Incompatible strains associated with defects in nematic elastomers

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In a nematic elastomer the deformation of the polymer network chains is coupled to the orientational order of the mesogenic groups. Statistical arguments have derived the so-called neoclassical free energy that models this coupling. Here we show that the neoclassical model supplemented by the usual Frank energy predicts incompatible network strains associated with the formation of standard nematic textures. The incompatibility is measured by the Riemann curvature tensor, which we find to be nonzero for both radial hedgehog defects and escaped disclinations of strength +1 in circular cylinders. Analogous problems for conventional nonlinearly elastic solids do not possess solutions with such incompatibilities. Compatibility in nematic elastomers would require either more complicated nematic textures in elastomers than in conventional (polymeric and low molecular weight) liquid crystals or a free-energy density more complicated than the neoclassical expression. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2149857]

I. INTRODUCTION

Liquid-crystal elastomers are a new class of materials with unusual properties made possible by interactions between the elasticity of the polymer network chains and the orientational order of the mesogenic groups. For example, nematic elastomers exhibit phase transitions, spontaneous shape changes, response to external fields, and microstructures such as striped domains.[1]

To model nematic elastomers, Warner *et al.*[2] and Bladon *et al.*[3] extended classical Gaussian network theory to allow for anisotropic distributions of the end-to-end vectors of the polymer chains and derived the contribution to the free-energy density due to elastic distortions of the network. The resulting expression, called neoclassical, is nonlinear, properly invariant, and depends on the deformation gradient and a symmetric and positive-definite tensorial measure of the polymer chain shape, called the step-length (or conformation) tensor.

Defects in the orientational order are common in traditional nematic liquid crystals and have also been reported for nematic elastomers. Here we use the neoclassical model supplemented with the usual Frank energy to study the compatibility of network strains associated with the formation of defects in the orientational order of uniaxial nematic elastomers. We find that for the formation of both radial hedgehog defects and escaped disclinations from an isotropic reference configuration, the fourth-order Riemann curvature tensor is nonzero. The network strain associated with the defects in orientational order is therefore incompatible, and the deformed uniaxial configuration cannot be obtained solely by a deformation from the reference configuration. These results appear surprising and suggest either inadequacies in the assumed defect model or in the assumed form of the free-energy density.

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II. NEOCLASSICAL MODEL

We consider a material prepared such that the alignment of the mesogens in the reference configuration is random. We also restrict attention to situations where the molecular conformation in the deformed configuration is uniaxial. In the absence of interactions between the network and a solvent, the free-energy density ψ of a nematic elastomer consists of three contributions differing in their physical origin and form:

$$\psi = \psi_n + \psi_e + \psi_f. \tag{1}$$

The nematic contribution ψ_n accounts for interactions between the mesogens and determines the phase stability of the reference configuration. The contribution ψ_e is due to the entropic distortion of the network and accounts for the coupling between the nematic order and the stretching of the cross-linked polymer chains. The remaining contribution ψ_f is the Frank energy and accounts for the spatial inhomogeneity in the distribution of the mesogens.

For the uniaxial case that we consider here, the nematic contribution ψ_n is a function of the nematic scalar order parameter S. For simplicity, we view S as a tunable control parameter (as opposed to an independent field quantity) that vanishes in the isotropic reference configuration and takes a specified value in the deformed configuration. We do not specify ψ_n other than to assume that both the isotropic reference and deformed uniaxial configurations are stable.

The statistical theory of Bladon *et al.*[3] yields the so-called neoclassical expression

$$\psi_e = \tfrac{1}{2}\mu(\mathrm{tr}(\boldsymbol{A}^{-1}\boldsymbol{F}\boldsymbol{A}_*\boldsymbol{F}^{\mathsf{T}}) - \log\det(\boldsymbol{A}^{-1}\boldsymbol{A}_*) - 3), \tag{2}$$

where $\boldsymbol{A}$ is the molecular conformation in the spatial configuration, $\boldsymbol{F}$ is the macroscopic distortion of the network, $\boldsymbol{A}_*$ is the molecular conformation in the reference configuration, and $\mu > 0$ is the constant shear modulus. The conformation tensors $\boldsymbol{A}_*$ and $\boldsymbol{A}$ are symmetric and positive-definite. $\boldsymbol{A}$ is a

