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Enhancement of magnetoelectric coupling in multiferroic composites via FEM simulation

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ABSTRACT

In this paper, the theoretical background of linear constitutive multifield behavior as well as the Finite Element implementation are presented. The developed tools enable the prediction of the electromagnetomechanical properties of materials and structures and supply useful tools for the optimization of multifunctional composites. First, linear three-field coupling is presented within the context of a Finite Element implementation. Then, a homogenization technique is applied to describe the macroscopic behavior. A numerical examples focus on the prediction of the magneto-electric (ME) effect for different composite arrangements.

1. INTRODUCTION

Multiferroic magnetoelectric materials, which simultaneously exhibit ferroelectricity and ferromagnetism, have recently stimulated a sharply increasing number of research activities. As novel multifunctional devices, they contain a significant technological potential. These materials are much desired, because of the presence of the interaction between electric and magnetic fields. It is important to note, that this interaction appears as a material property (*ME-effect*) and is not following from the Maxwell-equations (*EM-Effect*). The coupling of magnetic and electrical fields may occur due to the physical properties of a crystal or can be artificially produced in a smart composite. The application spectra of these materials are novel multifunctional devices, such as sensors, transducer, etc… One of the most promising applications is the efficient storage of data in ferroelectric devices controlled by magnetic fields.

Nevertheless, after the discovery of this phenomenon, it has become unattended. The reason for this is that native multiferroic single-phase compounds are rare and their magnetoelectric responses are either relatively weak or occur at temperatures which are too low for practical applications. After the discovery of multiferroic composites the potential of the ME-Effect became larger. Nowadays, it is possible to develop multifunctional composites as micro- or nano-structures (e.g. $\mathrm{BaTiO_4 - CoFe_2O_3}$). In

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contrast to the native materials, the multiferroic composites, which incorporate both ferroelectric and ferromagnetic phases, typically yield much larger magnetoelectric coupling response at room temperature (Fiebig 2005). Thus, the attention to the multiferroic composites with ME-coupling was increased.

In composites the ME-effect is induced by the strain field converting electrical and magnetic energies based on the ferroelectric and magnetostrictive effects as shown in Fig. 1.

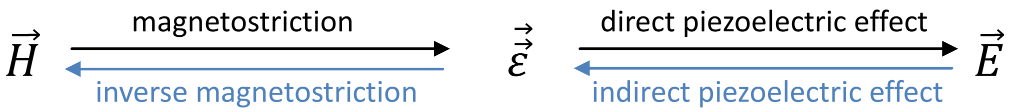

Fig. 1 Magnetoelectric (ME-) effect: coupling between magnetic and electric fields in composite materials. The (inverse) magnetostrictive and (in-) direct piezoelectric effects are coupled by deformation of matrix.

2. THEORETICAL FRAMEWORK OF LINEAR CONSTITUTIVE BEHAVIOR

2.1 Constitutive equations

The four Maxwell equations describe all of the phenomena of the classical electrodynamics (Jackson 1998):

$$\begin{aligned} \mathrm{rot}\,\vec{H} &= \dot{\vec{D}} + \vec{j}, \\ \mathrm{rot}\,\vec{E} &= -\dot{\vec{B}}, \end{aligned} \tag{1}$$

$$\begin{aligned} \mathrm{div}\,\vec{B} &= 0, \\ \mathrm{div}\,\vec{D} &= w_v, \end{aligned} \tag{2}$$

where $\vec{H}$, $\vec{E}$, $\vec{B}$, $\vec{D}$, $\vec{j}$ and w_v denote magnetic field, electrical field, magnetic induction, electric displacement, electrical current density and specific charge density. The first two equations couple electric and magnetic fields in transient systems. However, in multifunctional materials the fields are predominantly coupled via the constitutive equations.

