Universal states in nematic elastomers

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Outline

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Controllable and universal deformations

• A deformation $y$ maps material points $x$ into points $y(x)$ of space.

- A deformation $y$ is controllable if it can be maintained in equilibrium by the action of surface tractions alone:
  \[
  \text{div} T = 0 \quad \text{on } B_*, \quad \quad T\nu = \tau \quad \text{on } \partial B_*. 
  \]

- A deformation $y$ is universal within a class of materials if it is controllable for every material in that class.
**Universal deformations in conventional elastomers**

- In equilibrium, a conventional elastomer can be usefully modeled as a homogeneous, incompressible, and isotropic hyperelastic material.

- For a homogeneous, incompressible, and isotropic hyperelastic material the Cauchy stress tensor has the form

\[
T = -p1 + 2\frac{\partial \tilde{w}(I_1, I_2)}{\partial I_1} B - 2\frac{\partial \tilde{w}(I_1, I_2)}{\partial I_2} B^{-1},
\]

where \( p \) is a constitutively indeterminate pressure and \( \tilde{w} \) gives the strain-energy density \( w \) as a function of \( I_1 = \text{tr} B \) and \( I_2 = \frac{1}{2}(\text{tr}^2 B - \text{tr}(B^2)) \). (Here, \( B = FF^\top \) and \( F = \text{Grad} \ y \).)

- The problem of determining the complete set of universal deformations for the class of homogeneous, incompressible, and isotropic solid was first addressed by *Ericksen* (1954).
Experimental importance of Ericksen’s problem

To predict the behavior of any particular elastomer requires a characterizing function $\tilde{w}$ (or its derivatives). To determine one:

- Begin with a model expression, posited on molecular or phenomenological grounds, involving one or more free parameters.
- Perform various (e.g., stress vs. strain) experiments and a regression to fit the parameters, yielding a closed model.
- Perform a final independent experiment to test the fidelity of the closed model.

Because infidelity may occur, the experiments cannot be designed with reference to the original model expression.

Allowance should be made for the need to revise the original model and any subsequent improvements upon that model.

Experiments to identify $\tilde{w}$ for a given elastomer should, therefore, be designed to produce deformations that are universal in the class of homogeneous, incompressible, isotropic solids.
Status of Ericksen’s problem

- Families of universal deformations:

  1. Homogeneous deformations.
  2. Bending, stretching, and shearing of a rectangular block.
  3. Straightening, stretching, and shearing of a sector of a hollow cylinder.
  4. Inflation, bending, torsion, extension, and shearing of an annular wedge.
  5. Inflation or eversion of a sector of a spherical shell.
  6. Inflation, bending, extension, and azimuthal shearing of an annular wedge.

- Completeness open ... Families with $I_1$ and $I_2$ constant ...
**Nematic elastomers**

- A nematic elastomer is a rubber-like solid formed by cross-linking a nematic polymeric liquid-crystalline fluid.

- The kinematical description of a nematic elastomer involves:
  - $y$, the *deformation*, a macroscopic measure of the distortion of the network;
  - $Q$, the *nematic order tensor*, a macroscopic measure of the orientation of the nematic molecules; and
  - $A$, the *conformation*, a macroscopic measure of the distortion of the chain shape induced by nematic ordering.
• Free-energy density

\[ \psi = \psi_e(F, A; A_*) + \psi_n(Q), \]

with \( A_* \) the conformation in the reference configuration.

○ Under a rigid rotation \( H \) of the reference configuration:

\[ F \mapsto FH, \quad A \mapsto A, \quad A_* \mapsto H^\top A_* H, \quad Q \mapsto Q \]

○ Under a rigid rotation \( R \) of the deformed configuration:

\[ F \mapsto RF, \quad A \mapsto RA R^\top, \quad A_* \mapsto A_*, \quad Q \mapsto RQR^\top \]

○ Symmetry and objectivity require that

\[ \psi_e(F, A; A_*) = \tilde{\psi}_e(B, A, FA_* F^\top), \]

with \( \tilde{\psi}_e \) isotropic, and

\[ \psi_n(Q) = \tilde{\psi}_n(\text{tr}(Q^2), \text{tr}(Q^3)). \]
• When the eigenvalues of $Q$ are constant, so that the eigenvectors can only rotate, $\psi_n$ is constant.

