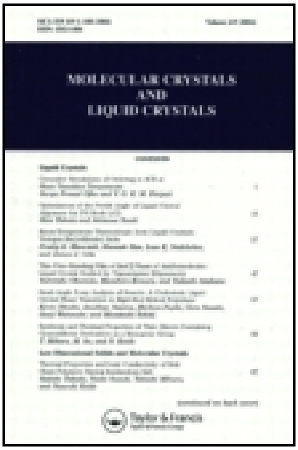

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Stability of B_7 Fibers

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Recent experimental studies have found that fluid smectic phases of bent core liquid crystals form stable free standing fibers with extremely high slenderness ratios. In [1] a model was proposed to explain their formation based on the distinctive packing properties of bent core molecules in the SmCG phase. Here we develop this model further to include smectic energy terms so as to allow more general configurations and fields. We show that the relative size of the energy's elasticity constants can be used to determine the stability (instability) of circular fibers.

Keywords: B_7 phase; bent core liquid crystals; fiber; smectic

INTRODUCTION

In this article we describe our analysis of a model for free standing fibers formed from concentric, smectic layers of kinked liquid crystal mesogens. Smectic liquid crystal molecules self organize into layers by molecular packing, taking into account both their physical and electrostatic portraits. For the case of the kinked molecules considered here (double tilted $SmCG$ mesogens in the B_7 phase) the layers further organize concentrically about a thread-like core into a fluid fiber of extremely high slenderness ratio. In Bailey *et al.* [1] formulated and studied a model so as to explain the fiber formation they found in their

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experiments and those of others. Our work further analyzes this model. A more detailed version of this analysis is in [2].

In a number of experiments [3,4] it is observed that the cross-section of a fiber drawn in air is independent of its length. Assuming this the problem of describing a fiber is posed as a free boundary problem for the cross-section with its free energy per unit length based on the distinctive elastic (molecular packing) and electrostatic properties of bent core liquid crystals. The analysis in [1] was limited to fibers with circular cross-sections and cylindrically constant fields. In the present paper we further develop this model by including energy terms for smectic layer formation, so as to allow for more general cross-sections, fields, and smectic layer structures. This is relevant since fibers with circular cross-sections as well as ones with small surface undulations are observed [3–5].

In our formulation the inner core and outer fiber surface are free boundaries. We prove the existence of equilibrium configurations with circular cross-sections and investigate their stability relative to general variations of the fiber cross-section. The liquid crystalline material is in a smectic *C* phase. As such there is an energetic cost to smectic layer bending. If the term in the energy corresponding to this is comparable to the Frank–Oseen energy we prove that the circular configurations are stable. If however the smectic layer energy is the dominant elastic contribution we prove that circular equilibria become unstable relative to deformations with small surface undulations.

THE MODEL

Bent core liquid crystal molecules are banana-shaped as in Figure 1 and are described by two orthonormal vectors $\mathbf{n}$ and $\mathbf{p}$. We view the molecule as two dimensional lying in the molecular plane spanned by $\mathbf{n}$ and $\mathbf{p}$. Here $\mathbf{n}$ is parallel to the chord connecting the two ends of the molecule and $\mathbf{p}$ points toward the kink in the molecule from the midpoint of the chord. Because of its shape the molecule carries a microscopic polarization parallel to $\mathbf{p}$. We do not distinguish between the microscope fields $\mathbf{n}$ and $\mathbf{p}$ and the corresponding macroscopic ones representing their local averages. We denote $\mathbf{n}$ as the *director* and $\mathbf{p}$ as the *polarization* vector.

Consider a cylindrical fiber with axis $\mathbf{e}_{x3}$, cross section Σ, and core $\Omega_0 \subset\subset \Sigma \subset x_1x_2$ plane. The region $\Omega := \Sigma\backslash\bar{\Omega}_0$ is an *annular domain*; that is Σ and Ω_0 are open bounded simply connected sets with smooth boundaries $\Gamma_0 := \partial\Omega_0$ and $\Gamma_1 := \partial\Sigma$. Here Ω represents the cross-section of the smectic region, and $\mathbf{n}, \mathbf{p} : \bar{\Omega} \to \mathbb{S}^2$ (See Figure 2).

