Instability of radial hedgehog configurations in nematic liquid crystals under Landau–de Gennes free-energy models

E. C. Gartland, Jr. * and S. Mkaddem
Department of Mathematics and Computer Science, Kent State University, Kent, Ohio 44242
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We consider radial hedgehog equilibrium configurations of the tensor order parameter in spherical droplets of nematic liquid crystals modeled by free energies of the Landau–de Gennes type. We show that such configurations must become unstable at sufficiently low temperatures in droplets of sufficiently large radii for all but a very limited range of elastic-constant ratios, which are very near the limit where the elastic-energy terms in the model cease to be positive definite. The analysis is complicated by the fact that no analytical solution is available for the hedgehog configuration. Nevertheless, using a combination of analytical bounds and numerical computation, we are able to construct a perturbation to which we can show that the spherically symmetric ground state becomes unstable. [S1063-651X(99)02901-3]

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I. INTRODUCTION

The study of equilibrium structures and defects of confined liquid crystals has been an area of interest for some time. Here we consider spherical droplets of a nematic with radial strong-anchoring conditions modeled by a Landau–de Gennes tensor-order-parameter model. We are motivated primarily by the succession of papers [1–4]. In [1] Schopohl and Sluckin illustrated for this model the structure of a radial hedgehog configuration with an isotropic core. Penzenstadler and Trebin [2] then showed that the core should instead broaden to a small ring (or loop) disclination (of 180° or strength 1/2). This was validated numerically by Sonnet, Kilian, and Hess in [3]. Rosso and Virga [4] argued that the radial hedgehog solution remained at least metastable over a certain range. Related results are presented in [5–7].

Motivated by this, we have undertaken a detailed numerical investigation of this system, on which we will report elsewhere. In the numerical modeling of the full system, we have imposed *rotational symmetry*. There, in addition to the (spherically symmetric) radial hedgehog and the (axially symmetric) ring disclination solutions, we found a new, metastable configuration, which also is axially symmetric and which consists of a ''split core'' with two isotropic points narrowly separated by a disclination line segment along its symmetry axis. The three solutions, which coexist over a broad range of parameters, are depicted in Fig. 1. These structures all are quite small, existing within a distance from the center of the droplet that is on the order of tens of units of the size of the hedgehog core.

We have computed a bifurcation diagram, Fig. 2, that indicates how these solutions are connected to each other. Since all three of these tensor fields are forced by symmetry to be *uniaxial* along their rotational symmetry axes (in particular at the center of the droplet), the nature of their orientational order at the *center* can be characterized by the value of the *scalar order parameter* there. We have used this as a parameter to distinguish the branches of equilibrium solutions in Fig. 2. This figure is reflective of the common situation of an ''imperfect bifurcation,'' in which the transcritical bifurcation point of the true (continuous) problem has separated under discretization into two nearby but distinct branches.

The diagram indicates that at a certain *critical temperature*, the radial hedgehog solution branch becomes unstable, and off of it bifurcate two branches, which break the spherical symmetry. The lower, metastable branch corresponds to the *split core* solution, which is uniaxial with a *negative* order parameter at the center; while the upper branch corresponds to the *ring disclination*, which is uniaxial with a *positive* order parameter at the center, and which does not become metastable until the radius of the ring grows to sufficient size.

In this note, we give a direct argument that the hedgehog solution must become unstable for sufficiently low temperature provided that the radius of the droplet is sufficiently large and that the elastic constants in the model are not too close to a certain limiting point of their admissible values. We mention that while our numerical modeling of the full system imposes rotational symmetry, the analysis that follows here is general and does not make any such assumption.

II. MODEL AND SCALINGS

Consider a Landau–de Gennes free-energy functional of the tensor order parameter $\boldsymbol{Q}$ in the form

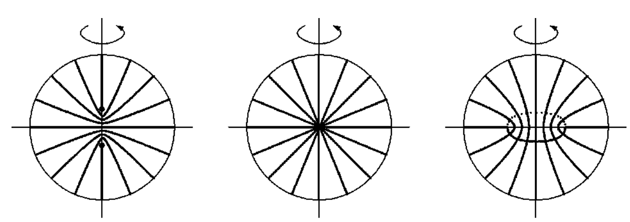

FIG. 1. Three equilibrium director profiles in a radially aligned spherical nematic droplet.

