Uniaxial and biaxial soft deformations of nematic elastomers

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We give a geometric interpretation of the soft elastic deformation modes of nematic elastomers, with explicit examples, for both uniaxial and biaxial nematic order. We show the importance of body rotations in this nonclassical elasticity and how the invariance under rotations of the reference and target states gives soft elasticity (the Golubovic and Lubensky theorem). The role of rotations makes the polar decomposition theorem vital for decomposing general deformations into body rotations and symmetric strains. The role of the square roots of tensors and that of finding explicit forms for soft deformations (the approach of Olmsted) are discussed in this context.

I. INTRODUCTION

Nematic elastomers display three unique and related phenomena not found in conventional elasticity—large spontaneous deformations, very large optical-mechanical response, and soft elasticity. The last, shape change with little or no energy cost, is the subject of this paper. We give a geometric interpretation of the soft modes described using fractional powers of tensors. We then discuss the soft modes of biaxial nematic elastomers. Since in contrast to conventional elastic solids, rotations play an essential role in the elasticity of nematic rubber, we conclude by discussing the related questions of breaking finite strains into symmetric shears and rotations, the polar decomposition theorem for tensors, and the nature of square roots of tensors.

Nematic elastomers have an internal, orientational degree of freedom in addition to those of ordinary elastic bodies. The anisotropy of molecular orientation induces shape anisotropy in the polymers that make up the elastomer. The switching on and off of this molecular shape anisotropy, either by temperature change [1] or by illumination [2], causes large ($\approx 400\%$) mechanical shape changes.

Rotation of this anisotropy, by imposed mechanical strains, is the extreme optomechanical effect that is observed. When accompanied by subsidiary shears and contractions that accommodate the changes of molecular distributions, the cost of the originally imposed strain is rendered to zero. This is what we call soft elasticity [3].

II. SOFTNESS IN LINEAR CONTINUA

One can explore the related ideas of anisotropy, rotation and soft elasticity initially for small deformations and rotations, that is within the linear continuum theory of a uniaxial body with a mobile director $\mathbf{n}$ that characterises the anisotropy direction [4]. The free energy density F is

$$F = \tfrac{1}{2}\widetilde{B}(\mathrm{Tr}[\underset{\approx}{\widetilde{\varepsilon}}])^2 + \tfrac{1}{2}\kappa\,\mathrm{Tr}[\underset{\approx}{\widetilde{\varepsilon}}](\mathbf{n}\cdot\underset{\sim}{\varepsilon}\cdot\mathbf{n}) + \tfrac{1}{2}\mu_0(\mathbf{n}\cdot\underset{\sim}{\varepsilon}\cdot\mathbf{n})^2$$

$$+ \tfrac{1}{2}\mu_1[\mathbf{n}\times\underset{\sim}{\varepsilon}\times\mathbf{n}]^2 + \tfrac{1}{2}\mu_2([\mathbf{n}\times\underset{\sim}{\varepsilon}]\cdot\mathbf{n})^2$$

$$+ \tfrac{1}{2}D_1[(\mathbf{\Omega}-\boldsymbol{\omega})\times\mathbf{n}]^2 + \tfrac{1}{2}D_2\,\mathbf{n}\cdot\underset{\sim}{\varepsilon}\cdot[(\mathbf{\Omega}-\boldsymbol{\omega})\times\mathbf{n}], \quad (1)$$

where $\underset{\sim}{\varepsilon} = \underset{\approx}{\widetilde{\varepsilon}} - \tfrac{1}{3}\mathrm{Tr}[\underset{\sim}{\widetilde{\varepsilon}}]\underset{\sim}{\delta}$ is the traceless part of the linear symmetric strain $\widetilde{\varepsilon}_{ij} = \tfrac{1}{2}(\partial_j u_i + \partial_i u_j)$, and $\mathbf{\Omega} = \tfrac{1}{2}\mathrm{curl}\ \mathbf{u}$ is the antisymmetric part (the body rotation), $\mathbf{u}$ being the displacement field.

The latter terms are those of the relative rotation ($\mathbf{\Omega} - \boldsymbol{\omega})\times\mathbf{n}$ coupling: for small rotations, the director variation corresponds to a rotation $\boldsymbol{\omega} = \mathbf{n}\times\delta\mathbf{n}_{\boldsymbol{\omega}}$ or $\delta\mathbf{n}_{\boldsymbol{\omega}} = \boldsymbol{\omega}\times\mathbf{n}$, see Fig. 1. Similarly, a small rotation $\mathbf{\Omega}$ of the matrix causes a vector $\mathbf{v}$ in the body to suffer the change $\delta\mathbf{v}_{\mathbf{\Omega}} = \mathbf{\Omega}\times\mathbf{v}$.

The net rotation of $\mathbf{n}$ with respect to the matrix inflicted by the relative rotation $\mathbf{\Omega} - \boldsymbol{\omega}$ of the matrix ($\mathbf{\Omega}$) relative to the changing director ($\boldsymbol{\omega}$) accordingly gives a relative change in $\mathbf{n}$ of

$$\delta\mathbf{n} = (\mathbf{\Omega} - \boldsymbol{\omega})\times\mathbf{n}.$$

The directors $\mathbf{n}$ and $\mathbf{n}_0$ before and after application of strain are not distinguished in the first terms of equation (1) for small director rotation, but clearly must be in the relative rotation coupling terms in F.

The tensor $\underset{\sim}{\widetilde{\varepsilon}}$ is the strain before it has been made traceless, that is, it still has volume changes $\mathrm{Tr}(\underset{\sim}{\widetilde{\varepsilon}})$ in it. Since rubber is a soft material, its deformations are at constant volume and we neglect the first two terms in Eq. (1) since they involve volume change. The geometry of the remaining strains is vital and shown in Fig. 2.

The de Gennes [4] relative rotation couplings are unique to nematic networks because they require an independent, rotational degree of freedom, i.e., the rotations $\boldsymbol{\omega}$ of the director $\mathbf{n}$. Its motion in the medium is coupled to the body
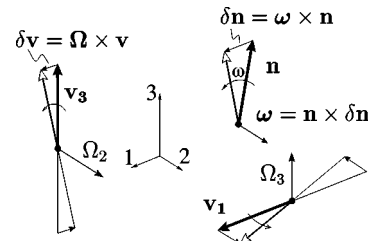

FIG. 1. Rotation of $\mathbf{n}$ about an axis $\boldsymbol{\omega}$, and body rotations $\mathbf{\Omega}$ rotating vectors $\mathbf{v}$. Only Ω_2 and Ω_1 (not shown) have any effect on vectors parallel to $\mathbf{n}$.
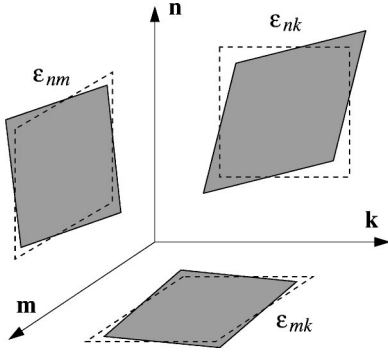
FIG. 2. The elements of strain in a uniaxially anisotropic medium. Dashed lines show the undistorted state, whereas the strained elements are shown by shaded areas. They divide into stretches ε_{nn} along $\mathbf{n}$, stretches ε_{mm} and ε_{kk} and distortions ε_{mk} in the plane perpendicular to $\mathbf{n}$, and distortions ε_{nm} and ε_{nk} encompassing $\mathbf{n}$ and the perpendicular plane.

rotation $\boldsymbol{\Omega}$. Hence, the elastic energy unusually involves *antisymmetric* components of shear strain. In Sec. VI, we accordingly show how to extract the rotational component from any finite shear. The first coupling D_1 purely resists the rotation of the director relative to a rotating, undeformed matrix. The D_2 term couples $\mathbf{n}$ to the symmetric part of shear in the plane that involves $\mathbf{n}$ (e.g., ε_{zx} if $n_0 = n_z$ in equilibrium). These are the shears ε_{nm} and ε_{nk} in Fig. 2. Since infinitesimal, incompressible symmetric shear is equivalent to stretch along one diagonal and compression along another, it is reasonable that prolate molecules will reduce the cost of distortion by rotating their ordering direction to being as much as possible along the elongation diagonal, depicted in Fig. 3. Oblate elastomers rotate their director toward the compression diagonal to achieve the appropriate elastic accommodation. The spheroid represents the anisotropic shape distribution of the crosslinked polymers from which the network is composed.

