1st Seminar on Ferroic Functional Materials

TU Dortmund, Germany
21–22 November, 2012
Scientific Objectives

Due to their unique properties, multifunctional materials continue to attract attention from both the research and the industrial application communities. Research on these materials has included experimental investigation, modeling, and simulation of their complex response features, such as multi-physical couplings, nonlinearity, inelasticity, and anisotropy. The seminar places a special focus on the investigation of multiferroics, i.e. materials that exhibit ferroelectricity, ferromagnetism or ferroelasticity, and their respective couplings.

Of particular interest is the characterization and modeling of ferroic functional response on multiple length scales, e.g. the mechanical and field-induced evolution of microstructures and domain patterns. The computation and optimization of effective material properties requires the generalization of homogenization and scale-bridging techniques that have been established in computational mechanics for classical materials. Advanced algorithmic strategies are continuously being developed to robustly and efficiently solve systems of coupled field equations and simulate complex initial boundary value problems on all scales. The calibration of these models on the basis of reliable experimental data is of key importance. Generalized notions of material stability for coupled responses also remain to be defined.

The goal of this meeting is to bring together researchers from mechanics, material science, solid state physics and applied mathematics to discuss current developments in the field. The seminar will place special emphasis on theoretical, numerical, and experimental aspects. Typical examples for classes of ferroic functional materials include piezo- and ferroelectric ceramics, ferromagnetic and magnetostrictive materials, and particularly also their composites.

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DFG-Research Unit 1509
# Program

## Wednesday, 21 November

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<td>13:20–13:30</td>
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| 13:30–14:15| E. Patoor: Electromechanical modeling for ferroelectric and ferroelastic ceramics  
              | ceramics (→)                                                         |
| 14:15–14:40| F. Wendler: Magnetic and elastic response of ferromagnetic shape memory alloys studied with a phase-field model (→) |
| 14:40–15:05| M. Labusch: A computational framework for the two-scale homogenization of non-dissipative magnetostrictive solids (→) |
| 15:05–15:30| G. Ethiraj: Microsphere model for finite deformation modeling of magneto-sensitive elastomers (→) |
| 15:30–16:00| coffee break/posters                                                 |
| 16:00–16:45| H. Beige: Lineare und nichtlineare elastische, dielektrische und elektromechanische Eigenschaften von Dielektrika (→) |
| 16:45–17:10| A. Nazrabi: Experimental methods for the measurement of strain in the nanometer regime (→) |
| 17:10–17:35| F. Endres: Towards molecular simulation of ferroelectric materials (→) |
| 17:35–18:00| D. Schrade: Interpretation of material parameters in phase field models for ferroelectrics (→) |
| 20:00      | dinner                                                              |

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<td>09:00–09:25</td>
<td>I. Anusca: Core/shell Nanoparticles: different synthetic routes of multiferroic cobalt ferrite/barium titanate and characterisation (→)</td>
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<td>09:25–09:50</td>
<td>M. Etier: Magnetoelectric properties of 0.2 CoFe$_2$O$_4$-0.8 BaTiO$_3$ composite prepared by the organosol route (→)</td>
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<td>09:50–10:15</td>
<td>B. Xu: Phase Field Simulation on the Influence of Point Defects with Domain Structures in Ferroelectrics (→)</td>
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<td>10:15–10:40</td>
<td>A. Bratskikh: Modeling and simulation of mechanical damage behavior of polycrystalline ferroelectric ceramics (→)</td>
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Abstracts
Electromechanical modeling for ferroelectric and ferroelastic ceramics

