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Elastic Response and Ward Identities in Stressed Nematic Elastomers

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Abstract
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Elastic response and Ward identities in stressed nematic elastomers

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Nematic elastomers exhibit a rich elastic response to external stresses. Of particular interest is the semisoft response of elastomers with an anisotropy direction (z) frozen in by a double cross-linking process. This response is characterized by a stress-strain curve for stresses along x perpendicular to z that rises initially, exhibits a nearly flat plateau between two critical values of strain, and then rises again. This paper explores elastic response in semisoft elastomers as a function of externally applied strain. It derives general Ward identities for elastic moduli and shows that the elastic modulus measuring response to xz shears vanishes at the boundaries of the semisoft plateau whereas moduli measuring response to shears perpendicular to the xz plane do not. It then calculates all relevant moduli in a simple model of elastomers and verifies the general Ward-identity predictions.

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I. INTRODUCTION

Nematic Elastomers [1], which combine the orientational properties of nematic liquid crystals with the large-strain mechanical response of rubber, exhibit unusual nonlinear elastic properties brought about by the orientational ordering of their constituent mesogens. They also provide ideal model systems to explore general properties of nonlinear elasticity. Of particular interest is the semisoft [1–5] elastic response of systems with a frozen-in anisotropy direction, which we take to be along z, induced by a second crosslinking [6] under uniaxial strain. This response is characterized by a stress-strain curve shown schematically in Fig. 1 in which the engineering stress σ^I_{xx} as a function of strain $\bar{\Lambda}_{xx}$ (defined to be equal to one when σ^I_{xx} is zero) perpendicular to the direction of nematic order along z first rises linearly, then between critical strains $\bar{\Lambda}_- < \bar{\Lambda}_+$ exhibits a nearly flat plateau, and finally rises again with a larger slope for strains larger than $\bar{\Lambda}_+$. In this paper, we will normally measure strain in terms of the deformation tensor $\underline{\underline{\Lambda}}$ relative to the isotropic reference state that can only be accessed when the anisotropy induced by the second crosslinking is turned off. Thus, when $\sigma^I_{xx}=0$, the system has uniaxial symmetry and a deformation $\underline{\underline{\Lambda}}^r$ relative to the isotropic reference state with nonvanishing components $\Lambda^r_{xx}=\Lambda^r_{yy}\neq\Lambda^r_{zz}$. As σ^I_{xx} is increased from zero, $\bar{\Lambda}_{xx}=\Lambda_{xx}/\Lambda^r_{xx}$ increases from one. The semisoft plateau is associated with a second-order transition [5] in which there is spontaneous development of shear in the xz plane characterized by a nonvanishing Λ_{xz}. Thus, on the plateau, the only nonvanishing equilibrium components of $\underline{\underline{\Lambda}}$ are Λ_{xx}, Λ_{yy}, Λ_{zz}, and Λ_{xz}.

Direct measurements of the stress-strain curve [7,8] of appropriately prepared elastomers exhibit the semisoft plateau. Measurements of anisotropic shear moduli in the presence of strain along x should also provide evidence of the phase transition to semisoft behavior [9]. Extensive measurements of the frequency-dependent shear moduli of nematic elastomers in the absence of strain along x have been carried out [10–16], but very few experiments on elastomers under such strain have been reported [17,18]. A recent paper, however, reports dynamic light scattering experiments [19] on stretched samples that provide measurements of elastic moduli in the semisoft regime. The purpose of this paper is to explore properties, as a function of $\bar{\Lambda}_{xx}$, of the elastic tensor $K'_{ijkl}=\partial^2 f/\partial\Lambda'_{ij}\partial\Lambda'_{kl}$, where f is the free-energy density, measuring response to strains Λ'_{ij} relative to a state with equilibrium strains in the presence of σ^I_{xx}, and related elastic tensors, K^{σ}_{ijkl}, in which various components of stress rather than strain are fixed. K'_{ijkl} is evaluated in the equilibrium state, i.e., at $\Lambda'_{ij}=\delta_{ij}$. Because the states we consider are anisotropic, K'_{ijkl} has many components. In addition because the imposed strain requires a stress that breaks rotational symmetry, K'_{ijkl} has components that are not symmetric under

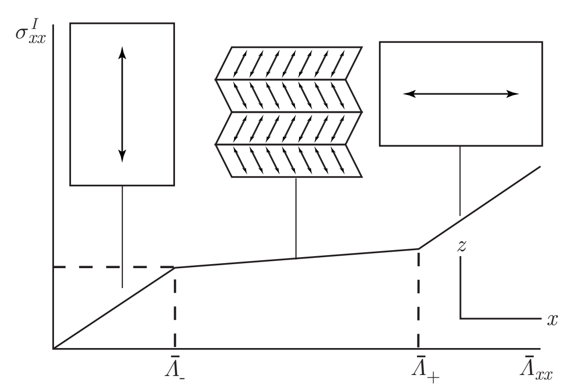

FIG. 1. Schematic of the engineering or first PK stress as a function of stretch $\bar{\Lambda}_{xx}$ showing the nearly flat semisoft plateau between critical values $\bar{\Lambda}_-$ and $\bar{\Lambda}_+$. In the plateau region, Λ_{xz} is nonzero and samples adopt a polydomain structure [9], determined by boundary conditions, with coexisting regions with opposite signs for Λ_{xz}. The double arrows in this figure indicate the direction of the principal strain anisotropy axis.

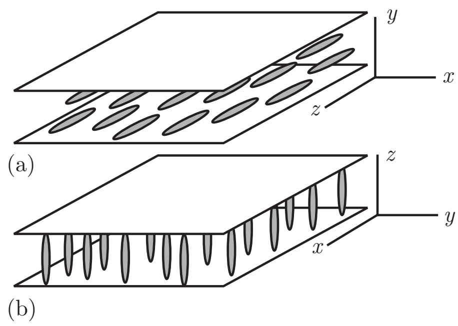

FIG. 2. Geometry for rheology measurements of elastic moduli in films of nematic elastomers. z is the direction of initial nematic anisotropy and x the direction of applied stress. In the parallel geometry of figure (a), the nematic director lies in the plane of the film. This configuration allows measurement of the moduli K'_{xyxy} and K'_{zyzy}. In the perpendicular geometry of figure (b), the director lies normal to the plane of the film. This configuration allows measurement of $K^{y\sigma}_{xzxz}$ and K'_{yzyz}. Clamps used to prevent rotations about axes in the plane of the films force deformations Λ'_{yx} and Λ'_{yz} to be zero in (a) and Λ'_{zy} and Λ'_{zx} to be zero in (b). In addition, boundary conditions at the upper and lower surfaces prevent deformations in the plane of the films, i.e., $\Lambda'_{xz}=\Lambda'_{zx}=0$ in (a) and $\Lambda'_{xy}=\Lambda'_{yx}=0$ in (b).

interchange of i and j or of k and l. Experiments are most easily performed on films, in what we call the parallel geometry, in which the nematic director **n** originally lies in the plane of the film as shown in Fig. 2(a). In this geometry, displacements along y and strains in the xz plane are prohibited, implying that $\Lambda'_{yx}=\Lambda'_{yz}=\Lambda'_{xz}=\Lambda'_{zx}=0$. Experiments, in what we call the perpendicular geometry, in which **n** is originally aligned perpendicular to the plane as shown in Fig. 2(b), though difficult, can also be carried out [18]. In this geometry, displacements along z and strains in the xy plane are prohibited, implying $\Lambda'_{zx}=\Lambda'_{zy}=\Lambda'_{xy}=\Lambda'_{yx}=0$. In the parallel geometry, strains Λ'_{zy} and Λ'_{xy} can be produced by displacing the top surface of the film relative to its bottom surface, allowing measurements of K'_{zyzy} and K'_{xyxy}, which in the literature [10–16] are usually called $G_{\parallel}$ and $G_{\perp}$, respectively. In the perpendicular geometry, strains Λ'_{xz} and Λ'_{yz} are produced in a similar fashion allowing measurements of K'_{xzxz} and K'_{yzyz}. In the absence of external stress σ^J_{xx}, the rotational symmetry in the xy plane guarantees that $K'_{xzxz}=K'_{yzyz}$. The presence of σ^J_{xx} breaks this symmetry and induces difference between K'_{xzxz} and K'_{yzyz}. Measurements of K'_{zxzx} and K'_{yxyx} would involve out-of-plane strains, which would be difficult to produce in the geometries shown in Fig. 2. Sound velocity measurements might, however, be able to probe these moduli.

Our principal result is that the $xyxy$, $yzyz$, and $xzxz$ components of the elastic modulus tensor exhibit the general forms shown in Fig. 3. These forms follow from Ward identities that apply to any model that exhibits a semisoft phase, and they exhibit the same characteristic features found in calculations based on other models [20,21]. In particular, K'_{xyxy} and $K'_{yzyz}=K'_{zyzy}$ are relatively insensitive to the semisoft plateau, though they do exhibit changes in slope, which are likely to be diminished at finite frequency, at the plateau boundaries. The $xzxz$ modulus, on the other hand, vanishes at the boundaries $\bar{\Lambda}_{xx}=\bar{\Lambda}_{-}$ and $\bar{\Lambda}_{+}$ and rises to a maximum in the vicinity of $\bar{\Lambda}_{xx}=(\bar{\Lambda}_{-}+\bar{\Lambda}_{+})/2$. It should be no surprise that this modulus vanishes at the plateau boundaries. As mentioned earlier, the plateau is really a broken-symmetry phase characterized by the spontaneous development of a nonzero Λ_{xz}. The transition to this phase is signaled by the vanishing of the rigidities K'_{xzxz} associated with Λ_{xz}.

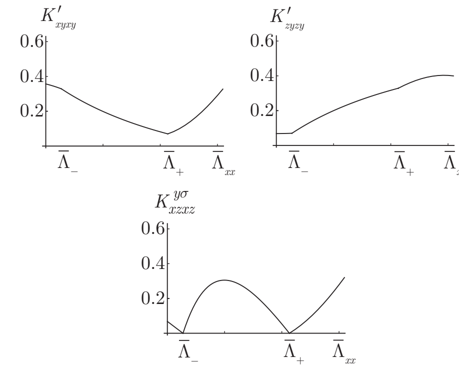

FIG. 3. Elastic moduli K'_{xyxy}, $K'_{zyzy}=K'_{yzyz}$, and $K^{y\sigma}_{xzxz}$ as a function of $\bar{\Lambda}_{xx}$. K'_{xyxy}, and K'_{zyzy} are measured at fixed $\underline{\underline{\Lambda}}'$. $K^{y\sigma}_{xzxz}$ is measured at $\sigma^J_{yy}=0$ and all deformations except Λ'_{yy} at their equilibrium value. These curves were calculated using the minimal model discussed in Sec. V with $\tilde{r}=0.08$, $\tilde{h}=0.05$ and $w/v=1$. The unit of these elastic constants is w.