A theorem of Golubovic and Lubensky (GL) [5] shows that on symmetry grounds any solid with an internal degree of freedom which is also capable of reaching an isotropic
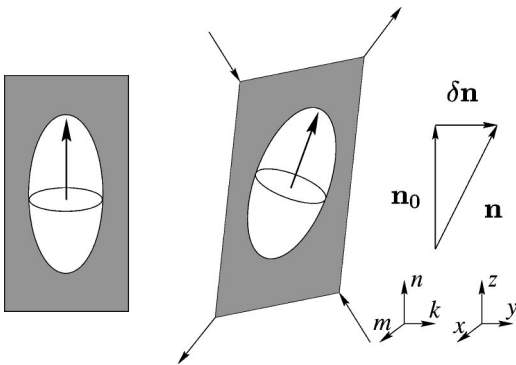

FIG. 3. Symmetric shear induces director rotation toward the elongation diagonal. The respective elongations and compressions along the diagonals are shown. The chain shape distribution also rotates. When mechanical shape changes of a network accommodate the rotations of the chain distribution without distortions, such shape changes can take place at minimal energy cost.

reference state must be invariant under the double set of rotations of both the reference and target states when considering elastic deformations. This has the remarkable consequence that some elastic deformations must be soft. This was discovered independently and in a superficially different form when studying at finite deformations the elastic response of nematic elastomers [3]. We sketch how this can be understood for elastomers in order to explain the complex soft modes we shall later discuss.

An effective shear modulus arises for an imposed elastic strain, when the nematic director is free to evolve optimally. The elastic modulus μ_2 is reduced; by the symmetry argument it must vanish to give no overall energy cost. If the local rotational component $\boldsymbol{\Omega}$ of the deformation of the elastic matrix and the rotation of the director are coaxial, i.e., $\boldsymbol{\Omega}$ and $\boldsymbol{\omega}$ are parallel and uniform, the argument is simple. Minimizing the relative-rotation part of the energy density (1), $\frac{1}{2}D_1[(\boldsymbol{\Omega}-\boldsymbol{\omega})\times\mathbf{n}]^2 + \frac{1}{2}D_2\,\mathbf{n}\cdot\underline{\underline{\varepsilon}}\cdot[(\boldsymbol{\Omega}-\boldsymbol{\omega})\times\mathbf{n}]$, one obtains the optimal relative rotation for a given shear strain $\underline{\underline{\varepsilon}}$:

$$[(\boldsymbol{\Omega}-\boldsymbol{\omega})\times\mathbf{n}] = -\frac{D_2}{2D_1}(\mathbf{n}\cdot\underline{\underline{\varepsilon}})\ \ \text{or}$$

$$(\boldsymbol{\Omega}-\boldsymbol{\omega}) = -\frac{D_2}{2D_1}[\mathbf{n}\cdot\underline{\underline{\varepsilon}}\times\mathbf{n}]. \tag{2}$$

We now substitute this back into the energy density and obtain for the rotation-strain terms:

$$\frac{1}{2}D_1[(\boldsymbol{\Omega}-\boldsymbol{\omega})\times\mathbf{n}]^2 + \frac{1}{2}D_2\,\mathbf{n}\cdot\underline{\underline{\varepsilon}}\cdot[(\boldsymbol{\Omega}-\boldsymbol{\omega})\times\mathbf{n}]$$

$$= -\frac{D_2^2}{8D_1}[\mathbf{n}\cdot\underline{\underline{\varepsilon}}\times\mathbf{n}]^2 \equiv -\frac{D_2^2}{8D_1}(\varepsilon_{xz}^2 + \varepsilon_{yz}^2). \tag{3}$$

The last expression is written in the specific coordinate frame where the initial director $\mathbf{n}_0$ is parallel to z axis. We can now unite this expression with the rest of the elastic energy density, Eq. (1) and thus obtain the effective rubber-elastic energy that depends only on strains; the director does not appear. For instance, in the specific coordinates of Fig. 2,

$$F = \frac{1}{2}\mu_0\,\varepsilon_{zz}^2 + \frac{1}{2}\mu_1(\varepsilon_{xx}^2 + 2\varepsilon_{xy}^2 + \varepsilon_{yy}^2)$$

$$+ \frac{1}{2}\left(\mu_2 - \frac{D_2^2}{4D_1}\right)(\varepsilon_{xz}^2 + \varepsilon_{yz}^2). \tag{4}$$

The modulus μ_2 is renormalized to $\mu_2 - D_2^2/(4D_1)$ which by the GL theorem must be zero, thus establishing a relation between the constants μ_2, D_1, and D_2. The molecular model, required below for finite deformations, produces linear continuum limiting values that also give

$$\mu_2^{\mathrm{R}} = \mu_2 - \frac{D_2^2}{4D_1} \to 0. \tag{5}$$

Olmsted [6] first proposed this continuum mechanism behind the Golubovic-Lubensky theorem: shape depends on the orientation of an internal (nematic) degree of freedom, the rotation of which causes a natural shape change at zero cost for suitable solids. A general discussion of the GL argument

and its extension to semisoftness and thresholds to rotation is given in Ref. [7]. The experimental evidence for mechanically soft distortions is also discussed there.

One can picture the continuous rotation generating mechanical distortion at low energy cost, see Fig. 3. The natural long axis of the body, as defined by the principal axis of molecular shape, rotates in an attempt to follow the apparent extension axis. To the extent that macroscopic shape change can thus be imitated, there is no real accompanying distortion of polymer shape. We quantify this picture below when considering nonlinear elasticity theory.

III. FINITE SOFT ELASTICITY

A simple extension of classical rubber elasticity theory to nematic elastomers gives for the free energy density

$$F = \tfrac{1}{2}\mu \mathrm{Tr}(\underline{\underline{\ell}}_0 \cdot \underline{\underline{\lambda}}^{\mathrm{T}} \cdot \underline{\underline{\ell}}^{-1} \cdot \underline{\underline{\lambda}}), \tag{6}$$

where μ is the rubber elastic shear modulus in the isotropic phase and $\lambda_{ij} = \partial R_i / \partial R_{0j}$ is the homogeneous deformation (gradient). If we take $\underline{\underline{\lambda}}$ to be $\underline{\underline{\lambda}} = \underline{\underline{\delta}} + \underline{\underline{u}}$, then the symmetric part of $\underline{\underline{u}}$ is identical to $\underline{\underline{\tilde{\varepsilon}}}$ of Eq. (1) in the infinitesimal limit.

