Isotropic-cholesteric transition of a weakly chiral elastomer cylinder

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When a chiral isotropic elastomer is brought to the low-temperature cholesteric phase, the nematic degree of freedom tends to order and form a helix. Due to the nematoelastic coupling, this also leads to elastic deformation of the polymer network that is locally coaxial with the nematic order. However, the helical structure of nematic order is incompatible with the energetically preferred elastic deformation. The system is therefore frustrated and appropriate compromise has to be achieved between the nematic ordering and the elastic deformation. For a strongly chiral elastomer whose pitch is much smaller than the system size, this problem has been studied by Pelcovits and Meyer, as well as by Warner. In this work, we study the isotropic-cholesteric transition in the weak-chirality limit, where the pitch is comparable to or much larger than system size. We compare two possible solutions: a helical state as well as a double-twist state. We find that the double-twist state very efficiently minimizes both the elastic free energy and the chiral nematic free energy. On the other hand, the pitch of the helical state is strongly affected by the nematoelastic coupling. As a result, this state is not efficient in minimizing the chiral nematic free energy.

I. INTRODUCTION

An isotropic chiral elastomer can be synthesized by cross-linking a chiral nematic polymer melt in the isotropic phase. When such a system is brought into the low-temperature cholesteric phase,[1] the nematic degree of freedom orders locally and tends to form a helical structure. Due to the nematoelastic coupling, the polymer network tends to stretch along the direction of the local nematic order, which continuously rotates along the helical axis. For a system with cylindrical shape, this leads to strain deformation which increases linearly with the cylinder radius, as illustrated in Fig. 2(b) below. Its elastic energy cost is formidably high, when the cylinder radius is much larger than the helical pitch. This frustration due to competition between network elasticity and liquid crystalline ordering makes it nontrivial to find the ground state of the system in the cholesteric phase.

This problem was first studied by Pelcovits and Meyer [1] using linear elasticity theory. In the limit of infinitely strong chirality, it is clear that the system should first satisfy the chirality by forming a planar helix along the cylinder axis. On the other hand, to avoid large strain energy, the solid can only deform uniaxially, which implies that the nematoelastic coupling can only be partially satisfied. Such a state, as illustrated in Figs. 2(a) and 2(c), is called a planar helix state in Ref. [1] and a transverse cholesteric state in Ref. [2]. As the chirality is tuned to be weaker, a conical helix state, where the processing director has nonvanishing component along the helical axis, may constitute a better solution. The associated solid deformation as well as director pattern for this conic state are illustrated in Figs. 2(d) and 2(e), respectively. Warner [2] carried out a nonlinear analysis of the same problem using neoclassical elasticity theory [3,4]. Nontrivial dependence of the phase boundary on the magnitude

of the nematic order was identified. The multicritical point associated with the planar-conic transition (where the first- and second-order transition lines meet) was also analyzed.

It is implicitly assumed in both Ref. [1] and Ref. [2] that the pitch of the corresponding nematic liquid crystal system (typically ≤ 0.1 μm) is much smaller than the system size, e.g., the radius of the cylinder. That is, the elastomer is in the strong-chirality limit. This is certainly correct for many cases. Nevertheless, the cholesteric pitch can be continuously tuned by changing the concentration of chiral chemical groups during polymerization. In particular, it can be tuned to be comparable to macroscopic length scales, e.g., the system size. This is especially true if the system has the shape of a thin cylinder or wire. It is therefore interesting and relevant to study the isotropic-cholesteric transition in the weak-chirality limit. In this work, we carry out a nonlinear elasticity analysis of this problem using variational methods. We find that in this regime a double-twist state has lower free energy than the usual helix director pattern. The results obtained by Pelcovits and Meyer [1] and Warner [2], as well as the authors in this work are summarized by the "phase diagram" of a chiral nematic cylinder in Fig. 1.
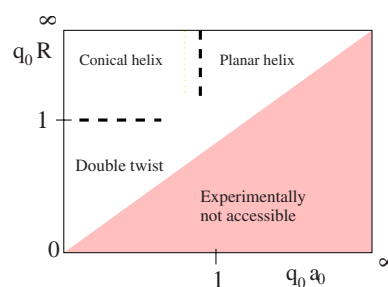

FIG. 1. (Color online) Phase diagram of a chiral cylinder in the nematic phase. The parameters a_0, q_0, and R are defined in Sec. II. The transition between the planar and conical helix states may be continuous or discontinuous, while the transition between the conical helix and double-twist states is expected to be discontinuous.