The curves in Fig. 3 were calculated using the minimal model of Ref. [4], which will be explored in detail in Sec. IV, in which the constraint Tr $\underline{\underline{\Lambda}}^T\underline{\underline{\Lambda}}=3$ rather than the true incompressible constraint det $\underline{\underline{\Lambda}}=1$ is applied. On the plateau, Λ_{xz} is nonzero, and changes in Λ'_{xz} from zero induce linear changes in Tr $\underline{\underline{\Lambda}}^T\underline{\underline{\Lambda}}=\Lambda^2_{xx}+\Lambda^2_{yy}+\Lambda^2_{zz}+\Lambda^2_{xz}$ if the diagonal components of $\underline{\underline{\Lambda}}$ are fixed. As a result the only way to satisfy the Tr $\underline{\underline{\Lambda}}^T\underline{\underline{\Lambda}}=3$ constraint is to allow some or all of the diagonal components of $\underline{\underline{\Lambda}}$ to relax in response to an imposed Λ'_{xz}. This can be accomplished by fixing some or all of diagonal components of stress rather than those of $\underline{\underline{\Lambda}}$. In the perpendicular geometry of Fig. 2(b), it is natural to fix Λ_{xx} and Λ_{zz} and to fix the stress σ^J_{yy} at zero, allowing Λ'_{yy} to relax in response to Λ'_{xz}. We denote the constant-σ^J_{yy} elastic tensor by $K^{y\sigma}_{ijkl}$. $K^{y\sigma}_{xzxz}$ is plotted in Fig. 3. Figure 4 plots the elastic moduli K^{σ}_{xyxy}, K^{σ}_{zyzy}, K^{σ}_{yzyz}, and K^{σ}_{xzxz} in which all relevant stresses rather than strains are fixed. Note that in this case, $K^{\sigma}_{zyzy}\neq K^{\sigma}_{yzyz}$ because K^{σ}_{zyzy} is measured in the parallel geometry in which Λ'_{xy} is not fixed at zero and coupling between K'_{zyzy} and K'_{xyxy} is allowed, whereas K^{σ}_{yzyz} is measured in the perpendicular geometry in which both Λ'_{xy} and Λ'_{yx} are fixed to zero.

As just discussed, the general behavior of the elastic moduli shown in Figs. 3 and 4 is really a consequence of the broken continuous symmetry of the semisoft state, and it follows from Ward identities that are essentially model independent. In Sec. III, we derive Ward identities [22] for elastic moduli that are valid for any strain-dependent free energy that consists of an isotropic part f_{iso}, invariant with respect both to arbitrary rotations of the reference space of initial positions of the sample and to arbitrary rotations of the distorted sample, and an anisotropic part arising from the sec-

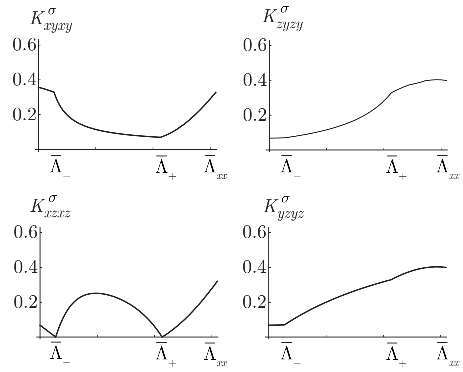

FIG. 4. Elastic moduli K^{σ}_{xzxz}, K^{σ}_{yzyz}, K^{σ}_{xyxy}, and K^{σ}_{zyzy} as a function of $\bar{\Lambda}_{xx}$. These moduli are measured at σ^{J}_{xx} fixed and all other stress except those fixing rotational and in-plane deformations [See Fig. 2] equal to zero. These curves were calculated using the minimal model discussed in Sec. V with $\tilde{r}=0.08$, $\tilde{h}=0.05$ and $w/v=1$. The unit of these elastic constants is w. Note that on the stress plateau K^{σ}_{zyzy} is different from $K^{\sigma}_{yzyz}(=K'_{yzyz})$.

ond crosslinking, which we model as a stress coupled linearly to the strain along z [3,4]. We then calculate in Sec. IV, the moduli of the minimal model introduced in reference [3,4]. In this model, the free energy is expressed as a function of the Cauchy-Saint-Laurent strain tensor $\underline{\underline{u}}=\frac{1}{2}(\underline{\underline{\Lambda}}^T\cdot\underline{\underline{\Lambda}}-\underline{\underline{\delta}})$, where $\underline{\underline{\delta}}$ is the unit matrix, and fluctuations in the trace of $\underline{\underline{u}}$ are suppressed by an energy $\frac{1}{2}B(\mathrm{Tr}\,\underline{\underline{u}})^2$ in the limit $B\to\infty$. This model thus depends only on the traceless part of $\underline{\underline{u}}$. In the appendix, we discuss relaxing the $B\to\infty$ constraint.

The organization of this paper is as follows. Section II establishes notation and derives relations between the elastic tensor K'_{ijkl}, K^{σ}_{ijkl}, and the more familiar elastic tensor C'_{ijkl} measuring response to Cauchy-Saint-Laurent strains u'_{ij}. Section III derives Ward identities [22] for both C'_{ijkl} and K'_{ijkl}, and Section IV treats the minimal model. The Appendix considers the minimal model when $B\neq\infty$ and the related model in which minimal-model compression energy is replaced by the true compression energy $\frac{1}{2}B(\delta V/V_0)^2$ where $\delta V=V-V_0$ is the volume change and V_0 is the volume of the equilibrium reference state.

II. STRAINS, ENERGIES, AND ELASTIC TENSORS

Distortions of an elastic medium are described by mappings of mass points $\mathbf{x}$ of its undistorted reference state to points $\mathbf{R}(\mathbf{x})$ of its distorted state. We will refer to the set of points $\mathbf{x}$ as the reference space S_R and the set of points $\mathbf{R}(\mathbf{x})$ as the target space S_T. Slowly varying distortions of the medium are described by the Cauchy deformation tensor $\underline{\underline{\Lambda}}$ with components

$$\Lambda_{ij}=\frac{\partial R_i(\mathbf{x})}{\partial x_j}, \tag{2.1}$$

where $i,j=x,y,z$ in the three-dimensional space, we consider here. In the absence of external forces defining specific directions in S_T, elastic energies are invariant under rigid rotations of the distorted object, and elastic energies are commonly expressed in terms of the Lagrangian nonlinear strain tensor,

$$\underline{\underline{u}}=\frac{1}{2}(\underline{\underline{\Lambda}}^T\cdot\underline{\underline{\Lambda}}-\underline{\underline{\delta}}), \tag{2.2}$$

where $\underline{\underline{\delta}}$ is the unit matrix, which is invariant under these rotations. Since u_{ij} is symmetric, it has six independent components u_{xx}, u_{yy}, u_{zz}, u_{xy}, u_{xz}, and u_{yz}.

In general, the elastic free energy density f of a material can be decomposed into an isotropic part $f_{\rm iso}$ that is invariant with respect to all rotations in S_R (i.e., with respect to rotations of $\mathbf{x}$) and an anisotropic part $f_{\rm ani}$ that is not:

$$f(\underline{\underline{u}})=f_{\rm iso}(\underline{\underline{u}})+f_{\rm ani}(\underline{\underline{u}}). \tag{2.3}$$

In a crystalline solid, $f_{\rm ani}$ is invariant under the point group operation of the crystal. In a liquid crystalline elastomer, $f_{\rm ani}$ reflects the anisotropy frozen in via a second cross-linking operation [6]. In the presence of a second Piola-Kirchhoff (PK) stress [23] σ_{ij},

$$\frac{\partial f}{\partial u_{ij}}=\sigma_{ij}, \tag{2.4}$$

equilibrium is determined by minimizing the Gibbs free energy per unit volume of the reference space

$$g(\underline{\underline{u}})=f(\underline{\underline{u}})-\sigma_{ij}u_{ij}. \tag{2.5}$$

The equilibrium strain $\underline{\underline{u}}^0$ may be nonzero either because of nonzero σ_{ij} or because of a phase transition from the original reference state in which it is zero. The energy of elastic distortions relative to the equilibrium state can be obtained by expanding g in powers of $\delta\underline{\underline{u}}=\underline{\underline{u}}-\underline{\underline{u}}^0$. To harmonic order in $\delta\underline{\underline{u}}$,

$$\delta\mathcal{G}\equiv\int_{V_0}d^3x\,\delta g=\frac{1}{2}\int_{V_0}d^3x\,C_{ijkl}\delta u_{ij}\delta u_{kl}, \tag{2.6}$$

where the integrals are over the volume V_0 of the reference state and where

$$C_{ijkl}=\frac{\partial^2 f}{\partial u_{ij}\,\partial u_{kl}} \tag{2.7}$$

is the elastic tensor, which is symmetric under the interchanges $i\leftrightarrow j$, $k\leftrightarrow l$, and $ij\leftrightarrow kl$. Note the differentiations in Eqs. (2.4) and (2.7) are, however, taken by treating $\underline{\underline{u}}$ as an arbitrary tensor. The free energy can alternatively be expressed in terms of the Voigt notation [24] in which $\underline{\underline{u}}$ is represented by its six independent components, u_α, $\alpha=1,\cdots,6$, which, respectively, correspond to $u_{xx},u_{yy},u_{zz},u_{xz},u_{yz},u_{xy}$. In this case, the elastic tensor,

$$C^{v}_{\alpha\beta}=\frac{\partial^2 f}{\partial u_\alpha\,\partial u_\beta}, \tag{2.8}$$

becomes a symmetric 6×6 matrix with

$$C^{v}_{\alpha\beta}=C_{aabb}\quad\text{for}\quad\alpha,\beta=1,2,3; \tag{2.9a}$$

$$C^{v}_{\alpha\beta}=2C_{aacd}\quad\text{for}\quad\alpha=1,2,3,\quad\beta=4,5,6; \tag{2.9b}$$

$$C^{v}_{\alpha\beta} = 4C_{abcd} \quad \text{for} \quad \alpha,\beta = 4,5,6, \tag{2.9c}$$

where C_{ijkl} is the elastic tensor given in Eq. (2.7). The counting factors of 2 in Eq. (2.9b) and 4 in Eq. (2.9c) are a simple consequence of the symmetry of u_{ij} under interchange of i and j.

The equilibrium state with strain $\underline{\underline{u}}^0$ has a particular orientation in space and is characterized by a deformation tensor $\underline{\underline{\Lambda}}^0$ with $\underline{\underline{u}}^0 = \frac{1}{2}(\underline{\underline{\Lambda}}^{0T} \cdot \underline{\underline{\Lambda}}^0 - \underline{\underline{\delta}})$. Mass points that were originally at positions $\mathbf{x}$ in the reference space are now at positions

$$x_i' = \Lambda^0_{ij} x_j. \tag{2.10}$$

Deformations relative to the reference space S_R' of points $\mathbf{x}'$ are described by the tensor $\Lambda'_{ij} = \partial R_i / \partial x_j'$, and

$$\Lambda_{ij} = \Lambda'_{ik}\Lambda^0_{kj}. \tag{2.11}$$

This allows us to express $\delta\underline{\underline{u}}$ in terms of the Lagangian strain $\underline{\underline{u}}' = \frac{1}{2}(\underline{\underline{\Lambda}}'^T \cdot \underline{\underline{\Lambda}}' - \underline{\underline{\delta}})$ relative to S_R':

$$\delta\underline{\underline{u}} = \frac{1}{2}(\underline{\underline{\Lambda}}^T \cdot \underline{\underline{\Lambda}} - \underline{\underline{\delta}}) - \frac{1}{2}(\underline{\underline{\Lambda}}^{0T} \cdot \underline{\underline{\Lambda}}^0 - \underline{\underline{\delta}}) = \underline{\underline{\Lambda}}^{0T} \cdot \underline{\underline{u}}' \cdot \underline{\underline{\Lambda}}^0. \tag{2.12}$$

With the aid of this relation, the harmonic energy of Eq. (2.6) can be expressed in terms of $\underline{\underline{u}}'$:

$$\delta\mathcal{G} = \frac{1}{2}\int_{V_0'} d^3x' \, C'_{ijkl} u'_{ij} u'_{kl}, \tag{2.13}$$

where the integral is over the volume $V_0' = \det \underline{\underline{\Lambda}}^0 V_0$ of the equilibrium distorted material and

$$C'_{ijkl} = \frac{1}{\det \underline{\underline{\Lambda}}^0} \Lambda^0_{ip}\Lambda^0_{kr} C_{pqrs} \Lambda^{0T}_{qj}\Lambda^{0T}_{sl}. \tag{2.14}$$

We consider here only deformations in which $\underline{\underline{\Lambda}}$ is spatially uniform. In real systems, domain structures such as those depicted in Fig. 1 form [9,25] in response to boundary conditions that are incompatible with a uniform $\underline{\underline{\Lambda}}$. The domain walls between these domains cost additional energy, which can be studied [9] by adding Frank elastic energy of the director to the elastic energy of the elastomer. The transition to the semi-soft striped state is, however, driven by the bulk elastic energy, and both the energy and strain in the region between the domain walls, which are separated by distances large compared to their width, is well described by the uniform deformation theory described here.

