

**FIBER-REINFORCED ELASTOMERS:
MACROSCOPIC PROPERTIES,
MICROSTRUCTURE EVOLUTION,
AND STABILITY**

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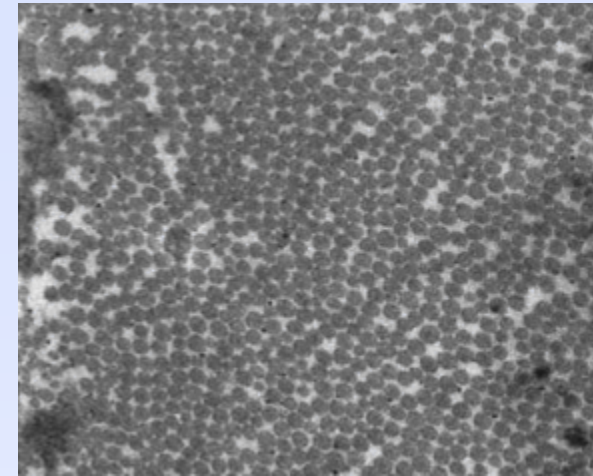
State University of New York, Stony Brook



Soft solids reinforced with fibers



Rubber reinforced with carbon-black and fabric



TEM of Collagen Fibrils in Human Brain Arteries

Thermoplastic Elastomers (Self-Assembled Nanodomains)

BCC



~ 50 nm

HX-Cyl.

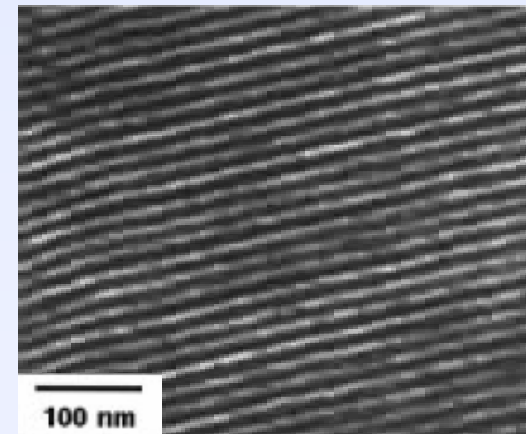


f_{PS}

Gyroid

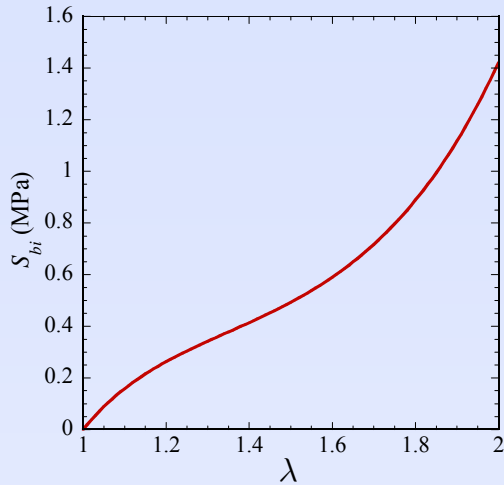


Lamellar

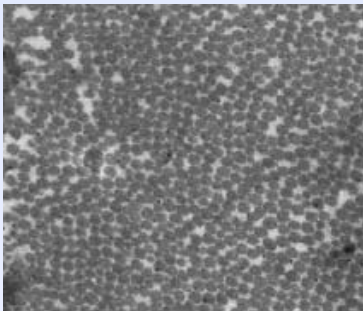


TEM of a triblock copolymer with cylindrical morphology

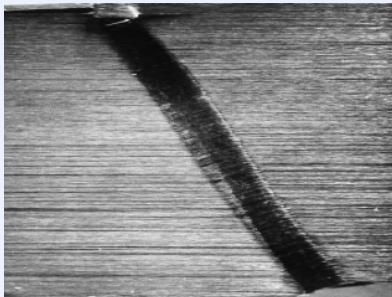
Issues in constitutive modeling of FREs



- **Nonlinear** constitutive matrix phase and fibers



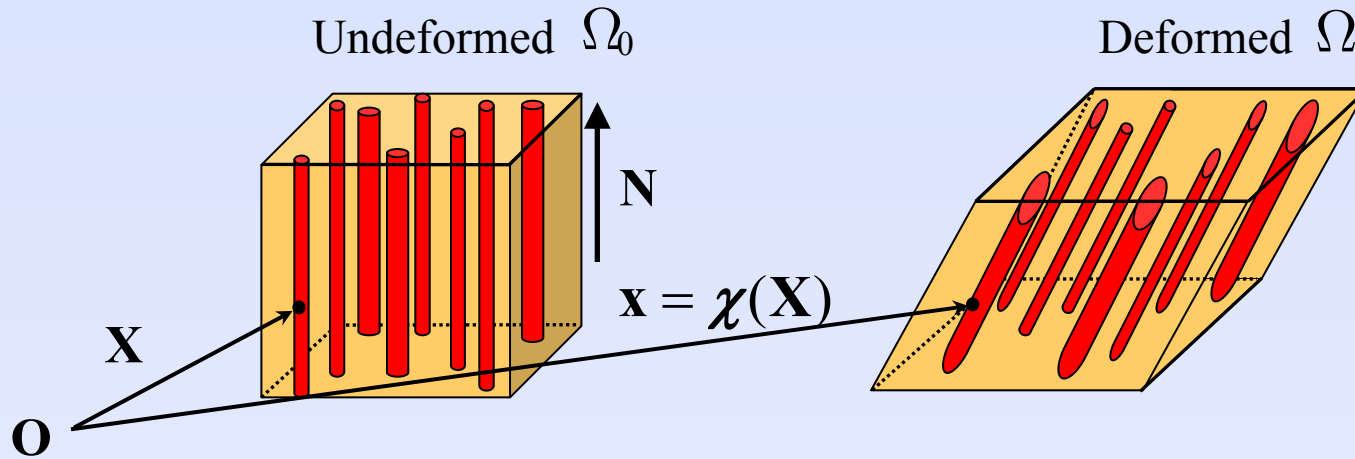
- **Complex** initial microstructure



- **Microstructure** evolution (geometric nonlinearity)
- **Development** of instabilities

Problem setting: Lagrangian formulation

Kinematics



Deformation
Gradient

$$\mathbf{F} = \frac{\partial \mathbf{x}}{\partial \mathbf{X}}$$

Right CG
Deformation Tensor

$$\mathbf{C} = \mathbf{F}^T \mathbf{F}$$

Local constitutive behavior

$$\mathbf{S} = \frac{\partial W}{\partial \mathbf{F}}(\mathbf{X}, \mathbf{F})$$

where

$$W(\mathbf{X}, \mathbf{F}) = \sum_{r=1}^2 \chi_0^{(r)}(\mathbf{X}) W^{(r)}(\mathbf{F})$$

Stored-energy function of the **matrix**

$$W^{(1)}(\mathbf{F})$$

Stored-energy function of the **fibers**

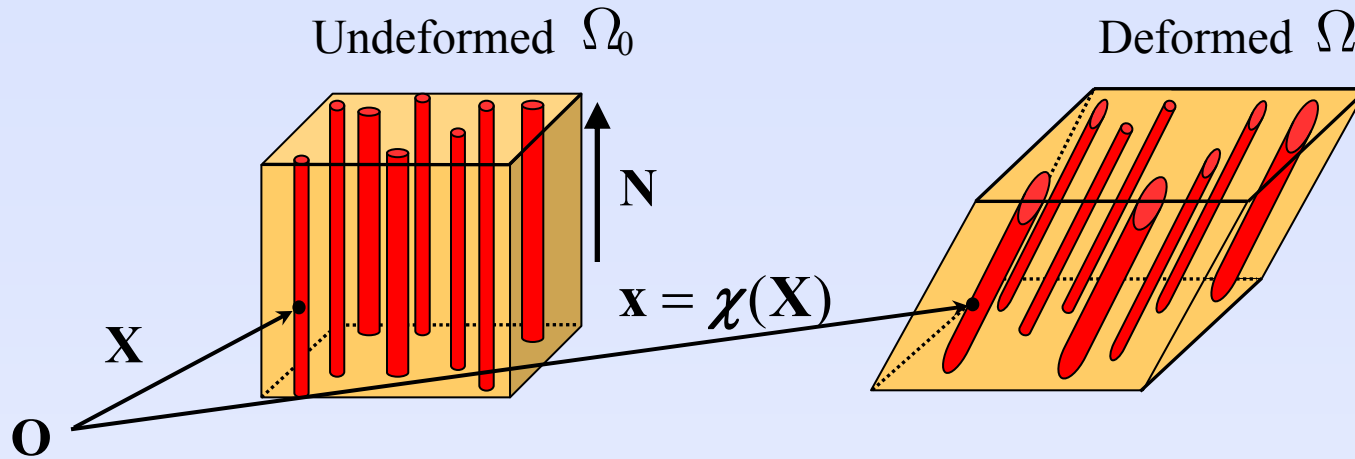
$$W^{(2)}(\mathbf{F})$$

Random variable characterizing the
microstructure

$$\chi_0^{(r)}(\mathbf{X}) = \begin{cases} 1 & \text{if } \mathbf{X} \in \text{phase } r \\ 0 & \text{otherwise} \end{cases}$$

Problem setting: Lagrangian formulation

Kinematics



**Deformation
Gradient**

$$\mathbf{F} = \frac{\partial \mathbf{x}}{\partial \mathbf{X}}$$

**Right CG
Deformation Tensor**

$$\mathbf{C} = \mathbf{F}^T \mathbf{F}$$

Local constitutive behavior

$$\mathbf{S} = \frac{\partial W}{\partial \mathbf{F}}(\mathbf{X}, \mathbf{F})$$

Isotropic matrix

$$W^{(1)}(\mathbf{F}) = \Psi^{(1)}(I_1, I_2)$$

Transversely isotropic fibers

$$W^{(2)}(\mathbf{F}) = \Psi^{(2)}(I_1, I_2, I_4, I_5)$$

where

$$W(\mathbf{X}, \mathbf{F}) = \sum_{r=1}^2 \chi_0^{(r)}(\mathbf{X}) W^{(r)}(\mathbf{F})$$

where

$$I_1 = \text{tr} \mathbf{C} \quad I_2 = \frac{1}{2} \left[(\text{tr} \mathbf{C})^2 - \text{tr} \mathbf{C}^2 \right]$$

$$I_4 = \mathbf{N} \cdot \mathbf{C} \mathbf{N} \quad I_5 = \mathbf{N} \cdot \mathbf{C}^2 \mathbf{N}$$

Problem setting: macroscopic response

Definition: relation between the volume averages of
the stress and deformation gradient over RVE

$$\bar{\mathbf{S}} = \frac{1}{\Omega_0} \int_{\Omega_0} \mathbf{S} \, d\mathbf{X} \quad \bar{\mathbf{F}} = \frac{1}{\Omega_0} \int_{\Omega_0} \mathbf{F} \, d\mathbf{X}$$

