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A two-scale homogenization scheme for the simulation of micro-heterogeneous magneto-electric composites

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Micro Abstract
We present the simulation of two-phase composites, consisting of a ferroelectric and a magnetostrictive phase, which generate a magneto-electric coupling. A two-scale finite element homogenization approach is performed. The typical hysteresis loops of the phases are approximated by considering the switching behavior of the spontaneous polarizations and the implementation of a Preisach operator.

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Introduction

Ferroic materials are characterized by particular coupling behaviors, such as electro-mechanical interactions in ferroelectric materials or magneto-mechanical interactions in magnetostrictive materials. They provide many applications in modern technical devices, which are used for sensors, actuators or data storage media. A further special phenomenon, which could improve these technical devices is the magneto-electric (ME) coupling. It is denoted by an interaction between magnetic fields and electric polarization or electric fields and magnetization. Such ME multiferroics enable Magneto-Electric Random Access Memory (MERAM) devices, see e.g. [1,5]. However, due to physical reasons, only very few natural materials with magneto-electric properties exist. They show their weak ME coupling far below room temperature. Magneto-electric two-phase composites, consisting of a ferroelectric matrix material with magnetostrictive inclusions, are a promising alternative. Figure 1 depicts the design of such composites. The idea of such composites is to generate the desired ME effect as a strain-induced product property, see [6]. In the case of ME composites, we distinguish between the direct and converse ME effect. The direct effect characterizes magnetically induced electric polarization, where an applied magnetic field yields a deformation of the magnetostrictive phase. These deformations are transferred to the ferroelectric phase, which result into a strain-induced polarization due to its electro-mechanical properties. The converse effect characterizes an electrically activated magnetization.

1 Two-scale Homogenization Scheme

The main idea of the FE^2-Method is the homogenization of microscopic quantities to obtain macroscopic constitutive equations instead of defining a macroscopic material model. Therefore, the macroscopic strains, electric and magnetic fields at each macroscopic integration point are localized to an underlying representative volume element ($\mathcal{RVE}$). The microscopic weak forms of the balance equations have to be solved. A following homogenization step determines the macroscopic quantities which are transferred to the associated points on the macroscale. Finally, the macroscopic boundary value problem is solved, whereby the entire procedure is repeated until an equilibrium state on both scales is reached, see [4].

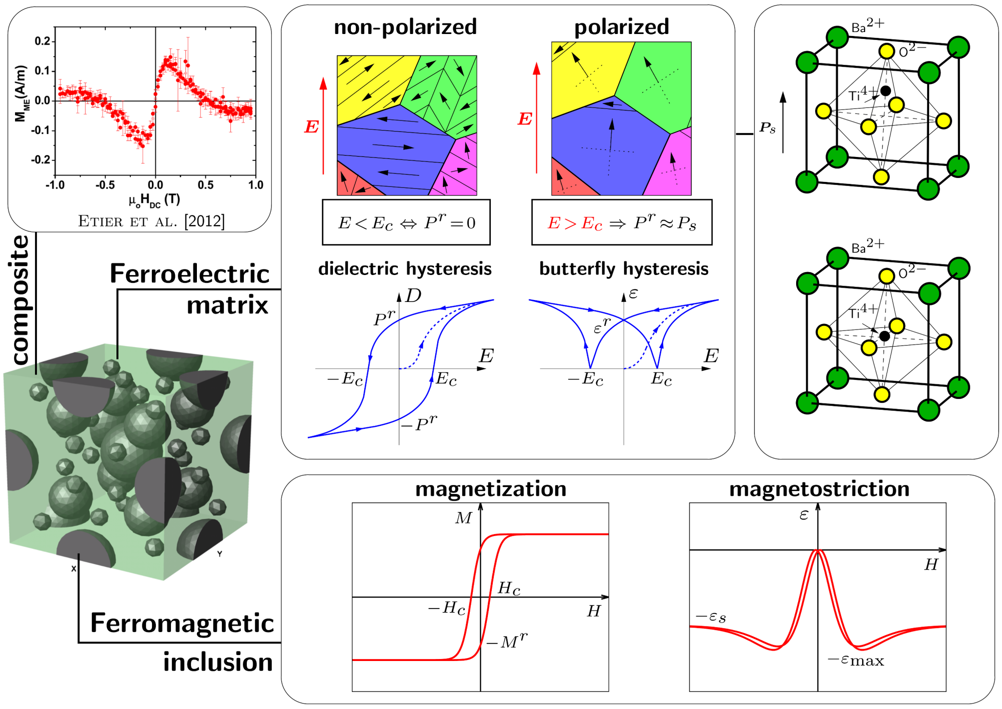

Figure 1. Design of magneto-electric two-phase composites. The typical polarization, magnetization and strain hysteresis curves of the corresponding phases as well as the resulting ME behavior are depicted.

