Instabilities in liquid crystal elastomers (supplementary information)

L. Angela Mihai[†] Alain Goriely[*]

[†]*School of Mathematics, Cardiff University, Cardiff, UK*
[*]*Mathematical Institute, University of Oxford, Oxford, UK}*

March 25, 2021

1 Stochastic model parameters

We refer to for extensive reviews on the information-theoretic approach in stochastic elasticity, and to the various papers cited in the main text for numerous examples and applications of continuum models with stochastic parameters. Here, we adopt the following hypothesis required by the stochastic models for nematic liquid crystal elastomers (LCEs) presented in [9]: For any given finite deformation, at any point in the material, the shear modulus $\mu > 0$, the shape parameter $a > 0$, and their inverse, $1/\mu$ and $1/a$, respectively, are second order random variables, i.e., they have finite mean value and finite variance. For the shear modulus μ (and similarly for a), to construct a prior probability law, we note that this assumption is guaranteed by setting the following mathematical expectations:

$$\begin{cases} E\left[\mu\right] = \underline{\mu} > 0, \\ E\left[\log\ \mu\right] = \nu, \quad \text{such that } |\nu| < +\infty. \end{cases} \tag{1}$$

The first constraint in (1) specifies the mean value for the random shear modulus μ, while the second constraint provides a condition from which it follows that $1/\mu$ is a second order random variable. Then, by the maximum entropy principle, the shear modulus μ with mean value $\underline{\mu}$ and standard deviation $\|\mu\| = \sqrt{\text{Var}[\mu]}$ (defined as the square root of the variance, $\text{Var}[\mu]$) follows a Gamma probability distribution with shape and scale parameters $\rho_1 > 0$ and $\rho_2 > 0$ respectively, such that

$$\underline{\mu} = \rho_1\rho_2, \qquad \|\mu\| = \sqrt{\rho_1}\rho_2. \tag{2}$$

The corresponding probability density function takes the form

$$g(\mu;\rho_1,\rho_2) = \frac{\mu^{\rho_1-1}e^{-\mu/\rho_2}}{\rho_2^{\rho_1}\Gamma(\rho_1)}, \qquad \text{for } \mu > 0 \text{ and } \rho_1,\rho_2 > 0, \tag{3}$$

where $\Gamma : \mathbb{R}_+^* \to \mathbb{R}$ is the complete Gamma function

$$\Gamma(z) = \int_0^{+\infty} t^{z-1}e^{-t}\text{d}t. \tag{4}$$

The word 'hyperparameters' is often used for ρ_1 and ρ_2 to distinguish them from μ and other material constants.

When $\mu = \mu_1 + \mu_2$, setting a fixed constant value $b > -\infty$, such that $\mu_i > b$, $i = 1, 2$ (e.g., $b = 0$ if $\mu_1 > 0$ and $\mu_2 > 0$, although b is not unique in general), we define the auxiliary random variable

$$R_1 = \frac{\mu_1 - b}{\mu - 2b}, \tag{5}$$

such that $0 < R_1 < 1$. Then, the random model parameters can be expressed equivalently as follows,

$$\mu_1 = R_1(\mu - 2b) + b, \qquad \mu_2 = \mu - \mu_1 = (1 - R_1)(\mu - 2b) + b. \tag{6}$$

It is reasonable to assume

$$\begin{cases} E\left[\log\ R_1\right] = \nu_1, & \text{such that } |\nu_1| < +\infty, \\ E\left[\log(1 - R_1)\right] = \nu_2, & \text{such that } |\nu_2| < +\infty, \end{cases} \tag{7}$$

in which case, the random variable R_1 follows a standard Beta distribution, with hyperparameters $\xi_1 > 0$ and $\xi_2 > 0$ satisfying

$$\underline{R}_1 = \frac{\xi_1}{\xi_1 + \xi_2}, \qquad \mathrm{Var}[R_1] = \frac{\xi_1 \xi_2}{(\xi_1 + \xi_2)^2\,(\xi_1 + \xi_2 + 1)}, \tag{8}$$

