A new mode of large electrostrictive actuation, based on 90° domain switching in ferroelectric crystals subjected to combined electromechanical loading, has recently been experimentally demonstrated. In this paper, we develop a model for this phenomenon by assuming a reasonable arrangement of domain walls and formulating equations of motion for these walls. The model captures most of the features observed in the experiments, reveals the significant role of friction at the interfaces between the loading frame and the crystal surfaces, and predicts that a reduction of friction will allow larger strains at lower mechanical loads.

Keywords: Ferroelectrics; Actuation; Electromechanical Loading; Friction

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1. Introduction

Ferroelectric perovskites are widely used for electro-mechanical actuation. Typically the actuation strain is small, and increasing it has been the subject of recent research (Park and Shrout, 1997; Liu and Lynch, 2003; Bhattacharya and Ravichandran, 2003). One such attempt, that of Burcsu et al. (2000; 2004), considered the 90° domain switching in single crystals of barium titanate, BaTiO$_3$. They subjected single crystals of BaTiO$_3$ to a constant dead load and cyclic electric field, and observed strains of almost equal to the c/a ratio due to 90 degree domain switching. While many of the observations followed the theoretical expectations (e.g., Shu and Bhattacharya, 2001; Tadmor et al., 2002 and Zhang and Bhattacharya, 2005), some puzzling aspects of the experiment remain open, and this motivates the work presented in this paper.

BaTiO$_3$ has a perovskite structure with cubic symmetry and no spontaneous electric polarization above the Curie temperature of 120°C. On cooling from above, it undergoes a phase transformation to a spontaneously electrically polarized tetragonal state. However, because of the reduction in symmetry, it can exist in six symmetry-related and energetically equivalent variants or states, which is shown in Fig. 1(a). In fact, a typical crystal contains numerous variants arranged in domains of individual variant with constant polarization and strain separated by domain walls. The strain and the polarization jump across the domain wall, but in a compatible manner. Consequently, the domain walls have a well-defined crystallographic orientation and the spontaneous polarization changes through a specific angle across the domain wall. In particular, a material that undergoes cubic to tetragonal transformation such as BaTiO$_3$ is limited to the so-called 180° and 90° domain boundaries, as shown in Fig. 1(b). When subjected to an electric field or mechanical stress, domain walls may move as one domain expands at the cost of the other or new domains may be nucleated. This process is collectively
known as domain switching. Domain switching leads to a change in the macroscopic polarization, and in the case of 90° switching also to a change in macroscopic strain. This strain associated with 90° domain switching can be quite large - up to 1.09% for BaTiO₃. Therefore, domain switching can form the basis of large strain actuation. We refer the reader to Damjanovic (1998) for a recent overview of ferroelectric materials.

Fig. 1. (a) The cubic (high temperature) to tetragonal (room temperature) transformation in BaTiO₃. (b) A typical specimen consists of the subgranular structure of domains, or regions of constant polarization, separated by 90° or 180° boundaries or domain walls.

The potential of the 90° domain switching was recently demonstrated by Burcsu et al. (2000; 2004), who obtained up to 0.9% strain in BaTiO₃ single crystals by subjecting them to constant
uniaxial compression load and cyclic electric field. Specifically, they took 5mm x 5mm x 1mm plates of single crystals BaTiO$_3$ with the pseudo-cubic axes oriented normal to the face of the crystal, placed them between glass platens which was subjected to a dead-load using a cantilever arrangement. Electrodes sandwiched between the crystal and the platens were used to apply a cyclic electric field. Their experimental set up allowed a clear optical path through the crystal, and polarized-light optical microscopy showed that this large strain was accompanied by 90° domain switching. At zero applied voltage, the applied stress forces the polarization to be in-plane (e.g., the [100] direction). As the voltage is increased, the electric field tries to align the polarization along the out-of-plane direction (i.e. the [001] direction), but this is resisted by the stress. There is an exchange of stability at a critical voltage and the polarization switches to [001] by the motion of 90° domain walls, which is accompanied by large strain. Finally, as the voltage is decreased, the polarization reverts back to in-plane, recovering the strain. Thus, cyclic actuation is obtained.

