Nanometric pitch in modulated structures of twist-bend nematic liquid crystals

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Abstract

The extended Frank elastic energy density is used to investigate the existence of a stable periodically modulate structure that appears as a ground state exhibiting a twist-bend molecular arrangement. For an unbounded sample, we show that the twist-bend nematic phase $N_{TB}$ is characterized by a heliconical structure with a pitch in the nano-metric range, in agreement with experimental results. For a sample of finite thickness, we show that the wave vector of the stable periodic structure depends not only on the elastic parameters but also on the anchoring energy, easy axis direction, and the thickness of the sample.

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I. INTRODUCTION

Modulated materials are remarkable examples of how structure defines macroscopic properties in condensed matter. From naturally occurring phononic activity in crystals to synthetic superconductor superlattices [1], modulated structures are key elements and fine tailored in cutting edge applications. They occur naturally in calamitic liquid crystals such as layered arrangements (in smectics), helical organization in chiral nematics or even defect lattice structures in blue phase materials [2]. Such organization defines, for example, exciting photonic properties like the ones observed in chiral nematic and blue phases. These are not, however, the only possible modulated phases in liquid crystals. Newly synthesized materials open up possibilities for new structures quite often in the liquid crystals field. A new class of materials has been recently discovered in the liquid crystals (LC) field to show molecular organizations that permits one to characterize them as new orientational phases. The twist-bend nematic (NTB) is one of these phases. It has a double degenerate ground-state with a periodically modulated heliconical structure exhibiting a nanoscale pitch favoured by achiral bent-shaped molecules. This phase has been experimentally observed very recently in bent molecular dimers [3, 5, 6], trimers [7] and, more recently yet, in rigid bent-core materials [8] and in chiral dimers as well [9]. In a very short time, such materials have already found applications, as reported in [10, 11]. It has been theoretically predicted years ago [12–14] together with a different orientational, modulated structure designed as splay-bend phase [13]. From the theoretical point of view, besides the initial predictions [13, 14], it has been argued [15] that flexoelectricity could be the driving force for the formation of the modulated phase. Nonetheless, recent studies indicate that the flexoelectric model cannot explain the large compression modulus of the NTB phase and that the driving force for the NTB phase comes from steric interactions [16].

One remarkable point regarding the NTB phase is the fact that it presents, in the ground state, spontaneous twist and bend deformations, exhibiting an oblique helicoidal structure with a nanometric pitch, which is directly related to a bend elastic constant $K_{33}$ much smaller than both $K_{22}$ (twist) and $K_{11}$ (splay). However, such ratios of the elastic constants have not been detected experimentally in the ground state region. To date, experimental investigations of elastic properties in the NTB phase have been reported by using the extrapolation technique to extract $K_{33}$, $K_{22}$ and $K_{11}$ elastic constants of those materials in the
region close to the phase transition. Thus, the approach to explore these elastic constants is still based on the standard elastic energy density, obtaining the threshold of the Fréedericksz transitions in splay, bend and twist geometries [17].

In this paper, we use a recently proposed elastic model [18, 19] to investigate the possibility of a ground state of periodic modulations exhibiting heliconical orientational ordering. For an infinite sample, we show that the pitch of the modulated structure may be in the nanometric range, as experimentally found. For a finite-length situation, when the interaction between the surface and the twist deformation is negligible, by means of the transversality condition, we demonstrate that the mechanical torque transmitted to the surface in equilibrium vanishes. In this case the wave vector of the periodic modulation coincides with the one found for the infinite sample. In the presence of an anchoring energy at the surface, we show that this wave vector depends on surface properties like easy axis angle and anchoring strength as well as on the bulk properties like the elastic parameters, cone angle, and the thickness of the sample, a behavior expected in heliconical structures, as observed in cholesterics [2, 20]. The paper is organized as follows: In Secs. II and III the elastic model for the nematic twist-bend is revisited in order to show that the conditions for the stability of the phase supports the existence of a pitch in the nanometric range, as experimentally verified. In Sec. IV the analysis focuses on a nematic twist-bend phase confined to a sample of finite thickness to explore the surface effects on the wave vector of the modulated structure. Some concluding remarks are drawn in Sec. V.

