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International Journal of Solids and Structures 43 (2006) 697–712
INTERNATIONAL JOURNAL OF SOLIDS and STRUCTURES
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Rate dependent properties of perovskite type tetragonal piezoelectric materials using micromechanical model

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Received 6 October 2004; received in revised form 18 April 2005
Available online 23 June 2005

Abstract

In this paper, a three-dimensional micromechanical model is presented for simulation of the rate dependent properties of certain perovskite type tetragonal piezoelectric materials. The model is based on linear constitutive, nonlinear domain switching, and linear kinetics theories. The simulation starts with a virgin bulk material of randomly oriented grains. Then the material is electrically loaded with an alternating voltage of various frequencies, which are in the order of 0.01 Hz to 1 Hz. An energy equation in combination with a probability function is used to determine the onset of the domain switching inside the grains. Such a probability function leads to a better phenomenological model for the domain switching even for electrical loadings, which are in a range far below the coercive fields. The propagation of the domain wall during the domain switching process in grains is modeled by means of linear kinetics relations after domain nucleation. The response of the bulk ceramic is predicted by averaging the response of individual grains using Euler angles for the transformation from local coordinates of the grains to global coordinate. Electric displacement hysteresis loops for different frequencies and amplitudes of the alternating electric fields are simulated. A simple micromechanical model without the probabilistic approach is compared with the one that takes it into account. Both models give important insights into the rate dependency of piezoelectric materials, which was observed in some experiments reported in the literature.

Keywords: Piezoelectricity; Rate dependence; Micromechanical modeling; Domain switching; Probabilistic approach

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doi:10.1016/j.ijsolstr.2005.04.030

1. Introduction

Ferroelectric and piezoelectric materials, which are commonly made of ceramics, have become a highly interesting part of smart materials due to their applications in intelligent systems, especially in the electronics and mechatronics engineering area. Vibration damping and noise control, precision positioning & cutting, injection mechanism in common rail systems with piezo control in automobiles, ink-jet printers, commercial stack actuators and sensors are some examples for applications of these materials in recent years. Although they have manufacturing difficulties, ferroelectric and piezoelectric microelectromechanical systems (MEMS) are also used in ferroelectric thin films, FRAM (ferroelectric random access memories), piezoelectric accelerometers and micromirrors (Yee et al., 2001). Piezoceramic materials can be classified into two types as soft and hard piezoceramics according to their crystal lattice. In general, soft piezoceramic materials have higher piezoelectric constant. $BaTiO_3$, PZT and PLZT (lead lanthanum zirconate titanate) are some examples of perovskite type tetragonal piezoceramic materials. The microstructure of $BaTiO_3$ is shown in Fig. 1. The fundamentals of piezoelectric materials are explained by Jaffe et al. (1971).

It is known that the lattice structure of piezoceramics is temperature dependent. When the temperature is reduced below the Curie temperature (T_C), the phase of piezoceramic materials changes from simple cubic to a tetragonal or a rhombohedral structure depending on the composition of the material (Fig. 1). For most PZT materials the Curie temperature is about 150–300 °C. Piezoceramic materials behave almost linearly when the electromechanical loading is small. Below the Curie temperature they possess a spontaneous polarization, the direction of which can change when they are loaded with a high electromechanical field. This change in direction of the spontaneous polarization in the microstructural level leads to a nonlinear behavior of the material. In literature various types of experiments were performed in order to determine non-linear characteristics of piezoelectric and ferroelectric materials under high electromechanical loadings. In these experiments, the piezoceramic sample is usually subjected to a quasi-static cyclic electric and/or mechanical loading (Hwang et al., 1995; Cao and Evans, 1993; Schaeufele and Haerdtl, 1996; Lynch, 1996; Lu et al., 1999). With these experiments, rate independent effects depending on the composition and concentrations of elements of some PLZT materials are investigated. Both (hard and soft) type of piezoceramics are examined and the electric displacement and mechanical strain are measured as functions of the applied electric field or mechanical stress. In these experiments the composition of the material and the concentrations of different elements are varied. This allows not only the investigation of different directions of polarization, but also the different phase changes like tetragonal to rhombohedral and vice versa.