The experiments discussed above were motivated by the theoretical analysis of Shu and Bhattacharya (2001). This analysis was based on energy minimization and could only describe the equilibrium states as a function of stress and electric field, but not the transition between these states. Tadmor et al. (2002) developed a continuum model by coarse-graining an ab-initio descriptions of ferroelectrics, and used it to study these experiments above. However, their model had difficulties with the electrostatic fields and boundary conditions, and thus predicted the need for unrealistically large electric fields for switching. By reformulating the widely used time-dependent Devonshire-Ginzburg-Landau model (e.g., Hu and Chen, 1998) to real space, Zhang and Bhattacharya (2005) were able to apply realistic electric boundary conditions, and were able to capture domain switching and reproduce many features of the experiments.
However, they were unable to accurately describe the details of the experimental butterfly curves (strain as a function of electric field). In particular, they fail to explain why the obtained actuation strains (0.9%) are smaller than the theoretically predicted value of 1.09%.

A key deficiency of the models described above is that they do not account for the friction that is present between the glass platens and the ferroelectric crystal. As the domain walls move, the normal strain in the thickness direction is accompanied by lateral strain which causes the crystal to elongate or contract laterally. This lateral motion is resisted by the friction between the crystal and the glass platens. Addressing this deficiency is one of the main motivations for this current work. We show below, that friction indeed played a key role in the Burcsu et al. (2004) experiments, and can explain the reduced actuation strains.

A second motivation is to introduce a model for the motion of discrete domain walls. Instead of introducing a phase-field and resolving the details of the domain wall, we treat individual domain walls as defect-surfaces and track their evolution. This is very much in the spirit of sharp interface models of martensitic phase transformations (see, e.g., Abeyaratne and Knowles, 1993) or discrete dislocation dynamics (see, e.g., van der Giessen and Needleman, 1995; Tang et al., 1998) simulations that have been used in recent years.

We develop our model in a simple idealized setting. Based on the experimental observations, we identify a simple unit process consisting of two domain walls and two variants of the tetragonal materials. We make a further ansatz that the strains and polarization are piecewise constant. Therefore, the model reduces to a single ordinary differential equation describing the motion of the domain wall as a function of applied stress, applied electric field and the pair of variants. Since frictional forces can change directions, and since we have a number of choices of variant pairs, we get multiple branches and have to exercise care in distinguishing between them.
We develop our model in Section 2. We identify the potential energy and the dissipation energy associated with the experimental set up of Burcsu et al. Specifically we consider the system to consist of the ferroelectric crystal as well as the glass platens. We then specialize to the idealized unit process, and identify the driving force on the domain walls to be the force conjugate to the domain wall velocity in the dissipation. We postulate a simple kinetic law consisting of a critical driving force for the motion of the domain wall and linear kinetics beyond. Section 3 presents simulations based on this model, and show that it captures most of the features observed in the measured butterfly and polarization curves. Analysis of these simulations reveals the important role of friction at the interfaces between the loading platens and the crystal surfaces. It is predicted that a reduction of the friction coefficient will lead to a significant increase of the actuation strains at lower applied stresses. We conclude in Section 4 with a discussion.

2. Theoretical model

2.1 Potential Energy

Consider a ferroelectric in the shape of a flat plate with electrodes on two faces as shown in Fig. 2a subjected to a (dead loading) stress $\sigma$ and an electric field $E$. Following the analysis of Shu and Bhattacharya (2001), the potential energy of the system may be written as

$$U = \int_\Omega GdV$$

(1)

where $\Omega$ is the region occupied by the ferroelectric crystal,
\[ G = W(P, \varepsilon) - E \cdot P - \sigma \cdot \varepsilon \]  

(2)

is the total (Gibbs) energy density and \( W \) is the free energy density of the crystal. We treat the glass platens as rigid and non-polar, and thus they do not contribute to the potential energy. The energy density \( W \) depends on the strain \( \varepsilon \) and polarization \( P \), and has multiple energy wells corresponding to the crystallographically equivalent ground states of the crystal. At room temperature, BaTiO\(_3\) is spontaneously polarized in one of six equivalent \(<100>\) directions and tetragonally distorted. These six states are described by the following six combinations of spontaneous polarization vectors and strain tensors:

\[
\varepsilon^{(100)} = \begin{pmatrix}
\eta_1 & 0 & 0 \\
0 & -\eta_2 & 0 \\
0 & 0 & -\eta_2
\end{pmatrix}
\]