Variational Characterization

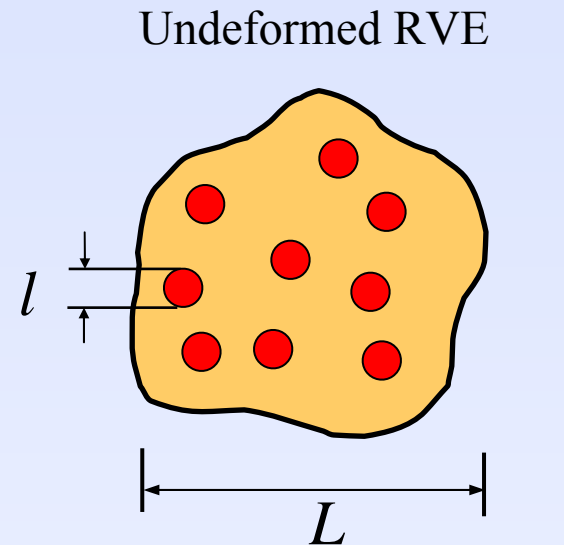
$$\bar{\mathbf{S}} = \frac{\partial \bar{W}}{\partial \bar{\mathbf{F}}}(\bar{\mathbf{F}})$$

where

$$\bar{W}(\bar{\mathbf{F}}) = \min_{\mathbf{F} \in K(\bar{\mathbf{F}})} \frac{1}{\Omega_0} \int_{\Omega_0} W(\mathbf{X}, \mathbf{F}) \, d\mathbf{X}$$

and

$$K(\bar{\mathbf{F}}) = \left\{ \mathbf{F} \mid \mathbf{F} = \nabla \mathbf{x} \text{ in } \Omega_0, \text{ and } \mathbf{x} = \bar{\mathbf{F}}\mathbf{X} \text{ on } \partial\Omega_0 \right\}$$



separation of
length-scales $\frac{l}{L} \ll 1$

Existing analytical approaches

- **Phenomenological models**

Formulated on the basis of invariants

$$\bar{W}(\bar{\mathbf{F}}) = F_{iso}(\bar{I}_1, \bar{I}_2) + G_{ani}(\bar{I}_4, \bar{I}_5)$$

Merodio, Horgan, Ogden,

Pence, Rivlin, Saccomandi,

among others

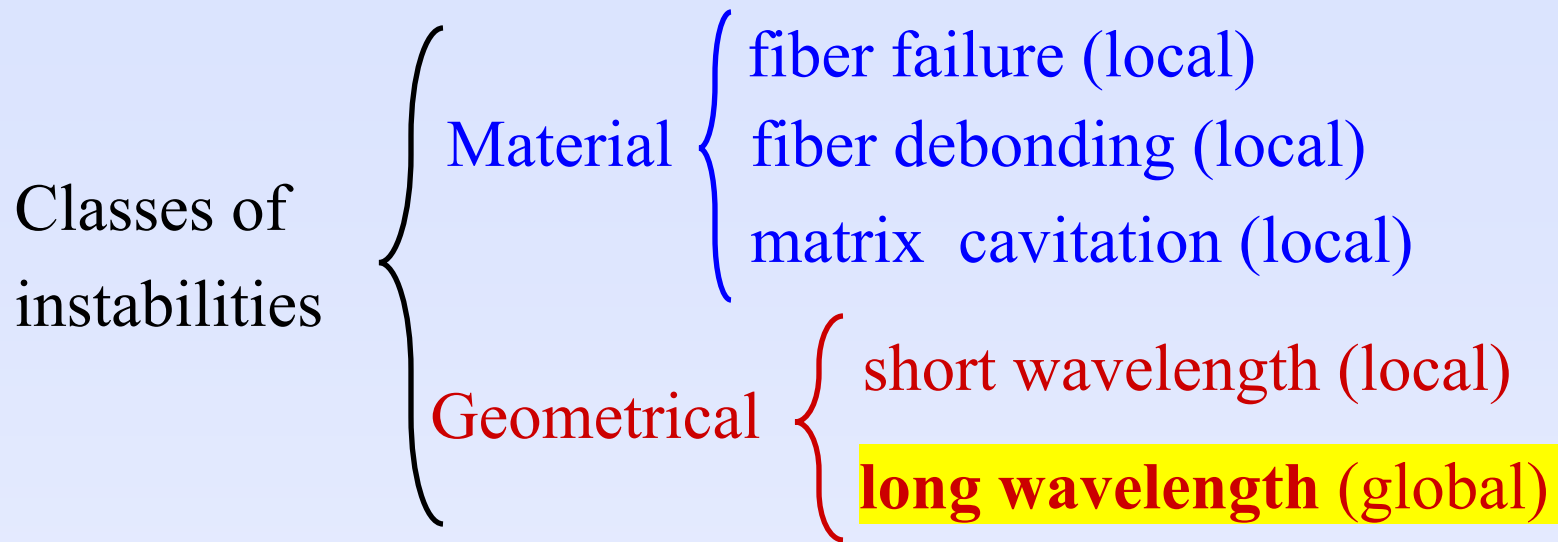
- **Homogenization/Micromechanics models**

Incorporate direct information from microscopic properties

– Estimates for special loading conditions, and special matrix and fiber constituents (He et al., 2006; deBotton et al., 2006)

– “Linear comparison” variational estimates for general loading conditions and isotropic constituents (LP & Ponte Castañeda 2006a,b)

Stability and failure



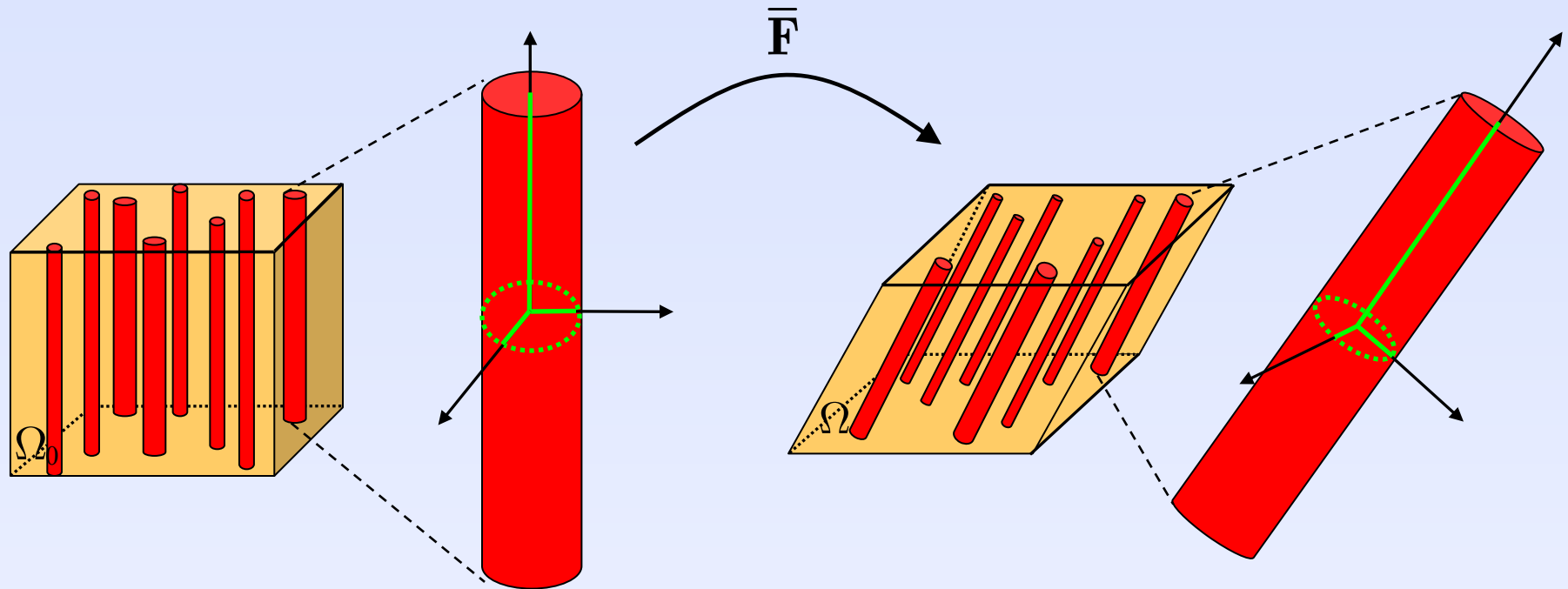
- The loss of strong ellipticity of the macroscopic response of the fiber-reinforced elastomer, as characterized by the effective stored-energy function $\bar{W}(\bar{\mathbf{F}})$, denotes the onset of long wavelength instabilities

$$B(\bar{\mathbf{F}}) = \min_{\substack{\mathbf{u}, \mathbf{v} \\ \|\mathbf{u}\| = \|\mathbf{v}\| = 1}} \left\{ v_i v_k \frac{\partial^2 \bar{W}}{\partial \bar{F}_{ij} \partial \bar{F}_{kl}}(\bar{\mathbf{F}}) u_j u_l \right\} = 0$$

Microstructure evolution

Undeformed

Deformed



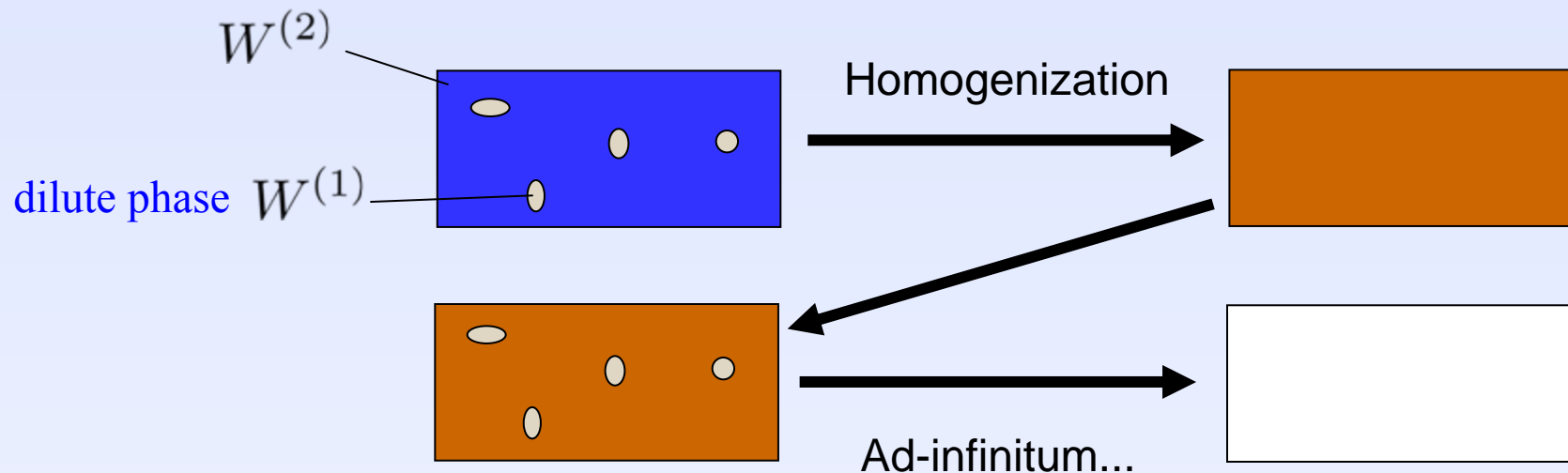
Information on microstructure evolution is important to identify and understand the **microscopic mechanisms** that govern the macroscopic behavior. For that we need information about the **local fields** $\mathbf{F}(\mathbf{X})$

**New Approach:
Iterated Homogenization**

Iterated dilute homogenization

Strategy: Construct a particulate distribution of fibers ($\chi_0^{(r)}(\mathbf{X})$) within a hyperelastic material for which it is possible to compute exactly the effective stored-energy function \bar{W}

