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RESEARCH ARTICLE

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Stress induced polarization switching and coupled hysteretic dynamics in ferroelectric materials

Abstract The dynamic responses of ferroelectric materials upon external mechanical and electrical stimulations are inherently nonlinear and coupled. In the current paper, a macroscopic differential model is constructed for the coupled hysteretic dynamics via modeling the orientation switching induced in the materials. A non-convex potential energy is constructed with both mechanic and electric field contributions. The governing equations are formulated as nonlinear ordinary differential equations by employing the Euler-Lagrange equation, and can be easily recast into a state space form. Hysteresis loops associated with stress induced polarization switching and butterfly-shaped behavior in ferroelectric materials are also successfully captured. The effects of mechanical loadings on the electrically induced switching are numerically investigated, as well as the mechanically-induced switching with various bias electric fields.

Keywords differential model, state space, electromechanical switching, butterfly effects, hysteresis

1 Introduction

Electro-mechanical materials have been extensively used in many engineering applications as sensors and actuators due to their capability of converting energy between electrical and mechanical types. The key property responsible for a wide range of current and potential applications is the intrinsic coupling effects between the electric and mechanic fields in the materials. It has been experimentally observed that, ferroelectric materials exhibit much more pronounced electromechanical coupling effects compared to other piezoelectric materials because of the characteristic ability to switch polarizations orientations in response to external electric or mechanical loadings. But, the constitutive laws of the ferroelectric materials are also much more complex. A common feature of the coupled dynamics of the ferroelectric materials is the presence of hysteresis loops in the E-P (electric field-electric polarization) curves under large dynamic loadings, and the presence of the butterfly-shaped curve in the E-ε (electric field-strain) relations. Furthermore, the hysteresis loops and butterfly-shaped curves are affected by both the mechanic and electric loadings and by their variation rates [1,2].

To enhance the applicability of the ferroelectric materials and to improve the application performance, it is essential to accurately model the hysteretic behaviors of these materials [3–5]. In the current paper, a phenomenological macroscopic differential model is constructed by simulating the orientation switching dynamics. The hysteresis loops and butterfly-shaped behaviors are treated as macroscale illustrations of the polarization orientation switching. A non-convex potential energy is constructed to characterize different polarization orientations in the materials at macro-scale. Each local equilibrium of the potential energy function is associated with one of the stable orientations. System state (polarization orientations) can be switched upon external loadings (electrical or mechanical) from one stable orientation to another, and the dynamics thus can be modeled by investigating the switching dynamics of the system states from one equilibrium state to another [6–8]. Governing equations for the switching dynamics are formulated by employing the Euler-Lagrange equation, and the damping effects of the switching process are also included in the model. The

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model is expressed as nonlinear differential equations.

We present results of numerical simulations where hysteretic dynamics and butterfly-shaped behaviors are successfully captured by the proposed model. The effects of mechanical loadings on the electrically induced orientation switching are numerically investigated, and the effects of biased electrical fields on the mechanically induced orientation switching are also numerically analyzed.

2 Orientation switching and hysteretic dynamics

The hysteretic dynamics can be regarded as a consequence of the polarization orientation switching induced in the materials. Due to the intrinsic coupling effects between the electric and mechanical fields in the ferroelectric materials, the system response (polarization and strain) are both affected by mechanical and electrical inputs. For the purpose of modeling of the coupled hysteretic dynamics, it is rather natural to construct a model on the basis of the description of the orientation switching.

At temperatures below the Curier temperature, the polarization orientations could be switched by mechanical or electrical loads. For cubic-tetragonal switching, there are six tetragonal variants in the three dimensional description, as sketched in Fig. 1(a). In Fig. 1, there are three tetragonal, each tetragonal has two possible orientations which are opposite as indicated by the arrows.

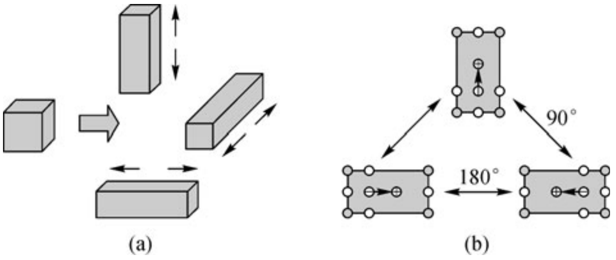

Fig. 1 Schematic of polarization orientation switching. (a) The cubic and tetragonal (6 variants) in the three dimensional system; (b) the simplified one-dimensional analog with only three orientations

For clarity, the three dimensional switching is simplified into a one-dimensional analog, as sketched in Fig. 1(b). It has been verified that the simplified one-dimensional analog is capable of accounting for the hysteretic behavior of the material in the one-dimensional system [8–9]. By doing this, the mechanically induced 90° orientation switching is also incorporated into the modeled by simulating the switching between the horizontal and vertical rectangles [7–10].