with \( P^{(100)}/(\overline{100}) = \pm P_0[1,0,0] \)

\[
\varepsilon^{(010)} = \begin{pmatrix}
-\eta_2 & 0 & 0 \\
0 & \eta_1 & 0 \\
0 & 0 & -\eta_2
\end{pmatrix}
\]

with \( P^{(010)}/(\overline{010}) = \pm P_0[0,1,0] \)

\[
\varepsilon^{(001)} = \begin{pmatrix}
-\eta_2 & 0 & 0 \\
0 & -\eta_2 & 0 \\
0 & 0 & \eta_1
\end{pmatrix}
\]

with \( P^{(001)}/(\overline{001}) = \pm P_0[0,0,1] \)  

(3)
Fig. 2. (a) A schematic representation of the experiments of Burcsu et al (2004). The ferroelectric crystal is loaded using two glass platens and subjected to an electric field as shown. The figure also shows the idealized domain wall arrangement. (b) A perspective view of the idealized domain wall arrangement and frictional force.

where $\eta_1 = 0.67\%$, $\eta_2 = 0.42\%$ and $P_0 = 0.26$ C/m$^2$ (Jona and Shirane, 1962). The strain tensors are defined with respect to the dimensions of the cubic unit cell. The energy density $W$ is minimized at these states.

In many ferroelectric crystals such as BaTiO$_3$ it turns out that the energy wells are fairly steep at the temperature of interest, i.e. room temperature. Consequently, in practice $P$ and $\varepsilon$ are located at the bottom of one of the wells, i.e. one of the six states described in Eq. (3). To do so, the crystal arranges itself in domains in which $P$ and $\varepsilon$ match one of the states described in Eq. (3). Different domains (having different state $P$ and $\varepsilon$) are separated by very narrow domain walls (Shilo et al., 2004; Franck et al., 2006) in which $P$ and $\varepsilon$ gradually change from one preferred state to another.
The energy density $W$ is the same for the six preferred states and hence the constraint that $P$ and $\varepsilon$ belong to one of these states cause $W$ to be constant. According to Eq. (2), an application of external electric field or mechanical stress causes a preference of some of the states upon the others. In the flat plate experiment described above the external electric field and stress tensor are described by

$$E = E_0 \begin{bmatrix} 0 \\ 0 \\ 1 \end{bmatrix}, \quad \sigma = -\sigma_0 \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 1 \end{bmatrix}. \quad (4)$$

Under these conditions, the (100)/(1000) and (010)/(0010) states are energetically equivalent and will be referred to collectively throughout this paper as the ‘(100) oriented’ or ‘in-plane polarized’ state. The (001) and (0010) states will likewise often be referred to collectively as the ‘out-of-plane polarized’ state though they need not be energetically equivalent. A substitution of Eqs. (3-4) into Eq. (2) shows that the total energy per unit volume of the in-plane polarized state is,

$$G^{(100)} = W_0 - \sigma_0 \eta_2. \quad (5)$$

Likewise, the total energy per unit volume of the out-of-plane polarized states is,

$$G^{(001)} = W_0 - E_0 P_0 + \sigma_0 \eta_1, \quad G^{(001T)} = W_0 + E_0 P_0 + \sigma_0 \eta_1. \quad (6)$$
It is clear that different values of applied electric field and stress favor different states. Therefore, one is able to switch from one state to another by an appropriate choice of electromechanical loading. In the experiments of Burcsu et al. (2004), the applied stress was held constant while the electric field was cycled. As the field is changed slowly, the switching between these different states occurs by the nucleation of new domains separated from the old domains by domain walls, and subsequently by the motion of domain the walls causing the expansion of one domain at the cost of the other.

2.2 Domain wall motion

A complete description of the entire process of domain switching is quite complicated and requires the solution of a coupled set of nonlinear field equations (see for example, Zhang and Bhattacharya (2005) and the references therein). In this work, we follow a different approach that builds on the experimental observations of Burcsu et al. (2004). In their experiments, the process was quite involved with different parts of the crystal transforming differently. Yet, polarized light microscopy revealed a unit process of the kind shown in Fig. 2 where one has domain walls separating in-plane and out-of-plane states go across the crystal. Therefore we make the ansatz that at any instant of time, one has two 90° domain walls located symmetrically in the crystal.

