Elastomers Filled with Liquid Inclusions:

Theory, Numerical Implemetation, and Some Basic Results

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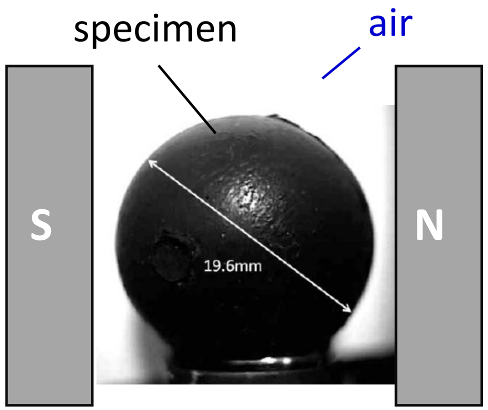
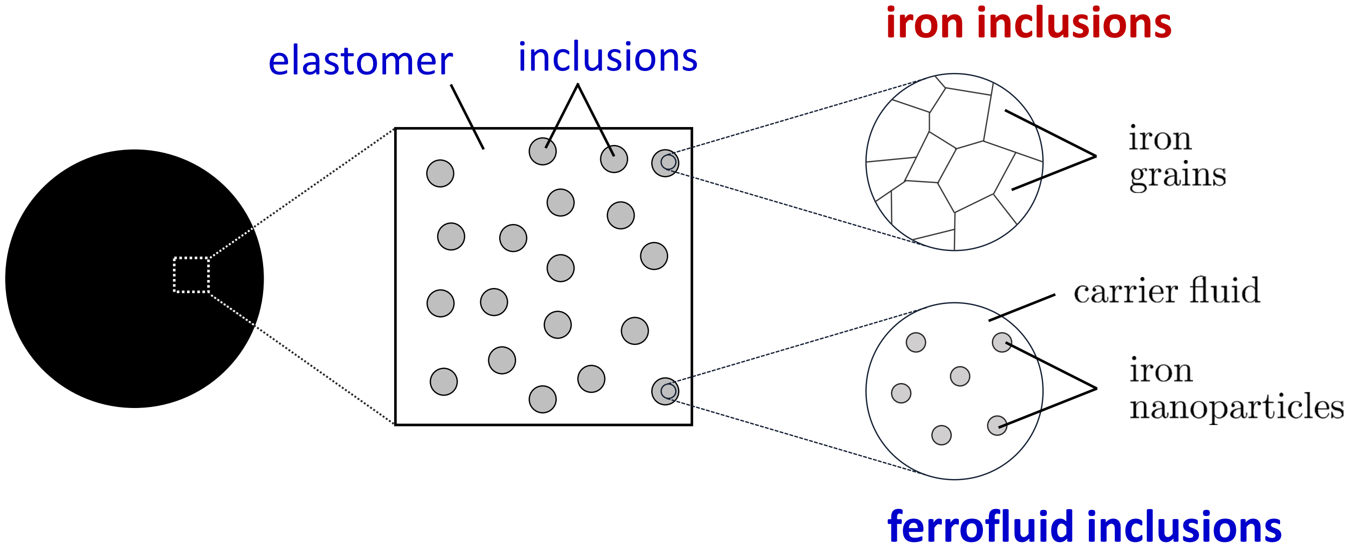
Liquid *vs*. solid inclusions: *(i) increased deformability*

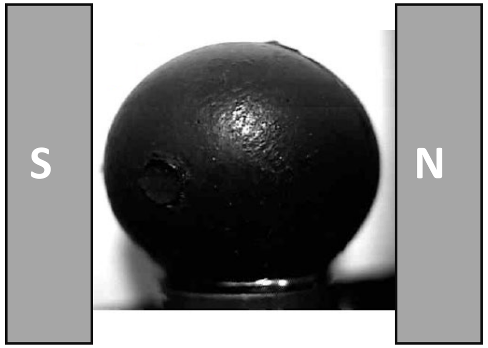
Conventional inclusions are typically much stiffer than elastomers. Example: ***iron inclusions*** in MREs



Magnetic field ***off***

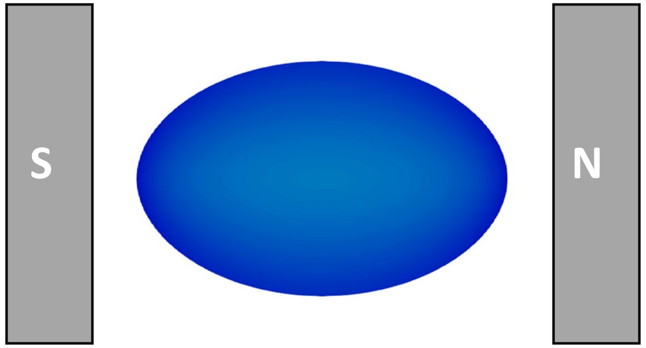


MRE with ***iron inclusions***



Magnetic field ***on***

MRE with ***ferrofluid inclusions***



Magnetic field ***on***

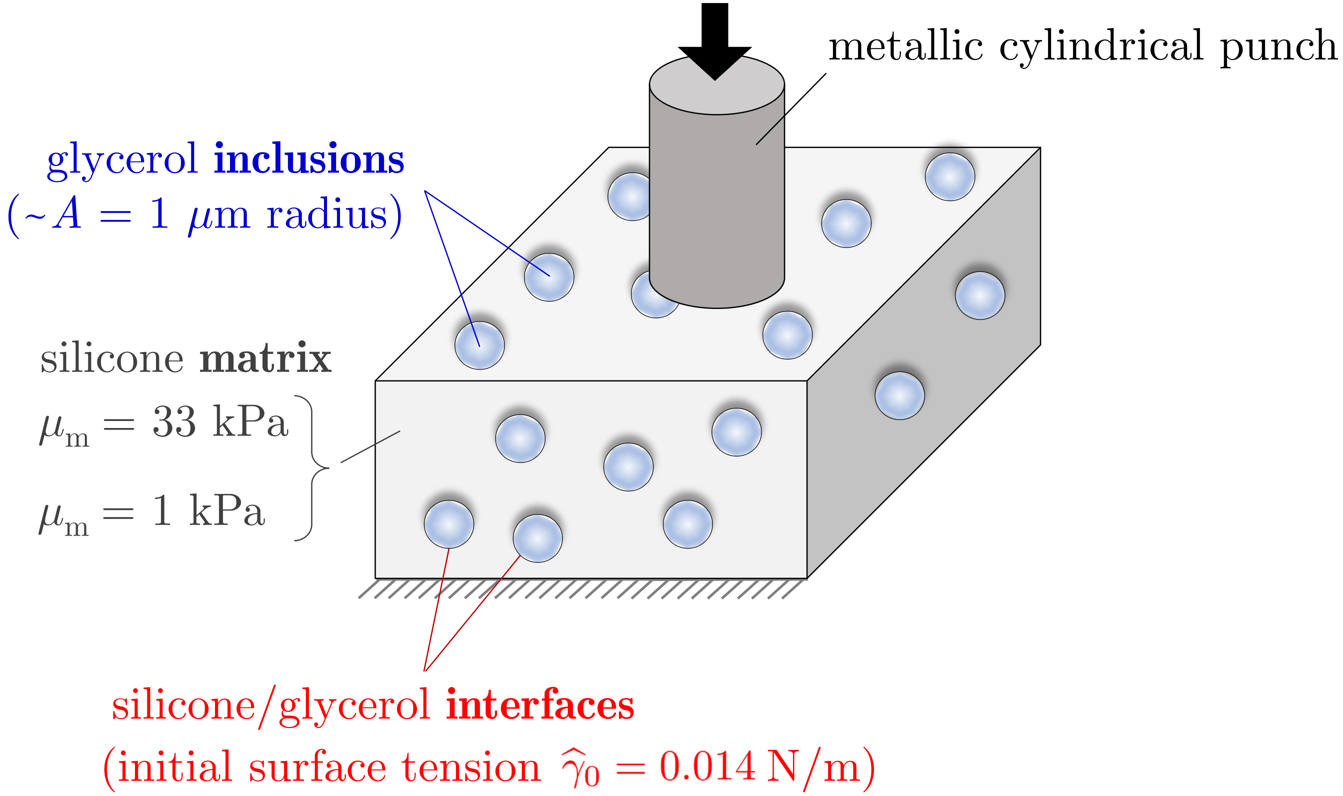
In contrast to the addition of *conventional (stiff) inclusions*,

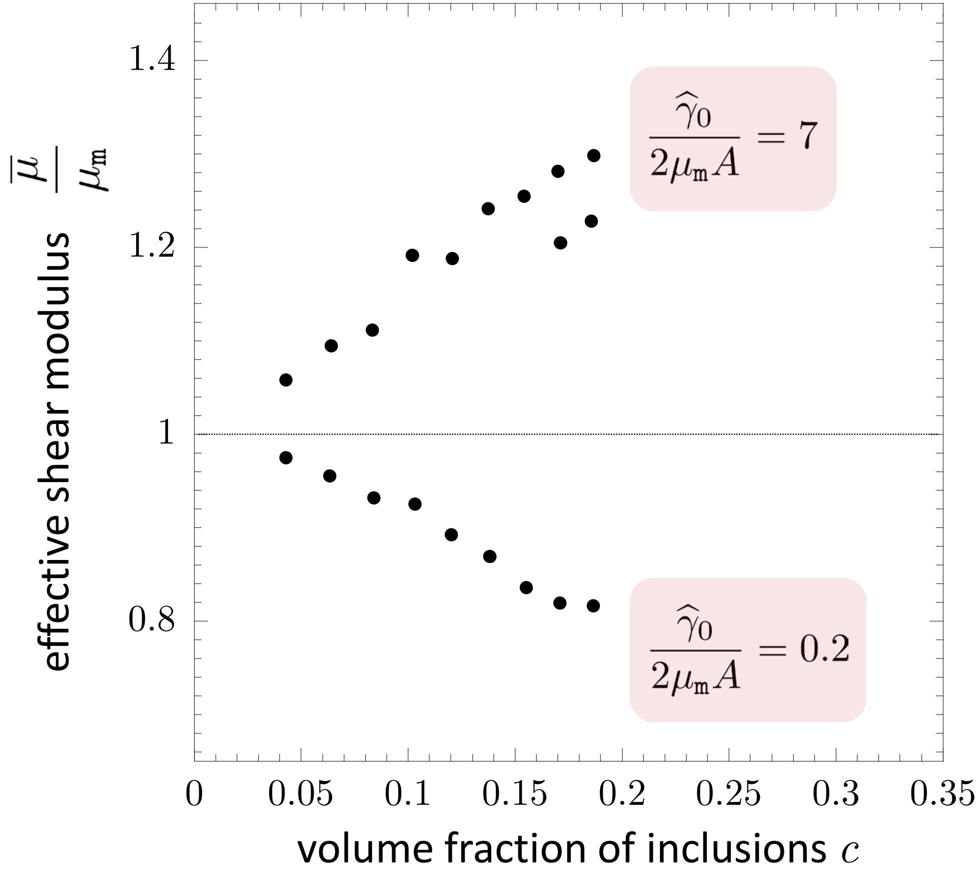
the addition of *liquid inclusions* to elastomers increases the overall deformability

Diguet (2010); Lopez-Pamies *JMPS* (2014); Lefèvre, Danas, Lopez-Pamies *JMPS* (2017)

Liquid *vs*. solid inclusions: *(ii) interface mechanics/physics may show up*

Indentation of silicone elastomers filled with glycerol droplets of roughly *monodisperse* size





Interface mechanics (e.g., elasto-capillarity) can be negligible, but it can also have a dominant effect on macroscopic properties

Style et al. (2015)

