Supplementary Information for "Normal stress anisotropy and marginal stability in athermal elastic networks"

Jordan Shivers,^{1,2} Jingchen Feng,² Abhinav Sharma,³ and F. C. MacKintosh^{1,2,4}

¹Department of Chemical and Biomolecular Engineering, Rice University, Houston, TX 77005, USA

²Center for Theoretical Biological Physics, Rice University, Houston, TX 77030, USA

³Leibniz-Institut für Polymerforschung Dresden, 01069 Dresden, Germany

⁴Departments of Chemistry and Physics & Astronomy, Rice University, Houston, TX 77005, USA

Affinely deforming isotropic network models

Consider a filament segment with initial orientation $\hat{\mathbf{n}}$, undergoing deformation described by the tensor $\mathbf{\Lambda}(\gamma)$. The deformation changes the filament's length and orientation, resulting in a tension τ directed along its new orientation. Treating the filament as a linear elastic element with stretching modulus μ and initial length $l_0 = 1$, the tension vector is $\boldsymbol{\tau} = \tau \mathbf{\Lambda} \hat{\mathbf{n}} / |\mathbf{\Lambda} \hat{\mathbf{n}}|$. For the spring model, we utilize a linear Hookean force-extension relation in which the filaments support both tension and compression: $\tau = \tau_s$ where $\tau_s = \mu(|\mathbf{\Lambda} \hat{\mathbf{n}}| - 1)$. For the rope model, we instead use a one-sided force-extension relation that is only finite under tension: $\tau = \tau_r$, with

$$\tau_r = \begin{cases} \mu(|\mathbf{A}\hat{\mathbf{n}}| - 1) & (|\mathbf{A}\hat{\mathbf{n}}| > 1) \\ 0 & (|\mathbf{A}\hat{\mathbf{n}}| \le 1) \end{cases}.$$
(1)

Taking the average, over all initial filament orientations, of the product of the *i* component of the tension, $\tau \Lambda_{il} n_l / |\mathbf{A}\hat{\mathbf{n}}|$, and the line density of filaments crossing the *j* plane after the deformation, $\frac{\rho}{\det \Lambda} \Lambda_{jk} n_k$, yields the stress tensor [1, 2],

$$\sigma_{ij} = \frac{\rho}{\det \mathbf{\Lambda}} \Big\langle \tau \frac{\Lambda_{il} n_l \Lambda_{jk} n_k}{|\mathbf{\Lambda} \hat{\mathbf{n}}|} \Big\rangle. \tag{2}$$

Since we consider only volume-conserving simple shear, det $\Lambda = 1$. Thus, for filaments in 3D with initial polar angle θ and azimuthal angle φ , the stress tensor is

$$\sigma_{ij} = \frac{\rho}{4\pi} \int_{\varphi} \int_{\theta} d\theta d\varphi \sin \theta \left[\tau \frac{\Lambda_{il} n_l \Lambda_{jk} n_k}{|\Lambda \hat{\mathbf{n}}|} \right], \tag{3}$$

in which the deformation tensor for simple shear in 3D is

$$\mathbf{\Lambda}(\gamma) = \begin{pmatrix} 1 & 0 & \gamma \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}$$
(4)

and the transformed orientation vector is

$$\mathbf{\Lambda}\mathbf{\hat{n}} = \begin{pmatrix} \sin\theta\cos\varphi + \gamma\cos\theta\\ \sin\theta\sin\varphi\\ \cos\theta \end{pmatrix}.$$
 (5)