External stresses, resulting for example from stretching a sample between two clamps, are conveniently described by the engineering or first PK stress tensor σ^I_{ij} specifying the force per unit area in direction i across a surface of the reference material oriented with normal along j. This is a mixed tensor whose i index responds to rotations in S_T and whose j index responds to rotations in S_R. It defines a direction, and thus breaks rotational invariance, in S_T. In the presence of first PK stresses, distortions of the sample cannot be described by the rotationally invariant Lagrangian strain tensor alone. Elastic energies relative to such broken-symmetry states must be expressed in terms of the nine independent components of the nonsymmetric deformation tensor $\underline{\underline{\Lambda}}$. The Gibbs energy density of such systems is

$$g^I = f(\underline{\underline{u}}) - \sigma^I_{ij}\Lambda_{ij}, \tag{2.15}$$

where f depends only on $\underline{\underline{u}}$, which in turn depends on $\underline{\underline{\Lambda}}$. Minimizing this energy yields the equation of state for Λ,

$$\sigma^I_{ij} = \frac{\partial f}{\partial \Lambda_{ij}} = \Lambda_{ik}\frac{\partial f}{\partial u_{kj}}. \tag{2.16}$$

Let Λ^0 be the solution to Eq. (2.16) and define $\delta\underline{\underline{\Lambda}} = \underline{\underline{\Lambda}} - \underline{\underline{\Lambda}}^0$. To harmonic order in $\delta\underline{\underline{\Lambda}}$,

$$\delta\mathcal{G}^I \equiv \int_{V_0} d^3x \, \delta g^I = \frac{1}{2}\int_{V_0} d^3x K_{ijkl}\delta\Lambda_{ij}\delta\Lambda_{kl}, \tag{2.17}$$

where

$$K_{ijkl} = \left.\frac{\partial^2 f}{\partial \Lambda_{ij}\, \partial \Lambda_{kl}}\right|_{\underline{\underline{\Lambda}} = \underline{\underline{\Lambda}}^0} \tag{2.18a}$$

$$= \delta_{ik}\sigma_{jl} + \Lambda^0_{ip}\Lambda^0_{kr}C_{pjrl}, \tag{2.18b}$$

where $\sigma_{ij} = (\Lambda^0)^{-1}_{ik}\sigma^I_{kj}$ is the second PK stress tensor of Eq. (2.4) and C_{ijkl} is the elastic tensor of Eq. (2.7). Note that K_{ijkl} is symmetric under the interchange $ij \leftrightarrow kl$, but not under the interchanges $i \leftrightarrow j$ or $k \leftrightarrow l$. Then, using Eq. (2.11), $\delta\mathcal{G}^I$ can be expressed as

$$\delta\mathcal{G}^I = \frac{1}{2}\int_{V_0'} d^3x' K'_{ijkl}\delta\Lambda'_{ij}\delta\Lambda'_{kl}, \tag{2.19}$$

where $V_0' = (\det \underline{\underline{\Lambda}}_0)V_0$, $\delta\underline{\underline{\Lambda}}' = \underline{\underline{\Lambda}}' - \underline{\underline{\delta}}$, and

$$K'_{ijkl} = \frac{1}{\det \underline{\underline{\Lambda}}^0} K_{iqks}\Lambda^{0T}_{qj}\Lambda^{0T}_{sl} \tag{2.20a}$$

$$= \delta_{ik}\sigma'_{jl} + C'_{ijkl} \tag{2.20b}$$

with $\sigma'_{jl} = (\det \underline{\underline{\Lambda}}^0)^{-1}\Lambda^0_{jq}\sigma_{qs}\Lambda^{0T}_{sl} = (\det \underline{\underline{\Lambda}}^0)^{-1}\partial f / \partial u'_{jl}$ and C'_{ijkl} given by Eq. (2.14). Thus, K'_{ijkl} consists of a part C'_{ijkl} that is symmetric under $i \leftrightarrow j$ and $k \leftrightarrow l$ and a part $\delta_{ik}\sigma'_{jl}$ that is not. The latter part leads to restoring forces for rotations away from the preferred direction defined by σ^I_{ij}. Consider, for example, an external stress σ^I_{xx}, arising from a force along the x direction across surfaces perpendicular to x in the reference space, that produces a diagonal deformation tensor with components Λ^0_{xx}, Λ^0_{yy}, and Λ^0_{zz}. The elastic constants associated with deformations Λ'_{xz} and Λ'_{zx} in the xz plane are $K'_{xzxz} = K'_{xzzx} = K'_{zxxz} = C'_{xzxz}$ and $K'_{zxzx} = \sigma'_{xx} + C'_{xzxz}$, and the associated harmonic energy is

$$\delta\mathcal{G}^I = \frac{1}{2}\int_{V_0'} d^3x'[\sigma'_{xx}(\delta\theta_y - \epsilon_{xz})^2 + 4C'_{xzxz}\epsilon^2_{xz}], \tag{2.21}$$

where $\delta\theta_y = (\Lambda'_{xz} - \Lambda'_{zx})/2$ is the rotation angle about the y axis and $\epsilon_{xz} = (\Lambda'_{xz} + \Lambda'_{zx})/2$ is the linearized symmetric strain tensor.

To keep notation more compact, in what follows we will replace $\underline{\underline{\Lambda}}^0$ by $\underline{\underline{\Lambda}}$ in expressions for C'_{ijkl} and K'_{ijkl} arising from Eqs. (2.14) and (2.20a).

In the particular systems we consider, the only nonvanishing components of the equilibrium deformation tensor are Λ_{xx}, Λ_{yy}, Λ_{zz}, and Λ_{xz}, and the only nonvanishing component of σ'_{ij} is $\sigma'_{xx}=\Lambda_{xx}^2\sigma_{xx}/\det\underline{\underline{\Lambda}}$. In this case,

$$K'_{ijkl}=\delta_{ik}\delta_{jx}\delta_{lx}\sigma'_{xx}+C'_{ijkl}, \tag{2.22}$$

implying that K'_{ijkl} and C'_{ijkl} only differ in their $xxxx$, $yxyx$, and $zxzx$ components and then by σ'_{xx}. Here, we consider only the geometries shown in Fig. 2 in which the top plate is displaced relative to the bottom plate to produce deformations Λ'_{xy} and Λ'_{zy} in the parallel geometry of Fig. 2(a) and Λ'_{xz} and Λ'_{yz} in perpendicular geometry of Fig. 2(b).

To conclude this section, we discuss the definition of the constant stress elastic tensor, K^{σ}_{ijkl}, in which all relevant components of stress rather than those of strain are fixed. In the parallel geometry, the relevant components of strain are the diagonal components of $\underline{\underline{\Lambda}}'$ and the off-diagonal components $\Lambda'_{xy},\Lambda'_{zy}$; in the perpendicular geometry, the relevant strain components are Λ'_{xz}, Λ'_{yz}, and the diagonal components. When these components are not fixed by boundary conditions, they are determined by

$$K'_{\alpha\beta}\delta\Lambda'_{\beta}=\delta\sigma'^{I}_{\alpha}, \tag{2.23}$$

and $\delta\Lambda'_{\alpha}=S'_{\alpha\beta}\delta\sigma'^{I}_{\beta}$ where $S'_{\alpha\beta}=(K'^{-1})_{\alpha\beta}$ is the compliance tensor. The response of $\delta\Lambda'_{\alpha}$ to a stress change whose only nonvanishing component is $\delta\sigma'^{I}_{\alpha}$, is then

$$K^{\sigma}_{\alpha\alpha}\equiv\frac{\delta\sigma'^{I}_{\alpha}}{\delta\Lambda'_{\alpha}}=\frac{1}{S'_{\alpha\alpha}}, \tag{2.24}$$

where no summation over α is implied.

III. WARD IDENTITIES

The rotational invariances of $\underline{\underline{u}}$ in S_T and of f_{iso} in S_R give rise to Ward identities [22] relating various components of the elastic tensors. These relations establish the conditions of external stress under which certain elastic constants vanish for particular forms of f_{ani} independent of the form of f_{iso}. They thus provide powerful almost model-independent insights. In this section, we will explore Ward identities for C_{ijkl} and K_{ijkl} for the particular case in which

$$f_{\text{ani}}=-h_{ij}u_{ij}\equiv-hu_{zz}. \tag{3.1}$$

This form (which is equivalent to an external second PK stress) for f_{ani} is the simplest one describing the effects of a second crosslinking of a nematic elastomer under uniaxial stress in the z direction. Note that it establishes a preferred direction in the reference space but remains invariant under rotations in the target space; it is not equivalent to the energy arising from the application of a load along the z direction in the target space [26].

In what follows, we will analyze in detail systems subjected to external stress $\sigma^{I}_{xx}=\Lambda_{xx}\sigma_{xx}$ along x. We will derive a number of properties of the elastic tensors of systems subject to this stress that are independent of the form of f_{iso}: In particular, $C'_{xzxz}=K'_{xzxz}$ vanishes but $C'_{xyxy}=K'_{xyxy}$ and $C'_{yzyz}=K'_{yzyz}$ remain finite at the boundaries of the stress plateau.

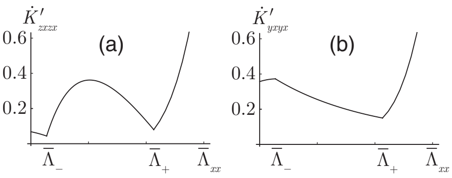

FIG. 5. (a) $\mathring{K}'_{zxzx}$ and (b) $\mathring{K}'_{yxyx}$, in units of w as a function of $\bar{\Lambda}_{xx}$, for $\tilde{r}=0.08$, $\tilde{h}=0.05$ and $w/v=1$. These moduli measure rotations away from the x direction defined by σ^{I}_{xx} and have a σ_{xx} contribution. Thus, at the plateau boundaries, K'_{zxzx} is equal to σ'_{xx} rather than zero.

The fact that f_{ani} is linear in u_{zz} implies that the equilibrium value of $\underline{\underline{u}}$ is nonzero even when σ_{xx} is zero. Physical quantities like stresses and elastic constants are usually measured as a function of deformation $\bar{\underline{\underline{\Lambda}}}$ relative to the equilibrium reference state at $\sigma_{xx}=0$ for which $\underline{\underline{\Lambda}}^{r}=\underline{\underline{\Lambda}}(\sigma_{xx}=0)$. Then $\underline{\underline{\Lambda}}=\bar{\underline{\underline{\Lambda}}}\cdot\underline{\underline{\Lambda}}^{r}$, and in particular $\Lambda_{xx}=\Lambda^{r}_{xx}\bar{\Lambda}_{xx}$. Figures 3–5 show components of the elastic tensors as a function of $\bar{\Lambda}_{xx}$.