II. THE ELASTIC MODEL

The general elastic free energy is characterized by a director $n$ and a helix axis $t$ related to the chiral twisted collective arrangement [18]. One assumes furthermore no polar order, in such a manner that $n$ is the usual nematic director. As shown previously [18, 21], the free energy density compatible with the symmetry of the phase, in absence of an external field, may be written as

$$f = f_0 - \frac{1}{2} \eta (n \cdot t)^2 + \kappa_1 t \cdot [n \times (\nabla \times n)] + \kappa_2 n \cdot (\nabla \times n) + \kappa_3 (n \cdot t)(\nabla \cdot n) + \frac{1}{2} K_{11} (\nabla \cdot n)^2$$
$$+ \frac{1}{2} K_{22} [n \cdot (\nabla \times n)]^2 + \frac{1}{2} K_{33} (n \times \nabla \times n)^2 - (K_{22} + K_{24}) \nabla \cdot (n \nabla \cdot n + n \times \nabla \times n)$$
\[ + \mu_1[t \cdot (n \times \nabla \times n)]^2 + \nu_1[t \cdot \nabla(t \cdot n)]^2 + \nu_2[t \cdot \nabla(n \cdot t)(\nabla \cdot n)] + \nu_3[\nabla(t \cdot n)]^2 + \nu_4[(t \cdot \nabla)n]^2 \\
+ \nu_5[\nabla(n \cdot t) \cdot (t \cdot \nabla)n] + \nu_6[\nabla \cdot (\nabla \times n)]. \]  

(1)

In Eq. (1), \( \kappa_i \), for \( i = 1, 2, 3 \) are the elastic parameters connected with the spontaneous splay, twist and bend, respectively, while \( K_{11}, K_{22}, K_{33} \) and \( K_{24} \) are the Frank elastic constants. The new elastic parameters are \( \eta \), which is a measure of the coupling strength between \( n \) and \( t \), \( \mu_1 \) and \( \nu_i \), for \( i = 1, 2, \ldots, 6 \).

In the N_{TB} phase, the director \( n \) forms an oblique helicoid around the direction \( t \), with a constant tilt angle \( \theta_0 \) fixed by molecular interaction forces. By taking \( t = u_z \), the nematic director in this case is

\[ n = [\cos \phi(z) u_x + \sin \phi(z) u_y] \sin \theta_0 + \cos \theta_0 u_z. \]  

(2)

As shown in Ref. [18], this director configuration simplifies the problem in such a way that only the following terms will contribute to energy density, namely:

\[ f_d = f_1 - \frac{1}{2} \eta(n \cdot t)^2 + \frac{1}{2} K_{22}[n \cdot (\nabla \times n) + q_0]^2 \\
\quad + \frac{1}{2} K_{33}(n \times \nabla \times n)^2 + \nu_4[t \times (\nabla \times n)]^2, \]  

(3)

where \( f_1 = f_0 - (1/2) K_{22}q_0^2 \) is a constant, and \( \nu_4[(t \cdot \nabla)n]^2 = \nu_4[t \times (\nabla \times n)]^2 \), because \( (t \cdot \nabla)n = \nabla(n \cdot t) - (t \times \nabla \times n) \), and \( n \cdot t = \cos \theta_0 \), which is constant. Furthermore, in Eq. (3), \( q_0 = \kappa_2/K_{22} \) is the wave vector of the cholesteric phase. The free energy density per unit area corresponding to (2) becomes:

\[ f(\phi', x) = f_1 - \frac{1}{2} \eta (1 - x) + \frac{1}{2} K_{22}(\phi'x - q_0)^2 \\
\quad - \frac{1}{2} K_{33}\phi'^2(x^2 - xb_0), \]  

(4)

in which \( x = \sin^2 \theta_0 \), \( b_0 = (1 + 2\nu_4/K_{33}) \), and \( \phi' = d\phi(z)/dz \). In a perfectly aligned nematic phase, \( x = 0 \) and therefore no bend distortion occurs. In the cholesteric phase, \( x = 1 \), and the medium will have no extra distortion only if \( b_0 = 1 \). On the other hand if \( b_0 \neq 1 \), for \( x = 1 \), and \( \phi = qz \), the free energy density (1) becomes

\[ f = f_1 + \frac{1}{2} K_{22}(q - q_0)^2 + \nu_4 q^2, \]  

