Nonlinear Electroelastic Deformations of Dielectric Elastomer Composites

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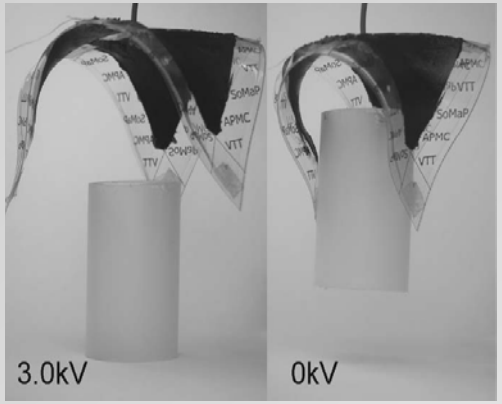
Work supported by the National Science Foundation (DMS, CMMI)

Dielectric elastomers

Energy Harvesters

Actuators



. . .

Examples: (pretty much) any elastomer

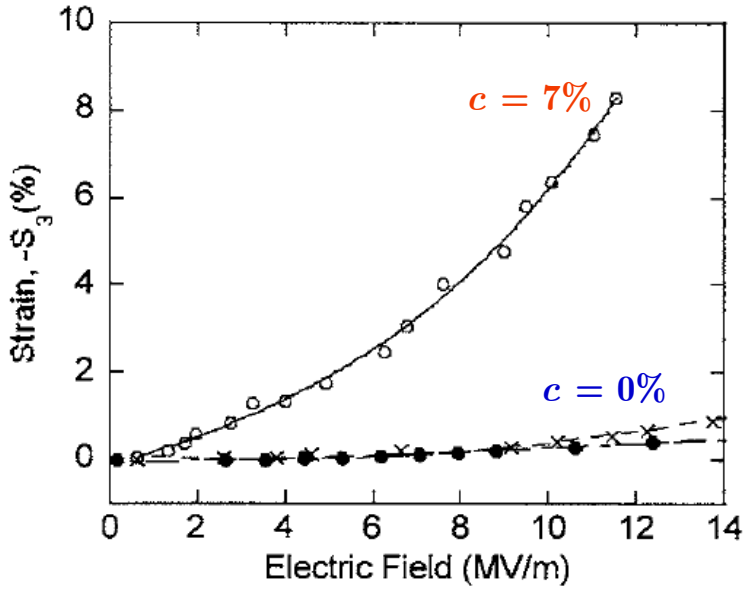
Advantages: finitely deformable — short response times — light — inexpensive — biocompatible

Disadvantage: { **require very large electric fields for actuation** (in the order of 100 MV/m)

Idea: add high-dielectric (or conducting) particles to **make dielectric composites** that can finitely deform under moderate electric fields

SRI (2009); Kofod et al. (2007); Carpi et al. (2010); Aschwanden & Stemmer (2006)

Dielectric elastomer composites



Matrix: **PU** { dielectric constant: **10**; shear modulus: **10 MPa**

Particles: **PAA decorated o-CuPc** { dielectric constant: **10^4**; shear modulus: **1 GPa**

Microstructure: **isotropic distribution of roughly spherical nano-particles at 7% vol. fraction**

Competition of effects: adding high-dielectric particles increases the overall dielectric constant of the composite, but it may also make it stiffer, and hence less deformable

- Initial conjecture in the literature (Li, 2003; Tian et al., 2012): **Enhancement is due to the heightened role of field fluctuations due to the nonlinearity of elastomers**

- Alternative conjecture (Lewis, 2004; Lopez-Pamies et al., 2014): **Enhancement is due to interphasial phenomena including free charges**

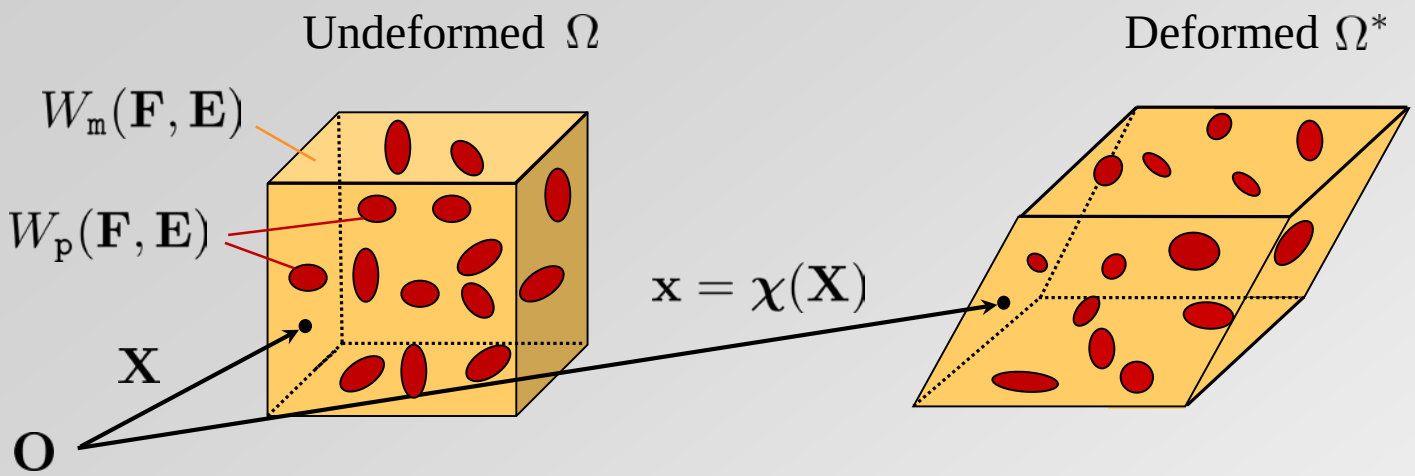
Huang et al. (2005)

Problem Formulation

(elastic dielectrics)

Problem setting: 2-phase particulate material

Kinematics



Deformation Gradient

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

Lagrangian Electric Field

$$\mathbf{E}$$

Local constitutive behavior

Total nominal stress

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

Lagrangian electric displacement

$$\mathbf{D} = -\frac{\partial W}{\partial \mathbf{E}}(\mathbf{X},\mathbf{F},\mathbf{E})$$

where

$$W(\mathbf{X},\mathbf{F},\mathbf{E}) = [1-\theta(\mathbf{X})]W_{\mathtt{m}}(\mathbf{F},\mathbf{E}) + \theta(\mathbf{X})W_{\mathtt{p}}(\mathbf{F},\mathbf{E})$$

Indicator function characterizing the initial microstructure

$$\theta(\mathbf{X}) = \begin{cases} 1 & \text{if } \mathbf{X} \in \text{particle} \\ 0 & \text{otherwise} \end{cases}$$

Dorfmann & Ogden (2005)

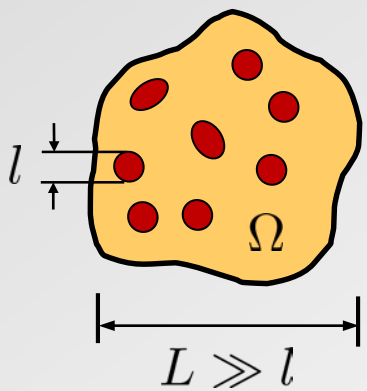
Toupin (1956)

Problem setting: macroscopic response

Definition: relation between the volume averages of $\mathbf{S}$, $\mathbf{D}$ and $\mathbf{F}$, $\mathbf{E}$

