

On the modeling of microscopic switching phenomena in tetragonal ferroelectric materials

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Abstract

Ferroelectric materials play a key role for the functionality of many high-tech electro-mechanically coupled devices, as for instance in actuator and sensor technology, and are therefore of great interest for both industry and science. In general, these materials are characterized by a pronounced electro-mechanical coupling. However, when applied to high electrical or mechanical fields, the material response becomes highly nonlinear. This results in an electro-mechanical hysteretic behavior, which is due to polarization switching in the microstructure of the material. Under electric loading, the well-known ferroelectric butterfly and dielectric hysteresis loops can be observed. On the other hand, polarization switching can also be induced when the material is exposed to high mechanical loads, which results in ferroelastic-type hystereses. Such nonlinear electro-mechanically coupled behavior is crucial for the prediction and the analysis of ferroelectric devices. Therefore, reliable models for the mathematical description of the complex, ferroelectric material behavior have to be formulated, which account for the mentioned nonlinearities. The present work will discuss a microscopically motivated switching criterion that is capable of reflecting the ferroelectric and ferroelastic hysteresis from a microscopic view point.

Keywords: anisotropy, coupled fields, finite element methods, microstructures, solid mechanics, smart materials

1. Introduction

Switching phenomena in ferroelectric materials have extensively been studied both with experimental and theoretical methods and are still part of current research. For a general overview of the ferroelectric effect the reader is referred e.g. to the standard text books JAFFE ET AL. [2] and LINES & GLASS [7]. A review paper on the constitutive modeling of the ferroelectric and ferroelastic effect as well as the basic properties of ferroelectric ceramics is given by KAMLAH [3].

In general, ferroelectric materials as e.g. BaTiO₃ have a crystalline structure. The main characteristic of such ferroelectric materials is their inherent *spontaneous polarization* which is a microscopic property of the material. In case of BaTiO₃, this spontaneous polarization is the result of a phase transition from a cubic to a tetragonal phase which occurs at a certain temperature, the so-called Curie temperature. Below this temperature barrier, the tetragonal unit cells of the crystalline ferroelectric possess a spontaneous polarization and are therefore termed spontaneously polarized. However, it has to be distinguished between the above mentioned microscopic properties and the resulting overall macroscopic properties of the underlying material, in this context see also SCHRÖDER & KEIP [9]. Since each microscopic spontaneous polarization is characterized by a certain amount and a certain direction in space, the resulting macroscopic polarization can strongly differ from the corresponding microscopic counterpart.

When dealing with the constitutive modeling of such ferroelectric materials, it has to be distinguished between microscopically and macroscopically based formulations of the ferroelectric material behavior. The macroscopic material models understand the polarization as a macroscopic property and are thus mostly formulated based on a macroscopic, phenomenological theory, see e.g. LANDIS [6] and KLINKE [5]. A phenomenological,

mesoscopically motivated model is given by SCHRÖDER & ROMANOWSKI [10]. On the other hand, microscopic models refer to the polarization as a discrete microscopic property and are therefore formulated by means of discrete switching criteria, see e.g. HWANG ET AL. [1].

The present work focuses on the description of polarization switching as a microscopic phenomenon. The microscopic formulation will be given in terms of a coordinate-invariant electric enthalpy function for tetragonal ferroelectric crystals.

2. Electro-mechanically coupled boundary value problem

Let \mathcal{B} be the body of interest that is parameterized in x . Furthermore, let \mathbf{u} denote the displacement field and ϕ the scalar electric potential. The governing balance equations for the electro-mechanical boundary value problem are the balance of momentum and Gauß's law

$$\operatorname{div} \boldsymbol{\sigma} + \mathbf{f} = \mathbf{0} \quad \text{and} \quad \operatorname{div} \mathbf{D} = \rho_f \quad \text{in } \mathcal{B}, \quad (1)$$

where $\boldsymbol{\sigma}$ denotes the mechanical stress tensor, \mathbf{f} the vector of body forces, \mathbf{D} is the vector of electric displacements, and ρ_f is the density of free charge carriers. The basic kinematical relations are given by the mechanical linear strain tensor and the electric field vector

$$\boldsymbol{\varepsilon} = \frac{1}{2}(\operatorname{grad} \mathbf{u} + \operatorname{grad}^T \mathbf{u}) \quad \text{and} \quad \mathbf{E} = -\operatorname{grad} \phi. \quad (2)$$