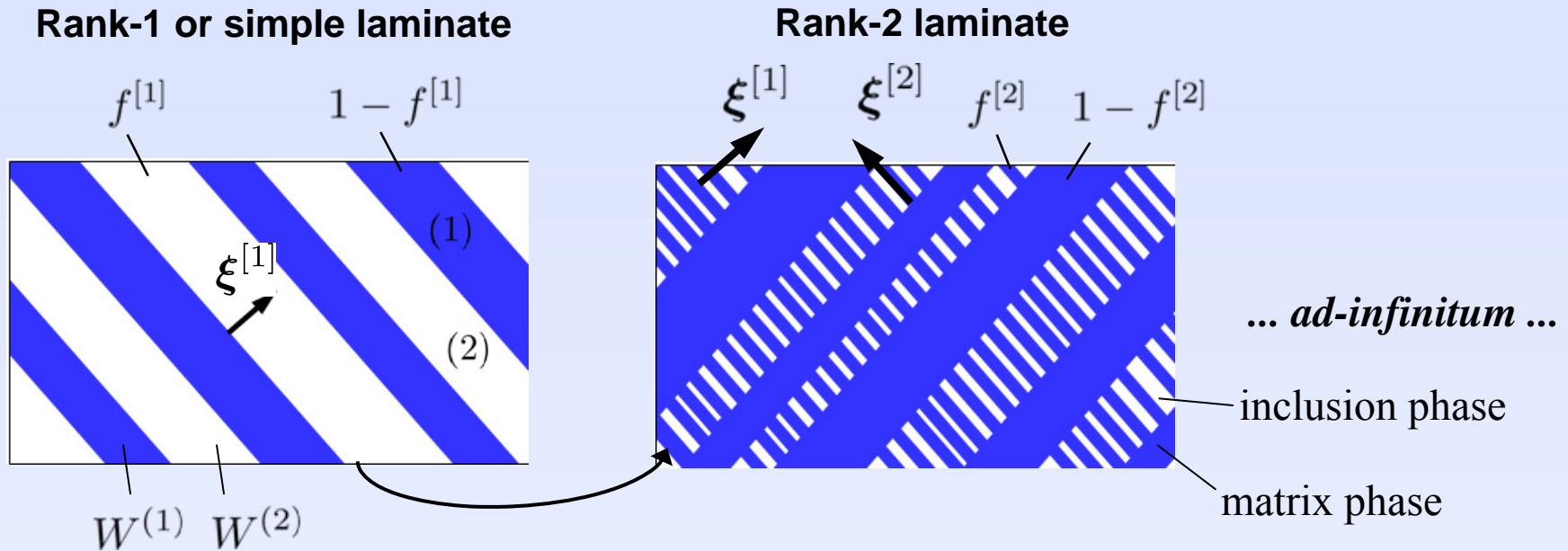
Step 1: iterated-dilute homogenization



$$c_0 \frac{\partial \bar{W}}{\partial c_0} - H \left[W^{(1)}, \bar{W}; \bar{\mathbf{F}} \right] = 0, \quad \bar{W}(\bar{\mathbf{F}}, 1) = W^{(2)}$$

Auxiliary dilute problem: sequential laminates

Step 2: sequential laminates



When the **matrix** phase is dilute:

$$H \left[W^{(1)}, \bar{W}, \bar{\mathbf{F}} \right] = \bar{W} + \max_{\omega} \int_S \left[\omega \cdot \frac{\partial \bar{W}}{\partial \bar{\mathbf{F}}} \xi - W^{(1)} \left(\bar{\mathbf{F}} + \omega \otimes \xi \right) \right] \underbrace{\nu(\xi)}_{\text{distributional function}} dS$$

distributional function related to the two-point statistics of fiber distribution in the undeformed configuration

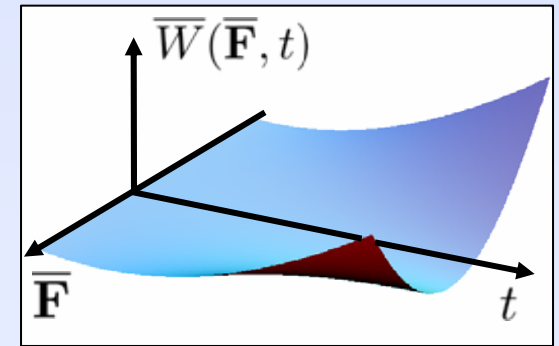
Iterated homogenization framework

- The **effective stored-energy function** \bar{W} can be finally shown to be given by the following **Hamilton-Jacobi equation**

$$c_0 \frac{\partial \bar{W}}{\partial c_0} - \bar{W} - \max_{\omega} \int_S \left[\omega \cdot \frac{\partial \bar{W}}{\partial \bar{\mathbf{F}}} \xi - W^{(1)}(\bar{\mathbf{F}} + \omega \otimes \xi) \right] \nu(\xi) dS = 0$$

subject to the initial condition

$$\bar{W}(\bar{\mathbf{F}}, 1) = W^{(2)}(\bar{\mathbf{F}})$$



the **time** variable is $t \doteq -\ln c_0$ (initial fiber concentration)

the **spatial** variable is $\bar{\mathbf{F}}$

the **Hamiltonian** is

$$H \left[W^{(1)}, \bar{W}, \bar{\mathbf{F}} \right] = \bar{W} + \max_{\omega} \int_S \left[\omega \cdot \frac{\partial \bar{W}}{\partial \bar{\mathbf{F}}} \xi - W^{(1)}(\bar{\mathbf{F}} + \omega \otimes \xi) \right] \nu(\xi) dS$$

Iterated homogenization framework: local fields

- Consider the following **perturbed problem** for \bar{W}_τ

$$c_0 \frac{\partial \bar{W}_\tau}{\partial c_0} - \bar{W}_\tau - \max_{\boldsymbol{\omega}} \int_S \left[\boldsymbol{\omega} \cdot \frac{\partial \bar{W}_\tau}{\partial \bar{\mathbf{F}}} \boldsymbol{\xi} - W^{(1)}(\bar{\mathbf{F}} + \boldsymbol{\omega} \otimes \boldsymbol{\xi}) \right] \nu(\boldsymbol{\xi}) \, dS = 0$$

subject to the initial condition

$$\bar{W}(\bar{\mathbf{F}}, 1) = W^{(2)}(\bar{\mathbf{F}}) + \tau U(\bar{\mathbf{F}})$$

Then, the following identity is true

$$\frac{1}{\Omega_0^{(2)}} \int_{\Omega_0^{(2)}} U(\mathbf{F}(\mathbf{X})) \, d\mathbf{X} = \frac{1}{c_0} \frac{\partial \bar{W}_\tau}{\partial \tau} \Big|_{\tau=0}$$

$U(\cdot)$ can be any function of interest, e.g., the deformation gradient

Remarks on the iterated homogenization approach

- The proposed IH method provides solutions for \bar{W} in terms of $W^{(1)}$ and $W^{(2)}$ and the **one-** and **two-point statistics** of the random distribution of fibers
- In the limit of small deformations as $\bar{\mathbf{F}} \rightarrow \mathbf{I}$, the IH formulation **reduces to the HS lower bound** for fiber-reinforced random media
- In the further limit of dilute fiber concentration $c_0 \rightarrow 0$, the IH formulation recovers the **exact result of Eshelby** for a dilute distribution of ellipsoidal fibers
- The proposed IH method provides access to **local fields**, which in turn permits the study of the **evolution of microstructure** and the **onset of instabilities**
- The computations amount to solving appropriate **Hamilton-Jacobi equations**, which are fairly tractable

**Application to
Fiber-Reinforced
Neo-Hookean Solids**

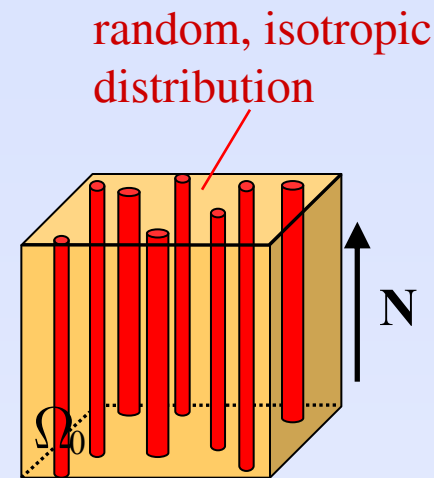
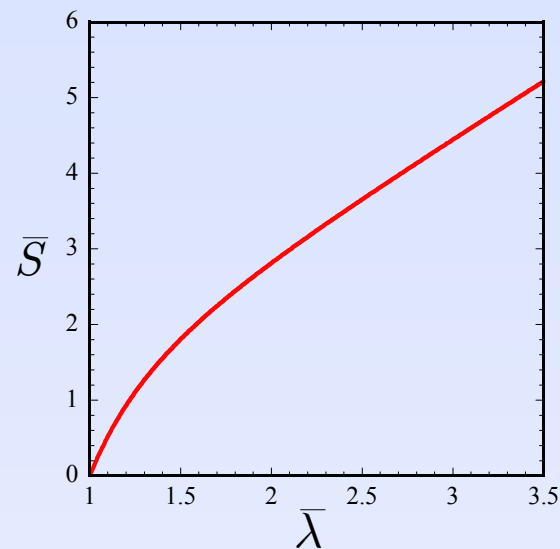
Fiber-reinforced Neo-Hookean solids

Neo-Hookean matrix

$$W^{(1)}(\mathbf{F}) = \Psi^{(1)}(I_1) = \frac{\mu^{(1)}}{2} (I_1 - 3)$$

Stiffer Neo-Hookean fibers

$$W^{(2)}(\mathbf{F}) = \Psi^{(2)}(I_1) = \frac{\mu^{(2)}}{2} (I_1 - 3)$$



Macroscopic stored-energy function

$$\bar{W}(\bar{\mathbf{F}}, c_0) = \bar{\Psi}(\bar{I}_1, \bar{I}_2, \bar{I}_4, \bar{I}_5, c_0)$$

Hamilton-Jacobi equation

$$c_0 \frac{\partial \bar{\Psi}}{\partial c_0} + \frac{\mu^{(1)}}{2} (\bar{I}_1 - 3) - \bar{\Psi} + \mu^{(1)} \left(\bar{I}_1 - \bar{I}_4 - \frac{2}{\sqrt{\bar{I}_4}} \right) \left(\frac{1}{2} - \frac{1}{\mu^{(1)}} \frac{\partial \bar{\Psi}}{\partial \bar{I}_1} \right)^2 = 0$$

subject to the initial condition $\bar{\Psi}(\bar{I}_1, \bar{I}_2, \bar{I}_4, \bar{I}_5, 1) = \Psi^{(2)}(\bar{I}_1, \bar{I}_4)$

Overall stress-strain relation

Closed-form solution for \bar{W}

$$\bar{W}(\bar{\mathbf{F}}) = \bar{\Psi}(\bar{I}_1, \bar{I}_4) = f(\bar{I}_1) + g(\bar{I}_4)$$

where

$$f(\bar{I}_1) = \frac{\tilde{\mu}}{2}(\bar{I}_1 - 3) \quad \text{and}$$

$$g(\bar{I}_4) = \frac{\bar{\mu} - \tilde{\mu}(\sqrt{\bar{I}_4} + 2)(\sqrt{\bar{I}_4} - 1)}{2\sqrt{\bar{I}_4}}$$

with

$$\bar{\mu} = (1 - c_0)\mu^{(1)} + c_0\mu^{(2)} \quad \text{and}$$

$$\tilde{\mu} = \frac{(1 - c_0)\mu^{(1)} + (1 + c_0)\mu^{(2)}}{(1 + c_0)\mu^{(1)} + (1 - c_0)\mu^{(2)}} \mu^{(1)}$$