*Author to whom correspondence should be addressed. Also at Chemical Physics Interdisciplinary Program, Liquid Crystal Institute, Kent State University, Kent, OH. Electronic address: gartland@mcs.kent.edu

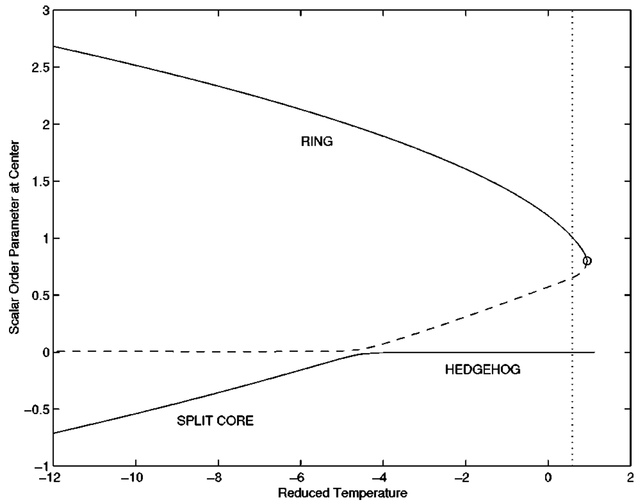

FIG. 2. Bifurcation diagram of discretized model for split core, radial hedgehog, and ring disclination configurations. The solid line indicates *locally stable* (stable or metastable) equilibria; the dashed line indicates *unstable*. The vertical dotted line indicates *transition temperature*: below this temperature, the *ring disclination* (uppermost branch) has minimum free energy; while above it, the free energy of the *radial hedgehog* (isotropic at the origin) is the global minimum.

$$\mathcal{F} := \int (f_{\mathrm{el}} + f_{\mathrm{v}})\, dV,$$

where the *elastic* and *bulk* free-energy densities are given by

$$f_{\mathrm{el}} := \frac{L_1}{2} Q_{\alpha\beta,\gamma} Q_{\alpha\beta,\gamma} + \frac{L_2}{2} Q_{\alpha\beta,\beta} Q_{\alpha\gamma,\gamma} + \frac{L_3}{2} Q_{\alpha\beta,\gamma} Q_{\alpha\gamma,\beta},$$

$$f_{\mathrm{v}} := \frac{a}{2} \mathrm{tr}(\boldsymbol{Q}^2) - \frac{b}{3} \mathrm{tr}(\boldsymbol{Q}^3) + \frac{c}{4} \mathrm{tr}(\boldsymbol{Q}^2)^2.$$

We nondimensionalize this model in terms of the length scale $\xi := \sqrt{27cL_1/b^2}$ and rescaled variables

$$\tilde{\boldsymbol{r}} := \frac{\boldsymbol{r}}{\xi}, \quad \tilde{\boldsymbol{Q}} := \sqrt{\frac{27c^2}{2b^2}} \boldsymbol{Q}, \quad \tilde{\mathcal{F}} := \sqrt{\frac{27c^3}{4b^2 L_1^3}} \mathcal{F}.$$

In terms of these (after dropping the tildes), the densities take the form

$$f_{\mathrm{el}} = \frac{1}{2} Q_{\alpha\beta,\gamma} Q_{\alpha\beta,\gamma} + \frac{\eta_2}{2} Q_{\alpha\beta,\beta} Q_{\alpha\gamma,\gamma} + \frac{\eta_3}{2} Q_{\alpha\beta,\gamma} Q_{\alpha\gamma,\beta},$$

$$f_{\mathrm{v}} = \frac{t}{2} \mathrm{tr}(\boldsymbol{Q}^2) - \sqrt{6}\, \mathrm{tr}(\boldsymbol{Q}^3) + \frac{1}{2} \mathrm{tr}(\boldsymbol{Q}^2)^2,$$

where

$$\eta_2 := \frac{L_2}{L_1}, \quad \eta_3 := \frac{L_3}{L_1}, \quad t := \frac{27ac}{b^2}.$$

The important dimensionless parameters, then, are the *elastic-constant ratios* η_2 and η_3, *reduced temperature* t, and the *radius* of the droplet in units of ξ, which we shall denote by R.