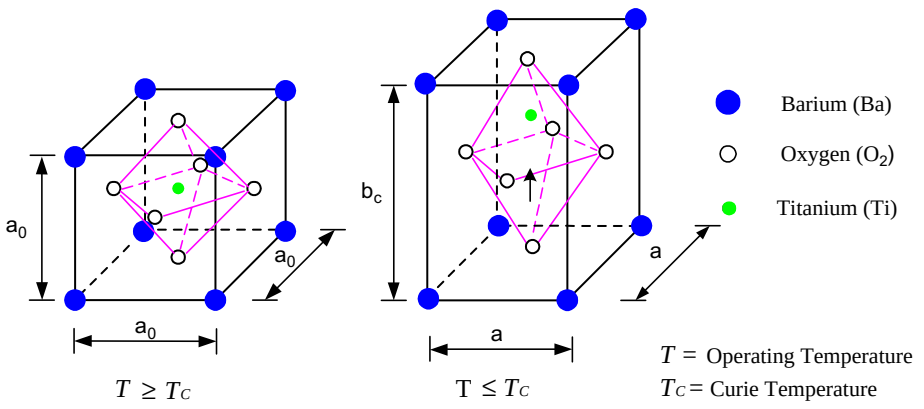

Fig. 1. Lattice structure of cubic and tetragonal elements ($BaTiO_3$).

Simulations of the nonlinear behavior of piezoelectric materials based on physical insights of the material are performed by using micromechanical models. In these models usually bulk piezoelectric materials are assumed to be composed of elements that are randomly oriented. It is supposed that the behavior of each element is that of a microcrystal. Linear constitutive equations are used to calculate the effects of the applied electric field and the applied mechanical stress until domain switching occurs. The models assume that the electric field or the mechanical stress in the material is uniform. An energy equation is introduced for the threshold of domain switching. Analytical averaging over all elements is implemented in order to get the macroscopic properties according to the micromechanical model. Hwang et al. (1995) proposed a similar model like the one described above. They used Preisach hysteresis model to describe the electric displacement and strain simulation of each grain of the PLZT. The simulations are compared with experimental results, which were performed with a PLZT 8/65/35. Lu et al. (1999) used an orientation distribution function which is the major difference compared to the other approaches for the simulation of perovskite type tetragonal ferroelectric ceramics under various electromechanical loadings. Further similar micromechanical models have been developed in order to be able to predict better the hysteresis and butterfly curves under quasi-static loading by means of some assumptions (Hwang et al., 1998; Chen and Lynch, 1998). There are other attempts to model the mechanical stress effect subject to a cyclic electrical loading in the simulations. In these models different energy equations are introduced in order to simulate hysteresis and butterfly curves under constant mechanical stress (Lynch, 1996; Lu et al., 1999; Chen and Lynch, 1998; Hwang and McMeeking, 1998a,b).

The phenomenological modeling of the nonlinear behavior is another approach which is mostly based on a thermodynamic framework. Different interesting phenomenological models, which investigate macroscopically history dependent nonlinearities, dielectric hysteresis, butterfly hysteresis due to domain switching, thermo-electromechanical coupling properties and time dependent effects in the simulations for ferroelectric materials are presented by Kamlah et al. (1997), Kamlah and Jiang (1997), Kamlah and Tsakmakis (1999) and Kamlah and Böhle (2001). Similarly, McMeeking and Landis (2002) presented a phenomenological constitutive model for the ferroelectric switching subject to multi-axial mechanical and electrical loadings.

The finite element method is one of the more useful tools, which can be used effectively for the simulation of piezoelectric materials. Allik and Hughes (1970) gave the basics of the finite element formulation of such materials. Li and Fang (2004) developed a three-dimensional finite element model for the non-linear modeling of ferroelectric materials with tetragonal microstructures. In their model the energy barrier for 90° domain switching is assumed to be one quarter of that for the 180° domain switching. Therefore, two successive 90° domain switchings occur instead of one 180° domain switching. Besides constant uni-axial loading, they applied a multi-axial loading for the investigation of the change of butterfly and hysteresis curves. In literature there are further attempts to use finite element methods for solving complex thermodynamical equations by using phenomenological models (Hwang et al., 1998; Kamlah, 2001). The finite element method can also be implemented in micromechanical models though, in general, they need much more time for simulations compared to the simple micromechanical models which are explained above (2002).