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[1] Certain precautions have to be taken in order that the system does not end up with a polydomain state where there is only short-range chiral nematic order.

II. MODEL

The total free energy per unit volume of a chiral liquid crystalline elastomer cross linked in the isotropic phase is given by

$$f = f_{\text{el}} + f_{\mathbf{Q}}, \qquad (1)$$

where f_{el} is the neoclassical elastic free energy

$$f_{\text{el}} = \frac{1}{2} \mu \text{Tr} \mathbf{\Lambda}^{\text{T}} l^{-1} \mathbf{\Lambda} - \frac{3}{2} \mu, \qquad (2)$$

with

$$\Lambda_{ia} = \frac{\partial r_i}{\partial x_a} \qquad (3)$$

the deformation gradient matrix defined relative to the isotropic reference state $\vec{r} = \vec{x}$, which is subject to the incompressibility constraint

$$\det \mathbf{\Lambda} \equiv 1.$$

The vector $\vec{x}$ hence coincides with the position of the mass point in the isotropic reference state and is usually referred to as the Lagrangian coordinate. The vector $\vec{r}$, on the other hand, describes the position of the mass point in the chiral nematic reference state (the ground state that minimizes the total free energy), and is usually referred to as the Eulerian coordinate. As a general property of nonlinear elasticity theory, it is essentially important to distinguish these two coordinates properly. The symmetric and positive definite tensor l in the neoclassical elastic free energy Eq. (2) is called the step length tensor [3,4] of the current state, or deformed state,[2] and describes the statistical conformation of polymer chains in the current state. It is related to the nematic order parameter $\mathbf{Q}$ by

$$l = a\mathbf{I} - b\mathbf{Q}, \qquad (4)$$

where a and b are some microscopic constants. In this work we shall always normalize l such that it has determinant 1. This is always possible as long as the magnitude of nematic order is saturated, as it always is deep in the nematic phase. In the principal coordinate system of the nematic order parameter, the step length tensor l can be represented as a matrix:

$$l = \begin{pmatrix} 1/\zeta & 0 & 0 \\ 0 & 1/\zeta & 0 \\ 0 & 0 & \zeta^2 \end{pmatrix} = (\zeta^2 - \zeta^{-1})\hat{n}\hat{n} + \zeta^{-1}\mathbf{I}, \qquad (5)$$

where ζ is a monotonically increasing function of the magnitude of the nematic order S, whose detailed functional form is irrelevant to our study. For an achiral nematic elastomer, ζ turns out to be the ratio of spontaneous stretch along the direction of the nematic director when the system enters the

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[2]Strictly speaking, there is also a step length tensor l_0 in the reference preparation state, which appears in the neoclassical free energy, in front of $\mathbf{\Lambda}^T$. However, since the reference state is isotropic, l_0 is proportional to the identity tensor and therefore can be eliminated by redefinition of l.

nematic phase from the isotropic phase [3,4]. Due to the incompressibility constraint, the system shrinks by a factor of $1/\sqrt{\zeta}$ in the perpendicular directions. Finally, we note that in Eq. (2) a constant term $-3\mu/2$ is introduced so that the elastic free energy vanishes in the isotropic reference state where $\mathbf{\Lambda} = l = \mathbf{I}$.

The second part $f_{\mathbf{Q}}$ in Eq. (1) is the Landau–de Gennes free energy for a chiral nematic liquid crystal. Assuming that the nematic order is well saturated with fixed magnitude S in the cholesteric state, the relevant nematic free energy is the Frank free energy for chiral nematic liquid crystals [5,6]:

$$f_{\text{Frank}} = \frac{1}{2} K_1 (\vec{\nabla} \cdot \hat{n})^2 + \frac{1}{2} K_2 (\hat{n} \cdot \vec{\nabla} \times \hat{n} - q_0)^2$$

$$+ \frac{1}{2} K_3 (\hat{n} \times \vec{\nabla} \times \hat{n})^2 + K_{24} \vec{\nabla} \cdot (\hat{n} \cdot \vec{\nabla}\hat{n} - \hat{n}\vec{\nabla} \cdot \hat{n}),$$