The Continuum Point of View

Elastomers filled with liquid inclusions: The continuum point of view

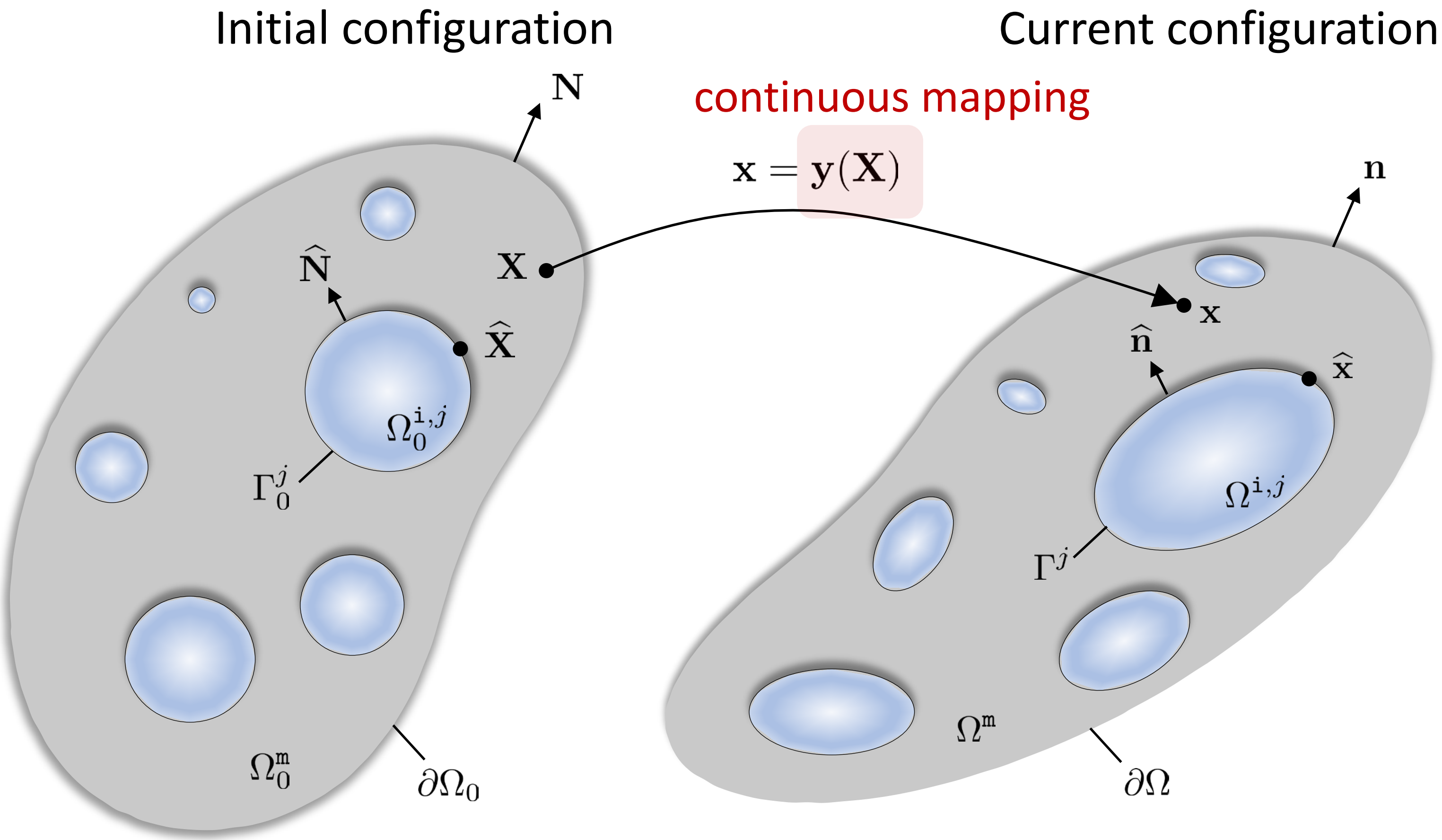
Three ingredients:

1. Initial configuration and kinematics
2. Balance of bulk, surface, and interface forces
3. Bulk and interface constitutive behaviors

Gurtin & Murdoch (1975); Ghosh & Lopez-Pamies *JMPS* (2022)

Elastomers filled with liquid inclusions: The continuum point of view

1. Initial configuration and kinematics
2. Balance of bulk, surface, and interface forces
3. Bulk and interface constitutive behaviors



- Material points on the interfaces

$$\widehat{\mathbf{X}} = \mathbf{X}, \quad \mathbf{X} \in \Gamma_0$$

- Deformation gradient in the bulk

$$\mathbf{F}(\mathbf{X}) = \nabla \mathbf{y}(\mathbf{X}) = \frac{\partial \mathbf{y}}{\partial \mathbf{X}}(\mathbf{X})$$

- Interface deformation gradient

$$\widehat{\mathbf{F}}(\widehat{\mathbf{X}}) = \widehat{\nabla}\mathbf{y}(\widehat{\mathbf{X}}) = \mathbf{F}(\widehat{\mathbf{X}})\widehat{\mathbf{I}}$$

where $\widehat{\mathbf{I}} = \mathbf{I} - \widehat{\mathbf{N}} \otimes \widehat{\mathbf{N}}$

- Continuity of deformation field $\mathbf{y}(\mathbf{X})$

$$[\![\mathbf{F}(\widehat{\mathbf{X}})]\!]\widehat{\mathbf{I}} = \mathbf{0} \quad \text{Hadamard condition}$$

where $[\![\mathbf{F}(\widehat{\mathbf{X}})]\!] := \mathbf{F}^{\mathtt{i}}(\widehat{\mathbf{X}}) - \mathbf{F}^{\mathtt{m}}(\widehat{\mathbf{X}})$

Gurtin & Murdoch (1975); Ghosh & Lopez-Pamies *JMPS* (2022)

Elastomers filled with liquid inclusions: The continuum point of view

1. Initial configuration and kinematics
2. Balance of bulk, surface, and interface forces
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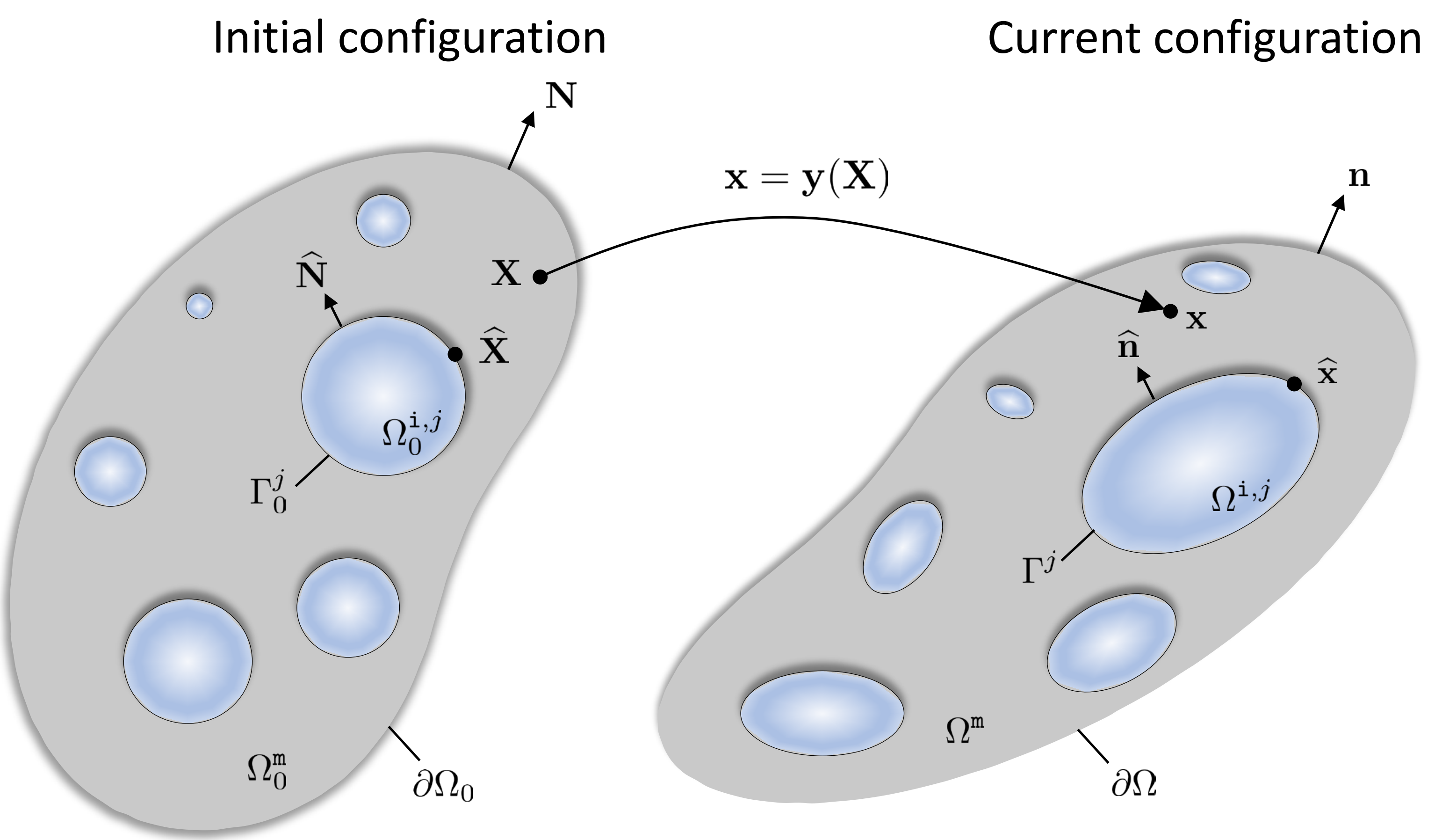

Notable relations:

$$\widehat{\mathbf{F}}^{-1}\widehat{\mathbf{F}} = \widehat{\mathbf{I}} \quad \text{and} \quad \widehat{\mathbf{F}}\widehat{\mathbf{F}}^{-1} = \widehat{\mathbf{i}}$$

where

$$\widehat{\mathbf{i}} = \mathbf{I} - \widehat{\mathbf{n}} \otimes \widehat{\mathbf{n}}$$

with

$$\widehat{\mathbf{n}} = \frac{J}{\widehat{J}}\mathbf{F}^{-T}\widehat{\mathbf{N}} \quad \text{where} \quad \widehat{J} = |J\mathbf{F}^{-T}\widehat{\mathbf{N}}|$$

It follows that

$$\boxed{\widehat{\mathbf{F}}^{-1} = \mathbf{F}^{-1}\widehat{\mathbf{i}}}$$

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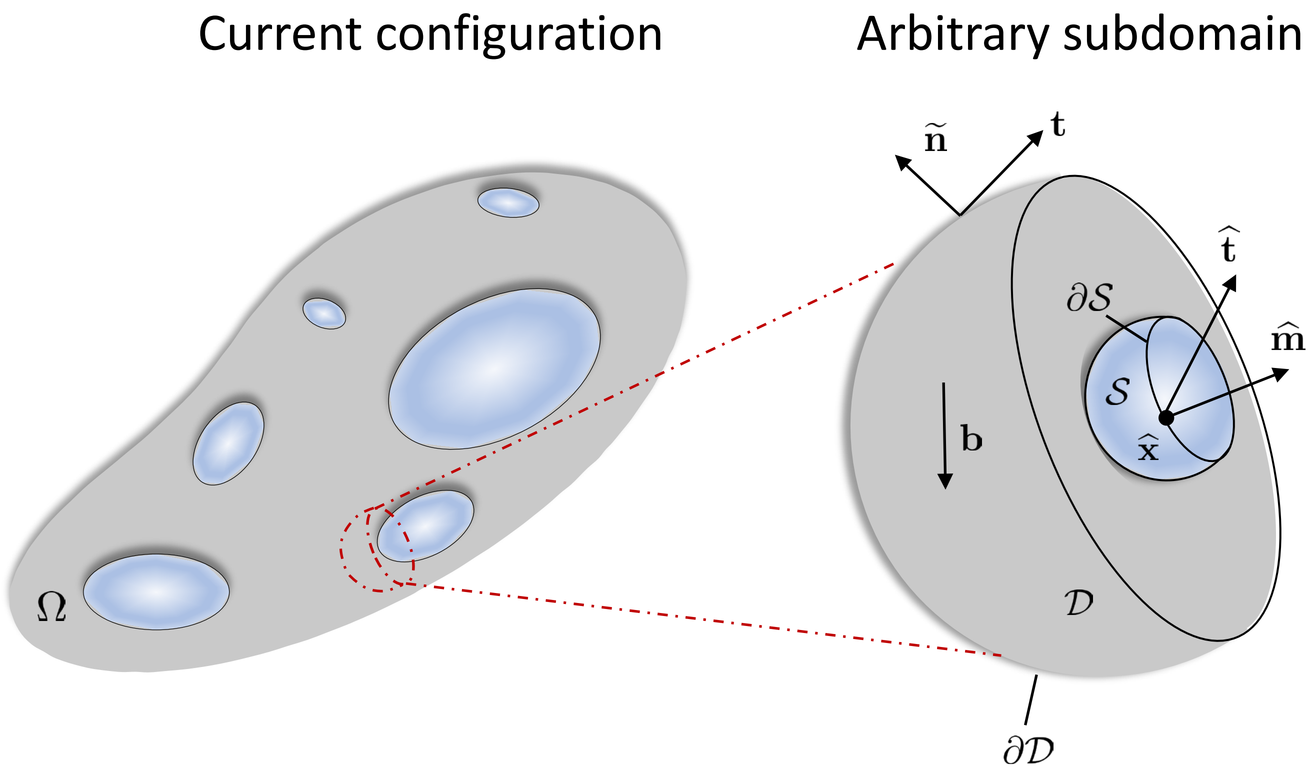