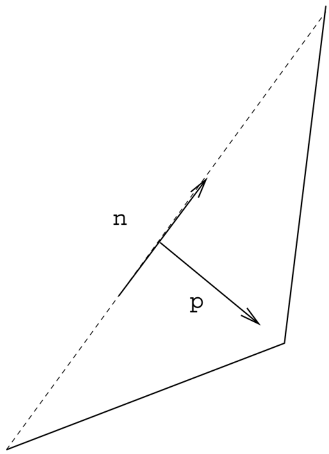

FIGURE 1 Bent core molecule.

Let $\mathbf{k}$ be a unit layer normal at a given point $\mathbf{x}_0$ and $\mathbf{u}$ a unit vector perpendicular to $\mathbf{k}$. Then the molecular orientation at $\mathbf{x}_0$ given through $\mathbf{n}(\mathbf{x}_0)$ and $\mathbf{p}(\mathbf{x}_0)$ is expressed in terms of three angles θ, α, and ϕ relative to $\mathbf{k}$ and $\mathbf{u}$ as depicted in Figure 3. Here $0 < \theta < \pi$ is the tilt angle between $\mathbf{n}$ and $\mathbf{k}$; $|\alpha| \leq \pi$ is the angle obtained by rotating $\mathbf{k} \times \mathbf{n}$ to $\mathbf{p}$, about $\mathbf{n}$ using a right-hand rule; α measures the

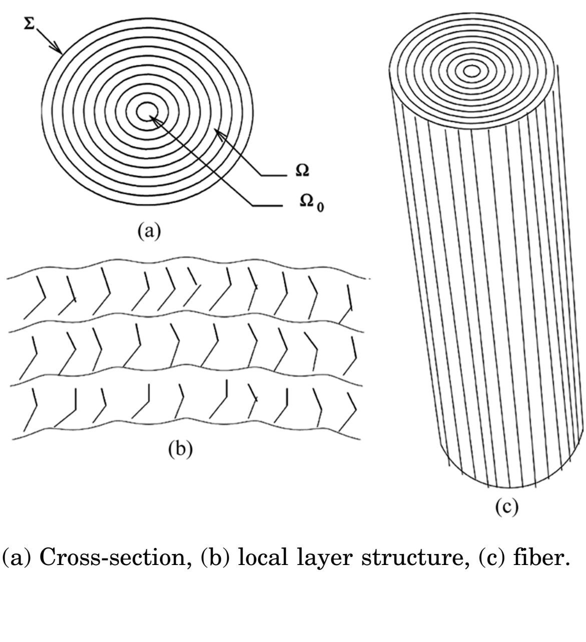

FIGURE 2 (a) Cross-section, (b) local layer structure, (c) fiber.

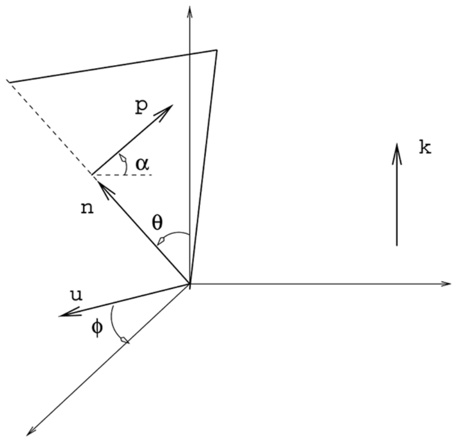

FIGURE 3 Molecular angles.

tilt of the molecular plane away from $\mathbf{k} \times \mathbf{n}$; ϕ mod 2π is the azimuthal angle measuring the rotation of the molecule about $\mathbf{k}$ away from $\mathbf{u}$ toward $\mathbf{k} \times \mathbf{n}$. Note that varying θ and α affects the molecule's height (layer thickness) while varying ϕ does not. Because of this θ and α are called *stiff* variables while ϕ is labeled a *soft* variable.

