Stability Against the Odds: The Case of Chromonic Liquid Crystals

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Abstract
The ground state of chromonic liquid crystals, as revealed by a number of recent experiments, is quite different from that of ordinary nematic liquid crystals: it is twisted instead of uniform. The common explanation provided for this state within the classical elastic theory of Frank demands that one Ericksen’s inequality is violated. Since in general such a violation makes Frank’s elastic free-energy functional unbounded below, the question arises as to whether the twisted ground state can be locally stable. We answer this question in the affirmative. In reaching this conclusion, a central role is played by the specific boundary conditions imposed in the experiments on the boundary of rigid containers and by a general formula that we derive here for the second variation in Frank’s elastic free energy.

Keywords Chromonic liquid crystals · Local stability · Frank’s free-energy functional · Ericksen’s inequalities · Elastic frustration

1 Introduction

Liquid crystals are anisotropic fluids that fall basically into two broad categories: they are either thermostropic or lyotropic, depending on whether it is temperature or concentration, respectively, responsible for driving the formation of these fascinating intermediate phases of soft condensed matter, which are birefringent like crystals and flow like liquids.
Chromonic liquid crystals (CLCs) are lyotropic. They are composed of plank-like molecules with a poly-aromatic core and polar peripheral groups, aggregated in columnar stacks resulting from noncovalent attractions between the poly-aromatic cores. CLCs are formed by certain dyes, drugs, and short nucleic-acid oligomers in aqueous solutions (Dickinson et al. 2009; Tam-Chang and Huang 2008; Mariani et al. 2009; Zanchetta et al. 2008; Nakata et al. 2007; Fraccia and Jia 2020). Their properties are reviewed in the following papers (Lydon 1998, a, 2010, 2011; Dierking and Neto 2020).

Lydon (2011) identifies three distinct phases in the history of CLCs. The first consists of isolated, episodical reports about novel nematic patterns observed in aqueous solutions of various substances, such as phenanthrene sulfonic acid, in the pioneering work (Sandqvist 1915), or diverse dyes. The second phase saw a growth of detailed studies devoted to the anti-asthmatic drug, disodium cromoglycate (DSCG). The final phase is identified with the realization that CLCs are widespread among drugs, dyes, nucleic acids, and similar water-soluble, aromatic compounds.

At low concentrations, supramolecular columns are not long enough to induce local nematic order. Upon increasing concentration, a nematic phase takes eventually over, although the critical concentration turns out to be much lower (see, for example, (Nayani et al. 2015)) than that predicted by Onsager’s excluded volume theory (Onsager 1949). Several unconventional models for columnar organization, envisioning the possibility that molecular stacks be either Y-shaped or side-slipped (Xiao et al. 2014; Park et al. 2008), have been put forward to try and explain this discrepancy. Upon further increasing concentration, the nematic phase gives way to a phase, called the M phase, as it is similar (that is, it has a herringbone texture similar) to the middle phases of conventional amphiphile systems (Lydon 2011).

CLCs are odd nematic phases. They do not seem to possess the same ground state as ordinary nematics. When a low-molecular liquid crystal in the nematic phase is left to itself, in the absence of either external disturbing agencies or confining boundaries, the nematic director \( \mathbf{n} \), which represents on a macroscopic scale the average orientation of the elongated molecules that constitute the medium, tends to be uniform in space, in a randomly chosen direction. This is not what a CLC does.

Experiments in capillary tubes with lateral boundary so smooth as to ensure degenerate planar anchoring for \( \mathbf{n} \), have shown that the spontaneous distortion is not the alignment along the cylinder’s axis, which is the only uniform one compatible with the boundary conditions; rather, it is a twisted orientation escaped along the cylinder’s axis, swinging away from it on the cylinder’s lateral boundary (see Fig. 1). Such an escaped-twist (ET) distortion\(^2\) is very similar (but not completely identical) to the double twist (DT), which, when energetically favored in cholesteric liquid crystals, gives rise to their blue phases\(^3\).

\(^1\) That is, with \( \mathbf{n} \) free to rotate in the local tangent plane.

\(^2\) This is the same as the escaped-twisted structure considered in Ondris-Crawford et al. (1993) that had been called twist-bend (Cladis and Kléman 1972) or escaped in the third dimension (Meyer 1973) before.

\(^3\) Blue phases, which owe their name to the color of their appearance, are exhibited by cholesteric liquid crystals in the proximity of their transition to the isotropic phase. They are attributed to a lattice of line defects that orderly traverse the medium (in different spatial arrangements in the phases labeled BPI and BPII) and to a disordered network in the amorphous phase BPIII (see pp. 68 and 407 of (Kleman and...
ET distortions come with two types of handiness: the director may wind either clockwise or anticlockwise as we progress radially outwards from the cylinder’s axis. Being both helicities equally energetic, they are seen with equal probability, and so either singular (point) defects (Davidson et al. 2015) or regular domain walls (Nayani et al. 2015) may arise where two ET domains with opposite chirality come together.

The issue as to whether ET distortions may or may not embody the ground state of chromonics is tackled in Sect. 2. There, we shall make contact with a notion of elastic frustration, which arises naturally from the geometric incompatibility of ET distortions. The latter were first described analytically by Burylov (1997); they exist as solutions to the pertinent Euler–Lagrange equations only when the uniform orientation along the cylinder’s axis ceases to be locally stable. We elaborate on this in Sect. 4, after having provided in Sect. 3 a general formula for the second variation of the classical Frank elastic free-energy functional for nematic liquid crystals. Finally, in Sect. 5, we collect our conclusions. The paper is closed by two Appendices: in the first we construct a dynamical analogy for the equilibrium director configurations in a capillary tube and give a phase space representation of ET distortions; in the second we record a number of accessory results that should assist the reader in navigating the main text.

2 Ground State

The classical Frank’s theory (Frank 1958) for low molecular weight nematics has been applied to chromonics as well. Frank’s theory is variational in nature; it is based on a celebrated formula for the elastic free-energy density (per unit volume) which measures the distortional cost \( W_F \) produced by a deviation from a uniform director field \( n \).

\[
W_F(n, \nabla n) := \frac{1}{2} K_{11} (\text{div} n)^2 + \frac{1}{2} K_{22} (n \cdot \text{curl} n)^2 + \frac{1}{2} K_{33} |n \times \text{curl} n|^2
\]

Lavrentovich 2003) for a quick, but effective introduction to this topic, and (Pišljar et al. 2022) for one of the latest most illuminating models). The name double twist was coined in Meiboom et al. (1981) (see also (Meiboom et al. 1983) for a fuller presentation of the elastic theory of blue phases), but the spatial arrangement of the director field that gives rise to it had already been precognized in Saupe (1969). The difference between ET and DT distortions rests simply on the fact that in the latter the lateral rotation of the director must be tuned appropriately, so that cylindrical tubes with axes at right angles can be glued together generating a network of disclinations.

Footnote 3 continued

For which there were antecedents in the works of Zocher (1933) and Osee (1933).
Here $K_{11}$, $K_{22}$, $K_{33}$, and $K_{24}$ are Frank’s elastic constants, material moduli characteristic of each liquid crystal. They are often referred to as the splay, twist, bend, and saddle-splay constants, respectively, by the features of different orientation fields, each with a distortion energy proportional to a single term in (1). While the elastic modes associated with $K_{11}$, $K_{22}$, and $K_{33}$ can be independently excited in the whole space, albeit not necessarily uniformly (that is, not with the same local energy everywhere), the saddle-splay mode driven by $K_{24}$ can only be discerned locally from the other elastic modes.\footnote{One can exhibit an orientation field whose energy density consists only in the $K_{24}$ term just at a point in space (see, for example, Chap. 5 of (Virga 1994)).}