E. Patoor
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Piezoceramics exhibit an electro-mechanical coupling which enables them to produce a proportional electric charge when subjected to a mechanical stress or to deform in presence of an electric field. This behaviour is used in many applications such as actuators or sensors. In the case of severe loadings, the response is no longer linear and it is characterized by the switching of uniformly polarized domains. Thus, ferroelectricity and ferroelasticity induce irreversible polarization and strain. This non-linear effect produces greater values of displacements. However, the complexity of the electromechanical coupling has to be overcome to design structures involving such materials. In the first part of this presentation an electro-micromechanical model is presented to examine the switching process that occurs in ferroelectric and ferroelastic single crystals under electromechanical loadings. Ferroelectrics undergoing a cubic to tetragonal phase transition are considered. Three kinds of energy are considered at the single crystal level: elastic energy, electric energy and the electro-elastic interaction energy due to incompatibilities of the spontaneous strain and electric displacement fields between domains. In the second part of the presentation the development of a macroscopic phenomenological constitutive model is considered and implemented in the finite element software Abaqus.
Magnetic and elastic response of ferromagnetic shape memory alloys studied with a phase-field model

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Magnetic shape memory alloys (MSMA) offer a combination of large recoverable strains combined with relatively high reaction rates, enabling switching frequencies up to 1000 Hz. Together with the inherent pseudoplasticity, these alloys are intensely investigated for actuation and damping applications. As the focus of recent research lies mainly on optimizing macroscopic material properties by variations in chemical composition (atomistic scale), additionally a better understanding is needed about the essential kinetic pathways leading to the observed twin structure on a mesoscopic length scale. Based on a (diffusionless) ferroelastic transition from the parent austenite phase to the martensitic state, the magnetic shape memory effect (MSME) relies on the intrinsic coupling of the magnetic and elastic order parameters. This allows for the easy re-orientation of twin boundaries separating martensitic variants with a moderate applied magnetic field. We apply a multi-phase field model to simulate the MSME in the Heusler alloy Ni$_2$MnGa under different external loading conditions and field configurations, where non-conserved order parameters are related to the eigenstrain of the tetragonal 5M variants. The approach is based on an interpolation of free energies extended by micromagnetic contributions including Zeeman, demagnetization, exchange and anisotropy energy, and the linearized elastic energy, which realizes the coupling between displacement, magnetization and order parameter fields. The origin and the numerical methods to solve for the dynamic equations (finite differences, FFT techniques, geometric integration) are briefly presented. Concerning the displacement field, either a dynamic wave equation with damping term is applied, or mechanical equilibrium is assumed. Limitations in computational size are imposed by the necessity to resolve domain and twin boundaries. Hence, we restrict ourselves to single crystalline samples with periodic geometry for 2D and 3D, connected to the externally applied mechanical and magnetic fields by a simple RVE approach. We present the result of simulation series for stress-controlled uniaxial deformation without external applied magnetic field, where the twinning stress level and slope of the transition plateau strongly depends on stress rate, damping coefficient and twin boundary tension. Also, the magnetic microstructure has a clear contribution to the macroscopic mechanical response. Diagrams of integral strain and magnetization vs. external field show the observed stress dependent hysteresis. The current limits and restrictions of the applied model, and necessary extensions to simulate fast transitional dynamics are discussed as an outlook.

References

A computational framework for the two-scale homogenization of non-dissipative magnetostrictive solids

Matthias Labusch, Marc-André Keip, and Jörg Schröder
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Magnetostrictive materials are characterized by magneto-mechanical coupling, which can be exploited in technical applications. The effective magneto-mechanical coupling of a heterogeneous magnetostrictive solid depends significantly on its microstructure and the coupling phenomena arising on the smaller scales. In order to compute the overall macroscopic material response, computational homogenization schemes can be used. One such scheme is the FE²-method, which is based on the two-scale simulation of a boundary value problem in consideration of the constitutive response of a microscopic representative volume element (RVE). The RVE is attached to each macroscopic material point and has to be chosen in such a way that it represents the microstructure of the material both constitutively and geometrically in a reasonable manner.

