decided to reduce it to a single parameter. Further,

$$f_2(\phi, \phi', \omega', S) = \frac{9}{4} L_1 S^2 \left\{ \left(1 + \frac{L_2}{2L_1}\right) \phi'^2 + \sin^2\phi \left(1 + \frac{L_2}{2L_1}\cos^2\phi\right) \omega'^2 \right\} \tag{4}$$

is the classic Frank elastic term and

$$f_3(\phi, \phi', S, S') = -\frac{3}{8} L_2 \sin(2\phi)\ \phi' S S' \tag{5}$$

gives rise to the S variation-induced subsurface deformation of the tilt angle ϕ [5]. Note that the free energy density depends on ω', appearing in the Frank term (f_2), but not on ω itself. If we considered chiral nematics which form a structure twisted spontaneously already in the unperturbed ground state, the only changes in the free energy density appear in the Frank term (4). Then, apart from an ω-independent term, a term linear in ω' has to be added, and the f_2 contribution to the free energy density is still ω-independent. Considering chiral nematics would not change the analysis substantially, therefore it will be restricted only to nonchiral nematics where the twist is imposed by confining surfaces.

In the strong anchoring case the total free energy per unit surface is given by

$$F = \int_{-d/2}^{d/2} f[\phi(z), \phi'(z), \omega'(z), S(z), S'(z)]\ dz. \tag{6}$$

Minimizing (6) with respect to $S(z)$, $\phi(z)$, and $\omega(z)$ yields the following Euler-Lagrange equations (ELE)

$$\frac{\partial f}{\partial S} - \frac{d}{dz}\frac{\partial f}{\partial S'} = 0, \qquad \frac{\partial f}{\partial \phi} - \frac{d}{dz}\frac{\partial f}{\partial \phi'} = 0, \qquad \frac{\partial f}{\partial \omega} - \frac{d}{dz}\frac{\partial f}{\partial \omega'} = 0. \tag{7}$$

Since in f there is no explicit dependence on ω (also for chiral nematics), the last of the above ELE can be rewritten as

$$\frac{\partial f}{\partial \omega'} = \frac{\partial f_2}{\partial \omega'} = \alpha = \text{const.} \tag{8}$$

Taking into account Eq. (4) we find that

$$\alpha = \frac{9}{2} S^2 \sin^2\phi \left\{ L_1 + \frac{L_2}{2}\cos^2\phi \right\} \omega', \tag{9}$$

which, once integrated with respect to z over the whole slab, results in

$$\omega(d/2) - \omega(-d/2) = \alpha \int_{-d/2}^{d/2} \frac{dz}{\frac{9}{2} S^2 (L_1 + \frac{L_2}{2}\cos^2\phi)\sin^2\phi}. \tag{10}$$

If we choose $\omega(d/2) = \omega(-d/2)$, it follows that $\alpha = 0$ must hold and from (9) also that $\omega' = 0$ everywhere in the sample. Hence in this case the twist deformation is absent and the director is lying in a plane (the problem is degenerate with respect to ω). This case has been examined in Ref. [5] in detail to investigate the coupling between the tilt angle ϕ and the scalar order parameter S. The results show that if there is a variation of S which is localized to a thin subsurface layer of thickness $\sim \xi$, it is accompanied by a variation of ϕ localized to a layer of the same thickness. If, however, $\omega(d/2) \neq \omega(-d/2)$, $\alpha = 0$ no longer holds and the twist deformation is present. For such a twisted nematic slab the ELE (7) must be solved numerically. As weak anchoring would only reduce the deformation, we here concentrate our attention to the strong anchoring boundary conditions, where the actual surface values of S, ϕ, and ω cannot deviate from those imposed by the substrate.

Let us first consider S-profiles in a twisted nematic sample. The scalar order parameter always relaxes monotonously from the surface value S_0 to the bulk value S_b which can be calculated by minimizing the free energy given by f_0 (2) alone. The variation occurs over a distance characterized by the nematic correlation length $\xi = \sqrt{L_1/a(T - T^*)}$ [1] (see Fig. 1) and is hence localized to a thin subsurface layer. For our choice of a, L_1, and $T - T^*$ its thickness is about 20 nm. S-profiles in presence of the twist deformation ($\alpha \neq 0$) are very similar to those reported in Ref. [5], where the twist deformation was absent ($\alpha = 0$). In general, any variation of ϕ or ω affects S-profiles only very weakly since the free energy contributions associated with the elastic deformation (f_1, f_2, and f_3) are considerably smaller than the homogeneous one (f_0).