Recently, Selinger (2018) has reinterpreted the classical Frank’s energy (1) by decomposing the saddle-splay mode into a set of other independent modes. The starting point of this decomposition is a novel representation of $\nabla n$ (see also (Machon and Alexander 2016)),

$$\nabla n = -b \otimes n + \frac{1}{2} T W(n) + \frac{1}{2} S P(n) + D,$$

where $b := -(\nabla n)n = n \times \text{curl } n$ is the bend vector, $T := \text{curl } n \cdot n$ is the twist, $S := \text{div } n$ is the splay, $W(n)$ is the skew-symmetric tensor that has $n$ as axial vector, $P(n) := I - n \otimes n$ is the projection onto the plane orthogonal to $n$, and $D$ is a symmetric tensor such that $Dn = 0$ and $\text{tr } D = 0$. By its own definition, $D \neq 0$ admits the following biaxial representation,

$$D = q(n_1 \otimes n_1 - n_2 \otimes n_2),$$

where $q > 0$ and $(n_1, n_2)$ is a pair of orthogonal unit vectors in the plane orthogonal to $n$, oriented so that $n = n_1 \times n_2$. It is argued in Selinger (2022) that $q$ should be given the name tetrahedral splay, to which we would actually prefer octupolar splay for the role played by a cubic (octupolar) potential on the unit sphere (Pedrini and Virga 2020) in representing all independent distortion characteristics $(S, T, b_1, b_2, q)$, but $T$. Here $b_1$ and $b_2$ are the components of the bend vector $b$ in the distortion frame $(n_1, n_2, n)$,

$$b = b_1 n_1 + b_2 n_2.$$  

By the use of the following identity,

$$2q^2 = \text{tr}(\nabla n)^2 + \frac{1}{2} T^2 - \frac{1}{2} S^2,$$

we can easily give Frank’s formula (1) the equivalent form

$$W_F(n, \nabla n) = \frac{1}{2} (K_{11} - K_{24}) S^2 + \frac{1}{2} (K_{22} - K_{24}) T^2 + \frac{1}{2} K_{33} B^2 + 2 K_{24} q^2,$$
where \( B^2 := b \cdot b \). Since \( (S, T, b, D) \) are all independent measures of distortion, it readily follows from (6) that \( W_F \) is positive semi-definite whenever
\[
K_{11} \geq K_{24} \geq 0, \quad K_{22} \geq K_{24} \geq 0, \quad K_{33} \geq 0,
\]
which are the celebrated Ericksen’s inequalities (Ericksen 1966). If these inequalities are satisfied in strict form, the global ground state of \( W_F \) is attained on the uniform director field, characterized by
\[
S = T = B = q = 0.
\]

More generally, it has been shown (Virga 2019) that besides (8) the only uniform distortions, that is, director fields that fill three-dimensional Euclidean space, having everywhere the same distortion characteristics, are only those for which
\[
S = 0, \quad T = \pm 2q, \quad b_1 = \pm b_2 = b,
\]

(9)
corresponding to Meyers’s heliconical distortions (Meyer 1976) characterizing the ground state of the twist–bend nematic phases identified experimentally in Cestari et al. (2011).\(^6\)

The experimental evidence gathered in Davidson et al. (2015) and Nayani et al. (2015) tells us that CLCs within a cylinder with degenerate planar anchoring on the lateral wall acquire either of ET distortions (see Fig. 1). This shows that the uniform distortion in (8) is not the ground state of chromonics, and neither are (9), as we shall show, thus entailing a degree of elastic frustration in the ground state.

For \( \mathcal{B} \) a region in space occupied by the material, the total elastic free energy stored in \( \mathcal{B} \) is given by the functional
\[
F_F[n] := \int_{\mathcal{B}} W_F(n, \nabla n) \, dV,
\]

(10)
where \( V \) is the volume measure. It is well known since the seminal paper of Ericksen (1962) that the saddle-splay term in \( W_F \) plays no role in the minimization of \( F_F \) in the class \( \mathcal{A}(n_0) \) of admissible distortions for which \( n \) is prescribed on the boundary \( \partial \mathcal{B} \),
\[
\mathcal{A}(n_0) := \left\{ n \in H^1(\mathcal{B}, \mathbb{S}^2) : n_0 \in H^1(\mathcal{B}, \mathbb{S}^2) \text{ is the trace of } n \text{ on } \partial \mathcal{B} \right\}.
\]

(12)
Formally \( \mathcal{A}(n_0) \) is defined as
\[
\mathcal{A}(n_0) := \left\{ n \in H^1(\mathcal{B}, \mathbb{S}^2) : n_0 \in H^1(\mathcal{B}, \mathbb{S}^2) \text{ is the trace of } n \text{ on } \partial \mathcal{B} \right\}.
\]

where \( H^1(\mathcal{B}, \mathbb{S}^2) \) is the closed subset of the Sobolev space of square integrable, \( \mathbb{R}^3 \)-valued functions subject (almost everywhere) to the constraint \( n \cdot n = 1 \).

\( ^6 \) In (9), \( q \) is positive and \( b \) arbitrary. As shown in Virga (2019), if \( q \) vanishes also does \( b \) and both forms of uniform distortions reduce to the standard uniform orientation in (8).
why $K_{24}$ is irrelevant for all $n \in \mathcal{A}(n_0)$ resides in the identity
\[ \text{tr}(\nabla n)^2 - (\text{div } n)^2 = \text{div } [(\nabla n)n - (\text{div } n)n], \tag{13} \]
which readily leads us to
\[ \int_B \left[ \text{tr}(\nabla n)^2 - (\text{div } n)^2 \right] \, dV = \int_{\partial B} \left[ (\nabla s n)n - (\text{div } s n)n \right] \cdot \nu \, dA \tag{14} \]
where $\nu$ is the outer unit normal to $\partial B$, $\nabla s$ and $\text{div } s$ denote the surface gradient and divergence (where only tangential derivatives on $\partial B$ are taken into account), and $A$ is the area measure.

As a consequence of (14), the saddle-splay energy contributes a constant to $F_F$, whose value is determined only by the boundary condition $n_0$, and so it is unable to affect the energy minimizers. This is why the saddle-splay energy is also called a null Lagrangian.

Besides the strong anchoring condition in (11), another boundary condition gives the $K_{24}$-energy a special form: this is the planar degenerate anchoring, for which,
\[ n \cdot \nu \equiv 0 \text{ on } \partial B. \tag{15} \]
In the latter case, as remarked in Koning et al. (2014), the $K_{24}$-integral can be rewritten as
\[ -K_{24} \int_{\partial B} \left( \kappa_1 n_1^2 + \kappa_2 n_2^2 \right) \, dA, \tag{16} \]
where $\kappa_1$ and $\kappa_2$ are the principal curvatures of $\partial B$, and $n_i$ are the components of $n$ along the corresponding principal directions of curvature.\footnote{We write the curvature tensor as $\nabla_s \nu = \kappa_1 e_1 \otimes e_1 + \kappa_2 e_2 \otimes e_2$, where $e_1$ and $e_2$ are unit vectors along the principal directions of curvature of $\partial B$.} It is clear from (16) that for $K_{24} > 0$, which is the strong form of (7), whenever (15) applies the saddle-splay energy would locally tend to orient $n$ on $\partial B$ along the direction of maximum (signed) curvature. For a region $B$ whose boundary $\partial B$ has bounded principal curvatures, the $K_{24}$-energy is then always finite, and so the following reduced Ericksen’s inequalities suffice to guarantee that $F_F$ is bounded below,
\[ K_{11} \geq 0, \quad K_{22} \geq 0, \quad K_{33} \geq 0, \tag{17} \]
see also (Long and Selinger 2022).

For $B$ a circular cylinder, we now describe the ET distortion that minimizes the free-energy functional $F_F$ subject to (15). Here, we essentially follow Burylov’s work.
Let $R$ be the radius of the cylinder $B$ and $L$ its height. We assume that in the frame $(e_r, e_\vartheta, e_z)$ of cylindrical coordinates $(r, \vartheta, z)$, with $e_z$ along the axis of $B$, $\mathbf{n}$ is represented as

$$ \mathbf{n} = \sin \beta(r) e_\vartheta + \cos \beta(r) e_z, \quad (18) $$

where the polar angle $\beta \in (-\pi, \pi)$, which $\mathbf{n}$ makes with the cylinder’s axis, depends only on the radial coordinate $r$. This approach is much in the spirit of Palais’ principle of symmetric criticality, which states that each critical symmetric point is also a symmetric critical point (Palais 1979).

Standard computations show that

$$ \nabla \mathbf{n} = -\frac{\sin \beta}{r} e_r \otimes e_\vartheta + \cos \beta' e_\vartheta \otimes e_r - \sin \beta' e_z \otimes e_r, \quad (19) $$

where a prime $'$ denotes differentiation with respect to $r$. As a result, the distortion characteristics $(S, T, q)$ of the director field (18) are given by

$$ S = 0, \quad (20a) $$

$$ T = \beta' + \frac{\cos \beta \sin \beta}{r}, \quad (20b) $$

$$ q = \frac{1}{2} \left| \beta' - \frac{\cos \beta \sin \beta}{r} \right|, \quad (20c) $$

the latter of which follows directly from (5). A direct computation also yields

$$ b = \frac{1}{r} \sin^2 \beta e_r. \quad (20d) $$

To extract from (20d) the remaining distortion characteristics $(b_1, b_2)$, we need to identify the distortion frame $(\mathbf{n}_1, \mathbf{n}_2, \mathbf{n})$ of a generic field in (18). Letting

$$ \mathbf{n}_\perp := e_r \times \mathbf{n} = \sin \beta e_z - \cos \beta e_\vartheta \quad (21) $$

and adopting the representation

$$ \mathbf{n}_1 = \cos \alpha \mathbf{n}_\perp + \sin \alpha e_r, \quad \mathbf{n}_2 = \cos \alpha e_r - \sin \alpha \mathbf{n}_\perp, \quad (22) $$