function of S and the director $\boldsymbol{n}$ in the deformed configuration, and $\boldsymbol{A}_*$ is a function of S_* and the director $\boldsymbol{n}_*$ in the referential configuration. The macroscopic distortion $\boldsymbol{F}$ maps an infinitesimal material line element $d\boldsymbol{x}_*$ in the reference configuration into a line element $d\boldsymbol{x}=\boldsymbol{F}d\boldsymbol{x}_*$ in the deformed configuration and is therefore a two-point tensor field. Consistent with the incompressibility of rubberlike materials, $\det \boldsymbol{F}=1$. Importantly, the statistical theory leading to (2) is not predicated on the assumption that $\boldsymbol{F}$ is the gradient of a deformation. It merely assumes that the network distortion is a two-point tensor-valued mean field.

As the alignment of the mesogens in the reference configuration is random, $\boldsymbol{A}_*$ is spherical. Without loss of generality, we may take

$$\boldsymbol{A}_*=\boldsymbol{I}. \tag{3}$$

We can represent $\boldsymbol{A}$ in the generic uniaxial form

$$\boldsymbol{A}=a_\perp \boldsymbol{I}+(a_\parallel-a_\perp)\boldsymbol{n}\otimes\boldsymbol{n}, \quad a_\perp>0, \quad a_\parallel>0, \tag{4}$$

where $\boldsymbol{n}$ is the director in the deformed configuration and $a_\perp$ and $a_\parallel$ are functions of the nematic order parameter S. The three cases $a_\perp>a_\parallel$, $a_\perp=a_\parallel$, and $a_\perp<a_\parallel$ correspond, respectively, to oblate, isotropic, and prolate alignments.

Furthermore, using (3) and (4) in (2), we obtain

$$\begin{aligned}\psi_e &= \tfrac{1}{2}\mu(\operatorname{tr}(\boldsymbol{A}^{-1}\boldsymbol{B})+\log\det\boldsymbol{A}-3)\\ &= \tfrac{1}{2}\mu(a_\perp^{-1}\operatorname{tr}\boldsymbol{B}-(a_\parallel^{-1}-a_\perp^{-1})\boldsymbol{n}\cdot\boldsymbol{B}\boldsymbol{n}+\log(a_\perp^2 a_\parallel)-3),\end{aligned} \tag{5}$$

where $\boldsymbol{B}=\boldsymbol{F}\boldsymbol{F}^{\mathsf{T}}$ denotes the Finger[4] (or left Cauchy-Green) strain tensor. Since $\det\boldsymbol{F}=1$, ψ_e as given by (5) serves as a measure of free-energy per unit volume in both reference and deformed configurations. Furthermore, we emphasize that $\boldsymbol{B}$ maps infinitesimal line elements in the deformed configuration into infinitesimal line elements in the deformed configuration. Like the spatial conformation $\boldsymbol{A}$, $\boldsymbol{B}$ is therefore a spatial tensor field. In view of these observations, it is both legitimate and natural to view ψ_e as a measure of free-energy per unit volume in the deformed configuration. For simplicity, we neglect spatial variations in S and follow Terentjev *et al.*[5] and Osborne and Terentjev[6] and use the usual Frank[7] expression

$$\begin{aligned}\psi_f &= \tfrac{1}{2}k_1(\operatorname{div}\boldsymbol{n})^2+\tfrac{1}{2}k_2(\boldsymbol{n}\cdot\operatorname{curl}\boldsymbol{n})^2+\tfrac{1}{2}k_3|\boldsymbol{n}\times\operatorname{curl}\boldsymbol{n}|^2\\ &\quad+\tfrac{1}{2}(k_2+k_4)(\operatorname{tr}((\operatorname{grad}\boldsymbol{n})^2)-(\operatorname{div}\boldsymbol{n})^2),\end{aligned} \tag{6}$$

with constant splay, twist, bend, and saddle-splay moduli k_1, k_2, k_3, and k_4 assumed to obey the inequalities $2k_1>k_2+k_4$, $k_2>|k_4|$, and $k_3>0$ set forth by Ericksen[8] to ensure that $\psi_f\geqslant 0$. The spatial derivatives are with respect to the deformed configuration.