- Balance of linear momentum (***Eulerian***)

$$\int_{\mathcal{D}} \mathbf{b}(\mathbf{x})\,\mathrm{d}\mathbf{x} + \int_{\partial\mathcal{D}} \mathbf{t}(\mathbf{x},\tilde{\mathbf{n}})\,\mathrm{d}\mathbf{x} + \int_{\partial\mathcal{S}} \widehat{\mathbf{t}}(\widehat{\mathbf{x}},\widehat{\mathbf{m}})\,\mathrm{d}\widehat{\mathbf{x}} = \mathbf{0}$$

on use of Cauchy's postulate and the bulk and interface divergence theorems

$$\int_{\mathcal{D}} \frac{\partial(\cdot)}{\partial x_j}\,\mathrm{d}\mathbf{x} = \int_{\partial\mathcal{D}} (\cdot) n_j\,\mathrm{d}\mathbf{x} + \int_{\mathcal{S}} [\![\cdot]\!]\widehat{n}_j\,\mathrm{d}\mathbf{x}$$

$$\int_{\mathcal{S}} \frac{\partial(\cdot)}{\partial x_l}\widehat{i}_{kl}\,\mathrm{d}\mathbf{x} = \int_{\partial\mathcal{S}} (\cdot)\widehat{m}_k\,\mathrm{d}\mathbf{x} + \int_{\mathcal{S}} \frac{\partial \widehat{n}_p}{\partial x_q}\widehat{i}_{pq}(\cdot)\widehat{n}_k\,\mathrm{d}\mathbf{x}$$

we can rewrite

$$\begin{cases} \operatorname{div}\mathbf{T} + \mathbf{b} = \mathbf{0}, & \mathbf{x}\in\Omega\setminus\Gamma \\ \widehat{\operatorname{div}}\,\widehat{\mathbf{T}} - [\![\mathbf{T}]\!]\widehat{\mathbf{n}} = \mathbf{0}, & \mathbf{x}\in\Gamma \end{cases}$$

- Balance of angular momentum (***Eulerian***)

$$\begin{cases} \mathbf{T}^T = \mathbf{T}, & \mathbf{x}\in\Omega\setminus\Gamma \\ \widehat{\mathbf{T}}^T = \widehat{\mathbf{T}}, & \mathbf{x}\in\Gamma \end{cases}$$

Elastomers filled with liquid inclusions: The continuum point of view

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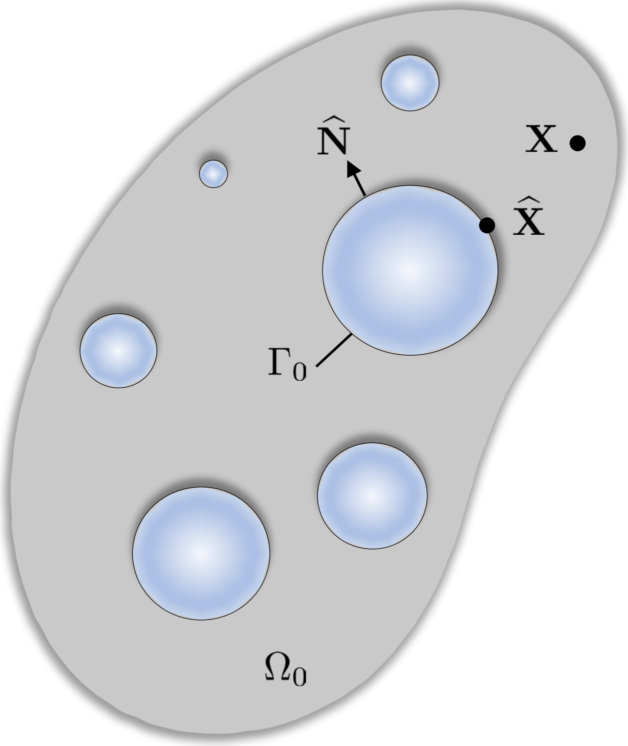
Initial configuration



- Balance of linear momentum (***Lagrangian***)

$$\begin{cases} \mathrm{Div}\,\mathbf{S} + \mathbf{B} = \mathbf{0}, & \mathbf{X} \in \Omega_0 \setminus \Gamma_0 \\ \widehat{\mathrm{Div}}\,\widehat{\mathbf{S}} - [\![\mathbf{S}]\!]\widehat{\mathbf{N}} = \mathbf{0}, & \mathbf{X} \in \Gamma_0 \end{cases}$$

- Balance of angular momentum (***Lagrangian***)

$$\begin{cases} \mathbf{S}\mathbf{F}^T = \mathbf{F}\mathbf{S}^T, & \mathbf{X} \in \Omega_0 \setminus \Gamma_0 \\ \widehat{\mathbf{S}}\widehat{\mathbf{F}}^T = \widehat{\mathbf{F}}\widehat{\mathbf{S}}^T, & \mathbf{X} \in \Gamma_0 \end{cases}$$

Here:

1st Piola-Kirchhoff stress tensor

$$\mathbf{S} = J\mathbf{T}\mathbf{F}^{-T}$$

Interface 1st Piola-Kirchhoff stress tensor

$$\widehat{\mathbf{S}} = \widehat{J}\widehat{\mathbf{T}}\widehat{\mathbf{F}}^{-T}$$

Elastomers filled with liquid inclusions: The continuum point of view

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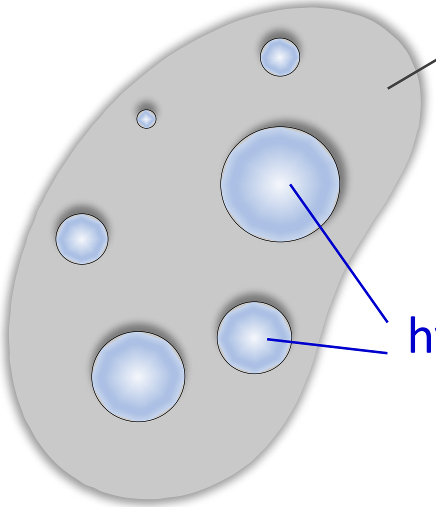
- **The bulk**: Assuming negligible dissipative phenomena, in the elastomeric matrix and liquid inclusions we take



hyperelastic solid: $W_{\mathtt{m}}(\mathbf{F}) = \frac{\mu_{\mathtt{m}}}{2}\left[\mathbf{F}\cdot\mathbf{F} - 3\right] - \mu_{\mathtt{m}} \ln J + \frac{\Lambda_{\mathtt{m}}}{2}(J-1)^2$

residual stress

compressibility (Lamé) parameter

hyperelastic fluid: $W^j_{\mathtt{i}}(\mathbf{X}, J) = r^j_{\mathtt{i}}(\mathbf{X})J + \frac{\Lambda_{\mathtt{i}}}{2}(J-1)^2 \quad j = 1, 2, ..., M$

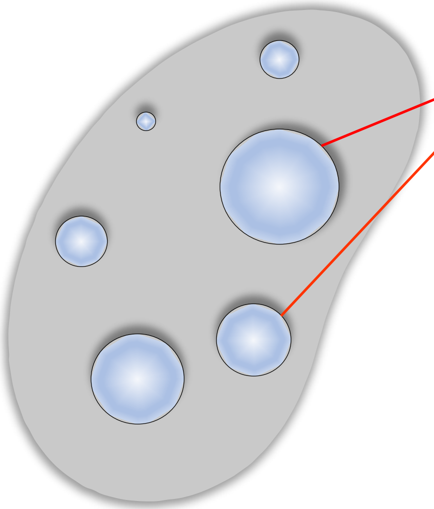
Compactly, we write $W(\mathbf{X},\mathbf{F}) = r_{\mathtt{i}}(\mathbf{X})J + \frac{\mu(\mathbf{X})}{2}\left[\mathbf{F}\cdot\mathbf{F} - 3\right] - \mu(\mathbf{X}) \ln J + \frac{\Lambda(\mathbf{X})}{2}(J-1)^2$ and hence

$$\boxed{\mathbf{S}(\mathbf{X}) = \frac{\partial W}{\partial \mathbf{F}}(\mathbf{X},\mathbf{F}) = r_{\mathtt{i}}(\mathbf{X})J\mathbf{F}^{-T} + \mu(\mathbf{X})\left(\mathbf{F} - \mathbf{F}^{-T}\right) + \Lambda(\mathbf{X})(J-1)J\mathbf{F}^{-T}, \quad \mathbf{X} \in \Omega_0 \setminus \Gamma_0}$$

Elastomers filled with liquid inclusions: The continuum point of view

1. Initial configuration and kinematics
2. Balance of bulk, surface, and interface forces
3. Bulk and interface constitutive behaviors

- **The interfaces**: Assuming, as for the bulk, negligible dissipative phenomena, in the interfaces we take



hyperelastic interfaces: $$\widehat{W}(\widehat{\mathbf{F}}) = \underbrace{\widehat{\gamma}_0}_{\text{initial surface tension}} \widehat{J} + \frac{\widehat{\mu}}{2}\left[\widehat{\mathbf{F}}\cdot\widehat{\mathbf{F}} - 2\right] - \widehat{\mu}\ln\widehat{J} + \frac{\widehat{\Lambda}}{2}(\widehat{J}-1)^2$$

The interface 1st Piola-Kirchhoff stress is hence given by

$$\boxed{\widehat{\mathbf{S}}(\mathbf{X}) = \frac{\partial\widehat{W}}{\partial\widehat{\mathbf{F}}} = \widehat{\gamma}_0\,\widehat{J}\widehat{\mathbf{F}}^{-T} + \widehat{\mu}(\widehat{\mathbf{F}} - \widehat{\mathbf{F}}^{-T}) + \widehat{\Lambda}(\widehat{J}-1)\widehat{J}\widehat{\mathbf{F}}^{-T}, \quad \mathbf{X}\in\Gamma_0}$$

Note: this constitutive relation generalizes in two counts the basic constitutive relation of constant surface tension

$$\widehat{\mathbf{T}}(\mathbf{x}) = \widehat{J}^{-1}\widehat{\mathbf{S}}\widehat{\mathbf{F}}^T = \widehat{\gamma}_0\,\widehat{\mathbf{i}} + \widehat{\mu}(\widehat{J}^{-1}\widehat{\mathbf{F}}\widehat{\mathbf{F}}^T - \widehat{\mathbf{i}}) + \widehat{\Lambda}(\widehat{J}-1)\widehat{\mathbf{i}}, \quad \mathbf{x}\in\Gamma$$

where we recall that $\widehat{\mathbf{i}} = \mathbf{I} - \widehat{\mathbf{n}}\otimes\widehat{\mathbf{n}}$

Elastomers filled with liquid inclusions: The continuum point of view

Putting all the three ingredients together, we end up with the following *Governing Equations*:

$$
\begin{cases}
\mathrm{Div}\left[r_{\mathtt{i}}(\mathbf{X})J\nabla\mathbf{y}^{-T}+\mu(\mathbf{X})\left(\nabla\mathbf{y}-\nabla\mathbf{y}^{-T}\right)+\Lambda(\mathbf{X})(J-1)J\nabla\mathbf{y}^{-T}\right]+\mathbf{B}=\mathbf{0}, \quad \mathbf{X}\in\Omega_0\setminus\Gamma_0 \\
\widehat{\mathrm{Div}}\left[\widehat{\gamma}_0\,\widehat{J}\widehat{\nabla}\mathbf{y}^{-T}+\widehat{\mu}(\widehat{\nabla}\mathbf{y}-\widehat{\nabla}\mathbf{y}^{-T})+\widehat{\Lambda}(\widehat{J}-1)\widehat{J}\widehat{\nabla}\mathbf{y}^{-T}\right]- \\
[\![r_{\mathtt{i}}(\mathbf{X})J\nabla\mathbf{y}^{-T}+\mu(\mathbf{X})\left(\nabla\mathbf{y}-\nabla\mathbf{y}^{-T}\right)+\Lambda(\mathbf{X})(J-1)J\nabla\mathbf{y}^{-T}]\!]\,\widehat{\mathbf{N}}=\mathbf{0}, \quad \mathbf{X}\in\Gamma_0 \\
\mathbf{y}(\mathbf{X})=\overline{\mathbf{y}}(\mathbf{X}), \quad \mathbf{X}\in\partial\Omega_0^{\mathcal{D}} \\
\left[\mu_{\mathtt{m}}\left(\nabla\mathbf{y}-\nabla\mathbf{y}^{-T}\right)+\Lambda_{\mathtt{m}}(J-1)J\nabla\mathbf{y}^{-T}\right]\mathbf{N}=\overline{\mathbf{s}}(\mathbf{X}), \quad \mathbf{X}\in\partial\Omega_0^{\mathcal{N}}
\end{cases}
$$

for the deformation field

$\mathbf{y}(\mathbf{X})$

In the initial configuration, prior to the applications of boundary conditions and body forces when, $\mathbf{y}(\mathbf{X})=\mathbf{X}$, **these equations reduce to**

$$
\begin{cases}
\nabla r_{\mathtt{i}}(\mathbf{X})=\mathbf{0}, \quad \mathbf{X}\in\Omega_0\setminus\Gamma_0 \\
r_{\mathtt{i}}(\mathbf{X})=-\mathrm{tr}\,\widehat{\nabla}\widehat{\mathbf{N}}\,\widehat{\gamma}_0, \quad \mathbf{X}\in\Gamma_0
\end{cases}
$$

No solution exists unless the inclusions have constant mean curvature

For spherical inclusions of initial radius A_j : $r_{\mathtt{i}}(\mathbf{X})=-\displaystyle\sum_{j=1}^{M}\theta_0^{\mathtt{i},j}(\mathbf{X})\frac{2\,\widehat{\gamma}_0}{A_j}$