8 The same results arrived at in Burylov (1997) were independently reobtained in Davidson et al. (2015).

9 More technically worded, this principle requires a smooth functional $J$ over a Banach manifold $M$, of maps of a set $X$ into a manifold $Y$ to be invariant under the action of a compact Lie group $G$, whose set of equivariant sections of $M$ we denote $\Sigma$. A theorem of Palais (p. 22, Palais 1979) (see also (Kobayashi and Ôtani 2004)) says that $\Sigma$ is a smooth submanifold of $M$ and if $u \in \Sigma$ is a critical point of $J|_\Sigma$ then $u$ is a critical point of $J$. In our case, $X = B$, $Y = S^2$, $G$ is the group $SO(e_z)$ of all rotations about $e_z$ (which also leaves (15) unchanged), $J$ is $\mathcal{F}_r$, and $\Sigma$ is the set of mappings represented by (18). Of course, the principle of symmetric criticality does not say that all critical points are symmetric.
so that by (20d)

\[ b_1 = \frac{1}{r} \sin^2 \beta \sin \alpha \quad \text{and} \quad b_2 = \frac{1}{r} \sin^2 \beta \cos \alpha, \]

we readily obtain from (3) and (20c), with the aid of (21) and (22), that

\[ \alpha = \text{sgn} \left( \frac{1}{r} \sin \beta \cos \beta - \beta' \right) \frac{\pi}{4}. \]  

(24)

By changing the variable \( r \) into

\[ \rho := \frac{r}{R}, \]

which ranges in \([0, 1]\), and making use of (20) in (6), we arrive at the following reduced functional, \( \mathcal{F}[\beta] \), which is an appropriate dimensionless form of Frank’s free-energy functional,

\[
\mathcal{F}[\beta] := \frac{\mathcal{F}[n]}{2\pi K_{22}L} = \int_0^1 \left( \frac{\rho \beta'^2}{2} + \frac{1}{2\rho} \cos^2 \beta \sin^2 \beta + \frac{k_3}{2\rho} \sin^4 \beta \right) d\rho \\
+ \frac{1}{2} (1 - 2k_{24}) \sin^2 \beta (1),
\]

(26)

where the boundary term clearly echoes\(^{10}\) (16) and the following scaled elastic constants have been introduced,

\[ k_3 := \frac{K_{33}}{K_{22}}, \quad k_{24} := \frac{K_{24}}{K_{22}}, \quad \text{with} \quad K_{22} > 0. \]  

(27)

We defer the reader to Appendix A for a reinterpretation of the functional \( \mathcal{F} \) in (26) as the action of an effective dynamical system. For the integral in (26) to be convergent, \( \beta \) must be subject to the condition

\[ \beta (0) = 0, \]  

(28)

which amounts to require that \( n \) is along \( e_z \) on the cylinder’s axis.\(^{11}\) We seek the functions \( \beta = \beta (\rho) \) of class \( C^2 \) on \([0, 1] \) that satisfy (28) and make (26) stationary.

The Euler–Lagrange equations for the functional \( \mathcal{F} \) read as

\[
\cos \beta \sin \beta \left[ 1 + 2(k_3 - 1) \sin^2 \beta \right] - \beta' - \rho \beta'' = 0, \quad \rho \in (0, 1),
\]

(29a)

\(^{10}\) To be more precise, an additional boundary term besides the \( K_{24} \)-energy arises in (26) from an integration by parts of a component of the twist-energy.

\(^{11}\) Actually, the convergence requirement would also be satisfied by enforcing the more general condition \( \sin \beta (0) = 0 \); however, our choice is not restrictive, it rather rests on the nematic symmetry, for which \( n \) and \( -n \) are physically equivalent.
subject to (28). Clearly, the constant $\beta \equiv 0$ is a trivial solution to these equations. Two other nonuniform solutions could also exist, depending on the value of $k_{24}$. To show this, we multiply both sides of (29a) by $\rho \beta'$ and easily see that this equation has a first integral,

$$
(\rho \beta')^2 - \sin^2 \beta \left( \cos^2 \beta + k_3 \sin^2 \beta \right) = c,
$$

where $c$ is an arbitrary constant. The regularity assumption entails that $|\beta'(0)| < \infty$, and so $c$ must vanish for (28) to be valid. Two branches of solution thus emanate from $\rho = 0$, depending on whether $\beta'(0)$ is positive or negative: they are one opposite to the other and both are obtained by integrating the equation

$$
\beta' = \frac{\sin \beta \cos \sqrt{1 + k_3 \tan^2 \beta}}{\rho}.
$$

We shall focus on the increasing (positive) branch. Evaluating (31) for $\rho = 1$ and making use of it into the boundary condition (29b), we obtain the following equation for $\beta(1)$,

$$
\sin \beta(1) \cos \beta(1) \left[ \sqrt{1 + k_3 \tan^2 \beta(1)} + (1 - 2k_{24}) \right] = 0,
$$

which admits a non-vanishing solution only for $k_{24} > 1$; that is,

$$
\beta(1) = \beta_1 := \arctan \left( \frac{2\sqrt{k_{24}(k_{24} - 1)}}{\sqrt{k_3}} \right).
$$

Solving (31) by quadrature yields

$$
\ln \rho = \int \frac{d\beta}{\sin \beta \cos \beta \sqrt{1 + k_3 \tan^2 \beta}} = \frac{1}{2} \ln g(\beta) + C,
$$

where

$$
g(\beta) := \frac{1}{2} - \frac{1}{\sqrt{1 + k_3 \tan^2 \beta} + 1}
$$

and $C$ is an arbitrary constant to be determined so as to satisfy (33). With a different constant $C$, we can rewrite (34) as

$$
\rho^2 = C \left( 1 - \frac{2}{\sqrt{1 + k_3 \tan^2 \beta} + 1} \right),
$$

which is valid for all $\rho \in [0, 1]$ only if $C > 1$. By requiring that (36) agrees with (33), we determine $C$ and arrive at the following solution

$$
\beta_{ET}(\rho) := \arctan \left( \frac{2\sqrt{k_{24}(k_{24} - 1)} \rho}{\sqrt{k_3} \left[ k_{24} - (k_{24} - 1) \rho^2 \right]} \right),
$$
Fig. 2 Graphs of the function $\beta_{ET}$ against $\rho$ for different values of the elastic parameters $k_{24}$ and $k_3$, increasing according to the orientation of the arrow. The twist angle is enhanced for increasing $k_{24}$ and depressed for increasing $k_3$, which together with its opposite $-\beta_{ET}$ represent the two variants of the ET distortion (with opposite chiralities). The function $\beta_{ET}$ is plotted against $\rho$ in Fig. 2, which illustrates the antagonistic roles played by the elastic parameters $k_{24}$ and $k_3$. Upon increasing the saddle-splay constant $K_{24}$, the effective anchoring energy (16) becomes stronger and drives $n$ closer to the circles on the boundary of the cylinder along which the curvature is the largest. This tendency is counteracted by an increase in the bending constant $K_{33}$, which drives $n$ closer to the cylinder’s axis $e_z$ to reduce the bending content of the elastic energy.

By the use of both (30) and (34) in (26), we express $F[\beta_{ET}]$ in the form,

$$F_{ET} := F[\beta_{ET}] = \int_0^{\beta_1} \sin \beta \sqrt{\cos^2 \beta + k_3 \sin^2 \beta} \, d\beta + \frac{1}{2} (1 - k_{24}) \sin^2 \beta_1,$$

which by (33) can be written explicitly in terms of the reduced elastic constants only,

$$F_{ET} = \begin{cases} 
1 - k_{24} + \frac{1}{2} \frac{k_3}{\sqrt{1 - k_3}} \arctanh \left( \frac{2\sqrt{1-k_3}(k_{24}-1)}{k_3+2(k_{24}-1)} \right), & k_3 \leq 1, \\
1 - k_{24} + \frac{1}{2} \frac{k_3}{\sqrt{k_3-1}} \arctan \left( \frac{2\sqrt{k_3-1}(k_{24}-1)}{k_3+2(k_{24}-1)} \right), & k_3 \geq 1,
\end{cases}$$

an expression that, for $k_{24} > 1$, can be shown to be negative in both instances, as the following inequalities hold true,

$$1 - k_{24} \leq F_{ET} \leq - \frac{2(k_{24} - 1)^2}{2k_{24} - 1} < 0,$$

for $0 \leq k_3 \leq 1$. \footnote{That is, the decreasing solution branch of (31).}
\[
- \frac{(2k_{24} - 1)^2}{2k_{24} - 1} \leq \mathcal{F}_{\text{ET}} < 0, \quad \text{for } k_3 \geq 1. \tag{41}
\]

\(\mathcal{F}_{\text{ET}}\), as given by (39) as a function of \(k_3\), is continuous along with its derivatives at \(k_3 = 1\).\(^{13}\) Moreover, the same formula is also valid for the energy of the mirror image \(-\beta_{\text{ET}}\) of \(\beta_{\text{ET}}\). Thus, whenever the ET distortion is permitted, that is, for \(k_{24} > 1\), it possesses less elastic free energy than the uniform alignment \(n \equiv e_z\), and so it becomes eligible for the ground state of CLCs, at least within the cylindrical confinement investigated experimentally.

