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Phase-field simulation of domain walls in rhombohedral ferroelectric single crystals

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ABSTRACT

Domain engineered ferroelectric relaxor-$PbTiO_3$ single crystals exhibit ultrahigh piezoelectric coefficients. A 10th order Landau-Devonshire energy function for rhombohedral PIN-PMN-PT ferroelectric single crystals was implemented in a phase-field model to study the behavior of domain walls under external electric field. The domain formation and domain wall evolution were simulated. A new way to apply periodic boundary conditions was implemented to accommodate nonzero strain during the domain formation and evolution. It was found that 71 and 109 domain walls reacted differently to external electric field along the [110] direction. A domain wall broadening effect was observed in the 71 domain walls when the electric field was below the coercive field. When the electric field exceeded the coercive field, homogeneous polarization switching occurred with no motion of the 71 domain walls. Heterogeneous polarization switching occurred by the sweeping of the 109 domain walls. The evolution mechanism of the two types of domain walls helps explain the behavior of engineered domain structures subjected to external electric field.

1. Introduction

Piezoelectric single crystals $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PMN-PT) exhibit ultrahigh piezoelectric coefficients and electromechanical coupling coefficients and thus are widely used as a substitute for conventional piezoelectric ceramics to improve the performance of sensors and actuators. The ternary compound $Pb(In_{1/2}Nb_{1/2})O_3$-$Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ (PIN-PMN-PT) [1] was developed to increase the phase transition temperature and Curie temperature of PMN-32PT without sacrificing the exceptional piezoelectric properties. These ferroelectric relaxor-$PbTiO_3$ single crystals are usually grown in composition ranges close to a morphotropic phase boundary (MPB) that results in rhombohedral symmetry. Then they are cut and poled along certain orientations to achieve enhanced piezoelectric properties, a process described as domain engineering [2–4].

The engineered domain structures are very important in understanding the material behavior and the mechanism of domain evolution under external excitations. The phase-field method is a powerful approach to modeling mesoscale microstructural evolution governed by the time-dependent Ginzburg-Landau (TDGL) equation. Chen et al. developed a semi-implicit Fourier-spectral method [5] to solve the TDGL equation and applied it to phase-field modeling of ferroelectric domain formation in three-dimensional space [6] and domain structures in ferroelectric thin films [7–10]. Using the same computational method, Wang et al. simulated ferroelectric and ferroelastic polarization switching [11] and studied the effect of mechanical strain [12] and defects (cracks and notches) [13–15]. Zhang and Bhattacharya modeled domain switching subjected to mechanical stress and cyclic electric field using a finite difference framework and attributed the large strain actuation to 90° domain switching [16]. A finite element implementation of a phase-field model was established by Schrade et al. [17] and Landis et al. [18]. In addition to single crystals, the phase-field method has been extended to polycrystals to look into the effect of grains [19–21]. Recently, the phase-field method was used to investigate the flexoelectric effect on the domain structures in nanoscale ferroelectrics [22–24].

Even though the phase-field method has proved to be a powerful tool for understanding the mechanism of microstructural evolution, most of the modeling work has addressed materials with a tetragonal structure such as $BaTiO_3$, $PbTiO_3$ and certain compositions of PZT with high Ti content. This is because the phase-field method requires input of a Landau-Devonshire

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energy function that describes the dielectric, piezoelectric and ferroelectric properties of a single crystal in the single domain state, but this type of energy function was not available for the new relaxor-$PbTiO_3$ single crystals. Recently Lv and Lynch developed a 10th order Landau-Devonshire energy function for rhombohedral PIN-PMN-PT ferroelectric single crystals [25,26]. This energy function was implemented in a phase-field model used in the work presented here. The results showed the effects of domain formation and evolution and their contributions to the material properties. Special attention was given to two types of domain walls that were observed in the rhombohedral crystals, the 71 domain wall and the 109 domain wall. The next section reviews the phase-field formulation and finite element implementation. This is followed by the simulations of 71 domain walls and 109 domain walls resulting in homogeneous and heterogeneous responses, respectively. Then the minimal domain wall motion in the engineered domain structures under the electric field is discussed based on the simulation results.