A. Identities for C_{ijkl}

Under rotation in S_R in which $x_i\to U^{a}_{ij}x_j$, where $\underline{\underline{U}}^{a}$ is a rotation matrix for rotations about an arbitrary axis a, $\mathbf{R}(\mathbf{x})\to\mathbf{R}(\underline{\underline{U}}\cdot\mathbf{x})$, and $\underline{\underline{u}}\to\underline{\underline{U}}\cdot\underline{\underline{u}}\cdot\underline{\underline{U}}^{-1}$. For infinitesimal rotations through θ_a about an axis $a=x,y,z$,

$$U^{a}_{ij}=\delta_{ij}-\theta_a\epsilon_{aij}, \tag{3.2}$$

and

$$u_{ij}\to u_{ij}+\theta_a(\epsilon_{aki}u_{kj}+\epsilon_{akj}u_{ik}). \tag{3.3}$$

Since f_{iso} is invariant under $\underline{\underline{U}}^{a}$, we have

$$\frac{df}{d\theta_a}=\frac{\partial f}{\partial u_{ij}}\frac{\partial u_{ij}}{\partial\theta_a}=\frac{df_{\text{ani}}}{d\theta_a}=-h_{ij}\frac{\partial u_{ij}}{\partial\theta_a}, \tag{3.4}$$

from which we obtain

$$(\epsilon_{ari}u_{rj}+\epsilon_{arj}u_{ir})(\sigma_{ij}+h_{ij})=0, \tag{3.5}$$

where $\sigma_{ij}=\partial f/\partial u_{ij}$. This is the fundamental Ward identity from which we will obtain relations among elastic constants. It applies to arbitrary tensors $\underline{\underline{u}}$ transforming according to Eq. (3.2) under rotations including ones that may not be symmetric or ones that are symmetric and traceless. The latter observation will be of importance in our discussion in Sec. IV of a simple model with a traceless constraint on $\underline{\underline{u}}$.

We consider first what Eq. (3.5) says about stresses and strains when $h_{ij}=h\delta_{iz}\delta_{jz}$ and $\sigma_{ij}=\sigma_{xx}\delta_{xi}\delta_{xj}$. For $a=x,y,z$ we obtain, respectively,

$$2u_{yz}h=0,\quad a=x \tag{3.6a}$$

$$2u_{xz}(h-\sigma_{xx})=0,\quad a=y \tag{3.6b}$$

$$2u_{xy}\sigma_{xx}=0,\quad a=z. \tag{3.6c}$$

Thus when h and σ_{xx} are nonzero, both u_{yz} and u_{xy} must be zero. In addition, u_{xz} must be zero as long as $\sigma_{xx}-h\neq0$.

When $\sigma_{xx}=h$, however, u_{xz} may be nonzero, and, indeed, this has been verified in detail in a specific model [3] as will be reviewed in Sec. IV. Thus in equilibrium, we need only consider nonvanishing u_{xz} and diagonal components of u_{ij}.

To obtain Ward identities for elastic constants, we merely differentiate Eq. (3.5) with respect to u_{kl}:

$$2\epsilon_{ari}u_{rj}C_{ijkl} + \epsilon_{aki}(\sigma_{il} + h_{il}) + \epsilon_{ali}(\sigma_{ik} + h_{ik}) = 0. \quad (3.7)$$

Again these equations also apply to a traceless strain tensor, in which case the elastic tensor satisfies traceless constraints $C_{iikl}=C_{ijkk}=0$. We are interested in equilibrium values of C_{ijkl}, so strictly speaking we should use $\underline{\underline{u}}^0$ in these equations. To keep notation more compact, however, we will continue to use $\underline{\underline{u}}$ rather than $\underline{\underline{u}}^0$ in equations like this when there is little room for ambiguity. Setting $a=x,y,z$, $\sigma_{ij}=\sigma_{xx}\delta_{ix}\delta_{jx}$ and $h_{ij}=h\delta_{iz}\delta_{hz}$ in Eq. (3.7), we obtain three equations relating elastic constants:

$$C_{yzkl} = \frac{1}{2}\frac{h}{u_{zz}-u_{yy}}I_{kl}^{yz} - \frac{u_{xz}}{u_{zz}-u_{yy}}C_{xykl}, \quad (3.8a)$$

$$C_{xzkl} = \frac{1}{2}\frac{h-\sigma_{xx}}{u_{zz}-u_{xx}}I_{kl}^{xz} - \frac{u_{xz}}{u_{zz}-u_{xx}}(C_{xxkl}-C_{zzkl}), \quad (3.8b)$$

$$C_{xykl} = \frac{1}{2}\frac{\sigma_{xx}}{u_{xx}-u_{yy}}I_{kl}^{xy} - \frac{u_{xz}}{u_{xx}-u_{yy}}C_{yzkl}, \quad (3.8c)$$

where $I_{kl}^{rs}=\delta_{kr}\delta_{ls}+\delta_{ks}\delta_{lr}$. It will be important to remember in the calculations of specific components of C_{ijkl} that follow that u_{xz} is nonzero only when $h=\sigma_{xx}$. When $h\neq\sigma_{xx}$, Eqs. (3.8) imply

$$C_{xyxy} = \frac{1}{2}\frac{\sigma_{xx}}{u_{xx}-u_{yy}}, \quad (3.9a)$$

$$C_{yzyz} = \frac{1}{2}\frac{h}{u_{zz}-u_{yy}}, \quad (3.9b)$$

$$C_{xzxz} = \frac{1}{2}\frac{h-\sigma_{xx}}{u_{zz}-u_{xx}}. \quad (3.9c)$$

All other components of C_{xykl}, C_{yzkl}, and C_{xzkl} other than those obtained from C_{xyxy}, C_{yzyz}, and C_{xzxz} by trivial symmetry operation are zero when $h\neq\sigma_{xx}$. These equations imply that C_{xzxz} approaches zero as $\sigma_{xx}\to h$ when $u_{zz}\neq u_{xx}$. In addition, $(u_{zz}-u_{yy})C_{yzyz}$ approaches $(u_{xx}-u_{yy})C_{xyxy}$ in this limit.

When $h=\sigma_{xx}$, u_{xz} can be nonzero, and

$$C_{xyyz} = -C_{yzyz}\frac{u_{xz}}{u_{xx}-u_{yy}} = -C_{xyxy}\frac{u_{xz}}{u_{zz}-u_{yy}}, \quad (3.10a)$$

$$C_{xyxy} = \frac{u_{zz}-u_{yy}}{u_{xx}-u_{yy}}C_{yzyz} \quad (3.10b)$$

$$= \frac{1}{2}h\frac{(u_{zz}-u_{yy})}{(u_{xx}-u_{yy})(u_{zz}-u_{yy})-u_{xz}^2}, \quad (3.10c)$$

$$C_{yzyz} = \frac{1}{2}h\frac{(u_{xx}-u_{yy})}{(u_{xx}-u_{yy})(u_{zz}-u_{yy})-u_{xz}^2}, \quad (3.10d)$$

$$C_{xzxz} = (C_{zzzz}-2C_{xxzz}+C_{xxxx})\frac{u_{xz}^2}{(u_{zz}-u_{xx})^2}, \quad (3.10e)$$

$$C_{xxxz} = (C_{xxzz}-C_{xxxx})\frac{u_{xz}}{u_{zz}-u_{xx}}, \quad (3.10f)$$

$$C_{zzxz} = (C_{zzzz}-C_{xxzz})\frac{u_{xz}}{u_{zz}-u_{xx}}. \quad (3.10g)$$

Note that $C_{yzyz}(u_{xx},u_{zz})=C_{xyxy}(u_{zz},u_{xx})$.

Our ultimate interest is the elastic tensor C'_{ijkl}. Equation (2.14) determines C'_{ijkl} in terms of C_{ijkl} and the equilibrium deformation tensor. As discussed earlier, to keep our notation compact in what follows, we will denote this tensor by $\underline{\underline{\Lambda}}$ rather than $\underline{\underline{\Lambda}}^0$. u_{xz} is nonzero if either Λ_{xz} or Λ_{zx} is nonzero along with the diagonal components Λ_{xx}, Λ_{yy}, and Λ_{zz}. As we shall see in the next subsection, Λ_{zx} must be zero in equilibrium for the first PK stress σ_{xx}^I along x. Thus, we are left with Λ_{xz} as the only nonvanishing off-diagonal component of $\underline{\underline{\Lambda}}$. From Eq. (2.14) we have $C'_{xyxy}=(\Lambda_{xx}^2C_{xyxy}+2\Lambda_{xz}\Lambda_{xx}C_{xyyz}+\Lambda_{xz}^2C_{yzyz})\Lambda_{yy}^2/\det\underline{\underline{\Lambda}}$, $C'_{yzyz}=\Lambda_{yy}^2\Lambda_{zz}^2C_{yzyz}/\det\underline{\underline{\Lambda}}$, and $C'_{xzxz}=(\Lambda_{xx}^2C_{xzxz}+2\Lambda_{xz}\Lambda_{xx}C_{xzzz}+\Lambda_{xz}^2C_{zzzz})\Lambda_{zz}^2/\det\underline{\underline{\Lambda}}$. Finally expressing u_{ij} in terms of the components of Λ_{ij}, we obtain

$$C'_{xyxy} = \frac{1}{\det\underline{\underline{\Lambda}}}\frac{\Lambda_{xx}^2\Lambda_{yy}^2\sigma_{xx}}{\Lambda_{xx}^2-\Lambda_{yy}^2}, \quad (3.11a)$$

$$C'_{yzyz} = \frac{1}{\det\underline{\underline{\Lambda}}}\frac{\Lambda_{yy}^2\Lambda_{zz}^2h}{\Lambda_{zz}^2-\Lambda_{yy}^2}, \quad (3.11b)$$

$$C'_{xzxz} = \frac{1}{\det\underline{\underline{\Lambda}}}\frac{\Lambda_{zz}^2\Lambda_{xx}^2(h-\sigma_{xx})}{\Lambda_{zz}^2-\Lambda_{xx}^2}, \quad (3.11c)$$

when $h\neq\sigma_{xx}$ and

$$C'_{xyxy} = \frac{\Lambda_{yy}^2[\Lambda_{xx}^2(\Lambda_{zz}^2-\Lambda_{yy}^2)-\Lambda_{yy}^2\Lambda_{xz}^2]}{\det\underline{\underline{\Lambda}}\Delta}h, \quad (3.12a)$$

$$C'_{yzyz} = \frac{\Lambda_{yy}^2\Lambda_{zz}^2(\Lambda_{xx}^2-\Lambda_{yy}^2)}{\det\underline{\underline{\Lambda}}\Delta}h, \quad (3.12b)$$

$$C'_{xzxz} = \frac{\Lambda_{zz}^2\Lambda_{xz}^2}{\det\underline{\underline{\Lambda}}[(\Lambda^2)_{zz}-\Lambda_{xx}^2]^2}\times[(\Lambda^2)_{zz}^2C_{zzzz}-2\Lambda_{xx}^2(\Lambda^2)_{zz}C_{xxzz} + \Lambda_{xx}^4C_{xxxx}], \quad (3.12c)$$

where $\Delta=(\Lambda_{xx}^2-\Lambda_{yy}^2)(\Lambda_{zz}^2-\Lambda_{yy}^2)-\Lambda_{yy}^2\Lambda_{xz}^2$ and $(\Lambda^2)_{zz}=\Lambda_{zz}^2+\Lambda_{xz}^2$, when $h=\sigma_{xx}$. Note that at the boundaries of the stress plateau where $\Lambda_{xz}=0$ and $h=\sigma_{xx}$, $C'_{yzyz}(\Lambda_{xx},\Lambda_{zz})=C'_{xyxy}(\Lambda_{zz},\Lambda_{xx})$.

We have expressed C'_{ijkl} in Eqs. (3.11) and (3.12) in terms of $\underline{\underline{\Lambda}}$ rather than $\bar{\underline{\underline{\Lambda}}}$. The transformation to $\bar{\underline{\underline{\Lambda}}}$ is trivial. Eventually, we will use equations of state to express Λ_{zz}, Λ_{yy}, and Λ_{xz} as a function of $\bar{\Lambda}_{xx}=\Lambda_{xx}/\Lambda_{xx}^r$.