We compute the *xz*, *xx*, and *zz* components of the stress tensor for the 3D case as follows:

$$\sigma_{xz} = \frac{\rho}{4\pi} \int_{\varphi} \int_{\theta} d\theta d\varphi \sin \theta \left[\tau \frac{(\sin \theta \cos \varphi + \gamma \cos \theta) \cos \theta}{|\mathbf{\Lambda} \hat{\mathbf{n}}|} \right]$$
(6)

$$\sigma_{xx} = \frac{\rho}{4\pi} \int_{\varphi} \int_{\theta} d\theta d\varphi \sin \theta \left[\tau \frac{(\sin \theta \cos \varphi + \gamma \cos \theta)^2}{|\mathbf{\Lambda} \hat{\mathbf{n}}|} \right]$$
(7)

$$\sigma_{zz} = \frac{\rho}{4\pi} \int_{\varphi} \int_{\theta} d\theta d\varphi \sin \theta \left[\tau \frac{\cos^2 \theta}{|\mathbf{\Lambda} \mathbf{\hat{n}}|} \right]$$
(8)

The integrals are taken over the ranges $0 \le \theta \le 2\pi$ and $0 \le \varphi \le \pi$. To compare with our results for the 3D FCC network, we use $\mu = 1$ and initial line density $\rho = \frac{12}{\sqrt{2}}$, corresponding to the fully-connected FCC lattice with $l_0 = 1$ [3].

We repeat the same process for the 2D case, in which the deformation tensor for simple shear is

$$\mathbf{\Lambda}(\gamma) = \begin{pmatrix} 1 & \gamma \\ 0 & 1 \end{pmatrix} \tag{9}$$

and the transformed orientation vector is

$$\mathbf{\Lambda}\mathbf{\hat{n}} = \begin{pmatrix} \cos\theta + \gamma\sin\theta\\ \sin\theta \end{pmatrix}.$$
 (10)

The resulting components of the 2D stress tensor are calculated as follows:

$$\sigma_{xz} = \frac{\rho}{2\pi} \int_{\theta} d\theta \left[\tau \frac{(\cos\theta + \gamma \sin\theta)\sin\theta}{|\mathbf{\Lambda}\hat{\mathbf{n}}|} \right]$$
(11)

$$\sigma_{xx} = \frac{\rho}{2\pi} \int_{\theta} d\theta \left[\tau \frac{(\cos \theta + \gamma \sin \theta)^2}{|\mathbf{A}\hat{\mathbf{n}}|} \right]$$
(12)

$$\tau_{zz} = \frac{\rho}{2\pi} \int_{\theta} d\theta \left[\tau \frac{\sin^2 \theta}{|\mathbf{\Lambda} \hat{\mathbf{n}}|} \right]$$
(13)

Here, the integrals are taken over the range $0 \le \theta \le 2\pi$.

Principal strain axes for simple shear

For simple shear with deformation gradient tensor

$$\mathbf{\Lambda}(\gamma) = \begin{pmatrix} 1 & 0 & \gamma \\ 0 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix},\tag{14}$$

we can decompose Λ into a combination of a pure stretch Uand a rigid body rotation R, satisfying $\Lambda = RU$. From the right Cauchy-Green tensor $C = U^2 = \Lambda^T \Lambda$, we determine

$$U = \frac{1}{\sqrt{4 + \gamma^2}} \begin{pmatrix} 2 & 0 & \gamma \\ 0 & \sqrt{4 + \gamma^2} & 0 \\ \gamma & 0 & 2 + \gamma^2 \end{pmatrix}.$$
 (15)

The eigenvalues of U are

$$\lambda_1 = \frac{\gamma + \sqrt{\gamma^2 + 4}}{2}, \quad \lambda_2 = \frac{-\gamma + \sqrt{\gamma^2 + 4}}{2}, \quad \lambda_3 = 1$$
 (16)

with corresponding eigenvectors

$$\boldsymbol{v}_1 = \left(\frac{1}{2}\left(-\gamma + \sqrt{4 + \gamma^2}\right), 0, 1\right)$$
$$\boldsymbol{v}_2 = \left(\frac{1}{2}\left(-\gamma - \sqrt{4 + \gamma^2}\right), 0, 1\right)$$
$$\boldsymbol{v}_3 = (0, 1, 0)$$

 λ_1 and λ_2 correspond to the elongation l/l_0 along the axes of maximum extension and compression, respectively.