According to the Landau theory of phase transformations, various polarization orientations involved in the polarization and orientation switchings can be characterized by an appropriately constructed free-energy function (the Landau free energy function), which should be non-convex and has multiple local minima (local equilibria). Each of the local equilibria can be associated with one of the polarization orientations involved. Since the purpose of the current paper is to construct a macro-scale model for the hysteretic dynamics, one can construct a potential energy for the dynamical system by mimicking the methodology of the Landau theory for phase transformations. Therefore, the potential energy for the one-dimensional analog of polarization switching can be constructed in the following form by taking into account the energy contributions from both the electric and mechanical fields.

$$W(P,\varepsilon)=\frac{a_2}{2}P^2+\frac{a_4}{4}P^4+\frac{a_6}{6}P^6+\frac{k}{2}\varepsilon^2+\frac{b}{2}\varepsilon P^2, \tag{1}$$

where ε is the elastic strain, P is the polarization. For characterizing different polarization orientations, P is chosen the order parameter.

The first three terms in Eq. (1) give the Landau type energy function, which is a six order polynomial and has only even terms of the polarization. One can easily see that it may have three minima out of the five extrema. Since the function is even, one of the minima must be $P = 0$, and the other two minima are symmetric. It has been verified that the above potential energy function can be tuned to handle orientation switching by choosing suitable coefficient values for a_2, a_4, and a_6 [11].

3 Differential model

For the modeling of the macro-scale hysteretic dynamics of ferroelectric materials, it is reasonable to assume that the strain, polarization, electric field, and stress are all uniform in the materials being discussed here. The equations of the motion can be set up by using the Lagrange's equations. For this purpose, the kinetic energy of the system can be written as

$$U=\frac{1}{2}\left(I_P\left(\frac{\mathrm{d}P}{\mathrm{d}t}\right)^2+I_\varepsilon\left(\frac{\mathrm{d}\varepsilon}{\mathrm{d}t}\right)^2\right), \tag{2}$$

where I_P and I_ε are material-specific constants related to the generalized inertial effects during orientation switching in the electric and mechanical fields, respectively.

In order to take into account the damping effects during the switching processes, a dissipation effect is introduced here, which is proportional to the square of the change rate of the system states.

$$R=\frac{1}{2}\left(\tau_P\left(\frac{\mathrm{d}P}{\mathrm{d}t}\right)^2+\tau_\varepsilon\left(\frac{\mathrm{d}\varepsilon}{\mathrm{d}t}\right)^2\right), \tag{3}$$

where τ_P and τ_ε are material-specific constants related to the generalized friction in the electric and mechanical fields, respectively.

By employing the Euler-Lagrange equation with the given potential energy, kinetic energy, and dissipation energy, the governing equations of the considered dynamics under mechanical loadings (stress σ) and electric loadings (electric field E) can be formulated as the following two equations:

$$\begin{aligned} &\tau_P \frac{\mathrm{d}P}{\mathrm{d}t} + a_2 P + a_4 P^3 + a_6 P^5 + b\varepsilon P - E = 0, \\ &\tau_\varepsilon \frac{\mathrm{d}\varepsilon}{\mathrm{d}t} + k\varepsilon + \frac{bP^2}{2} - \sigma = 0. \end{aligned} \tag{4}$$

In the above equations, the generalized inertial effects are ignored by setting the corresponding coefficients to zero, which makes the governing equations the first order differential equations. The simplification could be justified by the fact that, in most experimental observations, the inertial effects in the switching processes at macro-scale are much smaller compared to the damping effects and can be ignored in the dynamic analysis.

In the proposed model given by Eq. (4), there are seven model parameters which need to be estimated using experimental data. The details of parameter estimation was presented in Refs. [6,10], where the model was also validated by comparing simulation results and experimental data. It was illustrated that both the hysteresis loops in the E-P curves and the butterfly-shaped E-ε curves are successfully captured by the nonlinear ordinary differential equations, provided with suitable model parameter values.

4 Electric field induced switching

For the purpose of illustration, the dynamics of the considered system under a 1 Hz sinusoidal electric field with different constant stresses are presented in Fig. 2 (E-P curves) and Fig. 3 (E-ε curves). The applied stresses are set as $\sigma = -60, 0, 60$ MPa, in which the negative sign indicates that the applied stress is a tensile stress, whilst the positive values indicate compressive stresses. The non-dimensional parameter values are taken as follows: $\tau_P = 0.01$, $a_2 = -1.06$, $a_4 = 51001.48$, $a_6 = -1797.95$, $b = -2580.16$, $k = 65.42$, $\varepsilon_0 = 1.006$ [6,10].

