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Elastic continuum theory: Towards understanding of the twist-bend nematic phases

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The twist-bend nematic phase, N_{TB}, may be viewed as a heliconical molecular arrangement in which the director **n** precesses uniformly about an extra director field, **t**. It corresponds to a nematic ground state exhibiting nanoscale periodic modulation. To demonstrate the stability of this phase from the elastic point of view, a natural extension of the Frank elastic energy density is proposed. The elastic energy density is built in terms of the elements of symmetry of the new phase in which intervene the components of these director fields together with the usual Cartesian tensors. It is shown that the ground state corresponds to a deformed state for which $K_{22} > K_{33}$. In the framework of the model, the phase transition between the usual and the twist-bend nematic phase is of second order with a finite wave vector. The model does not require a negative K_{33} in agreement with recent experimental data that yield $K_{33} > 0$. A threshold is predicted for the molecular twist power below which no transition to a twist-bend nematic may occur.

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Conical structures in liquid crystals pose an interesting gateway for scientific challenges and industrial utilization. Its aspects have indeed been predicted, as in [1–3], and recently experiments have shown it has immediate applications [4]. The recently discovered twist-bend nematic phase, N_{TB}, has been theoretically predicted by Meyer [5] and Dozov [6], and has been experimentally evidenced by a number of studies [7–19]. By speculating that the bend elastic constant, K_{33}, may become negative, Dozov [6] employed a simple fourth-order model for the bulk free energy to predict the existence of two different periodic one-dimensional textures in the nematic phase. The first one was called splay-bend texture, characterized by a local bend that periodically changes its sign. The other one is a continuous conical twist-bend (TB) texture, in which the director **n** rotates along an axis forming a revolution cone with aperture θ. In an achiral system, this latter texture is twofold degenerated, allowing both right-hand and left-hand twists. By using techniques of small-angle x-ray scattering, modulated differential scanning calorimetry, it was recently concluded that the low-temperature mesophase of CB7CB is a new uniaxial nematic phase whose director distribution is composed of twist-bend deformations [7]. Transmission electron (TEM) and polarized optical microscopy have been used to experimentally demonstrate the existence of the TB nematic phase [19]. Structural observations confirming the existence of a TB ground state have also been carried out by TEM, together with measurements of the director cone angle and the full pitch of the director helix, indicating a strong coupling between the molecular and the director bend [8]. These experimental identifications permit one to describe this nematic order as a true liquid-crystal phase with a new type of order. It is then a twist-bend modulated phase formed by achiral molecules, in which the director follows an oblique helicoid, maintaining a constant oblique angle $0 \leqslant \theta \leqslant \pi/2$, with the helix axis [19]. If the helix axis is along z, then the director **n** may be written as $\mathbf{n} = (\sin\theta\ \cos\varphi, \sin\theta\ \sin\varphi, \cos\theta)$, where φ is the azimuthal angle, given by $\varphi = qz = (2\pi/p)z$, where q is the modulus of the wave vector and p is the nanoscale pitch of the helix. In this way, this new ground state may be faced as an intermediate structural phase between the (usual) uniaxial and the chiral nematic phases [19]. It can also be faced as a nonuniform ground state, characterized by the presence of a local spontaneous bend and a very low value of the bend elastic constant K_{33}. To account for the elastic properties of the phase, Virga [20] proposed an intrinsically quadratic elastic theory to describe N_{TB} phases with an extra director field, in which there are two variants of the helical nematic phase, as predicted in Ref. [6], with a helix axis **t**: one in which the helix winds upward and another one in which it winds downward, as shown in Fig. 1. This theory does not require a negative bend elastic constant, as suggested by the quartic theory proposed in Ref. [6] or the existence of a locally ferroelectric phase, as required by the theory proposed in Refs. [21,22], which is also quadratic but deals with an effective bend constant arising from the flexoelectric coupling. Katz and Lededev [23] propose a Landau description, which includes fluctuations, for the phase transition from the conventional nematic into the conical helical orientationally nonuniform structure formed by "banana" shaped molecules. Their main result is that fluctuations convert the phase transition to first order. Greco *et al.* [24] proposed a more molecular model by invoking a

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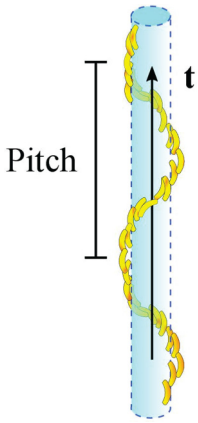

FIG. 1. (Color online) One of the variants of the helical nematic phase in which the helix can wind upwards or downwards. The vertical axis defines the direction of **t**.

generalized Maier-Saupe theory for the transition to N_{TB} phase.