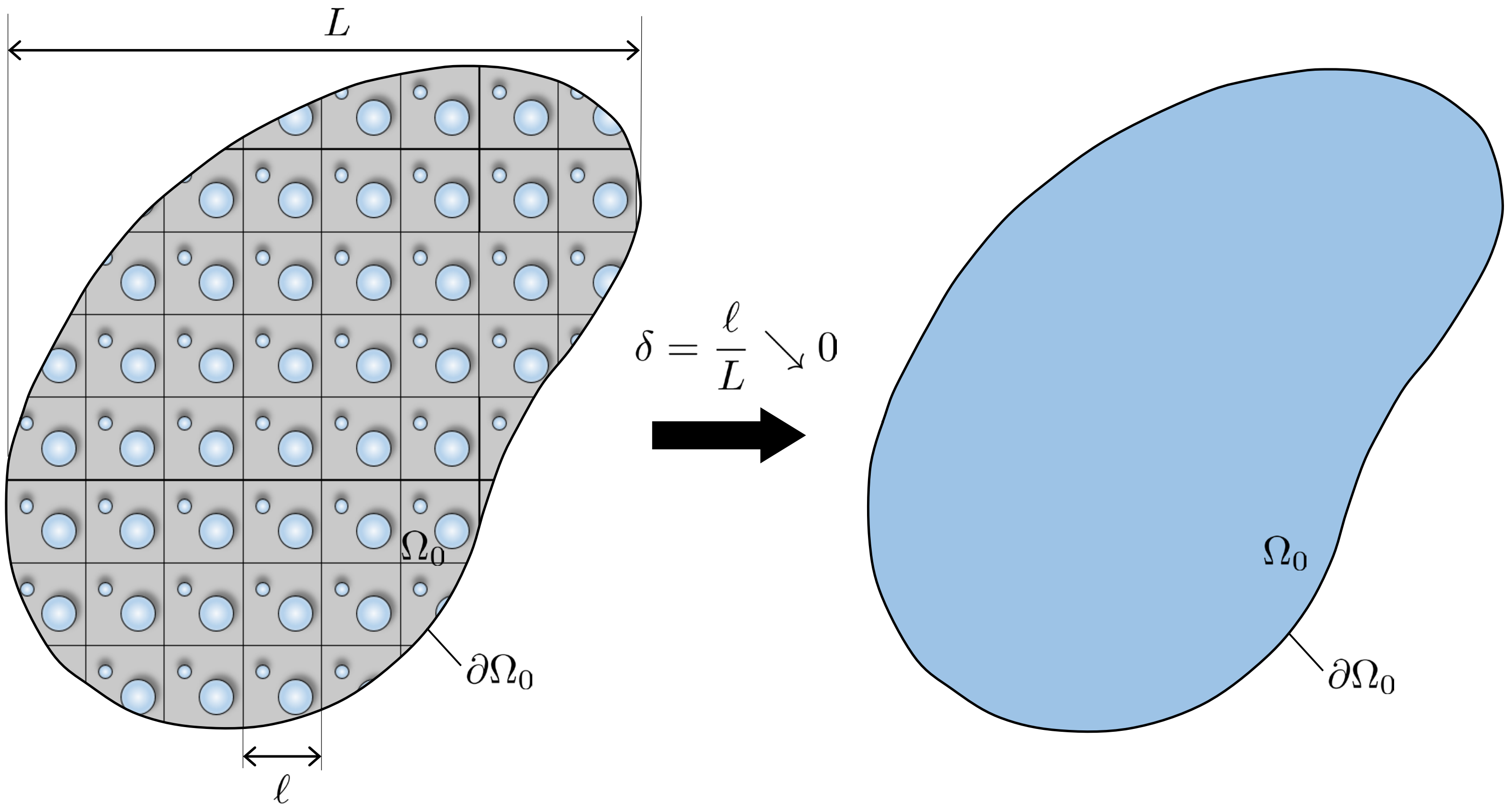
The presence of inclusions that are *not* initially spherical implies that the elastomer (and not just the liquid inclusions) have a residual stress

Ghosh & Lopez-Pamies *JMPS* (2022)

Homogenization

Elastomers filled with *spherical* liquid inclusions: Homogenization

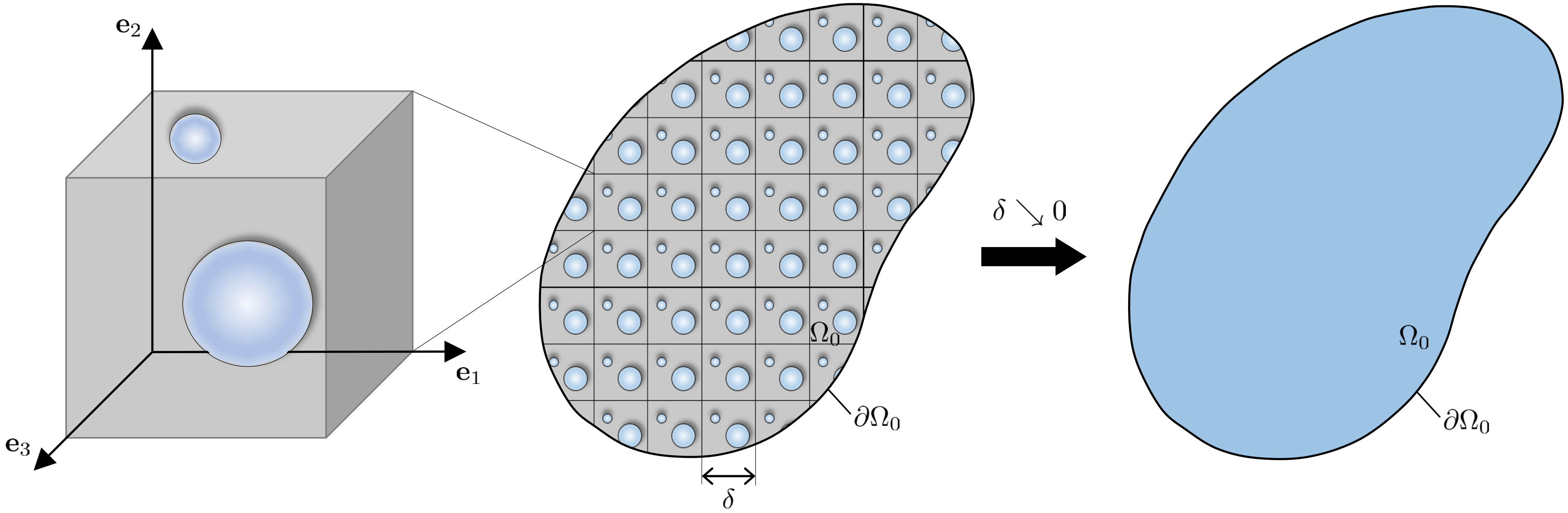
Among all possible boundary-value problems that we can now investigate, let us consider the one where we have: a statistically uniform distribution of small (relative to specimen size) spherical liquid inclusions



Elastomers filled with *spherical* liquid inclusions: Homogenization

Neglecting body forces and considering for definiteness Dirichlet boundary conditions and periodic microstructures of period δ, we have that for fixed δ:

$$
\left\{
\begin{array}{l}
\mathrm{Div}\left[r^{\sharp}_{\mathtt{i}}(\delta^{-1}\mathbf{X})J^{\delta}\nabla\mathbf{y}^{\delta^{-T}}+\mu^{\sharp}(\delta^{-1}\mathbf{X})\left(\nabla\mathbf{y}^{\delta}-\nabla\mathbf{y}^{\delta^{-T}}\right)+\Lambda^{\sharp}(\delta^{-1}\mathbf{X})(J^{\delta}-1)J^{\delta}\nabla\mathbf{y}^{\delta^{-T}}\right]=\mathbf{0},\quad \mathbf{X}\in\Omega_0\setminus\Gamma_0 \\[2ex]
\widehat{\mathrm{Div}}\left[\widehat{\gamma}_0\,\widehat{J}^{\delta}\widehat{\nabla}\mathbf{y}^{\delta^{-T}}+\widehat{\mu}(\widehat{\nabla}\mathbf{y}^{\delta}-\widehat{\nabla}\mathbf{y}^{\delta^{-T}})+\widehat{\Lambda}(\widehat{J}^{\delta}-1)\widehat{J}^{\delta}\widehat{\nabla}\mathbf{y}^{\delta^{-T}}\right]- \\[1ex]
\left[\!\left[r^{\sharp}_{\mathtt{i}}(\delta^{-1}\mathbf{X})J^{\delta}\nabla\mathbf{y}^{\delta^{-T}}+\mu^{\sharp}(\delta^{-1}\mathbf{X})\left(\nabla\mathbf{y}^{\delta}-\nabla\mathbf{y}^{\delta^{-T}}\right)+\Lambda^{\sharp}(\delta^{-1}\mathbf{X})(J^{\delta}-1)J^{\delta}\nabla\mathbf{y}^{\delta^{-T}}\right]\!\right]\widehat{\mathbf{N}}=\mathbf{0},\quad \mathbf{X}\in\Gamma_0 \\[2ex]
\mathbf{y}^{\delta}(\mathbf{X})=\overline{\mathbf{y}}(\mathbf{X}),\quad \mathbf{X}\in\partial\Omega_0
\end{array}
\right.
$$



Elastomers filled with *spherical* liquid inclusions: Homogenization

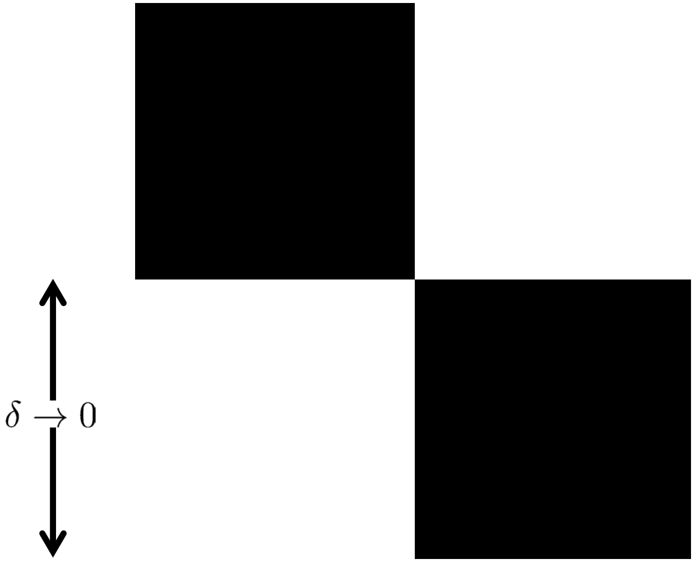
Neglecting body forces and considering for definiteness Dirichlet boundary conditions and periodic microstructures of period δ, we have that for fixed δ:

$$\begin{cases} \mathrm{Div}\left[r^{\sharp}_{\mathtt{i}}(\delta^{-1}\mathbf{X})J^{\delta}\nabla\mathbf{y}^{\delta^{-T}} + \mu^{\sharp}(\delta^{-1}\mathbf{X})\left(\nabla\mathbf{y}^{\delta} - \nabla\mathbf{y}^{\delta^{-T}}\right) + \Lambda^{\sharp}(\delta^{-1}\mathbf{X})(J^{\delta}-1)J^{\delta}\nabla\mathbf{y}^{\delta^{-T}}\right] = \mathbf{0}, \quad \mathbf{X}\in\Omega_0\setminus\Gamma_0 \\ \widehat{\mathrm{Div}}\left[\widehat{\gamma}_0\,\widehat{J}^{\delta}\widehat{\nabla}\mathbf{y}^{\delta^{-T}} + \widehat{\mu}(\widehat{\nabla}\mathbf{y}^{\delta} - \widehat{\nabla}\mathbf{y}^{\delta^{-T}}) + \widehat{\Lambda}(\widehat{J}^{\delta}-1)\widehat{J}^{\delta}\widehat{\nabla}\mathbf{y}^{\delta^{-T}}\right] - \\ \left[\!\left[r^{\sharp}_{\mathtt{i}}(\delta^{-1}\mathbf{X})J^{\delta}\nabla\mathbf{y}^{\delta^{-T}} + \mu^{\sharp}(\delta^{-1}\mathbf{X})\left(\nabla\mathbf{y}^{\delta} - \nabla\mathbf{y}^{\delta^{-T}}\right) + \Lambda^{\sharp}(\delta^{-1}\mathbf{X})(J^{\delta}-1)J^{\delta}\nabla\mathbf{y}^{\delta^{-T}}\right]\!\right]\widehat{\mathbf{N}} = \mathbf{0}, \quad \mathbf{X}\in\Gamma_0 \\ \mathbf{y}^{\delta}(\mathbf{X}) = \overline{\mathbf{y}}(\mathbf{X}), \quad \mathbf{X}\in\partial\Omega_0 \end{cases}$$

The name of the game is to pass to the limit as $\delta \searrow 0$ in these equations...