Macroscopic **deformation gradient** & **electric field**

$$\overline{\mathbf{F}} = \frac{1}{|\Omega|}\int_\Omega \mathbf{F}\,\mathrm{d}\mathbf{X} \qquad \overline{\mathbf{E}} = \frac{1}{|\Omega|}\int_\Omega \mathbf{E}\,\mathrm{d}\mathbf{X}$$



BC's

$$\Phi = -\overline{\mathbf{E}}\cdot\mathbf{X}$$
$$\mathbf{x} = \overline{\mathbf{F}}\mathbf{X}$$

Macroscopic **nominal stress** & **electric displacement**

$$\overline{\mathbf{S}} \doteq \frac{1}{|\Omega|}\int_\Omega \mathbf{S}\,\mathrm{d}\mathbf{X} \qquad \overline{\mathbf{D}} \doteq \frac{1}{|\Omega|}\int_\Omega \mathbf{D}\,\mathrm{d}\mathbf{X}$$

Variational Characterization

$$\left.\begin{aligned} \overline{\mathbf{S}} &= \frac{\partial \overline{W}}{\partial \overline{\mathbf{F}}}(\overline{\mathbf{F}},\overline{\mathbf{E}},c) \\ \overline{\mathbf{D}} &= -\frac{\partial \overline{W}}{\partial \overline{\mathbf{E}}}(\overline{\mathbf{F}},\overline{\mathbf{E}},c) \end{aligned}\right\} \quad \overline{W}\left(\overline{\mathbf{F}},\overline{\mathbf{E}},c\right) = \min_{\mathbf{F}\in\mathcal{K}}\max_{\mathbf{E}\in\mathcal{E}}\int_\Omega W\left(\mathbf{X},\mathbf{F},\mathbf{E}\right)\mathrm{d}\mathbf{X}$$

Hill (1972)

Lopez-Pamies (2014)

Problem setting: macroscopic response

Definition: relation between the volume averages of $\mathbf{S}$, $\mathbf{D}$ and $\mathbf{F}$, $\mathbf{E}$

Macroscopic **deformation gradient** & **electric field**

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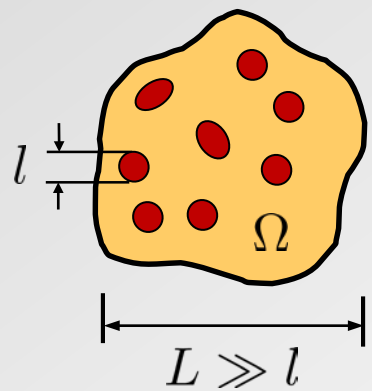

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Euler-Lagrange equations

$$\mathrm{Div}\,\mathbf{S} = \mathbf{0} \quad \text{and} \quad \mathrm{Div}\,\mathbf{D} = 0$$

Two strategies to generate exact solutions

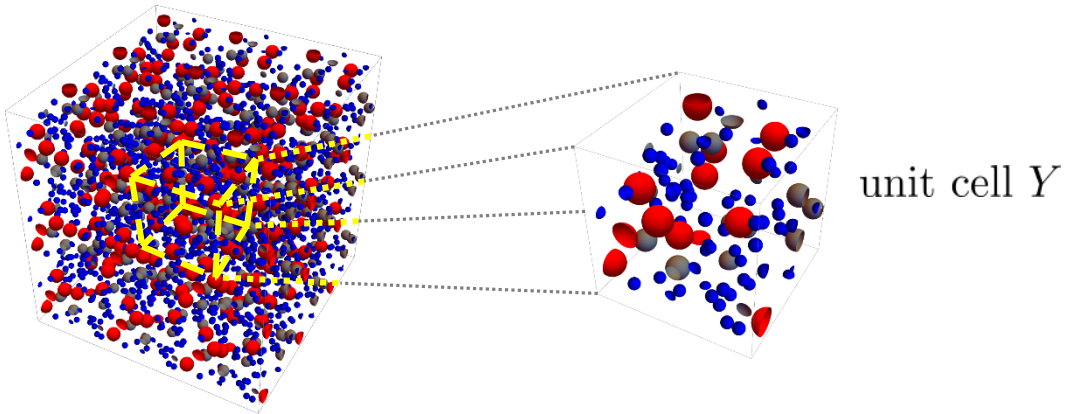
I. **Iterated homogenization** (Lopez-Pamies, 2014): for an *infinitely poly-disperse* particulate microstructure $\overline{W} = \overline{W}(\overline{\mathbf{F}}, \overline{\mathbf{E}}, c)$ is given implicitly by the **nonlinear first-order Hamilton-Jacobi pde**

$$\mathtt{c}\frac{\partial \overline{W}}{\partial \mathtt{c}} - \overline{W} - \left\langle \max_{\boldsymbol{\alpha}} \min_{\beta} \left[\boldsymbol{\alpha} \cdot \frac{\partial \overline{W}}{\partial \overline{\mathbf{F}}} \boldsymbol{\xi} + \beta \frac{\partial \overline{W}}{\partial \overline{\mathbf{E}}} \cdot \boldsymbol{\xi} - W_{\mathtt{m}} \left(\overline{\mathbf{F}} + \boldsymbol{\alpha} \otimes \boldsymbol{\xi}, \overline{\mathbf{E}} + \beta \boldsymbol{\xi} \right) \right] \right\rangle = 0$$

subject to the initial condition $\overline{W}(\overline{\mathbf{F}}, \overline{\mathbf{E}}, 1) = W_{\mathtt{p}}(\overline{\mathbf{F}}, \overline{\mathbf{E}})$

II. **Hybrid finite elements** (Lefèvre & Lopez-Pamies, 2016): periodic homogenization of an infinite medium made out of the repetition of a unit cell containing a random distribution of a *finite number* of particles

$$\overline{W}\left(\overline{\mathbf{F}}, \overline{\mathbf{E}}, c\right) = \min_{\mathbf{u} \in \mathcal{U}} \max_{\Phi \in \mathcal{F}} \max_{p \in \mathcal{P}} \int_Y \left\{ p[\det \mathbf{F}(\mathbf{u}) - 1] - \widehat{W}^*(\mathbf{X}, \mathbf{F}(\mathbf{u}), p, \mathbf{E}(\Phi)) \right\} \mathrm{d}\mathbf{X}$$



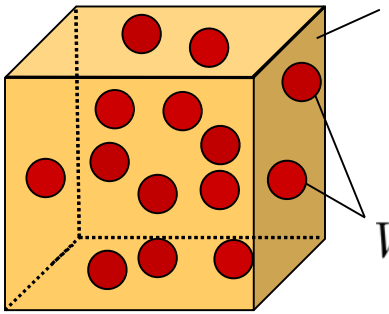
Solutions for

Ideal Elastic Dielectrics

with

Isotropic Microstructures

Isotropic ideal elastic dielectric composites



$$W_{\mathtt{m}}(\mathbf{F},\mathbf{E}) = \begin{cases} \dfrac{\mu}{2}[I_1 - 3] - \dfrac{\varepsilon}{2} I_5^E & \text{if} \quad J = 1 \\ +\infty & \text{otherwise} \end{cases}$$

$$W_{\mathtt{p}}(\mathbf{F},\mathbf{E}) = \begin{cases} \dfrac{\mu_{\mathtt{p}}}{2}[I_1 - 3] - \dfrac{\varepsilon_{\mathtt{p}}}{2} I_5^E & \text{if} \quad J = 1 \\ +\infty & \text{otherwise} \end{cases}$$

Here

$$\begin{cases} I_1 = \mathbf{F}\cdot\mathbf{F} \\ J = \det \mathbf{F} \\ I_5^E = \mathbf{F}^{-T}\mathbf{E}\cdot\mathbf{F}^{-T}\mathbf{E} \end{cases}$$