B. Identities for K'_{ijkl}

Though the Ward-identity relations among the components of K'_{ijkl} can be obtained from those for C'_{ijkl} and the Eq. (2.20b) relating K'_{ijkl} to C'_{ijkl}, it is instructive to derive these relations directly from Ward identities obtained via the rotations of $\underline{\underline{\Lambda}}$ rather than of $\underline{\underline{u}}$. $\underline{\underline{\Lambda}}$ responds independently to rotations in S_R and in S_T. Rotations in S_R are described by the rotation matrix $\underline{\underline{U}}^a$ of Eq. (3.2), and

$$\Lambda_{ij} \to \Lambda_{ik} U_{kj}^{a-1} \approx \Lambda_{ij} + \theta_a \Lambda_{ik} \epsilon_{akj}. \tag{3.13}$$

Rotations in S_T are described by a rotation matrix $\underline{\underline{O}}^a$ identical to $\underline{\underline{U}}^a$ but with θ_a replaced by another angle ϕ_a. Under $\underline{\underline{O}}^a$,

$$\Lambda_{ij} \to O_{ik}^a \Lambda_{kj} \approx \Lambda_{ij} - \phi_a \epsilon_{aik} \Lambda_{kj}. \tag{3.14}$$

$\underline{\underline{u}}$ is invariant under rotations in S_T, so f_{ani} is independent of ϕ_a but not of θ_a. The variation of f with respect to θ_a and ϕ_a yield

$$\frac{df}{d\theta_a} - \frac{df_{\text{ani}}}{d\theta_a} = \epsilon_{arj}(\sigma_{ij}^J + \delta_{jz}\Lambda_{iz} h)\Lambda_{ir} = 0, \tag{3.15a}$$

$$\frac{df}{d\phi_a} = - \epsilon_{air}\sigma_{ij}^J \Lambda_{rj} = 0, \tag{3.15b}$$

where as before $\sigma_{ij}^J = \partial f / \partial \Lambda_{ij}$. These equations are the basic Ward identities, and following procedures similar to those of the preceding subsection, they will be used both to determine which components of $\underline{\underline{\Lambda}}$ must be zero in equilibrium and to derive relations, after differentiation with respect to Λ_{kl}, among components of K_{ijkl}.

First consider constraints imposed by Eqs. (3.15) when the only nonvanishing components of σ_{ij}^J is σ_{xx}^J. In this case Eqs. (3.15) lead to five equations (a sixth equation arising from the variation of ϕ_x is simply zero):

$$\sigma_{xx}^J \Lambda_{xz} - h \Lambda_{ix} \Lambda_{iz} = 0, \quad (\theta_y), \tag{3.16a}$$

$$\sigma_{xx}^J \Lambda_{zx} = 0, \quad (\phi_y), \tag{3.16b}$$

$$h \Lambda_{iz} \Lambda_{iy} = 0, \quad (\theta_x), \tag{3.16c}$$

$$\sigma_{xx}^J \Lambda_{xy} = 0, \quad (\theta_z), \tag{3.16d}$$

$$\sigma_{xx}^J \Lambda_{yx} = 0, \quad (\phi_z), \tag{3.16e}$$

where the quantities in parenthesis indicate which angle was varied to produce the equation to the left. Equations (3.16d) and (3.16e) immediately imply that $\Lambda_{xy} = \Lambda_{yx} = 0$ in equilibrium. In addition, for the boundary conditions we are interested in [See Fig. 2], either Λ_{yz} or Λ_{zy} must be zero. Then from Eq. (3.16c) and $\Lambda_{xy} = 0$, we obtain $\Lambda_{yz}\Lambda_{yy} + \Lambda_{zy}\Lambda_{zz} = 0$ implying that Λ_{yz} must be zero if Λ_{zy} is zero and vice versa. Thus, both Λ_{yz} and Λ_{zy} are zero. Equation (3.16b) imposes $\Lambda_{zx} = 0$ as long as σ_{xx}^J is nonzero. Thus, Λ_{xz} is the only off-diagonal component of $\underline{\underline{\Lambda}}$ that can be nonzero in equilibrium. Equation (3.16a) then becomes

$$(\sigma_{xx}^J - h \Lambda_{xx}) \Lambda_{xz} = 0. \tag{3.17}$$

Thus, we conclude that either $\Lambda_{xz} = 0$ or $\sigma_{xx}^J - h\Lambda_{xx} = 0$. The latter condition is identical to the condition $\sigma_{xx} - h = 0$ obtained in the previous subsection.

To obtain relations among the components of K_{ijkl}, we differentiate Eq. (3.15) with respect to Λ_{kl} to obtain

$$\epsilon_{alj}(\sigma_{kj}^J + h\delta_{jz}\Lambda_{kz}) + \epsilon_{arj}(\Lambda_{ir}K_{ijkl} + h\delta_{jz}\delta_{lz}\Lambda_{kr}) = 0, \tag{3.18a}$$

$$\epsilon_{aik}\sigma_{il}^J + \epsilon_{air}\Lambda_{rj}K_{ijkl} = 0. \tag{3.18b}$$

Since a can take on three different values and kl nine, these two equations represent $2 \times 3 \times 9 = 54$ independent equations that can be expressed as six sets of equation for arbitrary kl:

$$\Lambda_{yy}K_{yzkl} - \Lambda_{zz}K_{zykl} = -(\delta_{kz}\delta_{ly}\Lambda_{zz} + \delta_{ky}\delta_{lz}\Lambda_{yy})h + \Lambda_{xz}(K_{xykl} - \delta_{kx}\delta_{ly}h) \tag{3.19a}$$

$$\Lambda_{zz}K_{yzkl} - \Lambda_{yy}K_{zykl} = 0 \tag{3.19b}$$

$$\Lambda_{zz}K_{zxkl} - \Lambda_{xx}K_{xzkl} = \delta_{kx}\delta_{lz}(h\Lambda_{xx} - \sigma_{xx}^J) + \delta_{kz}\delta_{lx}\Lambda_{zz}h + \Lambda_{xz}(\delta_{kx}\delta_{lx}h - K_{xxkl}) \tag{3.19c}$$

$$\Lambda_{xx}K_{zxkl} - \Lambda_{zz}K_{xzkl} = \delta_{kz}\delta_{lx}\sigma_{xx}^J - \Lambda_{xz}K_{zzkl} \tag{3.19d}$$

$$\Lambda_{xx}K_{xykl} - \Lambda_{yy}K_{yxkl} = \delta_{kx}\delta_{ly}\sigma_{xx}^J \tag{3.19e}$$

$$\Lambda_{yy}K_{xykl} - \Lambda_{xx}K_{yxkl} = -\delta_{ky}\delta_{lx}\sigma_{xx}^J + \Lambda_{xz}K_{yzkl}. \tag{3.19f}$$

When $\Lambda_{xz} = 0$, these equations are easily solved to yield

$$\frac{K_{yzyz}}{\Lambda_{yy}^2} = \frac{K_{zyyz}}{\Lambda_{zz}\Lambda_{yy}} = \frac{K_{zyzy}}{\Lambda_{zz}^2} = \frac{h}{\Lambda_{zz}^2 - \Lambda_{yy}^2} \tag{3.20a}$$

$$\frac{K_{xzxz}}{\Lambda_{xx}^2} = \frac{K_{xzzx}}{\Lambda_{zz}\Lambda_{xx}} = \frac{h - \sigma_{xx}^J/\Lambda_{xx}}{\Lambda_{zz}^2 - \Lambda_{xx}^2} \tag{3.20b}$$

$$K_{zxzx} = \frac{\Lambda_{zz}^2 h - \Lambda_{xx}\sigma_{xx}^J}{\Lambda_{zz}^2 - \Lambda_{xx}^2} \tag{3.20c}$$

$$K_{xyxy} = K_{yxyx} = \frac{\Lambda_{xx}}{\Lambda_{yy}} K_{xyyx} = \frac{\Lambda_{xx}\sigma_{xx}^J}{\Lambda_{xx}^2 - \Lambda_{yy}^2}. \tag{3.20d}$$

From these equations and Eq. (2.20a), it follows that

$$K'_{yzyz} = K'_{yzzy} = K'_{zyzy} = C'_{yzyz} \tag{3.21a}$$

$$K'_{xyxy} = K'_{xyyx} = C'_{xyxy} \tag{3.21b}$$

$$K'_{yxyx} = \sigma'_{xx} + C'_{xyxy} \tag{3.21c}$$

$$K'_{xzxz} = K'_{xzzx} = C'_{xzxz} \tag{3.21d}$$

$$K'_{zxzx} = \sigma'_{xx} + C'_{xzxz}, \tag{3.21e}$$

where C'_{ijkl} is given in Eq. (3.11), and as before, $\sigma'_{xx} = \Lambda_{xx}^2\sigma_{xx}/\det \underline{\underline{\Lambda}}$. When $\Lambda_{xz} \neq 0$, Eqs. (3.19) are more tedious to solve, and we will not display their solution here. As required, their solution yields results identical to Eq. (2.20b) with C'_{ijkl} satisfying Eqs. (3.11) and (3.12).

IV. SIMPLE MODEL

The Ward identities discussed in the preceding section provide general expressions for elastic constants for arbitrary f_{iso} in terms of the deformation tensor $\underline{\underline{\Lambda}}$, h, σ_{xx} (or σ^{J}_{xx}) and the elastic constants C_{xxxx}, C_{xxzz}, and C_{zzzz}. In a typical experiment, h (or more generally f_{ani}) is fixed in the preparation process, and stretch $\bar{\Lambda}_{xx}$ relative to the equilibrium state with zero stress is a control parameter. A given f_{iso} gives rise to particular equations of state that determine σ_{xx} and the other components of $\underline{\underline{\Lambda}}$ in terms of h and Λ_{xx}. Thus once f_{iso} is given, the elastic tensor can be determined in terms of h and $\bar{\Lambda}_{xx}$.

Here, we explore a simple model, which we call the minimal model, whose properties we explored extensively in references [3,4], in which the free energy density, $f=\mathring{f}+f_{\text{comp}}$, breaks up into a part $\mathring{f}$ that depends only on the traceless part of $\underline{\underline{u}}$,

$$\mathring{u}_{ij} = u_{ij} - \tfrac{1}{3}\delta_{ij}u_{kk} \tag{4.1}$$

and a part,

$$f_{\text{comp}} = \tfrac{1}{2}B\phi^2, \tag{4.2}$$

that depends only on $\phi \equiv \mathrm{Tr}\,\underline{\underline{u}}$ and that enforces $\phi=0$ in the limit $B\to\infty$. The constraint $\phi=0$ is equivalent to the true incompressible constraint $\det\underline{\underline{\Lambda}}=1$ only in the small strain limit about the reference state. Using f_{comp} rather than an energy that enforces $\det\underline{\underline{\Lambda}}=1$ greatly simplifies some algebra because the energies for ϕ and $\mathring{u}$ decouple in f. For $\mathring{f}$, we use the de-Gennes-Maier-Saupe energy for a nematic in an external field (with the nematic order tensor replaced by the traceless part of $\underline{\underline{u}}$):

$$\mathring{f} = \tfrac{1}{2}r\,\mathrm{Tr}(\underline{\underline{\mathring{u}}})^2 - w\,\mathrm{Tr}(\underline{\underline{\mathring{u}}})^3 + v[\mathrm{Tr}(\underline{\underline{\mathring{u}}})^2]^2 - h\mathring{u}_{zz}. \tag{4.3}$$

As long as boundary conditions do not prevent ϕ from relaxing to zero, physical properties will depend only on $\mathring{f}$ and $\underline{\underline{\mathring{u}}}$, but, as we show in the appendix, it is instructive to consider elastic tensors for finite B and to explore how they behave as $B\to\infty$.