These units were chosen to permit easy comparison with [1–4]. In terms of t, the critical values in the bulk are $t=0$ (below which the isotropic phase is unstable), $t=1$ (nematic-isotropic transition temperature), and $t=9/8$ (above which the ordered phase does not exist). The elastic constants L_1, L_2, and L_3 must satisfy certain inequalities in order for the elastic part of the free energy to be properly *positive definite* (see, for example, [8] or [9]); in terms of η_2 and η_3, these take the form

$$-1 < \eta_3 < 2, \quad 6 + 10\,\eta_2 + \eta_3 > 0. \tag{1}$$

III. HEDGEHOG SOLUTION

The radial hedgehog solution is distinguished by its complete spherical symmetry. It can be represented in spherical coordinates in the form

$$\boldsymbol{H}(r,\theta,\phi) = \sqrt{\tfrac{3}{2}}\, h(r) (\hat{\boldsymbol{e}}_r \otimes \hat{\boldsymbol{e}}_r - \tfrac{1}{3} \boldsymbol{I}). \tag{2}$$

Substituting this ansatz into the expression for the free energy and performing the integration with respect to $\sin\theta\, d\theta\, d\phi$ over the azimuthal and polar angles, one obtains the following expression for the free energy of a droplet of radius R:

$$\mathcal{F}(\boldsymbol{H}) = 4\pi \int_0^R \left\{ \left(1 + \frac{2}{3}\eta_{23}\right) \left[(h')^2 + \frac{6}{r^2} h^2 \right] + g(h) \right\} r^2 dr + 2\pi (2\eta_2 - \eta_3) R\, h(R)^2,$$

where

$$\eta_{23} := \eta_2 + \eta_3 = \frac{L_2 + L_3}{L_1} \quad \text{and} \quad g(h) := \frac{t}{2} h^2 - h^3 + \frac{1}{2} h^4.$$

Much of what develops with the solution $h(r)$ is driven by the *double-well potential* $g(h)$. This function depends on the reduced-temperature parameter t, and it has a negative relative minimum, at a certain value $h_- < 0$, for $t < 0$. For $t < 1$ its global minimum occurs at the *positive* value

$$h_+ := \frac{3 + \sqrt{9 - 8t}}{4}.$$

The Euler-Lagrange equation for equilibrium is given by

$$\left(1 + \frac{2}{3}\eta_{23}\right) \left(h'' + \frac{2}{r} h' - \frac{6}{r^2} h \right) - g'(h) = 0, \tag{3}$$

where $g'(h) = t\,h - 3h^2 + 2h^3$. Spherical symmetry forces $h(0) = h'(0) = 0$. We will seek solutions of this singular nonlinear ordinary differential equation that satisfy the additional condition $h(R) = h_+$ or the limiting form of this, $h(\infty) = h_+$.

An analytical solution for Eq. (3) is not available. However, it is not hard to obtain accurate numerical solutions to this equation, and these are utilized below. Also, one can use analytical techniques involving ''differential inequalities'' to obtain upper and lower bounds for the solution. These provide useful information about the behavior of the solution

$h(r)$, and they prove to be adequate to demonstrate its instability over a large portion of the parameter space. We develop this in greater detail in [10] and report here the main estimates that we require.

Guided by analysis of the local behavior $h(r)=O(r^2)$, as $r\to 0$, and $h(r)=h_+ - O(1/r^2)$, as $r\to\infty$, we can construct ''lower'' and ''upper solutions'' to Eq. (3) in the form

$$\alpha(r)=h_+\frac{r^2}{r^2+\lambda_\alpha^2} \quad \text{and} \quad \beta(r)=h_+\frac{r^2}{r^2+\lambda_\beta^2}, \tag{4}$$

with ''length-scale parameters''

$$\lambda_\alpha^2 := \left(1+\frac{2}{3}\eta_{23}\right)\frac{14}{-t},$$

$$\lambda_\beta^2 := \left(1+\frac{2}{3}\eta_{23}\right)\frac{6}{\sqrt{9-8t}}\,\frac{1}{h_+}.$$

These bound the true solution,

$$\alpha(r)\leq h(r)\leq \beta(r), \quad 0<r<\infty,$$

for the semi-infinite interval case: $h(\infty)=h_+$. The validity of this bracketing is established in [10], where the quality of the upper/lower bounding solutions also is illustrated.