Polarization switching and kinetics of it in the grains of piezoelectric materials are not independent on the frequency of the cyclic loading. In some applications of piezoelectric materials, rate dependent characteristics are important and have to be studied. The influence of the loading rate on the electric displacement and mechanical strain is observed by Zhou et al. (2001) by using different loading frequencies in experiments for PIC151 soft PZT piezoceramics. A macromechanical model for the time dependent hysteresis of piezoelectric materials at low frequencies is given by Smith et al. (2001). The model incorporates rate dependency into the basic quasi-static hysteresis model. Although the model does not accurately predict the change in the coercive field, it is able to reproduce the reduction in remnant polarization to increase the loading frequencies. A 3-D continuum model, which takes into account a switching criterion and the kinetics of polarization switching process has been successfully applied to 2-D ferroelectric ceramics by

Kim and Jiang (2002) using the finite element method. The model assumes that the material consists of various types of ferroelectric variants in which the behavior is characterized by the Helmholtz free energy. The start of nucleation of domains and the kinetics of switching are determined by the value of the electromechanical energy and the driving force between the different variants, respectively. A simple linear kinetics relation is used for the propagation of the new variant. The model explains the rate-dependent behavior in terms of the changes in mass fractions of different types of variants. The simulations are performed for hysteresis and butterfly curves for a cyclic loading with various frequencies and shows the increase in the hysteresis curve when the loading frequency is increased. Viehland and Chen (2000) presented an interesting experimental investigation of frequency dependent characteristics of a sample ferroelectric material with the chemical name $0.7Pb(Mg_{1/3}Nb_{2/3})O_3$–$0.3PbTiO_3$. The results of experiments and simulations can be well understood for such types of ferroelectrics. One example is that the polarization for high frequencies may increase even though the electric field is decreasing. This effect was observed in experiments with ferroelectric materials (Zhou et al., 2001; Zhou, 2003).

In this paper, the rate dependent properties of PIC151 piezoelectric materials are investigated using a three-dimensional micromechanical model (Seemann et al., 2004). The main difference of the model compared with existing models is the fact that domain switching occurs with the probability depending on an energy equation. Therefore, domain switching is not limited to electric and mechanical fields above some critical values but it may also happen at values which are below the coercive level. The paper is organized as follows: first, the model is described and quasi-static properties are given. Then the model is extended to cover the frequency and amplitude dependence for a cyclic loading of these materials. The probability functions that have been used together with the energy equation are assumed to be related to the applied electric field where the macroscopic mechanical stress is supposed to be zero. The influence of intergranular stresses can be modelled phenomenologically by using these probability functions. The results of the simulations are shown in hysteresis curves for the electric displacement versus the electric field. In the simulations the electric field is uniformly distributed and generated by an electric voltage, which is triangular in time. Simulations are performed for various frequencies and amplitudes.

2. Phenomena

As explained above piezoceramic materials at room temperature mostly exist in a phase with tetragonal or rhombohedral lattice structure depending on the constitutional elements of the material. The Curie temperature is the transition temperature for ferroelectric and piezoelectric materials at which they start to change their lattice structure. When the temperature is above the Curie temperature, the lattice structure is in the form of cubic elements, in which the net dipole moment is zero due to the equally spaced locations of atoms inside the lattice. This phase is called the paraelectric phase. The microstructure of these materials transforms into a different type when the temperature is reduced below the Curie temperature. The new phase occurring after this transition is called the ferroelectric phase, in which there is net non-zero dipole moment (Fig. 1). The polarization, which is the result of the net dipole moment per volume of the microstructure in the lattice structure during the phase transition, is called the spontaneous polarization (P_0). The direction of this polarization can change, when a large electric or mechanical load is applied. In addition to this, the motion of the atoms during the phase transition leads to a net mechanical strain, which is called the spontaneous strain. The model used in this paper assumes a piezoelectric material with a lattice structure of perovskite type tetragonal elements. Therefore, the spontaneous strain (S_0) in the loading can be calculated by

$$S_0 = \frac{(b_c - a_0)}{a_0} \tag{1}$$

Spontaneous strain depends on the dimensions of the unit cells of both phases (b_c and a_0).

