Soft Elasticity is Not Necessary for Striping in Nematic Elastomers

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The occurrence of striped domains in stretched nematic elastomers has been suggested as evidence for soft elasticity. Conversely, the neo-classical model of Bladon, Terentjev and Warner, which displays soft elasticity, predicts striping. Here we show that the postulated director rotations and shears in the domain regions are also predicted by more general constitutive models that do not involve any notion of softness. Striping in nematic elastomers may therefore be a more general phenomena that is not necessarily an indication of soft elasticity. Furthermore, constitutive models more general than the neo-classical model may also explain the behavior of some nematic elastomers that do not appear to exhibit striping.

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I. INTRODUCTION

Kundler & Finklemann [1] observed the formation of striped domains during the mechanical extension of nematic elastomeric sheets with the director initially aligned perpendicular to the extension. As a possible explanation of this striping instability, Verwey et al. [2] suggested the mechanism of soft elasticity in which certain elastic moduli are small so that rotations of the director can occur with little or no energy cost. Each domain is then interpreted as consisting of material with a uniformly rotated director. In fact, the neo-classical model of Bladon et al. [3–5] predicts such director rotations as minimizers of the free energy for a limited range of extensions. Conti et al. [6, 7] further used this idea of soft elasticity to numerically study the striping instability in nematic elastomeric sheets with clamped boundary conditions in order to better model the experiments.

There has been, however, some controversy over the existence of soft elasticity and the validity of the neo-classical model. For example, the recent rheological experiments of Martinoty et al. [8] appear to contradict the assumptions of soft elasticity. Furthermore, the experiments of Mitchell et al. [9] on nematic elastomers did not exhibit the striping instability, but rather an apparent jump discontinuity in the director orientation.

Here we analyze the uniform extension of nematic elastomers. In particular, we seek to determine whether the occurrence of striping in nematic elastomers can be used as evidence of soft elasticity. Our approach involves free-energy minimization using a simple constitutive model proposed by Fried & Sellers.[10] The model involves two strain tensors: the left Cauchy–Green strain that describes the overall macroscopic strain, and a microstructural relative strain that describes the strain of the microstructural degrees of freedom relative to the overall macroscopic strain. The model includes both the neo-Hookean and the neo-classical models as special cases. We may therefore use the model to study mechanical extension as a function of the material parameters entering the free-energy density and thereby determine the conditions under which striping may occur.

Our results show that the free-energy density we consider allows director rotations for a finite range of extensions without the need for any elastic modulus to be small as in soft (or semi-soft) elasticity. Striping in nematic elastomers therefore appears to be a phenomenon that may occur independent of any notion of softness. The range of extensions in which striping is allowed does, however, depend on the relative magnitudes of the two elastic moduli entering the free-energy density. This range decreases as the magnitude of the relative strain term in the free energy decreases, finally reaching the point where the transition can experimentally appear to be discontinuous. This may explain the apparent discontinuous transition observed by Mitchell et al. [9] in their stretching experiments.

II. CONSTITUTIVE MODEL

The constitutive model of Fried & Sellers [10] treats the nematic elastomer as a material with microstructure and uses a symmetric and positive-definite conformation tensor $\boldsymbol{A}$ to account for the influence of nematic ordering on the conformation of the polymer chains. A referential conformation tensor, denoted by $\boldsymbol{A}_*$, is used to account for any anisotropy in the reference state. The free-energy density is taken to depend on the macroscopic deformation gradient $\boldsymbol{F}$ and, in addition, on $\boldsymbol{A}$ and $\boldsymbol{A}_*$. Standard invariance arguments show that ψ must be of the form:[10]

$$\psi = \hat{\psi}(\boldsymbol{F}\boldsymbol{F}^{\top}, \boldsymbol{A}^{-1}\boldsymbol{F}\boldsymbol{A}_*\boldsymbol{F}^{\top}, \boldsymbol{A}). \tag{1}$$

The quantity $\boldsymbol{F}\boldsymbol{F}^{\top}$ is the left Cauchy–Green strain tensor, a common measure of macroscopic strain in isotropic

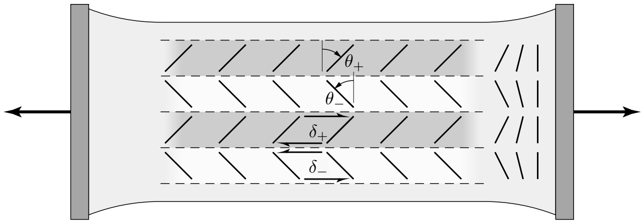

FIG. 1: Schematic of a uniaxially stretched nematic elastomeric specimen showing stripes consisting of bands with alternating pairs (θ_+, δ_+) and (θ_-, δ_-) of director orientations and shears.