To simplify algebra, it is convenient to rescale $\underline{\underline{\mathring{u}}}$ and coefficients in $\mathring{f}$ to set the coefficients of the third- and fourth-order terms in $\underline{\underline{\mathring{u}}}$ to unity via $\underline{\underline{\mathring{u}}}=(w/v)\underline{\underline{\tilde{u}}}$, $r=(w^2/v)\tilde{r}$, $h=(w^3/v^2)\tilde{h}$, and $\mathring{f}=(w^4/v^3)\tilde{f}$:

$$\tilde{f} = \tfrac{1}{2}\tilde{r}\,\mathrm{Tr}\,\underline{\underline{\tilde{u}}}^2 - \mathrm{Tr}\,\underline{\underline{\tilde{u}}}^3 + (\mathrm{Tr}\,\underline{\underline{\tilde{u}}}^2)^2 - \tilde{h}\tilde{u}_{zz}. \tag{4.4}$$

$\underline{\underline{\tilde{u}}}$ and $\underline{\underline{\mathring{u}}}$ have five rather than six independent components. Here we use the parametrization

$$\underline{\underline{\tilde{u}}} = \frac{v}{w}\underline{\underline{\mathring{u}}} = \begin{pmatrix} \tilde{u}_{xx} & \tilde{u}_{xy} & \tilde{u}_{xz} \\ \tilde{u}_{xy} & -\tilde{u}_{xx}-\tilde{u}_{zz} & \tilde{u}_{yz} \\ \tilde{u}_{xz} & \tilde{u}_{yz} & \tilde{u}_{zz} \end{pmatrix}, \tag{4.5}$$

in which $\mathring{u}_{xx}$ and $\mathring{u}_{zz}$ are treated as independent variables and $\mathring{u}_{yy}=-\mathring{u}_{xx}-\mathring{u}_{zz}$ is a dependent variable. In this representation, the independent components of $\mathring{u}_{ij}$ are identical to those of u_{ij} when $\phi=0$, i.e., when $i\neq j$, $u_{ij}=\mathring{u}_{ij}$, $u_{xx}=\mathring{u}_{xx}$, $u_{zz}=\mathring{u}_{zz}$, and $u_{yy}=\mathring{u}_{yy}=-u_{xx}-u_{zz}$. On the other hand $\partial\mathring{f}/\partial\mathring{u}_{xx}\neq\partial\mathring{f}/\partial u_{xx}$ and similarly for $\partial\mathring{f}/\partial\mathring{u}_{zz}$. This parametrization is appropriate to both the parallel and perpendicular geometries of Fig. 2 when the $xyxy$, $zyzy$, $yzyz$, and $xzxz$ components of the elastic modulus tensor are measured. In the parallel geometry, xy and zy strains couple to each other but not to other strains, and in particular not to strains that change $\mathrm{Tr}\,\underline{\underline{u}}$. Thus, the $xyxy$, $zyzy$, and $xyzy$ components of the elastic modulus tensor do not depend on whether xx, yy, and zz components of stress or strain are fixed (recall that $\Lambda'_{xz}=\Lambda'_{zx}=0$). However, as we shall see, Λ'_{xy} and Λ'_{zy} are coupled so that K'_{xyxy} differs from K^{σ}_{xyxy} in which σ^{J}_{zy} is fixed at zero, and K'_{zyzy} differs from K^{σ}_{zyzy} in which σ^{J}_{xy} is fixed at zero. In the perpendicular geometry, the condition $\sigma^{J}_{yy}=0$ allows u_{yy} to relax to its $\mathrm{Tr}\,\underline{\underline{u}}=0$ equilibrium of $u_{yy}=-u_{xx}-u_{zz}$.

Equilibrium in the presence of a nonvanishing stress σ^{J}_{xx} is determined by the following equations of state:

$$\frac{\partial\tilde{f}}{\partial\tilde{u}_{zz}} = 8(\tilde{u}_{xx}+2\tilde{u}_{zz})(\tilde{u}_{xx}^2+\tilde{u}_{xx}\tilde{u}_{zz}+\tilde{u}_{zz}^2+\tilde{u}_{xz}^2)+(\tilde{u}_{xx}+2\tilde{u}_{zz})(r + 3\tilde{u}_{xx}) - \tilde{h} - 3\tilde{u}_{xz}^2 = 0, \tag{4.6a}$$

$$\frac{\partial\tilde{f}}{\partial\tilde{u}_{xx}} = 8(\tilde{u}_{zz}+2\tilde{u}_{xx})(\tilde{u}_{xx}^2+\tilde{u}_{xx}\tilde{u}_{zz}+\tilde{u}_{zz}^2+\tilde{u}_{xz}^2)+(\tilde{u}_{zz}+2\tilde{u}_{xx})(r + 3\tilde{u}_{zz}) - 3\tilde{u}_{xz}^2 = \tilde{\sigma}_{xx}, \tag{4.6b}$$

$$\frac{\partial\tilde{f}}{\partial\tilde{u}_{xz}} = 16\tilde{u}_{xz}(\tilde{u}_{xx}^2+\tilde{u}_{xx}\tilde{u}_{zz}+\tilde{u}_{zz}^2+\tilde{u}_{xz}^2)+2\tilde{u}_{xz}(r-3\tilde{u}_{xx}-3\tilde{u}_{zz}) = 0, \tag{4.6c}$$

where $\tilde{\sigma}_{xx}=v^2/w^3\cdot\sigma_{xx}=v^2/w^3\cdot\sigma^{J}_{xx}/\Lambda_{xx}$ and the equilibrium values of $\tilde{u}_{xy}$ and $\tilde{u}_{yz}$ are zero as discussed in the preceding section. If $h\neq\sigma_{xx}$, $\tilde{u}_{xz}=0$ and Eq. (4.6a) with $\tilde{u}_{xz}=0$ determines $\tilde{u}_{zz}$ (and thus $\tilde{u}_{yy}=-\tilde{u}_{xx}-\tilde{u}_{xx}$) as a function of $\tilde{u}_{xx}$. When $h=\sigma_{xx}$, $\tilde{u}_{xz}$ is determined by Eq. (4.6c)

$$\tilde{r} - 3(\tilde{u}_{xx}+\tilde{u}_{zz}) + 8(\tilde{u}_{xx}^2+\tilde{u}_{xx}\tilde{u}_{zz}+\tilde{u}_{zz}^2) = -8\tilde{u}_{xz}^2, \tag{4.7}$$

which when used in either Eq. (4.6a) and (4.6b) yields

$$\tilde{r} - 3(\tilde{u}_{xx}+\tilde{u}_{zz}) + 24(\tilde{u}_{xx}+\tilde{u}_{zz})^2 = \tfrac{8}{3}\tilde{h}. \tag{4.8}$$

These equations imply

$$\tilde{u}_{xx}+\tilde{u}_{zz} = -\tilde{u}_{yy} = a_0 = \frac{1}{16}\left(1+\sqrt{1+\frac{256}{9}\tilde{h}-\frac{32}{3}\tilde{r}}\right) \tag{4.9}$$

and that

$$\tilde{u}_{xz}^2=(\tilde{u}_+-\tilde{u}_{xx})(\tilde{u}_{xx}-\tilde{u}_-), \tag{4.10}$$

where

$$\tilde{u}_\pm=\frac{a_0}{2}\pm\sqrt{\frac{9}{4}a_0^2-\frac{\tilde{h}}{3}}. \tag{4.11}$$

Thus, $\tilde{u}_{xz}$ is nonzero only for $\tilde{u}_{xx}$ between $\tilde{u}_-$ and $\tilde{u}_+$, which, respectively, mark the beginning and the end of the stress plateau. Equation (4.11) implies that $\tilde{u}_+ + \tilde{u}_- = a_0$ and consequently we have

$$\tilde{u}_{zz}=\tilde{u}_+ + \tilde{u}_- - \tilde{u}_{xx}. \tag{4.12}$$

Observe that $\tilde{u}_{xx}$ and $\tilde{u}_{zz}$ exchange values at the two ends of the stress plateau: at the beginning of the plateau, $\tilde{u}_{xx}=\tilde{u}_-$ and $\tilde{u}_{zz}=\tilde{u}_+$ whereas at the end of the plateau, $\tilde{u}_{xx}=\tilde{u}_+$ and $\tilde{u}_{zz}=\tilde{u}_-$.

Deviations of $\tilde{f}$ from these equilibrium conditions are determined by the "elastic" constant tensor $\tilde{C}_{ijkl}$. The relation between $\tilde{C}_{ijkl}$ and $\partial^2\tilde{f}/\partial\tilde{u}_\alpha\partial\tilde{u}_\beta$ is given in Eq. (2.9), where ij and kl (or α and β) run over the five independent subscripts, $xx=1$, $zz=3$, $xz=4$, $yz=5$, and $xy=6$. The nonvanishing components of $\tilde{C}_{ijkl}$ are

$$\tilde{C}_{xxxx}=2[\tilde{r}+3\tilde{u}_{zz}+12(\tilde{u}_{zz}^2+2\tilde{u}_{xx}\tilde{u}_{zz}+2\tilde{u}_{xx}^2)+8\tilde{u}_{xz}^2],$$

$$\tilde{C}_{zzzz}=2[\tilde{r}+3\tilde{u}_{xx}+12(\tilde{u}_{xx}^2+2\tilde{u}_{xx}\tilde{u}_{zz}+2\tilde{u}_{zz}^2)+8\tilde{u}_{xz}^2],$$

$$\tilde{C}_{xzxz}=\tfrac{1}{2}[\tilde{r}-3(\tilde{u}_{xx}+\tilde{u}_{zz})+8(\tilde{u}_{xx}^2+\tilde{u}_{xx}\tilde{u}_{zz}+\tilde{u}_{zz}^2)+24\tilde{u}_{xz}^2],$$

$$\tilde{C}_{xxzz}=\tilde{r}+6[\tilde{u}_{xx}+\tilde{u}_{zz}+4(\tilde{u}_{xx}+\tilde{u}_{zz})^2]+8\tilde{u}_{xz}^2,$$

$$\tilde{C}_{xxxz}=\tilde{u}_{xz}[8(\tilde{u}_{zz}+2\tilde{u}_{xx})-3],$$

$$\tilde{C}_{zzxz}=\tilde{u}_{xz}[8(\tilde{u}_{xx}+2\tilde{u}_{zz})-3],$$

$$\tilde{C}_{xyxy}=\tfrac{1}{2}[\tilde{r}+3\tilde{u}_{zz}+8(\tilde{u}_{xx}^2+\tilde{u}_{xx}\tilde{u}_{zz}+\tilde{u}_{zz}^2)+8\tilde{u}_{xz}^2],$$

$$\tilde{C}_{yzyz}=\tfrac{1}{2}[\tilde{r}+3\tilde{u}_{xx}+8(\tilde{u}_{xx}^2+\tilde{u}_{xx}\tilde{u}_{zz}+\tilde{u}_{zz}^2)+8\tilde{u}_{xz}^2],$$

$$\tilde{C}_{xyyz}=-\tfrac{3}{2}\tilde{u}_{xz}, \tag{4.13}$$

and those related trivially to these by symmetry. Using the equilibrium state equations given in the preceding paragraph, we can easily show that on the stress plateau $\tilde{C}_{xyxy}$, $\tilde{C}_{yzyz}$, and $\tilde{C}_{xzxz}$ are simplified to

$$\tilde{C}_{xyxy}=\tfrac{3}{2}\tilde{u}_{xx}+3\tilde{u}_{zz}=\tfrac{3}{2}(2a_0-\tilde{u}_{xx}), \tag{4.14a}$$

$$\tilde{C}_{yzyz}=\tfrac{3}{2}\tilde{u}_{zz}+3\tilde{u}_{xx}=\tfrac{3}{2}(a_0+\tilde{u}_{xx}), \tag{4.14b}$$

$$\tilde{C}_{xzxz}=8\tilde{u}_{xz}^2=8(\tilde{u}_+-\tilde{u}_{xx})(\tilde{u}_{xx}-\tilde{u}_-). \tag{4.14c}$$

These three equations imply that $\tilde{C}_{xyxy}$ decreases monotonically and $\tilde{C}_{yzyz}$ rises monotonically as $\tilde{u}_{xx}$ increases from $\tilde{u}_-$ to $\tilde{u}_+$ and that $\tilde{C}_{xzxz}$ vanishes at $\tilde{u}_{xx}=\tilde{u}_\pm$ and reaches a maximum in the vicinity of $\tilde{u}_{xx}=(\tilde{u}_+ + \tilde{u}_-)/2$. It is a straightforward exercise to verify that $\tilde{C}_{ijkl}$ obeys the Ward-identity relations, Eq. (3.10), on the stress plateau. Returning to the nonrescaled unit, we have

$$\mathring{C}_{ijkl}=\frac{\partial^2\mathring{f}}{\partial\mathring{u}_{ij}\,\partial\mathring{u}_{kl}}=\frac{w^2}{v}\tilde{C}_{ijkl}, \tag{4.15}$$

where it is understood that ij and kl are not equal to yy.