The free energy per unit length of the fiber, $\mathcal{F}$, is determined by taking into account elastic energy (measured by layer strain and compression), polar divergence, electric self-interaction, and surface tension:

$$\mathcal{F} := F_{Sm} + F_F + F_C + F_P + F_{El} + F_{Su}.$$

The cross-section of the fiber's smectic portion is described by Ω and a function $\omega(x_1, x_2)$ with $\omega = 0$ on the outer boundary and $\omega = \text{const.} < 0$ on the core boundary such that the cross-sections of the smectic layers foliating Ω correspond to level curves of ω (see Fig. 2a). Variations in layer thickness from material dependent ground state values are energetically expensive. In contrast to this the layers themselves can undergo deformations with a (relatively) small cost in energy. A liquid crystal is in the smectic A phase if the director is parallel to the layer normal within the bulk and in the smectic C phase if the angle, θ_0, between the normal and $\mathbf{n}$ is such that $0 < \theta_0 < \pi$. The liquid crystals studied here are in the smectic C phase. The smectic C material has bulk layer thickness d_0 and tilt angle θ_0. A Chen–Lubensky energy density [6], $f_{CL}(\psi, \mathbf{n})$ is used where ψ is the complex order parameter $\psi = e^{iq\omega(\mathbf{x})}$ where $q := \frac{2\pi}{d_0}$,

$$f_{CL} := \frac{a_\perp}{q^2}(D\cdot D_\perp\psi)^2 - c_\perp|D_\perp\psi|^2 + \frac{a_\|}{q^2}(D\cdot D_\perp\psi)^2 + c_\||D_\|\psi|^2 + \frac{q^2C_\perp^2}{4a_\perp}|\psi|^2.$$

Here $a_\perp$, $c_\perp$, $a_\|$, $c_\| > 0$ are such that

$$\sin^2\theta_0 = \frac{c_\perp}{2a_\perp}, \quad \nabla = (\partial_{x1}, \partial_{x2}, \partial_{x3}),$$

$D = \nabla - i\cos\theta_0 q\mathbf{n}$, $D_\| = (\mathbf{n}\cdot\Delta - i\cos\theta_0 q)\mathbf{n}$, and $D_\perp = D - D_\nu$. Evaluating f_{CL}

$$f_{CL} = a_\perp(\mathrm{div}(\nabla_\perp\omega))^2 + a_\|(\mathrm{div}(\nabla_\|\omega - \cos\theta_0\mathbf{n}))^2 + q^2[a_\perp(|\nabla_\perp\omega|^2 - \sin^2\theta_0)^2 + a_\||\nabla_\|\omega - \cos\theta_0\mathbf{n}|^4 + c_\||\nabla_\|\omega - \cos\theta_0\mathbf{n}|^2],$$

where $\nabla_\|\omega = (\mathbf{n}\cdot\nabla_\omega)\mathbf{n}$ and $\nabla_\perp\omega = \nabla\omega - \nabla_\|\omega$. Here q is understood to be large and one sees that part of the rational for the form of f_{CL} is to favor pure smectic C configurations, $(\omega, \mathbf{n})$ such that $\nabla_\|\omega \equiv \cos\theta_0\mathbf{n}$ and $|\nabla_\|\omega|^2 \equiv \sin^2\theta_0$. The quantity $|\nabla\omega(\mathbf{x})|$ is the ratio of the local layer spacing to that of the bulk at $\mathbf{x}$, [7]. A uniform state has no variation and as such $|\nabla\omega| \equiv 1$.

A bent core liquid crystal that energetically prefers an angle $0 < |\alpha_0| < \pi$, $|\alpha_0| \neq \frac{\pi}{2}$ in the bulk is labeled *SmCG*, [8]. This preference is a result of the molecular atomic structure and the geometry of molecular packing and is reflected in the free energy by the chiral energy density

$$f_{Ch} = jq^8((\nabla\omega\times\mathbf{n}\cdot\mathbf{p})^2(\nabla\omega\cdot\mathbf{n})^2 - x_o^2|\nabla\omega|^4)^2$$

where $j > 0$ and $\chi_0 = \sin\theta_0\cos\theta_0\cos\alpha_0$, [8]. We set $f_{Sm} = f_{CL} + f_{Ch}$ as the smectic energy density and

$$F_{Sm} = \int_\Omega f_{Sm}$$

as the smectic free energy per unit length.