The scalar electric and magnetic potentials (φ^{el} and φ^{m}) are motivated from the Maxwell equations, Eq. (1), for the electrostatical and magnetostatical case ($\dot{\vec{B}}, \dot{\vec{D}}, \vec{j} = 0$):

$$\begin{aligned} E_i &= -\varphi^{el}_{,i}, \\ H_i &= -\varphi^{m}_{,i}\,. \end{aligned} \tag{3}$$

With these definitions, the first Maxwell equations are trivially satisfied:

$$\vec{\nabla} \times \vec{E} = -\mathrm{rot}\;\mathrm{grad}\;\varphi^{el} = 0, \tag{4}$$

$$\vec{\nabla} \times \vec{H} = -\text{rot grad } \varphi^m = 0.$$

From here, only the analytical (index) notation is used implying summation and comma conventions.

With the help of the Eq. (3), the constitutive law of magnetoelectroelasticity is formulated. The relation between the thermodynamic potentials and the fields is obtained from thermodynamic analysis. The associated variables, i.e. stresses σ_{ij}, electric displacement D_i and magnetic induction B_i are obtained by partial differentiation of the thermodynamic potential

$$\Psi(\varepsilon_{ij}, E_i, H_i) = (\sigma_{ij}\varepsilon_{ij} - D_i E_i - B_i H_i)/2 \tag{5}$$

with respect to the independent variables, where ε_{ij} denotes the strain. The constitutive equations of linear magnetoelectroelasticity are then given by

$$\begin{aligned} \sigma_{ij} &= c_{ijkl}\varepsilon_{kl} - e_{lij}E_l - q_{lij}H_l, \\ D_i &= e_{ikl}\varepsilon_{kl} + \kappa_{il}E_l + g_{il}H_l, \\ B_i &= q_{ikl}\varepsilon_{kl} + g_{il}E_l + \mu_{il}H_l, \end{aligned} \tag{6}$$

with

$$\begin{aligned} c_{ijkl} &= \frac{\partial^2\Psi}{\partial\varepsilon_{ij}\partial\varepsilon_{kl}}, \qquad e_{lij} = -\frac{\partial^2\Psi}{\partial E_l \partial\varepsilon_{ij}}, \qquad q_{lij} = -\frac{\partial^2\Psi}{\partial H_l \partial\varepsilon_{ij}}, \\ \kappa_{il} &= -\frac{\partial^2\Psi}{\partial E_i \partial E_l}, \qquad \mu_{il} = -\frac{\partial^2\Psi}{\partial H_i \partial H_l}, \qquad g_{il} = -\frac{\partial^2\Psi}{\partial H_i \partial E_l}. \end{aligned} \tag{7}$$

The elastic properties are given by the fourth-order tensor c_{klij}, whereas the piezoelectric and magnetostrictive properties are denoted by third-order tensors e_{lij} and q_{lij}. The second-order tensors κ_{il}, μ_{il} and g_{il} represent the dielectric, magnetic permeabilities and magnetoelectric constants.

The constitutive Eqs. (6) describe all phenomena appearing in multifunctional materials. After the introduction of material coefficients with these equations, it is possible to describe the coupling between electrical, magnetical and mechanical phenomena.

To solve boundary value problems in a strict formulation, the balance equation of momentum has to be considered besides the Eqs. (2)

$$\sigma_{ij,j} + b_i = \rho\ddot{u}_i = 0 \tag{8}$$

where again the quasistatic limit is prescribed.

2.2 Finite Element formulation for the coupled field problem

An approximated solution can be obtained by applying the method of finite elements (FEM). The application of approximate methods requires the formulation of field equations in the weak form. This formulation is obtained e.g. from the generalized Hamilton's variational principle:

$$\delta S = \delta \int_{t_0}^{t_1} (K - \Psi)\, dt + \int_{t_0}^{t_1} \delta W^a\, dt = 0, \tag{9}$$

where K and δW^a denote kinetic energy and virtual work of the applied external forces. The weak formulation according to Eq. (9) is equivalent to the differential equations (2) and (8) as well as natural boundary conditions. For the static case, the generalized Hamilton's variational principle yields the principle of minimum of the total potential energy: $\delta \Pi = \delta(\Pi^i + \Pi^a) = 0$ with $\delta W^a = -\delta \Pi^a$ and $\Psi = \Pi^i$. It is important to note, that the inner and the external potentials Π^i and Π^a consist of mechanical, electric and magnetic parts. With the help of these equations, the field equations in the weak form are given as follows:

$$\begin{aligned} &\int_{(V)} \left(\sigma_{ij} \delta u_{i,j} + D_i \delta \varphi^{el}_{,i} + B_i \delta \varphi^{m}_{,i}\right) dV \\ &- \int_{(S_t)} \tilde{t}_i \delta u_i \, dS + \int_{(S_w)} \widetilde{w}^{el}_s \delta \varphi^{el} \, dS + \int_{(S_w)} \widetilde{w}^{m}_s \delta \varphi^{m} \, dS = 0. \end{aligned} \tag{10}$$

Volume charges w_v and forces b_i are neglected in Eq. (10). $\widetilde{w}^{el}_s$ and $\widetilde{w}^{m}_s$ are specific surface charges, where

$$\widetilde{w}^{m}_s = -B_i n_i \tag{11}$$

is formally following the definition for the electric charges without the physical presence of magnetic charges.

The calculation of finite element matrices is a very important part of FEM solution. Thereby, the most general and efficient technique is the application of isoparametric finite elements (Bathe 2006) which is applied here. Besides the weak formulation of magnetoelectroelastic field Eq. (10), the constitutive Eqs. (6) are required. The approximation of physical fields is performed within each single element with the following interpolations

$$\begin{aligned} u_i &= \sum_{\alpha=1}^{N} h^{\alpha}_u\, u^{\alpha}_i = [h_u]\{u_i\}, \\ \varphi^{el} &= \sum_{\alpha=1}^{N} h^{\alpha}_{el}\, \varphi^{el^{\alpha}} = [h_{el}]\{\varphi^{el}\}, \\ \varphi^{m} &= \sum_{\alpha=1}^{N} h^{\alpha}_{m}\, \varphi^{m^{\alpha}} = [h_m]\{\varphi^{m}\} \end{aligned} \tag{12}$$

where N is the number of nodes per element and $[h_u]$, $[h_{el}]$ and $[h_m]$ are isoparametric shape functions. After application of the fundamental lemma of variational calculus, the separated contributions to the generalized stiffness matrix (mechanical, electric, magnetic and the different mixed expressions) are obtained:

$$\begin{gathered}
[K_{uu}] = \int_{(V)} [B_u]^T [c] [B_u]\, dV\,, \qquad [K_{\varphi^{el}\varphi^{el}}] = -\int_{(V)} [B_{el}]^T [\kappa]^T [B_{el}]\, dV, \\
[K_{\varphi^m\varphi^m}] = -\int_{(V)} [B_m]^T [\mu]^T [B_m]\, dV, \\
[K_{u\varphi^{el}}] = \int_{(V)} [B_u]^T [e]^T [B_{el}]\, dV\,, \qquad [K_{u\varphi^m}] = \int_{(V)} [B_u]^T [q]^T [B_m]\, dV, \\
[K_{\varphi^{el}\varphi^m}] = -\int_{(V)} [B_{el}]^T [g] [B_m]\, dV.
\end{gathered} \tag{13}$$

Here, $[B_{el}]$ and $[B_m]$ relate the scalar potentials at nodes to the electric or magnetic field at the integration points. Of an element $[B_u]$ relates the mechanical displacement field to the strain field (Bathe 2006).

The calculation of the generalized stiffness matrix requires numerical integration, e.g. the GAUß quadrature. Based on the above equations, the boundary value problem is formulated as an algebraic system of equations $[K]\{U\} = \{R\}$:

$$\begin{bmatrix} [K_{uu}] & [K_{u\varphi^{el}}] & [K_{u\varphi^m}] \\ [K_{\varphi^{el}u}] & [K_{\varphi^{el}\varphi^{el}}] & [K_{\varphi^{el}\varphi^m}] \\ [K_{\varphi^m u}] & [K_{\varphi^m\varphi^{el}}] & [K_{\varphi^m\varphi^m}] \end{bmatrix} \begin{Bmatrix} [U_i] \\ [\varphi^{el}] \\ [\varphi^m] \end{Bmatrix} = \begin{Bmatrix} [F_s] \\ [Q_s^{el}] \\ [Q_s^m] \end{Bmatrix}, \tag{14}$$

where F_s, Q_s^{el} and Q_s^m denote the forces and generalized charges at nodes. The stiffness matrix as well as the displacement and the force vectors thus include mechanical, electric and magnetic contributions. Again, it is noted that "magnetic charges" Q_s^m are auxiliary quantities with no physical interpretation.