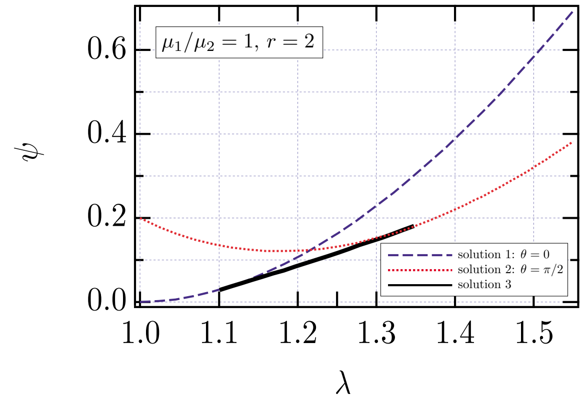

FIG. 2: The free energy corresponding to the 3 solutions with elastic modulus ratio $\mu_1/\mu_2 = 1$ and $r = 2$.

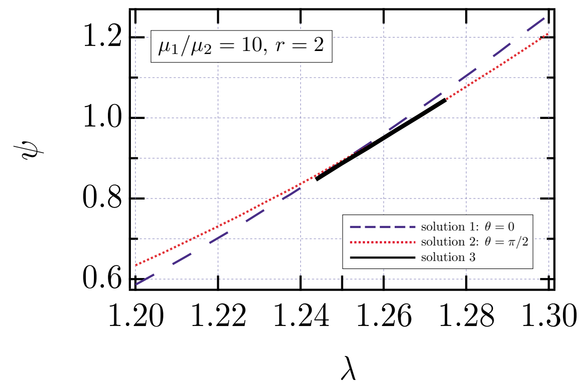

FIG. 3: The free energy corresponding to the 3 solutions with elastic modulus ratio $\mu_1/\mu_2 = 10$ and $r = 2$.

non-linear elasticity. The quantity $\boldsymbol{A}^{-1}\boldsymbol{F}\boldsymbol{A}_*\boldsymbol{F}^\top$ is a relative strain tensor that measures the strain of the microstructure relative to the overall macroscopic strain. When the microstructure convects with the macroscopic deformation—so that $\boldsymbol{A} = \boldsymbol{F}\boldsymbol{A}_*\boldsymbol{F}^\top$, the relative strain tensor reduces to the identity. In this model the total strain is therefore expressed by two strain measures: (i) the left Cauchy–Green strain for the macroscopic degrees of freedom; (ii) the relative strain for the microscopic degrees of freedom.

Here we assume that the nematic elastomer is incompressible, so that $\det\boldsymbol{F} = 1$, and we assume that the eigenvalues of $\boldsymbol{A}$ are unaffected by the deformation, so that $\boldsymbol{A}$ is a rotation of $\boldsymbol{A}_*$. This is approximately valid far from the nematic-isotropic transition temperature. In this case, a simple properly invariant expression for ψ linear in the two strain tensors is

$$\psi = \frac{\mu_1}{2}\mathsf{tr}\,(\boldsymbol{F}\boldsymbol{F}^\top - \boldsymbol{I}) + \frac{\mu_2}{2}\mathsf{tr}\,(\boldsymbol{A}^{-1}\boldsymbol{F}\boldsymbol{A}_*\boldsymbol{F}^\top - \boldsymbol{I}), \tag{2}$$

where the μ_i are non-negative elastic moduli. This expression reduces to the neo-classical free energy when μ_1 vanishes. Likewise, it reduces to the neo-Hookean expression when μ_2 vanishes. Here we do not assume that μ_1 is small relative to μ_2.