The price to pay to model mathematically the experimental observations with the ET distortion is to renounce one of Ericksen’s inequalities, thus accepting that Frank’s functional in (10) may be unbounded below, jeopardizing in general its coercivity. This has no noxious consequences whenever the degenerate boundary condition (15) is prescribed, as the integral in (16) is bounded below and the remaining terms in (1) are well-behaved, provided that the constants \(K_{11}, K_{22},\) and \(K_{33}\) are all positive. Instances are known in the literature where appropriate boundary conditions salvage a functional that in other, more general circumstances would fail to attain its minimum (see, for example, (Day and Zarnescu 2019)). Here a similar situation arises with the complicity of cylindrical symmetry. Fearing, however, a potentially latent pathology emerging from the lack of boundedness in the general setting, we find it especially appropriate to study the local stability of the ET distortion, albeit against perturbations that do not break cylindrical symmetry.

To this end, in the following section we shall derive a general formula for the second variation in \(\mathcal{F}_{\text{T}}\). Before that, we pause to illuminate further the ET field in terms of the distortion characteristics. By the use of (31) in (20), (23), and (24), we see that (for the positive branch)

\[
T = \frac{1}{r} \sin \beta \cos \beta (\sqrt{1 + \tan^2 \beta} + 1), \tag{42a}
\]

\[
q = \frac{1}{2r} |\sin \beta| \cos \beta (\sqrt{1 + \tan^2 \beta} - 1), \tag{42b}
\]

\[
\alpha = -\frac{\pi}{4}, \quad b_1 = -\frac{1}{\sqrt{2r}} \sin^2 \beta = -b_2, \tag{42c}
\]

where \(\beta = \beta_{\text{ET}}(\rho)\) as in (37) (with \(r = \rho R\)), whereas for \(\beta = -\beta_{\text{ET}}(\rho)\) (the negative branch), \(T\) changes its sign, as do \(\alpha, b_1,\) and \(b_2\), while \(q\) remains unchanged. Although the characteristics in (42) are all different from zero for \(r > 0\), in the limit as \(r \to 0\) they yield

\[
S = q = b_1 = b_2 = 0, \quad T = \frac{4\sqrt{k_{24} - 1}}{R \sqrt{k_3 k_{24}}}, \tag{43}
\]

suggesting that each ET distortion has a single non-vanishing characteristic only along the cylinder’s axis. Since such a single distortion mode is unable to fill space, it can be accommodated within the cylinder only by entraining along all other modes. We

\(^{13}\) Apart from a different scaling of the constant \(K_{24}\), the formula in (39) for \(k_3 \geq 1\) coincides with equation (5) of (Davidson et al. 2015).
may interpret such a lack of uniformity as resulting from an elastic frustration induced by cylindrical confinement, whose length scale appears explicitly in (43) through the radius $R$.

3 Second Variation

To compute the second variation in Frank’s elastic free-energy functional, we devise here a variant to the classical method: \(^\text{14}\) we represent a generic perturbation of a given director field $n$ via a path $n_t(x)$ on the unit sphere $S^2$ parameterized in $t \in (-\varepsilon, \varepsilon)$ and going through the point $n(x)$. The mapping $t \mapsto n_t(x)$ defines for each $x \in B$ a trajectory in $S^2$, which for $t = 0$ satisfies $n_0(x) = n(x)$. Thus, as the virtual time $t$ spans $(-\varepsilon, \varepsilon)$, the vector fields $n_t$ on $B$ are instantaneous realizations of diverse perturbations of $n$. The constraint of unimodularity for $n_t$ must be valid for all $t$, and so,

$$\dot{n}_t \cdot n_t = 0,$$

where a superimposed dot denotes differentiation with respect to $t$. We define the field

$$v := \dot{n}_t|_{t=0},$$

which by (44) is orthogonal to $n$,

$$v \cdot n = 0. \quad (46)$$

By further differentiating both sides of (44) with respect to $t$, we arrive at

$$\ddot{n}_t \cdot n_t + \dot{n}_t \cdot \dot{n}_t = 0. \quad (47)$$

The field $\ddot{n}_t|_{t=0}$ represents a second-order perturbation for $n$. It follows from (47) and (45) that

$$\ddot{n}_t|_{t=0} = w - v^2 n,$$

where $w$, precisely like $v$, is an arbitrary vector field orthogonal to $n$, and $v^2 = v \cdot v$. We say that $v$ and $w$ are the first- and second-order outer variations of $n$.

The first variation of $F_F$ at the field $n$ is a linear functional of $v$ defined as

$$\delta F_F[n][v] := \frac{d}{dt} F_F[n_t]|_{t=0} = \int_B \dot{W}_F(n_t, \nabla n_t)|_{t=0} dV. \quad (49)$$

An equilibrium director field $n$ makes $\delta F_F(n)[v]$ vanish for all $v$ satisfying (46). With the aid of (45) and (B2), we obtain that

$$\delta F_F[n][v] := \int_B \left\{ K_{11} \text{ div } n \text{ div } v + K_{22} n \cdot \text{ curl } n \cdot (v \cdot \text{ curl } n + n \cdot \text{ curl } v) \ight. \right.$$  

$$+ K_{33} (n \times \text{ curl } n) \cdot (v \times \text{ curl } n + n \times \text{ curl } v) \right\}$$

\(^\text{14}\) See also (Rosso et al. 2004; Gartland 2021) for allied methods leading to similar outcomes.
\[ + 2K_{24} \left( \text{tr}(\nabla n \nabla v) - \text{div} \ n \ \text{div} \ v \right) \} \ d V \]  

(50)

with \( v \) sufficiently regular.\(^{15}\)

Similarly, the second variation of \( \mathcal{F}_F \) at the field \( n \) is the functional defined by

\[ \delta^2 \mathcal{F}_F(n)[v, w] := \delta^2 \mathcal{F}_F(n)[w] \]

\[ + \int_{\mathcal{B}} \left\{ \left( K_{11} - 2K_{24} \right) \left[ (\text{div} \ v)^2 - v^2 (\text{div} \ n)^2 - (\text{div} \ n) \ n \cdot \nabla v^2 \right] \right. \]

\[ + K_{22} \left[ (v \cdot \text{curl} \ n + n \cdot \text{curl} \ v)^2 \right. \]

\[ + 2(n \cdot \text{curl} \ n)(v \cdot \text{curl} \ v - v^2 n \cdot \text{curl} \ n) \]

\[ + K_{33} \left[ (n \times \text{curl} \ n + n \times \text{curl} \ v)^2 \right. \]

\[ + (n \times \text{curl} \ n) \cdot (2v \times \text{curl} \ v - 2v^2 n \times \text{curl} \ n - \nabla v^2) \]

\[ + 2K_{24} \left[ \text{tr} (\nabla v)^2 - v^2 \text{tr} (\nabla n)^2 + n \times \text{curl} \ n \cdot \nabla v^2 \right] \} \ d V, \]

(52)

which reduces to only its quadratic component in \( v \) whenever \( n \) is an equilibrium field. In this latter case, with a slight abuse of notation, we shall simply set \( \delta^2 \mathcal{F}_F(n)[v] := \delta^2 \mathcal{F}_F(n)[v, 0] \).

### 3.1 Simple Applications

To put to the test the formula for the second variation of \( \mathcal{F}_F \) in (52), here we compute it for two special equilibrium fields.\(^{16}\)

#### 3.1.1 Uniform Field

We begin with a director field \( n \) uniformly constant in space and see how its stability is related to Ericksen’s inequalities (7). With no loss of generality, for a given Cartesian frame \( (e_1, e_2, e_3) \), we apply (52) to the director field \( n \equiv e_3 \),

\[ \delta^2 \mathcal{F}_F(e_3)[v] = \int_{\mathcal{B}} \left( (K_{11} - 2K_{24}) (\text{div} \ v)^2 \right. \]

\[ + \left. 2K_{24} \left( \text{tr}(\nabla n \nabla v) - \text{div} \ n \ \text{div} \ v \right) \right\} \ d V. \]

---

\(^{15}\) The reader is deferred to Appendix B for a number of ancillary results used here.