2.3 Homogenization procedures

Goal of the simulation is the investigation and finally optimization of multifunctional composites. Therefore, homogenization techniques need to be applied to describe the macroscopic behavior. On the macroscopic scale, the relations between the *effective* material tensors and the *averaged* fields are formulated as

$$\begin{aligned}
\langle \sigma_{ij} \rangle &= c^*_{ijkl} \langle \varepsilon_{kl} \rangle - e^*_{lij} \langle E_l \rangle - q^*_{lij} \langle H_l \rangle, \\
\langle D_i \rangle &= e^*_{ikl} \langle \varepsilon_{kl} \rangle + \kappa^*_{il} \langle E_l \rangle + g^*_{il} \langle H_l \rangle, \\
\langle B_i \rangle &= q^*_{ikl} \langle \varepsilon_{kl} \rangle + g^*_{il} \langle E_l \rangle + \mu^*_{il} \langle H_l \rangle.
\end{aligned} \tag{15}$$

To calculate the effective material tensors, we apply among others a generalized VOIGT approximation which is attended by the following boundary conditions: linear displacements (constant strains) and analogously constant electric and magnetic field

$$
\begin{aligned}
u_i &= \varepsilon_{ij}^0 x_j \quad \text{on } \partial V \text{ with } \quad \varepsilon_{ij}^0 = \text{const},\\
\varphi^{el} &= -E_i^0 x_i \quad \text{on } \partial V \text{ with } \quad E_i^0 = \text{const},\\
\varphi^{m} &= -H_i^0 x_i \quad \text{on } \partial V \text{ with } \quad H_i^0 = \text{const}.
\end{aligned} \tag{16}
$$

Thus, it follows:

$$
\langle \varepsilon_{ij} \rangle = \varepsilon_{ij}^0, \qquad \langle E_i \rangle = E_i^0, \qquad \langle H_i \rangle = H_i^0 \ . \tag{17}
$$

Consequently, constant strains, electric and magnetic fields are applied at the boundary of the RVE.

Following the prodecure outlined above, on the one hand normal displacements at constant E- and H- fields, electric potentials at constant displacements and H-fields or magnetic potentials at constant displacements and E-fields are imposed on a RVE. On the other hand, tangential displacements at constant E- and H-fields are imposed in order to calculate shear components. Subsequently, resulting stresses, B- and D-fields are calculated and all effective material constants are determined. Their definitions become obvious from Eqs.(6) and (7), in particular which quantities have to be kept constant by an appropriate choice of boundary conditions:

$$
\begin{aligned}
c_{ijkl} &= \left.\frac{\partial \sigma_{kl}}{\partial \varepsilon_{ij}}\right|_{E_i, H_j}, \qquad \kappa_{ij} = \left.\frac{\partial D_l}{\partial E_i}\right|_{\varepsilon_{ij}, H_j}, \qquad \mu_{ij} = \left.\frac{\partial B_l}{\partial H_i}\right|_{\varepsilon_{ij}, E_j},\\
-e_{lij} &= \left.\frac{\partial \sigma_{ij}}{\partial E_l}\right|_{\varepsilon_{ij}, H_l} = \frac{\partial^2 \Psi}{\partial E_l \partial \varepsilon_{ij}} = \frac{\partial}{\partial \varepsilon_{ij}}\left(\frac{\partial \Psi}{\partial E_l}\right) = -\left.\frac{\partial D_l}{\partial \varepsilon_{ij}}\right|_{E_l, H_l},\\
-q_{lij} &= \left.\frac{\partial \sigma_{ij}}{\partial H_l}\right|_{\varepsilon_{ij}, E_l} = \frac{\partial^2 \Psi}{\partial H_l \partial \varepsilon_{ij}} = \frac{\partial}{\partial \varepsilon_{ij}}\left(\frac{\partial \Psi}{\partial H_l}\right) = -\left.\frac{\partial B_l}{\partial \varepsilon_{ij}}\right|_{E_l, H_l},\\
g_{ij} &= \left.\frac{\partial D_j}{\partial H_i}\right|_{\varepsilon_{ji}, E_j} = -\frac{\partial^2 \Psi}{\partial H_i \partial E_j} = -\frac{\partial}{\partial E_j}\left(\frac{\partial \Psi}{\partial H_i}\right) = \left.\frac{\partial B_i}{\partial E_j}\right|_{\varepsilon_{ij}, H_i}.
\end{aligned} \tag{18}
$$