The rotation matrix R is determined as $R = \Lambda U^{-1}$,

$$\boldsymbol{R} = \frac{1}{\sqrt{4 + \gamma^2}} \begin{pmatrix} 2 & 0 & \gamma \\ 0 & \sqrt{4 + \gamma^2} & 0 \\ -\gamma & 0 & 2 \end{pmatrix}$$
(17)

The maximum stretch direction then corresponds to $v'_1 = \mathbf{R}v_1$. In the limit of small strains, v'_1 is oriented at $\theta = \pi/4$ above the *x*-axis, in the *x*-*z* plane.

- C. Storm, J. J. Pastore, F. C. MacKintosh, T. C. Lubensky, and P. A. Janmey, Nature 435, 191 (2005).
- [2] C. P. Broedersz, C. Storm, and F. C. MacKintosh, Physical Review E **79**, 1 (2009).

Bending interaction models

Prior work has shown that network models with bond-bending interactions (angle constraints between all nearest-neighbor bonds) exhibit the same strain-driven critical behavior as networks with freely-hinging crosslinks (angle constraints between only initially collinear nearest-neighbor bonds) [4, 5]. To emphasize that the details of the bending interactions do not influence our conclusions with regard to the normal stresses, we consider the mechanics of phantomized triangular networks with z = 3.4 and either freely-hinging crosslinks (bending along initially collinear fibers) or bond-bending interactions, both with $\tilde{\kappa} = 10^{-5}$. In Fig. S1, we show both K and the normal stresses σ_{ii}/γ^2 for each bending interaction type. We observe that the networks show qualitatively similar behavior in both cases, with $K \propto \tilde{\kappa}$ and $\sigma_{ii} \propto \tilde{\kappa} \gamma^2$ below the critical strain. The only apparent difference is that the magnitudes of K and the normal stresses are slightly higher for bond-bending networks than for networks with freely-hinging crosslinks in the bending-dominated regime. This is due to the additional angle constraints imposed by bondbending interactions.

Packing-derived networks with varying connectivity

In Fig. S2, we show both $N_1/(\sigma_{xz}\gamma)$ and K for 2D packingderived networks of size W = 100 with $\kappa = 10^{-5}$ and varying z. We observe that, for individual samples, peaks in $N_1/(\sigma_{xz}\gamma)$ occur at the z-dependent critical strain. On average, the Lodge-Meissner relation is satisfied.

- [3] A. J. Licup, A. Sharma, and F. C. MacKintosh, Physical Review E 93, 1 (2016).
- [4] A. Sharma, A. J. Licup, K. A. Jansen, R. Rens, M. Sheinman, G. H. Koenderink, and F. C. MacKintosh, Nature Physics 12, 584 (2016).
- [5] R. Rens, M. Vahabi, A. J. Licup, F. C. MacKintosh, and A. Sharma, Journal of Physical Chemistry B 120, 5831 (2016).



FIG. S1. Top: Differential shear modulus *K* vs. shear strain γ for 2D phantomized triangular networks with z = 3.4, $\tilde{\kappa} = 10^{-6}$ and $\delta_{\text{max}} = 0.4$, with freely-hinging crosslinks and bending interactions only along fibers (triangles) and with bond-bending interactions between all nearest-neighbor bonds (circles). Bottom: Normal stresses σ_{xx} and σ_{zz} , both normalized by γ^2 , as a function of γ for the same systems. Both *K* and σ_{ii} are higher in networks with bond-bending interactions than in the same networks with freely-hinging crosslinks, due to the additional angle constraints.



FIG. S2. In 2D packing-derived networks with $\kappa = 10^{-5}$ and varying *z*, we observe either upward or downward peaks in $N_1/(\sigma_{xz}\gamma)$ (top) for individual samples at the *z*-dependent critical strain γ_c , determined as the inflection point of *K* vs γ (bottom) plotted on a logarithmic scale. When the stress is averaged over all samples, the network ensemble approximately satisfies the Lodge-Meissner relation, such that $N_1/(\sigma_{xz}\gamma) = 1$ over the full strain range.