Phase-Field Modeling of Ferroelectric Ceramics: Identification of the Helmholtz Free Energy Density and Domain Simulation in Nanostructures

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Abstract

Mesoscopic phase-field modeling is the appropriate tool for simulating the evolution of domain structures in ferroelectric ceramics. All the information on the material is completely encoded in its Helmholtz Free Energy Density. In this paper, we present a physically sound method, to identify the coefficients in the Helmholtz Free Energy Density for Lead-Titanite and Lead-Zirconate-Titanite from physical properties obtained from project partners by ab initio and shell model potential methods. In addition, we present 3D finite element simulations of domain structures in free standing ferroelectric nanodots.

Keywords: coupled fields , homogenization, material properties, multiscale problems, nanomechanics, smart materials

1. Introduction

In technical sensor and actuator applications, the piezoelectric effect is usually exploited by means of ferroelectric materials. Ferroelectric materials are also candidates for realizing permanent memories and nano-electro-mechanical systems (NEMS). The electromechanical coupling properties of this class of materials are induced by their distinct microstructure and its evolution under electromechanical loading. Each grain in the polycrystal possesses a substructure of domains of uniform microdipol orientation characterized by the polarization vector. Prominent materials in this class are barium titanate, lead titanate and the solid solution lead zirconate-titanate which possesses optimum coupling properties.

2. Phase-Field Modeling of Ferroelectrics

The theory of phase-field modeling of ferroelectrics introduces the polarization of domains as continuous order parameter. Domain evolution is governed by minimizing the Total Helmholtz Free Energy Density of the ferroelectric system of a certain geometry for fixed strain and electric displacement. The heart of this theory is local energy density landscape, which encodes the crystallographic properties of the material. In particular, as a part of this energy density, the Landau energy as a higher order polynomial of polarization determines by its minima the spontaneous polarization states (Figure 1). A polarization gradient term enforces the occurrence of domains as localized regions with polarization changing in dependence on position. The evolution of polarization is governed by the time dependent Landau Ginzburg equation. Besides this PDE, mechanical equilibrium and Gauss' law has to be satisfied. Thus, phase-field modeling offers a physically sound way to understand microstructure evolution in ferroelectric materials.

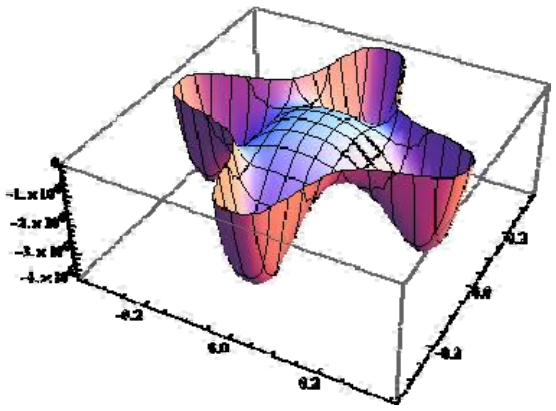

Figure 1: Landau Energy landscape

3. Helmholtz Free Energy Density

In the theory of phase-field modeling, the complete information on the material under consideration is encoded in a single function, namely in the Helmholtz Free Energy Density. Therefore, in order to obtain trustworthy simulation results, it is essential that the coefficients in this function are identified in a physically sound way. In this paper, we rely on physical properties of Lead-Titanite and Lead-Zirconate-Titanite obtained from project partners by ab initio and shell model potential methods [1]. Based on a sensitivity study, a strategy for the stepwise unique identification of all coefficients in the Helmholtz Free Energy Density has been developed to obtain

optimum adjustment to the physical properties calculated by ab initio and shell model potential methods.

4. Ferroelectric Nanostructures

In ferroelectric nanostructures with the dimensions on the order of magnitude of domain wall thickness, characteristic nanostructures may occur. A three-dimensional non-linear finite element has been developed for the implementation of the fully coupled electro-mechanical theory. Figure 2 shows a freestanding ferroelectric nanodot as a typical example for the kind of systems to be analyzed by such a tool [2].

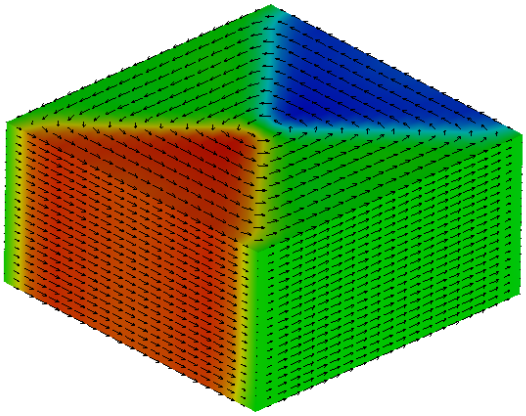

Figure 2: Freestanding ferroelectric nanodot

A flux closure structure of polarization occurs as a result of the competition between elastic energy, electric depolarization energy, and gradient energy and in response to the zero polarization boundary condition.

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References

[1] Voelker, B., Marton, P., Elsaesser, C., and Kamlah, M., Multiscale Modeling for ferroelectric materials: a transition from the atomic level to phase-field modeling, *Cont. Mech. Thermodyn.* (submitted), 2010.

[2] Wang, J., Kamlah, M., Domain control in ferroelectric nanodots through surface charges, *Appl. Phys. Lett.*, 93, 262904, 2008.