...this is currently out of reach from a rigorous point of view.

To gain insight, we begin by considering the **limit of small deformations** first



Homogenization:
The Small-Deformation Limit

Elastomers filled with *spherical* liquid inclusions: Homogenization

In the limit of small deformations, the above equations specialize to

$$\begin{cases} \mathrm{Div}\left[\mathbf{L}(\delta^{-1}\mathbf{X})\nabla\mathbf{u}^\delta\right] = \mathbf{0}, \quad \mathbf{X}\in\Omega_0\setminus\Gamma_0 \\ \widehat{\mathrm{Div}}\left[\delta\,\widehat{\mathbf{L}}\widehat{\nabla}\mathbf{u}^\delta\right] - [\![\mathbf{L}(\delta^{-1}\mathbf{X})\nabla\mathbf{u}^\delta]\!]\widehat{\mathbf{N}} = \mathbf{0}, \quad \mathbf{X}\in\Gamma_0 \\ \mathbf{u}^\delta(\mathbf{X}) = \overline{\mathbf{u}}(\mathbf{X}), \quad \mathbf{X}\in\partial\Omega_0 \end{cases}$$

for the displacement field $\mathbf{u}^\delta(\mathbf{X}) = \mathbf{y}^\delta(\mathbf{X}) - \mathbf{X}$

Here,

$$\mathbf{L}(\delta^{-1}\mathbf{X}) = \left(1-\theta(\delta^{-1}\mathbf{X})\right)\mathbf{L}^{(\mathtt{m})} + \sum_{I=1}^{\mathtt{N}}\theta_I(\epsilon^{-1}\mathbf{X})\left[3\Lambda^{(\mathtt{i})}\boldsymbol{\mathcal{J}} - \frac{2\widehat{\gamma}_0}{A_I}(\boldsymbol{\mathcal{A}}-\boldsymbol{\mathcal{K}}+2\boldsymbol{\mathcal{J}})\right]$$

and

$$\widehat{\mathbf{L}} = 2\,\widehat{\mu}\,\widehat{\boldsymbol{\mathcal{K}}} + 2(\widehat{\mu}+\widehat{\Lambda})\widehat{\boldsymbol{\mathcal{J}}} + \widehat{\gamma}_0\left(\widehat{\boldsymbol{\mathcal{A}}} - \widehat{\boldsymbol{\mathcal{K}}} + \widehat{\boldsymbol{\mathcal{J}}}\right)$$

where

$$\mathcal{A}_{ijkl} = \frac{1}{2}(\delta_{ik}\delta_{jl}-\delta_{il}\delta_{jk}), \quad \mathcal{K}_{ijkl} = \frac{1}{2}(\delta_{ik}\delta_{jl}+\delta_{il}\delta_{jk}) - \frac{1}{3}\delta_{ij}\delta_{kl}, \quad \mathcal{J}_{ijkl} = \frac{1}{3}\delta_{ij}\delta_{kl}$$

$$\widehat{\mathcal{A}}_{ijkl} = \delta_{ik}\widehat{I}_{jl} - \frac{1}{2}\left(\widehat{I}_{ik}\widehat{I}_{jl} + \widehat{I}_{il}\widehat{I}_{jk}\right), \quad \widehat{\mathcal{K}}_{ijkl} = \frac{1}{2}\left(\widehat{I}_{ik}\widehat{I}_{jl} + \widehat{I}_{il}\widehat{I}_{jk} - \widehat{I}_{ij}\widehat{I}_{kl}\right), \quad \widehat{\mathcal{J}}_{ijkl} = \frac{1}{2}\widehat{I}_{ij}\widehat{I}_{kl}$$

Ghosh & Lopez-Pamies *SIAP* (2022)

Elastomers filled with *spherical* liquid inclusions: Homogenization

In the limit of small deformations, the above equations specialize to

$$\begin{cases} \mathrm{Div}\left[\mathbf{L}(\delta^{-1}\mathbf{X})\nabla\mathbf{u}^{\delta}\right]=\mathbf{0}, & \mathbf{X}\in\Omega_0\setminus\Gamma_0 \\ \widehat{\mathrm{Div}}\left[\delta\,\widehat{\mathbf{L}}\widehat{\nabla}\mathbf{u}^{\delta}\right]-[\![\mathbf{L}(\delta^{-1}\mathbf{X})\nabla\mathbf{u}^{\delta}]\!]\widehat{\mathbf{N}}=\mathbf{0}, & \mathbf{X}\in\Gamma_0 \\ \mathbf{u}^{\delta}(\mathbf{X})=\overline{\mathbf{u}}(\mathbf{X}), & \mathbf{X}\in\partial\Omega_0 \end{cases}$$

for the displacement field $\mathbf{u}^{\delta}(\mathbf{X})=\mathbf{y}^{\delta}(\mathbf{X})-\mathbf{X}$

Here,

$$\mathbf{L}(\delta^{-1}\mathbf{X})=\left(1-\theta(\delta^{-1}\mathbf{X})\right)\mathbf{L}^{(\mathtt{m})}+\sum_{I=1}^{\mathtt{N}}\theta_I(\epsilon^{-1}\mathbf{X})\left[3\Lambda^{(\mathtt{i})}\boldsymbol{\mathcal{J}}-\frac{2\widehat{\gamma}_0}{A_I}(\boldsymbol{\mathcal{A}}-\boldsymbol{\mathcal{K}}+2\boldsymbol{\mathcal{J}})\right]$$

and

$$\widehat{\mathbf{L}}=2\,\widehat{\mu}\,\widehat{\boldsymbol{\mathcal{K}}}+2(\widehat{\mu}+\widehat{\Lambda})\widehat{\boldsymbol{\mathcal{J}}}+\widehat{\gamma}_0\left(\widehat{\boldsymbol{\mathcal{A}}}-\widehat{\boldsymbol{\mathcal{K}}}+\widehat{\boldsymbol{\mathcal{J}}}\right)$$

Remark 1: The moduli of elasticity $\mathbf{L}$ and $\widehat{\mathbf{L}}$ do *not* posses minor symmetry and are *not* positive definite

Remark 2: The Lax-Milgram theorem does not apply to prove existence of solution. A new theorem is needed!

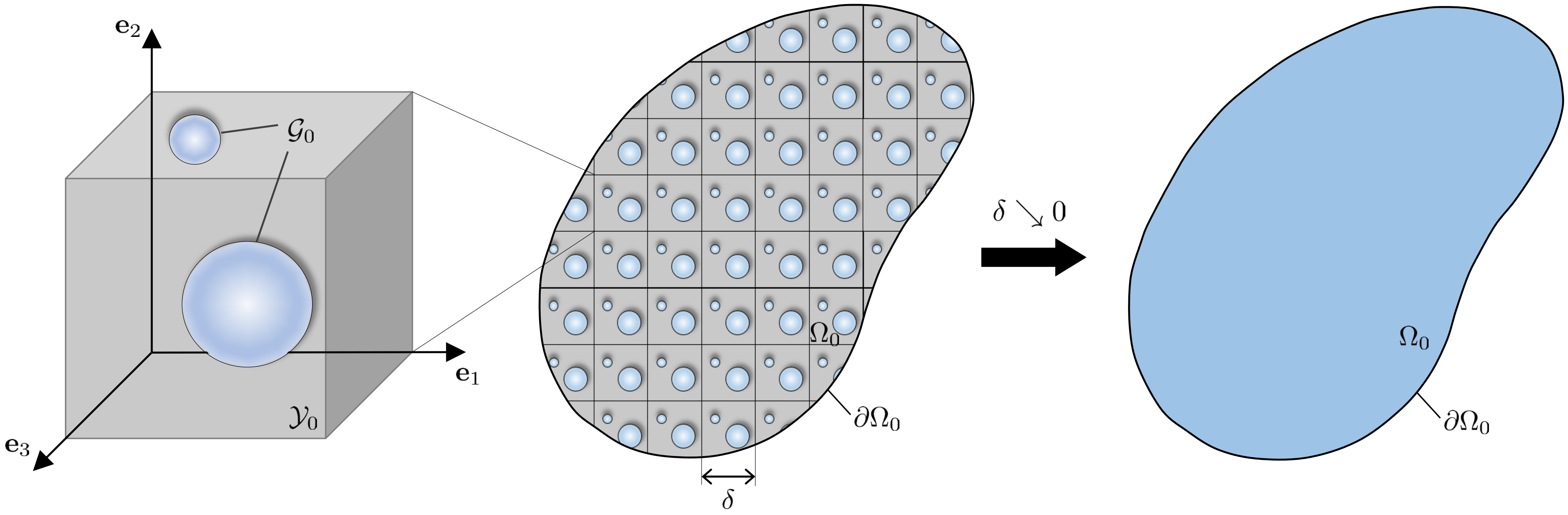
Remark 3: The expectation is that solutions $\mathbf{u}^{\delta}\in H^1(\Omega_0;\mathbb{R}^3)$ exist. We have examples!

Elastomers filled with *spherical* liquid inclusions: Homogenization

The limit $\delta \searrow 0$ by the method of two-scale asymptotic expansions. Assuming existence, we look for solutions of the form

$$u_i^\delta(\mathbf{X}) = u_i^{(0)}(\mathbf{X}, \delta^{-1}\mathbf{X}) + \delta\, u_i^{(1)}(\mathbf{X}, \delta^{-1}\mathbf{X}) + \delta^2 u_i^{(2)}(\mathbf{X}, \delta^{-1}\mathbf{X}) + \ldots = \sum_{s=0}^{\infty} \delta^s u_i^{(s)}(\mathbf{X}, \delta^{-1}\mathbf{X})$$

where the functions $\mathbf{u}^{(s)}(\mathbf{X}, \mathbf{Y})$ with $\mathbf{Y} = \delta^{-1}\mathbf{X}$ are $\mathcal{Y}_0$-**periodic** in their second argument.



Elastomers filled with *spherical* liquid inclusions: Homogenization

After some lengthy calculations, it can be shown that

$\mathbf{u}^{(0)}(\mathbf{X},\delta^{-1}\mathbf{X})=\mathbf{u}(\mathbf{X})$ where $\mathbf{u}(\mathbf{X})$ is solution of

$$\left\{\begin{array}{ll}\mathrm{Div}\left[\overline{\mathbf{L}}\,\nabla\mathbf{u}\right]=\mathbf{0}, & \mathbf{X}\in\Omega_0\\ \mathbf{u}(\mathbf{X})=\overline{\mathbf{u}}(\mathbf{X}), & \mathbf{X}\in\partial\Omega_0\end{array}\right.$$

with

$$\overline{L}_{ijkl}=\int_{\mathcal{Y}_0}L_{ijmn}(\mathbf{Y})\left(\delta_{mk}\delta_{nl}+\frac{\partial\omega_{mkl}}{\partial Y_n}(\mathbf{Y})\right)\mathrm{d}\mathbf{Y}+\int_{\mathcal{G}_0}\widehat{L}_{ijmn}\left(\delta_{mk}\widehat{I}_{nl}+\frac{\partial\omega_{mkl}}{\partial Y_p}(\mathbf{Y})\widehat{I}_{pn}\right)\mathrm{d}\mathbf{Y}$$

where $\boldsymbol{\omega}(\mathbf{Y})$ is the $\mathcal{Y}_0$-periodic function defined implicitly by the **unit-cell problem**

$$\left\{\begin{array}{l}\dfrac{\partial}{\partial Y_j}\left[L_{ijkl}(\mathbf{Y})\dfrac{\partial\omega_{kmn}}{\partial Y_l}(\mathbf{Y})\right]=-\dfrac{\partial L_{ijmn}}{\partial Y_j}(\mathbf{Y}),\quad \mathbf{Y}\in\mathcal{Y}_0\setminus\mathcal{G}_0\\ \dfrac{\partial}{\partial Y_q}\left[\widehat{L}_{ijkl}\dfrac{\partial\omega_{kmn}}{\partial Y_p}(\mathbf{Y})\widehat{I}_{pl}\right]\widehat{I}_{qj}-[\![L_{ijkl}(\mathbf{Y})\dfrac{\partial\omega_{kmn}}{\partial Y_l}(\mathbf{Y})]\!]\widehat{N}_j=-\dfrac{\partial}{\partial Y_q}\left[\widehat{L}_{ijkl}\delta_{km}\widehat{I}_{nl}\right]\widehat{I}_{qj}+[\![L_{ijkl}(\mathbf{Y})\delta_{km}\delta_{ln}]\!]\widehat{N}_j,\ \mathbf{Y}\in\mathcal{G}_0\\ \int_{\mathcal{Y}_0}\omega_{kmn}(\mathbf{Y})\mathrm{d}\mathbf{Y}=0\end{array}\right.$$

Note: In general, the solution to this unit-cell problem can only be work out numerically