The chains are no longer characterized by spherical Gaussian distributions as in the classical case, but in general by anisotropic distributions. Thus the effective step length tensors $\underline{\underline{\ell}}_0$ initially and $\underline{\underline{\ell}}$ currently after a distortion $\underline{\underline{\lambda}}$ are prolate (or oblate) spheroids defining the second moments that characterize the Gaussian distribution of chain spans **R** in the the network, that is, $\langle R_i R_j \rangle = \tfrac{1}{3}\ell_{ij}L$ where L is the arc length of a chain. Thus a measure of the mean size of a chain is $\underline{\underline{\ell}}^{1/2}$ where we shall soon define the roots of tensors more carefully. The tensor $\underline{\underline{\ell}}$ has one principal value $\ell_\parallel$ along **n** and $\ell_\perp$ perpendicular to **n**; thus

$$\underline{\underline{\ell}} = \begin{pmatrix} \ell_\perp & 0 & 0 \\ 0 & \ell_\perp & 0 \\ 0 & 0 & \ell_\parallel \end{pmatrix} = \ell_\perp \begin{pmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & r \end{pmatrix},$$

where $r = \ell_\parallel / \ell_\perp$. The extracted $\ell_\perp$ factor from $\underline{\underline{\ell}}_0$ cancels with the $1/\ell_\perp$ factor extracted from $\underline{\underline{\ell}}^{-1}$ when both tensors appear together in the trace formula (6) and we can henceforth just consider the $\underline{\underline{\ell}}$ tensors in their reduced form that simply depends on the intrinsic anisotropy r. Anisotropy varies between $r = 1.1 \rightarrow 60$. In this model of rubber elasticity the spontaneous elongation λ_{m}, on going from the isotropic to the nematic phase turns out to be $\lambda_{\mathrm{m}} = r^{1/3}$ and is thus a direct measure of r. Indeed spontaneous elongations in the range of $\lambda_{\mathrm{m}} \sim 1.03$–$4.00$ are observed.

Now distortions $\underline{\underline{\lambda}}$ are no longer small, but must continue to respect volume conservation, $\mathrm{Det}(\underline{\underline{\lambda}}) = 1$ in the nonlinear regime. The directors $\mathbf{n}_0$ and $\mathbf{n}$, of the initial and current nematic states, may be greatly rotated from each other.

The remainder of this paper is concerned with explaining the character of the soft modes within finite elasticity theory and extending this picture to that of soft modes in biaxial nematic elastomers. We explore the role of rotations and the connections between the isolation of rotational components

of strain at finite amplitude (the spherical decomposition theorem) and the relation of the roots of tensors to this picture.

IV. UNIAXIAL SOFTNESS

Consider the strain [6]

$$\underline{\underline{\lambda}} = \underline{\underline{\ell}}^{1/2} \underline{\underline{W}}_\alpha \underline{\underline{\ell}}_0^{-1/2}, \tag{7}$$

where $\underline{\underline{W}}_\alpha$ is an arbitrary rotation by an angle α. There are two continuous degrees of freedom describing the rotation connecting **n** and $\mathbf{n}_0$ and three degrees of freedom for the rotation $\underline{\underline{W}}_\alpha$. Hence, the strain $\underline{\underline{\lambda}}$ is described by five continuous degrees of freedom and thus represents a large set of deformations.

If we insert such a strain into the trace formula (6), as well as its transpose $\underline{\underline{\lambda}}^{\mathrm{T}}$ (equivalent to $\underline{\underline{\ell}}_0^{-1/2} \cdot \underline{\underline{W}}_\alpha^{\mathrm{T}} \cdot \underline{\underline{\ell}}^{1/2}$ since the $\underline{\underline{\ell}}$ are symmetric) we obtain

$$F_{\mathrm{el}} = \tfrac{1}{2}\mu \mathrm{Tr}(\underline{\underline{\ell}}_0 \cdot \underline{\underline{\ell}}_0^{-1/2} \cdot \underline{\underline{W}}_\alpha^{\mathrm{T}} \cdot \underline{\underline{\ell}}^{1/2} \cdot \underline{\underline{\ell}}^{-1} \cdot \underline{\underline{\ell}}^{1/2} \cdot \underline{\underline{W}}_\alpha \cdot \underline{\underline{\ell}}_0^{-1/2})$$

$$\equiv \tfrac{1}{2}\mu \mathrm{Tr}(\underline{\underline{\delta}}) = \frac{3}{2}\mu. \tag{8}$$

Canceling the middle section, $\underline{\underline{\ell}}^{1/2} \cdot \underline{\underline{\ell}}^{-1} \cdot \underline{\underline{\ell}}^{1/2} = \underline{\underline{\delta}}$, allows the $\underline{\underline{W}}$ terms to meet and gives $\underline{\underline{W}}^{\mathrm{T}} \cdot \underline{\underline{W}} = \underline{\underline{\delta}}$. Likewise disposing of the $\underline{\underline{\ell}}_0$ terms, one obtains the final expression $F_{\mathrm{el}} = \frac{3}{2}\mu$. This is identical to the free energy of an undistorted network. The nontrivial set of distortions $\underline{\underline{\lambda}}$ of this particular form, Eq. (7), have not raised the energy of nematic elastomer. From the results $\mathrm{Det}(\underline{\underline{A}} \cdot \underline{\underline{B}}) = \mathrm{Det}(\underline{\underline{A}})\mathrm{Det}(\underline{\underline{B}})$, $\mathrm{Det}(\underline{\underline{A}}^{-1}) = [\mathrm{Det}(\underline{\underline{A}})]^{-1}$, and $\mathrm{Det}(\underline{\underline{W}}) = 1$ for all rotations $\underline{\underline{W}}$, one can see that $\mathrm{Det}(\underline{\underline{\lambda}}) = \mathrm{Det}(\underline{\underline{\ell}}^{1/2} \cdot \underline{\underline{W}}_\alpha \cdot \underline{\underline{\ell}}_0^{-1/2}) = 1$, that is, these soft modes are volume preserving.

We saw pictorially in Fig. 3 that, on applying a stretch perpendicular to the initial director, rotation of the chain distribution is accommodated by the very elongation we have applied, together with a shear. Two remarkable consequences immediately follow from nematic elastomer response via rotation:

Impose an x extension λ_{xx}. All the accompanying distortions must be in the plane of rotation, that is, a transverse contraction λ_{zz} and shears λ_{xz} and λ_{zx} accommodate the rotation of the distribution. No shears perpendicular to this plane, that is, involving y direction ($\lambda_{yy}, \lambda_{yx}, \ldots$, etc.), are needed. For a classical isotropic elastomer $\lambda_{yy} = \lambda_{zz} = 1/\sqrt{\lambda_{xx}}$ is demanded by incompressibility, whereas in soft elasticity there is no shrinkage in the y direction ($\lambda_{yy} = 1$) and the appropriate Poisson ratio is zero.

Figure 4 shows the initial and final states of which the shear and partial rotation of Fig. 3 is an intermediate step. Softness must come to an end when the rotation is complete and the z dimension has diminished in the proportion $\lambda_{zz} = \sqrt{\ell_\perp / \ell_\parallel}$ and the x dimension extended in the proportion $\lambda_{xx} = \sqrt{\ell_\parallel / \ell_\perp}$. The original sizes $\sqrt{\ell_\parallel}$ and $\sqrt{\ell_\perp}$ have transformed to $\sqrt{\ell_\perp}$ and $\sqrt{\ell_\parallel}$, respectively. Thus softness would cease and director rotation be complete at $\lambda_{xx} = r^{1/2} \equiv \lambda_{\mathrm{m}}^{3/2}$. The strain $\lambda_{\mathrm{m}} = (\ell_\parallel / \ell_\perp)^{1/3}$ is the spontaneous extension suf-
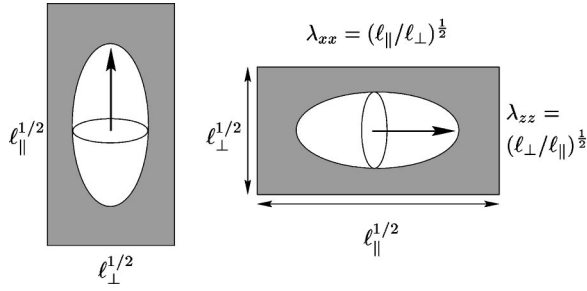
FIG. 4. Chain shape distribution before and after rotations. The extent of soft extensions (λ_{xx}) perpendicular to the initial director is set by the anisotropy of the molecular shape distribution. The macroscopic dimensions are shown changing affinely with the distribution, for instance, the x dimension changing from $\sqrt{\ell_\perp}$ to $\sqrt{\ell_\parallel}$.

fered on cooling to the nematic phase. Likewise one can imagine from an oblique form of Fig. 4 that if the initial director $\mathbf{n}_0$ (long axis of the shape ellipsoid) is not at 90° to the imposed strain, then rotation and softness is complete at a smaller $\lambda_{xx} < \lambda_m$.