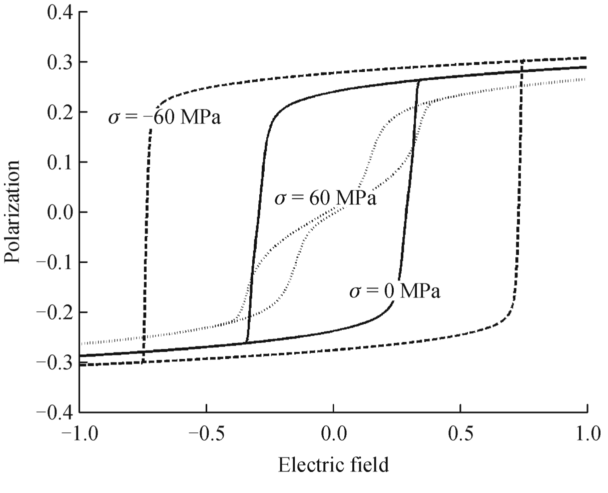

Fig. 2 Effects of stresses on the polarization switching in the ferroelectric materials (E-P curves)

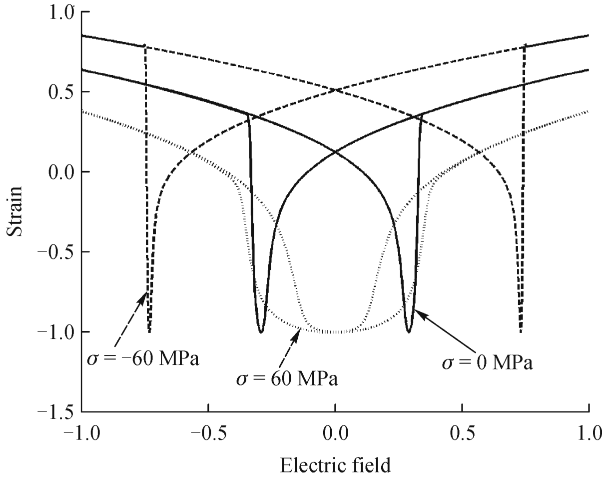

Fig. 3 Effects of the stresses on the polarization switching in the ferroelectric materials (E-ε curves)

It is clearly seen that the tensile stresses will enlarge the hysteresis loops by increasing the coercive field values and making the orientation more difficult to be switched. the tensile stresses also enlarge the butterfly-shaped E-ε curves of the material, the distance between the two branches of the butterfly-shaped E-ε curves are increased. While the compressive stress tends to make the orientation switching easier, the types of switching are different. In the one-dimensional description, all the orientation switchings caused by the electric fields are 180° switching, but the

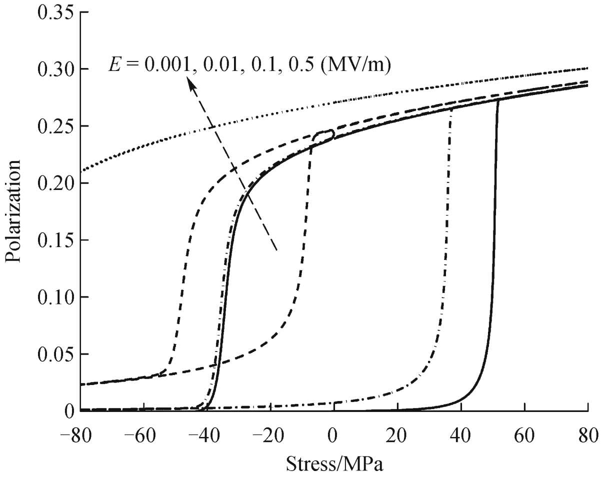

Fig. 4 Simulations of the effects of the biased electric fields on stress induced polarization switchings

switchings caused by the compressive stresses are always 90° switchings. The differences in the switching types are clearly illustrated by the different types of hysteresis loops in the *E-P* curves. The *E-P* curves associated with 180° switching always have a single hysteresis loop, while those *E-P* curves with 90° switching have two sub-hysteresis loops, which are symmetric if the two horizontal orientations are also symmetric.

5 Stress induced switching

Based on the proposed model, the effects of biased electric fields on the stress induced switching are also simulated. The applied stresses in these cases are chosen as a sinusoidal function $\sigma = 80\sin(2\pi t)$ MPa. At the same time, constant biased electric fields are applied at different values, in particular $E = 0.001$, 0.01, 0.1, 0.5 MV/m. The simulated orientation switchings are presented by plotting the σ-P curves in Fig. 4 and the σ-ε curves in Fig. 5. It is seen that, orientations in the considered ferroelectric structure can also be switched by the stress loadings when the electric loadings are weak. All the switchings are 90° because the polarization value never crosses the zero line in the plot.