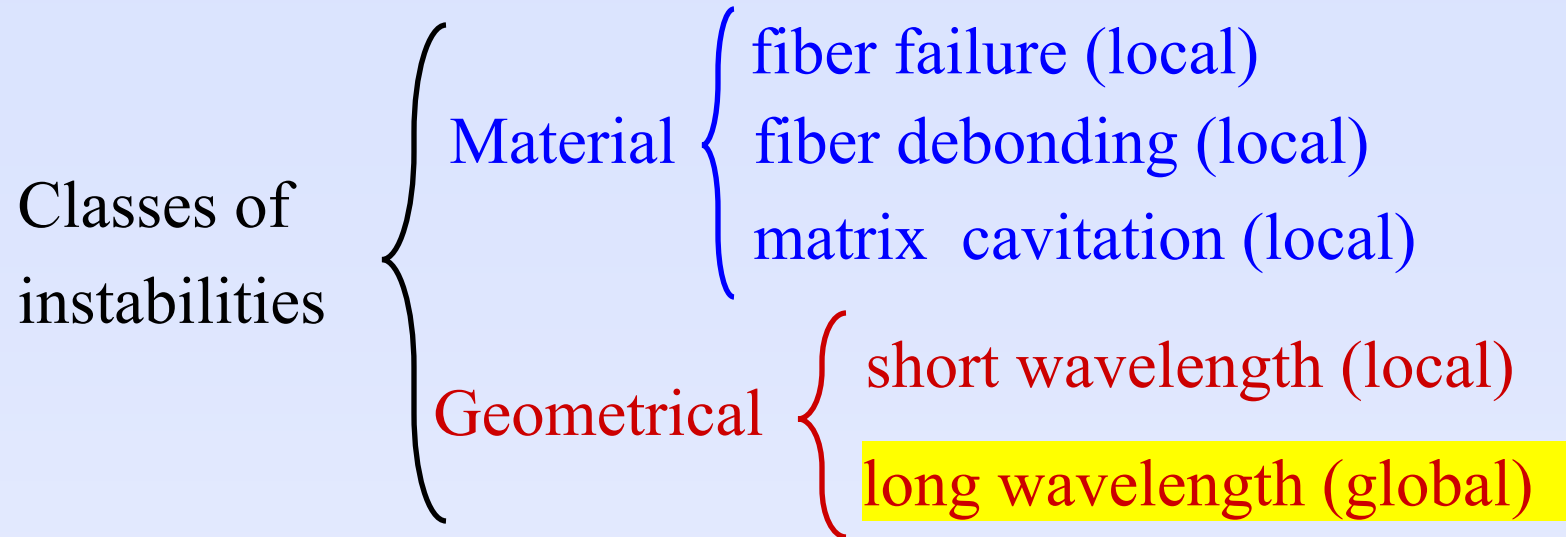
Note I: separable functional form

Note II: no dependence on the second \bar{I}_2 nor fifth \bar{I}_5 invariants

Overall stress-strain relation

$$\bar{\mathbf{S}} = \frac{\partial \bar{W}}{\partial \bar{\mathbf{F}}}(\bar{\mathbf{F}}) - \bar{p}\bar{\mathbf{F}}^{-T} = \tilde{\mu}\bar{\mathbf{F}} + (\bar{\mu} - \tilde{\mu})\left[1 - \bar{I}_4^{-3/2}\right]\bar{\mathbf{F}}\mathbf{N} \otimes \mathbf{N} - \bar{p}\bar{\mathbf{F}}^{-T}$$

Macroscopic Instabilities



Along an arbitrary loading path, the material first becomes unstable at a critical deformation $\bar{\mathbf{F}}_{cr}$ with $\bar{I}_4^{cr} = \bar{\mathbf{F}}_{cr} \mathbf{N} \cdot \bar{\mathbf{F}}_{cr} \mathbf{N}$ such that

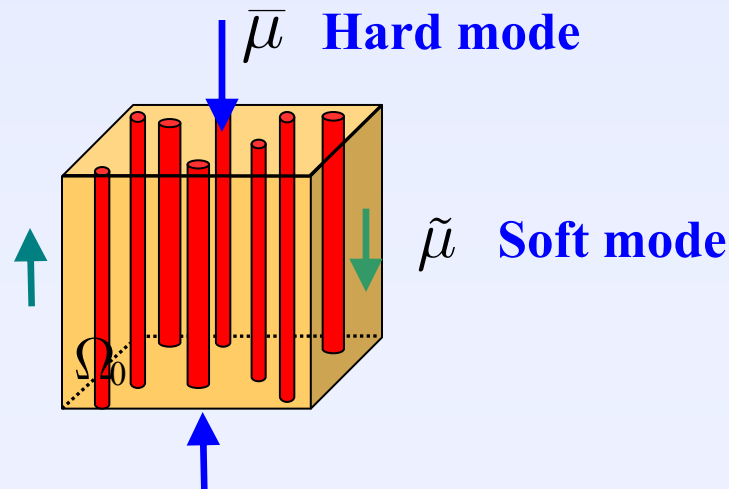
$$\bar{I}_4^{cr} = \left(1 - \frac{\tilde{\mu}}{\bar{\mu}} \right)^{2/3}$$

Note: $\tilde{\mu} \leq \bar{\mu} \Rightarrow \bar{I}_4^{cr} \leq 1$

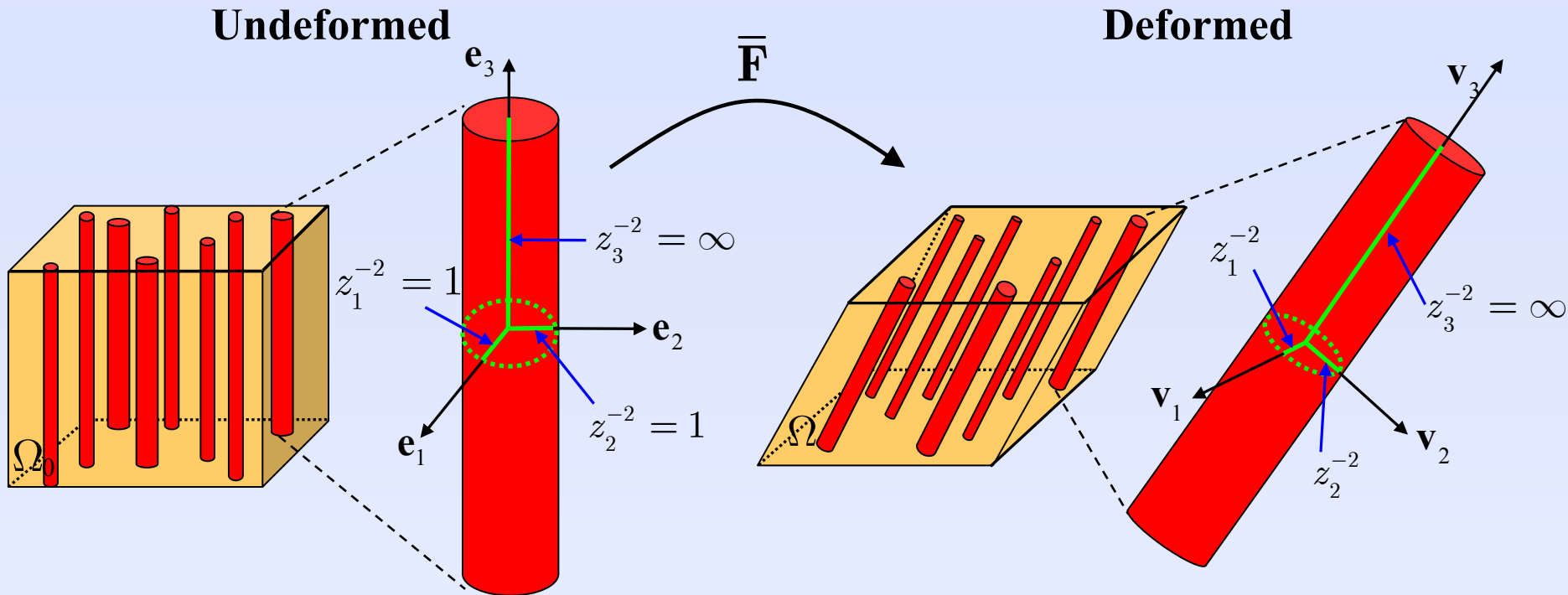
Macroscopic Instabilities

Observations

- Macroscopic instabilities may only occur when the deformation in the fiber direction, as measured by \bar{I}_4 , reaches a sufficiently large **compressive** value $\bar{I}_4^{cr} \leq 1$
- The condition states that instabilities may develop whenever the compressive deformation along the fiber direction reaches a critical value determined by the ratio of **hard-to-soft** modes of deformation



Microstructure evolution



The **average shape** and **orientation** of the fibers in the deformed configuration are characterized by the Eulerian ellipsoid

$$E = \left\{ \mathbf{x} \mid \mathbf{x} \cdot \mathbf{Z}^T \mathbf{Z} \mathbf{x} \leq 1 \right\}$$

where

$$\mathbf{Z} = \left(\mathbf{I} - \mathbf{N} \otimes \mathbf{N} \right) \left(\bar{\mathbf{F}}^{(2)} \right)^{-1}$$

Eigenvalues of $\mathbf{Z}^T \mathbf{Z}$

$$z_1, z_2, z_3$$

Eigenvectors of $\mathbf{Z}^T \mathbf{Z}$

$$\mathbf{v}_1, \mathbf{v}_2, \mathbf{v}_3$$

Microstructure evolution

$\bar{\mathbf{F}}^{(2)}$ is the **average deformation gradient in the fibers**. In the IH framework, it is solution of the pde

$$c_0 \frac{\partial \bar{\mathbf{F}}^{(2)}}{\partial c_0} - \frac{\partial \bar{\mathbf{F}}^{(2)}}{\partial \bar{\mathbf{F}}} \int_S \boldsymbol{\omega} \otimes \boldsymbol{\xi} \, dS = 0$$

subject to the initial condition $\bar{\mathbf{F}}^{(2)}(\bar{\mathbf{F}}, 1) = \bar{\mathbf{F}}$