The relation between the elastic constant tensor $\mathring{C}'_{ijkl}$ with respect to the reference space S'_R of points $\mathbf{x}'$ and $\mathring{C}_{ijkl}$ is the same as that [Eq. (2.14)] between C'_{ijkl} and C_{ijkl}. Of course $\mathring{C}'_{ijkl}$ can have components with ij or kl or both equal to yy even though $\mathring{C}_{ijkl}$ does not. To obtain $\mathring{K}'_{ijk}$, we treat Λ_{yy} as a dependent variable determined by Tr $\underline{\underline{\mathring{u}}}=0$. So long as there are no xy or yz components to $\mathring{\sigma}_{ij}=\partial\mathring{f}/\partial\mathring{u}_{ij}$, it is straightforward to show that

$$\mathring{K}'_{ijk}=\delta_{ik}\mathring{\sigma}'_{jl}+\mathring{C}'_{ijkl}. \tag{4.16}$$

Except for the trivial yy components, the nonvanishing components of $\underline{\underline{\mathring{C}}}'$ are the same as those of $\underline{\underline{\mathring{C}}}$. In the modified Voigt notation, with $xx=1$, $zz=3$, $xz=4$, $yz=5$, and $xy=6$,

$$\underline{\underline{\mathring{C}}}'=\begin{pmatrix}\mathring{C}'_{11} & \mathring{C}'_{13} & \mathring{C}'_{14} & 0 & 0\\ \mathring{C}'_{13} & \mathring{C}'_{33} & \mathring{C}'_{34} & 0 & 0\\ \mathring{C}'_{14} & \mathring{C}'_{34} & \mathring{C}'_{44} & 0 & 0\\ 0 & 0 & 0 & \mathring{C}'_{55} & \mathring{C}'_{56}\\ 0 & 0 & 0 & \mathring{C}'_{56} & \mathring{C}'_{66}\end{pmatrix}, \tag{4.17}$$

where $\mathring{C}'_{\alpha\beta}=\mathring{D}'_{\alpha\beta}/\det\underline{\underline{\Lambda}}$ and

$$\mathring{D}'_{11}=\Lambda_{xx}^4\mathring{C}_{xxxx}+\Lambda_{xz}^4\mathring{C}_{zzzz}+\Lambda_{xx}^2\Lambda_{xz}^2(4\mathring{C}_{xzxz}+2\mathring{C}_{xxzz}) + 4(\Lambda_{xx}^3\Lambda_{xz}\mathring{C}_{xxxz}+\Lambda_{xz}^3\Lambda_{xx}\mathring{C}_{zzxz}), \tag{4.18a}$$

$$\mathring{D}'_{13}=\Lambda_{zz}^4\mathring{C}_{zzzz}+\Lambda_{xx}^2\Lambda_{zz}^2\mathring{C}_{xxzz}+2\Lambda_{xx}\Lambda_{xz}\Lambda_{zz}^2\mathring{C}_{zzxz}, \tag{4.18b}$$

$$\mathring{D}'_{14}=\Lambda_{xx}^3\Lambda_{zz}\mathring{C}_{xxxz}+\Lambda_{xx}^2\Lambda_{xz}\Lambda_{zz}(2\mathring{C}_{xzxz}+\mathring{C}_{xxzz}) + 3\Lambda_{xx}\Lambda_{xz}^2\Lambda_{zz}\mathring{C}_{zzxz}+\Lambda_{xz}^3\Lambda_{zz}\mathring{C}_{zzzz}, \tag{4.18c}$$

$$\mathring{D}'_{33}=\Lambda_{zz}^4\mathring{C}_{zzzz}, \tag{4.18d}$$

$$\mathring{D}'_{34}=\Lambda_{xx}\Lambda_{zz}^3\mathring{C}_{zzxz}+\Lambda_{xz}\Lambda_{zz}^3\mathring{C}_{zzzz}, \tag{4.18e}$$

$$\mathring{D}'_{44}=\Lambda_{xx}^2\Lambda_{zz}^2\mathring{C}_{xzxz}+\Lambda_{xz}^2\Lambda_{zz}^2\mathring{C}_{zzzz}+2\Lambda_{xx}\Lambda_{xz}\Lambda_{zz}^2\mathring{C}_{zzxz}, \tag{4.18f}$$

$$\mathring{D}'_{55}=\Lambda_{yy}^2\Lambda_{zz}^2\mathring{C}_{yzyz}, \tag{4.18g}$$

$$\mathring{D}'_{56}=\Lambda_{xz}\Lambda_{yy}^2\Lambda_{zz}\mathring{C}_{yzyz}+\Lambda_{xx}\Lambda_{yy}^2\Lambda_{zz}\mathring{C}_{yzxy}, \tag{4.18h}$$

$$\mathring{D}'_{66}=\Lambda_{xx}^2\Lambda_{yy}^2\mathring{C}_{xyxy}+\Lambda_{xz}^2\Lambda_{yy}^2\mathring{C}_{yzyz}+2\Lambda_{xx}\Lambda_{xz}\Lambda_{yy}^2\mathring{C}_{yzxy}. \tag{4.18i}$$

From the expression for $\mathring{C}'_{44}$ (or $\mathring{C}'_{xzxz}$), we can see easily that it vanishes at the boundaries of the stress plateau as $\mathring{C}_{xzxz}$ does. As discussed in Eq. (2.22), when σ'_{xx} is the only nonzero equilibrium stress component, the only components of K'_{ijkl} that differ from $\mathring{C}'_{ijkl}$ are $\mathring{K}'_{xxxx}=\sigma'_{xx}+\mathring{C}'_{xxxx}$, $\mathring{K}'_{yxyx}=\sigma'_{xx}+\mathring{C}'_{xyxy}$, and $\mathring{K}'_{zxzx}=\sigma'_{xx}+\mathring{C}'_{xzxz}$, where $\sigma'_{xx}=(w^3/v^2)\tilde{\sigma}_{xx}\Lambda_{xx}^2/\det\underline{\underline{\Lambda}}$; for all the other components, $\mathring{K}'_{ijkl}=\mathring{C}'_{ijkl}$.

We now have all of the information we need to calculate $\mathring{K}'_{ijkl}$ (or $\mathring{C}'_{ijkl}$) as a function of $\bar{\Lambda}_{xx}=\Lambda_{xx}/\Lambda_{xx}^r$, where Λ_{xx}^r is the value of Λ_{xx} at $\sigma_{xx}=0$. Equations (4.12) and (4.10), along with the relations $\Lambda_{xx}^2=1+2(w/v)\tilde{u}_{xx}$, $\Lambda_{yy}^2=1+2(w/v)\tilde{u}_{yy}=1-2(w/v)(\tilde{u}_{xx}+\tilde{u}_{zz})$, $\Lambda_{zz}^2+\Lambda_{xz}^2=1+2(w/v)\tilde{u}_{zz}$, and $\Lambda_{xx}\Lambda_{xz}=2(w/v)\tilde{u}_{xz}$, allow us to express Λ_{zz}^2, Λ_{yy}^2, and Λ_{xz}^2 as a function of Λ_{xx} and thus of $\bar{\Lambda}_{xx}=\Lambda_{xx}/\Lambda_{xx}^r$:

$$\Lambda_{zz}^2=\Lambda_+^2+\Lambda_-^2-\Lambda_{xx}^2-\Lambda_{xz}^2(\Lambda_{xx}) \tag{4.19}$$

$$\Lambda_{xz}^2(\Lambda_{xx})=\frac{1}{\Lambda_{xx}^2}(\Lambda_+^2-\Lambda_{xx}^2)(\Lambda_{xx}^2-\Lambda_-^2), \tag{4.20}$$

where $\Lambda_\pm^2=1+2(w/v)\tilde{u}_\pm$.

We consider the parallel and perpendicular geometries separately. In the parallel geometry, boundary conditions preventing rotations of the sample and in-plane shears require $\Lambda'_{xz}=\Lambda'_{zx}=\Lambda'_{yz}=\Lambda'_{yx}=0$. Thus, the only components of $\mathring{K}'_{ijkl}$ relevant to this geometry are those with ij or kl equal to $xx\equiv 1$, $zz\equiv 3$, $zy\equiv 5$, and $xy\equiv 6$, where the numbers are the standard Voigt notation for strain components. Thus in the parallel geometry, $\underline{\underline{\mathring{K}}}'$ takes the form

$$\underline{\underline{\mathring{K}}}^{\parallel\prime}=\begin{pmatrix}\sigma'_{xx}+\mathring{C}'_{11} & \mathring{C}'_{13} & 0 & 0\\ \mathring{C}'_{13} & \mathring{C}'_{33} & 0 & 0\\ 0 & 0 & \mathring{C}'_{55} & \mathring{C}'_{56}\\ 0 & 0 & \mathring{C}'_{56} & \mathring{C}'_{66}.\end{pmatrix}. \tag{4.21}$$

Note that the shear components $\mathring{K}'_{55}$, $\mathring{K}'_{66}$, and $\mathring{K}'_{56}$ do not couple to the components involving compressions along the x, and z directions. As we show in the appendix, this result extends to the full K'_{ijkl} in which it is not assumed, as is the case here, that Λ_{yy} is always allowed to relax to satisfy the constraint $u_{ii}=0$. Thus, in this geometry, the shear moduli do not depend on whether Λ'_{xx}, Λ'_{yy}, Λ'_{zz}, or some combination of them is allowed to relax to satisfy the constraint $u_{ii}=0$. The constant displacement elastic moduli, plotted in Fig. 3, are simply

$$K'_{zyzy}=\mathring{C}'_{55},\quad K'_{xyxy}=\mathring{C}_{66}. \tag{4.22}$$

If on the other hand, the stresses rather than deformations are fixed, the elastic moduli become

$$\mathring{K}^{\sigma}_{zyzy}=\mathring{C}'_{55}-\frac{(\mathring{C}'_{56})^2}{\mathring{C}'_{66}}, \tag{4.23a}$$

$$\mathring{K}^{\sigma}_{xyxy}=\mathring{C}'_{66}-\frac{(\mathring{C}'_{56})^2}{\mathring{C}'_{55}}, \tag{4.23b}$$

and differ from their constant strain counterparts. These moduli are plotted in Fig. 4.

