Available online at www.sciencedirect.com
ScienceDirect
Comput. Methods Appl. Mech. Engrg. 196 (2007) 4365–4374
Computer methods in applied mechanics and engineering
www.elsevier.com/locate/cma

Domain evolution in ferroelectric materials: A continuum phase field model and finite element implementation

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Received 17 November 2006; received in revised form 3 May 2007; accepted 8 May 2007
Available online 25 May 2007

Abstract

A numerical tool for simulating the polarization distribution in ferroelectric materials is presented. Using the concept of a phase field, a continuum physics model is established which is descretized with finite elements. The main feature of the numerical implementation is an implicit time integration of the non-linear evolution equation. Representative examples and comparison to experimental measurements from the literature show the main features of the model.

Keywords: Phase field model; Ferroelectrics; Finite elements; Micromechanics; Configurational forces

1. Introduction

The electro-mechanical properties of ferroelectric materials are determined by the microstructure, i.e. mainly the domain structure, of these materials. Understanding the microscopic phenomena is essential in the analysis of these materials. The microstructure is subject to change during different loading situations, as for example the application of an external electric field or an external mechanical stress. The present paper focuses on making these microstructural changes accessible for a simulation technique. Due to the versatile applicability for different non-linearities and boundary conditions, the model is implemented into an implicit Finite Element Method. Main features and relevant microstructural situations are analyzed to illustrate the capabilities of the model. Phase field models are well-established in many fields of material physics, as e.g. to model martensitic or diffusive phase transitions (see among others [9,19,11]). In the field of ferroelectrics the application is more involved through the electro-mechanical coupling. Many authors have used a Ginzburg-Landau type theory to describe the statics and dynamics of ferroelectric domain structures (e.g. [4,1,2,16]). These approaches treat the total polarization as the primary order parameter and the strain as a secondary, dependent order parameter. The application of these theories to realistic scenarios is limited due to the fact that the polarization is the only independent variable. Ferroelectric and ferroelastic switching can only be achieved through artificial terms in the total potential. More recently, in Xiao et al. [20] and Zhang and Bhattacharya [22,21] the electric potential was introduced as an independent variable which allows for the solution of the associated electro-static field problem. Xiao et al. [20] use a finite element formulation but fail to give any information about the implementation, whereas Zhang and Bhattacharya [22,21] adopt a finite difference scheme. Other recent works include Bhattacharya and Ravichandran [3], Soh et al. [13], Wang and Zhang [17,18]. The implementations frequently use finite difference schemes (sometimes in combination with FFT-techniques) and explicit time integration. In order to derive a robust and flexible implementation with respect to boundary conditions, we follow a finite element scheme in combination with an implicit time integration. A similar approach was

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doi:10.1016/j.cma.2007.05.010

taken in Su and Landis [14,15]. All cited phase field modeling approaches use the total material polarization as the order parameter. In contrast, our model uses the *spontaneous* polarization as (primary) order parameter, from which the spontaneous strain and the piezoelectric constants are derived as secondary order parameters.

In conjunction with previous works, Gross et al. [6], Mueller et al. [10], Schrade et al. [12], this allows us to introduce defects in the simulation. However this is not the main focus of the present work.

2. Theory

In order to make the present paper self-contained we briefly recast the basics of ferroelectrics in a shortened way, appropriate for the following numerical implementation.

The body under consideration occupies a region $\mathscr{B}$ with boundary $\partial\mathscr{B}$ and is assumed to have ferroelectric and piezoelectric properties. Neglecting inertia terms, body forces, and volume charges, the equilibrium conditions for the mechanical stress $\boldsymbol{\sigma}$ and the balance of the electric displacement $\boldsymbol{D}$ read

$$\begin{aligned} &\operatorname{div}\boldsymbol{\sigma} = \boldsymbol{0} \quad \text{in } \mathscr{B}, \\ &\operatorname{div}\boldsymbol{D} = 0 \quad \text{in } \mathscr{B}. \end{aligned} \tag{1}$$

The corresponding boundary conditions for either the mechanical displacement $\boldsymbol{u}$ or the surface traction $\boldsymbol{\sigma n}$ are

$$\begin{aligned} &\boldsymbol{u} = \boldsymbol{u}^* \quad \text{on } \partial\mathscr{B}_u, \\ &\boldsymbol{\sigma n} = \boldsymbol{t}^* \quad \text{on } \partial\mathscr{B}_t. \end{aligned} \tag{2}$$

For the electric problem the corresponding boundary conditions are specified either for the electric potential φ or the surface charge density $\boldsymbol{D}\cdot\boldsymbol{n}$:

$$\begin{aligned} &\varphi = \varphi^* \quad \text{on } \partial\mathscr{B}_\varphi, \\ &\boldsymbol{D}\cdot\boldsymbol{n} = -Q^* \quad \text{on } \partial\mathscr{B}_Q. \end{aligned} \tag{3}$$