We give as a concrete example the set of soft distortions $\underline{\underline{\lambda}}_{\text{soft}} = \underline{\underline{\ell}}_\theta^{1/2} \cdot \underline{\underline{\ell}}_0^{-1/2}$, simplified by the absence of the arbitrary rotation matrix $\underline{\underline{W}}_\alpha$. They are simply characterized (parametrically) by the angle θ by which $\underline{\underline{\ell}}_0$ is rotated to $\underline{\underline{\ell}}_\theta$, that is, by which $\mathbf{n}_0$ is rotated to $\mathbf{n}$. Putting in the dyadic forms for $\underline{\underline{\ell}}_0^{-1/2}$ and $\underline{\underline{\ell}}_\theta^{1/2}$ into $\underline{\underline{\lambda}}_{\text{soft}}$ gives

$$\underline{\underline{\lambda}}_{\text{soft}} = [\underline{\underline{\delta}} + (\sqrt{r} - 1)\mathbf{n}\mathbf{n}^{\text{T}}] \cdot \left[\underline{\underline{\delta}} + \left(\frac{1}{\sqrt{r}} - 1\right)\mathbf{n}_0\mathbf{n}_0^{\text{T}}\right]$$

$$= \underline{\underline{\delta}} + (1/\sqrt{r} - 1)\mathbf{n}_0\mathbf{n}_0^{\text{T}} + (\sqrt{r} - 1)\mathbf{n}\mathbf{n}^{\text{T}}$$

$$+ (\mathbf{n}\mathbf{n}_0)(2 - \sqrt{r} - 1/\sqrt{r})\mathbf{n}\mathbf{n}_0^{\text{T}}. \qquad (9)$$

If $\mathbf{n}_0$ is along $\hat{\mathbf{z}}$ and is rotated by θ toward $\hat{\mathbf{x}}$, it becomes $\mathbf{n} = \hat{\mathbf{z}}\cos\theta + \hat{\mathbf{x}}\sin\theta$. We can write down a particular representation of $\underline{\underline{\lambda}}_{\text{soft}}$ (using the notation $s \equiv \sin\theta$ and $c \equiv \cos\theta$):

$$\underline{\underline{\lambda}}_{\text{soft}} = \hat{\mathbf{z}}\hat{\mathbf{z}}^{\text{T}}\left[1 - \left(1 - \frac{1}{\sqrt{r}}\right)s^2\right] + \hat{\mathbf{x}}\hat{\mathbf{x}}^{\text{T}}[1 + (\sqrt{r} - 1)s^2] + \hat{\mathbf{y}}\hat{\mathbf{y}}^{\text{T}}$$

$$+ \hat{\mathbf{x}}\hat{\mathbf{z}}^{\text{T}}\left(1 - \frac{1}{\sqrt{r}}\right)sc + \hat{\mathbf{z}}\hat{\mathbf{x}}^{\text{T}}(\sqrt{r} - 1)sc$$

$$\equiv \begin{pmatrix} 1 + (\sqrt{r} - 1)s^2 & 0 & (1 - 1/\sqrt{r})sc \\ 0 & 1 & 0 \\ (\sqrt{r} - 1)sc & 0 & 1 - (1 - 1/\sqrt{r})s^2 \end{pmatrix}. \qquad (10)$$

The soft modes are neither simple nor pure shear, but a mixture of the two. Accordingly, the soft modes have an element of body rotation in addition to elongations, compressions, and pure shears. The importance of body rotations is discussed before and after Eq. (2). The degree of body rotation is given in Sec. VI. Note that the extensional and compressional strains ($\lambda_{xx} - 1$) and ($1 - \lambda_{zz}$) are both proportional to
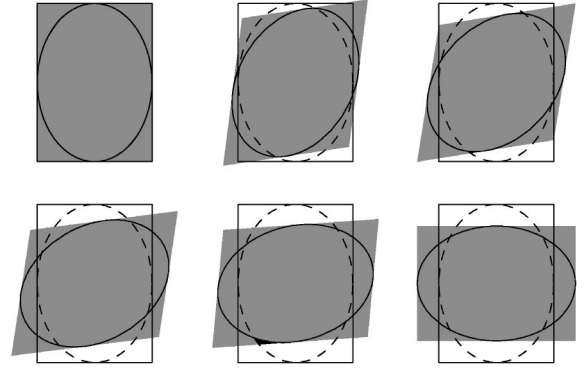

FIG. 5. Soft deformations of a nematic elastomer with anisotropy $r = 2$. The deformations correspond to director rotations of $\theta = 0$, $\pi/6$, $\pi/4$, $\pi/3$, $5\pi/12$, and $\pi/2$ that parametrically generate the distortions as discussed above. Note that the prolate spheroid characterizing the distribution of chains, embedded in the distorting solid (shaded and shown in section), when rotated by θ can be accommodated without distortion. The reference, undeformed body is shown in outline; the original distribution shown dashed.

$\sin^2\theta$. Thus the infinitesimal strains u_{zz} and u_{xx}, at small rotations θ, are proportional to θ^2. By contrast λ_{xz} and λ_{zx} are proportional to $\sin\theta\cos\theta$ and hence the infinitesimals u_{xz} and u_{zx} are proportional to θ—a lower order than u_{xx} and u_{zz}. There is no relaxation along y, perpendicular to the plane of rotation $\mathbf{n}$, that is, $\lambda_{yy} = 1$. Also note that in the isotropic limit ($r = 1$) this particular strain $\underline{\underline{\lambda}}_{\text{soft}} = \underline{\underline{\delta}}$ while the general soft deformation (7) reduces to the null strain $\underline{\underline{W}}_\alpha$, a simple body rotation. Both are the trivial cases evidently preserving the elastic energy at its minimum. The soft modes become nontrivial deformations when the material becomes a nematic elastomer.

The soft modes start at no strain, $\underline{\underline{\lambda}} = \underline{\underline{\delta}}$, and as the director θ rotates from 0 to $\pi/2$, they eventually end at

$$\underline{\underline{\lambda}} = \begin{pmatrix} \sqrt{r} & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 1/\sqrt{r} \end{pmatrix},$$

that is an extension $\lambda_{xx} = \sqrt{r}$ and a transverse contraction $\lambda_{zz} = 1/\sqrt{r}$.