III. MECHANICAL EXTENSION

We now consider a problem involving the mechanical extension of a uniaxial nematic elastomer. We assume that the elastomer is uniformly aligned, then stretched by the amount λ in the direction perpendicular to the director. We choose a Cartesian coordinate system with the direction of the extension along the x-axis and director initially aligned along the y-axis. The conformation tensor of the reference configuration then has the form

$$\boldsymbol{A}_* = \begin{bmatrix} 1 & 0 & 0 \\ 0 & r & 0 \\ 0 & 0 & 1 \end{bmatrix}, \tag{3}$$

where r is a material parameter that indicates the anisotropy of the elastomer: $r > 1$ corresponds to prolate chain shapes, $r < 1$ to oblate chain shapes, and $r = 1$ to isotropic chain shapes. For the conformation tensor in the deformed configuration, we allow for a director rotation with angle θ, so that

$$\boldsymbol{A}^{-1} = \begin{bmatrix} \cos^2\theta + r^{-1}\sin^2\theta & \frac{1}{2}(r^{-1}-1)\sin 2\theta & 0 \\ \frac{1}{2}(r^{-1}-1)\sin 2\theta & \sin^2\theta + r^{-1}\cos^2\theta & 0 \\ 0 & 0 & 1 \end{bmatrix}. \tag{4}$$

A choice of the deformation gradient that obeys $\det\boldsymbol{F} = 1$ and allows for an extension λ as well as a shear δ consistent with director rotations is given by

$$\boldsymbol{F} = \begin{bmatrix} \lambda & \delta & 0 \\ 0 & \lambda_{yy} & 0 \\ 0 & 0 & 1/(\lambda\lambda_{yy}) \end{bmatrix}. \tag{5}$$

Substitution of (3)–(5) into the free-energy density (2) yields:

$$\psi = \frac{\mu_1}{2}(\lambda^2 + \lambda_{yy}^2 + \lambda_{yy}^{-2}\lambda^{-2} + \delta^2 - 3) + \frac{\mu_2}{2}\big(\lambda^2 + \lambda_{yy}^2 + \lambda_{yy}^{-2}\lambda^{-2} + r\delta^2 - 3 - (r-1)(\lambda_{yy}\delta \sin 2\theta + (\lambda^2 r^{-1} - \lambda_{yy}^2 + \delta^2)\sin^2\theta)\big). \quad (6)$$

Equilibrium states minimize the free energy and satisfy the following three equations:

$$\left.\begin{aligned} \frac{\partial\psi}{\partial\delta} &= \mu_1\delta + \mu_2 r\delta - \mu_2(r-1)(\tfrac{1}{2}\lambda_{yy}\sin 2\theta + \delta\,\sin^2\theta) = 0, \\ \frac{\partial\psi}{\partial\theta} &= \mu_2(r-1)\big(\lambda_{yy}\,\delta(2\sin^2\theta - 1) - \tfrac{1}{2}(\lambda^2 r^{-1} - \lambda_{yy}^2 + \delta^2)\sin 2\theta\big) = 0, \\ \frac{\partial\psi}{\partial\lambda_{yy}} &= \mu_1(\lambda_{yy} - \lambda_{yy}^{-3}\lambda^{-2}) + \mu_2\big(\lambda_{yy} - \lambda_{yy}^{-3}\lambda^{-2} + (r-1)(\lambda_{yy}\,\sin^2\theta - \tfrac{1}{2}\delta\,\sin 2\theta)\big) = 0. \end{aligned}\right\} \quad (7)$$

In the above equations, λ is the imposed extension, and the quantities δ, θ, and λ_{yy} are to be determined.

There are exactly 3 physically-relevant classes of solutions to (7):

1. $\theta = 0, \quad \delta = 0, \quad \lambda_{yy} = \lambda^{-1/2}, \quad$ for $\lambda \geq 1$;
2. $\theta = \pi/2, \ \delta = 0, \ \lambda_{yy} = \left(\lambda_c^3/r\right)^{1/4}\lambda^{-1/2}$, for $\lambda \geq 1$;
3. $\theta = \theta_\pm(\lambda) = \pm\tan^{-1}\sqrt{\frac{r(\lambda^2-\lambda_c^2)}{\lambda_c^2(r-\lambda^2\lambda_c)}}$,
   $\delta = \delta_\pm(\lambda) = \pm\lambda^{-1}\sqrt{(\lambda^2-\lambda_c^2)(1/\lambda_c - \lambda^2/r)}$,
   $\lambda_{yy} = \lambda_c^{1/2}\lambda^{-1}$, $\quad$ for $\lambda_c \leq \lambda \leq (r/\lambda_c)^{1/2}$.