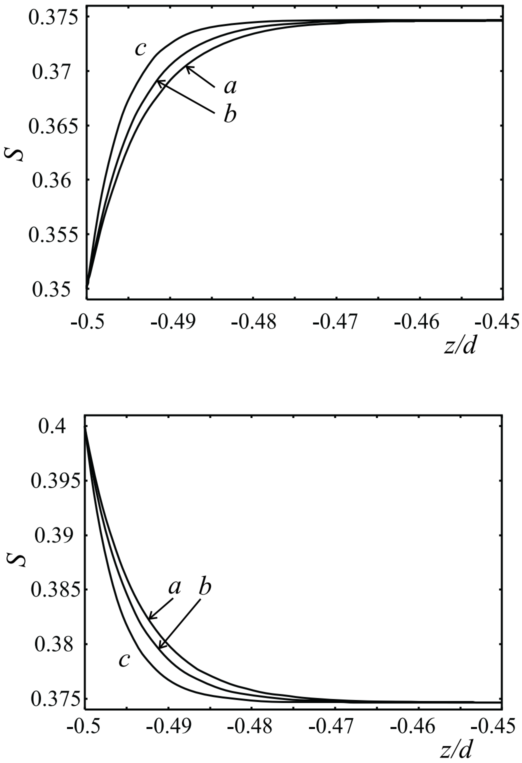


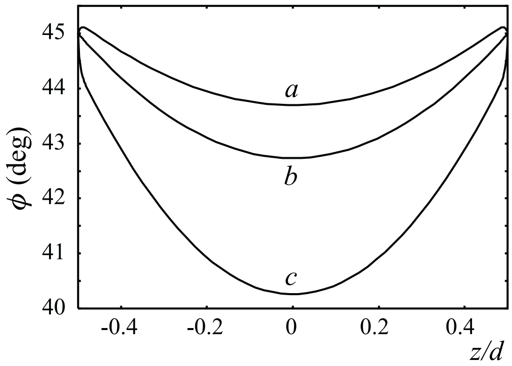
FIG. 1. $S(z)$-profiles in a twisted nematic slab with strong anchoring. $\phi(\pm d/2) = 45°$, $\omega(-d/2) = 0°$, $\omega(d/2) = 45°$, $S_b \approx 0.375$, $S_0 = 0.35$ (top figure) or $S_0 = 0.4$ (bottom figure), and $L_2 = L_1,\ 0,\ -L_1$ [curves (a), (b), and (c), respectively]. The sample thickness equals to 1 μm, $a = 0.13 \times 10^6$ J/m^{3}K, $B = 1.6 \times 10^6$ J/m^3, $C = 3.9 \times 10^6$ J/m^3, $T - T^* = 0.4$ K, and $L_1 = 10^{-11}$ N.

On the other hand, in a twisted nematic slab there is quite a significant change in tilt angle profiles $\phi(z)$ in comparison to the non-twisted case. These profiles show a considerable variation of ϕ which spreads over the whole slab (see Fig. 2) even in the symmetric anchoring case in which $\phi(-d/2) = \phi(d/2)$. The source of this delocalized deformation is the coupling between ω' and ϕ in the Frank elastic term f_2 (4). Namely, since the ω'^2-term, which is nonzero when the twist deformation is present, always gives a positive free energy contribution, the proportionality factor appearing in front of ω'^2 must be as low as possible, i.e., for $L_2 = 0, \pm L_1$ (as chosen for profiles plotted in Fig. 2) $|\phi|$ must decrease. However, this decrease is compensated by the ϕ'^2 term which is also present in f_2 and gives a positive free energy contribution as soon as ϕ varies. Note that because the ratio of the proportionality constants belonging to ω'^2 and ϕ'^2, respectively, is larger when $L_2 < 0$, in that case the decrease in $|\phi|$ can be larger than for $L_2 = 0$ (just the opposite holds for $L_2 > 0$). Further, it should be noticed that for $L_2 \neq 0$ the coupling between ϕ' and S' described by the f_3-term yields the localized S variation-induced subsurface deformation in $\phi(z)$ which behaves similarly as in the non-twisted case studied in Ref. [5].

