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An invariant formulation for phase field models in ferroelectrics

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ABSTRACT

This paper introduces an electro-mechanically coupled phase field model for ferroelectric domain evolution based on an invariant formulation for transversely isotropic piezoelectric material behavior. The thermodynamic framework rests upon Gurtin's notion of a micro-force system in conjunction with an associated micro-force balance. This leads to a formulation of the second law, from which a generalized Ginzburg–Landau evolution equation is derived. The invariant formulation of the thermodynamic potential provides a consistent way to obtain the order parameter dependent elastic stiffness, piezoelectric, and dielectric tensor. The model is reduced to 2d and implemented into a finite element framework. The material constants used in the simulations are adapted to meet the thermodynamic condition of a vanishing micro-force. It is found that the thermodynamic potential taken from the literature has to be extended in order to avoid a loss of positive definiteness of the stiffness and the dielectric tensor. The small-signal response is investigated in the presence and in the absence of the additional regularizing terms in the potential. The simulations show the pathological behavior of the model in case these terms are not taken into account. The paper closes with microstructure simulations concerning a ferroelectric nanodot subjected to an electric field, a cracked single crystal, and a ferroelectric bi-crystal.

1. Introduction

Ferroelectric materials are widely used in various industrial applications, such as sensor and actuator technology, non-volatile memory devices, micro-fluidics, transducers, and many more (Scott, 2007). Their macroscopic electro-mechanical properties depend on the ferroelectric domain structure which is subject to change under externally applied loading. The evolution of the domain structure is thus a key point in understanding and engineering the macroscopic properties of ferroelectric functional devices.

Phase field modeling provides a well-established and physically sound way to simulate evolving domain structures. The models currently in use are based on a continuum thermomechanics approach which incorporates an order parameter (generally the electrical polarization) as well as its first spatial gradient. The main differences between these models lie in the choice of the order parameter, the way in which the free energy is formulated, and in the numerical solution strategy. Traditionally the order parameter is the material polarization, and the free energy function is expanded to reflect cubic symmetry conditions with respect to the primary thermodynamic variables (strain and electric displacement), see e.g. the more recent publications (Völker and Kamlah, 2012; Xu et al., 2013; Chen, 2008; Su et al., 2011; Wang and Su, 2011) and the literature review given in Schrade et al. (2013). Since in these models there is no direct coupling between the electric displacement and the mechanical strain, the piezoelectric behavior is encoded in the Landau potential which is a polynomial in the order parameter. The resulting difficulties of fitting the small-signal response to the bulk properties of the material are addressed in detail in Völker et al. (2011, 2012) Another possibility is to meet the symmetry requirements of the spontaneously polarized state and thus to include the piezoelectric coupling terms in the free energy. The Landau free energy can then be used to adjust the properties of domain walls, see Schrade et al. (2013, 2008, 2007). As for numerical solution strategies, Fourier spectral methods appear to be quite time efficient (Chen and Shen, 1998) while imposing restrictions on geometry, applicable boundary conditions, and material inhomogeneities. Another approach based on a staggered solution algorithm with explicit time integration was taken in Zhang and Bhattacharya (2005a,b). The various shortcomings of such numerical implementations can be circumvented by finite element

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implementations (however at the expense of simulation time), see e.g. Su and Landis (2007), Schrade et al. (2007) and Miehe et al. (2012).

The cited publications (Schrade et al., 2013, 2008, 2007) are however limited to an isotropic mechanical stiffness and dielectric tensor. Motivated by Keip et al. (2014), in this article we intend to overcome these shortcomings by introducing a phase field model based on an invariant formulation for transverse isotropy which is adapted to fit into the existing phase field framework. The main problem in this adaption procedure is shown to be a loss of positive definiteness of the stiffness and the dielectric tensor. This problem is solved by extending the thermodynamic potential so that the material tangent remains positive definite for arbitrary polarization states. In a second step, the small-signal model response is compared with the input parameters of the simulation model. The input parameters are then adjusted under the condition of a vanishing micro-force. Numerical examples illustrate the necessity for extending the invariant description and adjusting the input parameters.

2. Phase field model

2.1. Basic equations and thermodynamical framework

We make use of Gurtin and Fried's notion of a generalized micro-force thermodynamics (Fried and Gurtin, 1993, 1994; Gurtin, 1996) and, following (Su and Landis, 2007), apply this theory to a phase field model. Since the current thermodynamical approach is already laid out in detail in Schrade et al. (2013), we will only summarize the main aspects in this regard while trying to keep the presentation self contained.