- In view of the **overall** (constitutive and geometric) **isotropy** and **incompressibility** of these composites, their effective free-energy function is of the form

$$\boxed{\overline{W}(\overline{\mathbf{F}},\overline{\mathbf{E}},c) = \begin{cases} \overline{W}(\overline{I}_1,\overline{I}_2,\overline{I}_4^E,\overline{I}_5^E,\overline{I}_6^E,c) & \text{if} \quad \overline{J} = 1 \\ +\infty & \text{otherwise} \end{cases}}$$

with

$$\overline{I}_1 = \overline{\mathbf{F}}\cdot\overline{\mathbf{F}} \qquad \overline{I}_4^E = \overline{\mathbf{E}}\cdot\overline{\mathbf{E}}$$

$$\overline{I}_2 = \overline{\mathbf{F}}^{-T}\cdot\overline{\mathbf{F}}^{-T} \qquad \overline{I}_5^E = \overline{\mathbf{F}}^{-T}\overline{\mathbf{E}}\cdot\overline{\mathbf{F}}^{-T}\overline{\mathbf{E}}$$

$$\overline{J} = \det\overline{\mathbf{F}} \qquad \overline{I}_6^E = \overline{\mathbf{F}}^{-1}\overline{\mathbf{F}}^{-T}\overline{\mathbf{E}}\cdot\overline{\mathbf{F}}^{-1}\overline{\mathbf{F}}^{-T}\overline{\mathbf{E}}$$

Solution from iterated homogenization

Let us rewrite the pde for $\overline{W} = \overline{W}(\overline{\mathbf{F}}, \overline{\mathbf{E}}, c)$ in the classical form of H-J equations

"time"

$$\frac{\partial \overline{W}}{\partial \mathsf{c}} + \mathcal{H}\left(\overline{\mathbf{F}}, \overline{\mathbf{E}}, \mathsf{c}, \overline{W}, \frac{\partial \overline{W}}{\partial \overline{\mathbf{F}}}, \frac{\partial \overline{W}}{\partial \overline{\mathbf{E}}}\right) = 0, \qquad \overline{W}(\overline{\mathbf{F}}, \overline{\mathbf{E}}, 1) = W_{\mathtt{p}}(\overline{\mathbf{F}}, \overline{\mathbf{E}})$$

"space"

with Hamiltonian

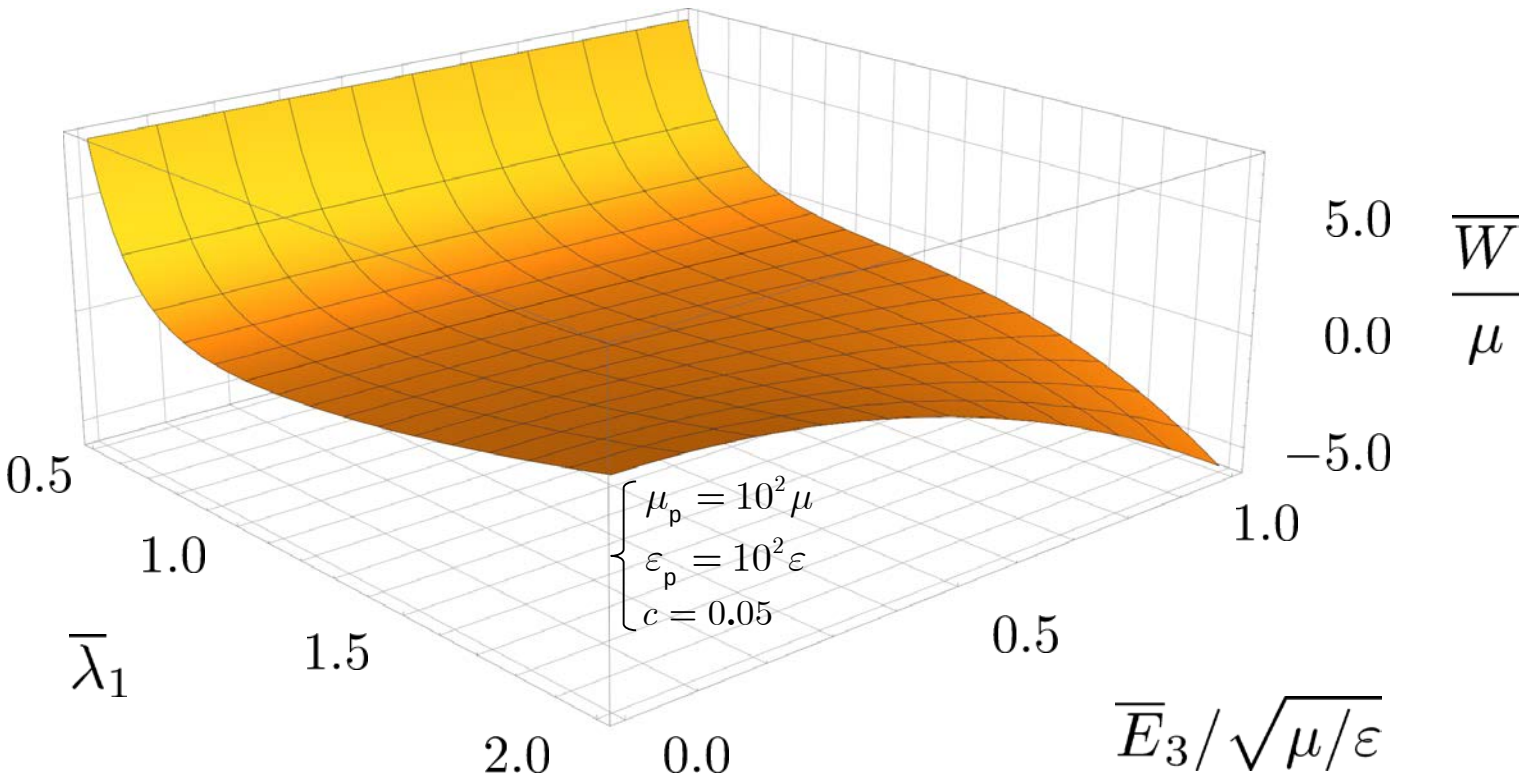
$$\mathcal{H} = -\frac{1}{\mathsf{c}}\overline{W} - \frac{1}{\mathsf{c}}\left\langle \max_{\boldsymbol{\alpha}} \min_{\beta} \left[\boldsymbol{\alpha} \cdot \frac{\partial \overline{W}}{\partial \overline{\mathbf{F}}}\boldsymbol{\xi} + \beta \frac{\partial \overline{W}}{\partial \overline{\mathbf{E}}} \cdot \boldsymbol{\xi} - W_{\mathtt{m}}\left(\overline{\mathbf{F}} + \boldsymbol{\alpha} \otimes \boldsymbol{\xi}, \overline{\mathbf{E}} + \beta\boldsymbol{\xi}\right)\right]\right\rangle$$

- H-J equations typically exhibit **non-uniqueness** of **non-smooth** (Lipschitz cont.) **solutions**
- The physically correct solution is the so-called **viscosity solution**
- Numerical scheme
  - **"space" discretization over a Cartesian grid making use of a *monotone scheme* and *WENO* finite difference *of fifth order***
  - **explicit "time" integration *of fifth order***

Crandall & Lions (1983, 1984)

Osher & Sethian (1988); Jiang & Shu (1996)

IH sample results: $c = 0.05,\ \mu_{\mathrm{p}} = 10^2\mu,\ \varepsilon_{\mathrm{p}} = 10^2\varepsilon$