The focus of this talk is to derive an FE²-formulation for the two-scale simulation of magnetostrictive solids, cf. [1]. As a first step, a non-dissipative material model for the description of the magnetostrictive coupling behavior based on the works [2,3] will be developed. The resulting model will then be implemented into an FE²-formulation in order to arrive at a two-scale method for the simulation of magneto-mechanical materials. The applicability of the formulation will be demonstrated by some numerical examples.

References


Microsphere model for finite deformation modeling of magnetosensitive elastomers

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In recent years, a number of industrial applications that make use of magnetosensitive materials have been developed. Magnetosensitive elastomers are a class of composite materials whose mechanical response may be altered by application of magnetic fields. Such materials with tunable mechanical properties find use in controllable stiffness devices or membranes, and applications for active control of structural components aimed at optimizing the performance of mechanical systems. In this work we present a novel approach to the modeling of magnetorheological elastomers (MREs) for finite deformations. Keeping in mind the composite nature at the microscale, we employ the microsphere model as an effective tool to capture the constitutive response of the material. The microsphere model has been successfully applied to the modelling of rubber-like materials. It may be described as a method based on homogenizing 1D models over a set of directions in order to obtain 3D constitutive material behavior. Here, we extend this approach by taking into account the effect of the magnetic dipole-dipole interactions on the orientation of the polymer chains by use of a multiplicative split in the isochoric part of the deformation gradient. Thus, the presented microsphere model is directly motivated by considering the underlying phenomena at the microscale level. We discuss details of the coupled magnetomechanical finite element implementation in a compact notation as well as aspects concerning the numerical computation of coupled tangent moduli. Finally the solution to application-oriented boundary value problems is presented.

References


Lineare und nichtlineare elastische, dielektrische und elektromechanische Eigenschaften von Dielektrika

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Experimental methods for the measurement of strain in the nanometer regime

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The characterization of the ferroic functional materials requires a more detailed description of its mechanical, electromechanical, and magneto-mechanical coupled as well as over all three quantities coupled constitutive laws. The response for the interaction of applied field in the magneto-electric composites is mediated by mechanical stress which is expected to be measurable as resulting strain. This talk gives an overview on the experimental methods for determining the strain in general and in the presence of electric and magnetic fields.
Towards molecular simulation of ferroelectric materials

F. Endres, P. Steinmann
Chair of Applied Mechanics, Friedrich-Alexander-Universität Erlangen-Nürnberg

In the past few years molecular models for the simulation of ferroelectric materials have been developed further and obtain accurate results for atomistic level simulations. These models such as the core-shell model calculate all different interactions in a ferroelectric crystal like material, e.g., barium titanate. However, applications on systems with macroscopic length scales are limited by the complexity of such simulation models. Especially molecular dynamics simulations have disadvantages not only due to the macroscopic length scales, but also of the very small time steps. An ansatz based on molecular statics is shown in [1]. The new method links the accuracy of the core-shell model of e.g, [2] with the efficiency of an atomic-scale finite element method. On the one hand some challenges like the handling of long range interactions still remain in molecular statics. On the other hand the calculation of a time independent energetic equilibrium is much more efficient with a static approach than with molecular dynamics. We discuss the core-shell model and its parameters for the simulation of barium titanate as well as the opportunities and challenges of quasi static molecular simulations.

References


Interpretation of material parameters in phase field models for ferroelectrics

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Phase field models have become an important tool for the simulation of ferroelectric domain structures. These models are based on an order parameter (usually the material polarization) which is introduced as an additional independent field variable. As a consequence of thermodynamic considerations, the evolution of the order parameter is governed by a time-dependent Ginzburg-Landau type equation. Traditionally the free energy is expanded at the ferroelectric phase transition from an assumed cubic (high-symmetry) parent phase to a low-symmetry phase exhibiting spontaneous polarization and strain. This results in a free energy satisfying the high-symmetry conditions so that piezoelectricity is not explicitly included in the free energy. We show how this shortcoming can be solved by expanding the free energy at the spontaneously polarized low-symmetry state. Similarly, we include dielectricity directly in the free energy. These extensions of the free energy lead to a slightly different interpretation of the order parameter but solve the difficult question of how to fit the coefficients of the Landau potential to achieve a desired electromechanical model response. In a next step we show how the key properties interface energy, width, and mobility can be input as material parameters by re-arranging the coefficients of a modified Landau potential and the gradient energy. In the end, all model parameters have a clear physical meaning, thus enhancing the usability of phase field models in the context of ferroelectrics. Finite element simulations are presented to illustrate the main results of the presented theoretical considerations.