These bounding functions give (rigorously) the scaling of the core (or inner layer) of the hedgehog solution as $O(1/\sqrt{-t})$, as $t\to-\infty$. We require sharp information about the behavior of the function $h(r)$ in this limit. Motivated by the above, we seek solutions of Eq. (3) in the scaled form

$$\hbar(\bar{r}) := \frac{h(r)}{h_+}, \quad \bar{r} := \sqrt{\frac{-t}{1+\frac{2}{3}\eta_{23}}}\, r. \tag{5}$$

Making this substitution and taking the limit as $t\to-\infty$, we obtain the ''limiting rescaled problem''

$$\hbar'' + \frac{2}{\bar{r}}\hbar' - \frac{6}{\bar{r}^2}\hbar + \hbar - \hbar^3 = 0, \quad 0<\bar{r}<\infty,$$

$$\hbar(0)=0, \quad \hbar(\infty)=1, \tag{6}$$

the solution of which we shall denote by $\hbar_\infty$. The uniqueness of the solution to Eq. (6) (within a certain restricted class) also is established in [10].

IV. STABILITY

We seek to demonstrate the instability of the radial hedgehog solution (for R and $-t$ sufficiently large) by explicitly constructing a perturbation to which it becomes unstable. Motivated by numerical evidence, the analyses of [2] and [4], and the desire to obtain a tractable problem, we consider tensor fields of the form $\boldsymbol{Q}=\boldsymbol{H}+\boldsymbol{P}$, where $\boldsymbol{H}$ is the tensor field of the radial hedgehog solution, as in Eq. (2), and $\boldsymbol{P}$ is a perturbation of the form

$$\boldsymbol{P}(r,\theta,\phi)=\sqrt{\tfrac{3}{2}}\,p(r)(\hat{\boldsymbol{e}}_z\otimes\hat{\boldsymbol{e}}_z-\tfrac{1}{3}\boldsymbol{I}).$$

Expanding $\mathcal{F}(\boldsymbol{H}+\boldsymbol{P})$ and integrating with respect to $\sin\theta\, d\theta\, d\phi$, one obtains the following weak form of the *second variation*:

$$4\pi\int_0^R \{(1+\tfrac{1}{3}\eta_{23})(p')^2+[t+\tfrac{14}{5}h^2(r)]p^2\}r^2 dr.$$

We will know that the hedgehog solution is unstable if we can construct a function $p(r)$ that produces a *negative* value for this integral.

We normalize and recast the *instability condition* as

$$\inf_p \frac{\int_0^R [(1+\frac{1}{3}\eta_{23})(p')^2+\frac{14}{5}h^2(r)\,p^2]\,r^2\,dr}{\int_0^R p^2 r^2\,dr} < -t,$$

where the infimum is taken over smooth functions p satisfying $p(R)=0$. Rescaling as before in Eqs. (5), renormalizing, and taking limits as $t\to-\infty$, we obtain the scaled limiting form of the instability condition:

$$\lambda_{\min}(\varepsilon) := \inf_{\bar{p}} \frac{\varepsilon\int_0^\infty (\bar{p}')^2\bar{r}^2\,d\bar{r} + \int_0^\infty \hbar_\infty^2(\bar{r})\,\bar{p}^2\bar{r}^2\,d\bar{r}}{\int_0^\infty \bar{p}^2\bar{r}^2\,d\bar{r}} < \frac{5}{7}, \tag{7}$$

where the ''coupling coefficient'' ε is given by

$$\varepsilon := \frac{5}{7}\,\frac{1+\frac{1}{3}\eta_{23}}{1+\frac{2}{3}\eta_{23}}.$$

This parameter is a *decreasing* function of η_{23}: $-3/2<\eta_{23}<\infty \leftrightarrow \infty>\varepsilon>5/14$. Here we have also extended the integration to the limit $R\to\infty$.