(5)
from which one derives that the wave vector, of the stable periodic structure, is renormalized by the presence of $\nu_4$, in such a way that

$$q = \frac{q_0 K_{22}}{K_{22} + 2 \nu_4}. \tag{6}$$

This corresponds to a stable structure only if $K_{22} + 2 \nu_4 > 0$. The energy density Eq. (1) describes the cholesteric phase for $\nu_4 = 0$. Thus $\lambda_C = \pi / q_0$ is the cholesteric pitch which is usually micrometric. It can be in the micrometric range even when $x \neq 1$, since a conical state is allowed for a small $K_{33}$ elastic constant [4, 10].

III. UNLIMITED SAMPLE: NANOMETRIC PITCH

In order to analyze the stability of the spontaneous periodic deformation in the N$_{TB}$ phase, we consider first an unlimited sample. By taking into account that there is a periodicity along the helix axis of the N$_{TB}$ phase, we may obtain $\phi(z)$ that minimizes the energy of one period [22], that means, the functional

$$\frac{F(\lambda)}{S} = \int_0^\lambda dz \left[ \frac{\gamma}{2} - K_{22} q_0 x \phi' + \frac{1}{2} \alpha \phi'^2 \right], \tag{7}$$

where $S$ is the area of the system in the $(x, y)$ plane, $\gamma = 2 f_1 - \eta (1 - x) + K_{22} q_0^2$, and $\alpha = (K_{22} - K_{33}) x^2 + K_{33} x b_0$ is an effective elastic constant. In (7), $\lambda$ is the wavelength of the periodic structure. The corresponding Euler-Lagrange equation permits to obtain a first integral in the form

$$-K_{22} q_0 x + \alpha \phi' = C, \tag{8}$$

where $C$ is an integration constant. For simplicity, we introduce the average energy density per period, $g$, that, by taking into account Eqs. (7) and (8), after some algebra, assumes the following form

$$g(C) = \frac{F(\lambda)}{S \lambda} = \frac{\gamma}{2} - \frac{1}{2 \alpha} (K_{22} q_0 x)^2 + \frac{1}{2} \frac{C^2}{\alpha}. \tag{9}$$

Minimization of $g$ with respect to $C$ yields $C = 0$. This result indicates that the mechanical torque present in the sample vanishes identically, i.e., $\partial f / \partial \phi' = 0$. The solution is of the form $\phi(z) = q z$, where
\[
\phi'(z) = \frac{K_{22}q_0x}{\alpha} = q_B.
\]

In this case, Eq. (10) assumes the form:

\[
f(q, x) = f_1 - \frac{1}{2}q(1 - x) + \frac{1}{2}K_{22}(q x - q_0)^2
- \frac{1}{2}K_{33}q^2(x^2 - x b_0).
\]

(11)

The stable configuration of the phase may be investigated by minimizing (11) in terms of the independent parameters \(q\) and \(x\) as \((\partial f/\partial q)_{q=q^*} = 0\) and \((\partial f/\partial x)_{x=x^*} = 0\), giving [18]:

\[
q^* = \pm \frac{\sqrt{\eta}}{\sqrt{b_0K_{33}}}
\]

(12)

for the wave vector, and

\[
x^* = -\frac{b_0K_{33} + K_{22}q_0\sqrt{b_0K_{33}/\eta}}{K_{22} - K_{33}}
\]

(13)

for the cone angle of \(n\) with \(t\). By combining (12) with (13), one easily shows that the equilibrium value \(q^* = q_B\) as determined by (10). Thus, the stable periodicity coincides with the one obtained from the minimization of the free energy, and the wavelength of the periodic structure, i.e., the pitch of the twist-bend phase, is simply given by

\[
\lambda_B = \frac{\pi}{q_B} = \lambda_C \frac{\alpha}{K_{22}x}.
\]