Elastomers filled with *spherical* liquid inclusions: Homogenization

1. The homogenized equations are those of the elastostatics of a linear elastic solid, albeit one whose modulus of elasticity depends on the size of the liquid inclusions

$$\overline{L}_{ijkl}=\int_{\mathcal{Y}_0} L_{ijmn}(\mathbf{Y})\left(\delta_{mk}\delta_{nl}+\frac{\partial\omega_{mkl}}{\partial Y_n}(\mathbf{Y})\right)\mathrm{d}\mathbf{Y}+\int_{\mathcal{G}_0}\widehat{L}_{ijmn}\left(\delta_{mk}\widehat{I}_{nl}+\frac{\partial\omega_{mkl}}{\partial Y_p}(\mathbf{Y})\widehat{I}_{pn}\right)\mathrm{d}\mathbf{Y}$$

$$\mathbf{L}(\mathbf{Y})=(1-\theta(\mathbf{Y}))\,\mathbf{L}^{(\mathtt{m})}+\sum_{I=1}^{\mathtt{N}}\theta_I(\mathbf{Y})\left[3\Lambda^{(\mathtt{i})}\boldsymbol{\mathcal{J}}-\boxed{\frac{2\widehat{\gamma}_0}{A_I}}(\boldsymbol{\mathcal{A}}-\boldsymbol{\mathcal{K}}+2\boldsymbol{\mathcal{J}})\right]$$

2. In contrast to the local behavior, the resulting homogenized solid is free of macroscopic residual stresses

$$\begin{cases}\mathrm{Div}\left[\boxed{\overline{\mathbf{L}}\,\nabla\mathbf{u}}\right]=\mathbf{0}, & \mathbf{X}\in\Omega_0\\ \mathbf{u}(\mathbf{X})=\overline{\mathbf{u}}(\mathbf{X}), & \mathbf{X}\in\partial\Omega_0\end{cases}$$

why? the average of the local residual stress and initial surface tension cancel each other out

$$-\int_{\mathcal{Y}_0}\sum_{I=1}^{\mathtt{N}}\theta_I(\mathbf{Y})\frac{2\widehat{\gamma}_0}{A_I}\mathbf{I}\,\mathrm{d}\mathbf{Y}+\int_{\mathcal{G}_0}\widehat{\gamma}_0\,\widehat{\mathbf{I}}\,\mathrm{d}\mathbf{Y}=\mathbf{0}$$

3. Remarkably, in spite of the lack of minor symmetry of $\mathbf{L}$ and $\widehat{\mathbf{L}}$, the resulting effective modulus of elasticity possesses the standard major and minor symmetries of a classical linear elastic solid

$$\boxed{\overline{L}_{ijkl}=\overline{L}_{klij}\quad\text{and}\quad\overline{L}_{ijkl}=\overline{L}_{jikl}=\overline{L}_{ijlk}}$$

Elastomers filled with *spherical* liquid inclusions: Homogenization

4. The **strain macro-variable** that emerges from the homogenization limit is given by

$$E_{ij}(\mathbf{X}) = \frac{1}{2}\left(\frac{\partial u_i}{\partial X_j}(\mathbf{X}) + \frac{\partial u_j}{\partial X_i}(\mathbf{X})\right)$$

5. The **stress macro-variable** that emerges from the homogenization limit is given by

$$S_{ij}(\mathbf{X}) = \int_{\mathcal{Y}_0} L_{ijkl}(\mathbf{Y})\left(\frac{\partial u_k}{\partial X_l}(\mathbf{X}) + \frac{\partial u_k^{(1)}}{\partial Y_l}(\mathbf{X},\mathbf{Y})\right)\mathrm{d}\mathbf{Y} + \int_{\mathcal{G}_0} \widehat{L}_{ijkl}\left(\frac{\partial u_k}{\partial X_p}(\mathbf{X}) + \frac{\partial u_k^{(1)}}{\partial Y_p}(\mathbf{X},\mathbf{Y})\right)\widehat{I}_{pl}\mathrm{d}\mathbf{Y} = \overline{L}_{ijkl}E_{kl}(\mathbf{X})$$

average of the local stress in the bulk   average of the local stress on the interface

6. The homogenized response is actually that of a hyperelastic solid with an **effective stored-energy function**

$$S_{ij} = \frac{\partial \overline{W}}{\partial E_{ij}}(\mathbf{E})$$

where

$$\begin{aligned}\overline{W}(\mathbf{E}) =& \frac{1}{2}\int_{\mathcal{Y}_0}\left(\frac{\partial u_i}{\partial X_j}(\mathbf{X}) + \frac{\partial u_i^{(1)}}{\partial Y_j}(\mathbf{X},\mathbf{Y})\right)L_{ijkl}(\mathbf{Y})\left(\frac{\partial u_k}{\partial X_l}(\mathbf{X}) + \frac{\partial u_k^{(1)}}{\partial Y_l}(\mathbf{X},\mathbf{Y})\right)\mathrm{d}\mathbf{Y} + \\ & \frac{1}{2}\int_{\mathcal{G}_0}\left(\frac{\partial u_i}{\partial X_p}(\mathbf{X}) + \frac{\partial u_i^{(1)}}{\partial Y_p}(\mathbf{X},\mathbf{Y})\right)\widehat{I}_{pj}\widehat{L}_{ijkl}\left(\frac{\partial u_k}{\partial X_q}(\mathbf{X}) + \frac{\partial u_k^{(1)}}{\partial Y_q}(\mathbf{X},\mathbf{Y})\right)\widehat{I}_{ql}\,\mathrm{d}\mathbf{Y} \\ =& \frac{1}{2}E_{ij}\overline{L}_{ijkl}E_{kl}\end{aligned}$$

Homogenization:
Finite Deformations

Elastomers filled with *spherical* liquid inclusions: Homogenization

Given the homogenization result in the small-deformation limit, the expectation is that one can pass to the limit as $\delta \searrow 0$ in the equations for finite deformation

$$\begin{cases} \mathrm{Div}\left[r_{\mathtt{i}}^{\sharp}(\delta^{-1}\mathbf{X})J^{\delta}\nabla\mathbf{y}^{\delta^{-T}} + \mu^{\sharp}(\delta^{-1}\mathbf{X})\left(\nabla\mathbf{y}^{\delta} - \nabla\mathbf{y}^{\delta^{-T}}\right) + \Lambda^{\sharp}(\delta^{-1}\mathbf{X})(J^{\delta}-1)J^{\delta}\nabla\mathbf{y}^{\delta^{-T}}\right] = \mathbf{0}, \quad \mathbf{X} \in \Omega_0 \setminus \Gamma_0 \\ \widehat{\mathrm{Div}}\left[\widehat{\gamma}_0\, \widehat{J}^{\delta}\widehat{\nabla}\mathbf{y}^{\delta^{-T}} + \widehat{\mu}(\widehat{\nabla}\mathbf{y}^{\delta} - \widehat{\nabla}\mathbf{y}^{\delta^{-T}}) + \widehat{\Lambda}(\widehat{J}^{\delta}-1)\widehat{J}^{\delta}\widehat{\nabla}\mathbf{y}^{\delta^{-T}}\right] - \\ \left[\!\left[r_{\mathtt{i}}^{\sharp}(\delta^{-1}\mathbf{X})J^{\delta}\nabla\mathbf{y}^{\delta^{-T}} + \mu^{\sharp}(\delta^{-1}\mathbf{X})\left(\nabla\mathbf{y}^{\delta} - \nabla\mathbf{y}^{\delta^{-T}}\right) + \Lambda^{\sharp}(\delta^{-1}\mathbf{X})(J^{\delta}-1)J^{\delta}\nabla\mathbf{y}^{\delta^{-T}} \right]\!\right] \widehat{\mathbf{N}} = \mathbf{0}, \quad \mathbf{X} \in \Gamma_0 \\ \mathbf{y}^{\delta}(\mathbf{X}) = \overline{\mathbf{y}}(\mathbf{X}), \quad \mathbf{X} \in \partial\Omega_0^{\mathcal{D}} \\ \left[\mu_{\mathtt{m}}\left(\nabla\mathbf{y}^{\delta} - \nabla\mathbf{y}^{\delta^{-T}}\right) + \Lambda_{\mathtt{m}}(J^{\delta}-1)J^{\delta}\nabla\mathbf{y}^{\delta^{-T}}\right]\mathbf{N} = \overline{\mathbf{s}}(\mathbf{X}), \quad \mathbf{X} \in \partial\Omega_0^{\mathcal{N}} \end{cases}$$

and show that the deformation field $\mathbf{y}^{\delta}(\mathbf{X})$ converges to a macroscopic deformation field $\mathbf{y}(\mathbf{X})$ solution of the **elastostatics equations for a hyperelastic solid:**

$$\begin{cases} \mathrm{Div}\left[\dfrac{\partial \overline{W}}{\partial \overline{\mathbf{F}}}(\nabla\mathbf{y})\right] = \mathbf{0}, \quad \mathbf{X} \in \Omega_0 \\ \mathbf{y}(\mathbf{X}) = \overline{\mathbf{y}}(\mathbf{X}), \quad \mathbf{X} \in \partial\Omega_0^{\mathcal{D}} \\ \left[\dfrac{\partial \overline{W}}{\partial \overline{\mathbf{F}}}(\nabla\mathbf{y})\right]\mathbf{N} = \overline{\mathbf{s}}(\mathbf{X}), \quad \mathbf{X} \in \partial\Omega_0^{\mathcal{N}} \end{cases}$$

Ghosh & Lopez-Pamies *JMPS* (2022)

Elastomers filled with *spherical* liquid inclusions: Homogenization

Moreover, the expectation is that **the effective stored-energy function $\overline{W}$ is given by the formula**

$$\overline{W}(\overline{\mathbf{F}})=\frac{1}{|\mathcal{Y}_0^{\mathbf{k}}|}\left(\int_{\mathcal{Y}_0^{\mathbf{k}}}\underbrace{W(\mathbf{Y},\nabla\boldsymbol{\chi})}_{\text{bulk energy}}\,\mathrm{d}\mathbf{Y}+\int_{\mathcal{G}_0^{\mathbf{k}}}\underbrace{\widehat{W}(\widehat{\nabla}\boldsymbol{\chi})}_{\text{interface energy}}\,\mathrm{d}\mathbf{Y}\right)$$

where $\boldsymbol{\chi}(\mathbf{Y})$ is a $\mathcal{Y}_0^{\mathbf{k}}$-periodic function solution of a **super-cell problem**

Remark: This generalizes the formula of Braides (1985)/Müeller (1987) to hyperelastic materials with residual stresses in the bulk and interfacial forces

1. The homogenized equations are those of the elastostatics of a hyperelastic solid, one whose elasticity depends on the size of the liquid inclusions

2. The resulting homogenized solid is free of macroscopic residual stresses $\dfrac{\partial\overline{W}}{\partial\overline{\mathbf{F}}}(\mathbf{I})=\mathbf{0}$

3. In the small-deformation limit, the resulting effective modulus of elasticity possesses the standard major and minor symmetries

$$\overline{L}_{ijkl}=\overline{L}_{klij}\quad\text{and}\quad\overline{L}_{ijkl}=\overline{L}_{jikl}=\overline{L}_{ijlk}$$

where

$$\overline{L}_{ijkl}:=\frac{\partial^2\overline{W}}{\partial\overline{F}_{ij}\partial\overline{F}_{kl}}(\mathbf{I})$$

Elastomers filled with *spherical* liquid inclusions: Homogenization

4. The above definition of homogenized behavior is consistent with the formal definition based on the relation between the **macroscopic stress**

$$\overline{\mathbf{S}} := \frac{1}{|\Omega_0|} \int_{\partial\Omega_0} \mathbf{S}\mathbf{N} \otimes \mathbf{X} \, \mathrm{d}\mathbf{X}$$

and the **macroscopic deformation gradient**

$$\overline{\mathbf{F}} := \frac{1}{|\Omega_0|} \int_{\partial\Omega_0} \mathbf{y} \otimes \mathbf{N} \, \mathrm{d}\mathbf{X}$$

when the body is subjected to **affine boundary conditions**

Sample Results for a Basic Case:

Isotropic Suspension of Monodisperse Incompressible Inclusions

Isotropic suspensions of monodisperse incompressible inclusions

Representative unit cell

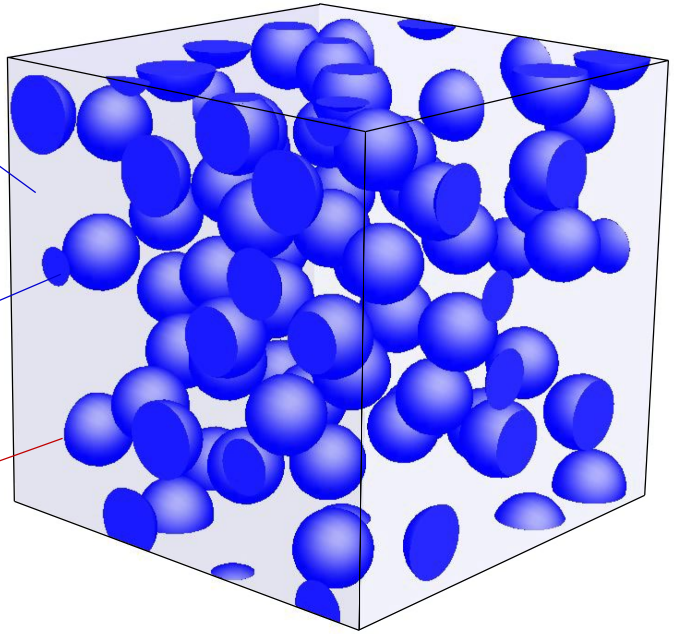
$$\Lambda_{\tt m} = +\infty$$

$$W_{\tt m}(\mathbf{F}) = \frac{\mu_{\tt m}}{2}\left[\mathbf{F}\cdot\mathbf{F} - 3\right] - \mu_{\tt m}\ln J + \frac{\Lambda_{\tt m}}{2}(J-1)^2$$

$$\Lambda_{\tt i} = +\infty$$

$$W_{\tt i}(J) = -\frac{2\widehat{\gamma}_0}{A}J + \frac{\Lambda_{\tt i}}{2}(J-1)^2$$

$$\widehat{W}(\widehat{\mathbf{F}}) = \widehat{\gamma}_0\,\widehat{J}$$



Inclusion radius: A

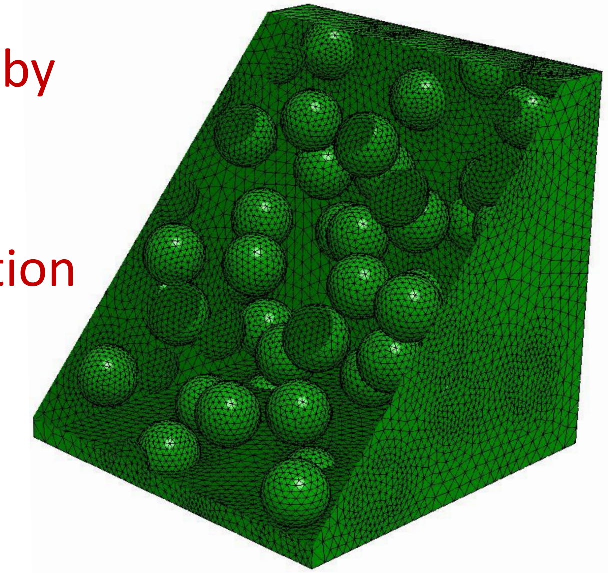
Inclusion volume fraction: c

In this basic case, one can define a unique ***initial* elasto-capillary number:**

$$\boxed{eCa := \frac{\widehat{\gamma}_0}{2\mu_{\tt m}A}}$$

We generate solutions numerically by a **hybrid finite-element scheme:**

Representative FE discretization



Ghosh & Lopez-Pamies *JMPS* (2022); Ghosh, Lefèvre, Lopez-Pamies (2022)

Sample results: the small-deformation limit

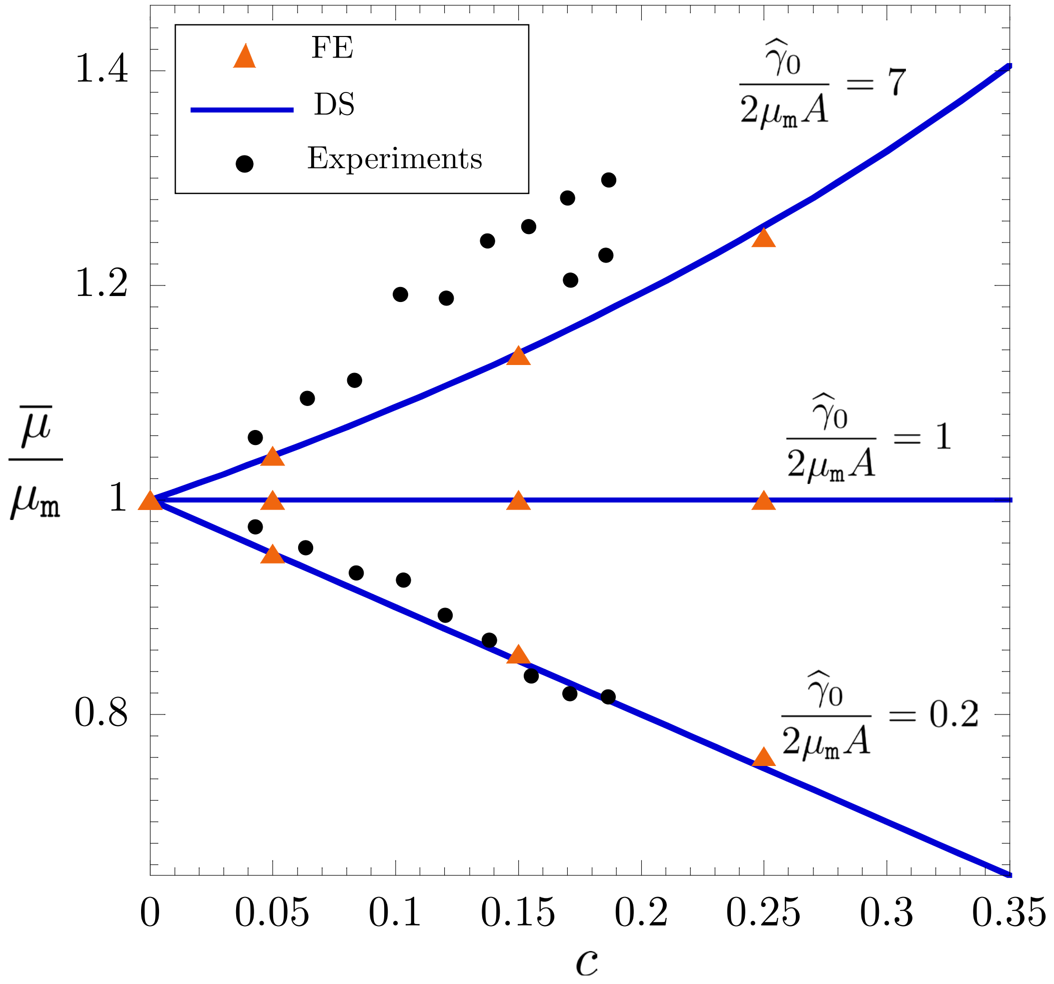
In the **limits of small deformations and dilute volume fractions of inclusions** it is possible to determine an analytical solution for the effective modulus of elasticity:

$$\overline{\mathbf{L}} = 2\,\overline{\mu}^{\,\mathrm{dil}}\,\mathcal{K} + \infty\mathcal{J} + O(c^2) \quad \text{with} \quad \boxed{\overline{\mu}^{\,\mathrm{dil}} = \mu_{\mathtt{m}} + \frac{5\,(eCa-1)}{3+5\,eCa}\,\mu_{\mathtt{m}}\,c}$$

This basic dilute solution can be exploited within a **generalized differential scheme** that accounts for residual stresses and interfacial forces to determine solutions for **suspensions with finite volume fraction of inclusions**. The simplest among this yields:

$$\overline{\mathbf{L}} = 2\,\overline{\mu}^{\,\mathrm{DS}}\,\mathcal{K} + \infty\mathcal{J} \quad \text{with} \quad \boxed{\overline{\mu}^{\,\mathrm{DS}} = \frac{\mu_{\mathtt{m}}}{(1-c)^{\frac{5(eCa-1)}{3+5eCa}}}}$$

Experiments: Glycerol droplets (A=1 μm) in two silicone elastomers ($\mu_{\mathtt{m}}$=1 & 33 kPa)



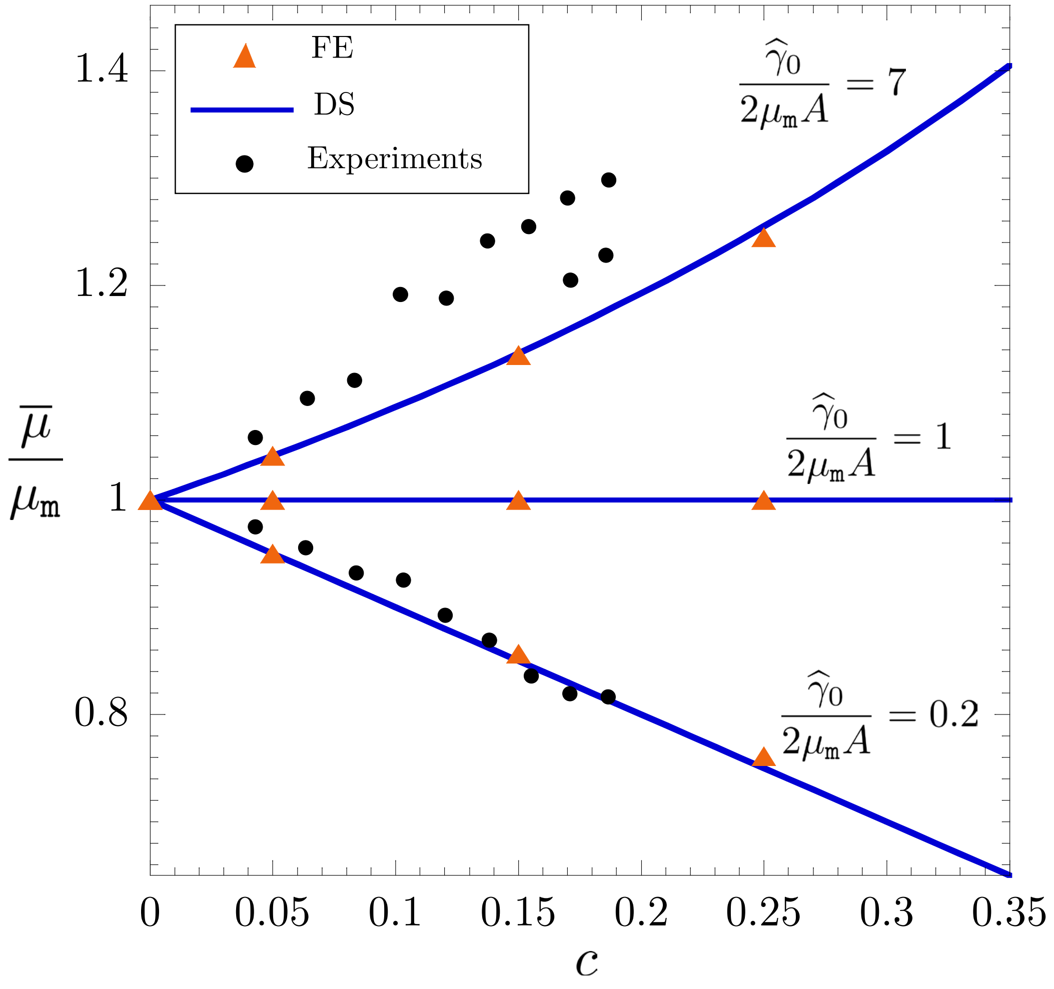
Style et al. (2015); Ghosh & Lopez-Pamies *JMPS* (2022)

Sample results: the small-deformation limit

The effective shear modulus is such that

$$\begin{cases} \overline{\mu} > \mu \quad \text{if } \ eCa > 1 & \text{stiffening} \\ \overline{\mu} = \mu \quad \text{if } \ eCa = 1 & \text{“cloaking”} \\ \overline{\mu} < \mu \quad \text{if } \ eCa < 1 & \text{softening} \end{cases}$$

Experiments: Glycerol droplets (A=1 μm) in two silicone elastomers ($\mu_{\mathtt{m}}$=1 & 33 kPa)



Sample results: finite deformations

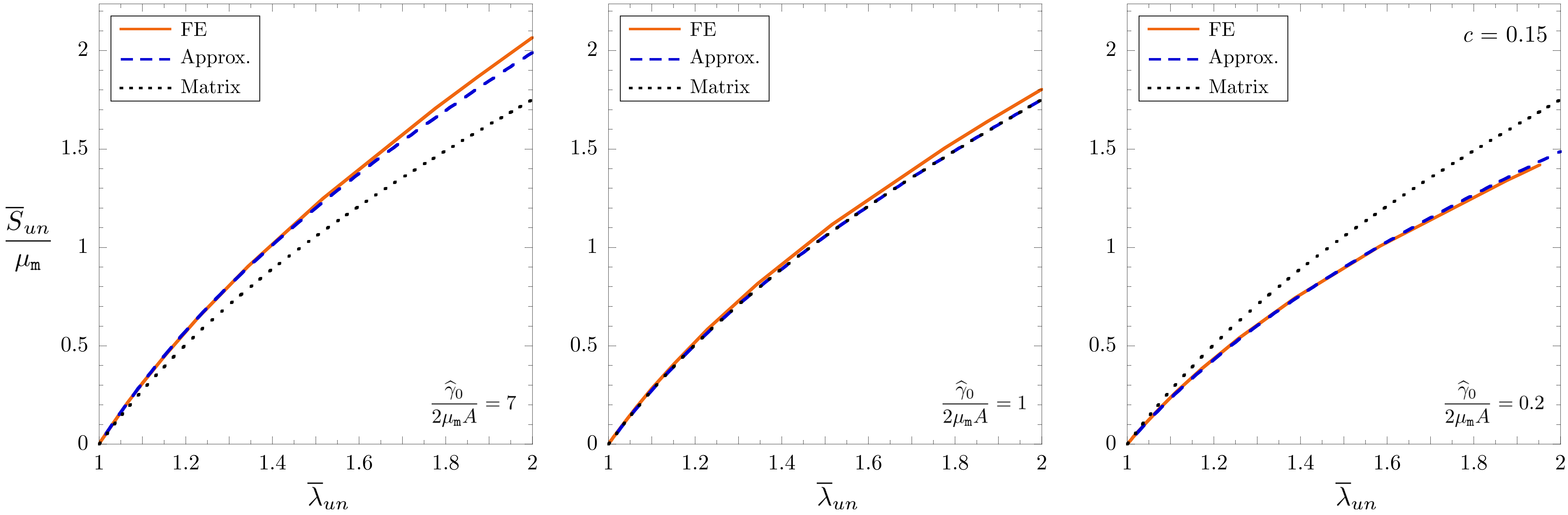
Stress-stretch response under **uniaxial tension**

$$\overline{\mathbf{F}} = \overline{\lambda}_{un}\mathbf{e}_1 \otimes \mathbf{e}_1 + \overline{\lambda}_{un}^{-1/2}(\mathbf{e}_2 \otimes \mathbf{e}_2 + \mathbf{e}_3 \otimes \mathbf{e}_3)$$

with $\overline{\mathbf{S}} = \overline{S}_{un}\mathbf{e}_1 \otimes \mathbf{e}_1$