where $\underline{R}_1$ is the mean value and $\mathrm{Var}[R_1]$ is the variance of R_1. The associated probability density function is

$$\beta(r; \xi_1, \xi_2) = \frac{r^{\xi_1 - 1}(1 - r)^{\xi_2 - 1}}{B(\xi_1, \xi_2)}, \qquad \text{for } r \in (0, 1) \text{ and } \xi_1, \xi_2 > 0, \tag{9}$$

where $B : \mathbb{R}_+^* \times \mathbb{R}_+^* \to \mathbb{R}$ is the Beta function

$$B(x, y) = \int_0^1 t^{x-1}(1 - t)^{y-1} dt. \tag{10}$$

Then, for the random coefficients given by (6), the corresponding mean values are

$$\underline{\mu}_1 = \underline{R}_1(\underline{\mu} - 2b) + b, \qquad \underline{\mu}_2 = \underline{\mu} - \underline{\mu}_1 = (1 - \underline{R}_1)(\underline{\mu} - 2b) + b, \tag{11}$$

and the variances and covariance take the form, respectively,

$$\mathrm{Var}\left[\mu_1\right] = (\underline{\mu} - 2b)^2 \mathrm{Var}[R_1] + (\underline{R}_1)^2 \mathrm{Var}[\mu] + \mathrm{Var}[\mu]\mathrm{Var}[R_1], \tag{12}$$

$$\mathrm{Var}\left[\mu_2\right] = (\underline{\mu} - 2b)^2 \mathrm{Var}[R_1] + (1 - \underline{R}_1)^2 \mathrm{Var}[\mu] + \mathrm{Var}[\mu]\mathrm{Var}[R_1], \tag{13}$$

$$\mathrm{Cov}[\mu_1, \mu_2] = \frac{1}{2}\left(\mathrm{Var}[\mu] - \mathrm{Var}[\mu_1] - \mathrm{Var}[\mu_2]\right). \tag{14}$$

2 Stress tensors for ideal nematic elastomers

We briefly recall the relations between the stress tensors of an ideal nematic elastomer and those of the underlying hyperelastic model. These relations were originally obtained in [10]. For an ideal incompressible nematic elastomer, the neoclassical strain-energy density function takes the general form

$$W^{(nc)}(\mathbf{F}, \mathbf{n}) = W(\mathbf{A}), \tag{15}$$

where the right-hand side represents the strain-energy function of a homogeneous isotropic incompressible hyperelastic material, depending only on the elastic deformation gradient $\mathbf{A}$. On the left-hand side, $\mathbf{n}$ is a unit vector for the localized direction of uniaxial nematic alignment in the present configuration; $\mathbf{F} = \mathbf{GA}$ is the deformation gradient tensor with respect to the reference isotropic state, with $\mathbf{G} = a^{-1/6}\mathbf{I} + \left(a^{1/3} - a^{-1/6}\right)\mathbf{n} \otimes \mathbf{n}$ the 'spontaneous' (or 'natural') deformation tensor and $\mathbf{A}$ the (local) elastic deformation tensor; $a > 0$ is a temperature-dependent, spatially-independent shape parameter; $\otimes$ denotes the tensor product of two vectors; and $\mathbf{I} = \mathrm{diag}(1, 1, 1)$ is the identity tensor.

The strain-energy function given by (15) takes the equivalent form

$$\mathcal{W}^{(nc)}(\lambda_1, \lambda_2, \lambda_3, \mathbf{n}) = W^{(nc)}(\mathbf{F}, \mathbf{n}), \tag{16}$$

where $\{\lambda_i^2\}_{i=1,2,3}$ are the eigenvalues of the tensor $\mathbf{F}\mathbf{F}^T$.

For the hyperelastic material described by the strain-energy function $W(\mathbf{A})$, the Cauchy stress tensor (representing the internal force per unit of deformed area acting within the deformed solid) is equal to

$$\mathbf{T} = (\det \mathbf{A})^{-1} \frac{\partial W}{\partial \mathbf{A}} \mathbf{A}^T - p\mathbf{I}, \tag{17}$$

where p denotes the Lagrange multiplier for the incompressibility constraint $\det \mathbf{A} = 1$.