1.1 Two-scale transition approach

The macroscopic body $\mathcal{B} \subset \mathbb{R}^3$ is parameterized in the Cartesian coordinates $\overline{\boldsymbol{x}}$. The fundamental balance laws are the balance of momentum and the Gauß's laws of electro- and magneto-statics

$$\mathrm{div}_{\overline{\boldsymbol{x}}}[\overline{\boldsymbol{\sigma}}] + \overline{\boldsymbol{f}} = \mathbf{0} \ , \quad \mathrm{div}_{\overline{\boldsymbol{x}}}[\overline{\boldsymbol{D}}] = 0 \quad \text{and} \quad \mathrm{div}_{\overline{\boldsymbol{x}}}[\overline{\boldsymbol{B}}] = 0 \tag{1}$$

where $\overline{\bullet}$ denotes the macroscopic quantities, $\overline{\boldsymbol{\sigma}}$ the stresses, $\overline{\boldsymbol{f}}$ the body forces, $\overline{\boldsymbol{D}}$ the dielectric displacement and $\overline{\boldsymbol{B}}$ the magnetic induction. The macroscopic strain $\overline{\boldsymbol{\varepsilon}}$, electric $\overline{\boldsymbol{E}}$, and magnetic field $\overline{\boldsymbol{H}}$ depend on the displacements $\overline{\boldsymbol{u}}$, the electric $\overline{\phi}$ and magnetic potential $\overline{\varphi}$ as

$$\overline{\boldsymbol{\varepsilon}} = \mathrm{sym}[\overline{\nabla}_{\overline{\boldsymbol{x}}}\overline{\boldsymbol{u}}] \ , \quad \overline{\boldsymbol{E}} = -\overline{\nabla}_{\overline{\boldsymbol{x}}}\overline{\phi} \quad \text{and} \quad \overline{\boldsymbol{H}} = -\overline{\nabla}_{\overline{\boldsymbol{x}}}\overline{\varphi} \tag{2}$$

with the macroscopic gradient operator $\overline{\nabla}$ with respect to $\overline{\boldsymbol{x}}$. The constitutive equations are

$$\begin{bmatrix} \Delta\overline{\boldsymbol{\sigma}} \\ -\Delta\overline{\boldsymbol{D}} \\ -\Delta\overline{\boldsymbol{B}} \end{bmatrix} = \underbrace{\begin{bmatrix} \overline{\mathbb{C}} & -\overline{\boldsymbol{e}}^T & -\overline{\boldsymbol{q}}^T \\ -\overline{\boldsymbol{e}} & -\overline{\boldsymbol{\epsilon}} & -\overline{\boldsymbol{\alpha}}^T \\ -\overline{\boldsymbol{q}} & -\overline{\boldsymbol{\alpha}} & -\overline{\boldsymbol{\mu}} \end{bmatrix}}_{\overline{\mathcal{Z}}} \begin{bmatrix} \Delta\overline{\boldsymbol{\varepsilon}} \\ \Delta\overline{\boldsymbol{E}} \\ \Delta\overline{\boldsymbol{H}} \end{bmatrix} \tag{3}$$

where the quantities on the left hand side are defined as volume integrals over the $\mathcal{RVE}$. The macroscopic fields of each integration point are localized on the $\mathcal{RVE}$s, where we have to apply suitable boundary conditions on the microscopic level. Therefore we assume that the individual parts of a generalized magneto-electro-mechanical Hill-Mandel condition have to be fulfilled independently [4]. Possible periodic boundary conditions can be obtained by assuming a decomposition of the microscopic strains, electric and magnetic fields in a macroscopic part and a fluctuation field. The overall tangent moduli $\overline{\mathcal{Z}}$ are calculated by a homogenization approach, with the generalized right-hand-sides $\underline{\boldsymbol{L}}$ and the global stiffness matrix $\underline{\boldsymbol{K}}$ as

$$\overline{\mathcal{Z}} = \frac{1}{V} \int_{\mathcal{RVE}} \mathcal{Z} dv - \frac{1}{V} \underline{\boldsymbol{L}}^T \underline{\boldsymbol{K}}^{-1} \underline{\boldsymbol{L}} \ . \tag{4}$$

For a more detailed derivation of the macroscopic material tangent we refer to [4].