An important aspect of the experiments of Burcsu et al. (2004) was the presence of friction between the ferroelectric crystal and loading device. The uniaxial compressive stress was applied by sandwiching the crystal between two glass platens which were squeezed together as shown in Fig. 2(a). As the domain walls move, the normal strain in the thickness direction is accompanied by lateral strain which causes the crystal to elongate or contract laterally. This
lateral motion is resisted by the friction between the crystal and the glass platens. The frictional tractions on the surface are shown in Fig. 2(b).

We are now in a position to derive an equation of motion for the position $x$ of the domain wall. We do so in a thermodynamically consistent manner by calculating the rate of dissipation. Since we have dissipation on the surface of the crystal due to friction, it is convenient to consider our system of interest to be the crystal $\Omega$ and the glass platens $G'$ and $G^b$. The dissipation is the difference between the rate at which external work is done to the system and the rate of change of stored energy in the system:

$$D = \frac{\int \mathbf{n} \cdot \mathbf{v} dA + \int \mathbf{P} \cdot \mathbf{E} dV - \frac{d}{dt} \int \mathbf{W} dV}{\Omega}$$

(7)

where $v$ is the particle velocity. The first term is the work done by the external mechanical forces, the second term is the work done by the external electric field and the third term is the rate of change of stored energy. We treat the glass platens to be rigid and non-polarizable so that the second and third terms are limited to the crystal. We can now apply the divergence theorem to the first term taking care to account for the sliding crystal glass interfaces and use mechanical equilibrium to obtain,

$$\int_{\partial(\Omega \cup G' \cup G^b)} \oint \mathbf{n} \cdot \mathbf{v} dA = - \int_{\partial\Omega'} \mathbf{n} \cdot \left[ \mathbf{f} \right] dA + \int_{\Omega} \mathbf{G} \cdot \mathbf{\varepsilon} dV$$

(8)

where $\partial\Omega'$ and $\partial\Omega^b$ denote the top and bottom surfaces of the crystal. Thus,
\[ D = -\int_{\partial \Omega' \cup \partial \Omega^b} \sigma \cdot \mathbf{n} \cdot \mathbf{h} \, dA + \int_{\Omega} \sigma \cdot \dot{\varepsilon} \, dV + \int_{\Omega} \dot{P} \cdot \mathbf{E} \, dV - \frac{d}{dt} \int_{\Omega} \mathbf{F} \, dV \]

\[
= -\frac{dU}{dt} - \int_{\partial \Omega' \cup \partial \Omega^b} \sigma \cdot \mathbf{n} \cdot \mathbf{h} \, dA
\]

\[
= -\frac{dU}{dx} \dot{x} - \int_{\partial \Omega' \cup \partial \Omega^b} \sigma \cdot \mathbf{n} \cdot \mathbf{h} \, dA
\]

(9)

Specializing now to the geometry shown in Fig. 2, we find that the jump in velocity across the crystal-glass interface is given by \( \varepsilon_0 \dot{x} \) and the traction along the direction of the velocity jump is \( -\text{sgn}(\dot{x}) \mu \sigma_0 \) and the area of each surface is \( wx \). \( \varepsilon_0 = \eta_1 + \eta_2 \) is the transformation strain (\( \varepsilon_0 = 1.09\% \) for BaTiO\(_3\)), \( \mu \) the coefficient of friction and \( w \) the width of the crystal. Therefore,

\[ D = F \dot{x} = \left( F_{\text{switch}} + F_{\text{fric}} \right) \dot{x} \]

(10)

where

\[
F_{\text{switch}} = -\frac{dU}{dx} = -\Delta G \, wh = \begin{cases} 
(\sigma_0 \varepsilon_0 - EP_0)wh & \text{for (001) to (100) switching} \\
(\sigma_0 \varepsilon_0 + EP_0)wh & \text{for (00\bar{1}) to (100) switching} \\
(-\sigma_0 \varepsilon_0 + EP_0)wh & \text{for (100) to (001) switching} \\
(-\sigma_0 \varepsilon_0 - EP_0)wh & \text{for (100) to (00\bar{1}) switching} 
\end{cases}
\]

(11)

with \( h \) the crystal thickness, and

\[ F_{\text{fric}} = 2 \text{sgn}(\dot{x}) \mu \sigma_0 \varepsilon_0 \, A = 2 \text{sgn}(\dot{x}) \mu \sigma_0 \varepsilon_0 \, wx \].