(14)

It may be rewritten by using the definition of \(\alpha\) given above in order to obtain

\[
\frac{\lambda_B}{\lambda_C} = x \left( 1 - \frac{K_{33}}{K_{22}} \right) + \frac{K_{33} + 2\nu_4}{K_{22}}.
\]

(15)

An initial estimation of the order of magnitude of the pitch can be obtained as follows. Since \(0 < \lambda_B/\lambda_C \leq 1\) and \(x << 1\) in the NTB phase, one observes that the values of \(\nu_4\) lie approximately in the interval \(-K_{33}/2 < \nu_4 < (K_{22} - K_{33})/2\), i.e., they can be also negative for \(K_{33} > 0\). The extrapolation of elastic constant gives an indication of the value of the bend coefficient in the twist-bend nematic, \(K_{33} \approx 0.5\) pN and \(K_{22} \approx 6.5\) pN, as reported by Adlem [6]. Since \(\theta_0 \approx 15^\circ\) [23], one obtains \(x = \sin^2\theta_0 \approx 0.06\) and \(K_{33}/K_{22} \approx 0.07\). If we take \(\nu_4 \approx -K_{33}/4\), we obtain \(\lambda_B/\lambda_C \approx 0.09\). This confirms that in the range in which \(\nu_4\) is
negative, even if small in absolute value, the pitch of the \( N_{\text{TB}} \) phase may be nanometric, as it is evidenced in experimental reports on \( N_{\text{TB}} \) material.

IV. FINITE-LENGTH SITUATION

The analysis will be now focused on the situation in which the sample is limited by two flat surfaces, parallel to the \( x, y \)-plane, placed at \( z = 0 \) and \( z = d \) of a Cartesian reference frame. We assume that the value of the tilt angle at the surface is fixed and is equal to the one imposed by intermolecular forces. In the case in which the anchoring is strong on the lower surface, i.e., \( \phi(0) = 0 \), and the upper surface imposes an easy axis \( \Phi \), with a finite azimuthal anchoring energy, \( W \), the total energy of the sample is

\[
F = \int_0^d f(\phi', x) \, dz + \frac{1}{2} W [\phi(d) - \Phi]^2. \tag{16}
\]

In Eq. (16), we have approximated the surface energy with a parabolic potential. Note that it is not a \( \pi \)-periodic potential. The Euler-Lagrange equation for this system is (see Eq. (10))

\[
\alpha \phi''(z) = 0, \tag{17}
\]

whose solution is again \( \phi(z) = qz \) and, at the upper surface, it is subjected to the boundary condition [21]:

\[
\left. \frac{\partial f}{\partial \phi'}(z) \right|_{z=d} = K_{22} x [\phi'(d)x - q_0] - K_{33} \phi'(d)(x^2 - x_0) = W[\phi(d) - \Phi]. \tag{18}
\]

When no interaction between the surface and the twist deformation is present, \( W = 0 \), the transversality condition \( \partial f / \partial \phi'|_{z=d} = 0 \), i.e., the mechanical torque transmitted to the surface in the equilibrium state vanishes. Thus, the wave vector of the periodic modulation coincides with the one found for the unlimited system: \( q = q_B \).

We consider now the more realistic case of a bounded sample characterized by two easy axes in the presence of a finite anchoring energy. Let \( \varphi(z) = \phi(z) - \Phi \), such that \( \varphi(0) = -\Phi \) and \( \varphi(d) = \phi(d) - \Phi \). The free energy density (16) becomes
\[ F = f_0 d + \frac{1}{2} \alpha \int_0^d \left[ \varphi'(z)^2 - 2q_B \varphi'(z) \right] dz + \frac{1}{2} W \varphi(d)^2. \]  

(19)

The Euler-Lagrange equation has a first integral in the form (see again Eq. (10))

\[ \varphi'(z) = q_B + \delta q = q. \]  

(20)

The solution becomes \( \varphi(z) = -\Phi + qz \), such that \( \varphi(d) = qd - \Phi \). Substitution of (20) into (19) yields the simple expression

\[ F = f_0 d + \frac{1}{2} \alpha (q^2 - 2q_B q)d + \frac{1}{2} W(qd - \Phi)^2, \]  