• The neo-classical molecular-statistical model (Warner, Gelling & Vilgis 1988; Bladon, Warner & Terentjev 1994) extends the Gaussian theory of rubber elasticity to give

$$\psi_e(F, A; A_*) = \frac{\mu}{2}(\text{tr}(A^{-1} F A_* F^\top) - 2 \det(A^{-1} F A_* F^\top) - 3),$$

with $\mu > 0$ the shear modulus. This model also presumes a functional relation between $A$ and $Q$. The free-energy density $\psi$ then becomes a function of $F$ and $Q$ (Uchida & Onuki, 1999) or $F$ and $A$ (F & Sellers 2004).

• One might wish to penalize spatial variations of $Q$ (and/or $A$) by including energetic dependence upon the gradients of $Q$ (and/or $A$) (Terentjev, Warner & Verwey 1996). Here, we neglect all such dependences.
Assumptions

- Dependence upon $Q$ can be replaced by dependence upon $A$, giving:

$$\psi = \tilde{\psi}(B, A, FA_*F^T).$$

- Constraints:
  - Macroscopic incompressibility:
    $$\det F = 1$$
  - Microscopic incompressibility:
    $$\det(A^{-1}A_*) = 1$$

Underlying the second constraint is the idea that, while the chain shape, as described by $A$ may change, the volume occupied by a generic polymer chain should not.
Materials isotropic in the reference state

When $A_*$ is spherical (cf. DeSimone & Dolzmann 2000),

$$
\psi = \tilde{\psi}(B, A) = \psi(\mathcal{I}),
$$

with $\mathcal{I} = (I_1, I_2, J_1, J_2, J_3, K_1, K_2, K_3, K_4)$ and

$$
I_1 = \text{tr} B, \quad I_2 = \frac{1}{2}(\text{tr}^2 B - \text{tr}(B^2)),
$$

$$
J_1 = \text{tr}(A^{-1}), \quad J_2 = \text{tr}(A^{-2}), \quad J_3 = \text{tr}(A^{-3}),
$$

$$
K_1 = \text{tr}(A^{-1}B), \quad K_2 = \text{tr}(A^{-1}B^2),
$$

$$
K_3 = \text{tr}(A^{-2}B), \quad K_4 = \text{tr}(A^{-2}B^2).
$$
Equations of equilibrium

• Vary $y$:

$$\text{div} \left( 2 \frac{\partial \psi(I)}{\partial B} B \right) = \text{grad} p$$

Macroscopic incompressibility renders controllable loading involving both conservative body forces and surface tractions.

• Vary $A$:

$$\frac{\partial \psi(I)}{\partial A} = qA^{-1}$$

The partial derivatives appearing in the above equations indicate differentiation on the constraint manifold.
St. Venant torsion  
(Conventional elastomers: *Rivlin* 1947)

- Consider a right-circular cylinder of radius $R$ and height $L$.
- St. Venant torsion is described by a deformation with gradient

$$F = 1 + \frac{\omega r}{L}e_\theta \otimes e_z,$$

where $\omega > 0$ is the angle of twist.

- Assume also that the conformation has the uniaxial form

$$A = as^{1/3}(1 + (1 - s)e_r \otimes e_r),$$

with $a = \det A = \det A_\star > 0$ and $s > 0$, the molecular anisotropy, constant. (Note: For $0 < s < 1$, $A$ is oblate about $e_r$; for $s = 1$, $A$ is spherical; for $s > 1$, $A$ is prolate about $e_r$.)
Alternatively, using rectangular Cartesian coordinates, we have $y$ and $A = as^{1/3}(1 + (1 - s)n \otimes n)$, with:

\[
\begin{align*}
y_1 &= x_1 \cos \left( \frac{\omega x_3}{L} \right) - x_2 \sin \left( \frac{\omega x_3}{L} \right), \\
y_2 &= x_1 \sin \left( \frac{\omega x_3}{L} \right) + x_2 \cos \left( \frac{\omega x_3}{L} \right), \\
y_3 &= x_3,
\end{align*}
\]

\[
\begin{align*}
n_1 &= \frac{y_1}{\sqrt{y_1^2 + y_2^2}}, \\
n_2 &= \frac{y_2}{\sqrt{y_1^2 + y_2^2}}, \\
n_3 &= 0.
\end{align*}
\]
Invariants for St. Venant torsion

- \( I_1 = 3 + \frac{\omega^2 R^2}{L^2} = I_2. \)