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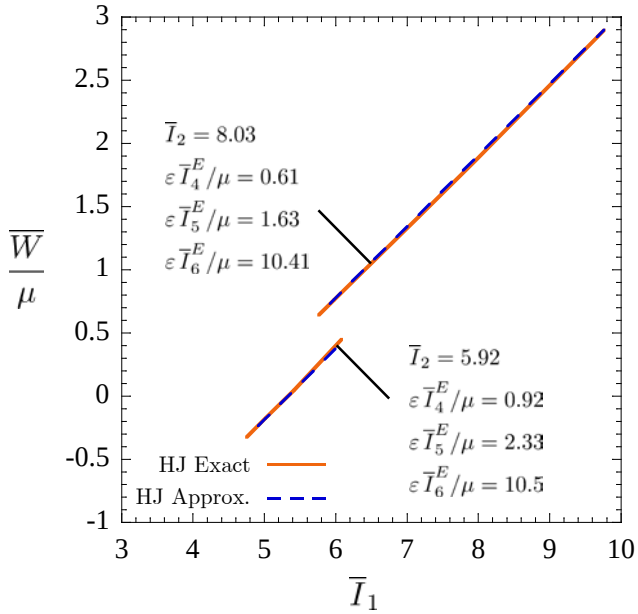
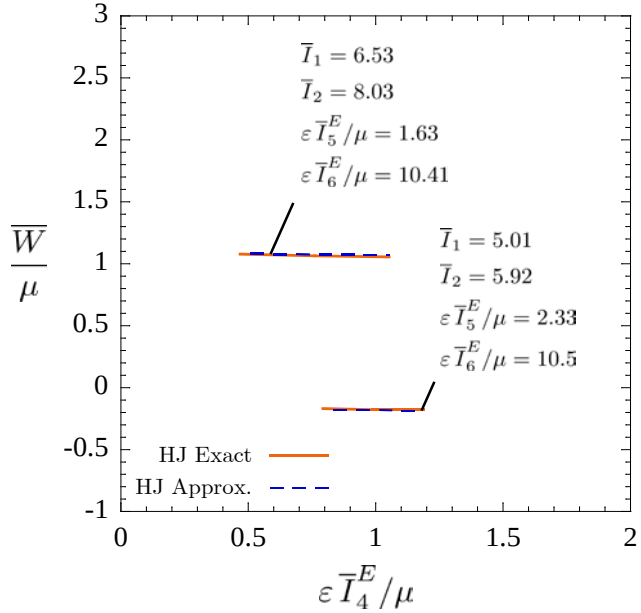
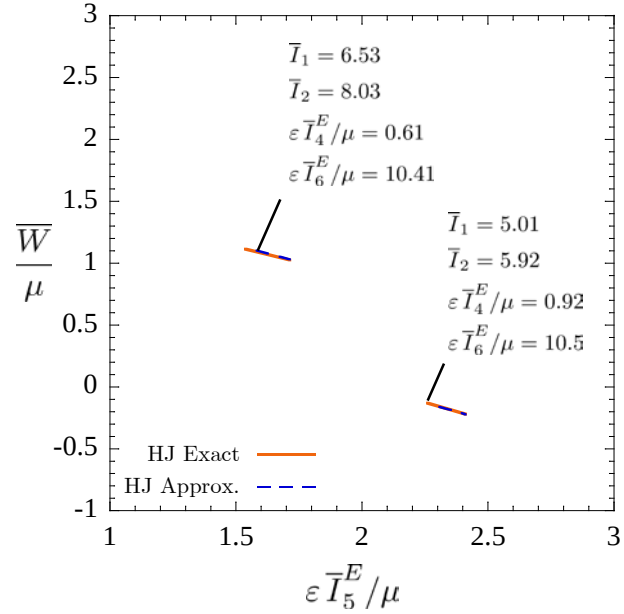
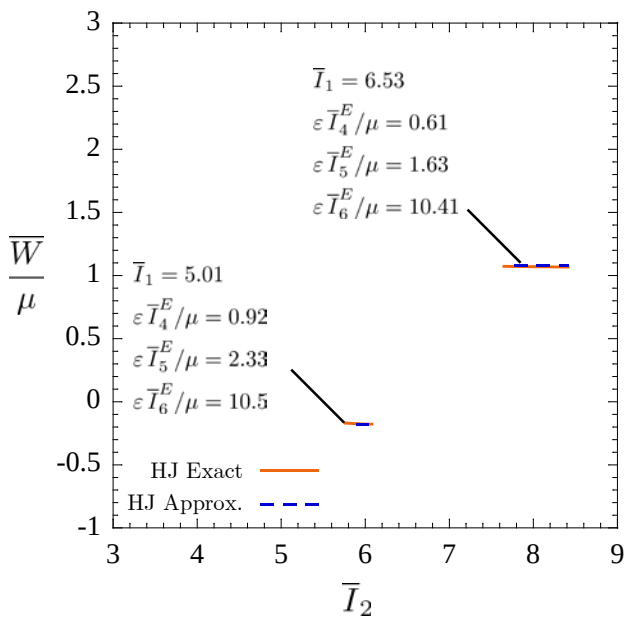
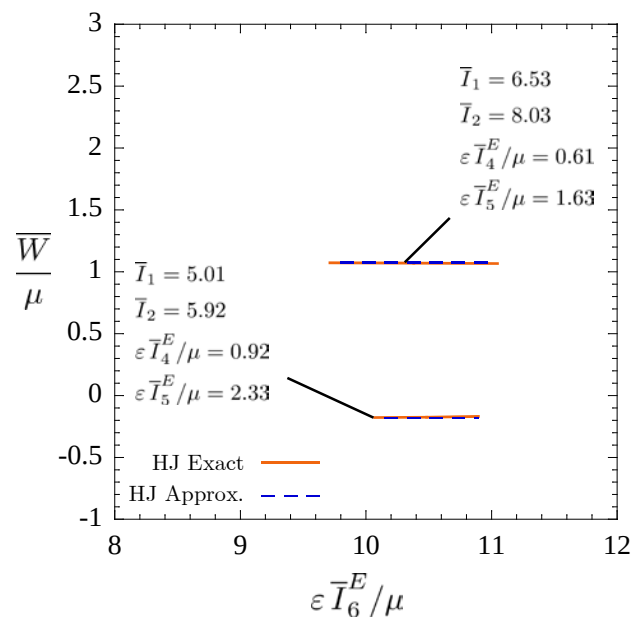










Linear dependence on

$\overline{I}_1,\ \overline{I}_4^E,\ \overline{I}_5^E$

independence of

$\overline{I}_2,\ \overline{I}_6^E$

for all $c,\ \mu,\ \varepsilon,\ \mu_{\mathrm{p}},\ \varepsilon_{\mathrm{p}}$

and all $\overline{\mathbf{F}},\ \overline{\mathbf{E}}$

Solutions via a hybrid FE formulation

With help of the partial Legendre transform

$$\widehat{W}^*(\mathbf{X},\mathbf{F},p,\mathbf{E}) \doteq \max_J \left\{ p(J-1) - \widehat{W}(\mathbf{X},\mathbf{F},J,\mathbf{E}) \right\}$$