Here

$$\lambda_c = \left(\frac{\mu_1 + \mu_2}{\mu_1 r^{-1} + \mu_2}\right)^{1/3}.$$

The first solution is the standard elastic solution with no rotation, so that the director remains perpendicular to the axis of extension. The second solution has the director rotated $\pi/2$ degrees, so that the director is parallel to the axis of extension. The shear δ vanishes for both the first and the second solutions. The third class of solutions describes two oppositely-oriented director orientations $\theta_\pm(\lambda)$ ranging from $\theta = 0$ at $\lambda = \lambda_c$ to $\theta = \pi/2$ at $\lambda = (r/\lambda_c)^{1/2}$ as well as two oppositely oriented shears $\delta_\pm(\lambda)$. For each value λ of the extension in this transition region, there are two possible pairs of director orientations and shears (θ_+, δ_+) and (θ_-, δ_-). These pairs can be used to construct striped solutions (Figure 1).

Figures (2) and (3) illustrate the free energy for the three solutions as a function of the extension λ. For small extensions, the first solution is the absolute minimizer of the free energy. For very large extensions, the second solution is the absolute minimizer. But there is also a transition region $\lambda_c \leq \lambda \leq (r/\lambda_c)^{1/2}$ where the third class of solutions are the absolute minimizers. As the extension is increased from λ_c to $(r/\lambda_c)^{1/2}$ in this transition region, the director continuously rotates from the initial perpendicular state to the final parallel state. In concert with this, a nontrivial shear develops.

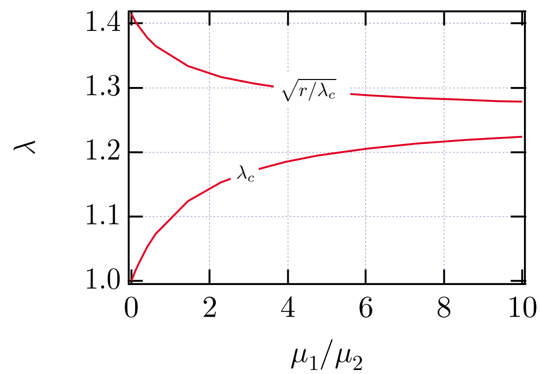

FIG. 4: As the ratio μ_1/μ_2 increases, the rotation of the director takes place over a more limited range of strains. The bounds both converge to $r^{1/3}$ in the limit $\mu_1/\mu_2 \to \infty$.

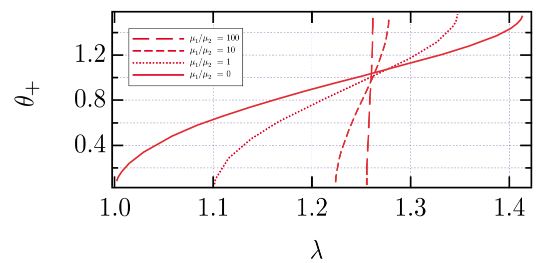

FIG. 5: The director orientation θ_+ as a function of extension λ.

Figure (4) shows how the bounds λ_c and $(r/\lambda_c)^{1/2}$ of the transition region vary with μ_1/μ_2. The model predicts that this transition region will always exist, but that the range of such extensions decreases as μ_1/μ_2 increases. If the ratio μ_1/μ_2 is significantly increased, the transition region will eventually shrink to the extent that it may appear experimentally unobservable. In this case, the first solution may appear to transform discontinuously to the second solution with a jump in the director. Were this the case, striping would not be observed. This might explain the observations of Mitchell et al. [9]

Figures (5) and (6) show the angle θ and shear δ of the third solution as the elastic modulus ratio μ_1/μ_2 varies. They clearly illustrate the change in the size of the transition region as a function of the ratio μ_1/μ_2 of the elastic moduli.