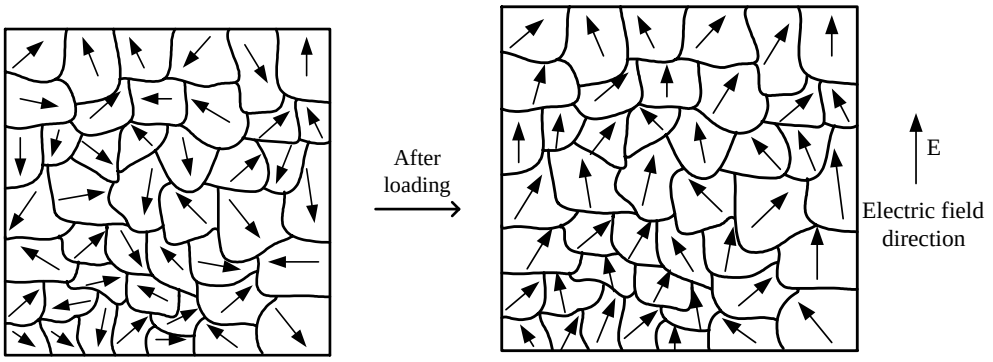

Fig. 2. Grains of the piezoceramic material before and after electrical loading (2-D view).

Looking closely at the microlevel of both piezoelectric and ferroelectric materials, it can be seen that the unpoled material is composed of randomly oriented grains. It is assumed that the bulk material is composed of many grains polarized randomly (Fig. 2). Therefore, the average polarization or mechanical strain of the bulk material is approximately zero. When the material is subject to a high external electric field, the directions of all spontaneous polarization vectors are aligned in the direction of electric field by means of domain switching of the grains. Since, the direction of the polarization vector after domain switching has to be in one of the local lattice axes, the polarization in general is not parallel to the electric field locally but has a component in direction of the electric field. The constitutive equations are used to calculate the electric displacement and mechanical strain values for each grain of the material taking into consideration the spontaneous polarization and spontaneous mechanical strain values.

$$D_k = \varepsilon_{kn} E_n + d_{klm}\sigma_{lm} + P_{0k} \tag{2}$$

$$S_{ij} = s_{ijlm}\sigma_{lm} + d_{nij}E_n + S_{0ij} \tag{3}$$

In these equations, D_k denotes the electric displacement, S_{ij} is the mechanical strain, E_n is the electric field, σ_{lm} is the mechanical stress, P_{0k} is the spontaneous polarization in the k direction, S_{0ij} is the spontaneous strain, d_{klm} is the piezoelectric constant, ε_{kn} is the dielectric constant and s_{ijlm} is the compliance tensor. The spontaneous polarization in direction of the polarization vector is P_0 while the components in transverse direction are zero. In Eq. (3) the spontaneous strain S_{0ij} is S_0 in direction of the polarization vector, $-S_0/2$ in transverse direction while S_{0ij} is zero for the shear strain.

For perovskite type tetragonal elements only two types of domain switching are possible. These are 90° and 180° switching, the names of which denote to the angle of rotation, which the position vector of the central atom undergoes during domain switching relative to its previous direction (Fig. 3).

3. Model description

In this paper, the bulk piezoceramic material is modeled by 1000 elements each is assumed to have the characteristics of an individual grain. The crystal axes and therefore the polarization direction in each grain do have a random orientation. These orientations of the grains are given statistically by using a random generator in the simulation. Randomness is given to every element by means of three Euler angles between 0 and 2π. A global coordinate system for the bulk material and a local coordinate system for each element are introduced in order to transform the values obtained by the calculations in the local coordinate systems to global coordinates and vice versa. Although it is clear that equally distributed Euler angles do not