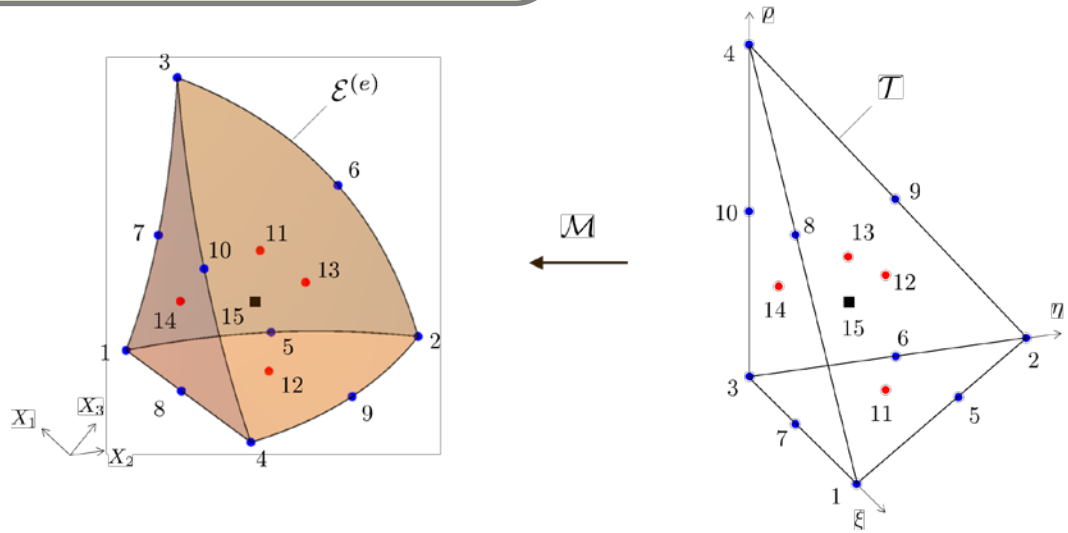
the effective free-energy function can be written as

$$\overline{W}\left(\overline{\mathbf{F}},\overline{\mathbf{E}},c\right) = \min_{\mathbf{u}\in\mathcal{U}} \max_{\Phi\in\mathcal{F}} \max_{p\in\mathcal{P}} \int_Y \left\{ p[\det \mathbf{F}(\mathbf{u}) - 1] - \widehat{W}^*(\mathbf{X},\mathbf{F}(\mathbf{u}),p,\mathbf{E}(\Phi)) \right\} \mathrm{d}\mathbf{X}$$

for Y-periodic microstructures

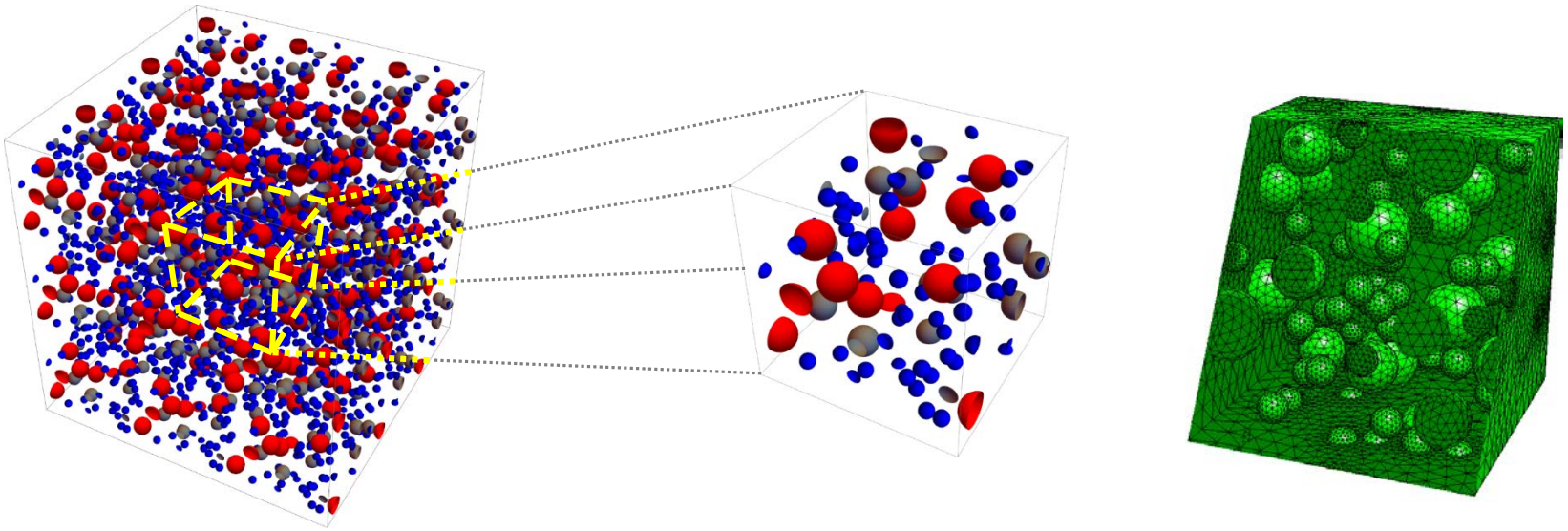
$$\begin{aligned}
&\mathbf{u}(\mathbf{X}) = (\overline{\mathbf{F}} - \mathbf{I})\mathbf{X} + \widetilde{\mathbf{u}}(\mathbf{X}), \text{where } \widetilde{\mathbf{u}} \text{ is } Y\text{-periodic}, \quad \widetilde{\mathbf{u}}(\mathbf{0}) = \mathbf{0},\\
&\Phi(\mathbf{X}) = -\overline{\mathbf{E}}\cdot\mathbf{X} + \widetilde{\Phi}(\mathbf{X}), \text{where } \widetilde{\Phi} \text{ is } Y\text{-periodic}, \quad \widetilde{\Phi}(\mathbf{0}) = 0,\\
&p \text{ is } Y\text{-periodic}
\end{aligned}$$

We look for FE solutions making use of **conforming Crouzeix-Raviart-type elements** — stable and convergent!