Even at finite deformations we have

$$\begin{cases} \overline{S} > S_{\mathtt{m}} & \text{if } eCa > 1 \quad \text{stiffening} \\ \overline{S} \approx S_{\mathtt{m}} & \text{if } eCa = 1 \quad \text{"cloaking"} \\ \overline{S} < S_{\mathtt{m}} & \text{if } eCa < 1 \quad \text{softening} \end{cases}$$



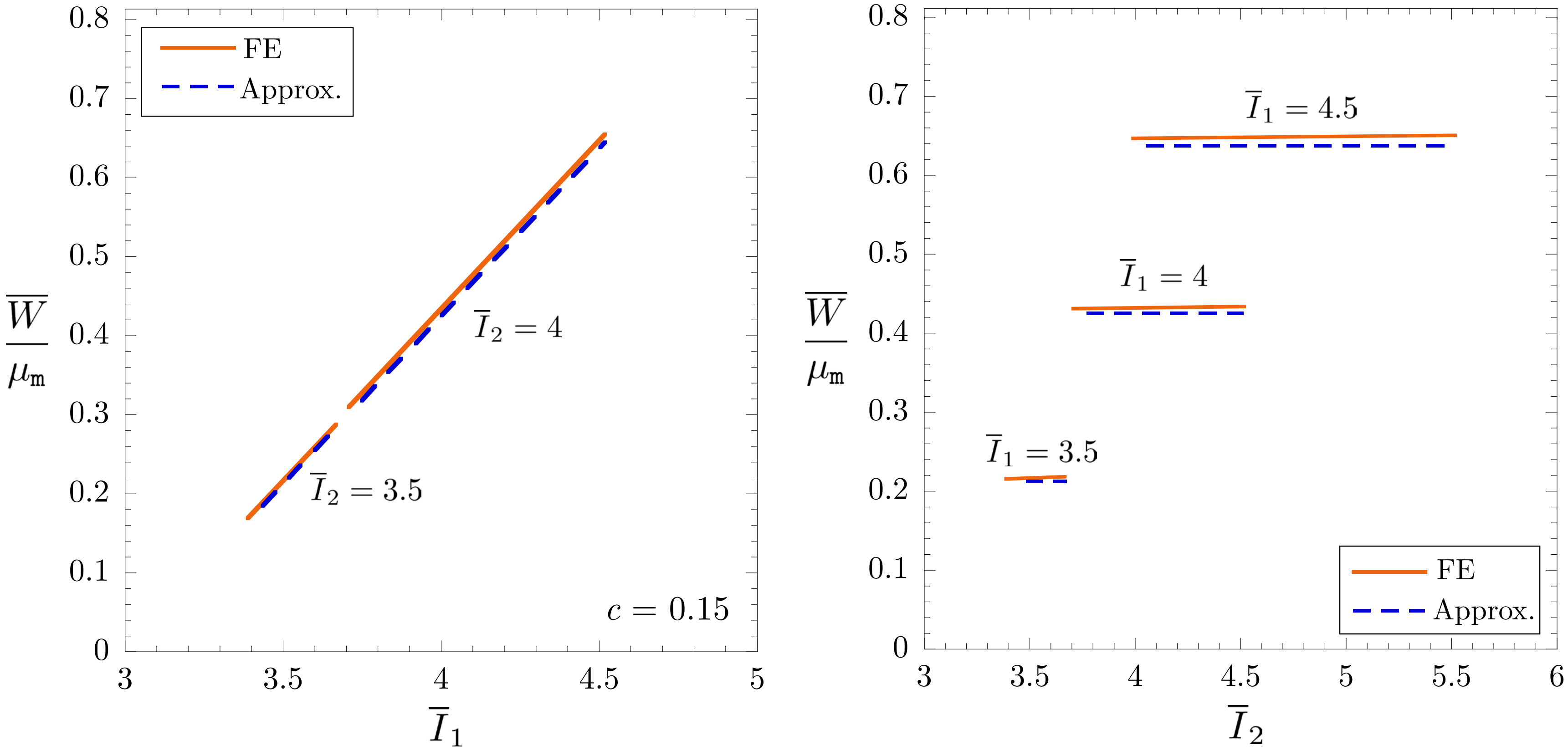
Sample results: finite deformations

A simple explicit approximation:

$$\overline{W}(\overline{\mathbf{F}}) = \begin{cases} \dfrac{\overline{\mu}}{2}\left[\overline{I}_1 - 3\right] & \text{if } \det \overline{\mathbf{F}} = 1 \\ +\infty & \text{otherwise} \end{cases}$$

with

$$\overline{\mu} = \frac{\mu_{\mathtt{m}}}{(1-c)^{\frac{5(eCa-1)}{3+5eCa}}}$$



Approximately **linear** in I_1

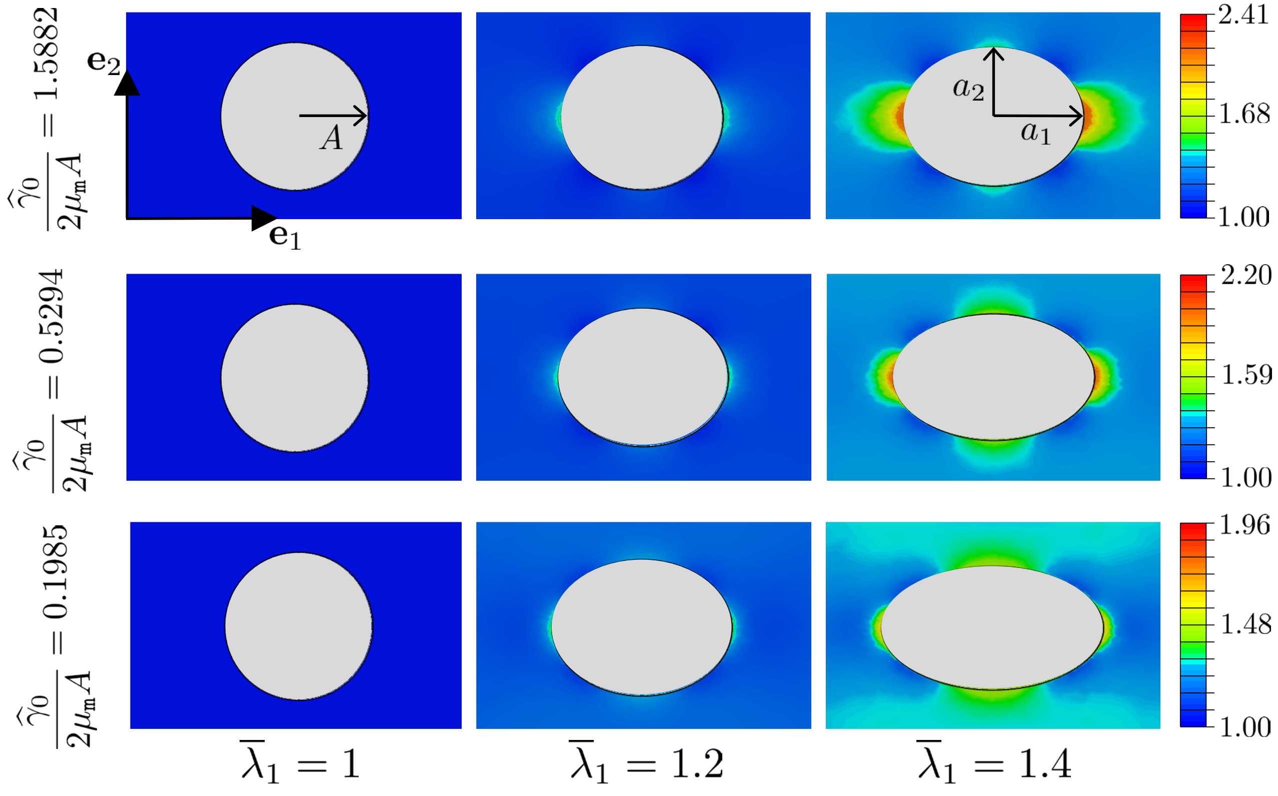
Approximately **independent** of I_2

This is the same behavior that isotropic Neo-Hookean composites exhibit in the absence of residual stresses and interface forces

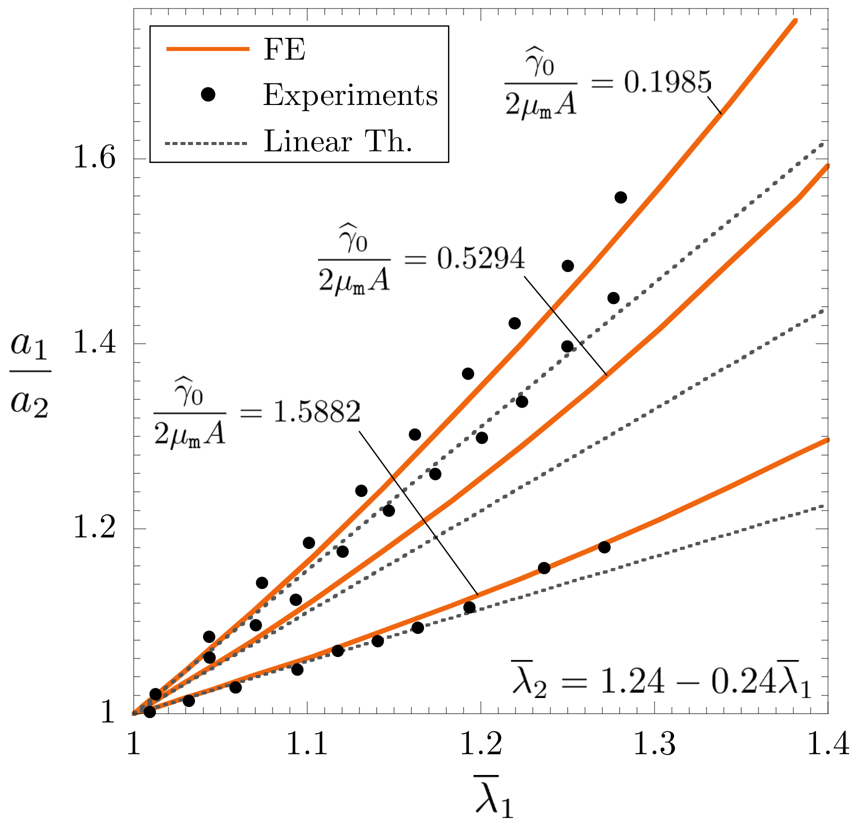
Ghosh & Lopez-Pamies *JMPS* (2022) ; Lopez-Pamies, Goudarzi, Danas *JMPS* (2013)

Sample results: finite deformations

Local deformation in and around isolated inclusions with three different values of *initial* elasto-capillary number



Experiments: Ionic-liquid droplets (A=2, 6, 16μm) in a silicone elastomer



Style et al. (2015); Ghosh & Lopez-Pamies *JMPS* (2022)

Summary and take-home messages

- We have formulated (neglecting dissipative phenomena) the homogenization problem that describes the mechanical response of elastomers filled with liquid inclusions under finite quasistatic deformations

- The resulting macroscopic behavior is that of a hyperelastic solid, albeit one that depends on the size of the inclusions

- Even though there are local residual stresses within the inclusions, the resulting macroscopic behavior turns out to be free of residual stresses

- Even though the initial local moduli do *not* possess minor symmetries, the resulting effective modulus does

- The equations point the way to new existence theorems (Lax-Milgram does *not* apply)

- Controlling the elastocapillarity of elastomers filled with liquid inclusions provides an exciting pathway to drastically change macroscopic behaviors (from softening to "cloacking" to stiffening)

- The surface tension at deforming solid/liquid interfaces is likely strongly nonlinear. Experiments are needed!