For the special case $\mu_1 = 0$ where (2) reduces to the neo-classical expression, $\lambda_c = 1$ and the 3 solutions above

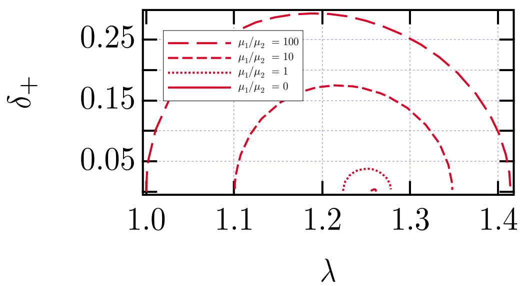

FIG. 6: The shear δ_+ as a function of extension λ

reduce to those given on page 159 of Warner & Terentjev [5].

IV. DISCUSSION

We have shown that, during the extension of a nematic elastomer modeled by the simple free-energy density (2), there exists a transition region in which the director continuously rotates from perpendicular alignment to parallel alignment regardless of the ratio μ_1/μ_2 of the elastic moduli. If we interpret the stripes observed in extensional experiments of the kind performed by Kundler & Finkelmann[1] as alternating domains (θ_+, δ_+) and (θ_-, δ_-), then a model based on (2) seems to predict striping as a general phenomenon that may occur independent of softness.

This result raises the question whether such transition regions are predicted by other free-energy densities. For example, one could also consider more general expressions involving higher order terms, such as a term proportional to the second invariant of $\boldsymbol{F}\boldsymbol{F}^{\top}$ as done in the Mooney energy density.[10] It is easily shown that such a free-energy density also predicts transition regions where an initially perpendicular director rotates upon extension.

In fact, as long as the free-energy density depends explicitly on the relative strain tensor $\boldsymbol{A}^{-1}\boldsymbol{F}\boldsymbol{A}_*\boldsymbol{F}^{\top}$, then one should expect the existence of solutions with transition regions. We can intuitively understand this idea as follows. A term proportional to the relative strain tensor penalizes deformations in which the microstructure does not convect with the macroscropic deformation—that is deformations for which $\boldsymbol{A} \neq \boldsymbol{F}\boldsymbol{A}_*\boldsymbol{F}^{\top}$. The neoclassical free-energy density is an example of a free-energy density which is linear in the relative strain, where the so-called soft deformations are simply deformations for which $\boldsymbol{A} = \boldsymbol{F}\boldsymbol{A}_*\boldsymbol{F}^{\top}$. On the other hand, terms involving the Cauchy–Green strain tensor penalize deformations in which the microstructure convects with the macroscopic deformation. For a free-energy density consisting of both such terms, such as (2), actual solutions will involve a competition between the terms tending to favor deformations in which the microstructure convects with the macroscopic deformation and those that penalize them. As the importance of the purely elastic terms increases, we would expect a diminishing tendency for the microstructure to convect with the macroscopic deformation, which is in fact illustrated in Figs (2) and (3).

