Electro-mechanical modeling of a ferroelastic relaxor single crystal

Mourad ELHADROUZ, Tarak BEN ZINEB and Etienne PATOOK
Laboratoire de Physique et Mécanique des Matériaux UMR CNRS 7554
Ecole Nationale Supérieure d'Arts et Métiers, Technopôle 2000, 4 rue Augustin Fresnel 57078 Metz Cedex, France.
TEL : +33 3 87 37 54 30 e-mail: mourad.elhadrouz@metz.ensam.fr
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This paper examines the switching process occurring in ferroelectric and ferroelastic relaxor single crystals under electro-mechanical loadings. Relaxors undergoing a cubic to tetragonal phase transition are considered. The single crystal energy has three origins: elastic, electric and the incompatibilities of the spontaneous strain and electric displacement fields between domains. The stress and the electric field fluctuate and present jumps at the domain walls. As a consequence, they induce electro-elastic interaction energy. Thus, it involves dissipation that the present work aims to capture through an electro-micromechanical approach.

Keywords : Ferroelectrics, Ferroelastic, Single crystals, Switching.

I. INTRODUCTION

Relaxors are widely used in a variety of applications including modulators, deflectors, optical memories imaging devices, non-volatile data storage elements, etc. Their applications in actuators and sensors in smart materials and structures have achieved much success. Ferroelastic relaxor crystals undergo a phase transition when cooling through the Curie point T_C (Fig. 1). Above it, the unit cell is cubic and it can be transformed into a tetragonal, orthorhombic or rhombohedral one. It results in a spontaneous polarisation and a lattice distortion. Then, a crystal is said to be ferroelectric when the spontaneous polarisation can be switched by an electric field. In the same manner, it is said ferroelastic when it can be switched by a mechanical stress. To minimise the energy associated with the spontaneous polarisation and strain, a crystal is divided into many uniformly polarised regions called domains that are separated by domain walls. Irreversible domain wall motions as well as nucleation and growth of domains with new orientations cause strong non-linearity in the macroscopic behavior.

This paper studies the non-linear macroscopic behavior of ferroelastic relaxor crystals under electric or mechanical loadings and it proposes a electro-micromechanical approach to capture it. Only PZT relaxor single crystals with 90° and 180° domains are considered; it means that the phase transition is a cubic to a tetragonal one. Some additional transitions may occur and lead to orthorhombic or rhombohedral configurations. It is an important assumption since it has consequences upon the kinematic variables that will be introduced later.

II. FREE ENERGY RELATED TO DOMAIN SWITCHING IN A SINGLE CRYSTAL

In this section, we consider a single crystal with a volume V and a boundary δV. It is composed of uniformly polarised regions or domains. Upon the cubic to tetragonal phase transition, the unit cell can take any of the six equivalent combinations of strain
and polarization (Fig.2). Both spontaneous strain and polarization are uniform in a given domain. Through an electric field or a mechanical stress, the direction of the spontaneous polarization can switch.

![PbTiO3 crystal structure](image)

Fig.1. Phase transition from a cubic unit cell to a tetragonal one when PZT is cooled through the Curie point.

![Unit cell](image)

Fig.2. The unit cell can take any of the six equivalent combinations of strain and polarization. The arrow indicates the direction of the polarization.

The microscopic constitutive laws in a domain \( n \) are with the Einstein notation, and summation implied over repeated indices:\(^5\)

\[
\begin{align*}
\varepsilon_{ij} &= S_{ijkl} \sigma_{kl} + d_{ij} E_k + \delta_{ij} = \varepsilon_{ij}^s + \varepsilon_{ij}^e \\
D_i &= d_{ik} \sigma_{kl} + \kappa_i E_k + P_i^s = P_i^{se} + P_i^e
\end{align*}
\]  

(1)  

(2)

where \( \varepsilon_{ij}, \sigma_{kl}, E_k \) and \( D_k \) are respectively the components of the total strain tensor, the local stress tensor, the electric field, the total polarization and the electric displacement vectors. \( S_{ijkl}, d_{ijkl} \) and \( \kappa_i \) are the components of the elastic compliance, the piezoelectric and dielectric permittivity tensors. We assume that the elastic compliance tensor and dielectric permittivity tensors possess isotropic symmetry with

\[
S_{ijkl} = \frac{1 + \nu}{E} \delta_{ik} \delta_{jl} - \frac{\nu}{E} \delta_{ij} \delta_{kl} \\
\kappa_{ij} = \kappa_{ji} \delta_{ij}
\]

(3)  