Let us finally consider also $\omega(z)$-profiles. These can be calculated by integrating Eq. (9) with respect to z. For simplicity, suppose first S and ϕ to be constant throughout the sample. The resulting profile is then a linear function of z, i.e.,

$$\omega(z) = \omega(-d/2) + \Delta\omega \left(\frac{z}{d} + \frac{1}{2} \right), \qquad (11)$$

where $\Delta\omega = \omega(d/2) - \omega(-d/2)$ is proportional to the constant α introduced in (8). If, however, either ϕ (with S=konst.; Frank solution) or both ϕ and S are allowed to vary with z, deviations $\delta\omega(z)$ from the linear profile given by (11) may occur. For example, global variations of ϕ appearing, e.g., when $\phi(-d/2) \neq \phi(d/2)$, or even in a symmetric case where ϕ varies due to the twist deformation, give rise to deviations $\delta\omega(z)$ that are global as well (see Fig. 3).



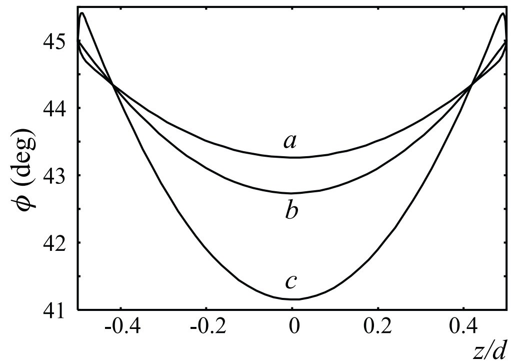


FIG. 2. $\phi(z)$-profiles in a twisted nematic slab with strong anchoring. Same parameters as in Fig. 1 have been used. $S_0 = 0.35$ (top figure) and $S_0 = 0.4$ (bottom figure).

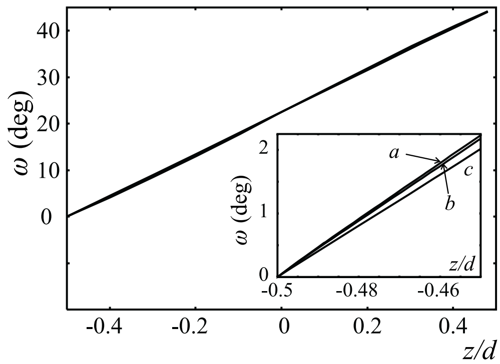


FIG. 3. $\omega(z)$-profiles in a twisted and strongly anchored nematic slab with a delocalized variation of ϕ. $S_0 = 0.4$, while all other parameters are same as in Fig. 1. In the inset $\omega(z)$ in the subsurface region is shown. The deviation $\delta\omega$ from the linear solution (11) spreads through the whole sample. Profiles for $S_0 = 0.35$ differ only negligibly from those plotted here.

On the contrary, a localized variation of $S(z)$ induces a variation of ω that is localized. Let us now explore this "subsurface deformation" in ω in more detail and compare it to the one appearing in the $\phi(z)$ profile. Let us, for sake of clarity, omit the $\phi(z)$-dependence by setting $\phi(d/2) = \phi(-d/2) = 90°$. In this case there is no source of the subsurface deformation in ϕ since the f_3-term (5) vanishes. Numerical solutions of the ELE (7) confirm indeed that then $\phi(z) = 90° =$ const. holds throughout the whole sample. Putting now $\phi(z) = 90°$ into (9) yields

$$\omega' = \frac{2\alpha}{9L_1 S^2(z)}. \tag{12}$$

Assume for the moment that the $S(z)$-profile is modeled by

$$S(z) = S_b - \Delta S \frac{\cosh(z/\lambda)}{\cosh(d/2\lambda)}, \tag{13}$$

representing the localized variation of S close to the confining walls with an amplitude $\Delta S = S_b - S_0$ and a characteristic length λ which is to be characterized by the nematic correlation length. The integration of (12) is particularly simplified if we further assume $|\Delta S| \ll S_b, S_0$. The resulting $\omega(z)$-profile is then given approximately by

$$\omega(z) \approx \omega(-d/2) + \frac{\Delta\omega}{2} + \Delta\omega \frac{z + 2\lambda \frac{\Delta S}{S_b} \frac{\sinh(z/\lambda)}{\cosh(d/2\lambda)}}{d + 4\lambda \frac{\Delta S}{S_b} \tanh(d/2\lambda)}. \tag{14}$$