Closed-form solution

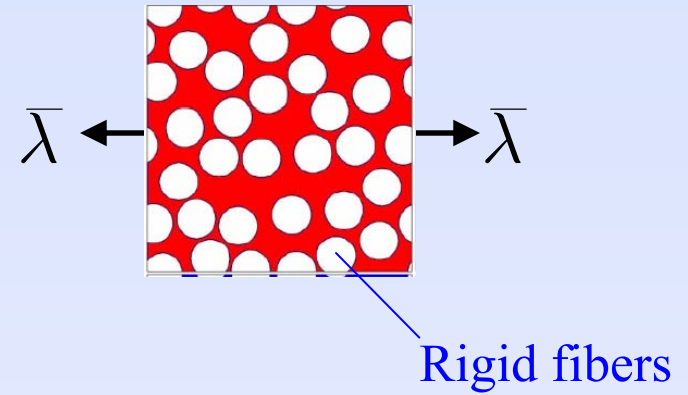
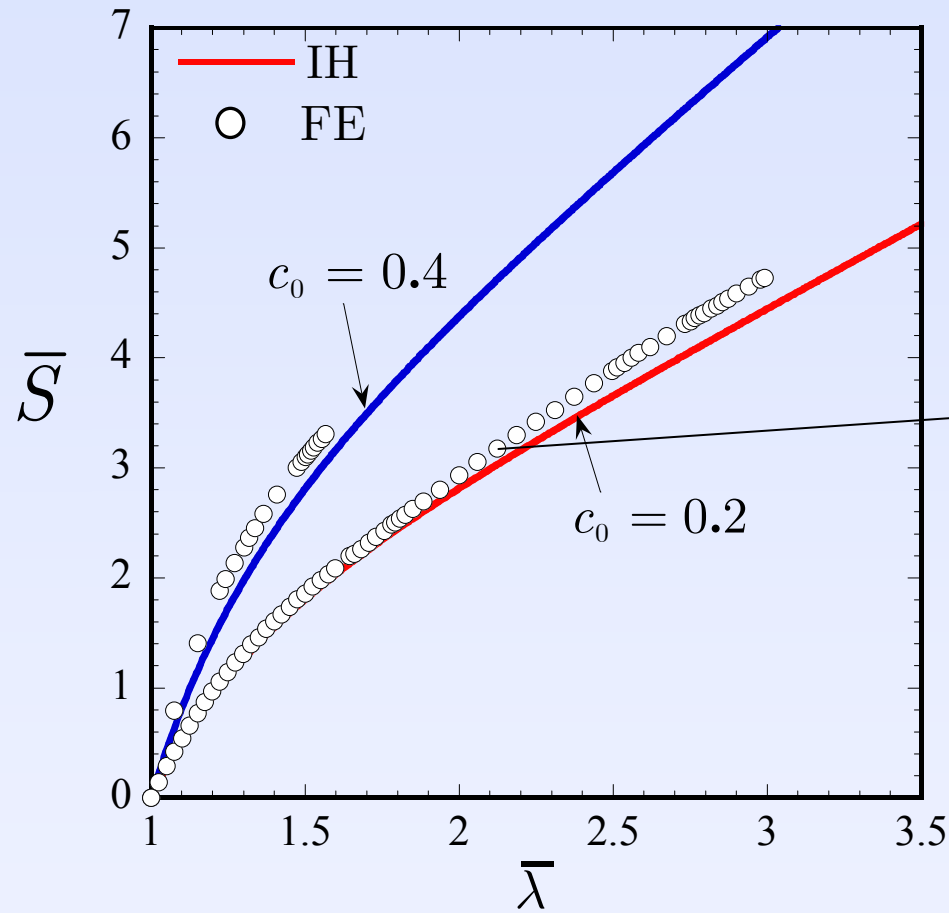
$$\bar{\mathbf{F}}^{(2)} = \gamma_1 [\bar{\mathbf{F}} - \bar{\mathbf{F}}\mathbf{N} \otimes \mathbf{N}] - \frac{2\bar{\nu} - \gamma_1}{\sqrt{\bar{I}_4}} [\bar{\mathbf{F}}^{-T} - \bar{\mathbf{F}}^{-T}\mathbf{N} \otimes \mathbf{N}] + \frac{2\bar{\nu} - \gamma_1}{\sqrt{\bar{I}_4}} \bar{\mathbf{F}}\mathbf{N} \otimes \mathbf{u} + \bar{\mathbf{F}}\mathbf{N} \otimes \mathbf{N}$$

where $\mathbf{u} = (\mathbf{I} - \mathbf{N} \otimes \mathbf{N})\bar{\mathbf{F}}^T \bar{\mathbf{F}}\mathbf{N}$, and

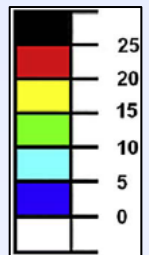
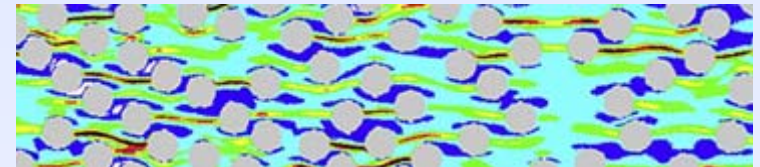
$$\gamma_1 = \bar{\nu} + \frac{\sqrt{\sqrt{\bar{I}_4} + \bar{\nu}^2 (\bar{I}_1 \bar{I}_4 - \bar{I}_5 - 2\sqrt{\bar{I}_4})}}{\sqrt{\bar{I}_1 \bar{I}_4 - \bar{I}_5 + 2\sqrt{\bar{I}_4}}}, \quad \bar{\nu} = \frac{\mu^{(1)}}{(1 + c_0)\mu^{(1)} + (1 - c_0)\mu^{(2)}}$$

Sample Results

IH vs. FEM: in-plane stress-strain response



$\bar{\lambda} = 2.1$



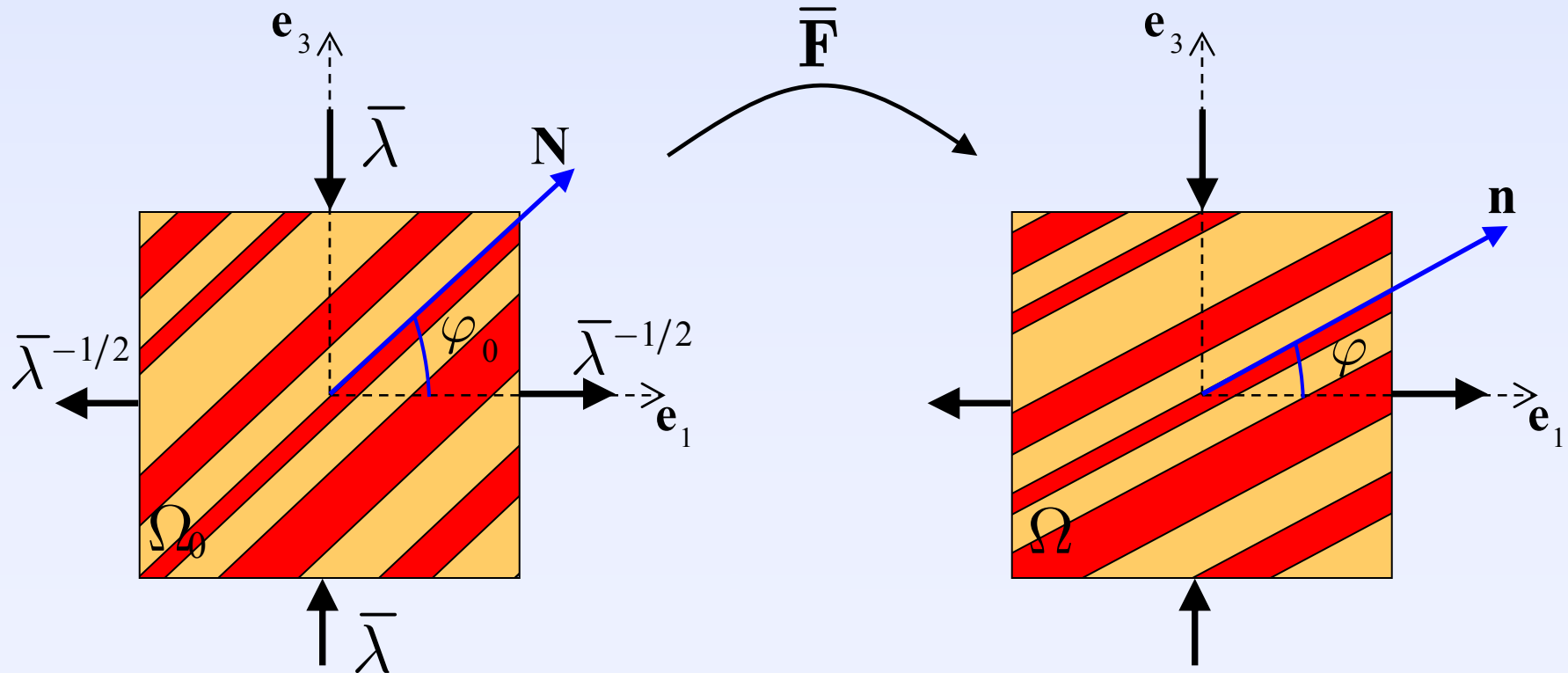
Axisymmetric compression at an angle φ_0

Applied Loading

$$\bar{\mathbf{F}} = \begin{pmatrix} \bar{\lambda}^{-1/2} & 0 & 0 \\ 0 & \bar{\lambda}^{-1/2} & 0 \\ 0 & 0 & \bar{\lambda} \end{pmatrix}$$

Initial fiber orientation

$$\mathbf{N} = \cos\varphi_0 \mathbf{e}_1 + \sin\varphi_0 \mathbf{e}_3$$



Axisymmetric compression at an angle φ_0

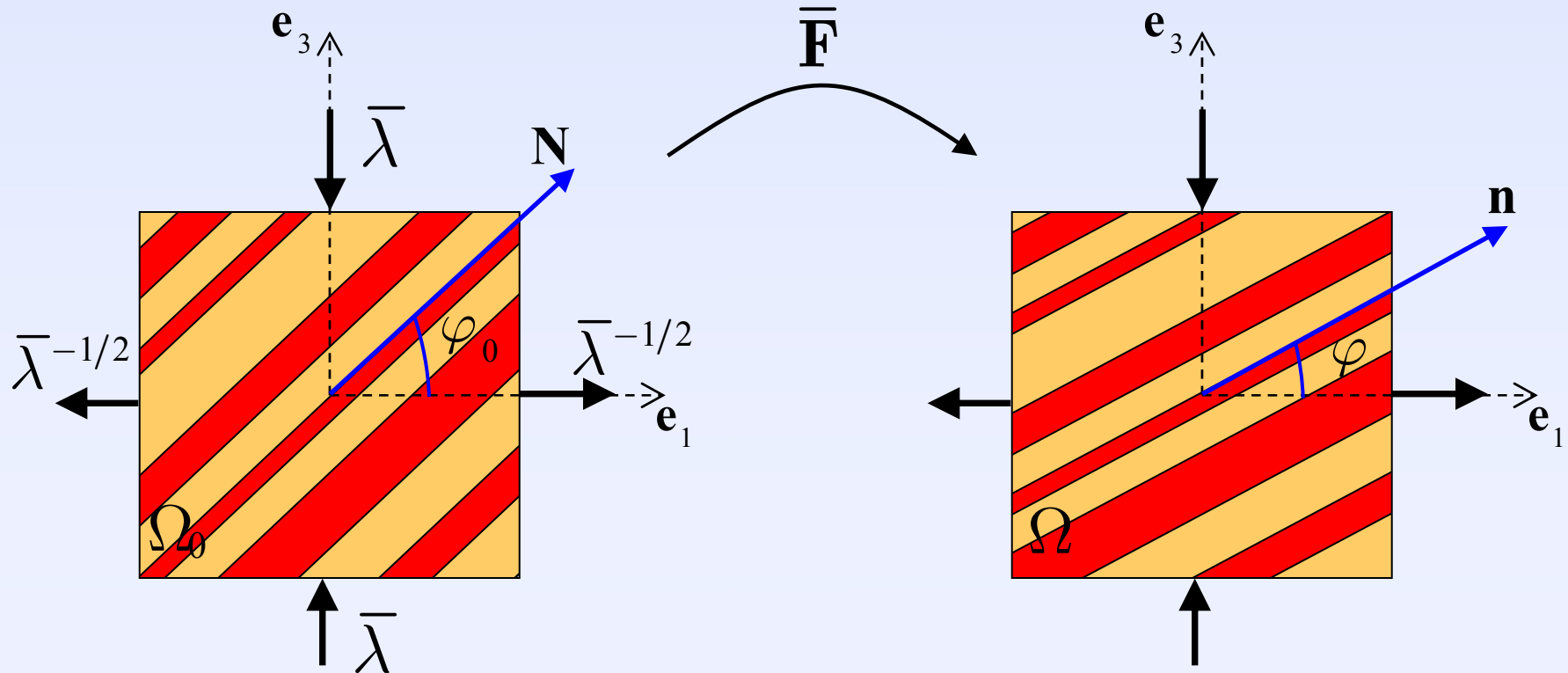
Applied Loading

$$\bar{\mathbf{F}} = \begin{pmatrix} \bar{\lambda}^{-1/2} & 0 & 0 \\ 0 & \bar{\lambda}^{-1/2} & 0 \\ 0 & 0 & \bar{\lambda} \end{pmatrix}$$