(12)
We can now identify the net driving force on interface to be the force conjugate to the domain wall velocity \( \dot{x} \). It is:

\[
F = F_{\text{switch}} + F_{\text{fric}}. \tag{13}
\]

We postulate that the velocity of the interface depends on this driving force. Specifically, we assume the following.

\[
\dot{x} = \begin{cases} 
\frac{F + F_c}{Bwh} & \text{if } F < -F \\
0 & \text{if } -F_c \leq F \leq F_c \\
\frac{F - F_c}{Bwh} & \text{if } F > F_c 
\end{cases} \tag{14}
\]

where \( F_c \) is the critical force for interface motion and \( B \) may be viewed as a viscosity coefficient per unit area of the domain wall. This provides a first order differential equation that can be solved analytically for each value of applied stress and voltage.

We now assume that the applied electric field to be cyclic,

\[
E_0 = E_0 \sin \omega t \tag{15}
\]

with a slight abuse of notation. If the cycling frequency is small, we can continue to use the equations derived above. Due to the multiple switching processes appearing in Eq. (11) and the condition appearing in Eq. (14), a careful determination of the process that occur in each time region during a complete cycle of the electric field has to be done.
Table 1 and Fig. 3 show a division of a half time period ($\pi/2 \leq \omega t \leq 3\pi/2$) into four regions in which different switching processes occur. Since it is imperative to start the simulation at a well defined polarization state, we chose starting at $\omega t = \pi/2$ (i.e. $E = E_0$). At this point $EP_0 \gg \sigma_0\varepsilon_0$ and hence we can assume that the polarization is completely along the (001) state. In region 1, the preferred state is (001) but the crystal is already completely polarized along this state and hence there is no change in the strain or polarization. In region 2, a switching from the (001) state to the (100) state occurs when $\sigma_0\varepsilon_0 - EP_0 > F_c$. A delicate point is the determination what happens when $E$ becomes negative. Here, two possible routes can be considered as described in Fig. 3. In the first, which is illustrated by the broken line in Fig. 3 (not appearing in Table 1), the switching from the (001) state to the (100) state continues until it completes and at some point another $90^\circ$ switching process starts, from the (100) state to the (00$\bar{1}$) state. Since this route occurs only by $90^\circ$ switching processes, at any time during this route a change of the polarization is accompanied by a change of the strain. In the second route, which is illustrated by a continuous line, at some point when $E < -E_{c^{180}}$, the remaining (001) domain switches to (00$\bar{1}$). This $180^\circ$ switching process is characterized by a fast change of the polarization with no strain change, as illustrated in Fig. 3. As is shown later, the measured curves of the actuation strain as a function of the polarization show a significant range in which a change of the polarization occurs without a change of the strain. This observation clearly indicates that the second route is the route that takes place in our case. This route is described in Table 1 and Fig. 3 by region 3. The rate of the $180^\circ$ domain switching is unknown and we simulate it as happening instantaneously.
Table 1. A division of the time interval $\pi/2 \leq \omega t \leq 3\pi/2$ into four regions in which different switching processes occur in accordance to Fig. 3.

<table>
<thead>
<tr>
<th>Ending time ($\omega t$)</th>
<th>Region 1</th>
<th>Region 2</th>
<th>Region 3</th>
<th>Region 4</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\pi - \arcsin \left( \frac{\sigma_0 \epsilon_0}{E_0 P_0} - \frac{E_c^{90}}{E_0} \right)$</td>
<td>$\pi + \arcsin \left( \frac{E_c^{180}}{E_0} \right)$</td>
<td>$\pi + \arcsin \left( \frac{\sigma_0 \epsilon_0}{E_0 P_0} + \frac{E_c^{90}}{E_0} \right)$</td>
<td>$\frac{3\pi}{2}$</td>
<td></td>
</tr>
<tr>
<td>Preferred state</td>
<td>(001)</td>
<td>(100)</td>
<td>(00 $\bar{T}$)</td>
<td>(00 $\bar{T}$)</td>
</tr>
<tr>
<td>Process</td>
<td>No switching</td>
<td>(001) $\rightarrow$ (100)</td>
<td>(001) $\rightarrow$ (00 $\bar{T}$)</td>
<td>(100) $\rightarrow$ (00 $\bar{T}$)</td>
</tr>
<tr>
<td>Strain</td>
<td>$\epsilon_0$</td>
<td>$\epsilon_0 (1 - 2x/L)$</td>
<td>No change</td>
<td>Strain at the end of the previous stage plus $\epsilon_0 (2x/L)$</td>
</tr>
<tr>
<td>Polarization</td>
<td>$P_0$</td>
<td>$P_0 (1 - 2x/L)$</td>
<td>The remaining polarization switch sign instantaneously</td>
<td>Polarization at the end of the previous stage minus $P_0 (2x/L)$</td>
</tr>
</tbody>
</table>