The ratio $\sinh(z/\lambda)/\cosh(d/2\lambda)$ appearing in (14) is nonzero only in the boundary layers and hence represents a localized subsurface variation of the twist angle, whose amplitude equals to

$$\delta\omega_0 = \frac{2\lambda \frac{\Delta S}{S_b}}{d + 4\lambda \frac{\Delta S}{S_b} \tanh(d/2\lambda)} \Delta\omega \approx 2\frac{\lambda}{d}\frac{\Delta S}{S_b}\Delta\omega, \tag{15}$$

taking into account $\lambda \ll d$ in the end. The approximate $\omega(z)$ profile can be close to the substrates ($z \to \pm d/2$) simplified to $\omega(z) \approx \omega(-d/2) + \Delta\omega(z/d + 1/2) \mp \delta\omega_0\{1 - \exp[(\pm z - d/2)/\lambda]\}$.

This simple calculation proves the existence of a localized variation also in the ω-profile. Its amplitude is rather small with respect to the overall variation of ω, e.g., for $\Delta\omega = 45°$, $\lambda \approx 0.01d$ ($d = 1\mu$m), $\Delta S \approx 0.025$, and $S_b \approx 0.375$ it is $\delta\omega_0 \approx 0.06°$. For comparison, the actual ω-profile in the slab geometry can be derived also from solving the ELE (7) for $\phi(\pm d/2) = 90°$ and the above parameters (see Fig. 4). The $\omega(z)$ dependence shows a localized deformation $\delta\omega(z)$ that is added to the linear solution (11) with $\Delta S = 0$, exhibiting a functional dependence similar to that predicted analytically by Eq. (14). The amplitude of $\delta\omega(z)$, $\delta\omega_0$, is of the same order of magnitude as estimated above. However, the $\delta\omega$ effect could become easily measurable considering a very thin highly twisted nematic cell ($d \sim 100$ nm, $\Delta\omega \sim 2\pi$) using strongly ordering substrates ($|\Delta S| \sim 0.1$) and adjusting the temperature close to the NI transition, where $\lambda \sim 10$ nm.

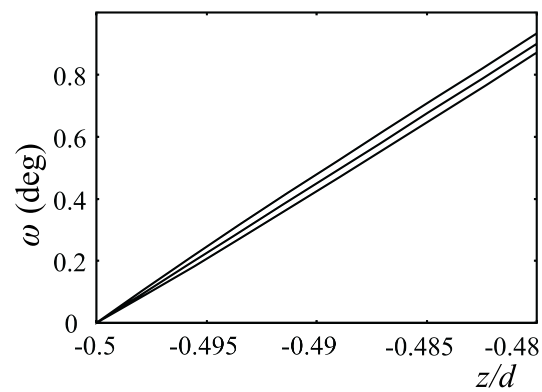


FIG. 4. $\omega(z)$-profiles in the subsurface region of a twisted nematic slab with a localized variation of S; $\phi(\pm d/2) = 90°$, $S_b \approx 0.375$, $\Delta S \approx 0.025$, 0, -0.025 (top, middle, and the bottom curve, respectively), $\omega(-d/2) = 0°$, $\omega(d/2) = 45°$; $L_2 = 0$ [for $\phi(\pm d/2) = 90°$ the problem is degenerate with respect to the value of L_2]. The profiles cross in the middle of the sample and exhibit a similar behavior at the opposite side, the top curve, e.g., now corresponding to the negative value of ΔS. The values for a, B, C, $T - T^*$, and L_1 are same as in Fig. 1.

Note again that the source of the subsurface variation of the twist angle $\delta\omega(z)$ is the Frank elastic term (f_2) and that the origin of this ω variation is different from that responsible for the variation of ϕ in Ref. [5] (in that case the source was the f_3 term). Both subsurface ϕ and ω variations follow from a localized variation of S, but the latter can exist only if a global distortion in ω is already present (since $\delta\omega \propto \Delta\omega$), while for the former no deformation in ϕ is necessary to assure its existence.

Further, note that in the Frank term (f_2) there is also a coupling between S and ϕ' which is mathematically equivalent to that between S and ω'. In samples where a delocalized deformation in ϕ is already present, e.g., in non-symmetric samples with $\phi(-d/2) \neq \phi(d/2)$, this can, in principle, induce an additional localized variation in ϕ, which has to be added to the one already analyzed in Ref. [5], where this kind of deformation was not discussed since only symmetric samples have been considered.

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