The director rotation is taken up by a shape change so that there is no entropically expensive deformation of the chain distribution as when a conventional elastomer deforms. The shape tensor's anisotropy $r = \ell_\parallel/\ell_\perp$ characterizes the ratio of the mean square size along the director to that perpendicular to the director. The square root of this ratio, $\sqrt{r}$, gives the characteristic ratio of average dimensions of chains in the network. During a soft deformation, the solid must change shape such that the rotating ellipsoid $\underline{\underline{\ell}}^{1/2}$, characterizing the physical dimensions of the distribution of chains, is accommodated without distortion. The ellipsoid is $\mathbf{R} \cdot \underline{\underline{\ell}}^{-1} \cdot \mathbf{R} = 1$, or in the principal frame $x^2/r + z^2 = 1$, that is, in section the ellipse has semimajor axes of $\sqrt{r}$ and 1. Figure 5 illustrates soft deformations of a nematic elastomer with chains of anisotropy $r = 2$. The distortions are parametrized by the direc-

tor rotation θ that ranges between 0 and $\pi/2$. The chain shape tensor $\underline{\underline{\ell}}^{1/2}$ rotates without distortion, just fitting in the solid into which it embedded. The soft deformations of Fig. 5 and Eqs. (9) and (10) of the body are in general nonsymmetric. Below we decompose these into pure shears plus rotations. One can think of $\underline{\underline{\lambda}}_{\mathrm{soft}} = \underline{\underline{\ell}}_\theta^{1/2} \cdot \underline{\underline{\ell}}_0^{-1/2}$ as converting the initial ellipsoid of Fig. 5 to a sphere by the action of the inverse $\underline{\underline{\ell}}_0^{-1/2}$ and then recreating an ellipsoid at an angle θ with the action of $\underline{\underline{\ell}}_\theta^{1/2}$. This is precisely the scheme of Refs. [8,9] who consider an isotropic reference state (the intermediate created after the action of $\underline{\underline{\ell}}_0^{-1/2}$). We discuss this rotational invariance again below.

V. BIAXIAL SOFTNESS

Biaxial nematic phases are rare. They have been found (Finkelmann *et al.* [10,11]) in nematic polymers since one has more complex molecular structural possibilities. In principle, such polymers could be used to make biaxially nematic elastomers. They would have rich mechanical properties.

The shape tensor of a biaxial polymer is

$$\underline{\underline{\ell}} = \begin{pmatrix} \ell_1 & 0 & 0 \\ 0 & \ell_2 & 0 \\ 0 & 0 & \ell_\parallel \end{pmatrix}.$$

Now the step lengths in the two directions perpendicular to **n**, ℓ_1, and ℓ_2, are distinguished. The tensor is reduced by taking out a factor of the mean perpendicular step length, $\ell_\perp = \frac{1}{2}(\ell_1 + \ell_2)$, to give

$$\underline{\underline{\ell}} = \begin{pmatrix} 1 + p/2 & 0 & 0 \\ 0 & 1 - p/2 & 0 \\ 0 & 0 & r \end{pmatrix}$$

$$\equiv r\mathbf{n}\mathbf{n}^{\mathrm{T}} + \left(1 + \frac{p}{2}\right)\mathbf{m}\mathbf{m}^{\mathrm{T}} + \left(1 - \frac{p}{2}\right)\mathbf{k}\mathbf{k}^{\mathrm{T}}$$

$$\equiv \underline{\underline{\delta}} + (r - 1)\mathbf{n}\mathbf{n}^{\mathrm{T}} + \frac{p}{2}\mathbf{m}\mathbf{m}^{\mathrm{T}} - \frac{p}{2}\mathbf{k}\mathbf{k}^{\mathrm{T}},$$

where the axes of the ellipsoid are **n**, **m**, and **k** (with the latter a third perpendicular axis given by $\mathbf{k} = \mathbf{n} \times \mathbf{m}$), see Fig. 6. The biaxiality is $p = 2[(\ell_1 - \ell_2)/(\ell_1 + \ell_2)] = (\ell_1 - \ell_2)/\ell_\perp$.

With fixed **n**, in the biaxial case one can rotate the anisotropic transverse section mk of the square root of the shape tensor $\underline{\underline{\ell}}^{1/2}$ and accommodate it without distortion by inducing shape changes in the mk plane. These distortions are soft for exactly the same reasons as in the uniaxial case when **n** suffered rotations in the nk or nm planes, see Figs. 5. Given that there are (now in the laboratory frame) λ_{xy} soft modes as well as the λ_{zx} and λ_{zy} forms (which are now no longer equivalent modes), the order of softness has become very much greater.
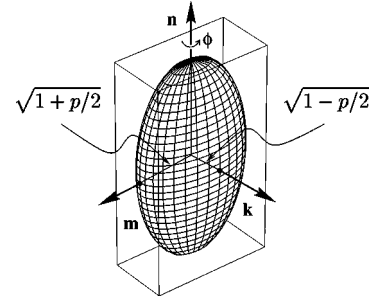

FIG. 6. The shape ellipsoid for a biaxially nematic polymer. The section perpendicular to **n** is not circular, but has semiaxes of length $\sqrt{1 \pm p/2}$. Rotations ϕ about $\mathbf{n} \equiv \mathbf{n}_0$ generate distortions in the x-y plane that softly accommodate the noncircular shape as it rotates.

A. Explicit examples of soft modes arising from biaxiality

The situation is exactly parallel to that of the soft shears (9) and (10). Now the soft modes are given by $\underline{\underline{\ell}}_\phi^{1/2} \underline{\underline{\ell}}_0^{-1/2}$, where ϕ is instead the angle of rotation about the principal director **n**. The tensor $\underline{\underline{\ell}}_0^{1/2}$ is

$$\underline{\underline{\ell}}_0^{1/2} = \begin{pmatrix} \sqrt{1 + p/2} & 0 & 0 \\ 0 & \sqrt{1 - p/2} & 0 \\ 0 & 0 & \sqrt{r} \end{pmatrix}, \qquad (11)$$

which will then be rotated by ϕ. By analogy with Fig. 5, the anisotropy in the x-y part of the matrix which is to be rotated is $\sqrt{r_\perp} = \sqrt{(1 + p/2)/(1 - p/2)} \equiv \sqrt{\ell_1/\ell_2}$. This measures the anisotropy of actual (root mean square) chain dimensions in the plane perpendicular to the director. The resulting soft shears in the x-y plane arising from rotations ϕ about $\mathbf{z} = \mathbf{n}_0$ are

$$\underline{\underline{\lambda}}_{\mathrm{biaxial}} = \begin{pmatrix} 1 - (1 - 1/\sqrt{r_\perp})s_\phi^2 & (1 - \sqrt{r_\perp})s_\phi c_\phi & 0 \\ (1/\sqrt{r_\perp} - 1)s_\phi c_\phi & 1 + (\sqrt{r_\perp} - 1)s_\phi^2 & 0 \\ 0 & 0 & 1 \end{pmatrix}. \qquad (12)$$

More complicated soft shears are possible if these shears are combined with those previously found in the uniaxial case. For example, in the uniaxial case, Sec. IV above, subsequent rotation about **n** after the rotation of **n** itself had no effect since there was symmetry about **n**. Now that this is lost, such rotations will reorient the perpendicular directors **m** and **k** and thereby induce additional soft shears.