\(^{16}\) Which are actually both universal equilibrium fields, as they solve the equilibrium equations for any frame-indifferent elastic free-energy density \( W(n, \nabla n) \), as shown in Ericksen (1967).
\[ + K_{22} (\mathbf{n} \cdot \text{curl} \, \mathbf{v})^2 + K_{33} |\mathbf{n} \times \text{curl} \, \mathbf{v}|^2 + 2K_{24} \text{tr} (\nabla \mathbf{v})^2 \} \, d \, V, \]

(53)

where \( \mathbf{v} \cdot \mathbf{e}_3 \equiv 0 \). It follows from this latter identity that

\[ \nabla \mathbf{v} := \mathbf{e}_1 \otimes \mathbf{a} + \mathbf{e}_2 \otimes \mathbf{c}, \]

(54)

with \( \mathbf{a} \) and \( \mathbf{c} \) arbitrary vector fields. Letting \( a_i \) and \( c_i \) be the components of \( \mathbf{a} \) and \( \mathbf{c} \) in the frame \( (\mathbf{e}_1, \mathbf{e}_2, \mathbf{e}_3) \), we easily compute

\begin{align*}
\text{div} \, \mathbf{v} &= a_1 + c_2, \\
\mathbf{e}_3 \cdot \text{curl} \, \mathbf{v} &= -a_3 e_1 - c_3 e_2, \\
\mathbf{e}_3 \times \text{curl} \, \mathbf{v} &= -a_2 e_1 - c_2 e_3, \\
\text{tr} (\nabla \mathbf{v})^2 &= a_1^2 + 2a_2 c_1 + c_2^2,
\end{align*}

(55)

which give (53) the following form,

\[ \delta^2 F_F(\mathbf{e}_3)[\mathbf{v}] = \int_{\mathcal{B}} \left\{ K_{11} \left( a_1^2 + c_2^2 \right) + 2 \left( K_{11} - 2K_{24} \right) a_1 c_2 + K_{22} \left( a_2^2 + c_1^2 \right) \\
- 2\left( K_{22} - 2K_{24} \right) a_2 c_1 + K_{33} \left( a_3^2 + c_3^2 \right) \right\} \, d \, V. \]

(56)

For a general domain \( \mathcal{B} \) with no boundary conditions for \( \mathbf{v} \), the integrand on the right-hand side of (56) is the sum of three independent quadratic forms, so that \( \delta^2 F_F(\mathbf{e}_3)[\mathbf{v}] \) is positive whenever all these forms are not negative, which is precisely when Ericksen’s inequalities (7) are satisfied.

### 3.1.2 Radial Hedgehog

The radial hedgehog is the director field that in the frame \( (\mathbf{e}_r, \mathbf{e}_\vartheta, \mathbf{e}_\phi) \) of spherical coordinates is represented as \( \mathbf{n} = \mathbf{e}_r \). Standard computations give \( \text{curl} \, \mathbf{e}_r = 0 \), \( \text{div} \, \mathbf{e}_r = 2/r \), and \( \text{tr}(\nabla \mathbf{e}_r)^2 = 2/r^2 \), so that (52) reduces to

\[ \delta^2 F_F(\mathbf{e}_r)[\mathbf{v}] = \int_{\mathcal{B}} \left\{ (K_{11} - 2K_{24}) \left[ (\text{div} \, \mathbf{v})^2 - \frac{2}{r^2} v^2 \right] + K_{22} (\mathbf{e}_r \cdot \text{curl} \, \mathbf{v}) \\
+ K_{33} |\mathbf{n} \times \text{curl} \, \mathbf{v}|^2 + 2K_{24} \left[ \text{tr} (\nabla \mathbf{v})^2 - \frac{2}{r^2} v^2 \right] \right\} \, d \, V. \]

(57)

For \( \mathcal{B} \) a sphere with center in the origin of the frame \( (\mathbf{e}_r, \mathbf{e}_\vartheta, \mathbf{e}_\phi) \) and under the strong anchoring condition \( \mathbf{v}|_{\partial \mathcal{B}} = 0 \), this formula coincides with equation (2.3) of (Kinderlehrer and Ou 1992), where the second variation (57) was proved to be positive in the class of perturbations \( \mathbf{v} \in H^1(B, \mathbb{R}^3) \cap L^\infty(B, \mathbb{R}^3) \) with compact support and satisfying \( \mathbf{v} \cdot \mathbf{e}_r = 0 \), whenever the following inequality is satisfied,

\[ 8 (K_{22} - K_{11}) + K_{33} \geq 0. \]

(58)
4 Local Stability

In this section, we study the local stability of the field $n_{ET}$, delivered by (18) when $\beta = \beta_{EF}$ as in (37). We shall focus our attention on this chiral variant of the ET field, as that with opposite chirality, represented by $\beta = -\beta_{ET}$, has by symmetry the same energy and the same stability character.

In the scaled radial coordinate $\rho$ introduced in (25), the perturbation field $v$, subject to $n_{ET} \cdot v = 0$, is represented in the frame $(e_r, e_\theta, e_z)$ of cylindrical coordinates as

$$v := f(\rho)e_r - g(\rho) \cos \beta_{ET}(\rho)e_\theta + g(\rho) \sin \beta_{ET}(\rho)e_z,$$

(59)

where $f$ and $g$ are absolutely continuous functions on $[0, 1]$. Here, we resort again to Palais’ principle of symmetric criticality recalled above, as we aim at proving that within the class of axisymmetric perturbations all critical values of $\delta^2 \mathcal{F}$ are positive. We shall actually prove that $\delta^2 \mathcal{F} > 0$ in this symmetric class. A stability result in a more general class of perturbations is presently out of our reach.

The validity of the degenerate planar anchoring condition (15) for the perturbed director field requires that

$$f(1) = 0,$$

(60)

while $g(1)$ remains completely arbitrary. Moreover, for the regularity of $v$ along the cylinder’s axis (and the integrability of the perturbed energy), we also request that

$$f(0) = g(0) = 0.$$

(61)

Making use in B of the identities (B5) recorded in Appendix B, we arrive at the following dimensionless form of the second variation $\delta^2 \mathcal{F}(n_{ET})[v]$ in the class of perturbation fields described by (59),

$$\delta^2 \mathcal{F}[f, g] := \frac{\delta^2 \mathcal{F}(n_{ET})[v]}{2\pi L K_{22}}$$

$$= 2 \int_0^1 \left\{ \left[ -2 \cos^2 \beta \sin^2 \beta + \frac{(-\sin^2 \beta + \cos^2 \beta)^2}{2} \right.ight.$$ 

$$+ k_3 (-\sin^2 \beta + 3 \cos^2 \beta) \sin^2 \beta \left[ \frac{g^2}{\rho} + \frac{\rho g'^2}{2} \right] \right\} d\rho$$

$$+ (1 - 2k_{24}) (-\sin^2 \beta + \cos^2 \beta) g^2(1)$$

$$+ \int_0^1 \left\{ \left[ k_1 - 2(\rho \beta' + \cos \beta \sin \beta)^2 \right. \right.$$ 

$$\left. + k_3 (\rho^2 \beta'^2 + \sin^2 \beta + 4 \cos \beta \sin \beta \beta' - 2 \sin^4 \beta) \right] \frac{f^2}{\rho} + k_1 \rho f'^2 \right\} d\rho,$$

(62)

where

$$k_1 := \frac{K_{11}}{K_{22}},$$

(63)

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while $k_3$ and $k_{24}$ are as in (27), and $\beta = \beta_{EF}$ as in (37). Integrations by parts have been performed in (62) by employing both (60) and (61).

In (62), the scalar perturbations $f$ and $g$ are decoupled and $\delta^2 F[f, g]$ can be regarded as the sum of two independent functionals. When $f = 0$ and $g \neq 0$, we say that $v$ is an azimuthal perturbation. On the contrary, when $f \neq 0$ and $g = 0$, we say that $v$ is a radial perturbation. We shall consider these perturbations separately below; the local stability of the field $n_{ET}$ requires that $\delta^2 F[f, g]$ be positive for both types of perturbation.

Our study will be limited to the case $k_3 \geq 1$, which according to the experimental measurements of (Zhou et al. 2012) and (Zhou et al. 2014) is the only relevant to chromonics, the materials of interest to us in this work.