$$(6)$$

where K_1, K_2, and K_3, are the splay, twist, and bending constants, respectively, while $q_0^{-1} = \ell_0$ is the cholesteric pitch for the corresponding chiral nematic liquid crystal. K_{24} is the saddle splay constant, which plays an important role in the physics of the blue phase [5,7,8]. Since the saddle splay density is a complete differential, its volume integral can be transformed into a surface integral by Gauss' theorem, and therefore scales in the same way as the surface anchoring of the nematic director field, which we shall not consider in this work. Nevertheless, it is rather straightforward to include this surface interaction. Here it is important to note that all the derivatives in Eq. (6), $\vec{\nabla}_i = \partial/\partial r_i$, etc., are with respect to the *Eulerian coordinates*, i.e., Cartesian coordinates of mass points in their deformed states. This is required by the liquid nature of the Frank free energy: at length scales where the Frank free energy becomes important, the system is essentially a liquid. The physical quantities of a liquid should be naturally expressed in terms of Eulerian rather than Lagrangian coordinates. To avoid confusion in notation, we shall use $\vec{\nabla}_i = \partial/\partial r_i$ for the derivative with respect to Eulerian coordinates and $\partial_a = \partial/\partial x_a$ to denote the partial derivative with respect to the Lagrangian coordinates.

Let us first have a qualitative discussion of the total free energy Eq. (1). Within the one-constant approximation of the Frank free energy, and ignoring the surface saddle splay term for a moment, there are three natural length scales in this problem. $q_0^{-1} = \ell_0$ is the chirality pitch, while $a_0 = \sqrt{K/\mu}$ is the nematic penetration length, i.e., the crossover length scale set by the competition between network elasticity and nematic director elasticity. The third length scale is the radius R of the cylinder. For most liquid crystalline elastomers, we estimate $K \sim (2-4) \times 10^{-12}$ N, while $\mu \sim 10^4 - 10^6$ Pa. Therefore $a_0 \sim 1-10$ nm, constituting the shortest length scale in our problem. On the other hand, the chirality pitch ℓ_0 can vary a lot; it is typically 0.1 μm or smaller for strongly chiral materials but may get much larger for weakly chiral materials. In particular, it can even be larger than the cylinder radius R. The regime $a_0/R \gg 1$ is clearly experimentally inaccessible.

Comparing these three length scales, we naturally are led to the following two distinct regimes: (1) the weak-chirality
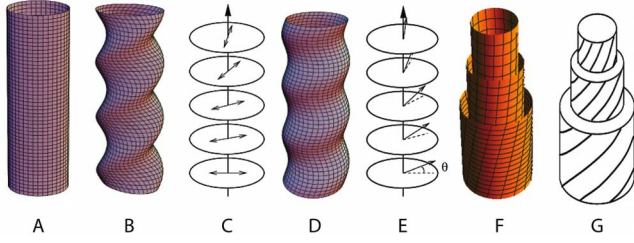
FIG. 2. (Color online) Elastic deformation and director pattern for various states. The grid lines in (a), (b), (d), and (f) are associated with the Lagrangian coordinate system and therefore characterize elastic deformations relative to the reference state (a). (a) Reference cylinder in the isotropic phase. (b) Elastic deformation of the planar helix state at the weak-chirality limit, studied in Sec. IV. (c) Nematic director pattern of the planar helix state. Note that the pitches in (c) and (d) are not in the same scale. This is only an artifact. (d) Elastic deformation of the conical helix state, studied in Refs. [1,2]. The pitch is exaggerated to make it clear. The real pitch should be much smaller than the cylinder radius. (e) Director pattern of the conical helix state; θ is the conical angle. (f) Elastic deformation of the double-twist state. (g) Director pattern of the double-twist state. The lines show the local direction of the nematic director.

regime $a_0 \ll R \ll \ell_0$; and (2) the strong-chirality regime $a_0, \ell_0 \ll R$. The strong-chirality limit has already been analyzed by Pelcovits and Meyer [1], as well as by Warner [2]. It is found that, as one tunes the dimensionless ratio a_0/ℓ_0 $= a_0 q_0$ to below a critical value of the order of unity, the system goes from a planar helix director pattern to a conical helix pattern. In this work, we shall mainly focus on the weak-chirality limit. Similar to Refs. [1,2], we shall use variational methods, proposing two kinds of candidate states with certain variational parameters and minimizing the total free energy over these parameters.