Meanwhile, the hysteresis loops in the σ-P curves are shifted and narrowed by the biased electric fields. This is easy to explain since the biased electric fields are constants. Consequently, the hysteresis loops in σ-ε curves are also shifted and narrowed by the biased electric fields. Furthermore, when the biased electric field is rather strong ($E = 0.5$ MV/m), the mechanically induced switching are completely suppressed, and there are no hysteresis loops in either σ-P curves or σ-ε curves. The simulation results can be easily explained by using Fig. 1(b) as follows. The vertical orientation has zero strain and polarization, which is favored by the compressive stresses. Whilst the horizontal orientations have non-zero polarization and strain, which is favored by the tensile stresses and electrical loadings. The overall response of the system under the combined electric-mechanical loadings is determined by the competition of the three involved orientation.

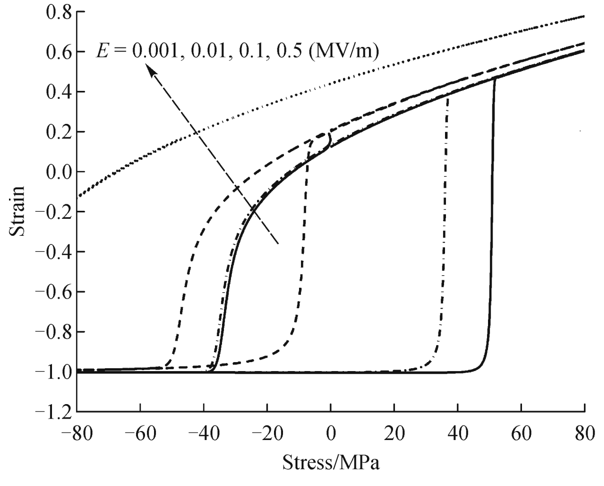

Fig. 5 Simulations of the effects of the biased electric fields on stress induced polarization switchings

6 Conclusions

In this paper, the coupled hysteretic dynamics of the ferroelectric materials under combined electro-mechanical loadings has been investigated by using a macroscopic differential model. The model has been constructed by employing a phenomenological theory for the polarization orientation switching. Hysteresis loops in the electric field and butterfly-shaped behaviors in the electro-mechanical coupling have been modeled as a consequence of polarizations and orientation switchings, together with a nonlinear electro-mechanical coupling. The effects of bias stresses on the polarization switchings have been analyzed. Stress induced polarization switchings have also been studied with the proposed model.

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References

1. Hall D A. Nonlinearity in piezoelectric materials. Journal of Materials Science, 2001, 36(19): 4575–4601
2. Yen H H, Shu Y C, Shieh J, Yeh J H. A study of electromechanical switching in ferroelectric single crystals. Journal of the Mechanics and Physics of Solids, 2008, 56(6): 2117–2135
3. Chen W, Lynch C S. A Micro-electro-mechanical model for polarization switching of ferroelectric materials. Acta Materialia, 1998, 46(15): 5303–5311
4. Ball B C, Smith R C, Kim S J, Seelecke S. A stress-dependent hysteresis model for ferroelectric materials. Journal of Intelligent Material Systems and Structures, 2006, 18(1): 69–88
5. Smith R C, Seelecke S, Dapino M, Ounaies Z. A unified framework for modeling hysteresis in ferroic materials. Journal of the Mechanics and Physics of Solids, 2006, 54(1): 46–85
6. Wang L X, Liu R, Melnik R V N. Modeling large reversible electric-field-induced strain in ferroelectric materials using 90° orientation switching. Science in China Series E: Technological Sciences, 2009, 52(1): 141–147
7. Wang L X. Hysteretic dynamics of ferroelectric materials under electromechanical loadings. In: Proceedings of SMASIS08 ASME Conference on Smart Materials, Adaptive Structures and Intelligent Systems, 2008,
8. Brown S A, Hom C L, Massuda M, Prodey J D, Bridger K, Shankar N, Winzer S R. Electromechanical testing and modeling of a PbO_3-

$PbTiO_3$-$BaTiO_3$ relaxor ferroelectric. Journal of the American Ceramic Society, 1996, 79(9): 2271–2282

9. Wang L X, Chen Y, Zhao W L. Macroscopic differential model for hysteresis and buttery-shaped behavior in ferroelectric materials, Advanced Materials Research, 2008, (47–50): 65–68
10. Wang L X, Melnik R. Control of coupled hysteretic dynamics of ferroelectric materials with a Landau-type differential model and feedback linearization. Smart Materials and Structures, 2009, 18(7): 074011
11. Falk F. Model free energy, mechanics, and thermomechanics of shape memory alloys. Acta Metallurgica, 1980, 28(12): 1773–1780