In the presence of volume forces $\boldsymbol{f}$ and volume charges ρ the ferroelectric body $\mathcal{B}$ under consideration obeys the mechanical and electrical field equations

$$\operatorname{div}\boldsymbol{\sigma}+\boldsymbol{f}=\boldsymbol{0}, \quad \operatorname{div}\boldsymbol{D}-\rho=0, \tag{1}$$

where $\boldsymbol{\sigma}$ is the stress tensor and $\boldsymbol{D}$ is the electric displacement. The linearized strain tensor $\boldsymbol{\varepsilon}$ and the electric field $\boldsymbol{E}$ are defined by the symmetrical and the negative gradient of the displacement field $\boldsymbol{u}$ and the electric potential φ:

$$\boldsymbol{\varepsilon}=\frac{1}{2}\left(\nabla\boldsymbol{u}+(\nabla\boldsymbol{u})^{\mathrm{T}}\right), \quad \boldsymbol{E}=-\nabla\varphi. \tag{2}$$

The thermodynamic framework consists of a micro-force system involving a micro-stress tensor $\boldsymbol{\Sigma}$ and the internal and external micro-force vector $\boldsymbol{g}$ and ζ, respectively. For an arbitrary control volume $\mathcal{R}$ with boundary $\partial\mathcal{R}$ and outer unit normal $\boldsymbol{n}$ the power expenditure of each of these quantities reads

$$\int_{\partial\mathcal{R}}(\boldsymbol{\Sigma}\boldsymbol{n})\cdot\dot{\boldsymbol{P}}\,\mathrm{d}a, \quad \int_{\mathcal{R}}\boldsymbol{g}\cdot\dot{\boldsymbol{P}}\,\mathrm{d}v, \quad \int_{\mathcal{R}}\zeta\cdot\dot{\boldsymbol{P}}\,\mathrm{d}v. \tag{3}$$

The physical meaning of the order parameter $\boldsymbol{P}(\boldsymbol{x},t)$ depends on the concrete choice of the thermodynamic potential (Schrade et al., 2013). As will be shown in Section 2.2, $\boldsymbol{P}(\boldsymbol{x},t)$ here is the material polarization less its dielectric and piezoelectric parts. Changes in the order parameter reflect reorganization in the microstructure. Such reorganization is accompanied by a dissipative process and by changes in the thermodynamic potential, both of which are taken into account by the internal micro-force. Referring to Gurtin (1996), we assume the local micro-force balance

$$\operatorname{div}\boldsymbol{\Sigma}+\zeta+\boldsymbol{g}=\boldsymbol{0}. \tag{4}$$

The second law of thermodynamics balances the power expenditure of external sources with changes in the Helmholtz free energy $\Psi=\tilde{\Psi}(\boldsymbol{\varepsilon},\boldsymbol{D},\boldsymbol{P},\nabla\boldsymbol{P})$; hence we have to include $(3)_1$ and $(3)_3$ in the second law:

$$\int_{\partial\mathcal{R}}\left((\boldsymbol{\sigma}\boldsymbol{n})\cdot\dot{\boldsymbol{u}}-\varphi\frac{\mathrm{d}}{\mathrm{d}t}(\boldsymbol{D}\cdot\boldsymbol{n})+(\boldsymbol{\Sigma}\boldsymbol{n})\cdot\dot{\boldsymbol{P}}\right)\mathrm{d}a + \int_{\mathcal{R}}\left(\boldsymbol{f}\cdot\dot{\boldsymbol{u}}+\varphi\dot{\rho}+\zeta\cdot\dot{\boldsymbol{P}}\right)\mathrm{d}v-\frac{\mathrm{d}}{\mathrm{d}t}\int_{\mathcal{R}}\Psi\,\mathrm{d}v\geqslant 0. \tag{5}$$

In accordance with current ferroelectric phase field modeling, electrostatic forces (cf. McMeeking and Landis, 2005) are not taken into account so that the Cauchy stress $\boldsymbol{\sigma}$ is symmetric. After a Legendre transform of the Helmholtz free energy according to

$$H=\Psi-\boldsymbol{D}\cdot\boldsymbol{E}, \tag{6}$$

we obtain (cf. Nowacki et al., 1979) the electric enthalpy $H=\tilde{H}(\boldsymbol{\varepsilon},\boldsymbol{E},\boldsymbol{P},\nabla\boldsymbol{P})$ and write

$$\int_{\partial\mathcal{R}}\left((\boldsymbol{\sigma}\boldsymbol{n})\cdot\dot{\boldsymbol{u}}+(\boldsymbol{D}\cdot\boldsymbol{n})\dot{\varphi}+(\boldsymbol{\Sigma}\boldsymbol{n})\cdot\dot{\boldsymbol{P}}\right)\mathrm{d}a + \int_{\mathcal{R}}\left(\boldsymbol{f}\cdot\dot{\boldsymbol{u}}-\rho\dot{\varphi}+\zeta\cdot\dot{\boldsymbol{P}}\right)\mathrm{d}v-\frac{\mathrm{d}}{\mathrm{d}t}\int_{\mathcal{R}}H\,\mathrm{d}v\geqslant 0. \tag{7}$$

