3D SIMULATION OF DOMAIN SWITCHING IN FERROELECTRICS BASED ON A PHASE FIELD MODEL

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Summary Ferroelectric ceramics are one kind of well developed functional materials and have been widely used e.g. in actuator, sensor and nonvolatile memory technologies. The application of ferroelectric ceramics relies on their favourable electro-mechanical coupling effect with high resolution and acceleration. The coupling effect is intrinsically based on variation of the polarization in the material. Polarization switching mechanism and the resultant microstructure evolution have been well recognized for ferroelectric ceramics. In this work, a phase field model for domain switching is formulated from a phase field potential which includes electrical enthalpy, domain separation energy and domain wall energy. Preliminary simulation results based on the finite element implementation of the model are documented to demonstrate the validity of the model.

INTRODUCTION

The development in the processing technology is accompanied by a widespread application of ferroelectric ceramics as actuators and sensors in micro-electro-mechanical systems, tunable microwave devices and nonvolatile memory technologies. As one of the typical functional materials, ferroelectric ceramics can respond to stimuli from different physics, namely the so-called multi-physics coupling effect. What makes the ferroelectrics favorable in application is primarily their large electro-mechanical coupling effect. This coupling is intrinsically based on its microstructure. In particular, perovskite ferroelectrics, one of the most important families in ferroelectrics, undergoes a cubic-to-tetragonal phase transformation when the material is cooled down below its Curie temperature. This phase transformation introduces spontaneous polarization and strain, with six variants determined by the crystallographic orientation of the polarization. Groups of unit cells with polarization of the same direction are called domains, which are separated by domain walls. The interaction of this micro domain structure with external electrical and/or mechanical loadings enables eventually the electro-mechanical coupling effect. Due to the heterogeneity and the micro scale, the switchable domain structure is not directly accessible to mesoscopic experimental measurements. Limited by the computation cost, atomistic simulation may also be insufficient for a fully understanding of the microstructure and its influences. The phase field theory has become an increasingly important simulation approach in the study of morphological evolution of heterogeneous materials and has lately been introduced to simulate the polarization and domain structure evolution of ferroelectrics. The method replaces the usual interface conditions by a partial differential equation for the evolution of the order parameter, i.e. the spontaneous polarization. The domain wall is therefore smoothed out with a finite thickness. The formulation is based on a free energy functional which includes the electrical enthalpy, the domain separation energy (polynomial of the order parameter) and the interfacial energy (the gradient term of the order parameter). The domain structure evolution is then treated as the consequence of the minimization process of the total free energy of the system, requiring no switching criteria and no prior assumption on domain geometries. In the following, we present a continuum phase field model and some preliminary simulations on spatial micro domain structures in single crystal ferroelectrics.

THE PHASE FIELD MODEL AND NUMERICS

Consider a ferroelectric body which occupies a region Ω with boundary $\partial\Omega$. The internal energy H in the body consists of three parts: the electrical enthalpy, the interfacial energy and the domain separation energy. The specific formulation is

$$\begin{aligned} H = \frac{1}{2}\big(\varepsilon_{ij} - \varepsilon^0_{ij}(P_m)\big)C_{ijkl}\big(\varepsilon_{kl} - \varepsilon^0_{kl}(P_m)\big) - \big(\varepsilon_{ij} - \varepsilon^0_{ij}(P_m)\big)\mathbb{b}_{kij}(P_m)E_k \\ -\frac{1}{2}A_{ij}E_iE_j - P_iE_i + \kappa\frac{G\epsilon}{P_0^2}P_{i,j}P_{i,j} + \gamma\frac{G}{\epsilon}\psi(P_m), \end{aligned} \tag{1}$$

in which ε_{ij} is the strain, C_{ijkl} the stiffness tensor, ε^0_{ij} the spontaneous strain, $\mathbb{b}_{kij}$ the piezoelectric tensor, E_k the electric field, A_{ij} the dielectric tensor, $P_{i,j}$ the gradient of the polarization, ψ the domain separation energy, G the domain wall energy, and ϵ the domain wall thickness, and P_0 the equilibrium polarization, and κ, γ the calibration constants. In particular, the spontaneous strain ε^0_{ij} and the piezoelectric tensor $\mathbb{b}_{kij}$ depend on the polarization [4], while the stiffness tensor C_{ijkl}, the dielectric tensor A_{ij}, the domain wall energy G and the domain wall thickness ϵ are assumed to be constant. Furthermore, ψ can be described by a polynomial function of the spontaneous polarization to form the domain