In this Rapid Communication, a quadratic elastic theory also based on the existence of an extra director, represented by the helix axis **t**, as in Ref. [20], is proposed. Here, however, this extra director plays the role of an internal local field that couples with the nematic director **n**. In this sense, it not only defines the conical organization of the phase but may be understood as an internal field that originates from the bend-shaped molecules, giving rise to a local twisting power, whose strength is measured by η. Differently from what is done in Ref. [20], in which the N_{TB} is assumed as existing, the reference state used here is the undeformed nematic, characterized only by the uniform part of the free energy density. The aim is to demonstrate that a phase like the N_{TB} may be stabilized, i.e., the ground state of the system corresponds to the minimum of a quadratic elastic theory built using only the existing elements of symmetry of the phase. Our model is predicting the N_{TB} phase as a ground state, stable phase. It can also predict the N_{TB} to regular nematic phase transition. Moreover, it describes the standard cholesteric phase and, as in [20], reduces to the usual Frank's theory for ordinary nematics if the extra director vanishes.

The starting point is to consider a crystal characterized by the nematic director **n** and the director of the torsion **t** related to the chiral twisted collective arrangement. One assumes no polar order, in such a manner that **n** is the usual nematic director. The elastic energy density of a crystal of this type, f, depends on the director field **n**. According to general rules [25]

$$f(\mathbf{n}) = f_0(\mathbf{n}) + L_{ij}\, n_{i,j} + \tfrac{1}{2} K_{ijkl} n_{i,j} n_{k,l}, \ldots, \tag{1}$$

where the dots are terms of higher order in $n_{i,j} = \partial n_i/\partial x_j$, and the summation convention has been assumed. The quantity $f_0(\mathbf{n})$, independent of $n_{i,j}$, can be decomposed, at the lowest order, as

$$f_0(\mathbf{n}) = f_1 - \tfrac{1}{2}\eta(\mathbf{n}\cdot\mathbf{t})^2 + \cdots, \tag{2}$$

where f_1 is the uniform part of the energy of the usual nematic phase, if one treats the vector **t** as a field, as proposed above. In this framework, the parameter η represents the intrinsic coupling between **n** and **t** [26–28].

The tensor of second order has to be decomposed in terms of the elements of symmetry of the phase, **n** and **t**, of the identity tensor and of the antisymmetric tensor of elements δ_{ij} and ε_{ijk}, respectively. Standard calculations give

$$\begin{aligned} L_{ij} = {} & A_1 n_i n_j + A_2 n_i t_j + A_3 t_i n_j + A_4 t_i t_j + A_5 \delta_{ij} \\ & + A_6 n_k \varepsilon_{kij} + A_7 \delta_{ij} n_k t_k. \end{aligned} \tag{3}$$

Since the medium is globally nonpolar, the condition

$$f(\mathbf{n}) = f(-\mathbf{n}) \tag{4}$$

holds. It requires that L_{ij} has to be odd in **n**, which implies that $A_1 = A_4 = A_5 = 0$. Furthermore, since $|\mathbf{n}| = 1$, i.e., $n_i n_i = 1$, it follows that $n_i n_{i,j} = 0$. Consequently, the term connected to A_2 does not play any role. The elastic energy density linear in the deformation, related to the spontaneous deformation of the phase under consideration, is

$$L_{ij}\, n_{i,j} = -\kappa_1 t_i\, n_j\, n_{i,j} + \kappa_2 n_k\, \varepsilon_{kij}\, n_{i,j} + \kappa_3 \delta_{ij} n_k t_k, \tag{5}$$

where the phenomenological constants appearing in Eq. (3) have been renamed as $\kappa_1 = -A_3$, $\kappa_2 = A_6$, and $\kappa_3 = A_7$. A simple calculation shows that

$$\begin{aligned} & t_i\, n_j\, n_{i,j} = -\mathbf{t}\cdot[\mathbf{n}\times(\nabla\times\mathbf{n})] \\ \text{and} \quad & n_k\, \varepsilon_{kij} n_{i,j} = \mathbf{n}\cdot(\nabla\times\mathbf{n}), \end{aligned} \tag{6}$$

and the linear term in the deformation of the elastic energy density can be rewritten in covariant form as

$$\begin{aligned} L_{ij}\, n_{i,j} = {} & \kappa_1\, \mathbf{t}\cdot[\mathbf{n}\times(\nabla\times\mathbf{n})] + \kappa_2\, \mathbf{n}\cdot(\nabla\times\mathbf{n}) \\ & + \kappa_3 (\mathbf{n}\cdot\mathbf{t})(\nabla\cdot\mathbf{n}). \end{aligned} \tag{7}$$