References


Core/shell Nanoparticles: different synthetic routes of multiferroic cobalt ferrite/barium titanate and characterisation

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Abstract The nanocomposite cobalt ferrite/barium titanate with a core-shell structure have been prepared using a multistep process that combines colloidal chemistry methods and a solid state reaction (sol-gel, co-precipitation, hydrothermal). The obtained samples were characterized by scanning electron microscopy (SEM), X-ray diffraction (XRD) and transmission electron microscopy (TEM). The core-shell structure BaTiO$_3$@SiO$_2$ or Al$_2$O$_3$ have been successfully prepared by microemulsion. TEM results indicate that the silica shell uniformly encapsulates the BaTiO$_3$ core particles.

References
Magnetoelectric properties of 0.2 CoFe$_2$O$_4$-0.8 BaTiO$_3$ composite prepared by the organosol route

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The multiferroic 0.2 CoFe$_2$O$_4$-0.8 BaTiO$_3$ nano powder core-shell composite was successfully synthesized combining co-precipitation and organosol method. The powder was sintered to form a 0-3 composite ceramic. The morphology and the structure of the powder and the ceramic were studied by x-ray diffraction, SEM, TEM and AFM. Magnetic, ferroelectric and magnetoelectric properties were studied using SQUID, Sawyer-Tower circuit, and modified SQUID susceptometer respectively. Converse magnetoelectric coefficient measured by modified SQUID susceptometer reaches to value $4.4 \cdot 10^{-12}$ s/m for the applied magnetic field $\mu_oH_{dc} = 0.15$ T at $T = 285$ K.
Phase Field Simulation on the Influence of Point Defects with Domain Structures in Ferroelectrics

B. X. Xu, Y. Zuo
Mechanics of Functional Materials, TU Darmstadt

Widely used in actuator and sensor technologies and memory devices, piezoelectric ceramics are known to have the performance degradation problem, which especially includes aging and fatigue phenomena. It has been well recognized that the interaction of point defects with domain structures plays significant role in aging and fatigue [1, 2]. However, controversies regarding the exact mechanisms remain still unsolved. In this work numerical simulations based on a phase field model [3] are conducted to investigate different aspects of point defects, e.g. defect dipole and space charge accumulation. Results show various influences of these aspects on the domain stability and the hysteresis behavior [4].

References
Modeling and simulation of mechanical damage behavior of polycrystalline ferroelectric ceramics

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A concept for multiscale modeling and simulation of mechanical damage behavior of polycrystalline ferroelectric ceramics under electro-mechanical load will be discussed. All known models for the description of damage and fatigue of ferroelectrics don’t consider concurrently: ferroelectric nonlinear material behavior, influence of domain switching in the real microstructure, three-dimensional microstructures, inter- or intragranular fracture. Therefore until now a realistic evaluation of experimental fracture data is not really possible. The long-term goal must be to develop a comprehensive damage model which is able to:

- include the essential microscopic phenomena (domain switching, grain anisotropy)
- represent interaction of this phenomena on the meso-level (polycrystal)
- consider the crack initiation and the crack growth in piezo- and ferroelectric macroscopic structures.

