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Simulations of gradual domain-switching in polycrystalline ferroelectrics using an optimization-based, multidomain-grain model

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ABSTRACT

An optimization-based computational model is proposed to study the constrained domain switching in polycrystalline ferroelectrics composed of numerous grains, each of which consists of multiple domains. Domain switching is realized by an optimization algorithm to minimize the free energy of each grain and is thus a gradual process with applied loading. Similar to phase field modeling, no *priori* domain-switching criterion is imposed in the proposed model and its computational efficiency is much higher. The domain textures evolution process can also be captured. Simulation results on both tetragonal and rhombohedral PZT ceramics illustrate the efficiency of this model.

1. Introduction

Ferroelectric materials, such as barium titanate ($BaTiO_3$), lead titanate zirconate (PZT), etc., are widely used as actuators, transducers, etc., due to their electromechanical coupling, ultra-fast response and compact size [1]. These types of materials show excellent linear response at low electric fields but under a large electric field or high stress shows significant nonlinearities due to domain switching [2]. In ferroelectric single crystals, complete domain switching can be achieved and a single domain state can exist after poling by a strong DC field. While this is not the case in ferroelectric ceramics where the crystallite axes arrange in a random way and multiple domain states still exist after complete poling. Strain measurements and X-ray diffraction studies have shown that after poling the volume fraction of completed 90° domain switching is only 10–12% in $BaTiO_3$ ceramics and about 50% in tetragonal PZT near the morphotropic phase boundary (MPB) [3]. This implies that a considerable amount of 90° domain switching has been constrained in polycrystalline ferroelectrics and cannot occur under conventional poling.

Modeling of domain switching in ferroelectric materials has received intensive attentions in recent years, including phenomenological models [4,5], micromechanical models [2,6] and phase field models [7–9]. The phenomenological models [4,5] are powerful in addressing the macroscopic nonlinear behavior of ferroelectrics, while they cannot predict the detailed domain switching process as no info of domains is included in this type of models. Theoretically, the micromechanical models [2,6] can capture both the macroscopic nonlinearities and the microscopic domain evolutions in ferroelectrics. However, a priori domain switching criterion must be prescribed in the micromechanical models and most of these models take the single-domain-grain assumption thus domain switching is always a sudden event instead of the gradual process as observed in polycrystalline ferroelectrics. In comparison, the phase field model (PFM) does not impose any *priori* domain switching criterion and domain switching is a natural process during minimizing the total free energy of the whole material system. PFM had achieved great success in modeling domain evolution in ferroelectric single crystals [7–9], while its applications in polycrystalline ferroelectrics are very few [10], probably because of the difficulties in addressing the complicated interactions between grains and the computational complexity in three dimensional cases. In our recent works [11,12], the elastic interactions in both tetragonal and rhombohedral ferroelectric ceramics are investigated by an analytical, constrained domain switching model. Note that in that model [11,12], a priori domain switching criterion and switching path must be prescribed and only uni-axial loading is allowed to get the analytical solution, thus it is not suitable for a general computational study of domain evolution in polycrystalline ferroelectrics under arbitrary loading.

In this work, we propose an optimization-based computational model to study the constrained domain switching in polycrystalline ferroelectrics composed of numerous grains, each of which consists of multiple domains. The total free energy in each grain is set as the optimization objective using the volume fractions of

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each type of domains as optimization variables. The optimization process is realized by the sequential quadratic programming (SQP) method for constrained optimization problem [13]. Like the phase field model (PFM), this computational model also does not impose any *priori* domain switching criterion. The computational complexity of this proposed model is fairly small compared to the PFM. Simulation results on both tetragonal and rhombohedral ferroelectric ceramics using this model can well fit the existing experiments. Also, the domain texture evolution process can be explicitly captured and presented step by step.

2. An optimization-based computational model

2.1. Material model

In this model, a polycrystalline ferroelectrics is made up of numerous randomly oriented grains each of which contains N types of domains (where $N = 6$ for the tetragonal case and $N = 8$ for the rhombohedral case), the volume fraction of each type of domains is denoted by f_i ($i = 1, 2, \ldots, N$) and obviously we have $\sum_{i=1}^{N} f_i = 1$. The interactions between grains are considered in a self-consistent inclusion manner. That is, each grain is regarded as an inclusion surrounded by an infinite large matrix with the materials properties same as the whole material, as shown in Fig. 1.