To account for the dissipative nature of the material, the mechanical strains and the electric displacements are additively splitted into a reversible, elastic and an irreversible, remanent part

$$\boldsymbol{\varepsilon} = \boldsymbol{\varepsilon}^e + \boldsymbol{\varepsilon}^r \quad \text{and} \quad \mathbf{D} = \mathbf{D}^e + \mathbf{P}^r, \quad (3)$$

where \mathbf{P}^r denotes the remanent polarization vector.

3. Electric enthalpy function in tetragonal setting

The description of the ferroelectric material will be realized by means of a coordinate-invariant formulation of the electric enthalpy function in tetragonal setting, see KEIP & SCHRÖDER [4]. In this context, the tetragonal structure of the material is defined by means of three perpendicular crystallographic axes $\mathbf{a}_1, \mathbf{a}_2$, and $\mathbf{a}_3 = \mathbf{c}$ with $\|\mathbf{a}_{(s)}\| = 1$, where \mathbf{c} represents the normalized preferred direction of the tetragonal unit cell. Based on that, the electric enthalpy function can be given as an additive function of a mechanical, piezoelectric, and dielectric part as

$$H = H_{mech} + H_{piezo} + H_{diel}, \quad (4)$$

where the individual terms are given as

$$\begin{aligned} H_{mech} &= \frac{1}{2} \lambda I_1^2 + \mu I_2 + \frac{1}{2} \alpha_1 I_5^2 + \alpha_2 I_1 I_5 \\ &\quad + \frac{1}{2} \alpha_3 (I_3^2 + I_4^2) + \frac{1}{2} \alpha_4 (I_5^2 + I_6^2 + I_7^2), \\ H_{piezo} &= \beta_1 K_1 + \beta_2 I_1 J_2 + \beta_3 I_5 J_2, \\ H_{diel} &= \frac{1}{2} \gamma_1 J_1 + \frac{1}{2} (\gamma_2 - \gamma_1) J_2^2 - J_2 P_s. \end{aligned} \quad (5)$$

The underlying principal and mixed invariants are defined as

$$\begin{aligned} I_1 &= \text{tr}[\boldsymbol{\varepsilon}^e], \quad I_2 = \text{tr}[(\boldsymbol{\varepsilon}^e)^2], \quad I_3 = \text{tr}[\boldsymbol{\varepsilon}^e \boldsymbol{\Xi}_1], \quad I_4 = \text{tr}[\boldsymbol{\varepsilon}^e \boldsymbol{\Xi}_2], \\ I_5 &= \text{tr}[\boldsymbol{\varepsilon}^e \mathbf{M}_{33}], \quad I_6 = \text{tr}[\boldsymbol{\varepsilon}^e \mathbf{M}_{11}], \quad I_7 = \text{tr}[\boldsymbol{\varepsilon}^e \mathbf{M}_{22}], \\ J_1 &= \text{tr}[\mathbf{E} \otimes \mathbf{E}], \quad J_2 = \text{tr}[\mathbf{E} \otimes \mathbf{c}], \quad K_1 = \text{tr}[\mathbf{E} \otimes (\boldsymbol{\Xi}_3 : \boldsymbol{\varepsilon}^e)], \end{aligned} \quad (6)$$

wherein the structural tensors denote

$$\begin{aligned} \mathbf{M}_{ij} &= \mathbf{a}_i \otimes \mathbf{a}_j, \\ \boldsymbol{\Xi}_1 &= (\mathbf{M}_{13} + \mathbf{M}_{31}), \quad \boldsymbol{\Xi}_2 = (\mathbf{M}_{23} + \mathbf{M}_{32}), \end{aligned} \quad (7)$$

$$\boldsymbol{\Xi}_3 = \sum_{i=1}^3 (\mathbf{a}_i \otimes \mathbf{a}_i \otimes \mathbf{c} + \mathbf{a}_i \otimes \mathbf{c} \otimes \mathbf{a}_i).$$