B. General biaxial softness

If, however, the axis of rotation does not coincide with a principal axis as in the previous examples, we discover soft modes of lower symmetry, which only lead back to an undeformed body after the chain distribution has been rotated by 2π. Consider, as an example of reduced symmetry, the situation when the chain distribution is rotated around a general axis in the x-y plane at an angle β to the x axis (Fig. 7): a rotation of π (Fig. 8) transfers the director **n** (along the z axis) into $-\mathbf{n}$, but the unit vectors **m** and **k** along the other
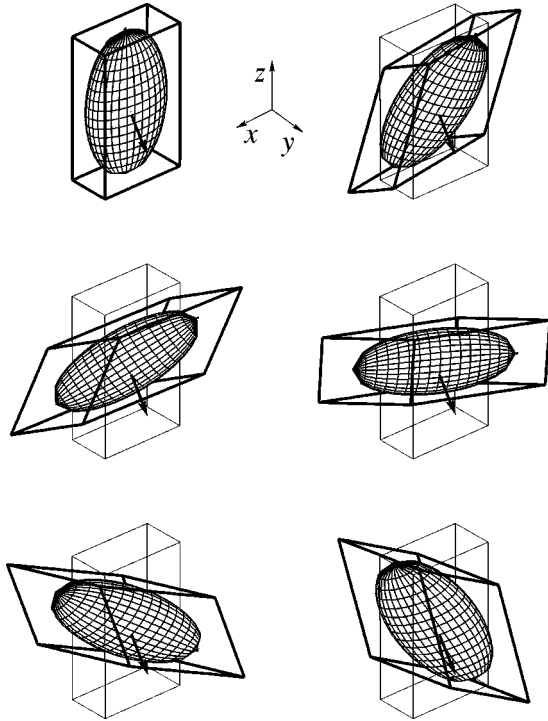
FIG. 7. Rotation of the chain distribution around an axis (arrow) in the x-y plane at an angle of $\pi/3$ with the x axis. The figure shows intermediate states of rotation at an angle of 0, π, $\frac{1}{6}\pi$, $\frac{1}{3}\pi$, $\frac{1}{2}\pi$, $\frac{2}{3}\pi$, and $\frac{5}{6}\pi$. Figure (8) shows the final stage of a rotation by an angle of π. The fine outline shows the initial body shape, whereas the highlighted outline illustrates the body deformation as the ellipsoid of the internal chain distribution rotates. Here, we have chosen $p=6/5$ and $r=200/45$, or, equivalently, the principal axes of the ellipsoid to be 6, 3, and 10, respectively.

two principal axes end up in a general position in the x-y plane. The same final position of the ellipsoid of chain distribution can be obtained by a rotation around the z axis by an appropriate angle. The corresponding body deformation leaves all z coordinates invariant, but transforms the (x,y) coordinates as if they have suffered a body rotation of 2β about z.

If we were to choose the axis of rotation in an arbitrary way, we would not observe this feature of invariant z com-
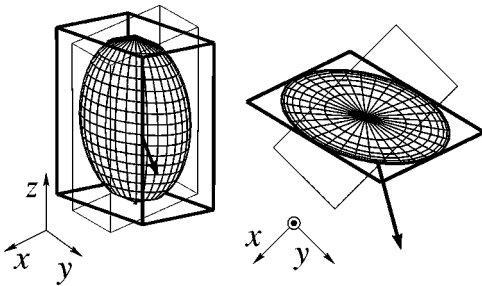

FIG. 8. Rotation of the chain distribution by π around an axis in the x-y plane at an angle of $\pi/3$ with the x axis. The figure shows two views of the same situation: the final ellipsoid together with the outlines of the bodies which indicate the corresponding soft mode deformation. The outline of the deformed body is highlighted. The parameters are the same as in Fig. 7.

ponents after a rotation of π. Only a full rotation of 2π would lead back into a state of higher symmetry, in this case, of course, the identity.

By rotating the ellipsoid of chain distribution, we create soft modes, which are closely related to the underlying symmetry, which is the crystal class of the biaxial ellipsoid, the dipyramidal orthorhombic class.

Comparing with Eq. (7), we see that so far we have set the rotation $\underline{W}_\alpha$ to be the identity. If, however, we were to allow a general rotation, we would find an even larger set of deformations, in fact described by 6 degrees of freedom: the additional rotations $\underline{W}_\alpha$ and the rotations connecting the principal frames of the initial and final ellipsoids each introduce 3 degrees of freedom.

VI. ROTATIONS, SYMMETRIC STRAINS, AND THE ROOTS OF TENSORS

We have seen in Eqs. (1)–(4) yielding softness that rotations are vital to understanding soft elasticity. The cartoon of Fig. 6 shows that soft shears are not symmetric and hence there must be a component of body rotation present. Here we explore the role of rotations in symmetry requirements. We then explicitly extract the rotational component from general distortions and from soft deformations in particular. The requirements for extracting the rotational components of the strain tensor are intimately related to finding the square roots of tensors that we discuss here since they are used in constructing soft deformations.

Any general matrix $\underline{M}$ can be broken down into products $\underline{S} \cdot \underline{Q}$ or $\underline{O} \cdot \underline{S}'$ of symmetric matrices $\underline{S}$ and $\underline{S}'$ and orthogonal matrices $\underline{Q}$ and $\underline{Q}'$, which can be restricted to proper rotations. In effect, one has a pure shear preceded or followed by a body rotation (the polar decomposition theorem [12]).

Note that in the infinitesimal case, we have $\underline{S} = \underline{1} + \underline{\delta S}$ and $\underline{Q} = \underline{1} + \underline{\delta O}$, hence $\underline{\delta S} = \underline{\delta S}^\mathrm{T}$ and $\underline{\delta O} = -\underline{\delta O}^\mathrm{T}$. Analogous statements hold for the matrices $\underline{S}'$ and $\underline{Q}'$. This shows that the rotation matrices $\underline{Q}$ and $\underline{Q}'$ are related to the antisymmetric part of the deformation in the infinitesimal limit.

Applied to the deformation $\underline{\lambda}$, we write

$$\underline{\lambda} = \underline{\lambda}^L \cdot \underline{V} \qquad \text{or} \qquad \underline{U} \cdot \underline{\lambda}^R, \qquad (13)$$

where the rotations are denoted by $\underline{V}$ or $\underline{U}$ depending upon whether they act on the reference or target spaces, respectively, of the deformation. The form of the accompanying symmetric deformations will depend on the order; they are denoted by $\underline{\lambda}^R$ and $\underline{\lambda}^L$, respectively, and yield the Cauchy-Green tensors $\underline{C}$ and $\underline{B}$, respectively: $\underline{C} = \underline{\lambda}^\mathrm{T} \cdot \underline{\lambda}$ and $\underline{B} = \underline{\lambda} \cdot \underline{\lambda}^\mathrm{T}$.

We summarize classical elasticity theory to highlight the differences with nematic rubber elasticity. The deformation (gradient) is

$$\lambda_{ij} = \partial R_i / \partial R_{0j}. \qquad (14)$$

If the target space (S_T) deforms under rotations represented

by the matrix $\underline{U}$, as $\mathbf{R}' = \underline{U} \cdot \mathbf{R}$, and the reference space (S_R) deforms under rotations $\underline{V}$ as $\mathbf{R}'_0 = \underline{V} \cdot \mathbf{R}_0$, then the deformation tensor deforms as

$$\lambda'_{ij} = U_{ik} \partial R_k / \partial R_{0l} V_{lj}^{\mathrm{T}}, \tag{15}$$

$$\underline{\underline{\lambda}}' = \underline{U} \cdot \underline{\underline{\lambda}} \cdot \underline{V}^{\mathrm{T}} \quad \text{or} \quad \underline{\underline{\lambda}} = \underline{U}^{\mathrm{T}} \cdot \underline{\underline{\lambda}}' \cdot \underline{V}. \tag{16}$$