III. DOUBLE TWIST OF NEMATIC DIRECTOR AND TWIST OF CYLINDER

Consider a cylindrical block of isotropic chiral elastomer of radius R, aligning along the z axis, as illustrated in Fig. 2(a). We need to find the nematic director field $\hat{n}(\vec{x})$ as well as the elastic deformation $\vec{r}(\vec{x})$ relative to the isotropic reference state that minimizes the total free energy. One possible low-energy configuration for the chiral Frank free energy is a double-twist texture,[3] as illustrated in Fig. 2(g). In chiral nematic liquid crystals, the double-twist configuration is energetically favorable if the saddle splay modulus K_{24} is positive and large enough [5,7,8]. According to the current understanding of the blue phase, these double-twist cylinders pack into a three-dimensional periodic structure with cubic symmetry. In liquid crystalline elastomers, due to the nema-

toelastic coupling, a double-twist nematic director texture necessarily induces a twist of the cylinder, together with a uniaxial stretch λ along the cylinder axis:

$$\vec{r}(\vec{x}) = \begin{pmatrix} 1/\sqrt{\lambda} & 0 & 0 \\ 0 & 1/\sqrt{\lambda} & 0 \\ 0 & 0 & \lambda \end{pmatrix} \mathbf{O}_z(\alpha z) \cdot \vec{x}, \qquad (7)$$

where

$$\mathbf{O}_z(\alpha z) = \begin{pmatrix} \cos \alpha z & -\sin \alpha z & 0 \\ \sin \alpha z & \cos \alpha z & 0 \\ 0 & 0 & 1 \end{pmatrix} \qquad (8)$$

is a z-dependent rotation about the z axis, i.e., a uniform twist. This deformation of the solid is illustrated in Fig. 2(f). Using the cylindrical coordinate system, the Lagrangian coordinates of a point $\vec{x}$ are given by the triplet (ρ, ϕ, z):

$$\vec{x} = \begin{pmatrix} \rho \cos \phi \\ \rho \sin \phi \\ z \end{pmatrix}. \qquad (9)$$

In the deformed state Eq. (7), the Eulerian coordinates $\vec{r}(\vec{x})$ are given by the triplet (r, φ, r_z), where

$$\vec{r}(\vec{x}) = \begin{pmatrix} r \cos \varphi \\ r \sin \varphi \\ r_z \end{pmatrix} = \begin{pmatrix} \dfrac{\rho}{\sqrt{\lambda}} \cos(\phi + \alpha z) \\ \dfrac{\rho}{\sqrt{\lambda}} \sin(\phi + \alpha z) \\ \lambda z \end{pmatrix}, \qquad (10)$$

where we have used Eqs. (7) and (9). Therefore we find

$$(r, \varphi, r_z) = \left(\frac{\rho}{\sqrt{\lambda}}, \phi + \alpha z, \lambda z \right). \qquad (11)$$

Note that the deformed cylinder has height $L\lambda$ and radius $R/\sqrt{\lambda}$.

Let $\hat{e}_\varphi$ be the unit vector associated with the Eulerian cylindrical coordinate φ:

$$\hat{e}_\varphi = \left| \frac{\partial \vec{r}}{\partial \varphi} \right|^{-1} \frac{\partial \vec{r}}{\partial \varphi} = \begin{pmatrix} -\sin \varphi \\ \cos \varphi \\ 0 \end{pmatrix}. \qquad (12)$$

In terms of the Eulerian coordinates, a double-twist texture of the nematic director is represented as

$$\hat{n}(\vec{r}) = \hat{e}_z \cos \theta(r) + \hat{e}_\varphi \sin \theta(r). \qquad (13)$$

Note that the twist angle $\theta(r) = \theta(\rho/\sqrt{\lambda})$ can be equally well represented as a function of the Lagrangian coordinate ρ. $\theta(r)$ satisfies the boundary condition $\theta(0) = 0$, since the nematic director is parallel to $\hat{z}$ on the center axis of the cylinder. On the outer surface of the cylinder $r = R/\sqrt{\lambda}$, $\theta(R/\sqrt{\lambda})$ is free to vary.