One can show that the instability condition for finite R and t depends continuously on (ε,R,t) (see [10]). We conclude that if the (scaled limiting) instability condition (7) is satisfied for some ε, then the finite instability condition will be satisfied for all $R>R_0$ and $t<t_0$, for some R_0, t_0, which may depend on ε.

The value $\lambda_{\min}(\varepsilon)$ corresponds to a spherically symmetric Schrödinger eigenvalue problem with potential $\hbar_\infty^2(\bar{r})$. The form of $\hbar_\infty(\bar{r})$ is illustrated in Fig. 3 together with the associated principal mode $\bar{p}(\bar{r})$, which produces the minimum value in Eq. (7). The value of $\lambda_{\min}(\varepsilon)$ increases with ε, ranging over $0<\lambda_{\min}<\infty$, for $0<\varepsilon<\infty$. We define ε^* via $\lambda_{\min}(\varepsilon^*)=5/7$. Then we will have $\lambda_{\min}(\varepsilon)<5/7$ if and only if $\varepsilon<\varepsilon^*$, in which case the hedgehog tensor field $\boldsymbol{H}$ will be unstable to a perturbation $\boldsymbol{P}$ in our class.

Lower bounds for ε^* can be derived by bounding $\hbar_\infty(\bar{r})$ from above by the limiting form of the rescaled upper bounding function from Eq. (4),

$$\hbar_\infty(\bar{r}) \leq \bar{\beta}_\infty(\bar{r}) = \frac{\bar{r}^2}{\bar{r}^2+3},$$

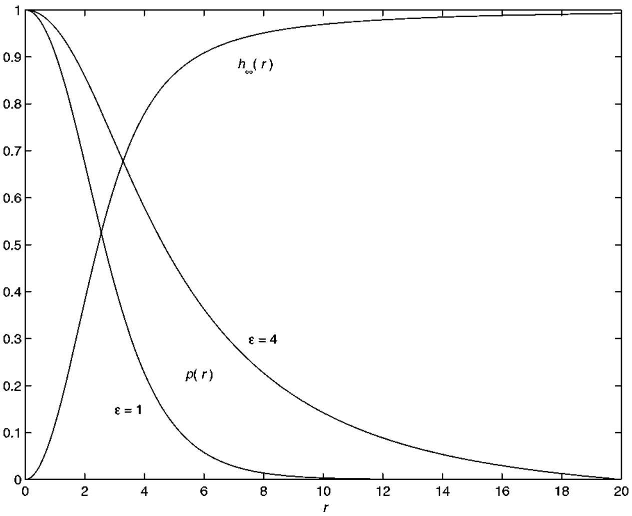

FIG. 3. Rescaled limiting form of radial hedgehog solution (''inner solution'') $\hbar_\infty(\bar{r})$ vs the minimum eigenmode $\bar{p}(\bar{r})$ in the instability criterion for two cases of the coupling coefficient: $\varepsilon=1$ and $\varepsilon=4$.

and evaluating the Rayleigh quotient in Eq. (7) on particular test functions $\bar{p}(\bar{r})$. After some trial and error, we obtain the function

$$\bar{p}(\bar{r}) = \frac{1}{(\bar{r}^2+12)^2},$$

which mimics the principal mode of Eq. (7) (with $\hbar_\infty$ replaced by $\bar{\beta}_\infty$ and $\varepsilon \approx 1$) and for which the integrals there take a simple form and give

$$\lambda_{\min}(\varepsilon) \leqslant \frac{\varepsilon \frac{3}{16} + \frac{28}{81}}{\frac{3}{4}}.$$

From this follows

$$\varepsilon^* \geqslant \tfrac{1724}{1701} \doteq 1.014 \quad \text{and} \quad \eta_{23}^* \leqslant -\tfrac{1527}{2233} \doteq -0.684.$$

These rigorous, purely analytical estimates are sufficient to treat the two-constant ($L_3=0$) model, for which we must have, by virtue of the conditions (1), $\eta_{23} > -3/5$ (and as a consequence $\varepsilon < 20/21$). That is, in the $L_3=0$ model (as considered, for example, in [2]), for any admissible values of the elastic constants L_1 and L_2, the radial hedgehog configuration must become unstable at sufficiently low temperature in a droplet of sufficiently large radius.