Thus $\underline{\underline{\lambda}}$ records the character of both the target and reference states, a property that will be essential in nonideal nematic elastomers where an isotropic reference state cannot be reached. The connection with both spaces is quite different in character from that of the Cauchy tensors. Thus the combination

$$\underline{\underline{C}} = \underline{\underline{\lambda}}^{\mathrm{T}} \cdot \underline{\underline{\lambda}} = \underline{V}^{\mathrm{T}} \cdot \underline{\underline{\lambda}}'^{\mathrm{T}} \cdot \underline{U} \cdot \underline{U}^{\mathrm{T}} \cdot \underline{\underline{\lambda}}' \cdot \underline{V}$$

$$= \underline{V}^{\mathrm{T}} \cdot \underline{\underline{\lambda}}'^{\mathrm{T}} \cdot \underline{\underline{\lambda}}' \cdot \underline{V} = \underline{V}^{\mathrm{T}} \cdot \underline{\underline{C}}' \cdot \underline{V} \tag{17}$$

is manifestly invariant under body rotations $\underline{U}$ of the final (target) space S_T and transforms as a second rank tensor in S_R. Since isotropic systems are invariant to rotations $\underline{V}$ of S_R, the system's final energy must be invariant to rotations of S_R. Thus F is a function of the rotational (in S_R) invariants of $\underline{\underline{C}}$ and is assured by the above of being invariant under rotations of S_T. As an example, the isotropic rubber elastic free energy, setting $\underline{\underline{\ell}}^0 = \underline{\underline{\ell}}^{-1} = \underline{\underline{\delta}}$ in Eq. (6) is

$$F = \tfrac{1}{2} \mu \mathrm{Tr}(\underline{\underline{C}}) = \tfrac{1}{2} \mu \mathrm{Tr}(\underline{V}^{\mathrm{T}} \cdot \underline{\underline{C}}' \cdot \underline{V})$$

$$= \tfrac{1}{2} \mu \mathrm{Tr}(\underline{\underline{C}}' \cdot \underline{V} \cdot \underline{V}^{\mathrm{T}}) = \tfrac{1}{2} \mu \mathrm{Tr}(\underline{\underline{C}}') \tag{18}$$

(by cyclical properties of the trace). Likewise, $\underline{\underline{B}} = \underline{\underline{\lambda}} \cdot \underline{\underline{\lambda}}^{\mathrm{T}}$ is invariant to rotations of the reference state and transforms like a second rank tensor in the target state.

Thus we see that "objectivity," frame indifference, is built into classical elasticity theory from the outset. Nematic elastomers are much more subtle. In continuum theory we have seen that rotations and symmetric shears enter, both separately (through the de Gennes D_1 term) and coupled (through the D_2 term). In finite elasticity, we see in the trace formula that the initial and final orientations of the solid and its directors enter via the tensors $\underline{\underline{\ell}}^0$ and $\underline{\underline{\ell}}^{-1}$ and there are not combinations like $\underline{\underline{B}}$ and $\underline{\underline{C}}$ which eliminate rotations. For instance, inserting $\underline{U} \cdot \underline{\underline{\lambda}}^R$ into the trace result, one obtains

$$F = \tfrac{1}{2} \mu \mathrm{Tr}(\underline{\underline{\ell}}_0 \cdot \underline{\underline{\lambda}}^{R\,\mathrm{T}} \cdot [\underline{U}^{\mathrm{T}} \cdot \underline{\underline{\ell}}^{-1} \cdot \underline{U}] \cdot \underline{\underline{\lambda}}^R), \tag{19}$$

where the $[\cdots]$ have been inserted to emphasize that a body rotation of S_T effectively adds to the rotation of $\mathbf{n}$. That is, a new $\underline{\underline{\ell}}'$ evolves: $\underline{\underline{\ell}}'^{-1} = \underline{U}^{\mathrm{T}} \underline{\underline{\ell}}^{-1} \underline{U}$. (The additional rotation is not necessarily coaxial with that which took $\mathbf{n}_0$ to $\mathbf{n}$.) Not unexpectedly, we see the effect of $\underline{U}$ compounded with the rotations implicit in $\underline{\underline{\ell}}^{-1}$ since they both live in the target space.

Other approaches have been taken [8,9] to soft elasticity, which apparently circumvent the necessity to follow orientations in both spaces, and restore objectivity. One can measure all deformations $\underline{\underline{\lambda}}$ from an isotropic reference state, that

is there is encoded into the $\underline{\underline{\lambda}}$ first a spontaneous deformation to current conditions of temperature, then a deformation imposed with respect to this intermediate state. Under these conditions the free energy must automatically be invariant under operations of $\underline{V}$, since the reference state is isotropic and without a director $\mathbf{n}_0$ to keep track of. Difficulties arise then when nematic elastomers are only *semisoft*, that is, they do not deform entirely at constant free energy, because they do not have a high temperature isotropic reference state. However, they do suffer director rotation, very low energy trajectories in $\underline{\underline{\lambda}}$ space, and a lack of λ_{yy} relaxation perpendicular to the plane of $\mathbf{n}$'s rotation [13]. In these cases, the complete cancellation rendering $\mu_2^{\mathrm{R}} = \mu_2 - (D_2^2/4D_1) \to 0$ fails, but nevertheless deformations are qualitatively soft and one has to keep track of both directions $\mathbf{n}_0$ and $\mathbf{n}$ as strain evolves.

A. Square roots of tensors and the polar decomposition theorem

We quote the classical conditions [12] for the square roots of tensors, that arise too in the all-important condition for polar decomposition that is required for nematic elastomers.

If $\underline{\underline{A}}$ is nonsingular, with μ distinct eigenvalues and with ν Jordan blocks, then $\underline{\underline{A}}$ has $\geq 2^\mu$ and $\leq 2^\nu$ nonsingular square roots. At least one of these roots is a polynomial in $\underline{\underline{A}}$.

The proof of the polar decomposition theorem (PDT) also offers a practical algorithm for decomposition: take a nonsingular $\underline{\underline{\lambda}}$ and construct the manifestly symmetric $\underline{\underline{\lambda}}^{\mathrm{T}} \underline{\underline{\lambda}}$. Take the square root $\underline{\underline{G}}$ of $\underline{\underline{\lambda}}^{\mathrm{T}} \underline{\underline{\lambda}}$ that is a polynomial in $\underline{\underline{\lambda}}^{\mathrm{T}} \underline{\underline{\lambda}}$. Since $\underline{\underline{\lambda}}^{\mathrm{T}} \underline{\underline{\lambda}}$ is symmetric, then so is $\underline{\underline{G}} = \sqrt{\underline{\underline{\lambda}}^{\mathrm{T}} \underline{\underline{\lambda}}}$. Then define $\underline{\underline{Q}} = \underline{\underline{G}}^{-1} \underline{\underline{\lambda}}$. Clearly, $\underline{\underline{Q}}^{\mathrm{T}} \underline{\underline{Q}} = \underline{\underline{\delta}}$, that is, $\underline{\underline{Q}}$ represents rotations. From this one recovers $\underline{\underline{\lambda}} = \underline{\underline{G}} \, \underline{\underline{Q}}$.

B. Symmetric strain and body rotation of soft modes

We investigate the special case of uniaxial soft modes in the x-z plane. They are achieved by rotating the chain distribution around the y axis. The resulting deformation keeps the y-components of the body constant. Hence these soft modes are effectively 2×2 and can be easily related to Eq. (10) and Fig. 5.