Fig. 3. Illustration of the strain ($\epsilon$) and polarization ($P$) as a function of the electric field during the time interval $\pi/2 \leq \omega t \leq 3\pi/2$. Four time regions are indicated in accordance to different switching processes as described in Table 1. The broken curves illustrate the response which would have happened in the absence of the 180° domain switching that occurs in region 3.
The model described in this section includes six material parameters. Amongst them, $P_0$ and $\varepsilon_0$ have been measured in many experiments (e.g., Jona and Shirane, 1962) and are known with good accuracy. The critical fields, $E_{c\theta}^{90}$ and $E_{c\theta}^{180}$, can be evaluated based on our experiments and on previous experiments (Li et al., 1992) to be in the range of $0.2 - 0.5$ kV/cm, where typically $E_{c\theta}^{90} > E_{c\theta}^{180}$. The friction coefficient between the two platinum thin films (one deposited on the crystal and the other on the glass plate) can be evaluated to be in the range of $0.15 < \mu < 0.7$, which is typical to a dry contact between two metal surfaces (Meriam and Kraig, 2002). If grease would have been applied between the surfaces, a value of $\mu = 0.05$ may be considered (Meriam and Kraig, 2002). The only parameter which is completely unknown and may have values within a range of several orders of magnitude is the viscosity coefficient $B$.

3. Simulations and analysis

Simulations for the strain and polarization as a function of time were obtained by solving Eqs. (11)-(14) interpreted appropriately as mentioned in Table 1. For example, in region 2 the following linear first order equation was obtained,

$$
\dot{x} + \omega' x = \frac{\sigma_0 \varepsilon_0 - E_{c\theta}^{90} P_0}{B} - \frac{E_0 P_0}{B} \sin \omega t
$$

(16)

where

$$
\omega' = \frac{2\mu \sigma_0 \varepsilon_0}{B h}.
$$

(17)
Similar equation appears in region 4 with only some sign differences. Equation (16) demonstrates that the frequency independence which was observed by Burcsu et al. (2004) means that in the range of frequencies applied in the experiments (i.e. up to 1 Hz) $\omega << \omega'$. This conclusion together with Eq. (17) means that $B << 4 \times 10^6 \text{ Ns/m}^3$. Further experiments at higher frequencies can provide more precise evaluation of $B$.

Figures 4 and 5 show simulated and measured curves of the strain as a function of electric field at the same five values of compressive stress. The material parameters substituted in all simulations are $P_0 = 0.26 \text{ C/m}^2$, $\varepsilon_0 = 1.09\%$, $E_{c90} = 0.3 \text{ kV/cm}$, $E_{c180} = 0.2 \text{ kV/cm}$, $\mu = 0.24$, and $\omega'/\omega = 50$. To avoid confusion we used two definitions for the strain. In the left axis the reference for the strain is chosen such that the minimal strain is zero (we term this strain as ‘actuation strain’) in accordance with Burcsu et al. (2004). In the right axis the strain is defined in accordance with the crystallographically ground states, such that $\varepsilon = 0$ relates to a complete (100) orientation and $\varepsilon = \varepsilon_0 = 1.09\%$ relates to a complete (001) or (00\overline{1}) orientation.