The associated first Piola-Kirchhoff stress tensor (representing the internal force per unit of undeformed area acting within the deformed solid) is then

$$\mathbf{P} = \mathbf{T}\mathrm{Cof}(\mathbf{A}), \tag{18}$$

where $\mathrm{Cof}(\mathbf{A}) = (\det \mathbf{A})\, \mathbf{A}^{-T}$ is the cofactor of $\mathbf{A}$.

2.1 Free director

When the nematic director is 'free' to rotate relative to the elastic matrix, $\mathbf{F}$ and $\mathbf{n}$ are independent variables, and the Cauchy stress tensor for the nematic material with the strain-energy function described by (15) is calculated as follows,

$$\begin{aligned} \mathbf{T}^{(nc)} &= J^{-1} \frac{\partial W^{(nc)}}{\partial \mathbf{F}} \mathbf{F}^T - p^{(nc)}\mathbf{I} \\ &= J^{-1}\mathbf{G}^{-1} \frac{\partial W}{\partial \mathbf{A}} \mathbf{A}^T \mathbf{G} - p^{(nc)}\mathbf{I} \\ &= J^{-1}\mathbf{G}^{-1}\mathbf{T}\mathbf{G}, \end{aligned} \tag{19}$$

where $\mathbf{T}$ is the Cauchy stress tensor defined by (17), $J = \det \mathbf{F}$, and the scalar $p^{(nc)}$ represents the Lagrange multiplier for the internal constraint $J = 1$.

The principal components $\left(T_1^{(nc)}, T_2^{(nc)}, T_3^{(nc)}\right)$ of the Cauchy stress defined by (19) are the solutions of the characteristic equation

$$\det\left(\mathbf{T}^{(nc)} - \Lambda\mathbf{I}\right) = 0. \tag{20}$$

Since

$$\det\left(\mathbf{T}^{(nc)} - \Lambda\mathbf{I}\right) = \det\left[\mathbf{G}^{-1}\left(J^{-1}\mathbf{T} - \Lambda\mathbf{I}\right)\mathbf{G}\right] = J^{-1}\det\left(\mathbf{T} - J\Lambda\mathbf{I}\right), \tag{21}$$

it follows that the principal Cauchy stresses for the underlying hyperelastic model satisfy

$$(T_1, T_2, T_3) = J\left(T_1^{(nc)}, T_2^{(nc)}, T_3^{(nc)}\right). \tag{22}$$

Therefore, if the Baker-Ericksen inequalities hold for the hyperelastic model, then the greater principal Cauchy stress occurs in the direction of the greater principal elastic stretch for the nematic model. We recall that, for a hyperelastic material, the Baker-Ericksen inequalities state that the greater principal stress occurs in the direction of the greater principal stretch [1, 4].

In the presence of a nematic field, the total Cauchy stress tensor $\mathbf{T}^{(nc)}$ given by (19) is not symmetric in general. In addition, the following condition is required,

$$\frac{\partial W^{(nc)}}{\partial \mathbf{n}} = \mathbf{0}, \tag{23}$$

or equivalently, by the principle of material objectivity,

$$\frac{1}{2}\left(\mathbf{T}^{(nc)} - \mathbf{T}^{(nc)T}\right)\mathbf{n} = \mathbf{0}, \tag{24}$$

where $\left(\mathbf{T}^{(nc)} - \mathbf{T}^{(nc)^T}\right)/2$ represents the skew-symmetric part of the Cauchy stress tensor.

The first Piola-Kirchhoff stress tensor for the nematic material is equal to

$$\mathbf{P}^{(nc)} = \mathbf{T}^{(nc)}\mathrm{Cof}(\mathbf{F}) = \mathbf{G}^{-1}\mathbf{T}\mathbf{A}^{-T} = \mathbf{G}^{-1}\mathbf{P}, \tag{25}$$

where $\mathbf{P}$ is the first Piola-Kirchhoff stress given by (18).