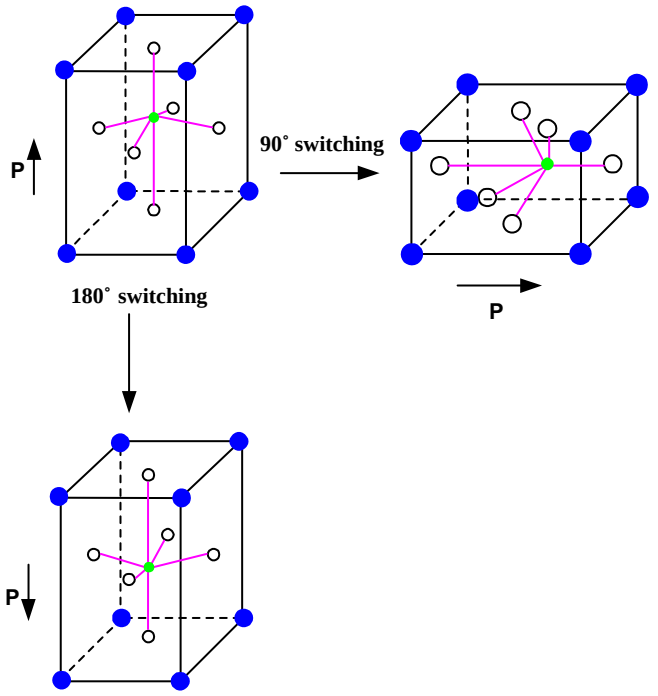

Fig. 3. The 90° and the 180° switching in the lattice structure of a perovskite type tetragonal element.

guaranty a uniform distribution of the polarization directions, it is common to use these equally distributed Euler angles for such simulations in micromechanical models. Polarization, strain and electric field given in local coordinates in a domain can be transformed to global coordinates or vice versa with the help of the corresponding transformations.

As explained before, material constants such as dielectric and piezoelectric constants influence the types of piezoelectric materials. For quasi-static simulations the results have been published before (Delibas et al., 2004). Parameters of soft PZT-51 material are chosen in order to compare with the hysteresis and butterfly curves presented in the experimental results (Lu et al., 1999). A cyclic, uni-axial electric field is applied with an amplitude of 2 kV/mm during the simulation. The starting point for the first cycle is at zero electric field for the unpoled bulk ceramic. The domain switching at each grain is determined by using the electromechanical energy criterion (Hwang et al., 1995), (Eq. (4)).

$$\Delta P_i E_i + \Delta S_{ij}\sigma_{ij} \geq 2P_c E_c. \tag{4}$$

In this relation E_i and σ_{ij} are the electric field and the mechanical stress, E_c is the coercive electric field, P_c is the critical value of the spontaneous polarization. Both E_c and P_c are material dependent constants. ΔP_i and ΔS_{ij} are the polarization and strain changes during the domain switching correspondingly. During the simulation the stress is assumed to be zero. Thereby, on the left side of the inequality (4), the second term vanishes. According to this criterion, domain switching occurs if the energy change is higher than a certain critical level. For 90° and 180° domain switchings in perovskite type tetragonal elements the same energy levels are adopted during the simulations. For quasi-static simulations the critical value of the spontaneous polarization (P_c) of 0.3 C/m^2 and 0.676 kV/mm for the coercive electric field (E_c) are used (Lu et al., 1999).

Intergranular effects between grains do have a significant influence on the nonlinear behavior of piezoceramic materials. Since all grains are oriented differently from each other, grain boundary stresses are