An explicit example of the PDT for decomposing a general x-z distortion $\underline{\underline{\lambda}}$ into a combination of symmetric distortion $\underline{\underline{\lambda}}^R$ followed by a body rotation $\underline{U}$, about the y axis by an angle α is

$$\underline{\underline{\lambda}}_{\mathrm{soft}} = \begin{pmatrix} \lambda_{xx} & \delta \\ \delta' & \lambda_{zz} \end{pmatrix} = \begin{pmatrix} c & s \\ -s & c \end{pmatrix} \begin{pmatrix} a & d \\ d & b \end{pmatrix} \equiv \underline{U} \cdot \underline{\underline{\lambda}}^R.$$

(with $s = \sin \alpha$ and $c = \cos \alpha$). One can confirm that the rotation is:

$$\tan \alpha = (\delta - \delta')/(\lambda_{xx} + \lambda_{zz}) \tag{20}$$

and thus, for instance, for $\sin \alpha$,

$$\sin \alpha = \frac{1}{\Delta}(\delta - \delta') \quad \text{with} \quad \Delta^2 = (\lambda_{xx} + \lambda_{zz})^2 + (\delta - \delta')^2.$$
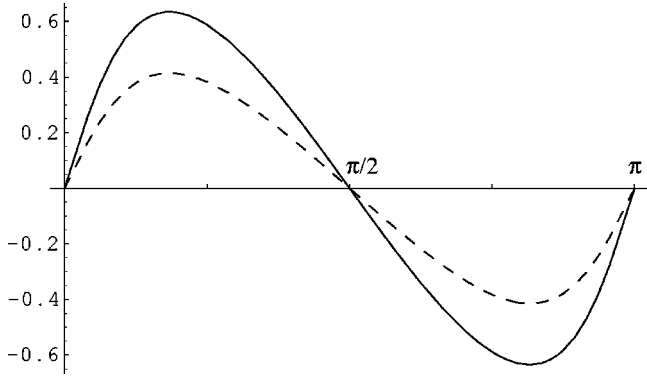
FIG. 9. Off-diagonal element d of the symmetric shear $\underline{\underline{\lambda}}^R$, and angle α of the rotation $\underline{\underline{U}}$ (dotted line) plotted against director rotation θ for the soft deformations of Fig. 5 for a nematic elastomer with anisotropy $r=20$.

Of course there is no body rotation for $\underline{\underline{\lambda}}$ symmetric, that is, $\delta = \delta'$. One can also confirm that the symmetric shear tensor is

$$\underline{\underline{\lambda}}^R = \frac{1}{\Delta}$$

$$\times \begin{pmatrix} \lambda_{xx}(\lambda_{zz}+\lambda_{xx})-\delta'(\delta-\delta') & \lambda_{xx}\delta+\delta'\lambda_{zz} \\ \lambda_{xx}\delta+\delta'\lambda_{zz} & \lambda_{zz}(\lambda_{zz}+\lambda_{xx})+\delta(\delta-\delta') \end{pmatrix}. \quad (21)$$

The results (20) and (21) break the soft modes down into a symmetric shear $\underline{\underline{\lambda}}^R$ followed by a body rotation $\underline{\underline{U}}$ through an angle α about the y axis. Thus the soft mode is $\underline{\underline{\lambda}}_{\text{soft}} = \underline{\underline{U}} \cdot \underline{\underline{\lambda}}^R$. We continue to parametrize them with the director rotation θ. The body rotation is through an angle α given by

$$\tan\alpha = \frac{\sin\theta\cos\theta(\sqrt{r}-1)^2}{2\sqrt{r}+\sin^2\theta(\sqrt{r}-1)^2}$$

$$= \frac{\tan\theta(\sqrt{r}-1)^2}{2\sqrt{r}+\tan^2\theta(r+1)}. \quad (22)$$

For small shear (small director rotation θ), the component of body rotation is small too and proportional to θ:

$$\alpha \approx \theta\frac{(\sqrt{r}-1)^2}{2\sqrt{r}}. \quad (23)$$

For large rotations, $\theta \approx \pi/2$, the rotation α vanishes as we have seen in Fig. 5. Thus it is only at first that body rotation plays a part in accommodating the rotating chains. As the rotation of chains approaches $\pi/2$, the body is simply extended or compressed along the principal axes of the original chain distribution.

The corresponding symmetric shear strain d, the off-diagonal component of $\underline{\underline{\lambda}}^R$, is

$$d = \frac{1}{\Delta}\frac{\sin\theta\cos\theta(r-1)}{\sqrt{r}},$$
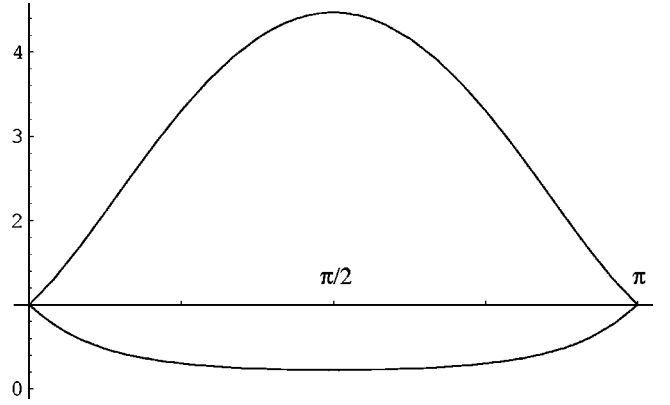

FIG. 10. The two eigenvalues of the pure shear $\underline{\underline{\lambda}}^R$ for a nematic elastomer with anisotropy $r=20$. For a director rotation of $\pi/2$, the soft mode deformation becomes a pure shear with extension and compression ratio $\sqrt{r}$ or $1/\sqrt{r}$, respectively.

which, for small distortions is also proportional to θ, bearing in mind that Δ is constant at first order in θ:

$$d = \frac{1}{\Delta}\frac{\theta(r-1)}{\sqrt{r}}. \quad (24)$$

As we have seen in Fig. 5, it also vanishes at $\theta = \pi/2$, where no further accommodation of the shape tensor by rotation is possible. In Fig. 9, we show the rotation and off-diagonal element of the symmetric shear occurring during soft deformations between $\theta = 0$ and π.

Notice that both quantities are linear in θ for small distortions. As the director of the chain distributions is rotated beyond $\pi/2$, the solid body rotation α and the off-diagonal element of the shear d become negative. At a director rotation of π, the original body shape is recovered, as it should be in an uniaxial nematic system under reflection of the director $\mathbf{n} \to -\mathbf{n}$.

Similarly, the behavior of the pure shear is instructive: for this purpose, we plot in Fig. 10 the two eigenvalues of $\underline{\underline{\lambda}}^R$, which serve as a good measure of the net presence of shear.

Note that, due to incompressibility, the product of the two eigenvalues is constant at 1. Both eigenvalues are linear for small θ, saturate at values $\sqrt{r}$ and $1/\sqrt{r}$, respectively, for a director rotation of $\pi/2$, where the soft mode becomes a simple compression and extension along the principal axes. At a rotation of π, we recover the eigenvalues of the identity matrix.

From Eq. (10), we see that the compression and extension in the x and y directions are quadratic in θ for small director rotations. In other words, u_{zz} and u_{xx} too are proportional to $\theta^2 \propto \alpha^2 \propto d^2$ [by Eqs. (24) and (23)].

VII. CONCLUSIONS

Soft deformations of nematic elastomers result from the rotation of the anisotropic chain shape distribution without distortion and therefore without rubber elastic free energy cost. This is by contrast to classical rubber, where distorsion of the distribution lowers entropy and raises the free energy.

Explicit forms of these soft modes are derived in the case of biaxial nematic elastomers using the Olmsted method of the square roots of tensors developed for the uniaxial case. For both cases we show geometrically what these deformations look like. They correspond to rotations of prolate distributions being accommodated by elastic strains of the body they are inscribed into.

The elasticity of nematic elastomers depends, unlike in classical elastomers, on body rotations (since these can be with respect to the underlying nematic director). At finite strains it is important to isolate the rotational component of the generally nonsymmetric deformations. We make contact with the polar decomposition theorem in this context. In doing so we discuss the roots of tensors that are employed in finding the manifold of soft deformation tensors.

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