Current fiber orientation

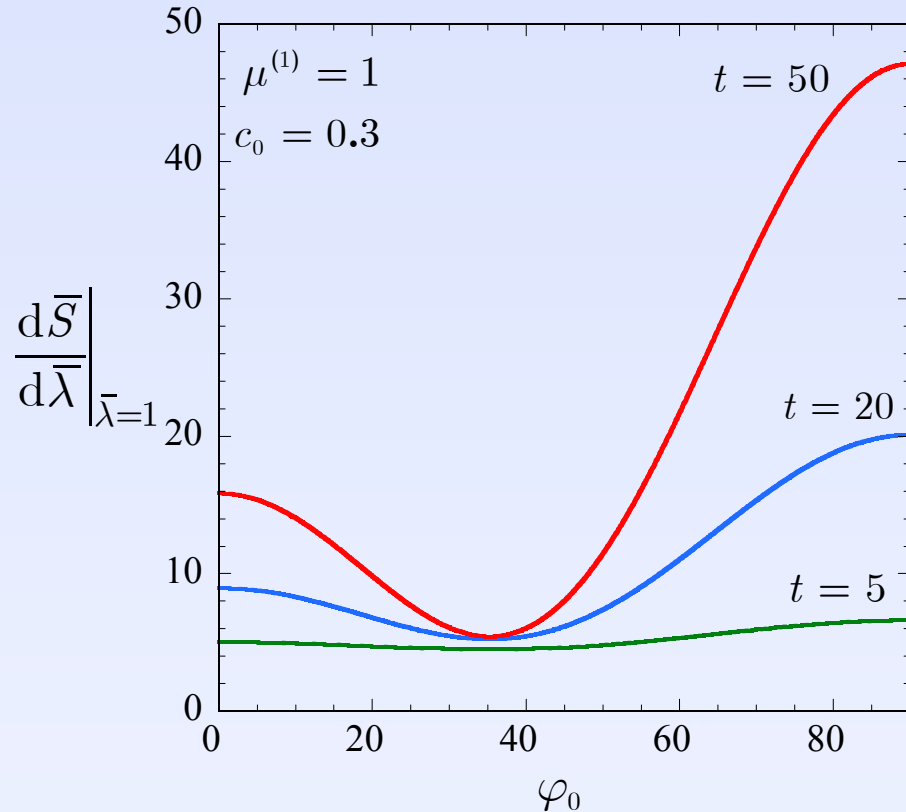
$$\mathbf{n} = \cos\varphi \mathbf{e}_1 + \sin\varphi \mathbf{e}_3$$

$$\varphi = \text{Arcos} \left[\frac{\cos \varphi_0}{\sqrt{\cos^2 \varphi_0 + \bar{\lambda}^3 \cos^2 \varphi_0}} \right]$$