expected to occur during the loading. Especially in the domain switching range these stresses are very important. Therefore, local values of the loading, which can be the electric field or the mechanical stress can be much different in neighboring grains, so that a domain switching can occur even at levels of the macroscopic loading, which are well below the coercive field. The corresponding nonlinearities can be observed even in a small electromechanical loading range. In order to take into account intergranular effects phenomenologically, a certain probability for domain switching is introduced in the described model. The probability for domain switching is taken to be a polynomial function of some degree (n) of the ratio of the absolute values of the actual applied free energy and the critical energy (Eqs. (5) and (6)). The value of the probability (p) is varying between 0 and 1 depending on the applied electromechanical energy. In this model n is a parameter, which can be chosen arbitrarily in order to fit the results to experimental data. In the simulations n was chosen to be two, three, four or five. Also other functions for the probability like exponential or hyperbolic functions may be used in the simulations. However, in our simulations we restrict to polynomials and other probability functions are not implemented in this paper. According to the probabilistic approach, domain switching only depends on Eq. (5), if the applied free energy is smaller than the critical energy. So, with this polynomial function the probability for a domain to switch its polarization direction is more and more increasing, if the applied electric field is approaching the level of the coercive electric field. For a loading higher than the coercive field, Eq. (4) is automatically satisfied, which means that the domain switching occurs in any case. For example, if the applied electric field E_i is 0.4 kV/mm and polarization change ΔP_i which is assumed to have same magnitude with polarization constant P_c is 0.3. When coercive field is 0.676 kV/mm and n is chosen 3, then $P(0.4)$ equals 0.026. So we have 2.6% of probability of domain switching. In our simulation code, random generators are implemented according to this result in order to give corresponding percent of domain switching. Therefore, 26 elements are estimated to have domain switching among 1000 elements when the applied electric field is 0.4 kV/mm.

$$p(E_i) = (E_i \Delta P_i / 2E_c P_c)^n \quad \text{for } \|E_i\| \leq \|E_{ci}\| \tag{5}$$

$$p(E_i) = 1 \quad \text{for } \|E_i\| > \|E_{ci}\| \tag{6}$$

Fig. 4 shows a polarization (electric displacement) versus electric field hysteresis curve for a quasi-static loading without considering a probability function in the simulation. As it can be seen in this figure the polarization curve has a sharp corner near the coercive electric field, which is the main difference of the model compared to experimental data. Fig. 5 is the curve from the model in which a fourth order polynomial ($n = 4$) for the probability function is implemented. Smoothness of the curves near the coercive electric field levels can be observed easily from the figures. A comparison of the simulation with experimental data which was measured by Lu et al. (1999) is also given (Fig. 6). The simulated results obtained with a fifth order ($n = 5$) probability function fit better to experimental results than those curves, which were simulated without using the probabilistic approach.

4. Rate dependent model

Early experiments to determine dependence of the electric field and the temperature on the switching time and the switching current were performed by Merz (1954) for $BaTiO_3$ single crystals. In addition he explained the switching mechanism as the nucleation of new domains and a growth process with domain wall motion. For an applied constant electric field, a reduction of the switching time and thus an increasing switching current was observed for increasing temperatures at which the experiments were performed. This means that the nucleation of domains and the growth process are faster at higher temperatures. Likewise, the dependence of the switching time and the current on the applied electric field and the coercive field at a fixed temperature was observed. One of the first publications on the simulation of the rate dependency of polarization curves for $BaTiO_3$ was given by Landauer et al. (1956). They used the observations of Merz

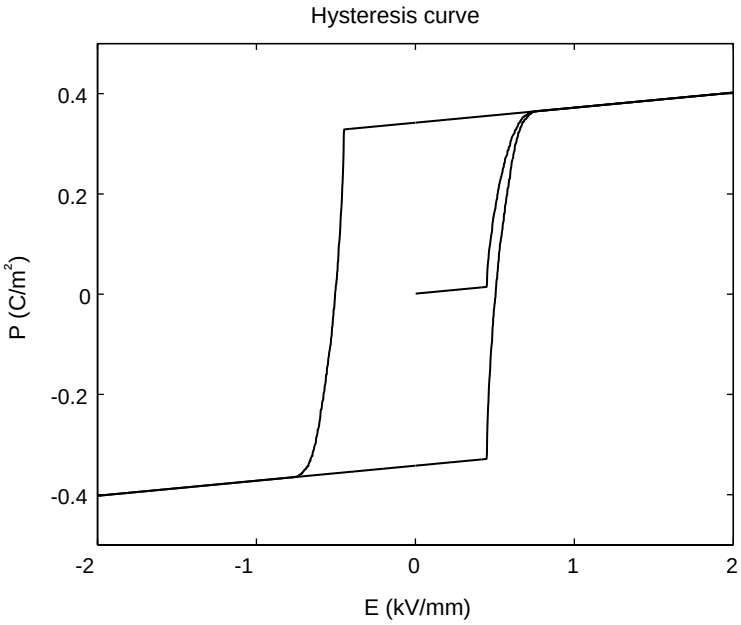

Fig. 4. Hysteresis curve without probability function.