4.1 Azimuthal Perturbations

To compute $\delta^2 F[0, g]$, we take advantage of the fact that $\beta_{ET}$ is a monotonically increasing function of $\rho$, ranging in the interval $[0, \beta_1]$, with $\beta_1 < \pi$ as in (33), as $\rho$ ranges in $[0, 1]$. This allows us to express $g$ as a function of $\beta$, and ultimately as a function, $U(u)$, of the variable

$$u := \frac{\sin \beta}{A},$$

which ranges in $[0, 1]$, as $A$ is defined by

$$A := \sin \beta_1 = \frac{2\sqrt{k_{24}(k_{24} - 1)}}{\sqrt{k_3 + 4k_{24}(k_{24} - 1)}}. \tag{65}$$

This cascade of changes of variables is characterized by the following relations involving their differentials,

$$\frac{d \rho}{\rho} = \frac{d \beta}{\sin \beta \sqrt{\cos^2 \beta + k_3 \sin^2 \beta}} = \frac{d u}{u \sqrt{1 - (Au)^2} \sqrt{1 + (k_3 - 1)(Au)^2}}. \tag{66}$$

Their use gives $\delta^2 F$ the following, more compact form,

$$\delta^2 F[U] = \int_0^1 \left[ \frac{\phi(k_3, A, u)}{\gamma(k_3, A, u)} U(u)^2 + \gamma(k_3, A, u) U'(u)^2 \right] du + q_0(k_3, A) U(1)^2, \tag{67}$$

where a prime now denotes differentiation with respect to $u$, we have set

$$\phi(k_3, A, u) := \left(1 - 2(Au)^2\right) \left(1 + 2(k_3 - 1)(Au)^2\right) + 4(k_3 - 1)(1 - (Au)^2)(Au)^2, \tag{68a}$$

$$\gamma(k_3, A, u) := u \sqrt{1 - (Au)^2} \sqrt{1 + (k_3 - 1)(Au)^2}, \tag{68b}$$

$$q_0(k_3, A) := - \frac{(1 - 2A^2) \sqrt{1 + (k_3 - 1)A^2}}{\sqrt{1 - A^2}}. \tag{68c}$$
and $U$ belongs to the admissible class

$$\mathcal{A} := \{ U \in AC[0, 1] : U(0) = 0 \}. \quad (69)$$

The elastic constants enter (67) through the pair of dimensionless parameters $(k_3, A)$. It follows from (65) that $A$ ranges in $(0, 1)$ for all $k_{24} \in (1, \infty)$ and $k_3 \in [1, \infty)$. Conversely, $k_{24}$ is expressed in terms of $(k_3, A)$ by

$$k_{24} = \frac{\sqrt{1 - A^2} + \sqrt{1 + (k_3 - 1)A^2}}{2\sqrt{1 - A^2}}, \quad (70)$$

so that the correspondence between the half-space $H := \{(k_3, k_{24}) : k_3 \in [1, \infty), k_{24} \in (1, \infty)\}$ and the strip $S := \{(k_3, A) : k_3 \in [1, \infty), k_{24} \in (1, \infty)\}$ is one-to-one as the Jacobian determinant of the transformation is positive,

$$\frac{\partial(k_3, k_{24})}{\partial(k_3, A)} = \frac{k_3}{2} \frac{A}{(1 - A^2)^{3/2}\sqrt{1 + (k_3 - 1)A^2}} > 0. \quad (71)$$

Here we show that for all $(k_3, A) \in S$ the functional in (67) is positive definite in the class $\mathcal{A}$. Our desired conclusion will be reached in two steps. First, we see that the functions

$$\tilde{U} := \sqrt{\gamma} U, \quad Q(u, \tilde{U}) := 1, \quad G(u, \tilde{U}) := \frac{3}{4} \frac{\gamma'}{\gamma} \tilde{U}^2 = \frac{3}{4} \gamma' U^2, \quad (72)$$

obey all three conditions of Lemma 1 in Appendix B for $U \in \mathcal{A}$ and $\gamma$ as in (68b). Thus, it follows from (B6) that

$$\int_0^1 \left[ (\sqrt{\gamma} U)' \right]^2 \, du \geq - \int_0^1 \left( \frac{3}{4} \frac{\gamma'^2}{\gamma} + \frac{3}{2} \gamma'' \right) U^2 \, du + \frac{3}{2} |\gamma'|_{u=1} U(1)^2. \quad (73)$$

Second, from the identity

$$[\sqrt{\gamma} U]' = \frac{\gamma'}{2\sqrt{\gamma}} U + \sqrt{\gamma} U', \quad (74)$$

we readily obtain that

$$\int_0^1 \gamma U'^2 \, du = \int_0^1 \left\{ \left[ (\sqrt{\gamma} U)' \right]^2 - \frac{\gamma'^2}{4\gamma} U^2 - \gamma' U U' \right\} \, du. \quad (75)$$

Making use of (73) in (75) and integrating by parts, we finally arrive at

$$\int_0^1 \gamma U'^2 \, du \geq \int_0^1 \left( - \frac{\gamma'^2}{\gamma} - \gamma'' \right) U^2 \, du + \gamma'|_{u=1} U(1)^2. \quad (76)$$
We are now in a position to conclude our proof, as combining (76) with (67) we estimate
\[
\delta^2 \mathcal{F}[U] \geq \int_0^1 \left( \frac{\phi}{\gamma} - \frac{\gamma'^2}{\gamma} - \gamma'' \right) U(u)^2 \, du + (q_0 + \gamma'|_{u=1}) \, U(1)^2,
\]
(77)
and so, for \( \delta^2 \mathcal{F} \) to be positive on \( \mathcal{A} \) it suffices that the following inequalities are valid
\[
\frac{\phi}{\gamma} - \frac{\gamma'^2}{\gamma} - \gamma'' \geq 0, \quad q_0 + \gamma'|_{u=1} \geq 0,
\]
(78)
which by (68) reduce to
\[
\frac{7A^2 u}{\sqrt{1 + (k_3 - 1)(Au)^2} \sqrt{1 - (Au)^2}} \left[ \frac{4}{7} + (k_3 - 1)(Au)^2 \right] \geq 0,
\]
\[
\frac{A^2 (1 - A^2)}{\sqrt{1 + (k_3 - 1)(Au)^2} \sqrt{1 - (Au)^2}} (k_3 - 1) \geq 0,
\]
(79)
and are easily seen to be valid for all \( u \in [0, 1] \) and \((k_3, A) \in S\).

4.2 Radial Perturbations

Now, we ignite the radial modes in the second variation \( \delta^2 \mathcal{F} \) in (62), while silencing the azimuthal ones,
\[
\delta^2 \mathcal{F}[f, 0] = \int_0^1 \left\{ k_1 - 2 \left( \rho \beta' + \cos \beta \sin \beta \right)^2 \right. \\
\left. + k_3 \left( \rho^2 \beta'' + \sin^2 \beta + 4 \cos \beta \sin \beta \beta' - 2 \sin^4 \beta \right) \right\} \frac{f^2}{\rho} + k_1 \rho f'^2 \, d \rho,
\]
(80)
where \( \beta = \beta_{ET} \) and \( f \) is subject to (61) and (60). Here, as suggested by the experimental evidence presented in Zhou et al. (2012, 2014), we assume that for chromonics \( k_1 \geq 1 \). This inequality, combined with the same changes of variables performed in Sect. 4.1, leads us to replace (67) with
\[
\delta^2 \mathcal{F}[U] \geq \int_0^1 \left[ \frac{\psi(k_3, A, u)}{\gamma(k_3, A, u)} U(u)^2 + \gamma(k_3, A, u) U'(u)^2 \right] du,
\]
(81)
where now
\[
\psi(k_3, A, u) := 1 + 2(k_3 - 2)(Au)^2 + (k_3 - 4)(k_3 - 1)(Au)^4 \\
+ 4(k_3 - 1)(Au)^2 \sqrt{1 - (Au)^2} \sqrt{1 + (k_3 - 1)(Au)^2}
\]
and \( U \in \mathcal{A} \) is subject to the additional condition that \( U(1) = 0 \).
Following the same lines of thought that in Sect. 4.1 established the lower bound (77), here we arrive at

\[ \delta^2 \mathcal{F}[U] \geq \int_0^1 \left( \frac{\psi}{\gamma} - \frac{\gamma'^2}{\gamma} - \gamma'' \right) U(u)^2 \, du \]  

(83)

and the following inequality suffices to render \( \delta^2 \mathcal{F} \) positive,

\[ \frac{\psi}{\gamma} - \frac{\gamma'^2}{\gamma} - \gamma'' = \frac{4A^2 u}{\sqrt{1 - (Au)^2}} \frac{\sqrt{1 + (k_3 - 1)(Au)^2}}{\sqrt{1 - (Au)^2}} 
\times \left[ (k_3 - 1) \sqrt{1 - (Au)^2} \sqrt{1 + (k_3 - 1)(Au)^2} 
+ \frac{1}{4} (Au)^2 (k_3 + 11)(k_3 - 1) - k_3 + 2 \right] \geq 0, \]  

(84)

which is indeed satisfied for all \( u \in [0, 1] \) and \( (k_3, A) \in \mathcal{S} \).

We thus conclude that the ET fields are locally stable, despite the violation of one Ericksen’s inequality.\(^\text{17}\) Here we considered only the case \( k_3 \geq 1 \); for completeness, the local stability of the ET fields has also been established in the case \( 0 < k_3 \leq 1 \) by use of a spectral method (Paparini 2022).