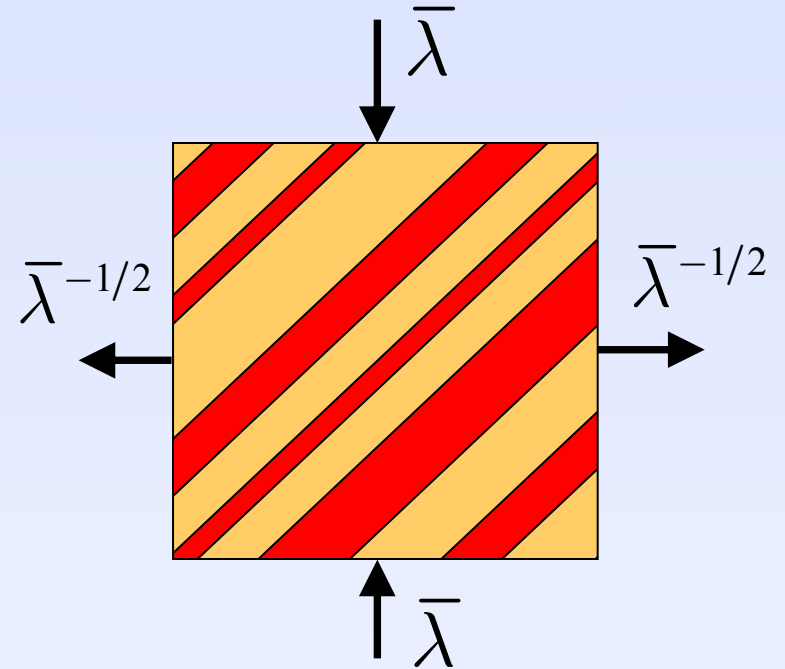


Axisymmetric compression at an angle φ_0

Small-deformation response

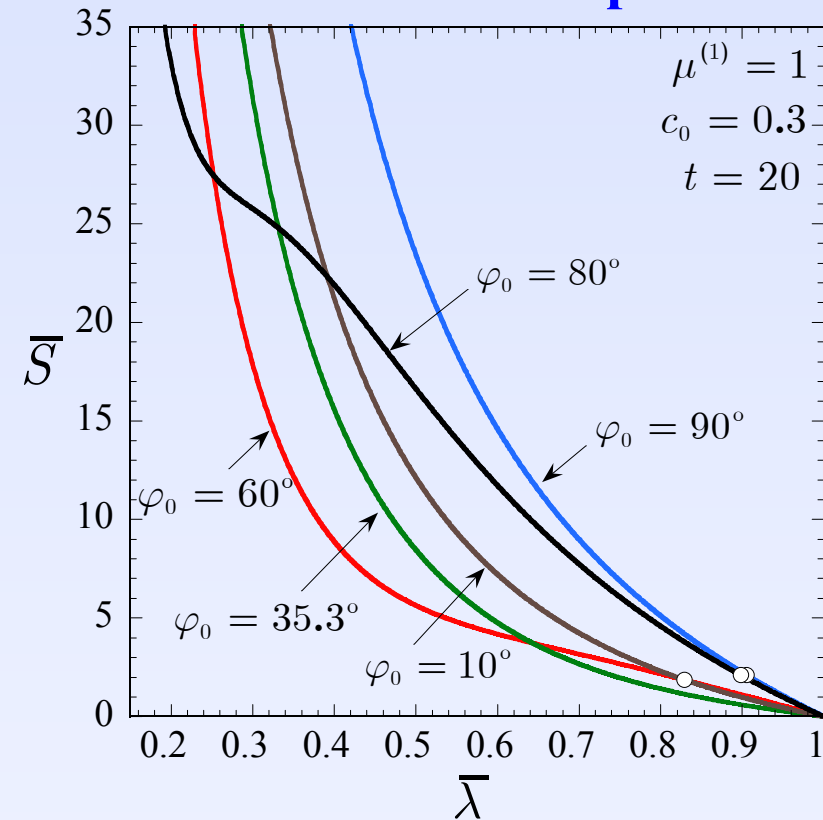


$$t \doteq \frac{\mu^{(2)}}{\mu^{(1)}} \quad \text{heterogeneity contrast}$$

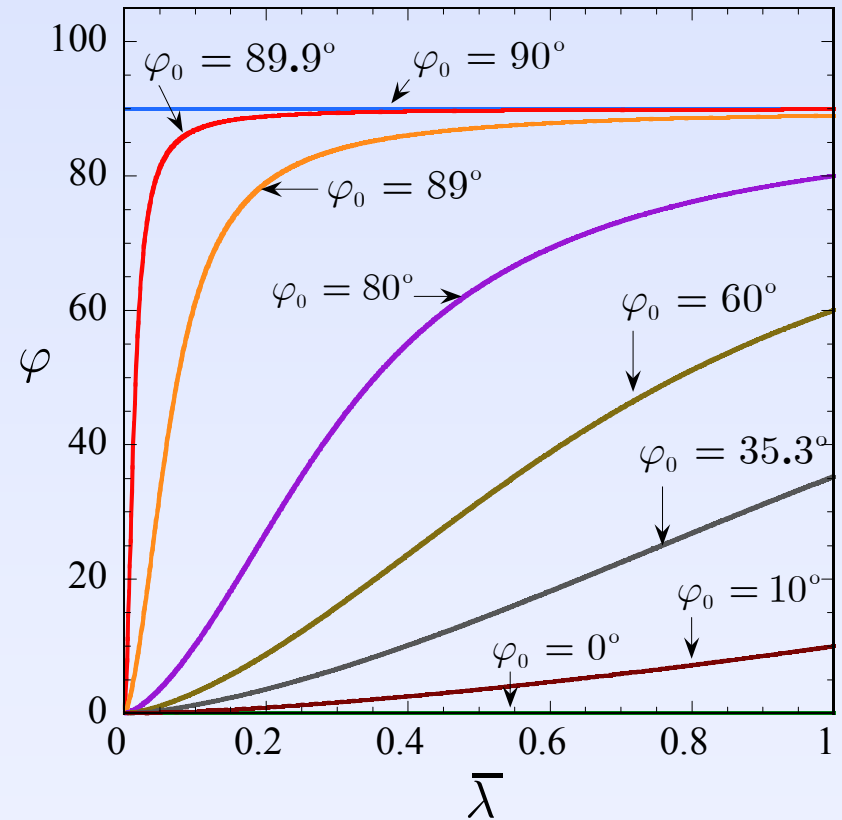


Axisymmetric compression at an angle φ_0

Large-deformation stress-strain response



Evolution of fiber orientation

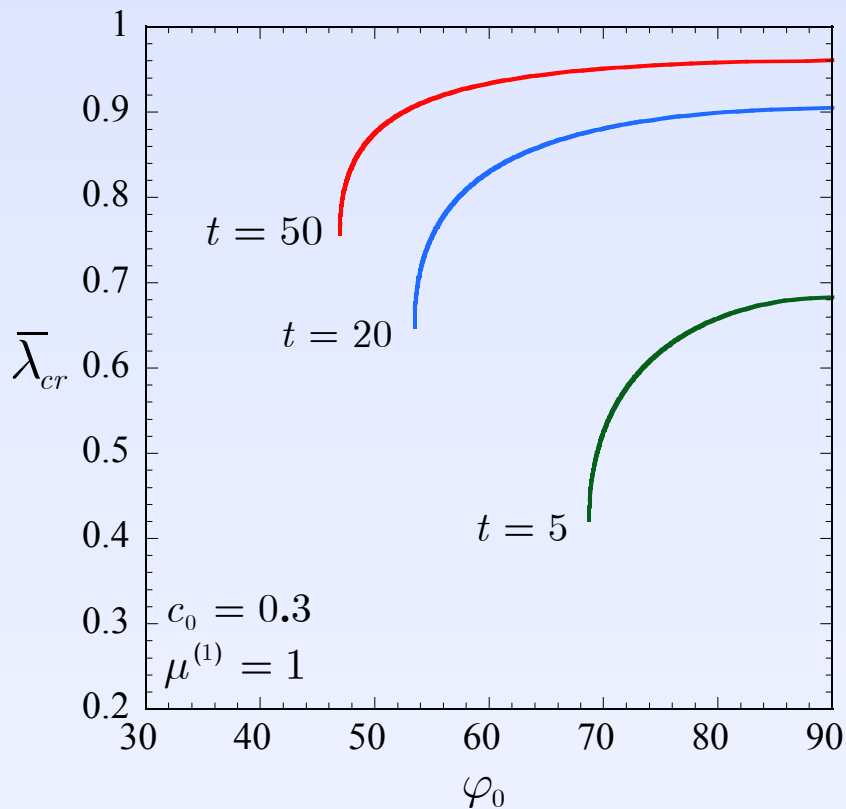


Note: Macroscopic instability at \circ

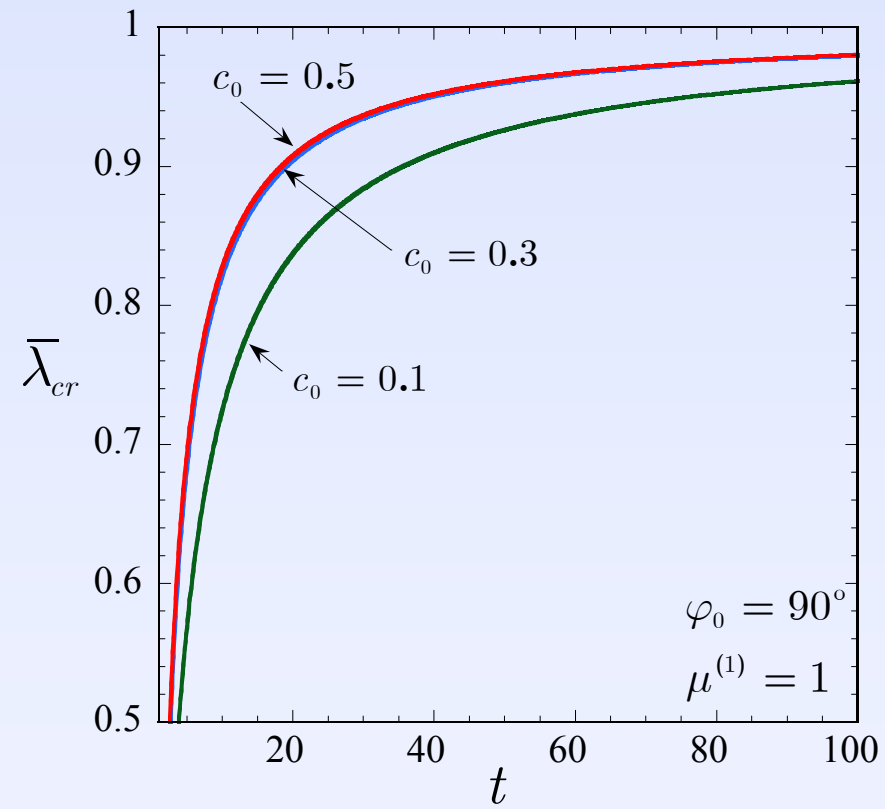
Axisymmetric compression at an angle φ_0

Onset of macroscopic instabilities at $\bar{\lambda}_{cr}$

Effect of fiber orientation



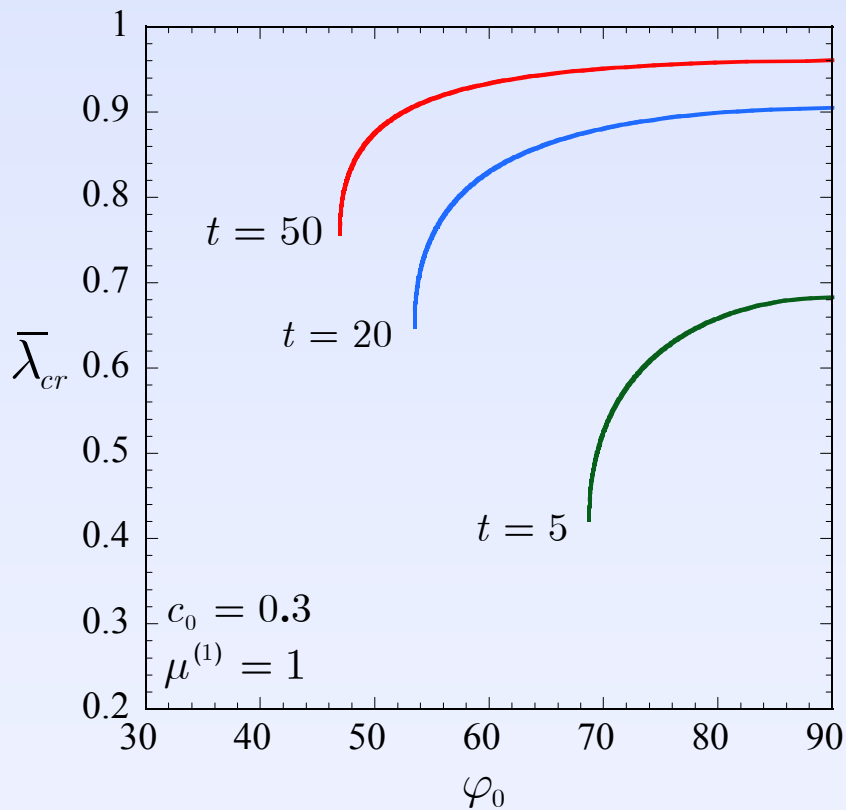
Effect of fiber-to matrix contrast



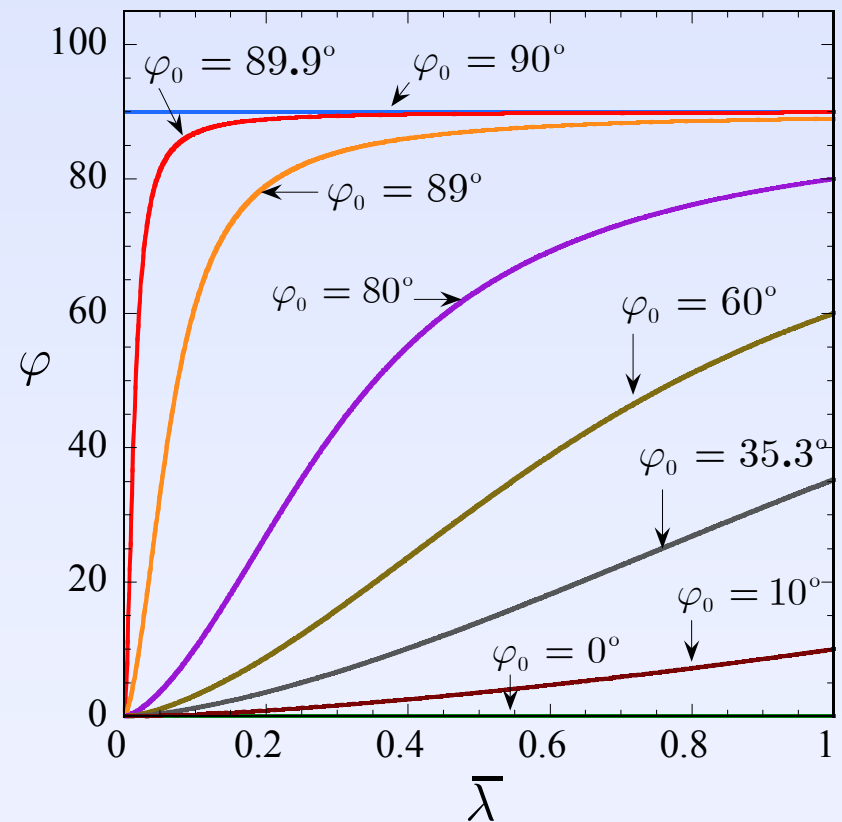
Axisymmetric compression at an angle φ_0

Onset of macroscopic instabilities at $\bar{\lambda}_{cr}$

Effect of fiber orientation



Evolution of fiber orientation



Remarks

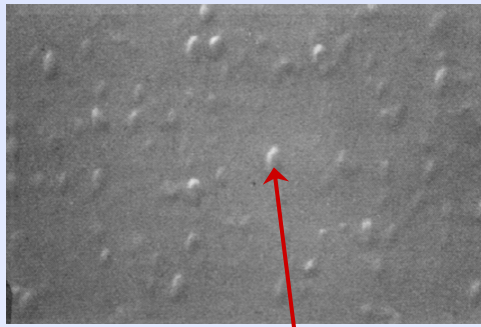
The **rotation of the fibers** — which depends critically on the relative orientation between the loading axes and the fiber direction — can act as a dominant **geometric softening mechanism**.

- It was found that the long axes of **the fibers rotate away from the axis of maximum compressive loading** towards the axis of maximum tension.
- Loadings with predominant compression along the fibers lead to larger rotation of the fibers, which in turn lead to larger geometric **softening of the constitutive response**, and in some cases — when the heterogeneity contrast between the matrix and the fibers is sufficiently high — also to the **loss of macroscopic stability**.

Remarks

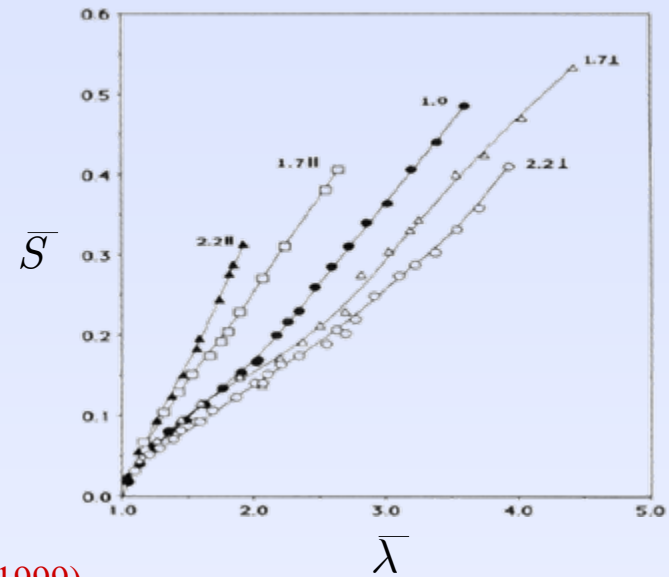
The results of this work can help understanding the behavior of many **other solids with oriented microstructures**

Reinforced Elastomers (Wang and Mark 1990)



2 μm

Polystyrene Ellipsoids

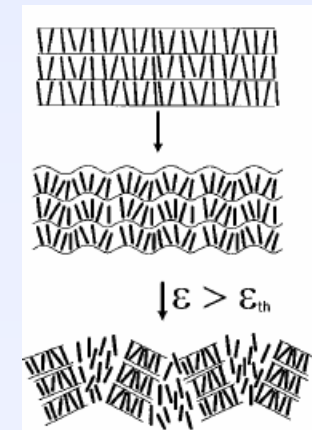
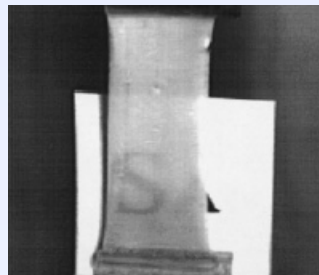


Liquid crystal elastomers (Nishikawa and Finkelmann 1999)