Calculation of the deformation gradient using Eq. (7) is a trivial and tedious matter. On the other hand, by substituting Eq. (13) into Eq. (5) we can calculate the step length tensor l. Substituting these results into Eq. (2), we find that the

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[3]It is called a double twist, because starting from the center, the nematic director is twisted along each of two directions perpendicular to the cylinder axis. This should be contrasted with the usual cholesteric state, where the nematic director is twisted only along one direction.

spatially dependent elastic free energy density for the proposed double-twist solution is given by

$$f_{el} = \frac{\mu}{4\zeta^2\lambda}[1 + \lambda^3\zeta^3 + \alpha^2\rho^2\zeta^3 + 3\zeta^3 + \lambda^3 + \alpha^2\rho^2$$
$$- (\zeta^3 - 1)(\lambda^3 - \alpha^2\rho^2 - 1)\cos(2\theta)$$
$$- 2\alpha(\zeta^3 - 1)\lambda^{3/2}\rho\sin(2\theta)]. \tag{14}$$

The spatially dependent Frank free energy density can be calculated by substituting Eq. (13) into Eq. (6), carefully noting that all derivatives are with respect to the Eulerian coordinates $\vec{r}$. The result is

$$f_{Frank} = \frac{1}{2}K_2\left(\frac{1}{2r}\sin 2\theta + \frac{d\theta}{dr} - q_0\right)^2$$
$$+ \frac{1}{2}K_3\frac{\sin^4\theta}{r^2} - K_{24}\frac{\sin 2\theta}{r}\frac{d\theta}{dr}, \tag{15}$$

which is identical to that for a chiral nematic liquid crystal in a double-twist cylinder [6].

In the weak-chirality limit, $q_0 R \ll 1$, we expect $\theta(r)$ to be small and linear in r. We can therefore expand the elastic free energy density in terms of ρ and $\theta(r)$:[4]

$$f_{el} = f_0 + f_2 + \text{(higher-order terms)}, \tag{16}$$

where

$$f_0 = \frac{\mu\lambda^2}{2\zeta^2} + \frac{\zeta\mu}{\lambda} - \frac{3}{2}\mu, \tag{17}$$

$$f_2 = \frac{\mu}{2\zeta^2\lambda}[\alpha^2\rho^2\zeta^3 + (\zeta^3 - 1)\theta^2(\lambda^3 - 1) - 2\alpha(\zeta^3 - 1)\theta\lambda^{3/2}\rho] \tag{18}$$

are terms of order of r^0 and r^2, respectively. We shall ignore all higher-order terms in the elastic free energy. Note that f_0 is exactly the free energy density for a monodomain nematic elastomer, with anisotropy ratio ζ, undergoing a uniaxial deformation coaxial with the step length tensor. Minimizing f_0 over λ, we obtain

$$\lambda = \zeta \rightarrow f_0 = 0, \tag{19}$$

as expected. Substituting this into Eq. (18), we find

$$f_2 \rightarrow \frac{\mu}{2\zeta^3}[(\zeta^3 - 1)\theta - \alpha\zeta^{3/2}\rho]^2, \tag{20}$$

which is a complete square. Since f_{Frank} is independent of α, and since θ is linear in ρ as will be shown below, Eq. (20) is minimized by

$$\alpha = \zeta^{-3/2}(\zeta^3 - 1)\frac{\theta}{\rho}, \tag{21}$$

$$f_2 = 0. \tag{22}$$

Hence, there is *no* elastic free energy cost for the double-

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[4]Remembering that $\rho = \sqrt{\lambda}r$ is proportional to r.

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twist state up to the order of $(\alpha R)^2$. As we shall show below, the parameter α is of the order of q_0. Hence $(\alpha R)^2$ is indeed a small parameter in the weak-chirality limit.

Similarly, we also expand the Frank free energy in terms of θ and r. To the leading order we find

$$f_{Frank} = \frac{1}{2}K_2\left(\frac{\theta}{r} + \frac{d\theta}{dr} - q_0\right)^2 - K_{24}\frac{2\theta}{r}\frac{d\theta}{dr}, \tag{23}$$

which depends only on θ. Note that the bending term is of higher order when compared to all other terms that we have kept.

We have to minimize the total Frank free energy density

$$F_{Frank} = 2\pi L\lambda\int_0^{R/\sqrt{\zeta}} f_{Frank} r\, dr \tag{24}$$

over $\theta(r)$ in order to determine the optimal director texture. Let us define $R_\zeta = R/\sqrt{\zeta}$ in order to streamline the notations below. Calculating the first variation of the Frank free energy, including the boundary terms at $r = R_\zeta$, we find

$$\frac{\delta F_{Frank}}{2\pi L\lambda} = K_2\int_0^{R_\zeta} dr\left(-r\theta''(r) - \theta'(r) + \frac{\theta(r)}{r}\right)\delta\theta(r)$$
$$+ [K_2(\theta(R_\zeta) + R_\zeta\theta'(R_\zeta) - R_\zeta q_0)$$
$$- 2K_{24}\theta(R_\zeta)]\delta\theta(R_\zeta). \tag{25}$$