(21)

which, after being minimized in \( q \), yields:

\[ q = \frac{q_B + (W/\alpha) \Phi}{1 + (W/\alpha) d}. \]  

(22)

This is the wave vector of the stable periodicity for a finite-length sample when the upper surface is characterized by a weak anchoring energy, \( W \), and imposes an easy axis, \( \Phi \). As it follows from Eq. (1), \( \alpha \) plays the role of an effective elastic constant for the considered distortion. Hence \( \ell = \alpha/W \) is the usual extrapolation length. From Eq. (22), in the limit of zero anchoring energy \( (d \ll \ell) \), one retrieves the unlimited sample situation. That means the wavevector of the TB-phase remains equal to \( q_B \). To go on further, we assume the following general expressions for the thickness of the sample and the easy axis, respectively:

\[ d = m\lambda_B + r\lambda_B \quad \text{and} \quad \Phi = m\pi + s\pi, \quad m = 0, 1, 2, \ldots, \]  

(23)

with \( 0 \leq r \leq 1 \) and \( 0 \leq s \leq 1 \). From Eq. (22), we get for the relative variation of the actual pitch \( q \) with respect to that in the infinite sample \( q_B \) the expression

\[ \frac{q - q_B}{q_B} = \frac{(s - r)\pi}{q_B \ell + (m + r)\pi}. \]  

(24)

The limit of an infinite sample is achieved for \( d \to \infty \), i.e., \( m \to \infty \). In this case, from Eq. (24) we deduce that \( q \to q_B \), as expected. The same limit is reobtained for \( W \to 0 \), i.e. \( \ell \to \infty \). For a finite-length sample in the limit of strong anchoring \( (W \to \infty, \text{and hence} \ell \to 0) \), we obtain
\[
\frac{q - q_B}{q_B} = \frac{s - r}{m + r}.
\]

(25)

However, since for a macroscopic sample \( m \gg 1 \), then \( q \sim q_B \). The conclusion is that for macroscopic sample the influence of the surface treatment on the actual pitch of the \( N_{TB} \) phase is small.

For infinite anchoring strength on both surfaces and parallel easy axes on them, if the thickness of the cell is not a semi-integer multiple of the pitch, one expects that the system will adapt by changing its natural wavevector to \( q = q_B + \delta q \). That means, the pitch variation \( \delta \lambda = \lambda_B/2N \), where \( N = \text{integer}[d/\lambda_B] \). If one considers an ultrathin film of some dozens of pitch thick, the expected pitch variation is of the order of a few percent of \( \lambda_B \). When the anchoring is sufficiently weak at least at one surface, in case that the thickness of the cell is not a semi-integer multiple of \( \lambda_B \), the system can accommodate an incomplete turn, that is, the wave vector could remain equal to \( q_B \).

The above analysis is similar to the problem for a cholesteric LC in a finite thickness cell with a preferred easy direction on the boundaries. In the latter case, the wavevector dependence on the anchoring and thickness of the cell, cholesteric pitch transition, and multistability have been extensively investigated \[24\,26\] under the presence of an external field. Of course our present analysis, in the case of a finite thickness TB phase, is elementary and it intends just to find an upper limit for the variation of the free wavevector \( q_B \).

V. CONCLUDING REMARKS

We have discussed twist-bend molecular organization by obtaining a link between the pitch of the \( N_{TB} \) phase and the pitch of the cholesteric phase. An estimation of the order of magnitude of this pitch was done by using the extrapolated values of the elastic parameters of the nematic twist-bend known in the literature. It shows that the elastic description we are using here predicts a stable ground state for heliconical twist-bend structure characterized by a nanometric pitch, as experimentally found. In addition, we have analyzed how the wave vector of the modulated phase depends on the thickness of the sample and also on the anchoring energy of the bounding surfaces. It is shown that it is, actually, independent of the surface treatment if the sample is macroscopic. The analysis is a further indication that this elastic model may stand as one of the pathways to enrich our understanding of
spontaneous modulation in nematic liquid crystals systems.

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