(4)

where \( \delta_{ij} \) is the Kronecker symbol, \( \kappa_{ji} \) is the dielectric permittivity, \( \nu \) and \( E \) are respectively Young's modulus and Poisson's ratio. The piezoelectric tensor is assumed to be transversely isotropic and has a principal direction that aligns with the direction of the polarization denoted \( e^p \). It is given by the expression

\[
d_{imn} = d_{33} e^p m e^p n + d_{15} \left( \delta_{in} - e^p_i e^p_n \right) e^p m + d_{15} \left( \delta_{mn} - e^p_m e^p_n \right) e^p i
\]

(5)

where \( d_{33}, d_{15} \) and \( d_{15} \) are the longitudinal, transverse and shear piezoelectric constants.\(^6\) In addition, the strain and the electric displacement are decomposed into a linear piezoelectric part with the superscript \( ^{se} \) and the spontaneous quantity with the superscript \( ^s \) with

\[
\varepsilon_{ij}^s = \frac{1}{2} \varepsilon^{se} \left( e^p_i e^p_j - \frac{1}{3} \delta_{ij} \right) \\
P_i^s = P^{se} e^p_i
\]

(6)  

(7)

Neglecting heat effects, the total differential of the internal energy is given by 'Ref.7.'

\[
dU = TdS + E_i dD_i^{se} + \sigma_{ij} d\varepsilon_{ij}^{se}
\]

(8)

where \( S \) and \( T \) denote respectively the entropy and temperature. The second term \( E_i dD_i^{se} \) is the work against the electric field and the third one \( \sigma_{ij} d\varepsilon_{ij}^{se} \) is the work against the mechanical stress. They are associated to the local stress and the electric field created by the loading conditions on \( \delta V \) and by the incompatibilities of the spontaneous strain and the electric displacement in each domain. We can change
the variables of the internal energy through a Legendre transformation. Then we introduce the Gibbs free energy \( \psi \) defined by

\[
\psi = U - TS - E_i D_i^e + \sigma_y \varepsilon_y^e
\]  
(9)

Thus, an integration by part of the differential form of (9) combined with (1), (2) and (8) provide the expression of the Gibbs free energy

\[
\psi = -\frac{1}{2} \sigma_y (\varepsilon_y - \varepsilon_y^t) - \frac{1}{2} E_i (D_i - P_i)
\]  
(10)

The global energy of the crystal is obtained by integrating over the volume the relation (10)

\[
\Psi = \frac{1}{V} \int_V \psi dV
\]  
(11)

It may be transformed using kinematical conditions inside \( V \)

\[
\varepsilon_y (r) = u_{i,j} = \varepsilon_y^e (r) + \varepsilon_y^t (r)
\]  
(12)

\[
E_i (r) = -\varphi_i (r)
\]  
(13)

where \( u \) and \( \varphi \) are respectively the mechanical displacement and the electric potential. In addition, homogeneous mechanical and electrical boundary conditions on \( \partial V \) have to be introduced

\[
u_i = E_y x_j
\]  
(14)

\[
\varphi = -E_i^0 x_i
\]  
(15)

where \( E_y \) and \( E_i^0 \) are the macroscopic strain and the electric field applied on \( \partial V \). From (12) and (13), it comes that

\[
\Psi = -\frac{1}{2V} \int_V \sigma_y (u_{i,j} - \varepsilon_y^t) - \varphi_i (D_i - P_i) dV
\]  
(16)

By partial integration and taking into account the boundary conditions (14) and (15), the mechanical and electrical equilibrium conditions

\[
\sigma_{y,j} = 0
\]  
(17)

\[D_{ij} = 0\]  
(18)

(10) is reduced to

\[
\Psi = -\frac{1}{2} \Sigma_y E_y - \frac{1}{2} E_i^0 \overline{D_i}
\]

\[-\frac{1}{2V} \int_V \sigma_y \varepsilon_y^t - \varphi_i P_i dV
\]  
(19)

with

\[
\Sigma_y = \frac{1}{V} \int_V \sigma_y dV
\]  
(20)

\[
\overline{D_i} = \frac{1}{V} \int_V D_i dV
\]  
(21)

that represent respectively the overall stress and the overall electric displacement. We decompose the stress \( \sigma \) into the macroscopic \( \Sigma \) and the internal stress \( \tau \) (only related to the \( \varepsilon^t \) field)

\[
\sigma_y (r) = \Sigma_y + \tau_y (r)
\]  
(22)

so that

\[
\tau_{y,i} (r) = 0
\]  
(23)

\[
\frac{1}{V} \int_V \tau_y (r) dV = 0
\]  
(24)

Similarly, the electric field \( E \) is decomposed into the macroscopic electric field \( E^0 \) and the internal electric field \( E^{in} \) with

\[
\frac{1}{V} \int_V E^{in} (r) dV = 0
\]  
(25)