Charge screening effect [14] in real ceramics is taken into account thus the depolarization electric field induced by polarization switching vanishes. The mechanical interactions between grains cannot be treated similarly and is considered in an elastic Eshelby inclusion manner [15]. Since the spherical inclusion case is the simplest and no evidence indicates that other shapes of inclusion are more accurate, we use the spherical inclusion case in this model. Furthermore, to grasp the major characteristics of domain switching and to make the computational complexity affordable, in the present model we also assume that a polycrystalline ferroelectric ceramic is dielectrically and elastically isotropic and shows linear dielectric and elastic behavior unless domain switching occurs (i.e., all nonlinear polarization and strain are caused by domain switching).

2.2. Free energy of a single grain

In the present model, as the interactions between grains have been considered in an Eshelby inclusion manner, we use the free energy of each grain as the optimization objective and sweep the optimization process over all grains to get the domain structures and properties of the whole material system. The free energy of a specific grain is expressed as

$$\begin{aligned} U(\mathbf{E},\boldsymbol{\sigma}) = &-\mathbf{E}\cdot\mathbf{P}^r - \boldsymbol{\sigma}:\boldsymbol{\varepsilon}^r + \frac{1}{2}k\mathbf{E}\cdot\mathbf{E} + \frac{1}{2}\frac{1}{2\mu}\boldsymbol{\sigma}:\boldsymbol{\sigma} - \frac{1}{2}\frac{v}{2\mu(1+v)} \\ &\times [tr(\boldsymbol{\sigma})]^2 + \frac{1}{2}\cdot 2\mu\frac{7-5v}{15(1-v)}(\boldsymbol{\varepsilon}^r - \bar{\boldsymbol{\varepsilon}}):(\boldsymbol{\varepsilon}^r - \bar{\boldsymbol{\varepsilon}}) \\ &+ f_{180}\cdot W_{180} + f_{non\text{-}180}\cdot W_{non\text{-}180} \end{aligned} \tag{1}$$

where $\mathbf{E}$, $\boldsymbol{\sigma}$ are the applied electric field vector and applied stress tensor, respectively. k is the isotropic dielectric constant, μ, v the isotropic shear moduli and Poisson ratio. $\bar{\boldsymbol{\varepsilon}}$ is the mean strain tensor of the whole material system or the matrix.

$\mathbf{P}^r$ and $\boldsymbol{\varepsilon}^r$ are the remanent polarization vector and remanent strain tensor of a grain which can be expressed as linear functions of the domain fractions

$$\mathbf{P}^r = \sum_{i=1}^{N} f_i\mathbf{P}^{(i)}, \quad \boldsymbol{\varepsilon}^r = \sum_{i=1}^{N} f_i\boldsymbol{\varepsilon}^{(i)} \tag{2}$$

Where $\mathbf{P}^{(i)}$ and $\boldsymbol{\varepsilon}^{(i)}$ are the spontaneous polarization vector and spontaneous strain tensor of the ith domain. The six types of domains in tetragonal ferroelectrics and eight types of domain in the rhombohderal are illustrated in Fig. 2.

For the tetragonal ferroelectric crystals, we have

$$\begin{aligned} &\mathbf{P}^{(1)} = -\mathbf{P}^{(2)} = P_0(0,0,1)^{\mathbf{T}}, \quad \mathbf{P}^{(3)} = -\mathbf{P}^{(4)} = P_0(1,0,0)^{\mathbf{T}}, \quad \mathbf{P}^{(5)} \\ &\quad = -\mathbf{P}^{(6)} = P_0(0,1,0)^{\mathbf{T}} \end{aligned} \tag{3}$$

$$\begin{aligned} &\boldsymbol{\varepsilon}^{(1)} = \boldsymbol{\varepsilon}^{(2)} = \frac{S_0}{3}\begin{bmatrix} -1 & & \\ & -1 & \\ & & 2 \end{bmatrix}, \quad \boldsymbol{\varepsilon}^{(3)} = \boldsymbol{\varepsilon}^{(4)} = \frac{S_0}{3}\begin{bmatrix} 2 & & \\ & -1 & \\ & & -1 \end{bmatrix}, \\ &\boldsymbol{\varepsilon}^{(5)} = \boldsymbol{\varepsilon}^{(6)} = \frac{S_0}{3}\begin{bmatrix} -1 & & \\ & 2 & \\ & & -1 \end{bmatrix} \end{aligned} \tag{4}$$