One notices that while the term related to $\mathbf{n}\cdot(\nabla\times\mathbf{n})$ is a chiral term, present in standard cholesteric liquid crystals, the first term is peculiar to the heliconical phase under consideration. In the case in which $\mathbf{t} = \mathbf{u}_z$, the director field **n** may be expressed as

$$\mathbf{n} = [\cos\varphi(z)\,\mathbf{u}_x + \sin\varphi(z)\,\mathbf{u}_y]\sin\theta + \cos\theta\,\mathbf{u}_z, \tag{8}$$

where $\mathbf{u}_x, \mathbf{u}_y, \mathbf{u}_z$ are the unit vectors along the Cartesian axes. As stated before, the director field given by Eq. (8) corresponds to a precession of **n** around z with constant orientation of **n** with respect to **t**. In this case, one easily shows that

$$\begin{aligned} & \mathbf{t}\cdot[\mathbf{n}\times(\nabla\times\mathbf{n})] = 0, \quad \nabla\cdot\mathbf{n} = 0, \\ \text{and} \quad & \mathbf{n}\cdot(\nabla\times\mathbf{n}) = -q\, \sin^2\theta \end{aligned} \tag{9}$$

are constant quantities.

The fourth-rank tensor K_{ijkl} in the elastic energy (1), may be decomposed by following the standard procedure (see, e.g.,

Ref. [25]), namely,

$$K_{ijkl} = k_5 n_j n_l \delta_{ik} + k_6 \delta_{ij}\delta_{kl} + k_7 \delta_{ik}\delta jl + k_8 \delta_{il}\delta_{jk} + \mu_1 n_l n_j t_i t_k + \nu_1 t_i t_j t_k t_l + \tfrac{1}{2}\nu_2(t_i t_j \delta_{kl} + t_k t_l \delta_{ij}) + \nu_3 t_i t_k \delta_{jl} + \tfrac{1}{2}\nu_4(t_i t_l \delta_{jk} + t_j t_k \delta_{il}) + \nu_5 t_j t_l \delta_{ik} + \nu_6 t_i \varepsilon_{jkl}. \quad (10)$$

The terms involving k_i yield the usual Frank contribution. The other terms are also decomposed in the usual manner and the total elastic energy density may be written as

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

For the physical state represented by the director given by Eq. (8), the terms connected with ν_1, ν_2, ν_3, ν_5, and ν_6 do not contribute, because $\mathbf{n}\cdot\mathbf{t} = \cos\theta$, which is independent of z. The only term that survives is the one connected with ν_4, because

$$[(\mathbf{t}\cdot\nabla)\mathbf{n}]^2 = q^2\sin^2\theta.$$

For what concerns the usual Frank terms, the nonvanishing ones are

$$(\mathbf{n}\times\nabla\times\mathbf{n})^2 = q^2\sin^2\theta\cos^2\theta$$
$$\text{and}\quad (\mathbf{n}\cdot\nabla\times\mathbf{n})^2 = q^2\sin^4\theta.$$

After collecting all the nonvanishing terms, one obtains for the elastic energy density the following expression:

$$f(q,x) = f_1 - \tfrac{1}{2}\eta(1-x) - \kappa_2 q x + \tfrac{1}{2}K_{22}q^2x^2 + \tfrac{1}{2}K_{33}q^2x(1-x) + \nu_4 q^2 x, \quad (12)$$

where $x = \sin^2\theta$ is the order parameter of the transition. If $x = 1$, that is, $\mathbf{t}\perp\mathbf{n}$, Eq. (12) reduces to energy density of a cholesteric phase, with an effective twist elastic constant equal to $K_{22} + 2\nu_4$. For $x < 1$, the procedure is now to minimize $f(q,x)$ with respect to x and q. By imposing $\partial f/\partial x = 0$ and $\partial f/\partial q = 0$, one obtains