2.2 Frozen director

If the nematic director is 'frozen', the Cauchy stress tensor for the nematic material takes the form

$$\widehat{\mathbf{T}}^{(nc)} = J^{-1}\mathbf{G}^{-1}\mathbf{T}\mathbf{G} - J^{-1}q\left(\mathbf{I} - \frac{\mathbf{F}\mathbf{n}_0 \otimes \mathbf{F}\mathbf{n}_0}{|\mathbf{F}\mathbf{n}_0|^2}\right)\mathbf{n} \otimes \frac{\mathbf{F}\mathbf{n}_0}{|\mathbf{F}\mathbf{n}_0|}, \tag{26}$$

where $\mathbf{T}$ is the Cauchy stress defined by (17), $J = \det \mathbf{F}$, $p^{(nc)}$ is the Lagrange multiplier for the volume constraint $J = 1$, and q is the Lagrange multiplier for the constraint

$$\mathbf{n} = \frac{\mathbf{F}\mathbf{n}_0}{|\mathbf{F}\mathbf{n}_0|}. \tag{27}$$

As the Cauchy stress tensor given by (26) is not symmetric in general, the following additional condition must hold,

$$\frac{\partial \widehat{W}^{(nc)}}{\partial \mathbf{n}} = \mathbf{0}, \tag{28}$$

or equivalently,

$$\frac{1}{2}\left(\widehat{\mathbf{T}}^{(nc)} - \widehat{\mathbf{T}}^{(nc)T}\right)\mathbf{n} = \mathbf{0}. \tag{29}$$

The corresponding first Piola-Kirchhoff stress tensor for the nematic material is equal to

$$\widehat{\mathbf{P}}^{(nc)} = \widehat{\mathbf{T}}^{(nc)}\mathrm{Cof}(\mathbf{F}). \tag{30}$$

3 Cavitation of a nematic sphere

The static and dynamic cavitation of homogeneous and radially inhomogeneous isotropic incompressible hyperelastic spheres with stochastic material parameters was analyzed in [8,11] where up-to-date reviews of the relevant literature are presented. For an inflated elastic sphere, the radially symmetric deformation takes the form

$$r = g(R), \qquad \theta = \Theta, \qquad \phi = \Phi, \tag{31}$$

where (R, Θ, Φ) and (r, θ, ϕ) are the spherical polar coordinates in the reference and current configuration, respectively, such that $0 \leq R \leq B$, and $g(R) \geq 0$ is to be determined. The corresponding deformation gradient is equal to $\mathbf{A} = \mathrm{diag}\,(\alpha_1, \alpha_2, \alpha_3)$, with

$$\alpha_1 = \frac{\mathrm{d}g}{\mathrm{d}R} = \alpha^{-2}, \qquad \alpha_2 = \alpha_3 = \frac{g(R)}{R} = \frac{r}{R} = \alpha, \tag{32}$$

where α_1 and $\alpha_2 = \alpha_3$ are the radial and hoop principal stretches, respectively, and $\mathrm{d}g/\mathrm{d}R$ denotes the derivative of g with respect to R. By (32),

$$g^2\frac{\mathrm{d}g}{\mathrm{d}R} = R^2, \tag{33}$$

hence,

$$g(R) = \left(R^3 + c^3\right)^{1/3}, \tag{34}$$

where $c \geq 0$ is a constant to be calculated. If $c > 0$, then $g(R) \to c > 0$ as $R \to 0_+$, and a spherical cavity of radius c forms at the center of the sphere, from zero initial radius, otherwise the sphere remains undeformed.