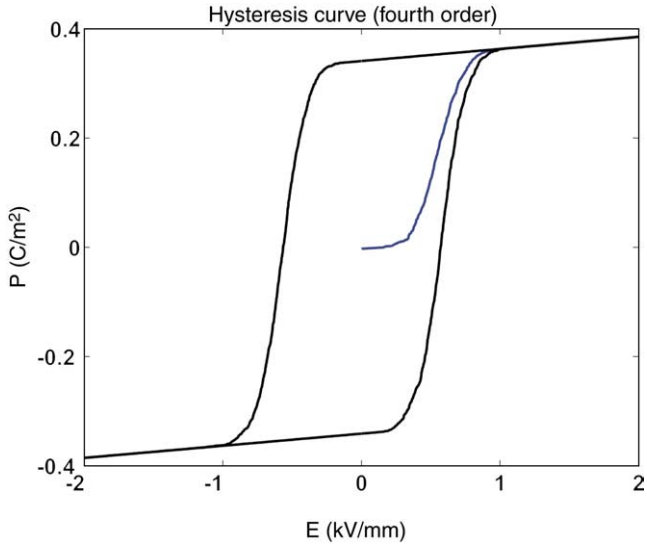

Fig. 5. Hysteresis curve with fourth order polynomial for the probability function.

(1954), in which the rate of switching was basically expressed as an exponential function of the applied electric field. Although the switching process was not determined clearly as a nucleation with further domain expansion, they managed to illustrate the change of the coercive field due to different loading rates. The minimum coercive electric field was found for the lowest loading rate. It is known from literature that

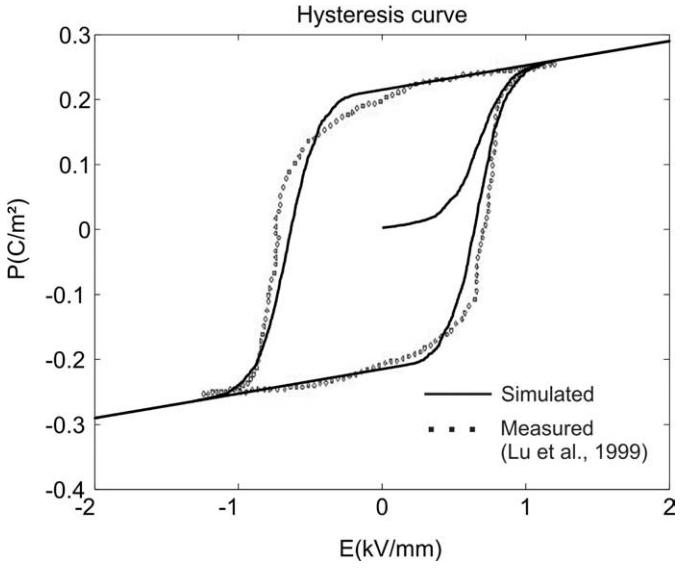

Fig. 6. Comparison between simulation (fifth order polynomial for the probability function) and measured data.

domain switching starts inside a domain of the piezoelectric material at a critical level of either the applied electrical field or the mechanical stress. The generation of the new phase during the domain switching process can be separated into two parts: nucleation and kinetics of the new phase (Abeyaratne et al., 1994). The beginning of the nucleation of the new phase is assumed to be taken at the threshold of the critical energy. The propagation of the new phase is dominated by the phase boundary between the old and the new nucleated phases (Arlt, 1996a,b, 1997). The propagation of the new phase is determined by so called kinetics relations. The completion of domain switching inside the domain requires a certain time period. The simulation of piezoelectric materials for quasi-static loading uses the assumption of complete domain switching inside the domains during each incremental loading step. When the loading is faster, the time between two successive loading steps is not adequate for a complete domain switching. Therefore, the overall electric displacement and strain versus electric field curves will change for cyclic loadings with increasing frequencies. For the propagation of the new phase, linear kinetics theory is used in this paper. Due to this theory, there is a critical time step (Δt_l) during the simulation. This limit time is the time, which the phase boundary needs to propagate completely through the domain whenever nucleation has been initiated. It is a material dependent parameter. If the time interval between two subsequent incremental steps is larger than this critical time period, the phase boundary can completely propagate through the domain, i.e. the switching process can be completed. If the time between two simulation steps is smaller than (Δt_l), then the switching process in the grain is not finished after one simulation step. For a simulation of the quasi-static case the time step in the simulation should be greater than (Δt_l). In the simulation the frequency of the applied electric field is given by