$$x^* = -\frac{K_{33} + 2\nu_4 \mp \kappa_2\sqrt{(K_{33}+2\nu_4)/\eta}}{K_{22}-K_{33}} \quad (13)$$

for the cone angle of $\mathbf{n}$ with $\mathbf{t}$, and

$$q^* = \pm\sqrt{\frac{\eta}{K_{33}+2\nu_4}} \quad (14)$$

for the wave vector. Note that, by replacing Eq. (14) with Eq. (13), one obtains the same x^* no matter the sign of q^*. Thus, both signs of q^* are likely, indicating that both right- and left-handed twist have the same chance of occurring. This result is in agreement with the experimental observations that both chiralities are present in a sample [8,29,30] in the form of domains. For the subsequent analysis we arbitrarily chose the (+) sign for the chirality without loss of generality. The director profile associated with x^* and q^* corresponds to a minimum of the free energy density of an unlimited nematic sample only if

$$\left(\frac{\partial^2 f}{\partial x^2}\right)_{x^*,q^*} \geqslant 0, \quad (15)$$

and the Hessian determinant defined by

$$H(x^*,q^*) = \left\{\frac{\partial^2 f}{\partial x^2}\frac{\partial^2 f}{\partial q^2} - \frac{\partial^2 f}{\partial x\partial q}\frac{\partial^2 f}{\partial q\partial x}\right\} > 0. \quad (16)$$

Simple calculations yield

$$\left(\frac{\partial^2 f}{\partial x^2}\right)_{x^*,q^*} = \eta\frac{K_{22}-K_{33}}{K_{33}+2\nu_4} \geqslant 0 \quad (17)$$

and

$$H(x^*,q^*) = \kappa_2\sqrt{\eta(K_{33}+2\nu_4)} - \eta(K_{33}+2\nu_4) > 0. \quad (18)$$

These are the requirements to be fulfilled in order to have an equilibrium state. We note here that the pseudoscalar κ_2 is a phenomenological elastic constant that quantifies the tendency of the molecules to twist (see, e.g., Ref. [31]). Therefore, it is different from zero for (i) chiral molecules as in a cholesteric phase, and (ii) for "achiral" molecules that may give helical structure as in the case of banana shaped molecules and dimers [8,19,29,30,32,33]. The Hessian gives a minimum as far as $\kappa_2 > \sqrt{\eta(K_{33}+2\nu_4)} = \kappa_c$, that is, for values of κ_2 larger than a critical value κ_c, otherwise no TB phase could be stabilized. Note that for $\kappa_2 = \kappa_c$, Eq. (13) gives $x^* = 0$, which is a nematic phase with $\mathbf{t}\|\mathbf{n}$. From Eqs. (17) and (18), one concludes that two scenarios are possible. Since q^* is real, one obtains that a stable ground state may exist if $\eta < 0$ and $K_{33} + 2\nu_4 < 0$. In this case, $\eta(K_{22}-K_{33})/(K_{33}+2\nu_4) \geqslant 0$ when $K_{22} > K_{33}$. On the other hand, another stable ground state may be obtained if $\eta > 0$ and $K_{33} + 2\nu_4 > 0$. Again, $\eta(K_{22}-K_{33})/(K_{33}+2\nu_4) \geqslant 0$ when $K_{22} > K_{33}$. Thus, both situations are physically possible, implying that the twist elastic constant is such that $K_{22} > K_{33}$, as observed experimentally (see, e.g., Ref. [34]). Now, the analysis can be cast in the framework of a simple Landau-like expansion of the free energy if one rewrites Eq. (12) in a still more compact form as

$$f(x) = f_1 - \frac{\eta}{2} + Ax + Bx^2, \quad (19)$$

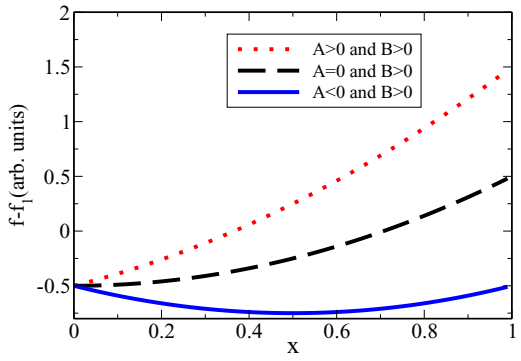

FIG. 2. (Color online) Elastic energy density $F = f - f_1$ (in arbitrary units), given by Eq. (19), as a function of x in three situations: (1) for $A > 0$ and $B > 0$ (dotted line), (2) $A = 0$ and $B > 0$ (dashed line), and (3) $A < 0$ and $B > 0$ (solid line). For illustrative purposes, the curves have been drawn for $\eta > 0$.