The elastic energy density accounts for steric molecular interactions not associated with layer formation. The density is given in terms of distortions of the fields $\mathbf{n}$ and $\mathbf{p}$ by a one constant Frank-Oseen energy density,

$$f_F = K(|\nabla\mathbf{n}|^2 + |\nabla\mathbf{p}|^2) \text{ where } K > 0$$

and we set

$$F_F = \int_\Omega f_F.$$

The core is assumed to be isotropic and is modeled by a constant bulk energy density $b = b(K, a_\perp)$ such that the core free energy is

$$F_C = \int_{\Omega_0} b = b|\Omega_0|.$$

Linear divergence of $\mathbf{p}$ contributes to the free energy through elastic and electrostatic interactions. The field $\mathbf{p}$ tends to splay in an effort to maximize molecular packing. In addition the energy due to electrostatic repulsion manifests itself as splay in the macroscopic polarization $\mathbf{P} = P_0\mathbf{p}$ where $P_0 = \text{const}$. We write $f_p = c_p$ div $\mathbf{p}$ where $c_p := c' + c'' P_0$ to account for both contributions and set

$$F_P = \int_\Omega f_P.$$

The diverging spontaneous polarization induces an electric field $\mathbf{E} = -\nabla\Phi\ (x_1, x_2)$. In addition free charges collect on Γ_1 that suppress the field outside of the fiber, so that $\Phi = 0$ in $\sum^c$. Gauss's law then reads

$$\begin{cases} \varepsilon_0\bar{\varepsilon}\Delta\Phi = \operatorname{div}(\mathbf{P}\chi_\Omega) & \text{in } \Sigma, \\ \Phi = 0 \text{ on } \delta\Sigma, \end{cases} \tag{1.1}$$

where $\varepsilon_0 > 0$ is the dielectric permittivity of free space, $\bar{\varepsilon} > 0$ is the relative dielectric constant for the liquid crystal, and χ_Ω is the characteristic function for Ω. Here $\mathbf{P}$ is set equal 0 in $\bar{\Omega}^c$. The energy density for these interactions is then

$$f_{El} = \frac{\varepsilon_0\bar{\varepsilon}}{2}|\mathbf{E}|^2 - \mathbf{P}\cdot\mathbf{E} = \frac{\varepsilon_0\bar{\varepsilon}}{2}|\nabla\Phi|^2 + \mathbf{P}\cdot\nabla\Phi \text{ in}\Sigma$$

Using (1.1) then we can write

$$F_{El} = \int_\Sigma f_{El} = 3\frac{\varepsilon_0\bar{\varepsilon}}{2}\int_\Sigma |\nabla\Phi|^2 = e\int_\Sigma |\nabla\Phi|^2$$

where $e := \frac{3\varepsilon_0\bar{\varepsilon}}{2}$.

The energy required to create the interfaces at Γ_0 and Γ_1 is taken as

$$F_{Su} = \int_{\Gamma_0} \sigma_0 + \int_{\Gamma_1} \sigma_1 = \sigma_0|\Gamma_0| + \sigma_1|\Gamma_1|$$

where σ_0, $\sigma_1 > 0$ are constants.