Acknowledgments

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1044	Panat, R., K. J. Hsia, and D. G. Cahill	Evolution of surface waviness in thin films via volume and surface diffusion—*Journal of Applied Physics* (submitted)	Mar. 2004
1045	Riahi, D. N.	Steady and oscillatory flow in a mushy layer—*Current Topics in Crystal Growth Research*, in press (2004)	Mar. 2004
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1048	Brown, E. N., S. R. White, and N. R. Sottos	Fatigue crack propagation in microcapsule toughened epoxy—*Journal of Materials Science* (submitted)	Apr. 2004
1049	Zeng, L., S. Balachandar, and P. Fischer	Wall-induced forces on a rigid sphere at finite Reynolds number—*Journal of Fluid Mechanics* (submitted)	May 2004
1050	Dolbow, J., E. Fried, and H. Ji	A numerical strategy for investigating the kinetic response of stimulus-responsive hydrogels—*Computer Methods in Applied Mechanics and Engineering* **194**, 4447–4480 (2005)	June 2004
1051	Riahi, D. N.	Effect of permeability on steady flow in a dendrite layer—*Journal of Porous Media*, in press (2004)	July 2004
1052	Cermelli, P., E. Fried, and M. E. Gurtin	Transport relations for surface integrals arising in the formulation of balance laws for evolving fluid interfaces—*Journal of Fluid Mechanics* (submitted)	Sept. 2004
1053	Stewart, D. S., and A. R. Kasimov	Theory of detonation with an embedded sonic locus—*SIAM Journal on Applied Mathematics* (submitted)	Oct. 2004
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1055	Ji, H., H. Mourad, E. Fried, and J. Dolbow	Kinetics of thermally induced swelling of hydrogels—*International Journal of Solids and Structures* (submitted)	Dec. 2004
1056	Fulton, J. M., S. Hussain, J. H. Lai, M. E. Ly, S. A. McGough, G. M. Miller, R. Oats, L. A. Shipton, P. K. Shreeman, D. S. Widrevitz, and E. A. Zimmermann	Final reports: Mechanics of complex materials, Summer 2004 (K. M. Hill and J. W. Phillips, eds.)	Dec. 2004
1057	Hill, K. M., G. Gioia, and D. R. Amaravadi	Radial segregation patterns in rotating granular mixtures: Waviness selection—*Physical Review Letters* **93**, 224301 (2004)	Dec. 2004
1058	Riahi, D. N.	Nonlinear oscillatory convection in rotating mushy layers—*Journal of Fluid Mechanics* (submitted)	Dec. 2004
1059	Okhuysen, B. S., and D. N. Riahi	On buoyant convection in binary solidification—*Journal of Fluid Mechanics* (submitted)	Jan. 2005
1060	Brown, E. N., S. R. White, and N. R. Sottos	Retardation and repair of fatigue cracks in a microcapsule toughened epoxy composite—Part I: Manual infiltration—*Composites Science and Technology* (submitted)	Jan. 2005
1061	Brown, E. N., S. R. White, and N. R. Sottos	Retardation and repair of fatigue cracks in a microcapsule toughened epoxy composite—Part II: *In situ* self-healing—*Composites Science and Technology* (submitted)	Jan. 2005
1062	Berfield, T. A., R. J. Ong, D. A. Payne, and N. R. Sottos	Residual stress effects on piezoelectric response of sol-gel derived PZT thin films—*Journal of Applied Physics* (submitted)	Apr. 2005
1063	Anderson, D. M., P. Cermelli, E. Fried, M. E. Gurtin, and G. B. McFadden	General dynamical sharp-interface conditions for phase transformations in viscous heat-conducting fluids—*Journal of Fluid Mechanics* (submitted)	Apr. 2005
1064	Fried, E., and M. E. Gurtin	Second-gradient fluids: A theory for incompressible flows at small length scales—*Journal of Fluid Mechanics* (submitted)	Apr. 2005
1065	Gioia, G., and F. A. Bombardelli	Localized turbulent flows on scouring granular beds—*Physical Review Letters*, in press (2005)	May 2005
1066	Fried, E., and S. Sellers	Orientational order and finite strain in nematic elastomers—*Journal of Chemical Physics* **123**, 044901 (2005)	May 2005
1067	Chen, Y.-C., and E. Fried	Uniaxial nematic elastomers: Constitutive framework and a simple application—*Proceedings of the Royal Society of London A* (submitted)	June 2005
1068	Fried, E., and S. Sellers	Incompatible strains associated with defects in nematic elastomers—*Physical Review Letters* (submitted)	Aug. 2005
1069	Gioia, G., and X. Dai	Surface stress and reversing size effect in the initial yielding of ultrathin films—*Journal of Applied Mechanics*, in press (2005)	Aug. 2005
1070	Gioia, G., and P. Chakraborty	Turbulent friction in rough pipes and the energy spectrum of the phenomenological theory—*arXiv:physics* 0507066 v1 8 Jul 2005	Aug. 2005
1071	Keller, M. W., and N. R. Sottos	Mechanical properties of capsules used in a self-healing polymer—*Experimental Mechanics* (submitted)	Sept. 2005
1072	Chakraborty, P., G. Gioia, and S. Kieffer	Volcán Reventador's unusual umbrella	Sept. 2005
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