1.2 Ferroelectric material model

To characterize the ferroelectric phase, we use a magneto-electric enthalpy function which represents the tetragonal symmetry of barium titanate, see [3]. The total strains and the electric displacement are decomposed into an elastic and a remanent part as

$$\boldsymbol{\varepsilon} = \boldsymbol{\varepsilon}^e + \boldsymbol{\varepsilon}^r \quad \text{and} \quad \boldsymbol{D} = \boldsymbol{D}^e + \boldsymbol{P}^r \quad \text{with} \quad \boldsymbol{P}^r = P_s\, \boldsymbol{c} \quad \text{and} \quad \boldsymbol{\varepsilon}^r = \frac{3}{2}\varepsilon_s \mathrm{dev}(\boldsymbol{c} \otimes \boldsymbol{c}) \tag{5}$$

with the spontaneous polarization P_s and strains ε_s. Depending on a switching criterion

$$\frac{\boldsymbol{E} \cdot \Delta \boldsymbol{P}_1^r}{\mathcal{W}^{diss}_{e,180^\circ}} \geq 1 \quad \text{and} \quad \frac{\boldsymbol{E} \cdot \Delta \boldsymbol{P}_i^r}{\mathcal{W}^{diss}_{e,90^\circ}} + \frac{\boldsymbol{\sigma} : \Delta \boldsymbol{\varepsilon}_i^r}{\mathcal{W}^{diss}_{m,90^\circ}} \geq 1 \tag{6}$$

based on [2], the preferred direction can change its direction. Thereby, the switching criterion for $i = 2, ..., 5$, checks if the change of free energy for possible switching options is larger than the dissipated work during a switching process. In each microscopic integration point an orientation distribution function representing the ferroelectric domain structure is attached.

1.3 Magnetostrictive material model

The magnetostrictive material behavior is described with a three-dimensional Preisach model. Therefore, we additively decompose the magnetic induction and the strains into an elastic part ($\bullet^e$) and a remanent part ($\bullet^r$), where the linear part is described by a transversely isotropic linear material law. The nonlinear remanent magnetization is described by the Preisach operator and the remanent strains depending on the current magnetization state are defined as

$$\boldsymbol{M}_r = \mathfrak{p}(\boldsymbol{H})\boldsymbol{a} \quad \text{and} \quad \boldsymbol{\varepsilon}_r = \frac{3}{2}\varepsilon_s \frac{1}{M_s^2}\left[\mathrm{dev}(\boldsymbol{M}_r \otimes \boldsymbol{M}_r) - \frac{1}{2}\mathrm{dev}(\boldsymbol{M}_r^{soft} \otimes \boldsymbol{M}_r^{soft})\right] , \tag{7}$$

with the saturation magnetization M_s and the saturation strain ε_s. Since the butterfly hysteresis curve of cobalt ferrite shows a softening behavior for higher magnetic fields, as can be seen in Figure 1, the remanent strains depend additionally on a virtual softening magnetization $\boldsymbol{M}_r^{soft} = \mathfrak{p}^{soft}(\boldsymbol{H})\boldsymbol{a}$. The remanent magnetization $\boldsymbol{M}_r$ is determined through the Preisach operator as

$$\mathfrak{p}(\boldsymbol{H}, \boldsymbol{a}) = \int_\beta \int_a lpha\omega(\alpha, \beta)\gamma(\alpha, \beta)\boldsymbol{H}(t) \cdot \boldsymbol{a}\,\mathrm{d}\alpha\mathrm{d}\beta \ . \tag{8}$$