2. Phase-field method

The phase-field method uses a free energy density functional with order parameters that describe dielectric, piezoelectric, ferroelectric and elastic properties. Polarization and strain are usually chosen as the order parameters in ferroelectric materials. The total free energy density of the system includes the Landau-Devonshire energy, gradient energy, elastic energy and electrical energy, Eq. (1).

$$f = f_{\mathrm{L-D}} + f_{\mathrm{grad}} + f_{\mathrm{elas}} + f_{\mathrm{elec}} \tag{1}$$

Here the Landau-Devonshire energy is expressed as a 10th order polynomial that was developed for PIN-PMN-PT single crystals with rhombohedral variants [25,26]. This energy was originally expressed in terms of three polarization components P_1, P_2 and P_3 along the axes of the pseudo-cubic unit cell as shown in Fig. 1(a). The associated coordinate system is called the "cubic" coordinate system. Since the polarization component perpendicular to the diagonal plane (−110) remains zero throughout the switching process under the electric field along the [110] direction [26], the whole system can be reduced to two dimensions by using two in-plane components p_y and p_z as defined in Fig. 1(b). The associated coordinate system is referred to as the "rhombohedral" coordinate system. The plane (−110) contains four spontaneous polarization directions corresponding to energy minima in the Landau-Devonshire energy and each of them is called a rhombohedral variant. By poling along the [110] direction with electric field, an engineered domain structure is obtained with only two variants,

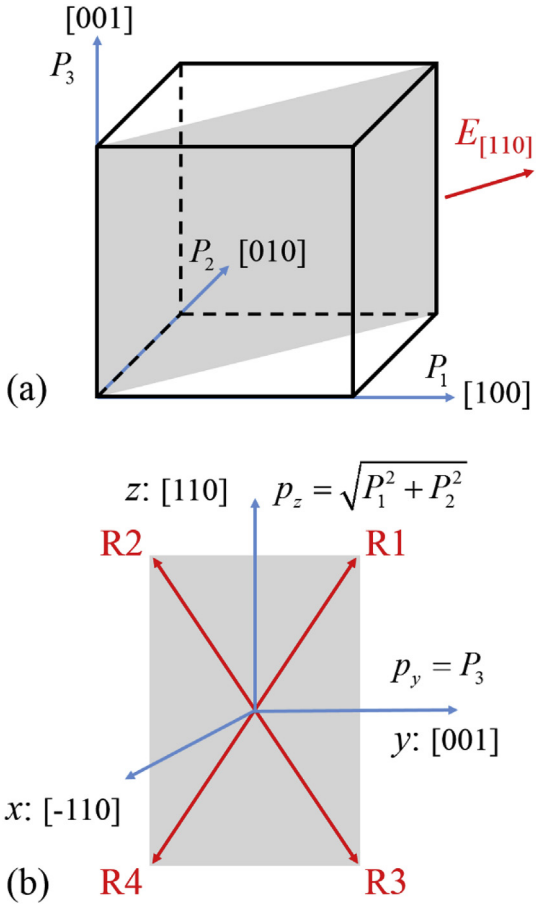

Fig. 1. (a) The "cubic" coordinate system used in Ref. [26]. The three axes are in the directions [100]-[010]-[001]. (b) The 2D "rhombohedral" coordinate system used in this work. The three axes are in the directions [-110]-[001]-[110]. The y-z plane is the diagonal plane (−110) in (a). It contains four rhombohedral variants, R1, R2, R3 and R4.

R1-R2 or R3-R4. This is called a two-rhombohedral-variant (2R) crystal. In this work, the engineered 2R domain structure is modeled in the 2D rhombohedral coordinate system in Fig. 1(b). All of the energy terms are expressed in this 2D system. Detailed derivations of the energies can be found in Su and Landis [18]. The 10th order Landau-Devonshire energy function with coefficients expressed in the rhombohedral coordinate system is given by Eq. (2).

$$\begin{aligned}
f_{\mathrm{L-D}} = {} & \alpha_1\left(p_y^2 + p_z^2\right) + \frac{1}{2}\alpha_{11}\left(2p_y^4 + p_z^4\right) + \frac{1}{4}\alpha_{12}\left(4p_y^2p_z^2 + p_z^4\right) + \frac{1}{4}\alpha_{111}\left(4p_y^6 + p_z^6\right) \\
& + \frac{1}{4}\alpha_{112}\left(4p_y^4p_z^2 + p_z^6 + 2p_y^2p_z^4\right) + \frac{1}{4}\alpha_{123}p_y^2p_z^4 + \frac{1}{8}\alpha_{1111}\left(8p_y^8 + p_z^8\right) \\
& + \frac{1}{8}\alpha_{1112}\left(2p_y^2p_z^6 + p_z^8 + 8p_y^6p_z^2\right) + \frac{1}{16}\alpha_{1122}\left(8p_y^4p_z^4 + p_z^8\right) \\
& + \frac{1}{4}\alpha_{1123}\left(p_y^4p_z^4 + p_y^2p_z^6\right) + \frac{1}{16}\alpha_{11112}\left(2p_y^2p_z^8 + p_z^{10} + 16p_y^8p_z^2\right) \\
& + \frac{1}{16}\alpha_{11223}\left(4p_y^4p_z^6 + p_y^2p_z^8\right) + \frac{1}{8}\alpha_{11123}\left(2p_y^6p_z^4 + p_y^2p_z^8\right)
\end{aligned} \tag{2}$$