For rhombohedral crystals, we have

$$\begin{aligned} &\mathbf{P}^{(1)} = -\mathbf{P}^{(2)} = \frac{P_0}{\sqrt{3}}(1,1,1)^{\mathbf{T}}, \quad \mathbf{P}^{(3)} = -\mathbf{P}^{(4)} = \frac{P_0}{\sqrt{3}}(1,-1,1)^{\mathbf{T}}, \\ &\mathbf{P}^{(5)} = -\mathbf{P}^{(6)} = \frac{P_0}{\sqrt{3}}(-1,1,1)^{\mathbf{T}}, \quad \mathbf{P}^{(7)} = -\mathbf{P}^{(8)} = \frac{P_0}{\sqrt{3}}(1,1,-1)^{\mathbf{T}} \end{aligned} \tag{5}$$

and

$$\begin{aligned} &\boldsymbol{\varepsilon}^{(1)} = \boldsymbol{\varepsilon}^{(2)} = \frac{S_0}{3}\begin{bmatrix} 0 & 1 & 1 \\ & 0 & 1 \\ sym & & 0 \end{bmatrix}, \quad \boldsymbol{\varepsilon}^{(3)} = \boldsymbol{\varepsilon}^{(4)} = \frac{S_0}{3}\begin{bmatrix} 0 & -1 & 1 \\ & 0 & -1 \\ sym & & 0 \end{bmatrix} \\ &\boldsymbol{\varepsilon}^{(5)} = \boldsymbol{\varepsilon}^{(6)} = \frac{S_0}{3}\begin{bmatrix} 0 & -1 & -1 \\ & 0 & 1 \\ sym & & 0 \end{bmatrix}, \quad \boldsymbol{\varepsilon}^{(7)} = \boldsymbol{\varepsilon}^{(8)} = \frac{S_0}{3}\begin{bmatrix} 0 & 1 & -1 \\ & 0 & -1 \\ sym & & 0 \end{bmatrix} \end{aligned} \tag{6}$$

where P_0 is the spontaneous polarization; S_0 is the single crystal deformation [12] in ferroelectric crystals, for the tetragonal, $S_0 = S_{lattice} = c/a - 1$ (where c and a are tetragonal lattice constants), while for the rhombohedral, we have [12]

$$(8/9)S_0 = S_{lattice} = d_{[111]}/d_{[11\bar{1}]} - 1 \tag{7}$$

Returning to Eq. (1), W_{180} and $W_{non\text{-}180}$ are the energy barrier (per volume) for 180° and non-180° domain switching, respectively.

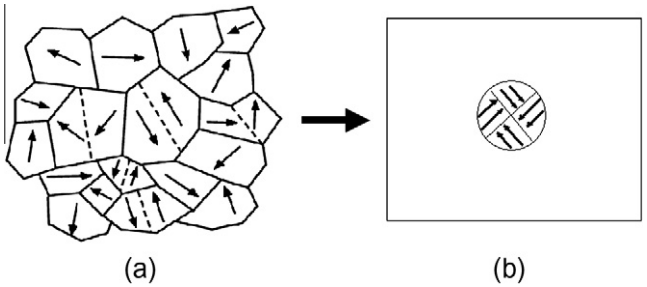

Fig. 1. 2-D illustration of material model for polycrystalline ferroelectrics.

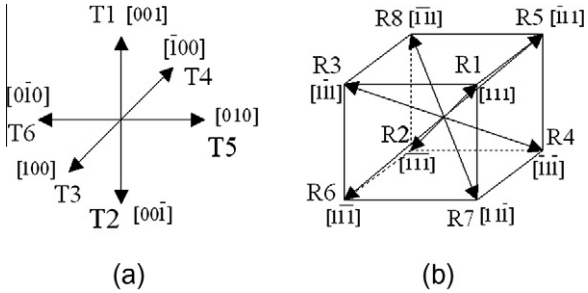

Fig. 2. Domains in (a) tetragonal and (b) rhombohderal ferroelectrics.