5 Conclusions

In this article, we addressed the problem posed by the peculiar ground state exhibited by chromonic liquid crystals. These, unlike ordinary nematic liquid crystals when confined within capillary tubes with degenerate boundary conditions do not acquire the uniform alignment with the director oriented along the cylinder’s axis, but develop a spontaneous twist, equally likely to have opposite chiralities. A commonly accepted explanation for such a behavior is that these materials have a twist elastic constant \( K_{22} \) smaller than the saddle-splay constant \( K_{24} \) of Frank’s elastic theory, a fact which has been confirmed by a number of experiments. The problem with this explanation is that assuming \( K_{22} < K_{24} \) violates one of Ericksen’s inequalities, which guarantee that Frank’s elastic free energy is bounded below.

Very recently, such a violation has also been investigated in Long and Selinger (2022), which proposed that the pure (double) twist mode that would characterize the ground state of chromonics,\(^\text{18}\) being non-uniform and so unable to fill space (Virga 2019) prompts the excitation of other elastic modes whose positive cost counterbalances the divergence to negative infinity of the total free energy. Here, concerned as we were by tackling a variational problem with indefinite energy, we gave a different, but complementary explanation. We showed that the boundary conditions prescribed on capillary tubes are indeed responsible for taming the unboundedness of the energy.

\(^{17}\) A violation nonetheless necessary for these fields to be equilibrium solutions.

\(^{18}\) With only \( T \neq 0 \).
and securing a solution to the variational problem in an admissible class of distortions with cylindrical symmetry.

The issue about the stability of such a solution then remained open, a question that was not idle to ask, given the wildness of the parent energy. To resolve this issue, we derived a general formula for the second variation of Frank’s elastic free-energy functional and applied it to the study of the (local) stability of the twisted ground state of chromonics. We concluded that this is stable.

The role played by boundary conditions in stabilizing the ground state of chromonics within the classical elastic theory of Frank poses at least two further questions. First, what is the most general class of anchoring conditions capable in rigid containers of preventing the energy from diverging to negative infinity? Second, and more subtly, would free boundaries, such as the ones arising in the equilibrium problem for a chromonic drop surrounded by its isotropic melt, keep the energy bounded below?

We have not tackled the first question. As for the second, preliminary explorations (Paparini 2022) have shown that the violation of Ericksen’s inequalities for either the twist or splay constants may result in a paradoxical disintegration process, which poses a serious threat to the applicability of Frank’s elastic theory to chromonics. We plan to explore further this issue in the near future.

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Appendix A: Dynamical Analogy

This Appendix is devoted to a dynamical interpretation of the free-energy functional $\mathcal{F}$ in (26). In this interpretation, the first integral (30) will be regarded as a conservation law and the qualitative features of the equilibrium solution (37) will be derived from a phase plane analysis. Of course, nothing will be added to what we already know about the ET fields, but we shall perhaps appreciate better their stance amid other equilibrium configurations with unbounded energy.

We transform functional (26) into a dynamical action by introducing the artificial time

$$t := -\ln \rho.$$  

(A1)

Thus, the axis of the cylinder $\mathcal{B}$ at $\rho = 0$ is approached as $t \to \infty$, while $t = 0$, the origin of time, corresponds to the surface of the lateral surface of $\mathcal{B}$. Correspondingly,
the angle $\beta$ becomes a function of $t$,

$$b(t) := \beta \left( e^{-t} \right),$$

(A2)

and $\mathcal{F}$ in (26) acquires the form of an *infinite horizon* action (Agrachev and Chittaro 2009),

$$\mathcal{F}[b] := \int_{0}^{\infty} \left( \frac{\dot{b}^2}{2} + \Phi(b) \right) dt - \Phi_0(b(0)),$$

(A3)

where, as customary, a superimposed dot denotes differentiation with respect to $t$ and

$$\Phi(b) := \frac{1}{2} \sin^2 b (\cos^2 b + k_3 \sin^2 b),$$

(A4)

$$\Phi_0(b) := -\frac{1}{2} (1 - 2k_{24}) \sin^2 b.$$  

(A5)

The associated Lagrangian $\mathcal{L}$ is the classical sum of a kinetic energy and a potential,

$$\mathcal{L} := \frac{1}{2} \dot{b}^2 + \Phi(b),$$

(A6)

in the single Lagrangian variable $b$. It is perhaps less typical of a classical dynamical system the *initial-value* potential $\Phi_0$. The *orbits* of the system are all solutions to the equation of motion

$$\frac{d}{dt} \frac{\partial \mathcal{L}}{\partial \dot{b}} - \frac{\partial \mathcal{L}}{\partial b} = 0,$$

(A7)

which here reads simply as

$$\ddot{b} - \Phi'(b) = 0.$$  

(A8)

This equation is subject to both initial and asymptotic conditions which stem from requiring stationarity of the action at the beginning and at the end of time,\(^{19}\)

$$\left( \frac{\partial \mathcal{L}}{\partial \dot{b}} + \frac{\partial \Phi_0(b)}{\partial b} \right)_{t=0} = 0,$$

(A9a)

$$\lim_{t \to \infty} \frac{\partial \mathcal{L}}{\partial \dot{b}} = 0.$$  

(A9b)

While the latter requires that

$$\lim_{t \to \infty} \dot{b}(t) = 0,$$

(A10)

the former identifies a locus in the phase plane $(b, \dot{b})$ for all admissible initial conditions,

$$\dot{b} = (1 - 2k_{24}) \sin b \cos b.$$  

(A11)

\(^{19}\) Alternatively, we may say that equations (A9) prescribe the sources of momentum at both ends of the time horizon.
Fig. 3 Phase portrait in the plane \((b, \dot{b})\) of the dynamical system with total action in (A3). Here \(k_3 = 30\), \(k_{24} = 7.5\), and \(c \geq -30\). The red (thick) curve is the locus of allowed initial conditions; the separatrix is blue, while closed and open orbits are depicted in green and gray, respectively. Arrows indicate the direction of flow as time elapses. Three trajectories have been highlighted in darker green, darker blue, and black, starting from the initial condition: they correspond to the following values of the parameter \(c\) that selects the orbits, \(c = -20\), \(c = 0\), and \(c = 20\), respectively (Color figure online)

A conservation law follows readily from (A8),

\[
\frac{1}{2} \dot{b}^2 = \Phi(b) + \frac{1}{2} c, \tag{A12}
\]

where \(c\) is an arbitrary constant. Since \(\Phi \leq k_3/2\), it readily follows from (A12) that the admissible values of \(c\) fall in the range \(c \geq -k_3\). The phase portrait of (A12) in the plane \((b, \dot{b})\) is illustrated in Fig. 3 for \(b \in [-\pi, \pi]\). Orbits are selected by the values of \(c\). For \(c = -k_3\), the corresponding orbits collapse in the points \((\pm \pi/2, 0)\). As \(c\) increases, orbits are inflated and stay closed until \(c = 0\) (see green trajectories in Fig. 3). This latter value corresponds to orbits that converge to the origin and to the points \((\pm \pi, 0)\) (blue trajectories). They collectively act as a separatrix in phase plane, bounding the domain of closed orbits. For \(c > 0\), orbits become unbounded (gray trajectories).

This qualitative analysis, based on the conservation law (A12), embraces all possible orbits. We are especially interested in those that start from the admissible locus (A11) in phase space and obey the asymptotic condition (A10). The former is represented by the red curve in Fig. 3, while the latter requires the orbit to approach the \(b\)-axis as \(t \to \infty\). Now, if \(c < 0\), the green trajectories are periodic orbits that keep crossing infinitely many times the \(b\)-axis, whereas if \(c > 0\) the gray trajectories are open orbits.
that never cross the $b$-axis. Only for $c = 0$, that is, along the separatrix, can an orbit approach the $b$-axis starting from an admissible initial condition.

As shown in Fig. 3, there is a critical value of $c$ corresponding to orbits tangent to the curve of admissible initial conditions,

$$c_{\text{max}} := \frac{4k_{24}^2(k_{24} - 1)^2}{k_3 + 4k_{24}(k_{24} - 1)}.$$  \hspace{1cm} (A13)

For $c > c_{\text{max}}$, the open orbits of the system are inadmissible. For $0 < c \leq c_{\text{max}}$ and $-k_3 \leq c < 0$, the orbits obey (A9a), but violate (A9b); their total action $\mathcal{F}$ in (A3), which by (A12) can be expressed as

$$\mathcal{F}[b] = \int_0^\infty \left[ 2\Phi(b) + \frac{c}{2} \right] dt + \Phi_0(b(0)),$$  \hspace{1cm} (A14)

is easily seen to be unbounded.

Contrariwise, for $c = 0$, the total action is finite and can be shown to equal the elastic free energy $\mathcal{F}_{\text{ET}}$ computed in (38). The rate at which time diverges as the origin in phase space is approached (on a the separatrix) can be estimated as

$$t \approx -2 \ln b \quad \text{as} \quad b \to 0,$$  \hspace{1cm} (A15)

see (Paparini 2022) for more quantitative details.