Since the twist angle $\theta(r)$ is free to vary on the boundary $r = R_\zeta$, we have to set both the integrand and the boundary term to zero in order to find the minimizing solution. This leads to the following two Euler-Lagrange equations:

$$r\theta''(r) + \theta'(r) + \frac{\theta(r)}{r} = 0, \tag{26}$$

$$(\theta(R_\zeta) + R_\zeta\theta'(R_\zeta) - R_\zeta q_0) - 2\eta\theta(R_\zeta) = 0, \tag{27}$$

where $\eta = K_{24}/K_2$ is a dimensionless ratio. Solving these two equations, we find

$$\theta(r) = \frac{q_0}{2(1-\eta)}r = \frac{q_0\rho}{2(1-\eta)\sqrt{\zeta}}, \tag{28}$$

which explicitly shows that $\theta(r)$ is indeed linear in r. The twist angle on the boundary is given by

$$\theta(R_\zeta) = \frac{q_0 R}{2(1-\eta)\sqrt{\zeta}},$$

which serves as a small parameter that controls the validity of the perturbative analysis. Substituting Eq. (28) into Eq. (21), we find the parameter α given by

$$\alpha = \frac{(\zeta^3 - 1)q_0}{2(1-\eta)\zeta^2}, \tag{29}$$

which is indeed a constant, of the same order as q_0, and independent of r. Substituting Eq. (28) into Eq. (23), we find the Frank free energy density, which is also the total free energy density [since the elastic free energy vanishes at the order of $(\alpha R)^2$], given by

$$f_{tot} = f_{Frank} = -\frac{K_2 \eta q_0^2}{2(1-\eta)}. \tag{30}$$

Summarizing Eqs. (28)–(30), we find that, if $\eta < 1$, our perturbative calculation is quantitatively good in the weak-chirality regime where $q_0 R / 2(1-\eta)\sqrt{\zeta} \ll 1$. The double-twist state is very efficient in minimizing both the elastic free energy and the Frank free energy. In particular, when the saddle splay constant K_{24} vanishes, $\eta = 0$, and therefore the total free energy Eq. (30) also vanishes. Note that the total free energy is positive definite if $\eta = 0$. Hence, the double-twist state is clearly the ground state, at least up to the order of $(q_0 R)^2$. By contrast, for a cholesteric liquid crystal with $K_{24} = 0$, the blue phase is clearly not the lowest-energy state, compared to the usual helical state. This shows that, unlike for the blue phase of cholesteric liquid crystal, the saddle splay constant K_{24} does not play an important role in the formation of the double-twist pattern in cholesteric elastomers. When $q_0 R / 2(1-\eta)\sqrt{\zeta}$ is comparable to or larger than unity, the higher-order terms of the elastic free energy and the Frank free energy cannot be neglected, and one has to minimize the full free energy Eqs. (14) and (15). Finally, if $\eta > 1$, a perturbative calculation in powers of αR is qualitatively incorrect, no matter how small the parameter $q_0 R$. We must minimize the full elastic free energy Eqs. (14) and (15).

IV. HELICAL STATE

In the weak-chirality regime that we are interested in, $q_0 a_0 \ll q_0 R \ll 1$, the elastic energy scale (per unit volume) μ is much larger than the chiral Frank energy scale $K q_0^2$. Therefore, the conical helix state studied in Refs. [1,2] can never be the ground state, as it only partially minimizes both the Frank free energy and the elastic free energy. There is, however, another potential candidate for the ground state, which can minimize the elastic free energy up to the leading order. Let us consider a planar helix director pattern along the cylinder axis, where the nematic director remains perpendicular to the cylinder z axis and rotates around this axis with pitch α:[5]

$$\hat{n}(z) = \hat{e}_x \cos \alpha z + \hat{e}_y \sin \alpha z = \mathbf{O}_z(\alpha z)\hat{e}_x. \tag{31}$$

In this work, we shall use both dyadic notation and matrix notation for tensor quantities. The corresponding local step length tensor is given by

$$\boldsymbol{l}(z) = \mathbf{O}_z(\alpha z)\boldsymbol{l}(z=0)\mathbf{O}_z(-\alpha z), \tag{32}$$

where

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[5]We note that α is the helical pitch measure by the Lagrangian coordinate $\vec{x}$. The physical value of the pitch, however, should be defined using the Eulerian coordinate and is therefore given by $\alpha\sqrt{\zeta}$.

$$\boldsymbol{l}(z=0) = \begin{pmatrix} \zeta^2 & 0 & 0 \\ 0 & 1/\zeta & 0 \\ 0 & 0 & 1/\zeta \end{pmatrix} \tag{33}$$

is the step length tensor at the plane $z=0$. This variational form of the nematic director field is the same as the planar helix state considered in Ref. [1].