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variants. It may have the form

$$\psi = a_1 + \frac{a_2}{P_0^2}(P_1^2 + P_2^2 + P_3^2) + \frac{a_3}{P_0^4}(P_1^4 + P_2^4 + P_3^4) + \frac{a_4}{P_0^4}(P_1^2 P_2^2 + P_2^2 P_3^2 + P_1^2 P_3^2) + \frac{a_5}{P_0^6}(P_1^6 + P_2^6 + P_3^6), \quad (2)$$

in which a_i are coefficients. A contour plot of ψ with respect to the polarization components is shown in Fig. 1. For more information on H readers are referred to the work [1, 2, 3]. In the model, the linear kinematic relation and the definition of the electric field is used

$$\varepsilon_{ij} = \frac{1}{2}(u_{i,j} + u_{j,i}), \qquad E_i = -\varphi_i, \quad (3)$$

in which u_i is the displacement component, and φ is the electric potential. The constitutive equations are given in

$$\sigma_{ij} = \frac{\partial H}{\partial \varepsilon_{ij}} = C_{ijkl}\left(\varepsilon_{kl} - \varepsilon_{kl}^0\right) - \mathbb{b}_{kij} E_k, \qquad D_k = -\frac{\partial H}{\partial E_k} = \left(\varepsilon_{ij} - \varepsilon_{ij}^0\right)\mathbb{b}_{kij} + A_{ik} E_i + P_k. \quad (4)$$

The governing equations for the electro-mechanical problem and the Ginzburg-Landau type evolution equation for the polarization are

$$\sigma_{ij,j} + f_i = 0, \qquad D_{k,k} - q = 0, \qquad \frac{1}{M}\dot{P_m} - \left(2\kappa \frac{G\epsilon}{P_0^2} P_{m,jj} - \frac{\partial H}{\partial P_m}\right) = 0, \quad (5)$$

in which f_i is the volume force component, q the volume charge, and M the mobility parameter. The boundary conditions for the mechanical and the electric part may be of the Neumann type or the Dirichlet type

$$\sigma_{ij} n_j = t_i, \text{ on } \partial\Omega^t \qquad u_i = \bar{u}_i, \text{ on } \partial\Omega^u \qquad D_k n_k = -Q, \text{ on } \partial\Omega^Q \qquad \varphi = \bar{\varphi}, \text{ on } \partial\Omega^\varphi, \quad (6)$$

in which t_i is the mechanical traction component, and Q is the surface charge density, $\bar{u}_i$ and $\bar{\varphi}$ the prescribed displacement and the electric potential, respectively. Generally, the polarization may have a Robin type boundary condition

$$P_{m,j} n_j + P_m/\lambda = 0, \quad (7)$$

λ is of a length scale. It can be seen that for $\lambda \longrightarrow 0$ the Robin boundary condition reduces to the zero polarization boundary condition, and for $\lambda \longrightarrow \infty$ it reduces to the zero flux boundary condition.

RESULTS AND DISCUSSION

A 3D implementation of the model was completed using the finite element method with the program FEAP. An implicit time integration is used for the evolution, and the Newton iteration is adopted for the nonlinear system of equations. As an example, the domain structure evolution of $BaTiO_3$ single crystal cube has been simulated, in which the volume force and charge are neglected. The nanodot is under traction free and charge free boundary conditions, and for the polarization the zero flux condition is adopted. Starting from an initial random distribution, the domain structure evolves eventually to a vortex structure, with all the polarizations parallel to the surface, see Fig. 2.

It is noted that the resultant domain structures can be largely influenced by the domain wall energy G. When G is large enough so that it is more expensive to form domain wall than to create non-compensated charges on the boundary, the resultant domain structure will be not any more a regular vortex, and the polarization may not be parallel to the surface. In an upcoming project, phase field parameters will be particularly identified through comparison with experimental measurements on perovskite single crystals e.g. the poling curve and the hysteresis of barium titanate.

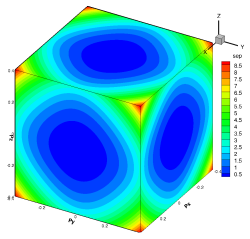

Figure 1. Domain separation energy

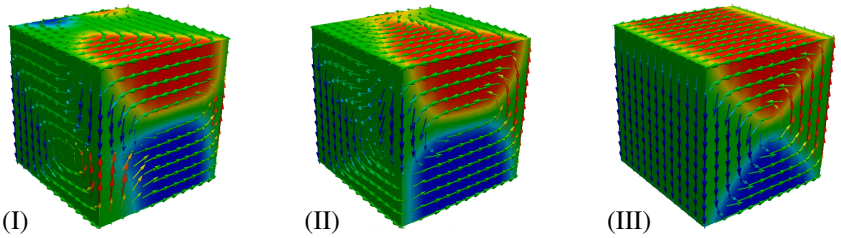

Figure 2. Domain structure evolution (I -> II -> III) of a free standing nanodot under charge free boundary condition

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