We have learned in Sect. 2 that the equilibrium solution $\beta_{\text{ET}}$ in (37) only exists for $k_{24} > 1$. This also arises from the dynamical analogy studied here and is revealed by a geometric feature of the phase portrait. As can be shown by an asymptotic expansion of both (A11) and (A12) near the origin, for $k_{24} < 1$, the curve of admissible initial conditions lies inside the separatrix, and so there is no admissible orbit with finite total action. Figure 4 illustrates graphically the situation; in particular, for $k_{24} = 0$ and $k_{24} = 1$, the curve of admissible initial conditions is (internally) tangent to the separatrix.

**Appendix B: Useful Identities and Inequalities**

We collect in this Appendix a number of identities and inequalities that are used in the main text.

For the mapping $t \mapsto n_t(x)$ introduced in Sect. 3, we compute the following derivatives with respect to the parameter $t$,

$$\frac{d}{dt} \text{tr} (\nabla n_t)^2 = \frac{d}{dt} \left( I \cdot \nabla n_t \right) = I \cdot (\nabla n_t) \cdot \nabla n_t + I \cdot \nabla n_t \cdot (\nabla n_t) = 2 \text{tr}(\nabla n_t \nabla n_t) \hspace{1cm} (B1)$$

and

$$\frac{d^2}{dt^2} \text{tr} (\nabla n(t))^2 = 2 \frac{d}{dt} (I \cdot \nabla n_t \nabla n_t) = 2 \text{tr} \left( \nabla n_t \nabla n_t + (\nabla n_t)^2 \right). \hspace{1cm} (B2)$$
Fig. 4 Separatrix (blue) and the curve of allowed initial conditions (red) are depicted on the phase plane $(b, \dot{b})$ for $k_3 = 1$ and several values of $k_{24}$. For $0 \leq k_{24} \leq 1$, no orbit is allowed with finite total action, as the curve of admissible initial conditions is fully enclosed by the separatrix (Color figure online).

By the use of these in (1), we readily obtain that

$$\dot{W}_F := \frac{d}{dt} W_F(n_t, \nabla n_t)$$

$$= K_{11} \text{div} n_t \text{div} \dot{n}_t + K_{22} (n_t \cdot \text{curl} n_t) (\dot{n}_t \cdot \text{curl} n_t + n_t \cdot \text{curl} \dot{n}_t)$$

$$+ K_{33} n_t \times \text{curl} n_t \cdot (\dot{n}_t \times \text{curl} n_t + n_t \times \text{curl} \dot{n}_t)$$

$$+ 2 K_{24} [\text{tr}(\nabla \dot{n}_t \nabla n_t) - \text{div} n_t \text{div} \dot{n}_t]$$

(B3)

and

$$\ddot{W}_F := \frac{d^2}{dt^2} W_F(n_t, \nabla n_t) = K_{11} \left( (\dot{n}_t \cdot \text{curl} n_t)^2 + \text{div} n_t \text{div} \dot{n}_t \right)$$

$$+ K_{22} \left( (\dot{n}_t \cdot \text{curl} n_t + n_t \cdot \text{curl} \dot{n}_t)^2 \right.$$}

$$+ n_t \cdot \text{curl} n_t (\ddot{n}_t \cdot \text{curl} n_t + 2 \dot{n}_t \cdot \text{curl} \ddot{n}_t + n_t \cdot \text{curl} \dot{n}_t) \bigg)$$

$$+ K_{33} \left( |\dot{n}_t \times \text{curl} n_t + n_t \times \text{curl} \dot{n}_t|^2 + n_t \times \text{curl} n_t \cdot (\ddot{n}_t \times \text{curl} n_t$$

$$+ 2 \dot{n}_t \times \text{curl} \ddot{n}_t + n_t \times \text{curl} \dot{n}_t) \bigg)$$

$$+ 2 K_{24} \left[ \text{tr} (\nabla \ddot{n}_t \nabla n_t) + \text{tr}(\nabla \dot{n}_t)^2 - (\text{div} \dot{n}_t)^2 - \text{div} n_t \text{div} \dot{n}_t \right].$$

(B4)
For a director field \( \mathbf{n} \) as in (18), whose gradient has been computed in (19), and for the perturbation field \( \mathbf{v} \) described in (59), the following computational ingredients were needed in the main text to arrive from the general formula for the second variation of Frank’s elastic free energy in (52) at the special form needed when the unperturbed field is \( \mathbf{n} = \mathbf{n}_{ET} \),

\[
\begin{align*}
\text{div } \mathbf{n} &= 0, \\
\mathbf{n} \cdot \text{curl } \mathbf{n} &= \frac{1}{R} \left( \beta' + \frac{\cos \beta \sin \beta}{\rho} \right), \\
\mathbf{n} \times \text{curl } \mathbf{n} &= \frac{1}{R} \left( \frac{\sin^2 \beta}{\rho} \right) \mathbf{e}_r, \\
\text{tr} (\nabla \mathbf{n})^2 &= \frac{1}{R^2} \left( -\frac{2 \cos \beta \sin \beta \beta'}{\rho} \right), \\
\nabla \mathbf{v} &= \frac{1}{R} \left[ f' \mathbf{e}_r \otimes \mathbf{e}_r + \frac{g \cos \beta}{\rho} \mathbf{e}_r \otimes \mathbf{e}_\theta + \left( g \sin \beta \beta' - \cos \beta g' \right) \mathbf{e}_\theta \otimes \mathbf{e}_r \\
&\quad \quad + \frac{f}{\rho} \mathbf{e}_\theta \otimes \mathbf{e}_\theta + \left( \sin \beta g' + g \cos \beta \beta' \right) \mathbf{e}_z \otimes \mathbf{e}_r \right], \\
\text{div } \mathbf{v} &= \frac{1}{R} \left( f' + \frac{f}{\rho} \right), \\
\mathbf{v} \cdot \text{curl } \mathbf{n} &= \frac{1}{R} \left( \frac{g \sin^2 \beta}{\rho} \right), \\
\mathbf{n} \cdot \text{curl } \mathbf{v} &= \frac{1}{R} \left[ -g' - \frac{g \cos^2 \beta}{\rho} \right], \\
\mathbf{v} \cdot \text{curl } \mathbf{v} &= \frac{1}{R} \left[ g^2 \left( \beta' - \sin \beta \cos \beta \right) \right], \\
\mathbf{v} \times \text{curl } \mathbf{n} &= \frac{1}{R} \left[ -g \left( \beta' + \frac{\sin \beta \cos \beta}{\rho} \right) \mathbf{e}_r \\
&\quad \quad - f \left( \frac{\sin \beta}{\rho} + \cos \beta \beta' \right) \mathbf{e}_\theta + f \sin \beta \beta' \mathbf{e}_z \right], \\
\mathbf{n} \times \text{curl } \mathbf{v} &= \frac{1}{R} \left[ g \left( \beta' - \frac{\sin \beta \cos \beta}{\rho} \right) \mathbf{e}_r \right], \\
(\mathbf{v} \times \text{curl } \mathbf{v}) \cdot \mathbf{e}_r &= \frac{1}{R} \left( gg' + \frac{g^2 \cos^2 \beta}{\rho} \right), \\
v^2 &= f^2 + g^2, \\
\nabla v^2 &= \frac{1}{R^2} \left( 2 ff' + 2 gg' \right), \\
\text{tr} (\nabla v)^2 &= \frac{1}{R^2} \left[ f'^2 + \frac{2 g \cos \beta}{\rho^2} \left( g \sin \beta \beta' - \cos \beta g' \right) \right],
\end{align*}
\]

where a prime denotes differentiation with respect to \( \rho \).
A family of inequalities has played a central role in Sect. 4. They stem from the adaptation to our setting of a Lemma proved in Li and Yeh (1995) (see Lemma 2.1).

**Lemma 1** Let \([a, b]\) be an interval in \(\mathbb{R}\) and let \(\tilde{U} = \tilde{U}(u) \in AC[a, b]\). If there are two functionals, \(Q(u, t)\) and \(G(u, t)\), satisfying the following conditions
1. \(t\) is in the range of the function \(\tilde{U} = \tilde{U}(u)\),
2. \(Q(u, \tilde{U}(u))\) is integrable for \(u \in [a, b]\),
3. \(G(u, \tilde{U}(u))\) is absolutely continuous for \(u \in [a, b]\),
then
\[
\int_{a}^{b} Q(u, \tilde{U}(u)) \tilde{U}'(u)^2 \, du \geq - \int_{a}^{b} \left[ Q(u, \tilde{U}(u))^{-1} G_{\tilde{U}}^2 \right.
+ 2G_{u} \, du + 2 \left[ G(b, \tilde{U}(b)) - G(a, \tilde{U}(a)) \right],
\]
(B6)

where \(G_{\tilde{U}} := \partial G(u, \tilde{U})/\partial \tilde{U}\) and \(G_{u} := [\partial G(u, t)/\partial u]_{t=\tilde{U}}\).

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