The constant fields are written behind the vertical bar. Due to the integrability condition, there are two alternative ways to define and finally calculate coupling coefficients.
Since magnetic fields are free of sources and free electric volume charges are commonly assumed not to be present in a dielectric material ($w_v = 0$), the balance equations (2) and (8) can be specified as

$$
\operatorname{div} \vec{\sigma} = 0, \qquad \operatorname{div} \vec{D} = 0, \qquad \operatorname{div} \vec{B} = 0 \qquad \text{in } V \tag{19}
$$

where inertia effects and volume forces have been neglected likewise. Thus, stresses, electric displacements and magnetic flux on the boundaries are equal to the respective average fields within the domain V. Therefore, Eqs. (17) and (18) supply relations for the effective properties of the homogenized medium:

$$
\begin{aligned}
c^*_{ijkl} = \left.\frac{\langle\sigma_{kl}\rangle}{\varepsilon^0_{ij}}\right|_{E_i,H_j}, \quad & \kappa^*_{il} = \left.\frac{\langle D_l\rangle}{E^0_i}\right|_{\varepsilon_{ij},H_j}, \quad \mu^*_{il} = \left.\frac{\langle B_l\rangle}{H^0_i}\right|_{\varepsilon_{ij},E_j}, \\
-e^*_{lij} &= \left.\frac{\langle\sigma_{ij}\rangle}{E^0_l}\right|_{\varepsilon_{ij},H_l} = -\left.\frac{\langle D_l\rangle}{\varepsilon^0_{ij}}\right|_{E_l,H_l}, \\
-q^*_{lij} &= \left.\frac{\langle\sigma_{ij}\rangle}{H^0_l}\right|_{\varepsilon_{ij},E_l} = -\left.\frac{\langle B_l\rangle}{\varepsilon^0_{ij}}\right|_{E_l,H_l}, \\
g^*_{il} &= \left.\frac{\langle D_j\rangle}{H^0_i}\right|_{\varepsilon_{ji},E_j} = \left.\frac{\langle B_i\rangle}{E^0_j}\right|_{\varepsilon_{ij},H_i}.
\end{aligned}
\tag{20}
$$

For example, the magnetoelectric tensor can be calculated either from the ratio between the averaged dielectric displacement and the applied magnetic field or from the averaged magnetic induction and the applied electric field as shown in Eq. (20). The corresponding magnetoelectric constant in x_1-direction g_{11} (same with e_{11}, q_{11}, κ_{11}, μ_{11}, e_{12}, q_{12}) is calculated as shown in Fig. 2.