Comparison of Fig. 4 to Fig. 5 shows that the simulated curves capture most of the features observed in the measured curves. Actually, the simulated and measured curves exhibit good agreement in the lower 2/3 of the butterfly curves, but in the upper 1/3 of the curve the measured strains increase more slowly than predicted by the simulations. Moreover, when the amplitude of the electric field decreases, the measured strain decreases even in the region where the out-of-plane polarized states are still preferred (e.g., region 1 in Table 1 and Fig. 3). This indicates the existence of another mechanism, which was not taken into account by the theoretical model. This mechanism not only resists the switching from the (100) states to the (001) or (00\overline{1}) states when the electric field increases, but also promotes the switching back from the (001) or (00\overline{1}) states to the (100) states when the electric field decreases. One possible explanation for
such behavior is a mechanism recently suggested by Ren (2004), in which a general symmetry conforming property of point defects provides a restoring force toward a preferred polarization state (the (100) state in our case).

![Simulations of the strain as a function of electric field for five values of compressive stress.](image)

Fig. 4. Simulations of the strain as a function of electric field for five values of compressive stress.
Fig. 5. Experimental observations of strain vs. electric field for five values of compressive stress. Reprinted from Burcsu et al. (2004) with permission from Elsevier.

Figures 6 and 8 show the polarization vs. electric field and the strain vs. polarization trajectories which corresponds to the simulations of Fig. 4 (the same material parameters were used). The corresponding experimental observations are shown in Figures 7 and 9 respectively. Comparison of experimental and simulated curves shows a satisfactory agreement. In fact, the main disagreement in the polarization vs. electric field curves arises from the fact that we simulate the 180° domain switching (region 3 in table 1 and Fig. 3) to be happening
instantaneously. In practice, this process takes some time and therefore the measured curves don’t exhibit an abrupt change of the polarization as observed in the simulations.

Fig. 6. Simulations of the polarization as a function of electric field for five values of compressive stress.
Fig. 7. Experimental observations of polarization vs. electric field for five values of compressive stress. Reprinted from Burcsu et al. (2004) with permission from Elsevier.
Fig. 8. Simulations of the strain as a function of polarization for five values of compressive stress.
4. Discussion

The results of all experiments, taken from three crystals (one having an in-plane initial orientation and two having an out-of-plane initial orientation), are collected in Fig. 10 together with their corresponding simulation results. This shows the amplitude of the actuation strain as a function of the applied compressive stress. The experimental values are calculated in the fifth cycle of each experiment after the response has reached a steady state. Two values are collected.
for each experiment: one for increasing electric field and one for the decreasing electric field. The qualitative behavior is similar for all three crystals, with little dependence on initial orientation. The actuation strain is negligible for zero stress, rises steadily with increasing stress, reaches a peak and then begins to decrease. The maximum recorded value of actuation strain amplitude is 0.9%. The simulation results exhibit good agreement with the experiments up to a stress of about 2 MPa. Beyond this stress the experimental results exhibit a decrease of the actuation strain while the simulations exhibit an asymptotic flattening toward a maximal actuation strain of about 0.9%. The decrease of actuation strain that appear in the experiments beyond 2 MPa is probably related to the mentioned mechanism that resists the switching from the (100) states to the (001) or (00 1) states at high electric fields.

An interesting result is that the simulations predict that even at the absence of this mechanism the actuation strains are restricted to a value of less than 0.9%. This restriction appears due to the friction at the interfaces between the loading frame and the crystal surfaces. One of the benefits of having a model that simulates a set of experiments is the ability of applying the simulations on conditions which were not studied by the experiments. An interesting condition is that of a significantly smaller friction coefficient, as would have been if grease was applied between the glass plates and the crystal surfaces. Figure 11 shows simulated curves of the strain as a function of electric field calculated using the same material parameters as in Fig. 4 except for the friction coefficient which was changed to $\mu = 0.05$. This set of simulations predicts that a reduction of the friction coefficient will lead to a significant increase of the actuation strains. Moreover, a smaller friction coefficient will allow reverting to the in-plane polarization by an application of a smaller stress. This factor is especially important as the
stress and the friction forces were observed to cause a mechanical damage in a form of cracks (Buresu et al., 2004).

Fig. 10. Amplitude of actuation strain vs. compressive stress for three different BaTiO₃ crystals and the corresponding simulation results.
Fig. 11. Simulations of the strain as a function of electric field for three values of compressive stress, calculated using the same material parameters as in Fig. 4 except for the friction coefficient which was changed to $\mu = 0.05$.

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Bhatta