In this model the preferred direction aligns with the direction of the microscopic magnetic field. This involves an update of the material tangent moduli as well as a switching of the corresponding Preisach relays. However, a direct switching of the orientation could lead to a back-and-forth switching of the preferred direction and the Preisach relays. In order to prevent this numerical instabilities a time dependent rotation process is implemented, where a fraction of the total angle θ_{total}, between the local microscopic magnetic field and the orientation, defines the rotation angle θ_{rot} during one load step with

$$\theta_{total} = \mathrm{acos}\left[\frac{\boldsymbol{a}_n \cdot \boldsymbol{H}}{||\boldsymbol{a}_n|| \cdot ||\boldsymbol{H}||}\right] , \quad \theta_{rot} = -\tanh[\kappa_\theta \cdot \tilde{t}] \cdot \theta_{total} \quad \text{and} \quad \boldsymbol{a}_{n+1} = \mathrm{R}(\theta_{rot})\boldsymbol{a}_n \tag{9}$$

where $\kappa_\theta = x/\mathrm{d}t$ denotes the rotation speed depending on the time step $\mathrm{d}t$ and $\boldsymbol{R}$ the rotation matrix for the transformation of the direction of the last time step $\boldsymbol{a}_n$ into the new direction $\boldsymbol{a}_{n+1}$. In order to adjust the magnetostrictive hysteretic behavior the piezomagnetic modulus depends on the current magnetization state as

$$\boldsymbol{q} = \left(2\frac{||\boldsymbol{M}_r||}{M_s}\left[1 - \left(\frac{||\boldsymbol{M}_r||}{M_s}\right)^2\right] - \frac{||\boldsymbol{M}^{soft}||}{M_s}\left[1 - \left(\frac{||\boldsymbol{M}_r^{soft}||}{M_s}\right)^2\right]\right)\hat{\boldsymbol{q}} \tag{10}$$

with the piezomagnetic coupling parameters in $\hat{\boldsymbol{q}}$

Simulations of magneto-electric composites

For the simulation of the magneto-electric coupling a composite consisting of a ferroelectric matrix with cylindrical magnetostrictive inclusions is considered. In a first step the matrix material is loaded with an electric field in order to polarize the material and to obtain electro-mechanical coupling properties. Several simulations with different electric field strengths demonstrate the dependence of the piezoelectric properties on the overall magneto-electric coupling. After this pre-polarization, an alternating magnetic field is applied on the composite, which causes the nonlinear deformations of the magnetostrictive inclusions. Due to the strain-induced interaction the magneto-electric coefficients for different polarization states are investigated, see Figure 2.

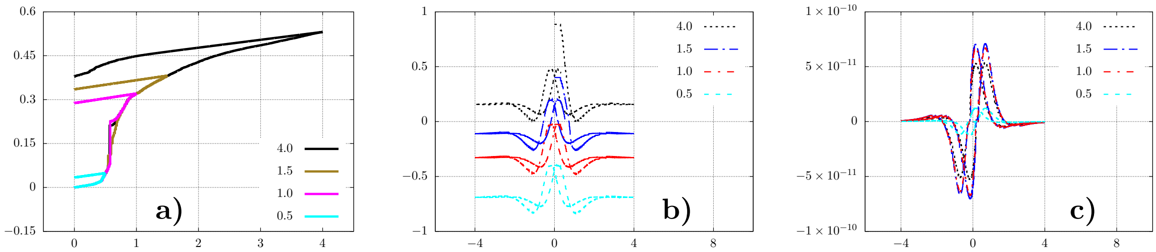

Figure 2. Resulting a) dielectric displacement ($\overline{E}_3$ kV/mm vs. $\overline{D}_3/P_s$), b) magnetostriction ($\overline{H}_3$ kA/mm vs. $\overline{\varepsilon}_{33}/\varepsilon_s$), and c) ME coupling ($\overline{\alpha}_{33}$ vs. $\overline{H}_3$ kA/mm) of a two-phase magneto-electric composite.

Conclusions

The FE^2-method is used to determine the effective properties of a two-phase magneto-electric composite. The desired magneto-electric coupling depends significantly on the the piezoelectric and magnetostrictive properties of the corresponding phases. Therefore, two nonlinear material models are implemented, which describe the hysteretic behaviors of the phases. Numerical simulations demonstrate that this approach is capable of describing the magneto-electric coupling in such composites. An accurate fitting of the material parameters and the investigation of different microstructures could lead to promising predictions of the ME coefficient.

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