in which $A = -\overline{\kappa}_2 + \eta/2 + \overline{K_{33}}/2$ and $B = \Delta/2$, with the following compact notation introduced: $\overline{\kappa}_2 = \kappa_2 q^*$, $\overline{K_{33}} = (K_{33} + 2\nu_4)(q^*)^2$, and $\Delta = (K_{22} - K_{33})(q^*)^2$. Simple calculations show that f is minimum for $x = -A/(2B)$, as given by (13) when (14) is taken into account. This corresponds to a stable state if $B > 0$, i.e., $\Delta > 0$ or $K_{22} > K_{33}$. The twist-bend phase, which corresponds to $x \neq 0$, is energetically favored only if $A < 0$ (see Fig. 2). Thus, there is a phase transition from the usual nematic phase ($x = 0$) to a twist-bend nematic $x \neq 0$ for $A = 0$, i.e., when $2\kappa_2 q^* = \eta + (K_{33} + 2\nu_4)(q^*)^2$, which, by using (14), becomes $\eta = \eta_c = \kappa_2^2/(K_{33} + 2\nu_4)$. For this critical value, the wave vector of the modulated phase assumes the value $q_c = |\kappa_2/(K_{33} + 2\nu_4)|$. In addition, $A = \eta - \sqrt{\eta\eta_c}$ may be used to drive the transition according to the values of η. Therefore, one can conclude that a second-order phase transition occurs when $\eta = \eta_c$, if $x = \sin^2\theta$ is used as the "order parameter" for the N_{TB} phase. This critical value of η separates the usual from the twist-bend nematic phase. In this way as well, the parameter η may play a role similar to the temperature such that $A > 0$ for $\eta > \eta_c$, and $A < 0$ for $\eta < \eta_c$.

Some concluding remarks are in order. The experimentally discovered twist-bend nematic phase may be described from the elastic point of view by means of a quadratic theory, which can be constructed as the usual one, i.e., by considering only the elements of symmetry enough to characterize the system. For the twist-bend phase, these elements are the director **n** and the helix vector **t**, the latter playing the role of an internal field. In addition to the usual Frank constants, the elastic theory requires the existence of three additional parameters, namely, η, which is the coupling constant between the nematic director and the director of the helical structure; κ_2, which is the coefficient of the twist distortion, peculiar to the heliconical phase; and ν_4, a parameter that renormalizes the elastic constant K_{33}. The model can be interpreted in analogy with the Landau phenomenological approach for phase transitions. This analogy permits one to treat the cone angle θ as the main ingredient for an "order parameter" defined by $x = \sin^2\theta$. The twist-bend nematic phase is then the one for which $x \neq 0$, and is energetically favored with respect to the usual nematic phase ($x = 0$) when η is larger than a critical value η_c pointing to a second-order transition between the two stable phases allowed by symmetry considerations, the uniform nematic and the twist-bend nematic. This scenario is characterized by the exigence that $K_{22} > K_{33}$ always, as experimentally evidenced. In addition, the possibility of having negative values of K_{33}, as argued by some authors, may not be excluded but it is not mandatory. Indeed, one of the possible stable phases may exist for $\eta < 0$ and $K_{33} + 2\nu_4 < 0$, but this condition may be satisfied also for $K_{33} > 0$ when the elastic parameter ν_4 is negative enough, i.e., when the term $\nu_4[(\mathbf{t}\cdot\nabla)\mathbf{n}]^2 = \nu_4 q^2 \sin^2\theta$ is energetically favored, which is a physically sound possibility. Finally, note that (i) our model explains why some bend-core molecules give rise to the N_{TB} phase and others do not. This is due to the existence of a threshold for the parameter $\kappa_2 = \kappa_c$ below which the molecular interaction is not strong enough to induce long range twisted structures. (ii) It does not imply a negative K_{33} in order to be stabilized the twist-bend phase, in agreement with recent experimental results which show that K_{33} goes through a positive minimum at the transition [34,35]. (iii) It gives two minima in the twist-bend nematic corresponding to (+) and (−) chirality domains in agreement with experimental results [8,9,18,19,30] and previous theoretical models [6,20].

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