In particular, for a static sphere of neo-Hookean material, if the surface of the cavity is traction-free, the radial component of the Cauchy stress is equal to (see [11] for a detailed derivation using the same notation)

$$T_{rr}(b) = \frac{2}{3}\int_{x^3+1}^{\infty} \mu\frac{1+u}{u^{7/3}}\mathrm{d}u, \tag{35}$$

where $x = c/B$. After evaluating the integral in (35), the required uniform dead-load traction at the outer surface, $R = B$, in the reference configuration, takes the form

$$P = \left(x^3+1\right)^{2/3} T_{rr}(b) = 2\mu \left[\left(x^3+1\right)^{1/3} + \frac{1}{4\left(x^3+1\right)^{2/3}}\right], \tag{36}$$

and increases as x increases. The critical dead load for the onset of cavitation is then

$$P_0 = \lim_{x\to 0_+} P = \frac{5\mu}{2}. \tag{37}$$

To analyze the stability of this cavitation, we study the behavior of the cavity opening, with radius c as a function of P, in a neighborhood of P_0. After differentiating the function given by (36), with respect to the dimensionless cavity radius $x = c/B$, we have

$$\frac{\mathrm{d}P}{\mathrm{d}x} = 2\mu x^2 \left[\frac{1}{\left(x^3+1\right)^{2/3}} - \frac{1}{2\left(x^3+1\right)^{5/3}}\right] > 0, \tag{38}$$

i.e., the cavitation is stable, regardless of the material parameter $\mu > 0$.

For a nematic sphere of neoclassical material with the strain-energy function given by (15) derived from the neo-Hookean hyperelastic model, when $\mathbf{F} = \mathrm{diag}\left(\lambda^{-2}, \lambda, \lambda\right)$ and $\mathbf{G} = \mathrm{diag}\left(a^{-1/3}, a^{1/6}, a^{1/6}\right)$, with $\lambda > a^{1/6} > 1$, the Cauchy stress is equal to that of the neo-Hookean sphere. Hence, $T_{rr}^{(nc)} = T_{rr}$, and the first Piola-Kirchhoff stress representing the critical dead load for cavitation in the nematic sphere is equal to $P_0^{(nc)} = a^{1/3} P_0 = 5a^{1/3}\mu/2$.

4 Inflation of a nematic spherical shell

Static and dynamic inflation instabilities of homogeneous and radially inhomogeneous isotropic incompressible hyperelastic spheres with stochastic material parameters were analyzed in [5–7]. For a thin hyperelastic spherical shell, such that $0 < \epsilon = (B - A)/A \ll 1$, where A and B represent the inner and outer radii of the reference shell, respectively, if the external pressure is equal to zero, then the internal pressure can be approximated as follows,

$$T = \frac{\epsilon}{\alpha^2} \frac{\mathrm{d}\mathcal{W}}{\mathrm{d}\alpha}, \tag{39}$$

where the deformation gradient for radially symmetric inflation is equal to $\mathbf{A} = \mathrm{diag}\left(\alpha^{-2}, \alpha, \alpha\right)$, with $\alpha = r/R > 1$, and $\mathcal{W}(\alpha) = W(\mathbf{A})$. The critical value of α where a limit-point instability occurs is obtained by solving for $\alpha > 1$ the following equation,

$$\frac{\mathrm{d}T}{\mathrm{d}\alpha} = 0, \tag{40}$$

where T is the radial component of the Cauchy stress given by (39).

For a nematic sphere of neoclassical material with the strain-energy function given by (15) derived from the Mooney-Rivlin hyperelastic model, when $\mathbf{F} = \mathrm{diag}\left(\lambda^{-2}, \lambda, \lambda\right)$ and $\mathbf{G} = \mathrm{diag}\left(a^{-1/3}, a^{1/6}, a^{1/6}\right)$, with $\lambda > a^{1/6} > 1$, the Cauchy stress is equal to that of the hyperelastic sphere. If the shell is thin, assuming zero external pressure, the internal pressure can be approximated as

$$T^{(nc)} = T = \frac{\epsilon}{\alpha^2} \frac{\mathrm{d}\mathcal{W}}{\mathrm{d}\alpha} = \frac{\epsilon a^{1/2}}{\lambda^2} \frac{\mathrm{d}\mathcal{W}^{(nc)}}{\mathrm{d}\lambda}. \tag{41}$$

Then, the critical value of λ where a limit-point instability occurs is found by solving for $\lambda > a^{1/3}$ the equation

$$\frac{\mathrm{d}T^{(nc)}}{\mathrm{d}\lambda} = 0, \tag{42}$$

with $T^{(nc)}$ given by (41).

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