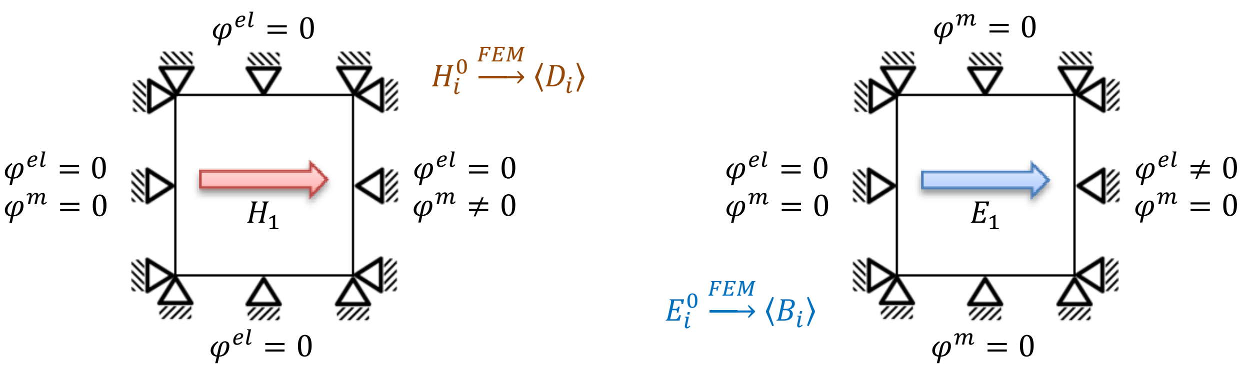

Fig. 2 Boundary conditions for the calculation of the effective magnetoelectric tensor

2.4 Results of the homogenization

We consider three different composites. The first consists of an elastic matrix which is dielectric and diamagnetic with embedded piezoelectric and magnetostrictive particles. The second composite has a piezoelectric matrix with embedded magnetostrictive particles and the third composite has a magnetostrictive matrix with embedded piezoelectric particles. The magnetoelectroelastic moduli of the three phases are presented in Table 1 (Tang & Yu 2009).

Table 1. Material properties of $BaTiO_3$, $CoFe_2O_4$, and epoxy

	$BaTiO_3$	$CoFe_2O_4$	Epoxy
c_{11} (GPa)	162	269.5	5.53
c_{12} (GPa)	77.5	170	2.97
c_{22} (GPa)	166	286	5.53
c_{23} (GPa)	76.6	173	2.97
c_{44} (GPa)	42.9	45.3	1.28
κ_{11} (C/Vm)	12.57E − 9	0.093E − 9	0.1E − 9
κ_{22} (C/Vm)	11.16E − 9	0.08E − 9	0.1E − 9
μ_{11} (Ns^2/C^2)	0.1E − 4	1.57E − 4	0.01E − 4
μ_{22} (Ns^2/C^2)	0.05E − 4	−5.9E − 4	0.01E − 4
e_{11} (C/m^2)	18.6	0	0
e_{12} (C/m^2)	−4.4	0	0
e_{24} (C/m^2)	11.6	0	0
q_{11} (N/Am)	0	700	0
q_{12} (N/Am)	0	580	0
q_{24} (N/Am)	0	550	0

The corresponding effective magnetoelectric constants in x_1-direction are calculated by using the homogenization technique described in section 2.3. This constant is obtained as the arithmetical average of the two approaches described in the Eq. (20). For all calculations, the same geometrical model has been used, just the material allocations are different. Our result is that the second composite (see Fig. 3) has the largest magnetoelectric coupling constant of $g_{11}^* = 0.111\text{E} - 9$ [Ns/VC] ($0.7BaTiO_3 - 0.3CoFe_2O_4$).