Using uniform elasticity and dielectricity, (19) is written

\[
\Psi = -\frac{1}{2} \Sigma_y (E_y - \varepsilon_y) - \frac{1}{2} E_i^0 (D_i - P_i)
\]

\[+ \frac{1}{2V} \int_V \tau_y \varepsilon_y^t dV + \frac{1}{2V} \int_V E^{in} P_i dV
\]  
(26)
with

$$
\overline{\varepsilon}_y = \frac{1}{V} \int \varepsilon_y^d dV = \sum_n f_n \varepsilon_y^{m-n} \\
\overline{P}_i = \frac{1}{V} \int P_i^d dV = \sum_n f_n P_i^{m-n}
$$

(27)

(28)

where $f_n$ is the volume fraction of the domain $n$ for which the polarisation has the direction $n$ among the six possible directions previously defined and $\varepsilon_{sn}$ and $P_{sn}$ are the corresponding spontaneous polarisation and strain. The Gibbs free energy has three origins: elastic, electric and the two last terms in (26) represents the interaction energy between domains since strain or polarisation are different from a domain to the neighboring ones. The sources of internal stresses and internal electric fields are located on the boundaries between domains or domain walls. As a consequence, the stress field and the electric field fluctuate, weakly inside the domains and present jumps across the domain wall. Therefore, it may be evaluated using interface operator techniques as it was previously made in the case of shape memory alloys. As a consequence, we assume that the interaction energy can be written

$$
W_{int} = -\frac{1}{2} \sum_n f_n H_{nm} f_m
$$

(29)

Depending on the loading, the volume fractions of these six variants may change. $H_{nm}$ is an interaction matrix analogous to the one developed by Sired by et al. that accounts for the interaction between different domains. Then (26) becomes

$$
\Psi = -\frac{1}{2} \sum_n \left( \varepsilon_y - \overline{\varepsilon}_y \right) - \frac{1}{2} \varepsilon_y^0 \left( \overline{D}_i - \overline{P}_i \right) - \frac{1}{2} \sum_n f_n H_{nm} f_m
$$

(30)

The driving force on the volume fraction $f_n$ is defined and given by

$$
F_n = \frac{\partial \Psi}{\partial f_n} = -\frac{1}{2} \sum_n \varepsilon_y^{m-n} + \frac{1}{2} E_i^0 P_i^{m-n} - \sum_m H_{nm} f_m
$$

(31)

For time independent behavior, the switching may progress on a current variant $n$ only if the driving force reaches a critical value $F_n^c$ that is assumed to be constant

$$
\frac{1}{2} \sum_n \varepsilon_y^{m-n} + \frac{1}{2} E_i^0 P_i^{m-n} - \sum_m H_{nm} f_m = F_n^c
$$

(32)

It is justified by the dissipation caused by switching. Once it is satisfied, the coherency condition

$$
\dot{F}_n = \dot{F}_n^c = \frac{\partial F_n}{\partial \varepsilon_y} \dot{\varepsilon}_y + \frac{\partial F_n}{\partial E_i} \dot{E}_i + \frac{\partial F_n}{\partial f_m} \dot{f}_m = 0
$$

(33)

provides the evolution of the switching rates for each variant as a function of the rate of the imposed control variables. In this case, (33) reduces to

$$
\frac{1}{2} \dot{\varepsilon}_y \varepsilon^{m-n} + \frac{1}{2} \dot{E}_i E_i^{m-n} = \sum_m H_{nm} \dot{f}_m
$$

(34)

The electro-mechanical constitutive equation for the single crystal is defined by the relations between the overall stress rate, the electric displacement rate, the strain rate and the electric field rate. From the piezoelectric constitutive equations, one gets

$$
\dot{\varepsilon}_y = C_{ijkl} \left( \dot{E}_{kl} - \overline{\varepsilon}_y^{k-l} \right) - \Gamma_{p_j} \dot{E}_p^0 - \dot{E}_p^0
$$

$$
\dot{D}_i = \Gamma_{kli} \left( \dot{E}_{kli} - \overline{E}_y^{k-l} \right) + \Gamma_{kli} \left( \dot{E}_{kli} - \overline{E}_y^{k-l} \right) \dot{E}_p^0
$$

(35)

(36)

where $C_{ijkl}$ and $\Gamma_{kli}$ are respectively the components of the overall elastic and the converse piezoelectric tensors. The overall converse piezoelectric tensor is linked to the overall piezoelectric tensor by the relation

$$
\Gamma_{p_j} = C_{ijkl} \tilde{d}_{ikl} = C_{ijkl} \sum_n f_n d_{ikl}^n
$$

(37)
where \( d_{\rho\delta}^n \) is the piezoelectric tensor for the domain \( n \). A Voigt approximation is adopted for the expression of the overall piezoelectric tensor. Then (35) becomes

\[
\dot{\Sigma}_y = C_{ijkl} \left( \dot{E}_{kl} - \sum_n \dot{j}_n \epsilon_{il}^0 \right) - \Gamma_{ij} \dot{E}_{ij}^0 - C_{ijkl} \sum_n \dot{j}_n d_{\rho\delta}^n E_{\rho}^0 \quad (38)
\]