The coordinate-invariant material parameters are given by $\lambda, \mu, \alpha_1, \alpha_2, \alpha_3, \alpha_4$ for the mechanical part, $\beta_1, \beta_2, \beta_3$ for the piezoelectric part, and γ_1, γ_2 for the dielectric part; P_s is the spontaneous polarization. Regarding the formulation of coordinate invariant energies for piezoelectric solids including the identification of material parameters the reader is referred to SCHRÖDER & GROSS [8].

4. Microscopic switching model

The present work utilizes a microscopically motivated, energy based ferroelectric/ferroelastic switching criterion for the modeling of the nonlinear switching effects in ferroelectric materials. The switching condition for the tetragonal unit cells is based on the change of the free energy as

$$d\mathcal{W} = d\mathcal{W}_{elec} + d\mathcal{W}_{mech} = \mathbf{E} \cdot d\mathbf{D} + \boldsymbol{\sigma} \cdot d\boldsymbol{\varepsilon}, \quad (8)$$

compare e.g. HWANG ET AL. [1]. Herein, the change of electric energy $d\mathcal{W}_{elec}$ can be approximated by the product of the electric field and the change of polarization during a discrete switching event, i.e.

$$\Delta\mathcal{W}_{elec} = \int d\mathcal{W}_{elec} \approx \mathbf{E} \cdot \Delta\mathbf{D} = \mathbf{E} \cdot \Delta\mathbf{P}^r, \quad (9)$$

supposed that the electric field stays constant and linear effects of the electric displacement are not taken into account during switching. On the other hand, supposed that the stresses stay constant during a discrete switching event, the change of mechanical energy $d\mathcal{W}_{mech}$ can be approximated by the product of the mechanical stresses and the change of remanent strains, i.e.

$$\Delta\mathcal{W}_{mech} = \int d\mathcal{W}_{mech} \approx \boldsymbol{\sigma} \cdot \Delta\boldsymbol{\varepsilon} = \boldsymbol{\sigma} \cdot \Delta\boldsymbol{\varepsilon}^r. \quad (10)$$

The electric work that dissipates during the switching process can be given by

$$\mathcal{W}_{e,180^\circ}^{diss} = 2 P_s E_c \quad \text{and} \quad \mathcal{W}_{e,90^\circ}^{diss} = \sqrt{2} P_s E_c \quad (11)$$

for 180° switching and 90° switching, respectively; E_c denotes the coercive electric field strength. Based on that, purely ferroelectric switching initiates whenever a critical energy barrier is reached, i.e. $\Delta\mathcal{W}_{elec} \geq \mathcal{W}_{e,180^\circ/90^\circ}^{diss}$. This results in five switching criteria for five variants of ferroelectric switching in a tetragonal unit cell

$$\mathbf{E} \cdot \Delta\mathbf{P}_{180^\circ}^r \geq \mathcal{W}_{e,180^\circ}^{diss} \quad \text{and} \quad \mathbf{E} \cdot \Delta\mathbf{P}_{i,90^\circ}^r \geq \mathcal{W}_{e,90^\circ}^{diss} \quad (12)$$

for $i = 1, \dots, 4$. In the present work, the polarization vector as well as the remanent strains are directly connected to the crystal lattice in the sense that $\mathbf{P}^r = P_s \mathbf{c}$ and $\boldsymbol{\varepsilon}^r = \frac{3}{2} \varepsilon_s \text{dev}(\mathbf{c} \otimes \mathbf{c})$, where ε_s denotes the saturation strain. Due to this fact, a change of polarization goes along with a change of the crystal lattice orientation resulting in a corresponding update of the constitutive mechanical, piezoelectric, and dielectric moduli which can be computed from the electric enthalpy function given in (5). For the onset of mechanically induced switching, similar switching criteria to the one formulated above can be derived, see e.g. HWANG ET AL. [1].

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