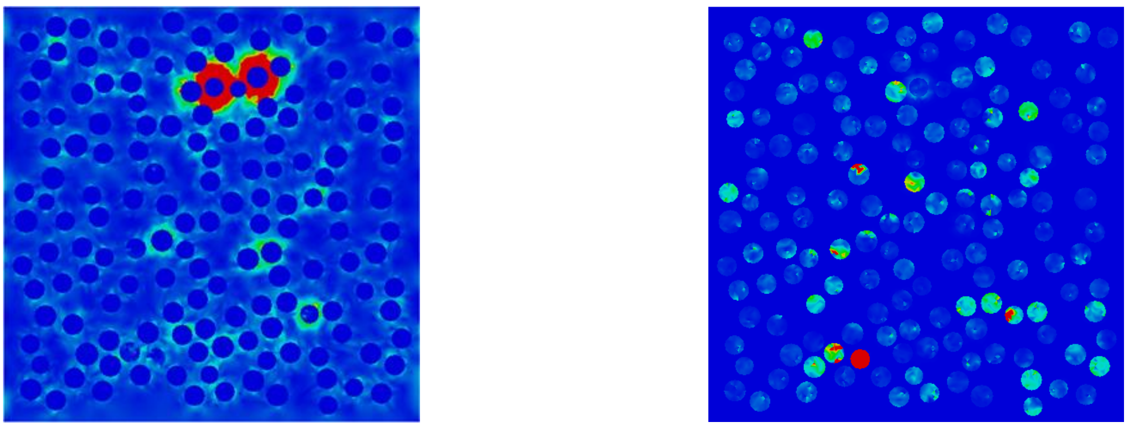

Fig. 3 Magnetoelectric particle composite (matrix: $BaTiO_3$ and particles: $CoFe_2O_4$) left: electric displacement and right: magnetic induction

The other effective constants are calculated likewise. The results are presented in Table 2.

Table 2. Effective material properties of the $0.7BaTiO_3 - 0.3CoFe_2O_4$ composite

	$0.7BaTiO_3 - 0.3CoFe_2O_4$ composite
c_{11}^* (GPa)	192.3
c_{12}^* (GPa)	95.9
c_{22}^* (GPa)	190.9
c_{44}^* (GPa)	45
κ_{11}^* (C/Vm)	6.97E − 9
κ_{22}^* (C/Vm)	6.70E − 9
μ_{11}^* (Ns^2/C^2)	0.165E − 4
μ_{22}^* (Ns^2/C^2)	0.107E − 4
e_{11}^* (C/m^2)	10.36
e_{12}^* (C/m^2)	−2.84
e_{24}^* (C/m^2)	7.26
q_{11}^* (N/Am)	22.17
q_{12}^* (N/Am)	15.58
q_{24}^* (N/Am)	−4.43
g_{11}^* (Ns/VC)	0.111E − 9
g_{22}^* (Ns/VC)	0.330E − 9

Now, if we make calculations for the second composite with all effective material constants, a magnetic induction of one Tesla yields an electric field strength of 8.01 V/mm. Comparing this result to the one for the inhomogeneous composite, the deviations along the boundary of the RVE are plotted in Fig. 4. The average deviation is not larger than $1{,}56\%$.

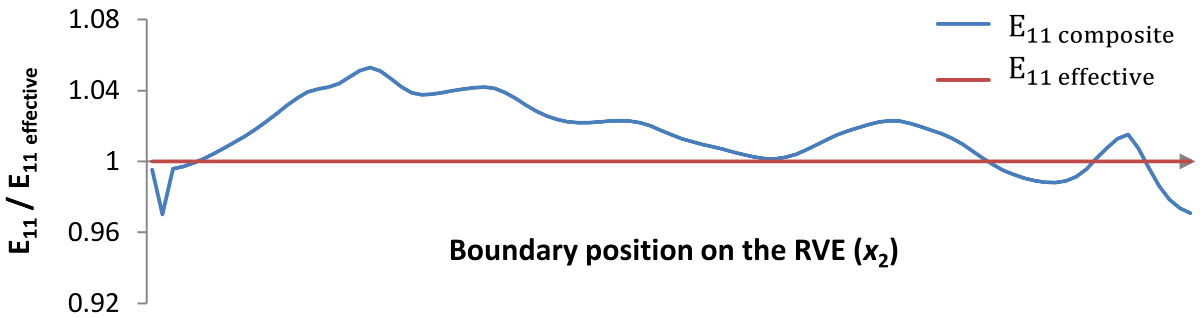

Fig. 4 Relative ratio of the electric field in x_1-direction between the $0.7BaTiO_3 - 0.3CoFe_2O_4$ composite and the body with effective properties

4. CONCLUSIONS

In this work were derived field equations, constitutive relations and the stiffness matrix of multifunctional materials. A subroutine USER-Element (UEL) for linear magneto-electroelastic behavior has been developed and implemented in a commercial FEM software ABAQUS. Further, were performed calculations with magnetoelectoelastic boundary value problems in association with homogenization procedures. The next step of our work is to include a nonlinear ferromagnetic and ferroelectric behavior in to the model. The goal is to evaluate mechanical stresses during the multifield poling process and to optimize the composite with respect to functionality and strength.

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