# PHASE FIELD MODELING HETEROGENEOUS MULTIFERROICS

Yong Ni[* a)], Linghui He[*] & Armen G. Khachaturyan[**]

[*a] *CAS Key Laboratory of Mechanical Behavior and Design of Materials, Department of Modern Mechanics, University of Science and Technology of China, Hefei, Anhui, 230026, P.R.China*

[**] *Department of Materials Science & Engineering, Rutgers University, 607 Taylor Road, Piscataway, NJ 08854, USA*

Summary Based on the equivalency method and the variational principle, a phase field model is proposed to solve the elastic strain, electrostatic and magnetostatic fields of a magnetoelectroelastic solid with arbitrary inhomogeneities under applied loadings. The effect of mesoscale inhomogeneities on the ferroic property, such as piezoelectricity and magnetoelectricity in heterogeneous multiferroics are discussed through some numerical results based on the model.

## INTRODUCTION

To obtain a large magnetoelectric (ME) effect in multiferroic materials with the polarization and magnetization coexisting currently attracts intense research efforts due to its potential for applications as multifunctional devices. The direct ME effect is usually weak in multiferroic single-phase compound. In contrast, the elastic interactions between the piezoelectric and piezomagnetic (or magnetostrictive) phases can lead to a giant indirect ME response of these multiferroic composites. The ME behavior in multiferroic composites is found to strongly depend on the spatial architecture of the composite characterized by the volume fractions of the phases, the shape and orientation of particles of these phases, their connectivity, and mutual positions. It also is critically dependent on the materials properties of the constituent phases such electronic/magnetic permeability and piezoelectric coefficients and magnetostriction constants[1]. Since the ME effect strongly depends on the composite architecture, it is of fundamental interest to investigate how the ME effect depends on the microstructure of the composite, and to find the optimal architecture that produces the largest ME effect. We therefore try to develop a general theoretic framework capable of systematically calculating the coupling magneto-electric-mechanical response in these multiferroic composites with arbitrary geometrical configurations. This approach allows us to address two problems, they are: (i) the finding of the two-phase architecture of a composite and the combination of intrinsic properties of the constituent phases that would optimize the ME coupling, and (ii) the realistic 3D prototyping of the evolution of the microstructure during the processing of bulk and laminar composites.

## METHOD

We proposed a generalized Phase-Field Microelasticity modeling[2] wherein a series of density functions fields (phase fields) are used to characterize the states of the multiferroic composite: a shape function, $\theta(\mathbf{r},t)$, for the arbitrary geometrical configuration; the effective eigenstrain, $\varepsilon_{ij}^0(\mathbf{r},t)$, for the elastic inhomogeneities; the effective polarization, $P_i^0(\mathbf{r},t)$, for the dielectric inhomogeneities; the effective magnetization, $M_i^0(\mathbf{r},t)$, for the magnetic inhomogeneities. Based on the equivalency approach, in which these fields at the equilibrium in the original heterogeneous system are exactly the same as those in an equivalent uncoupled homogeneous system with constant elastic moduli, dielectric permitivities and magnetic permeabilities having properly distributed effective eigenstrain, polarization and magnetization fields. Finding these effective fields fully solves the equilibrium elasticity, electrostatics and magnetostatics in the original heterogeneous solid. It is shown that minimizing a specific energy functional, $\Pi$, with respect to the eigenstrain, polarization, and magnetization phase field variables gives the exact equilibrium elastic, electrostatic, and magnetostatic fields in a given multiferroic composite with the inhomogeneities characterized by the coordinate dependent constitutive coefficients, such as, elastic, piezoelectric, piezomagnetic, electromagnetic coefficients, dielectric permitivities and magnetic permeabilities. The minimizing process is realized by numerically solving a set of time-dependent Ginzburg-Landau kinetic equations. The minimizers can be obtained as the steady solutions of these eqations.

$$\frac{\partial \varepsilon_{ij}^0(\mathbf{r},t)}{\partial t} = -L_{ijkl}^{elas}\frac{\delta\Pi}{\delta\varepsilon_{kl}^0(\mathbf{r},t)},\quad \frac{\partial P_i^0(\mathbf{r},t)}{\partial t} = -L_{ij}^{elect}\frac{\delta\Pi}{\delta P_j^0(\mathbf{r},t)},\quad \frac{\partial M_i^0(\mathbf{r},t)}{\partial t} = -L_{ij}^{mag}\frac{\delta\Pi}{\delta M_i^0(\mathbf{r},t)}, \tag{1}$$

$$\Pi = \Pi\left(\varepsilon_{ij}^0(\mathbf{r},t), P_i^0(\mathbf{r},t), M_i^0(\mathbf{r},t), \theta(\mathbf{r},t)\right) \tag{2}$$

[a)] Corresponding author. Email: yni@ustc.edu.cn

The microstructure evolution of the composite architecture is described by a Cahn-Hilliard kinetic equation[3].

$$\frac{\partial \theta(\mathbf{r},t)}{\partial t} = D\nabla^2 \frac{\delta \Pi}{\delta \theta} \tag{3}$$

## RESULTS

Based on the equivalency method and the variational principle, we proved that the elastic strain, electrostatic and magnetostatic fields of the heterogeneous multiferroic composite is identical to those of its equivalent uncoupled homogenous system subjected to the proper effective eigenstrain, polarization and magnetization, which are the steady solutions of the time-dependent Ginzburg-Landau kinetic equations. The numerical solutions of these equations (Eq. (1)-(3)) are easily calculated by using Fast Fourier Transform (FFT) algorithms. This could allow us to simultaneously take into account the crack growth and ferroic domain switch within a unified way. An incorporation of the phase field method into the homogenization process allows us to predict all the effective properties of the heterogeneous magnetoelectroelastic solid.

The configuration-dependent parts of the strain-induced, electrostatic and magnetostatic energies are the reason that the strain-mediated ME effect depends on the composite architecture. The first order ME effect is dominated by the pairwise long-range interaction, which is mainly attributed to the elastic interaction energy between piezoelectric and piezomagnetic phases.

The optimal architecture that produces the strongest ME effect is a plate-like configuration of constituent phases in the case of the ME effect determined by the pairwise long-range interaction. The value of this interaction depends on piezoelectric/piezomagnetic strains, which are related to the external electric/magnetic fields.

## CONCLUSIONS

A 3-dimentional phase field theory of the strain-mediated magnetoelectric coupling in piezoelectric-piezomagnetic composites with arbitrary spatial arrangement of particles of constituent phases is proposed.

The phase field theory can be integrated into optimizing the ME coupling constants through a simulated 3-D virtual evolution of the composite architecture with a driving force that is formulated to eventually lead to a configuration with the best ME coupling.

The configuration-dependent parts of the strain-induced, electrostatic and magnetostatic energies, can always be presented as a sum of the contributions from pairwise (dipole-dipole like), triplet and higher-order multi-particle interactions between finite elements of the piezoelectric and piezomagnetic phases. They are the reason that the strain-mediated ME effect strongly depends on the composite architecture.

The first order ME effect is dominated by the pairwise long-range interaction, which is mainly attributed to the elastic interaction energy between piezoelectric and piezomagnetic phases.

The analytical solution and PFM simulation demonstrate that the optimal architecture that peruses the strongest ME effect is plate-like configuration of constituent phases in the case of the ME effect determined by the pairwise long-range interaction.

## References

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