Due to the nematoelastic coupling, the polymer network prefers to stretch along the local nematic director. This, however, implies that the direction of local strain deformation rotates $\pi/2$ between two cross sections $\ell_0/4$ apart along the cylinder, which leads to additional strain energy density $\mu(\alpha r)^2$ that explicitly depends on the radius r. For a fat cylinder (or in the strong-chirality limit), $\alpha R \gg 1$ and this strain energy is prohibitively high. Indeed it is shown by Pelcovits and Meyer [1], as well as by Warner [2], that the system prefers a uniform uniaxial deformation along the cylinder axis in this regime. For a thin cylinder (or in the weak-chirality limit), however, $\alpha R \ll 1$ and this additional strain energy constitutes only a perturbation to the strain energy of the corresponding uniform deformation. Nevertheless, to reduce the additional strain energy at the order of $\mu(\alpha R)^2$, the system can globally twist in the direction opposite to the nematic helix. The overall nonuniform deformation, shown in Fig. 2(b), is represented by the Eulerian coordinates as functions of the Lagrangian coordinates:

$$\begin{aligned} \vec{r}(\vec{x}) &\equiv \widetilde{\boldsymbol{\Lambda}}(z) \cdot \vec{x} \\ &= \mathbf{O}_z(\alpha z)\begin{pmatrix} \lambda & 0 & 0 \\ 0 & 1/\sqrt{\lambda} & 0 \\ 0 & 0 & 1/\sqrt{\lambda} \end{pmatrix}\mathbf{O}_z[-(\alpha+\beta)z] \cdot \vec{x}, \end{aligned} \tag{34}$$

where β measures the global twist of the solid. We note that also, due to the additional inhomogeneous strain deformation, the inverse pitch of the director helix α is generically different from the inverse pitch p_0 of the corresponding liquid crystal system. This will be made clear in the calculation below.

It is important to note that the matrix $\widetilde{\boldsymbol{\Lambda}}$ defined through Eq. (34) explicitly depends on the coordinate z and is *not* the deformation gradient Λ_{ia}. The latter should be obtained by taking the partial derivative of Eq. (34) with respect to the Lagrangian coordinates x, y, z. Being derived in this way, the deformation gradient matrix naturally satisfies the following compatibility conditions:

$$\partial_a \Lambda_{ib} = \partial_b \Lambda_{ia}.$$

Substituting the deformation gradient and nematic order parameter Eq. (31) into Eq. (2), integrating over the reference volume of the cylinder, and dividing it by the total volume $\pi R^2 L$, we obtain the elastic free energy density for the proposed deformation gradient as

$$f_{el}[\alpha, \beta, \lambda] = f_0 + f_2, \tag{35}$$

$$f_0 = \mu\left(\frac{\zeta}{\lambda} + \frac{\lambda^2}{2\zeta^2}\right) - \frac{3}{2}\mu, \tag{36}$$

$$f_2 = \frac{\mu R^2}{8\zeta^2\lambda}[(\zeta^3 + \lambda^3)\beta^2 - 2(\zeta^3 - \lambda^{3/2})(\lambda^{3/2} - 1)\alpha\beta$$
$$+ \alpha^2(\zeta^3 + 1)(\lambda^{3/2} - 1)^2]. \qquad (37)$$

Note that f_0 is independent of the cylinder radius R and is identical to the free energy of an achiral nematic elastomer undergoing uniaxial and homogeneous deformation. By contrast, f_2 is proportional to R^2 and quadratic in α and β. f_2 is clearly due to the inhomogeneous deformation. The ratio between f_0 and f_2 scales as $(\alpha R)^2$ as discussed earlier. The dimensionless ratio αR characterizes the importance of chirality in this problem. In Refs. [1,2] this ratio is implicitly taken to be large at the very beginning. In this work, we shall assume it to be a small number. More precisely, we shall assume another dimensionless ratio $q_0 R \ll 1$. Also, we shall see below that for this proposed variational solution, the Frank free energy scales in the same way as f_2; hence, it is reasonable to minimize f_0 first and then the sum of f_2 and f_{Frank}. Minimization of f_0 leads to

$$\lambda = \zeta, \qquad (38)$$

$$f_0 \to 0. \qquad (39)$$

That is, the local elastic deformation is identical to that of a homogeneous achiral nematic elastomer. Inclusion of f_2 and f_{Frank} induces a small change of λ at the order of αR.