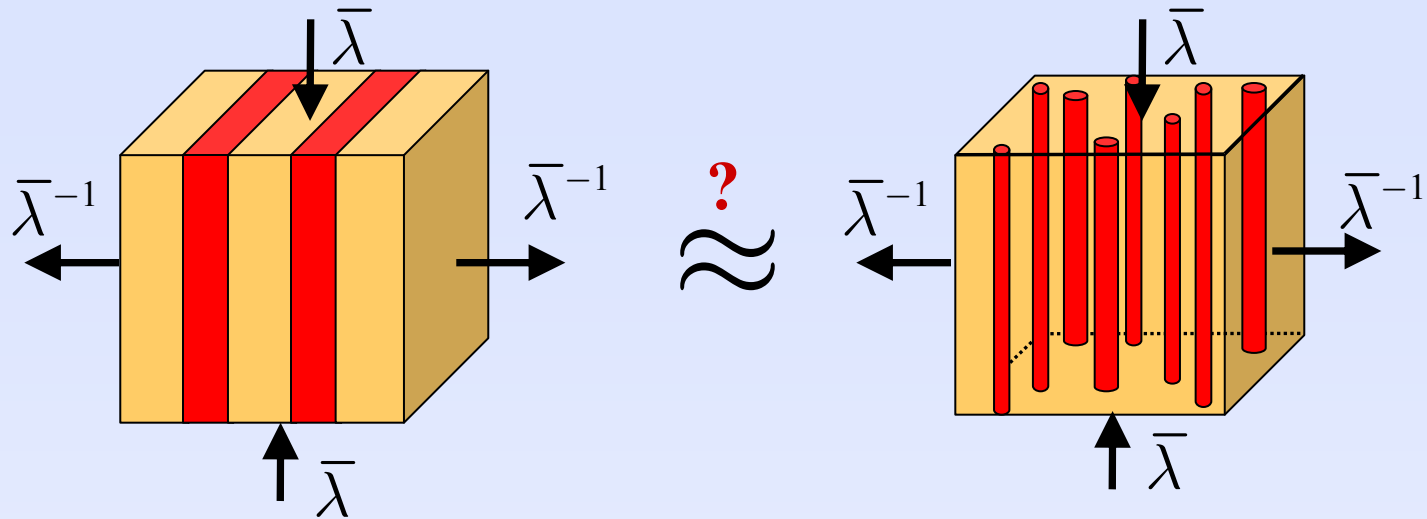
Transparent



Opaque



Contact with earlier work with laminates



- For the **aligned plane-strain loading of a laminate** the onset of macroscopic instabilities occurs at

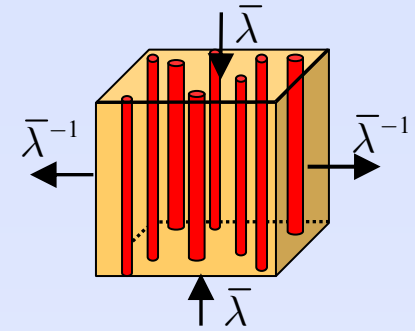
$$\bar{\lambda}_{cr}^{Lam} = \left(1 - \frac{\tilde{\mu}^L}{\bar{\mu}} \right)^{1/4}$$

where

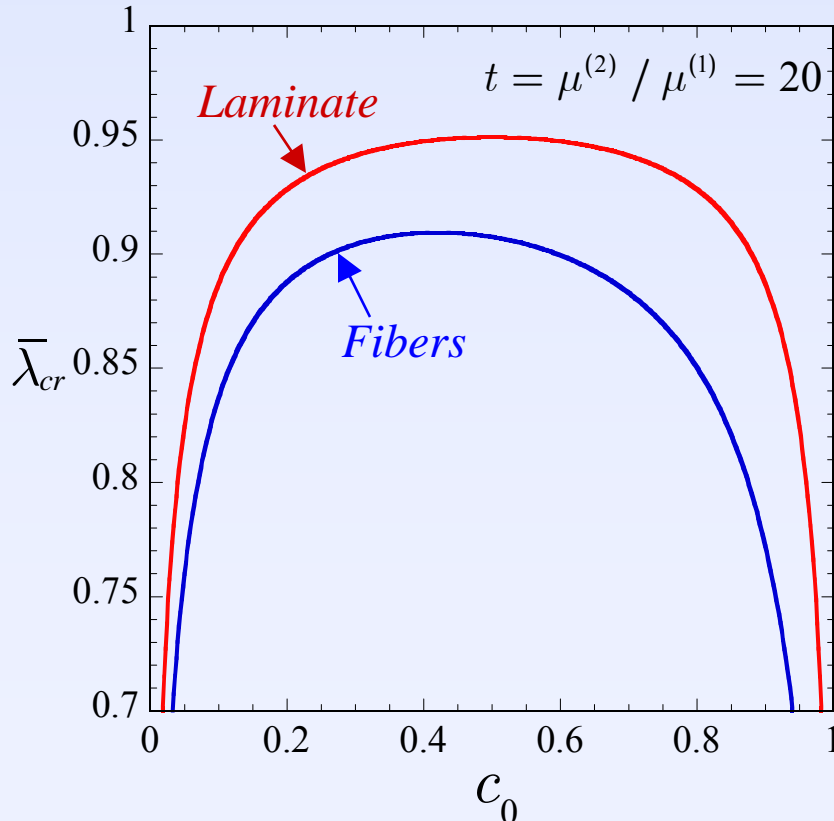
$$\bar{\mu} = (1 - c)\mu^{(1)} + c\mu^{(2)} \quad \text{and} \quad \tilde{\mu}^L = \left(\frac{1 - c}{\mu^{(1)}} + \frac{c}{\mu^{(2)}} \right)^{-1}$$

Contact with earlier work with laminates

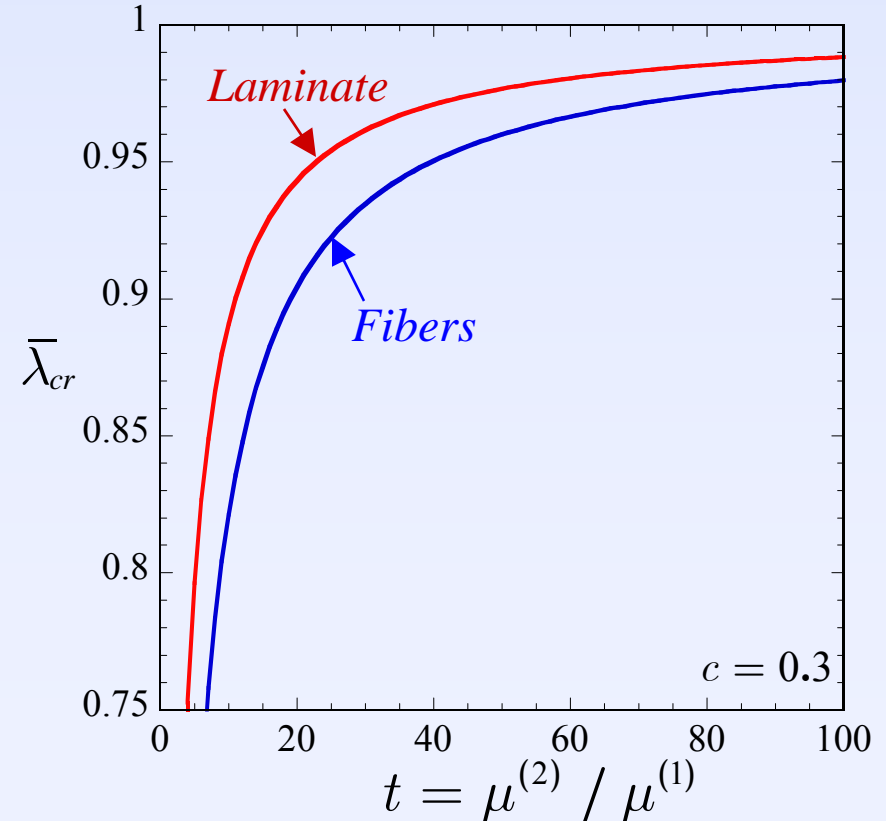
Aligned plane-strain loading conditions



Effect of **fiber concentration** c_0



Effect of **contrast** $t = \mu^{(2)} / \mu^{(1)}$



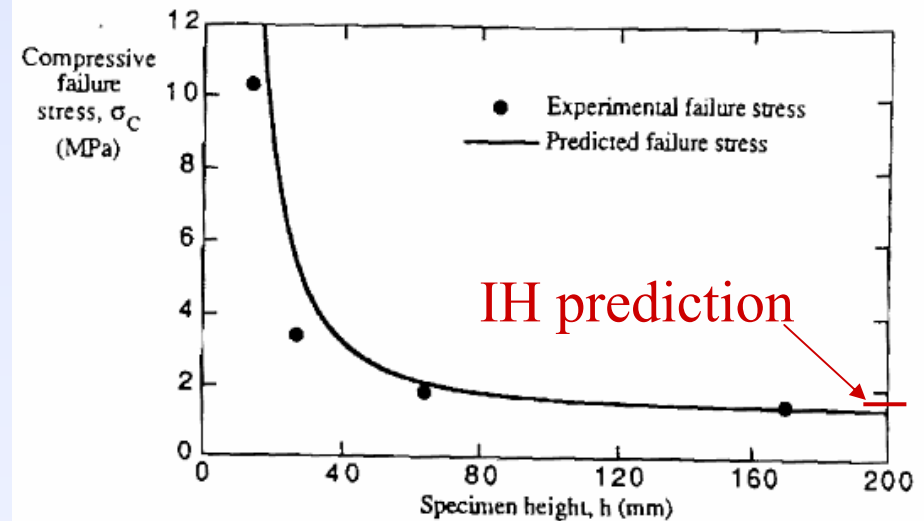
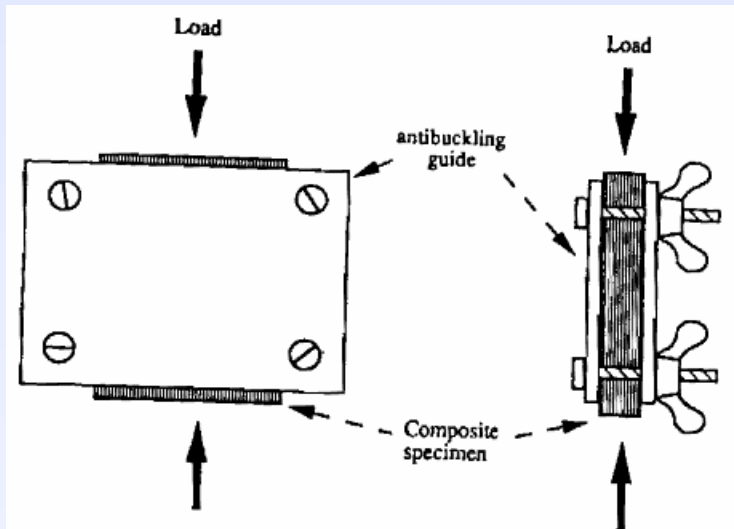
Contact with experiments

Matrix: **Silicone** — Young's Modulus $E^{(1)} = 2.9 \text{ MPa}$

Fibers: **Spaghetti** — Young's Modulus $E^{(2)} = 69 \text{ MPa}$

Volume fraction $c_0 = 31\%$

Experimental setup



Final remarks

- An iterated homogenization approach in finite elasticity has been proposed to construct exact (realizable) constitutive models for fiber-reinforced hyperelastic solids
- Because the proposed formulation grants access to local fields, it can be used to thoroughly study the onset of failure and the evolution of microstructure in fiber-reinforced soft solids with random microstructures
- The required analysis reduces to the study of tractable Hamilton-Jacobi equations
- As a first application, closed-form results were derived for fiber-reinforced Neo-Hookean elastomers
- These ideas can be generalized to more complex systems of soft heterogeneous media with random microstructures