The internal micro-force $\boldsymbol{g}$ is omitted in the second law as it is not part of the external power supply. Combining the local form of (7) with (4), one obtains by standard arguments of rational thermomechanics

$$\boldsymbol{\sigma}=\frac{\partial H}{\partial\boldsymbol{\varepsilon}}, \quad \boldsymbol{D}=-\frac{\partial H}{\partial\boldsymbol{E}}, \quad \boldsymbol{\Sigma}=\frac{\partial H}{\partial\nabla\boldsymbol{P}}. \tag{8}$$

Insertion of these relations in the local form of the second law yields the residual dissipation inequality

$$-(\boldsymbol{g}+\boldsymbol{\eta})\cdot\dot{\boldsymbol{P}}\geqslant 0, \tag{9}$$

where

$$\boldsymbol{\eta}=\frac{\partial H}{\partial\boldsymbol{P}} \tag{10}$$

is the micro-force vector. The term in the parenthesis is identified as the dissipative micro-force

$$\boldsymbol{g}_{\mathrm{dis}}=\boldsymbol{g}+\boldsymbol{\eta}. \tag{11}$$

The residual dissipation inequality (9) is satisfied (by means of a sufficient condition) by assuming

$$\boldsymbol{g}_{\mathrm{dis}}=-\boldsymbol{\beta}\dot{\boldsymbol{P}}, \tag{12}$$

where $\boldsymbol{\beta}$ is a positive semi-definite second order inverse mobility tensor (Gurtin, 1996). The dissipation $\mathcal{D}$ which takes place due to the evolving microstructure is then given by

$$\mathcal{D}=\int_{\mathcal{B}}-\boldsymbol{g}_{\mathrm{dis}}\cdot\dot{\boldsymbol{P}}\,\mathrm{d}v=\int_{\mathcal{B}}\dot{\boldsymbol{P}}\cdot(\boldsymbol{\beta}\dot{\boldsymbol{P}})\,\mathrm{d}v\geqslant 0. \tag{13}$$

We can now see that the internal micro-force

$$\boldsymbol{g}=\boldsymbol{g}_{\mathrm{dis}}-\boldsymbol{\eta}=-\boldsymbol{\beta}\dot{\boldsymbol{P}}-\frac{\partial H}{\partial\boldsymbol{P}} \tag{14}$$

has a dissipative and a non-dissipative contribution. An evaluation of the residual dissipation inequality leads to a generalized form of the Ginzburg–Landau equation (Schrade et al., 2013):

$$\boldsymbol{\beta}\dot{\boldsymbol{P}}=\operatorname{div}\boldsymbol{\Sigma}+\zeta-\frac{\partial H}{\partial\boldsymbol{P}}. \tag{15}$$

We limit ourselves to Dirichlet and Neumann boundary conditions and neglect the vacuum polarization outside of $\mathcal{B}$:

$$\begin{aligned}&\boldsymbol{u}-\boldsymbol{u}^{*}=\boldsymbol{0} \quad \text{on } \partial\mathcal{B}_u, \boldsymbol{\sigma}\boldsymbol{n}-\boldsymbol{t}^{*}=\boldsymbol{0} \quad \text{on } \partial\mathcal{B}_\sigma,\\ &\varphi-\varphi^{*}=0 \quad \text{on } \partial\mathcal{B}_\varphi, \boldsymbol{D}\cdot\boldsymbol{n}+\rho_{\mathrm{s}}^{*}=0 \quad \text{on } \partial\mathcal{B}_D,\\ &\boldsymbol{P}-\boldsymbol{P}^{*}=\boldsymbol{0} \quad \text{on } \partial\mathcal{B}_P, \boldsymbol{\Sigma}\boldsymbol{n}-\boldsymbol{\pi}^{*}=\boldsymbol{0} \quad \text{on } \partial\mathcal{B}_\Sigma,\end{aligned} \tag{16}$$

where $\boldsymbol{t}^{*}$, ρ_{s}^{*}, and $\boldsymbol{\pi}^{*}$ are prescribed surface tractions, surface charge densities, and polarization fluxes, respectively. Initial values for the order parameter are prescribed at every material point by