The Frank free energy density for the proposed director pattern Eq. (31) can also be easily calculated. Again we have to be careful with the derivatives in Eq. (6) with respect to the Eulerian coordinate $\vec{r}$. After some tedious calculation and replacing λ with ζ, we find

$$f_{\text{Frank}} = \frac{1}{2}K_2\alpha^2\zeta - K_2\alpha q_0\sqrt{\zeta} + \frac{1}{2}K_2 q_0^2, \qquad (40)$$

which is independent of the elastic constants K_1, K_3, and K_{24}.

We still need to minimize the sum of f_2, given in Eq. (35) and f_{Frank}, Eq. (40), over the remaining two variational parameters α, β. Since f_{Frank} does not depend on β, we minimize f_2 over β and find

$$\beta = \frac{\alpha(\zeta^{3/2} - 1)^2}{2\zeta^{3/2}}, \qquad (41)$$

$$f_2 = \frac{R^2\alpha^2(\zeta^3 - 1)^2\mu}{16\zeta^3}. \qquad (42)$$

We note that, as long as $\zeta \neq 1$ and $\alpha \neq 0$, the global spontaneous twist of solid β does not vanish. More importantly, unlike the double-twist state, the planar helix state considered here does cost elastic free energy at the order of $(\alpha R)^2$.

We can now minimize the sum of Eqs. (42) and (40) over α, which leads to

$$\alpha = \frac{8a_0^2 q_0 \zeta^{7/2}}{8a_0^2\zeta^4 + R^2(\zeta^3 - 1)^2}. \qquad (43)$$

Remembering that $a_0/R \leqslant 10^{-6}$ even for $R = 1$ mm, and $\zeta \neq 1$, the first term in the denominator can be safely ignored and α can be approximated as

$$\alpha = \frac{8a_0^2 q_0 \zeta^{7/2}}{R^2(\zeta^3 - 1)^2} \sim \left(\frac{a_0}{R}\right)^2 q_0 \ll q_0. \qquad (44)$$

This result indicates that the helix of the nematic director is strongly resisted by the nematoelastic coupling energy and the pitch becomes much longer than the corresponding value ℓ_0 in the nematic liquid crystal. Substituting Eq. (44) into Eqs. (42) and (40), we find

$$f_{\text{tot}} \approx -\frac{a_0^2}{R^2}\frac{4K_2 q_0^2 \zeta^4}{(\zeta^3 - 1)^2} + \frac{1}{2}K_2 q_0^2 \approx \frac{1}{2}K_2 q_0^2. \qquad (45)$$

The total free energy is therefore *positive*, in strong contrast with the double-twist state we considered in the preceding section. The planar helix state considered here is therefore not efficient in energy minimization. This is clearly due to the extra elastic free energy cost Eq. (42) caused by the nematoelastic coupling.

V. DISCUSSION AND CONCLUSION

We have shown in this work that, in the weak-chirality limit $q_0 a_0 \ll q_0 R \ll 1$, the double-twist state minimizes both the Frank free energy and the elastic free energy up to the order of $(q_0 R)^2$, and is therefore a good candidate for the real ground state. The planar helix state, on the other hand, is strongly influenced by the nematoelastic coupling, with the pitch much longer than the corresponding value in a cholesteric liquid crystal. As the dimensionless parameter $q_0 R$ becomes larger than 1, the elastic energy cost due to inhomogeneous strain, scaling as $\mu(q_0 R)^2$, dominates all other terms. When $q_0 R \gg 1$ and $q_0 a_0 \ll 1$, the ground state is likely to be the conical helix state with $\theta \approx \pi/2$, according to the studies in Refs. [1,2]. The conical state and the double-twist state are qualitatively different, and cannot be mutually accessed in a continuous fashion. Therefore the aforementioned two regimes are likely separated by a first-order phase transition. Study of this transition is technically challenging, mainly because in the transition region, $q_0 R \sim 1$, and neither the current analysis nor the analyses in Refs. [1,2] are valid. There is no small parameter that one can use to perform a perturbative calculation. Numerical analysis of this transition is feasible but is beyond the scope of this work.

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