- \( J_1, J_2, J_3, K_1, K_2, K_3, \) and \( K_4 \) are constant:

\[
\begin{align*}
J_1 &= \text{tr}(A^{-1}) = \frac{1 + 2s}{as}, & J_2 &= \text{tr}(A^{-2}) = \frac{1 + 2s^2}{a^2 s^2}, \\
J_3 &= \text{tr}(A^{-3}) = \frac{1 + 2s^3}{a^3 s^3}, \\
K_1 &= \text{tr}(A^{-1} B) = \frac{1 + 2s}{as}, & K_2 &= \text{tr}(A^{-1} B^2) = \frac{1 + 2s}{as}, \\
K_3 &= \text{tr}(A^{-2} B) = \frac{1 + 2s^2}{a^2 s^2}, & K_4 &= \text{tr}(A^{-2} B^2) = \frac{1 + 2s^2}{a^2 s^2}.
\end{align*}
\]
Equations of equilibrium for St. Venant torsion

• Balance arising from variations of \( y \) determines \( p \).
  
  o Note:

  \[
  \frac{\partial \psi(I)}{\partial B} = \left( \frac{\partial \psi(I)}{\partial I_1} I + \frac{\partial \psi(I)}{\partial K_1} A^{-1} + \frac{\partial \psi(I)}{\partial K_3} A \right) B
  
  - B^{-1} \left( \frac{\partial \psi(I)}{\partial I_1} I + \frac{\partial \psi(I)}{\partial K_2} A^{-1} + \frac{\partial \psi(I)}{\partial K_4} A \right)
  \]

• Balance arising from variations of \( A \) determines \( q \).

  o Key observation: \( A^{-1}B = A^{-1}, \ldots \)

  \[
  \therefore \quad \frac{\partial \tilde{\psi}(B, A)}{\partial A} \propto A^{-1}
  \]
Resultants

(Cf. Rivlin 1947)

• Axial force:

\[
N = \int_0^R \int_0^\infty T_{33} r \, dr \, d\theta
\]

\[
= -\frac{2\pi \omega^2}{L^2} \int_0^R \left( \frac{\partial \Psi(I)}{\partial I_1} + 2 \frac{\partial \Psi(I)}{\partial I_2} \right) r^3 \, dr
\]

• Axial torque:

\[
T = \int_0^R \int_0^\infty (y_1 T_{32} - y_2 T_{31}) (r) r \, dr \, d\theta
\]

\[
= \frac{4\pi \omega}{L} \int_0^R \left( \frac{\partial \Psi(I)}{\partial I_1} + \frac{\partial \Psi(I)}{\partial I_2} \right) r^3 \, dr
\]
Conclusions

- A uniaxial disclinated state of St. Venant torsion is universal for nematic elastomers.

- St. Venant torsion does not yield insight concerning how $\Psi$ depends upon the additional invariants.

- To determine the nature of any dependence of $\Psi$ upon the additional invariants requires additional universal solutions (i.e., experiments)!
Energetics of St. Venant torsion

- Restrict attention to neo-classical nematic elastomers.
- Normalize $\Psi$ to vanish in the reference state.
- The net free-energy $F$ per unit volume of the specimen is
  \[ F = \frac{\mu s^{1/3} \omega^2 R^2}{4L^2}. \]
- For a Gaussian rubber with shear modulus $\mu$,
  \[ F_{\text{iso}} = \frac{\mu \omega^2 R^2}{4L^2}. \]

For $0 < s < 1$ (oblate chain anisotropy), $F < F_{\text{iso}}$ and a uniaxial disclinated state of St. Venant torsion is energetically preferred over an isotropic state.
Open questions/conjectures/work in progress

• What can be said about the energetics of disclinated states of St. Venant torsion in a generic nematic elastomer?
• Can we obtain a classification of the families of universal states for nematic elastomers comparable to that available for conventional elastomers?
• Given a nontrivial deformation \( y \) that is universal for a conventional elastomer, how should we define \( A \) so that the pair \((y, A)\) is a universal state for a nematic elastomer?
  
  ◦ Conjecture: For \((y, A)\) to be a universal state it is necessary and sufficient for \( A \) to obey

\[
A^{-1}BA \propto 1 \quad (B \neq 1).
\]

  If \( A^{-1}BA \propto 1 \), the balance arising from variations of \( A \) is satisfied. Sufficiency is therefore obvious. Necessity???

• How does the inclusion of gradient energy alter the story? (Cf. Marris 1978, 1979.)