In the *full* model (with L_1, L_2, *and* L_3), the restriction on the elastic-constant ratios is $\eta_{23} > -3/2$, and so the range of admissible values for ε is $5/14 < \varepsilon < \infty$. Our instability condition must fail for ε large enough, i.e., for η_{23} sufficiently close to $-3/2$. We resort to numerical calculations to obtain sharper estimates. We calculate $\hbar_\infty(\bar{r})$ by solving numerically the problem (6). Using this, we then discretize the eigenvalue problem associated with Eq. (7),

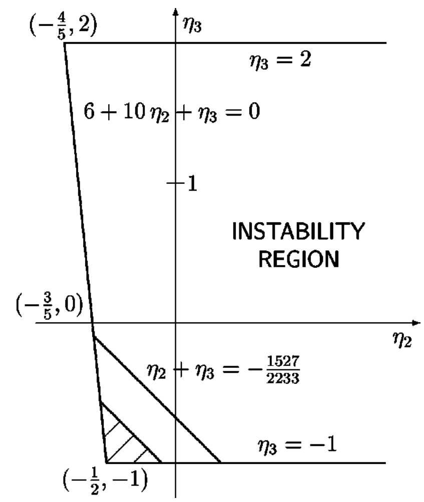

FIG. 4. Region of instability for radial hedgehog configuration in terms of elastic-constant ratios $\eta_2 = L_2/L_1$, $\eta_3 = L_3/L_1$. The hedgehog is proven here to be unstable throughout the entire admissible range ($-1 < \eta_3 < 2$, $6 + 10\eta_2 + \eta_3 > 0$) with the exception of the hashed region near the lower left corner above. Analytical bounds are sufficient to produce the outer enclosure $\eta_2 + \eta_3 \leqslant -1527/2233$ of this excluded portion.

$$\varepsilon\left(\bar{p}'' + \frac{2}{\bar{r}}\bar{p}'\right) + [\lambda - \hbar_\infty^2(\bar{r})]\bar{p} = 0, \quad 0 < \bar{r} < \infty,$$

$$\bar{p}'(0) = 0, \quad \bar{p}(\infty) = 0,$$

and calculate the minimum eigenvalue using library software. We obtain

$$\varepsilon^* \doteq 1.720 \quad \text{and} \quad \eta_{23}^* \doteq -1.107.$$

V. CONCLUSIONS

Thus we have shown that the hedgehog equilibrium solution must be unstable at sufficiently low temperatures and sufficiently large radii of the droplet for all but this small region of admissible elastic constants, $-1.5 < \eta_{23} \leqslant \eta_{23}^*$. These various regions of instability and limits are depicted in Fig. 4. This instability region is only a ''lower bound'' on the true range of elastic-constant ratios for which the hedgehog will be unstable; the region can only grow as one broadens the class of perturbations. The analysis suggests the possibility that the radial hedgehog may in fact become unstable for *all* admissible values of the elastic constants.

In considering the papers that we have cited previously, we find that our most direct comparisons can be made with [6] and [4]. In [6] Cohen and Taylor used a Frank elastic model, for which the radial hedgehog solution takes the form of the pure-splay distortion $\hat{\mathbf{n}}(r,\theta,\phi) = \hat{\mathbf{e}}_r$. They found, in terms of our parameters, that the hedgehog solution is metastable against the uniaxial perturbations admitted by Frank's model if and only if $\eta_{23} < 2/7$. Rosso and Virga [4] considered the same Landau–de Gennes model we have used here; however, they used a certain ''outer approximation'' for the (unknown) hedgehog solution. They then investigated metastability with respect to particular classes of perturbations, which differ somewhat from the ones we have used here.

They found the approximate hedgehog to be unstable (for a sufficiently large radius) if $1+2\eta_2+\eta_3>0$. The instability region we obtain here contains the regions from both of these papers.

As a final point, we mention that while the analysis was conducted on a certain limiting problem (in the doubly infinite limit $R\to\infty$, $t\to-\infty$), the reality is that these phenomena are observed numerically for rather modest values of these variables: $R\approx10$, $t\approx-5$ (as is seen in the figures).

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