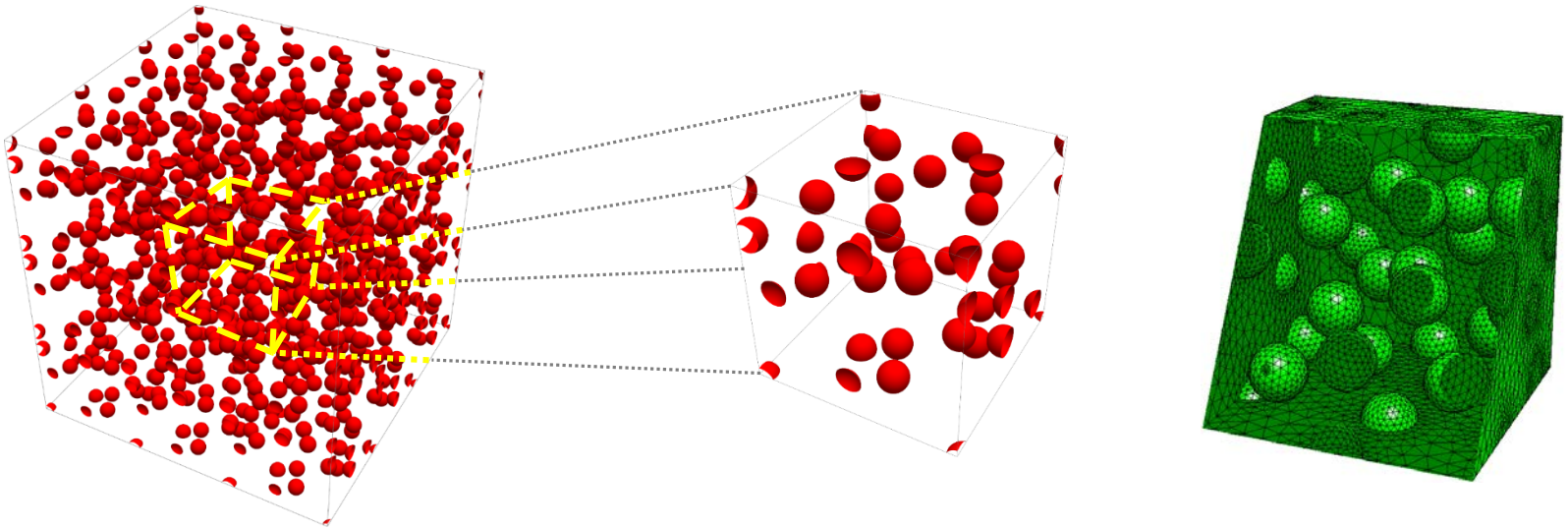


Sample "isotropic" microstructures and meshes

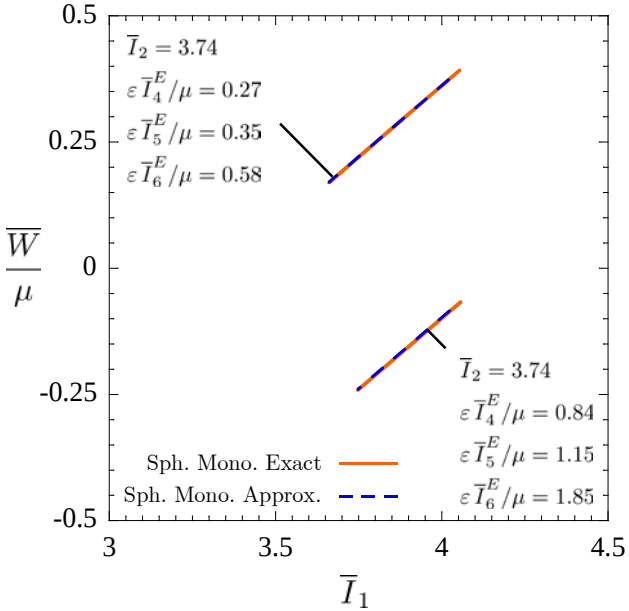
Example: isotropic distribution of 5% vol. fraction of three families of spherical particles

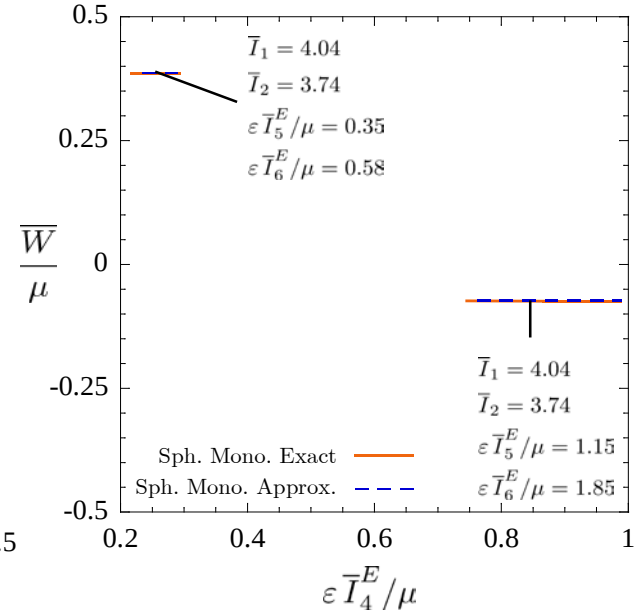


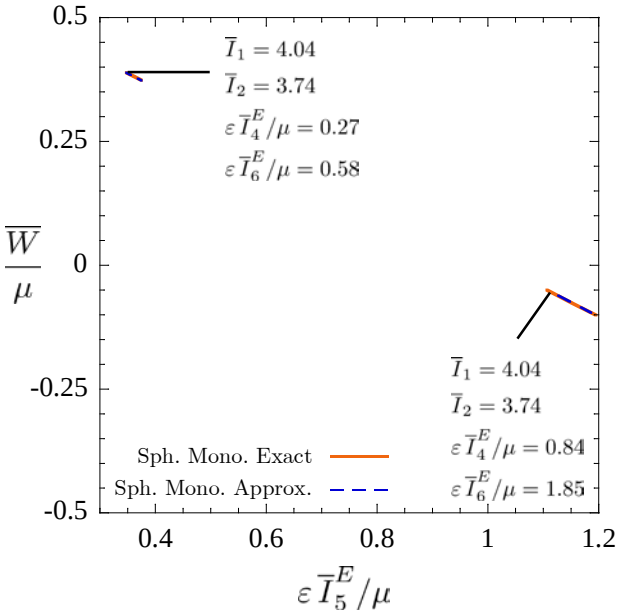
Example: isotropic distribution of 5% vol. fraction of monodisperse spherical particles

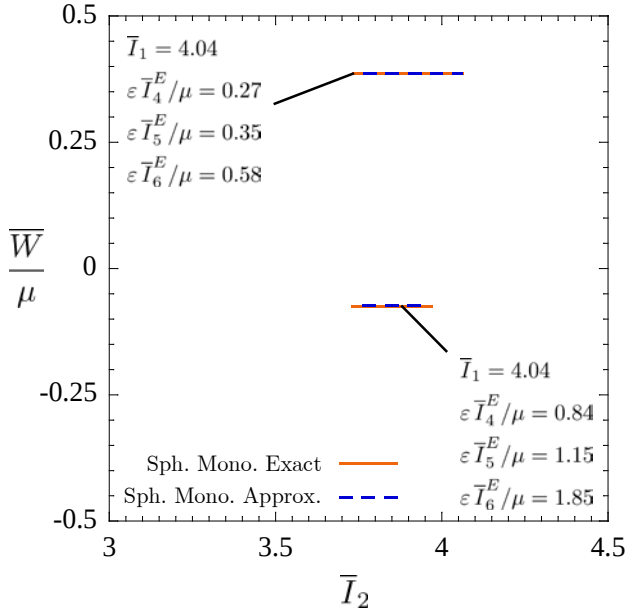


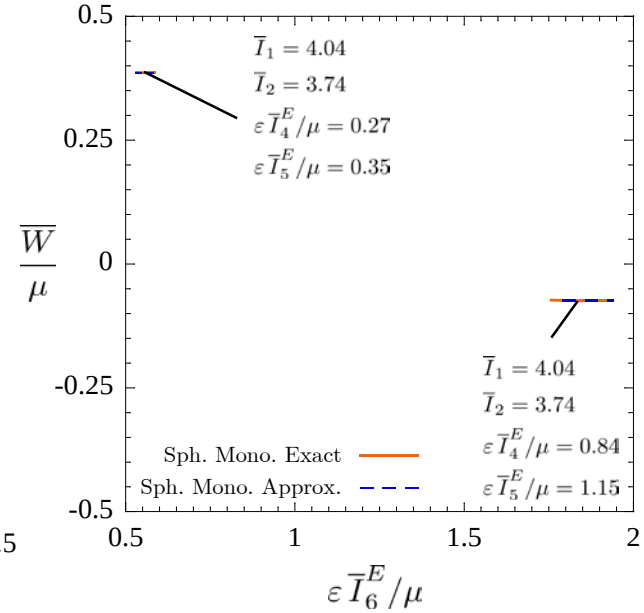
FE sample results: $c = 0.05,\ \mu_{\mathrm{p}} = 10^2\mu,\ \varepsilon_{\mathrm{p}} = 10^2\varepsilon$











Linear dependence on

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independence of

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for all $c,\ \mu,\ \varepsilon,\ \mu_{\mathrm{p}},\ \varepsilon_{\mathrm{p}}$

and all $\overline{\mathbf{F}},\ \overline{\mathbf{E}}$

An approximate closed-form solution

$$\overline{W}(\overline{\mathbf{F}},\overline{\mathbf{E}},c)=\begin{cases}\dfrac{\widetilde{\mu}}{2}\left[\overline{I}_1-3\right]+\dfrac{\widetilde{m}_K-\widetilde{\varepsilon}}{2}\overline{I}_4^E-\dfrac{\widetilde{m}_K}{2}\overline{I}_5^E & \text{if} \quad \overline{J}=1\\ +\infty & \text{otherwise}\end{cases}$$