Consider now uniaxial equilibrium states in the deformed configuration that minimize the total free-energy density ψ. We take the order parameter S as a specified control parameter and minimize over states subject to the constraints $\det\boldsymbol{F}=1$ and $|\boldsymbol{n}|=1$. The contribution ψ_e is then minimized by the state

$$\boldsymbol{B}=\kappa\boldsymbol{A}, \tag{7}$$

where

$$\kappa=(\det\boldsymbol{A})^{-1/3}=(a_\perp^2 a_\parallel)^{-1/3}. \tag{8}$$

Substitution into ψ_e yields

$$\psi_e(\kappa\boldsymbol{A},\boldsymbol{A})=\tfrac{3}{2}\mu(\kappa-\log\kappa-1)\geqslant 0, \tag{9}$$

which is a function of S through $a_\perp$ and $a_\parallel$. This provides an energy penalty for changes in $\det\boldsymbol{A}$, i.e., changes in volume occupied by a generic polymer chain.

Thus, for a nematic elastomer that is isotropic in the reference configuration and for which the spatial conformation is uniaxial, the neoclassical model shows that the entropic contribution to the free-energy density is minimized if and only if the Finger strain tensor is a scalar multiple of the spatial conformation tensor. Moreover, granted that $\boldsymbol{B}=\kappa\boldsymbol{A}$, the problem of minimizing the total free-energy $\int_{\mathcal{R}}\psi d\upsilon$ of a nematic elastomer occupying a region $\mathcal{R}$ reduces to the problem of minimizing the free-energy $\int_{\mathcal{R}}(\psi_f+\psi_n+(3/2)\mu(\kappa-\log\kappa-1))d\upsilon$. And since S is regarded as specified, the problem reduces to minimizing the Frank free-energy $\int_{\mathcal{R}}\psi_f d\upsilon$, which is also the free energy of a conventional uniaxial nematic liquid crystal. So any solution to a problem of static equilibrium in a conventional uniaxial nematic liquid crystal describes a corresponding equilibrium state in a nematic elastomer.

DeSimone and Dolzmann[9] and Conti *et al.*[10] studied the additional constraint $\det\boldsymbol{A}=\det\boldsymbol{A}_*=1$, which requires that $\kappa=1$, i.e., the volume occupied by a generic polymer chain remains unchanged regardless of shape changes. In this special case, $\boldsymbol{A}$ reduces to

$$\boldsymbol{A}=(a_\perp/a_\parallel)^{1/3}(\boldsymbol{I}+(a_\parallel/a_\perp-1)\boldsymbol{n}\otimes\boldsymbol{n}). \tag{10}$$

As shown by DeSimone and Dolzmann,[9] ψ_e as given in (5) is bounded below by zero:

$$\min_{\det\boldsymbol{A}=\det\boldsymbol{F}=1,|\boldsymbol{n}|=1}\psi_e=0. \tag{11}$$

For this special constrained case, we may therefore conclude that $\psi=\psi_n+\psi_f$ if and only if $\boldsymbol{B}=\boldsymbol{A}$.

III. INCOMPATIBLE STRAINS

For static equilibrium strains $\boldsymbol{B}$ determined by the above minimization to be compatible with a deformation from some reference region $\mathcal{R}_*$ to the region $\mathcal{R}$ occupied by the specimen, it is necessary and sufficient that the Riemann curvature tensor $\boldsymbol{\mathcal{R}}$ based on $\boldsymbol{B}^{-1}$ vanishes (Truesdell and Toupin,[11] Sec. 34). In components, $\boldsymbol{\mathcal{R}}$ is given by[12]

$$\begin{aligned}R_{lijk} &= \frac{1}{2}\left(\frac{\partial^2 B^{-1}_{lk}}{\partial x^i\partial x^j}+\frac{\partial^2 B^{-1}_{ij}}{\partial x^l\partial x^k}-\frac{\partial^2 B^{-1}_{lj}}{\partial x^i\partial x^k}-\frac{\partial^2 B^{-1}_{ik}}{\partial x^l\partial x^j}\right)\\ &\quad+B^{hm}([ij,m][lk,h]-[ik,m][lj,h])\end{aligned} \tag{12}$$

with

$$[ij,k]=\frac{1}{2}\left(\frac{\partial B^{-1}_{ik}}{\partial x^j}+\frac{\partial B^{-1}_{jk}}{\partial x^i}-\frac{\partial B^{-1}_{ij}}{\partial x^k}\right). \tag{13}$$