Multiplying (38) by \((\epsilon_{ij}^{sm}/2)\) yield

\[
\dot{j}_n = \frac{1}{2} \sum_{m} K_{\rho\delta}^{nm} \epsilon_{ij}^{sm} C_{ijkl} \dot{E}_{kl} + \frac{1}{2} \sum_{m} K_{\rho\delta}^{nm} \left( P_{ij}^{sm} - \epsilon_{ij}^{sm} \Gamma_{ij} \right) \dot{E}_{\rho}^0 \quad (39)
\]

where

\[
K_{\rho\delta}^{nm} = \left( H_{\rho\delta}^{nm} + \frac{1}{2} \epsilon_{ij}^{sm} C_{ijkl} \left( \epsilon_{ij}^{sm} + d_{\rho\delta}^n E_{\rho}^0 \right) \right)^{-1} \quad (40)
\]

Summations are made only on the potentially active variants, i.e., those satisfying the switching conditions (32) and (33). Introducing (39) into (35) and (36) yields

\[
\dot{\Sigma}_y = L_{ijpq} \dot{E}_{pq} + M_{ijl} \dot{E}_{ij}^0 \quad (41)
\]

\[
\dot{D}_l = N_{ij} \dot{E}_{ij}^0 + Q_{ij} \dot{E}_{ij}^0 \quad (42)
\]

with

\[
L_{ijpq} = C_{ijpq} - \frac{1}{2} \sum_{m} K_{\rho\delta}^{nm} \epsilon_{ij}^{sm} C_{ijkl} \left( \epsilon_{ij}^{sm} + d_{\rho\delta}^n E_{\rho}^0 \right) \quad (43)
\]

\[
M_{ijl} = -\Gamma_{ijl}
\]

\[
-\frac{1}{2} \sum_{n} \sum_{m} K_{\rho\delta}^{nm} C_{ijkl} \left( P_{ij}^{sm} - \epsilon_{ij}^{sm} \Gamma_{ij} \right) \left( \epsilon_{ij}^{sm} + d_{\rho\delta}^n E_{\rho}^0 \right) \quad (44)
\]

\[
N_{ij} = \Gamma_{ij}
\]

\[
+ \frac{1}{2} \sum_{n} \sum_{m} K_{\rho\delta}^{nm} \epsilon_{ij}^{sm} C_{ijkl} \quad (45)
\]

\[
Q_{ij} = \frac{1}{2} \sum_{n} \sum_{m} K_{\rho\delta}^{nm} \left[ P_{ij}^{sm} - \epsilon_{ij}^{sm} \Gamma_{ij} \right] \quad (46)
\]

III. RESULTS AND CONCLUSION

As an example, the evolution of the strain and the electric displacement during a compressive test is simulated (Fig.3 and Fig.4).

![Fig.3. Evolution of the strain in the case of a compressive test.](image1)

![Fig.4. Evolution of the electric displacement in the case of a compressive test.](image2)

The mechanical depolarisation is characterised by the decrease of the initial polarisation and the increase of the remanent polarisation with respect to the applied stress. The corresponding materials parameters are given in Table 1.
Table 1. Values chosen for to the material constants for the computations.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E$</td>
<td>3.3*10^4</td>
</tr>
<tr>
<td>$v$</td>
<td>0.3</td>
</tr>
<tr>
<td>$d_{33}$</td>
<td>1.52*10^6</td>
</tr>
<tr>
<td>$d_{31}$</td>
<td>-5.7*10^-7</td>
</tr>
<tr>
<td>$d_{15}$</td>
<td>1.856*10^-6</td>
</tr>
<tr>
<td>$\kappa_{33}$</td>
<td>6.7*10^-8</td>
</tr>
<tr>
<td>$\varepsilon^r$</td>
<td>0.27</td>
</tr>
<tr>
<td>$P^r$</td>
<td>2*10^-7</td>
</tr>
</tbody>
</table>

This paper describes the switching process during electrical and mechanical loadings in ferroelectric single crystals. It could be described by the interaction between domains. They are considered like variants having an orientation defined by the polarization direction. From this viewpoint, the volume fraction of each of them are used as internal variables so that the free energy is function of the macroscopic stress, electric field and the set of the volume fractions. These interactions induce fluctuating internal and multiaxial stresses and electric fields. The macroscopic stress and electric displacement depend on the microstructure the domain pattern resulting from these interactions. It appears also that this switching process leads to domain wall motions and it could be verified and characterized using techniques like scanning force microscopy. It means that the interaction between domains has to be described more accurately by expliciting the interaction matrix. Therefore, it may be evaluated using interface operator techniques. This work is under investigations and may account for a spatial organization of domains.

REFERENCES


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