Set

$$\begin{aligned}\mathcal{A} :=\{(\omega, \mathbf{n}, \mathbf{p}) : \omega \in W^{2,2}(\Omega) \text{ such that} \\ \omega = 0 \text{ on } \Gamma_1 \text{ and } \omega = \text{const.} \leq 0 \text{ on } \Gamma_0 \\ \mathbf{n}, \mathbf{p} \in W^{1,2}(\Omega; \mathbb{S}^2) \text{ such that } (\mathbf{n} \cdot \mathbf{p}) = 0 \text{ in } \Omega\}\end{aligned}$$

here $W^{k,2}(\Omega)$ has square integrable derivatives up to order k. We have

Theorem 1. *There exists a minimizer for $\mathcal{F}$ in $\mathcal{A}$.*

Given Ω the liquid crystalline structure of the fiber is determined by the triple $(\omega, \mathbf{n}, \mathbf{p}) \in \mathcal{A}$. Our problem is to investigate the stability of equilibria for $\mathcal{F}$ first with respect to $(\omega, \mathbf{n}, \mathbf{p}) \in \mathcal{A}$ (Ω) and then with respect to variations of the cross-section Ω. This is highly complex and we instead study a limiting case that reduces the number of unknowns but retains enough of the physics to investigate the stability of circular fibers.

THE LIMITING MODEL

We set $\mathcal{F} := \mathcal{F}_q$. The smectic layer thickness is very small relative to the fiber's diameter and this motivates the limiting problem obtained by sending $q \to \infty$. In general, if Γ_1 and Γ_0 are unrelated then these energies would diverge; singularities would form in the layers near the boundary as $q \to \infty$. For our case however the boundary is free, layer formation is promoted and the limit should be well defined. We form a limiting problem on a class of competing domains near circular cross-sections.

Set $\mathbb{T} = \mathbb{R}/2\pi\mathbb{Z}$. Let Γ_1 be described by a 2π periodic polar curve $\rho = \rho_1(\gamma) > 0$ of class $C^3(\mathbb{T}), \Gamma_1 : \tilde{\mathbf{x}}(\gamma) = (\rho_1(\gamma)\cos\gamma, \rho_1(\gamma)\sin\gamma)$. We label

$$\ell = (\rho_1^2 + {\rho_1}'^2)^{1/2} = |\tilde{\mathbf{x}}(\gamma)|, \ \mathbf{t} = \tilde{\mathbf{x}}'/\ell, \ \boldsymbol{\nu} = (t_2, -t_1)$$

where $\mathbf{t}$ is a unit tangent and $\boldsymbol{\nu}$ is the exterior normal to $\sum$ at Γ_1.

Definition. An annular domain, Ω is *ruled* if there is a 2π periodic function $d_0(\gamma) > 0$ of class $C^2(\mathbb{T})$ so that the parameterizations

$$\mathbf{x}(\gamma, \tau) = -\tau\boldsymbol{\nu}(\boldsymbol{\gamma}) + \tilde{\mathbf{x}}(\gamma) \tag{2.1}$$

for $\gamma \in \mathbb{T},\ 0 \le \tau \le d_0(\gamma)$ is a diffeomorphism onto $\bar{\Omega}$.

A ruled annular domain is depicted in Figure 4. Note that this notion is open, in that if a second region, Ω' is sufficiently close to a ruled annular domain Ω (i.e., Γ_1' is of class C^3 and $\partial\Omega'$ is close to $\partial\Omega$ in C^2) then Ω' is ruled as well. We are particularly interested in the case where Ω' is a perturbation of a circular annulus $B_r \backslash B_{r0}$, with d_0 close to $r - r_0$ and ρ_1 close to r.

Set

$$\mathcal{A}_\infty := \{(\omega, \mathbf{n}, \mathbf{p}) \in \mathcal{A} : |\nabla_\perp\omega|^2 = \sin^2\theta_0, \nabla_\| \omega = \cos\theta_0 \mathbf{n},$$
$$\text{and} (\mathbf{n} \times \nabla\omega \cdot \mathbf{p})^2 \cos^2\theta_0 = \chi_0^2 \quad \text{almost everywhere in } \Omega\},$$

and define

$$\mathcal{F}_\infty(\omega, \mathbf{n}, \mathbf{p}) := \int_\Omega a_\perp(\operatorname{div}(\nabla_\perp\omega))^2 + F_C + F_F + F_P + F_{El} + F_{Su}$$

for (ω, **n, p**) $\in \mathcal{A}_\infty$.