In the perpendicular geometry, $\Lambda'_{xy}=\Lambda'_{yx}=\Lambda'_{zy}=\Lambda'_{zx}=0$, and the only relevant components of $\mathring{K}'_{ijkl}$ are those with ij and kl equal to $xx\equiv 1$, $zz\equiv 3$, $xz\equiv 4$, and $yz\equiv 5$. Equation (4.17) then implies

$$\underline{\underline{\mathring{K}}}'^{\perp}=\begin{pmatrix}\sigma'_{xx}+\mathring{C}'_{11} & \mathring{C}'_{13} & \mathring{C}'_{14} & 0\\ \mathring{C}'_{13} & \mathring{C}'_{33} & \mathring{C}'_{34} & 0\\ \mathring{C}'_{14} & \mathring{C}'_{34} & \mathring{C}'_{44} & 0\\ 0 & 0 & 0 & \mathring{C}'_{55}\end{pmatrix}\equiv\underline{\underline{\mathring{K}}}'^{\perp}_{\parallel}\otimes\mathring{C}'_{55}. \tag{4.24}$$

Thus if Λ'_{xx} and Λ'_{zz} are fixed at one, with Λ'_{yy} left free to adjust, we find

$$K^{y\sigma}_{xzxz}=\mathring{C}'_{44},\quad K^{y\sigma}_{yzyz}=K'_{yzyz}=\mathring{C}'_{55}. \tag{4.25}$$

These moduli are plotted in Fig. 3. Other stresses rather than strains could also be fixed. If the stress σ^I_{xx}, σ^I_{yy} and σ^I_{zz} are fixed, the $xzxz$ component of the constant-stress elastic tensor at $\sigma_{yz}=0$ in the $B\to\infty$ limit is then

$$K^{\sigma}_{xzxz}=\frac{\det\underline{\underline{\mathring{K}}}'^{\perp}_{\parallel}}{\mathring{C}'_{33}(\sigma'_{xx}+\mathring{C}'_{11})-(\mathring{C}'_{13})^2}. \tag{4.26}$$

Because the modulus K'_{yzyz} does not couple to any others, its constant-stress counterpart K^{σ}_{yzyz} is thus identical to K'_{yzyz}. K^{σ}_{xzxz}, and K^{σ}_{yzyz} are plotted in Fig. 4.

The elastic matrix C_{ijkl} for finite B can be obtained from $\mathring{C}_{ijkl}$ as outlined in the appendix. The result is that C'_{ijkl} is a 6×6 block-diagonal matrix in the Voigt notation with a 4×4 block coupling indices 1 to 4 and a 2×2 block coupling indices 5 and 6. In the parallel geometry, $\underline{\underline{K}}'^{\parallel}$ becomes a 5×5 matrix with a 3×3 $xx-yy-zz$ block and a 2×2 $xy-zy$ block. In the perpendicular geometry, $\underline{\underline{K}}'^{\perp}$ becomes a 5×5 matrix with a 4×4 $xx-yy-zz-xz$ block and a 1×1 yz block. In the appendix, we outline how the finite B matrices reduce to those calculated here in the $B\to\infty$ limit.

The moduli $\mathring{K}'_{zxzx}=\mathring{\sigma}'_{xx}+\mathring{C}'_{xzxz}$ and $\mathring{K}'_{yxyx}=\sigma'_{xx}+\mathring{C}'_{xyxy}$ measure response to rotations of the plane of the film that are resisted by the stress σ^I_{xx}. They are plotted in Fig. 5.

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APPENDIX: THE MINIMAL MODEL AT FINITE COMPRESSIBILITY

It is instructive to investigate the minimal model when B is not infinite. In this case, we cannot impose the constraint Tr $\underline{\underline{u}}=0$ from the beginning. Differentiating $f=\mathring{f}+\frac{1}{2}B\phi^2$ with respect to u_{ij} twice yields the elastic tensor C_{ijkl}, which consists of two parts

$$C_{ijkl}=\overset{\circ\circ}{C}_{ijkl}+B\delta_{ij}\delta_{kl}, \tag{A1}$$

where $\overset{\circ\circ}{C}_{ijkl}=\partial^2\mathring{f}/\partial u_{ij}\partial u_{kl}$ is traceless with respect to contractions of i with j and of k with l. Using $\mathring{u}_{xx}=\frac{2}{3}u_{xx}-\frac{1}{3}(u_{yy}+u_{zz})$, $\mathring{u}_{yy}=\frac{2}{3}u_{yy}-\frac{1}{3}(u_{xx}+u_{zz})$, and $\mathring{u}_{zz}=\frac{2}{3}u_{zz}-\frac{1}{3}(u_{xx}+u_{yy})$, it is straightforward to express $\overset{\circ\circ}{C}_{ijkl}$ in terms of $\mathring{C}_{ijkl}$, which is defined as $\partial^2\mathring{f}/\partial\mathring{u}_{ij}\partial\mathring{u}_{kl}$ with ij and kl running over the five independent subscripts, xx, zz, xz, yz, and xy, and with $\mathring{u}_{yy}$ treated as a variable dependent on $\mathring{u}_{xx}$ and $\mathring{u}_{zz}$,

$$\overset{\circ\circ}{C}_{ijkl}=\mathring{C}_{ijkl}\quad ij,kl=xy,xz,yz \tag{A2a}$$

$$\overset{\circ\circ}{C}_{ijxx}=\tfrac{1}{3}(2\mathring{C}_{ijxx}-\mathring{C}_{ijzz})\quad ij=xy,xz,yz \tag{A2b}$$

$$\overset{\circ\circ}{C}_{ijyy}=-\tfrac{1}{3}(\mathring{C}_{ijxx}+\mathring{C}_{ijzz})\quad ij=xy,xz,yz \tag{A2c}$$

$$\overset{\circ\circ}{C}_{ijzz}=\tfrac{1}{3}(2\mathring{C}_{ijzz}-\mathring{C}_{ijxx})\quad ij=xy,xz,yz \tag{A2d}$$

$$\overset{\circ\circ}{C}_{zzzz}=\tfrac{1}{9}(4\mathring{C}_{zzzz}-4\mathring{C}_{xxzz}+\mathring{C}_{xxxx}) \tag{A2e}$$

$$\overset{\circ\circ}{C}_{xxxx}=\tfrac{1}{9}(4\mathring{C}_{xxxx}-4\mathring{C}_{xxzz}+\mathring{C}_{zzzz}) \tag{A2f}$$

$$\overset{\circ\circ}{C}_{yyyy}=\tfrac{1}{9}(\mathring{C}_{zzzz}+2\mathring{C}_{xxzz}+\mathring{C}_{xxxx}) \tag{A2g}$$

$$\overset{\circ\circ}{C}_{xxzz}=-\tfrac{1}{9}(2\mathring{C}_{zzzz}-5\mathring{C}_{xxzz}+2\mathring{C}_{xxxx}) \tag{A2h}$$

$$\overset{\circ\circ}{C}_{yyzz}=-\tfrac{1}{9}(2\mathring{C}_{zzzz}+\mathring{C}_{xxzz}-\mathring{C}_{xxxx}) \tag{A2i}$$

$$\overset{\circ\circ}{C}_{xxyy}=-\tfrac{1}{9}(2\mathring{C}_{xxxx}+\mathring{C}_{xxzz}-\mathring{C}_{zzzz}). \tag{A2j}$$

As required, $\overset{\circ\circ}{C}_{ijkl}$ satisfies the traceless constraints. Then transforming to the new equilibrium state, we have

$$C'_{ijkl}=\overset{\circ\circ}{C}'_{ijkl}+B\Lambda_{ip}\Lambda^T_{pj}\Lambda_{kr}\Lambda^T_{rl}. \tag{A3}$$

On the plateau where Λ_{xz} is nonzero and the other off-diagonal components are zero

$$C'_{xzxz}=\overset{\circ\circ}{C}'_{xzxz}+B(\Lambda_{xz}\Lambda_{zz})^2. \tag{A4}$$

Thus, even though C'_{xzxz} vanishes at the plateau boundaries, it becomes arbitrarily large on the plateau when B diverges. This is because the $(u'_{xz})^2$ term also contributes to the value of Tr $\underline{\underline{u}}$ at an equilibrium state with nonzero Λ_{xz}. On the other hand $K^{y\sigma}_{xzxz}$ and K^{σ}_{xzxz} are well behaved as a function of B and approach the limits calculated in Sec. IV in the $B\to\infty$ limit as can be seen by direct evaluation.

The anomaly of a diverging C'_{xzxz} does not occur if $\frac{1}{2}B\phi^2$ is replaced by a true volume compression energy

$$f_{\mathrm{comp}}=\frac{1}{2}B\left(\frac{V}{V_0}-1\right)^2=\frac{1}{2}B(\psi-1)^2, \tag{A5}$$

where $\psi=\det\underline{\underline{\Lambda}}=\det^{1/2}\underline{\underline{g}}$ with $\underline{\underline{g}}=\underline{\underline{\Lambda}}^T\cdot\underline{\underline{\Lambda}}$. The equation of state then becomes,

$$\frac{\partial\mathring{f}}{\partial u_{ij}}+B(\psi-1)\psi g^{-1}_{ij}=\sigma_{ij}, \tag{A6}$$

and with the aid of this equation of state, the elastic tensor becomes,

$$C'_{ijkl}=\overset{\circ\circ}{C}'_{ijkl}-B\psi(\psi-1)(\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk})+B(2\psi-1)\psi\delta_{ij}\delta_{kl}. \tag{A7}$$

The equation of state [Eq. (A6)] ensures that $B\psi(\psi-1)$ is of the order of the elastic constants that appear in $\mathring{f}$ rather than B and thus that the only deformations that cost the large compression energy measured by B are those with a nonvanishing Tr $\underline{\underline{u}}$ as expected, and C'_{xzxz} is perfectly finite in the limit $B\to\infty$. On the other hand, it is still true that because K'_{aaxz} for $a=x,y,z$ are nonzero, $K^{\sigma}_{xzxz}\neq K'_{xzxz}$.

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[26] The energy associated with the application of a stress in the direction i in the target space across a surface along direction j in the reference space is $\Delta F^{\text{ext}}=\int d^d x \sigma^I_{ij}\Lambda_{ij}$, where $\sigma^I_{ij}=\partial f/\partial\Lambda_{ij}$ is the first Piola-Kirchhoff stress tensor. σ^I_{ij} is in general not symmetric. External stresses can also be described by the Cauchy stress tensor σ^C_{ij} giving the force in direction i across a surface with normal along direction j in the target space. $\sigma^C_{ij}=(\det \underline{\underline{\Lambda}})^{-1}\Lambda_{ik}\sigma_{kl}\Lambda^T_{lj}$, where σ_{ij} is the second PK stress tensor of Eq. (2.4), is symmetric. The energy associated with the application of σ^C_{ij} is $\Delta F^{\text{ext}}=\int d^d R \sigma^C_{ij} u^C_{ij}$, where $u^C_{ij}=(1/2)(\partial u_i/\partial R_j+\partial u_j/\partial R_i)$. The strain u^C_{ij} is symmetric in ij because σ^C_{ij} is. To linear order in u_i, it is identical to the Eulerian strain variable, but it lacks the nonlinear term $-(1/2)(\partial u_k/\partial R_i)(\partial u_k/\partial R_j)$ required for rotational invariance. It is the absence of this nonlinear term in ΔF^{ext} that encodes its preferred direction in the target space and that ultimately distinguishes it from the energy of Eq. (3.1), which is linear in the strain u_{zz} that is rotationally invariant in target space. For more details, see Ref. [4]. A minus sign appears in Eq. (3.1) because it is in reality contribution to the Legendre transformed free energy, which after putting u_{zz} equal to its equilibrium value, depends on h and not u_{zz}.