This requires a ferroelectric material model for a single crystal (a domain configuration) and an efficient FE$^2$ method. The model from [1] can be taken for ferroelectric material description [2] and has to be extended to the rhomboedric phase as well as to the morphotropic phase. The model for a single crystal is prerequisite for the simulation of the material behavior of a polycrystal on the meso-level using statistically representative polycrystalline volume elements (RVE). A microscopic model is necessary because purely macroscopic mechanical approaches can not describe satisfactory the fracture toughness on micro-level [3]. Finally, for modeling of fatigue, damage, and crack growth in nonlinear electro-mechanical materials electro-mechanical cohesive laws are the particularly suitable approach.

References


Ferroelectricity, ferroelasticity, and charge transport in piezoceramics and their couplings: modeling and experiments

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The talk is divided into three parts. In the first part, we will talk about our work on phase-field modeling as part of a multi-scale simulation chain. The phase-field’s free energy densitity was adjusted to single domain properties computed by atomistic methods. Based on this, effective small and large signal properties of typical domain configurations were calculated as input for micromechanical models. In the second part, we discuss the significance of the presence of charge transport to the simulation of poling processes in piezoceramic components. The third part deals with the measurement of material parameters of ferroelectrics using the partial unloading method. This last part refers to some older work which is going to be continued now, and basically is meant as an offer for future cooperation.
A multi-scale computational procedure via coupling between finite-element and phase-field methods for microstructure evolution applications

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We have developed a multiscale computational procedure based on a multiscale constitutive model describing stressed microstructure evolution in polycrystalline materials [1]. The multiscale computational procedure is implemented in the Abaqus/Standard finite element program by writing a user material subroutine UMAT. A separate subroutine is also developed and embedded within the UMAT to calculate the microstructural evolution within a representative volume element (RVE) through phase-field simulations at mesoscale. Each finite-element integration point represents a macroscopic material point and its attached RVE. The macroscopic deformation gradient and macroscopic temperature at each finite-element integration point will be provided by the finite-element program. The respective mesoscopic counterparts of these macroscopic variables are then used to conduct phase-field simulations to determine the microstructure evolution within a RVE. We perform phase-field simulations by discretizing each RVE into a large number of subregions. Each subregion of a RVE has one grid point located at its centroid. The mesoscopic quantities e.g. free energy, stress, volume fractions of species, intensity variables etc. in a given subregion of a RVE are calculated at its grid point. The gradient and Laplacian of a given quantity are calculated using the finite-difference method. The numerical implementation of the constitutive model is then verified with respect to analytical solutions for some benchmark examples. We have also employed our coupled finite-element and phase-field modeling capability to simulate the phenomena of stressed microstructure and texture evolution as seen in the experiments [2]. With the aid of reasonably-made modeling assumptions, we are able to qualitatively and quantitatively describe the experimental evolution of crystallographic texture and grain size statistics as a result of grain boundary motion in polycrystalline copper thin films of different thicknesses subjected to the annealing process.

References


Ferroelectric nanogenerators coupled to an electric circuit for energy harvesting

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The direct transformation of ambient mechanical energy into electricity using ferroelectric nanogenerators is discussed within the context of usability for self-sustaining microelectronics. Thus, it is essential to store the generated electric energy within an accumulator or capacitor. However, the contact and charge status of the electric storage medium strongly influences the performance of the generator. This necessitates to couple the generator and the electric circuit to determine working points. Therefore, a phase field model for the ferroelectric generator is coupled with the response of a standard full-wave rectifier and a capacitor. Non-linear diode characteristics as well as energy losses are under consideration. The amount and the type of connections for the nanogenerators in the harvesting field are discussed to bridge from the nanoscale to electrical quantities for microelectronics. Naturally, the generators exhibit a favorable working point. For the design of nanogenerators we consider an appropriate substrate which prestresses the ferroelectric material by slightly different crystal lattice parameters to enforce energy conversion. The numerical simulation considers an electro-mechanical phase-field model with polarization as state variable. The complex boundary conditions can be considered within a finite element formulation. However, an enhanced numerical algorithm is required to handle the coupling to the electric circuit.

References
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