We have

Theorem 2. *Let Ω be a ruled annular domain with d_o = const. Then there exists $(\bar{\omega}, \bar{\mathbf{n}}, \bar{\mathbf{p}}) \in \mathcal{A}_\infty$ so that*

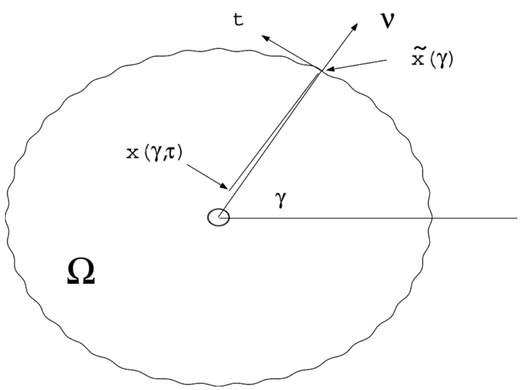

FIGURE 4 Coordinates for Ω.

$$\mathcal{F}_\infty(\bar{\omega}, \bar{\mathbf{n}}, \bar{\mathbf{p}}) = \lim_{q \longrightarrow \infty} (\inf_{\mathcal{A}} \mathcal{F}_q) = \inf_{\mathcal{A}\infty} \mathcal{F}_\infty.$$

Moreover $\bar{\omega}(x_1, x_2) = -dist.((x_1, x_2), \Gamma_1)$, *and for a subsequence minimizers for the q-problem,* $(\omega_{qi}, \mathbf{n}_{qi}, \mathbf{p}_{qi}) \longrightarrow (\bar{\omega}, \bar{\mathbf{n}}, \bar{\mathbf{p}})$ *as* $q_i \longrightarrow \infty$.

It follows that we can investigate the stability of a circular fiber with cross-section $B_r \backslash B_{ro}$ and a class of perturbed domains Ω using the limiting energy $\mathcal{F}_\infty$ and $\mathcal{A}_\infty$. The limiting problem is stiffer. The tilt angles θ_0 and α_0 are fixed as well as the layer structure. One can see the stiffness in the fields **n** and **p** as well. We have that $\{\boldsymbol{\nu}(\gamma), \mathbf{t}(\gamma), e_{x_3}\}$ is an orthonormal frame for Ω following the parameterizations (2.1). In terms of this frame we write

$$\mathbf{n} = n_1 \boldsymbol{\nu} + n_2 \mathbf{t} + n_3 \mathbf{e}_{x3},$$

where

$$[n_1, n_2, n_3] = [\cos\theta_0, \sin\theta_0 \cos\phi, \sin\theta_0 \sin\phi].$$

The polarization **p** satisfies pin, $\mathbf{p} \perp \mathbf{n}$,

$$\mathbf{p} = p_1 \boldsymbol{\nu} + p_2 \mathbf{t} + p_3 \mathbf{e}_{x3}$$

where

$$\begin{aligned}[p_1, p_2, p_3] =& [\sin\alpha_0 \sin\theta_0, -\cos\alpha_0 \sin\phi - \sin\alpha_0 \cos\theta_0 \cos\phi, \\ & \cos\alpha_0 \cos\phi - \sin\alpha_0 \cos\phi_0 \sin\phi].\end{aligned}$$

Thus $\mathbf{n} = \mathbf{n}(\phi\ (x_1, x_2))$, $\mathbf{p} = \mathbf{p}(\phi\ (x_1, x_2))$ and the analysis becomes much more tractable. We investigate $\mathcal{F}_\infty$ evaluated at admissible domains Ω near $B_r \backslash B_{ro}$ and fields $\mathbf{n}(\phi)$ and $\mathbf{p}(\phi)$ with ϕ near ϕ_0. These are characterized by triples $(d, \rho_1(\gamma), \tilde{\phi}(\gamma, u))$ where $\Gamma_1 : \rho = \rho_1(\gamma), \Gamma_0 :$ d— level set of dist(x, Γ_1), and $\phi(\tilde{\mathbf{x}}(\gamma, \tau)) = \phi(\gamma, \tau/d)$ such that $\tilde{\phi}(\gamma, u) \in W^{1,2}(\mathbb{T} \times (0, 1))$. We write

$$\Im_\infty(d, \rho_1, \tilde{\phi}) := \mathcal{F}_\infty(\omega, \mathbf{n}, \mathbf{p}; \Omega).$$

We use this to prove (as in [1]) the existence of circular fibers with radially constant fields. This done by minimizing $\Im$ with respect to constant d, ρ_1, and $\tilde{\phi}$. Moreover we show that these are equilibria with respect to general variations.