Furthermore, a condition necessary and sufficient for $\boldsymbol{F}$ to be the gradient of a deformation is that $\operatorname{curl}(\boldsymbol{F}^{-1})=\boldsymbol{0}$. Since $\boldsymbol{B}=\boldsymbol{F}\boldsymbol{F}^{\mathsf{T}}$ and $\boldsymbol{F}$ is nonsingular, the polar-decomposition theorem ensures the existence of a unique rotation $\boldsymbol{R}$ ($\boldsymbol{R}\boldsymbol{R}^{\mathsf{T}}=\boldsymbol{I}$, $\det\boldsymbol{R}=1$) such that $\boldsymbol{F}=\boldsymbol{B}^{1/2}\boldsymbol{R}$. So when $\boldsymbol{B}=\kappa\boldsymbol{A}$, we have $\boldsymbol{F}^{-1}=\kappa^{-1/2}\boldsymbol{R}^{\mathsf{T}}\boldsymbol{A}^{-1/2}$ and the condition $\operatorname{curl}(\boldsymbol{F}^{-1})=\boldsymbol{0}$ translates into a condition involving $\boldsymbol{R}$ and $\boldsymbol{n}$:

$$\operatorname{curl}(\boldsymbol{R}^{\mathsf{T}})=\left(1-\sqrt{\frac{a_{\perp}}{a_{\parallel}}}\right)\operatorname{curl}(\boldsymbol{R}^{\mathsf{T}}\boldsymbol{n}\otimes\boldsymbol{n}). \tag{14}$$

Given $\boldsymbol{n}$, (14) provides a differential equation for $\boldsymbol{R}$. The existence of a solution for $\boldsymbol{R}$ is then equivalent to the condition that $\boldsymbol{F}$ be the gradient of a deformation.

We now give two static equilibrium states that are free-energy minimizers, but that involve incompatible strains.

A. Hedgehog defects

We first consider a radially symmetric point defect—or hedgehog—located at the center of a spherical specimen of radius R. Specifically, consider a specimen occupying a spherical region $\mathcal{R}=\{\boldsymbol{x}:|\boldsymbol{x}|<R\}$ of space. Suppose that strong anchoring prevails on the boundary $\partial\mathcal{R}$ of the specimen, so that

$$\boldsymbol{n}(\boldsymbol{x})=\frac{\boldsymbol{x}}{R}\quad\text{when } |\boldsymbol{x}|=R. \tag{15}$$

Suppose also that $\partial\mathcal{R}$ is traction-free, so that the Cauchy stress tensor $\boldsymbol{T}=-p\boldsymbol{I}+(\partial\psi_e/\partial\boldsymbol{F})\boldsymbol{F}^{\mathsf{T}}-(\operatorname{grad}\boldsymbol{n})^{\mathsf{T}}(\partial\psi_f/\partial(\operatorname{grad}\boldsymbol{n}))$ obeys

$$\boldsymbol{T}(\boldsymbol{x})\boldsymbol{x}=\boldsymbol{0}\quad\text{when } |\boldsymbol{x}|=R. \tag{16}$$

Neglect all external body forces. Furthermore, assume that the director field has the form

$$\boldsymbol{n}(\boldsymbol{x})=\operatorname{grad}|\boldsymbol{x}|=\frac{\boldsymbol{x}}{|\boldsymbol{x}|} \tag{17}$$

and that $\boldsymbol{B}=\kappa\boldsymbol{A}$, with $\boldsymbol{A}$ as given in (4). It follows that $\operatorname{grad}\boldsymbol{n}=(\boldsymbol{I}-\boldsymbol{n}\otimes\boldsymbol{n})/|\boldsymbol{x}|$ and $\operatorname{curl}\boldsymbol{n}=\boldsymbol{0}$. Also, the strong-anchoring boundary condition (15) is satisfied trivially. Granted the above assumptions,