Here, $\widetilde{\mu}$, $\widetilde{\varepsilon}$, $\widetilde{m}_K$ denote the **effective shear modulus**, **permittivity**, and **electrostriction constant** in the ***limit of small deformations and moderate electric fields***

$$\widetilde{\mu}=\int_\Omega 2\mu(\mathbf{X})\mathcal{K}_{12rs}\Gamma_{r12,s}\mathrm{d}\mathbf{X}$$

$$\widetilde{\varepsilon}=\int_\Omega \frac{1}{3}\varepsilon(\mathbf{X})\gamma_{m,m}\mathrm{d}\mathbf{X}$$

$$\widetilde{m}_K=\int_\Omega 2\varepsilon(\mathbf{X})\Gamma_{r12,s}\mathcal{K}_{rspq}\gamma_{p,1}\gamma_{q,2}\mathrm{d}\mathbf{X}$$

where $\mathbf{\Gamma}$ and $\boldsymbol{\gamma}$ are solutions of the **one-way coupled linear pdes**

$$\begin{cases}\left[\mu(\mathbf{X})\mathcal{K}_{ijmn}\Gamma_{mkl,n}+\dfrac{1}{2}\delta_{ij}q_{kl}\right]_{,j}=0\\ \Gamma_{mkl,m}=0\end{cases} \qquad \text{for } \mathbf{X}\in\Omega, \quad \Gamma_{ikl}=\delta_{ik}X_l \text{ for } \mathbf{X}\in\partial\Omega$$

and

$$\left[\varepsilon(\mathbf{X})\gamma_{i,j}\right]_{,i}=0 \qquad \text{for } \mathbf{X}\in\Omega, \qquad \gamma_i=X_i \text{ for } \mathbf{X}\in\partial\Omega.$$

Tian, Tevet-Deree, deBotton, Bhattacharya (2012)

Lefèvre & Lopez-Pamies (2014); Spinelli, Lefèvre, Lopez-Pamies (2015)

Solutions for

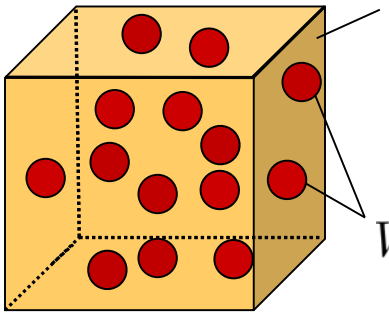
Non-Gaussian Elastomers

Isotropically Filled

with

Non-Linear Elastic Dielectric Particles

Non-Gaussian elastomers; non-linear particles



$$W_{\mathtt{m}}(\mathbf{F},\mathbf{E}) = \begin{cases} \Psi(I_1) - \dfrac{\varepsilon}{2} I_5^E & \text{if} \quad J = 1 \\ +\infty & \text{otherwise} \end{cases}$$

$$W_{\mathtt{p}}(\mathbf{F},\mathbf{E}) = \begin{cases} \dfrac{\mu_{\mathtt{p}}}{2}[I_1 - 3] - \mathcal{S}(I_5^E) & \text{if} \quad J = 1 \\ +\infty & \text{otherwise} \end{cases}$$

Here

$$\begin{cases} I_1 = \mathbf{F}\cdot\mathbf{F} \\ J = \det \mathbf{F} \\ I_5^E = \mathbf{F}^{-T}\mathbf{E}\cdot\mathbf{F}^{-T}\mathbf{E} \end{cases}$$

Examples:

(Lopez-Pamies, 2010)

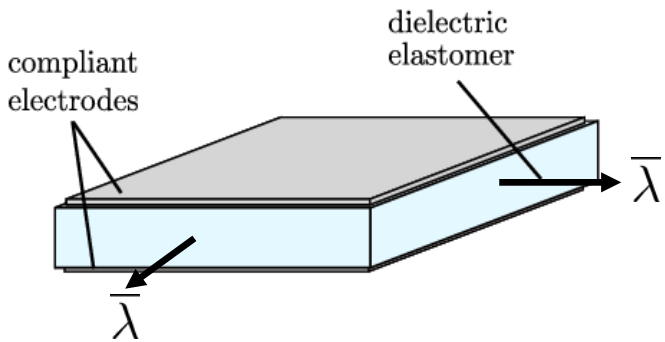
$$\Psi(I_1) = \frac{3^{1-\alpha_1}}{2\alpha_1}\mu_1[I_1^{\alpha_1} - 3^{\alpha_1}] + \frac{3^{1-\alpha_2}}{2\alpha_2}\mu_2[I_1^{\alpha_2} - 3^{\alpha_2}]$$

Langevin (1904); Debye (1929)

$$\mathcal{S}(I_5^E) = \frac{\varepsilon_0}{2} I_5^E + \frac{p_s^2}{3(\varepsilon_{\mathtt{p}} - \varepsilon_0)}\left[\ln\left(\sinh\left(\frac{3(\varepsilon_{\mathtt{p}} - \varepsilon_0)\sqrt{I_5^E}}{p_s}\right)\right) - \ln\left(\frac{3(\varepsilon_{\mathtt{p}} - \varepsilon_0)\sqrt{I_5^E}}{p_s}\right)\right]$$

- **Numerically**: *exact solutions* can be generated via the **HJ equation** & the **hybrid FE formulation**
- **Analytically**: *approximate solutions* can be generated via a **nonlinear comparison medium method**