In Eqs. (2) and (3) the vector $\boldsymbol{n}$ denotes the outer unit normal to the surface $\partial\mathscr{B}$. The phase field potential H of the system consists of three parts: the electric enthalpy H^{ent} (cf. [12]), a phase separation potential H^{sep}, and an interface energy H^{int}:

$$H = H^{\text{ent}} + H^{\text{sep}} + H^{\text{int}} \tag{4}$$

with

$$\begin{aligned} H^{\text{ent}} &= \frac{1}{2}(\boldsymbol{\varepsilon}-\boldsymbol{\varepsilon}^0) : [\mathbb{C}(\boldsymbol{\varepsilon}-\boldsymbol{\varepsilon}^0)] - (\boldsymbol{\varepsilon}-\boldsymbol{\varepsilon}^0) : \mathfrak{e}^{\mathrm{T}}\boldsymbol{E} \\ &\quad - \frac{1}{2}\boldsymbol{E}\cdot\boldsymbol{K}\boldsymbol{E} - \boldsymbol{P}\cdot\boldsymbol{E}, \\ H^{\text{sep}} &= \psi(\boldsymbol{P}), \\ H^{\text{int}} &= \frac{1}{2}\beta\|\nabla\boldsymbol{P}\|^2. \end{aligned} \tag{5}$$

Here $\boldsymbol{P}$ is the spontaneous polarization of the material, $\mathbb{C}$, $\mathfrak{e}$, and $\boldsymbol{K}$ denote the elastic stiffness, the piezoelectric coupling constants, and the dielectric tensor, respectively. The remanent strain $\boldsymbol{\varepsilon}^0$ is an inelastic eigenstrain which describes the "phase transition" from one polarization state to another. The scalar β is the scaling parameter of the interface energy. The mechanical strain $\boldsymbol{\varepsilon}$ and the electric field $\boldsymbol{E}$ are defined as

$$\begin{aligned} \boldsymbol{\varepsilon} &= \frac{1}{2}(\nabla\boldsymbol{u} + (\nabla\boldsymbol{u})^{\mathrm{T}}), \\ \boldsymbol{E} &= -\nabla\varphi, \end{aligned} \tag{6}$$

respectively.

The constitutive equations can be derived from the potential by standard arguments of rational thermodynamics. These arguments yield

$$\boldsymbol{\sigma} = \frac{\partial H}{\partial\boldsymbol{\varepsilon}} = \mathbb{C}(\boldsymbol{\varepsilon}-\boldsymbol{\varepsilon}^0) - \mathfrak{e}^{\mathrm{T}}\boldsymbol{E}, \tag{7}$$

and

$$\boldsymbol{D} = -\frac{\partial H}{\partial\boldsymbol{E}} = \mathfrak{e}(\boldsymbol{\varepsilon}-\boldsymbol{\varepsilon}^0) + \boldsymbol{K}\boldsymbol{E} + \boldsymbol{P}. \tag{8}$$

The time evolution of the phase field (order) parameter $\boldsymbol{P}$ is given by a Ginzburg-Landau type equation. Due to the appearance of the gradient of $\boldsymbol{P}$ in the potential the variational derivative appears in the evolution equation

$$\begin{aligned} \dot{\boldsymbol{P}} &= -\alpha\frac{\delta H}{\delta\boldsymbol{P}} = -\alpha\left(\frac{\partial H}{\partial\boldsymbol{P}} - \operatorname{div}\frac{\partial H^{\text{int}}}{\partial\nabla\boldsymbol{P}}\right) \\ &= -\alpha\left(\frac{\partial H^{\text{ent}}}{\partial\boldsymbol{P}} + \frac{\partial\psi}{\partial\boldsymbol{P}} - \beta\Delta\boldsymbol{P}\right), \end{aligned} \tag{9}$$

where α is another scalar parameter, which describes the mobility of the process. This lets the spontaneous polarization evolve such that the total potential is reduced in the course of the evolution. For details of the evaluation of the variation and the partial derivatives the reader is referred to Appendix A. The boundary conditions for the spontaneous polarization may be specified as

$$\begin{aligned} &\boldsymbol{P} = \boldsymbol{P}^* \quad \text{on } \partial\mathscr{B}_P, \\ &\nabla\boldsymbol{P}\boldsymbol{n} = \boldsymbol{\pi}^* \quad \text{on } \partial\mathscr{B}_\pi. \end{aligned} \tag{10}$$

The determination of the boundary conditions on the order parameter is not a trivial task. In the following we will tacitly assume homogeneous Neumann type boundary conditions, i.e. $\boldsymbol{\pi}^* = \boldsymbol{0}$ in (10). In view of their importance in the phase transition, the inelastic strain and the piezoelectric constants are chosen to depend on the spontaneous polarization. The eigenstrain is constructed to be purely deviatoric, and ε^0 resembles the strain for the fully poled state with respect to the paraelectric phase. With reference to Kamlah [8] we have

$$\boldsymbol{\varepsilon}^0(\boldsymbol{P}) = \frac{3}{2}\varepsilon^0\frac{\|\boldsymbol{P}\|}{P_0}\left\{\boldsymbol{e}\otimes\boldsymbol{e} - \frac{1}{3}\boldsymbol{1}\right\}, \tag{11}$$

where $\boldsymbol{e}$ is the unit vector in the poling direction, i.e. $\boldsymbol{e} = \boldsymbol{P}/\|\boldsymbol{P}\|$, and P_0 is the equilibrium polarization for zero applied loading. With this definition the spontaneous strain is linear in the spontaneous polarization. It is noted that