$$\boldsymbol{T}=-(p-\kappa\mu)\boldsymbol{I}+\frac{2k_1-k_2-k_4}{|\boldsymbol{x}|^2}(\boldsymbol{I}-\boldsymbol{n}\otimes\boldsymbol{n}), \tag{18}$$

whereby the traction-free boundary condition (16) implies that the pressure required to maintain the incompressibility of the medium must be given by $p=\kappa\mu$. For the hedgehog (17), the Frank free-energy $\int_{\mathcal{R}}\psi_f d\upsilon$ is stationary. Moreover, that energy is minimized if and only if the Frank moduli k_1, k_2, and k_3 obey the inequality[13]

$$8(k_2-k_1)+k_3>0. \tag{19}$$

We assume that the inequality (19) is satisfied. For this case, a direct but tedious calculation of the Riemann curvature tensor using (12) shows that it is nonzero. The Finger strain $\boldsymbol{B}$ associated with the formation of a hedgehog defect (energy minimizing or otherwise) from an isotropic state is therefore incompatible.

We also show explicitly that the corresponding distortion $\boldsymbol{F}$ is not the gradient of a deformation. Symmetry considerations dictate that the rotation $\boldsymbol{R}$ appearing in the representation $\boldsymbol{F}=\boldsymbol{A}^{1/2}\boldsymbol{R}$ for $\boldsymbol{F}$ must be constant and obey $\boldsymbol{R}\boldsymbol{n}=\boldsymbol{R}^{\mathsf{T}}\boldsymbol{n}=\boldsymbol{n}$. In this case,

$$\operatorname{curl}(\boldsymbol{F}^{-1})=\boldsymbol{0}\quad\text{if and only if }\operatorname{curl}(\boldsymbol{n}\otimes\boldsymbol{n})=\boldsymbol{0}. \tag{20}$$

Since $[\operatorname{curl}(\boldsymbol{n}\otimes\boldsymbol{n})]\boldsymbol{\upsilon}=[\operatorname{grad}\boldsymbol{n}](\boldsymbol{n}\times\boldsymbol{\upsilon})+(\boldsymbol{\upsilon}\cdot\operatorname{curl}\boldsymbol{n})\boldsymbol{n}$ for any vector $\boldsymbol{\upsilon}$, substitution of (17) shows that

$$[\operatorname{curl}(\boldsymbol{n}\otimes\boldsymbol{n})]\boldsymbol{\upsilon}=|\boldsymbol{x}|^{-1}\boldsymbol{n}\times\boldsymbol{\upsilon} \tag{21}$$

for all $\boldsymbol{\upsilon}$; hence, $\operatorname{curl}\boldsymbol{F}^{-1}\neq\boldsymbol{0}$.

B. Escaped disclinations

The previous case illustrated the incompatibility of the strain for the formation of a point defect. We also illustrate the possibility of incompatible strains with a line defect present. Consider, for example, an escaped disclination of strength +1 located on the axis of a circular cylindrical specimen of radius R and infinite height. Specifically, let $\{\boldsymbol{e}_1,\boldsymbol{e}_2,\boldsymbol{e}_3\}$ be a fixed orthonormal basis and consider a specimen occupying the cylinder

$$\mathcal{R}=\{\boldsymbol{x}:\sqrt{x_1^2+x_2^2}\leqslant R,-\infty<x_3<\infty\}, \tag{22}$$

with $x_i=\boldsymbol{x}\cdot\boldsymbol{e}_i$. Introduce cylindrical-polar coordinates (r,θ,z) via

$$r=\sqrt{x_1^2+x_2^2},\quad \theta=\arctan\left(\frac{x_2}{x_1}\right),\quad z=x_3, \tag{23}$$

and let $\{\boldsymbol{e}_r,\boldsymbol{e}_\theta,\boldsymbol{e}_z\}$ denote the associated physical basis. Suppose that radial strong anchoring prevails on the lateral surface of the specimen, so that

$$\boldsymbol{n}(r,\theta,z)=\boldsymbol{e}_r\quad\text{when } r=R. \tag{24}$$

Suppose also that the lateral surface of the specimen is traction-free, so that the Cauchy stress tensor obeys

$$\boldsymbol{T}(r,\theta,z)\boldsymbol{e}_r=\boldsymbol{0}\quad\text{when } r=R. \tag{25}$$