Theorem 3. *Let $0 \le \phi_0 < 2\pi$ minimize $(n_2^2 + p_2^2)(\phi)$ and assume that $\sigma_0 + c_P p_1 < 0$. Then there exist unique $0 < \bar{d} < \bar{r}$ such that $(\bar{d}, \bar{r}, \phi_0)$ minimizes $\Im$ among all constant states. Moreover this is an equilibrium for $\Im$ with respect to general variations $(d, \rho_1(\gamma), \tilde{\phi}(\gamma, u))$ near $(\bar{d}, \bar{r}, \phi_0)$.*

This determines a circular fiber with radius $\bar{r}$ and core radius $\bar{r} - \bar{d}$. The condition $\sigma_0 + c_p p_1 < 0$ ensure there is sufficient polar splay to balance the other terms in the energy that favor shrinking the cross-section. The main reason for forming the limiting energy is that we can explicitly calculate its second variation at $(\bar{d}, \bar{r}, \phi_0)$ with respect to general variations $(\bar{d} + \varepsilon h, \bar{r} + \varepsilon g(\gamma), \phi_0 + \varepsilon\eta(\gamma, u))$,

$$D^2\Im(\bar{d}, \bar{r}, \phi_0)[h, g, \eta] := \frac{d^2\Im}{d\varepsilon^2}(\bar{d} + \varepsilon h, \bar{r} + \varepsilon g, \phi_0 + \varepsilon\eta) \quad \text{at} \quad \varepsilon = 0.$$

The elasticity constants $a_\perp$ and K are much smaller than the other coefficients. As such we examine the case where both are small. We show however that the relative sizes of $a_\perp$ and K are particularly relevant. We prove in the case where $a_\perp$ and K are proportional and small that the circular configurations are stable with respect variations. We then consider a case where K is much smaller than $a_\perp$ and find instances where the circular configuration becomes unstable with respect to variations having small modulations.

Theorem 4. *Let $\mu > 0$ and assume that $\mu^{-1}K \le a_\perp \le \mu K$. Then there is a $\delta > 0$ such that if $a_\perp + K < \delta$ then*

$$D^2\Im(\bar{d}, \bar{r}, \phi_0)[h, g, \eta] \ge 0 \quad \textit{for all admissible} \quad (h, g, \eta).$$

Theorem 5. *Assume $\theta_0 \ne \theta_0 \ne \pi/2$. Let s be such that $0 < s < 1$ and $b = (a_\perp + K)^{-s}$. If $K \le \delta(a_\perp)^{2-s}$ for some $\delta > 0$. Then there exist g and η such that $D^2\Im(\bar{d}, \bar{r}, \phi_0)[0, g, \eta] < 0$.*

The perturbations g and η used in the theorem have oscillations, showing that a fiber with surface undulations and a polarization field with modulations in the layer plane has less energy than the circular fiber with cylindrically constant fields. Physically plausible values for the coefficients are $a_\perp = 10^{-9}N$, $K = 10^{-11}N$, and $b = 10^7$. The conditions above are consistent with these values if we take $s = \frac{7}{9}$.

CONCLUSIONS

In this article we generalize a model by Bailey *et al* [1] for cylindrical fibers to include smectic layer energy and general cross-sections. We then derive a limiting model freezing both molecular tilt angles (leaving the fields **n** and **p** with one degree of freedom) and layer thickness. Using the limiting energy we are able to show that circular fibers are stable if the layer bending constant $a_\perp \propto K$, the Frank constant. If K is much smaller than $a_\perp$ we show that circular fibers are unstable with respect to variations having surface undulations and modulations in the layer–plane component of the polarization.

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