Comparison with electrostriction experiments



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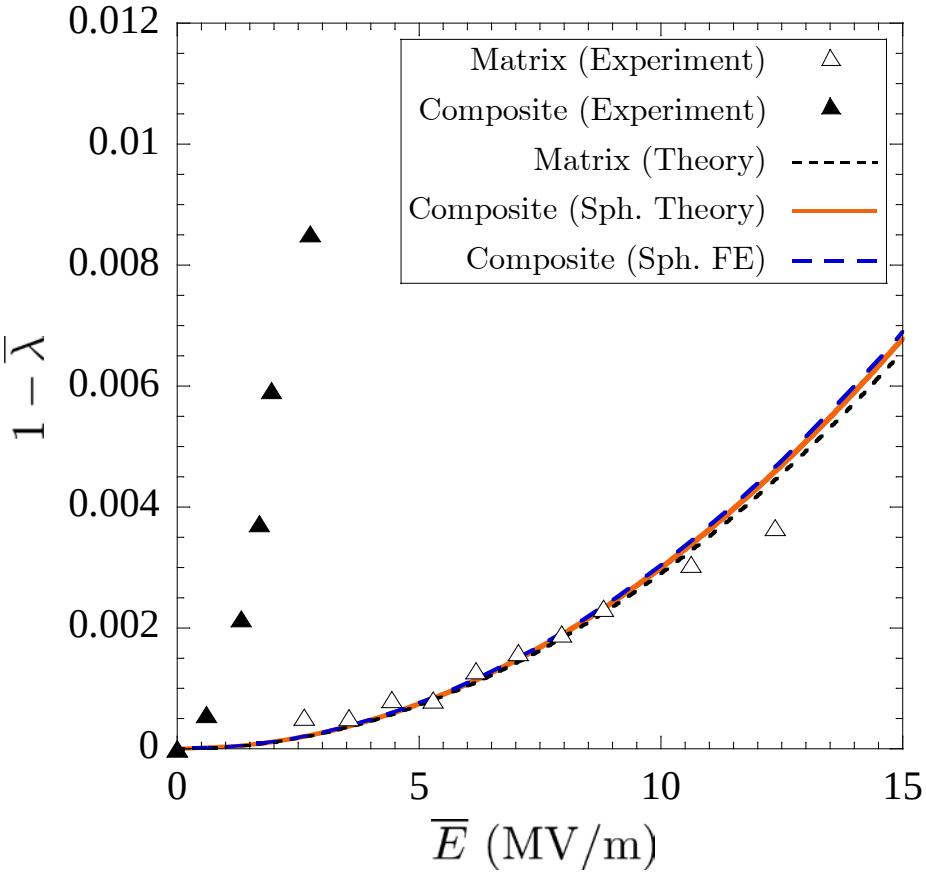
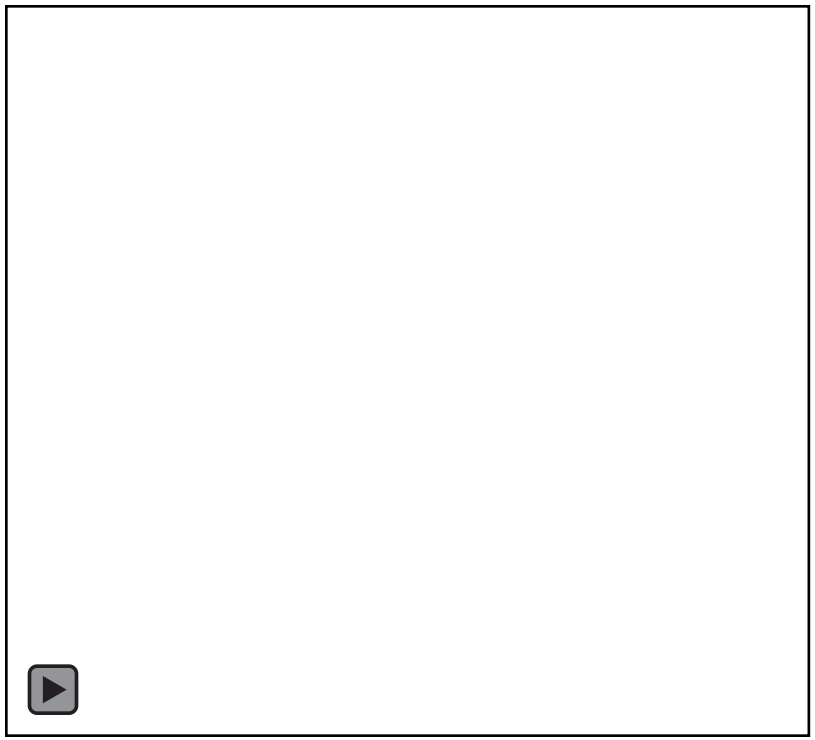
Particles: **PAA decorated o-CuPc** { dielectric constant: **10^4**; shear modulus: **1 GPa**

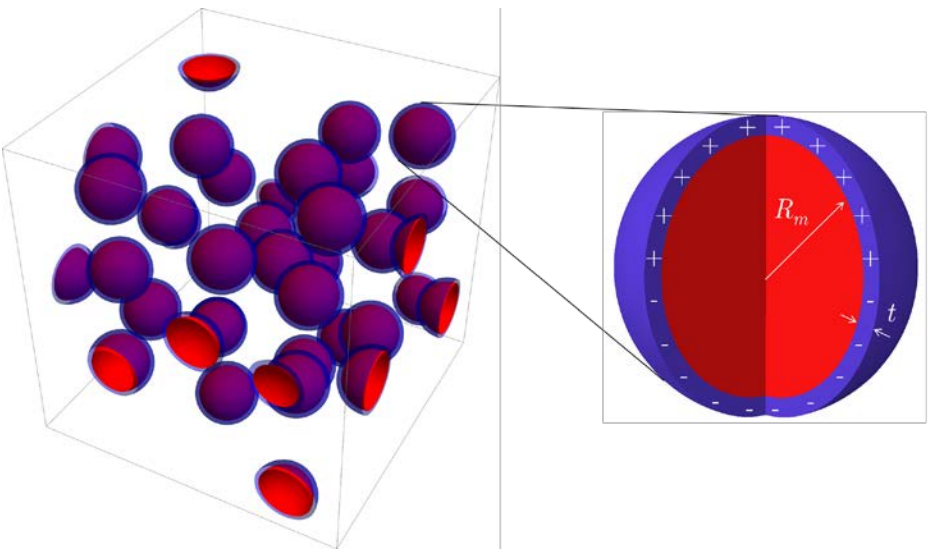
Microstructure: **isotropic distribution of roughly spherical nano-particles at 7% vol. fraction**

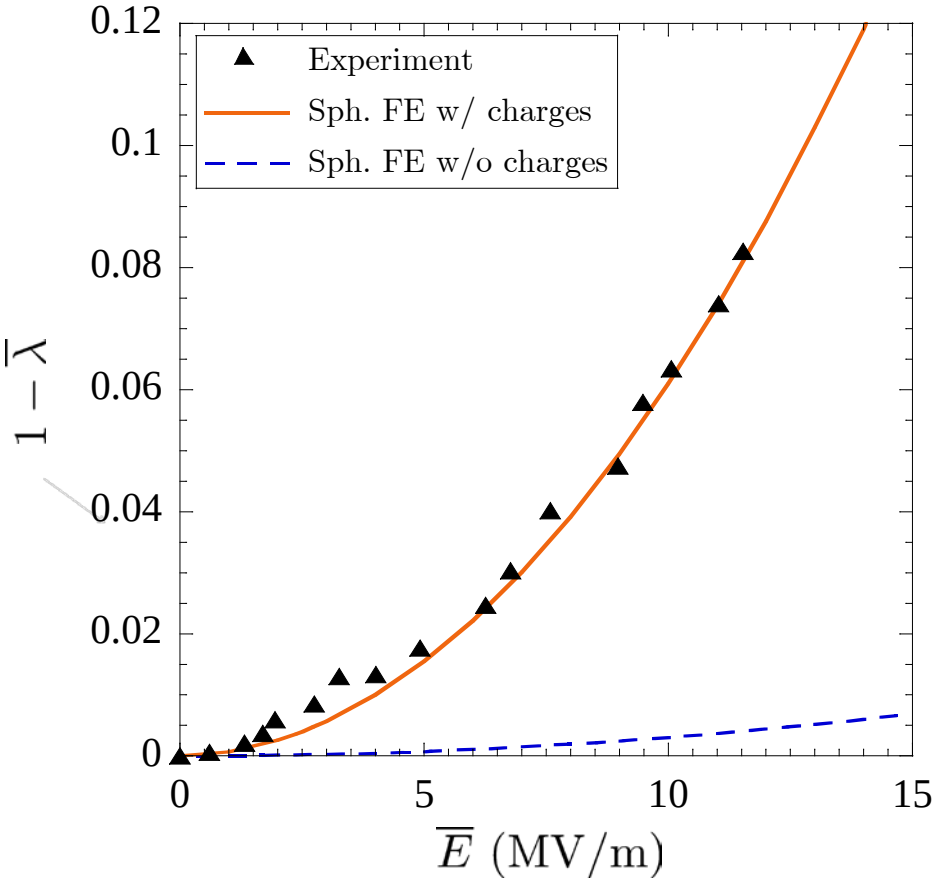




Huang et al. (2005); Lefèvre & Lopez-Pamies (2016)

Interphasial phenomena?





Lefèvre & Lopez-Pamies (2016)