Neglect all external body forces. Assume that the director has the specific form

$$\boldsymbol{n}=\cos\chi\boldsymbol{e}_r+\sin\chi\boldsymbol{e}_z, \tag{26}$$

where χ depends only on r and, consistent with the notion of escape to the third dimension and the anchoring condition (24), obeys

$$\chi(0)=\frac{\pi}{2}\quad\text{and}\quad\chi(R)=0, \tag{27}$$

respectively. Furthermore, assume that $\boldsymbol{B}=\kappa\boldsymbol{A}$ with $\boldsymbol{A}$ as given in (4). Then

$$\psi_f=\frac{k_1\sin^2\chi+k_3\cos^2\chi}{2}\left(\frac{d\chi}{dr}\right)^2+\frac{k_1\cos^2\chi}{r^2}-\frac{(k_1-k_2-k_4)\sin 2\chi}{2r}\frac{d\chi}{dr}. \tag{28}$$

For $k_2+k_4=0$ and $k_3>k_1$, Cladis and Kléman[14] and Meyer[15] show that, for the boundary conditions (27), the Euler-

Lagrange equation arising from the free-energy density (28) integrates to yield an expression relating r/R to χ in terms of elementary functions. Corresponding to that solution is the net free energy per unit length of the specimen,

$$2\pi\int_0^R \psi(r)rdr = \pi k_1\left(2+\frac{\arcsin\beta}{\beta\sqrt{1-\beta^2}}\right), \tag{29}$$

with $\beta=\sqrt{1-k_1/k_3}$. As is well known, this energy is finite and less than that corresponding to the alternative description of a straightline disclination of strength +1 in which the director field remains planar.

Again, a tedious but straightforward calculation of the Riemann curvature tensor $\mathcal{R}$ shows that the Finger strain $\boldsymbol{B}$ associated with the formation of the escaped disclination (26) is incompatible.

Furthermore, a similar result holds for the case

$$\boldsymbol{n} = \cos\chi\boldsymbol{e}_\theta + \sin\chi\boldsymbol{e}_z. \tag{30}$$

In this case, the results are identical to those considered above where the net free energy per unit length is again (29), but with $\beta=\sqrt{1-k_2/k_3}$ ($k_3>k_2$ is assumed).

IV. DISCUSSION

We have presented examples where the neoclassical theory of nematic elastomers yields incompatible strains of the polymer network chains for the formation of point and line defects in the orientational degrees of freedom of the mesogens. Moreover, the distortion $\boldsymbol{F}$ may fail to be the gradient of a deformation. Since the neoclassical theory yields the Finger strain $\boldsymbol{B}=\kappa\boldsymbol{A}$, inhomogeneous equilibrium states of the director field will typically yield incompatible strains.

It is important to emphasize that, for a strongly elliptic, isotropic, incompressible, hyperelastic solid, both problems analogous to those considered here possess unique solutions (up to arbitrary rigid rotations). These solutions involve no deformation and are therefore trivially compatible.

For nematic elastomers it is typically assumed, however, that strains are compatible. If we were to view the predicted strain incompatibilities as undesirable or even unphysical, it would then be necessary to modify either the assumed form of the defect or the constitutive model. Note that in the examples presented here we have neglected the variability of S as well as the variability of $a_\perp$ and $a_\parallel$. Taking into account the spatial variability of these parameters, as done by Fried and Todres[16] and Fried *et al.*,[17] for example, might eliminate the strain incompatibilities as the defect core can become isotropic. This procedure would imply that nematic textures in elastomers are more complicated than in conventional (polymeric and low molecular weight) liquid crystals. Alternatively, a generalization of the neoclassical free-energy density to include additional terms (e.g., Fried and Sellers[18]) would change the resulting strain so that the restrictive form (7) is no longer valid. Such additional terms would also eliminate the so-called soft solutions of the neoclassical model, solutions which have been questioned in the literature.[19] It is, however, not yet known how such modifications to the free-energy density affect the compatibility of the resulting strains. Further work is needed to clarify these points.

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