$$f_E = 1/T_E \tag{7}$$

where T_E is the period according to Fig. 7. The rate of change of the electric field ($E\cdot$) is given in (8) with the amplitude $\hat{E}$ of the electric field.

$$\dot{E} = \pm 2\hat{E}/(T/2) = \pm 4\hat{E}/T = \pm 4\hat{E}f_E \tag{8}$$

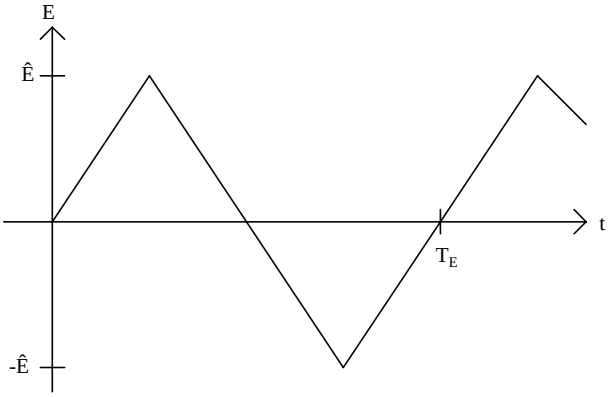

Fig. 7. Cyclic triangular electric field.

During the simulation the increments of the electric field ΔE between different loading steps are given explicitly. In Fig. 8 it can be seen that such an increment corresponds to a time step Δt_s for which

$$\Delta t_s = \Delta E/\dot{E} = \Delta E/(4\hat{E}f_E) \tag{9}$$

holds. This is the simulation time step.

Eq. (9) shows that the time step Δt_s is decreasing for increasing frequencies of the electric field. It has to be mentioned that ΔE and $\hat{E}$ is kept constant for the simulations. If Δt_s is smaller than Δt_l a domain which begins to change the direction of polarization does not change phase in the whole domain during one simulation period Δt_s. Instead, due to linear kinetics theory only a fraction ($\Delta t_s/\Delta t_l$) will have the new polarization direction. Especially, if Δt_s is very small due to a high frequency f_E, the domain switching process may take several simulation steps until the whole domain is switched. In order to determine the volume fraction ΔV of the switched part of the total volume V of the domain, the time Δt_p has to be determined, which has elapsed since the nucleation process begins.

$$\Delta V = V\frac{\Delta t_p}{\Delta t_l} \;\Rightarrow\; \frac{\Delta V}{V} = \frac{\Delta t_p}{\Delta t_l} \tag{10}$$

Note that Δt_p may be a multiple of Δt_s. If P_{0b} and P_{0a} denote the polarization together with the polarization direction of the whole domain before and after the switching process, then the polarization P_0 during the switching process after inserting the volume fractions is

$$P_0 = \left(1 - \frac{\Delta V}{V}\right)P_{0b} + \frac{\Delta V}{V}P_{0a} = \left(1 - \frac{\Delta t_p}{\Delta t_l}\right)P_{0b} + \frac{\Delta t_p}{\Delta t_l}P_{0a} \tag{11}$$

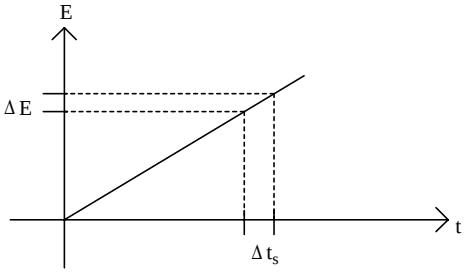

Fig. 8. Loading steps (ΔE, Δt_s).

As we discretize the time axis the polarization at one loading step after the beginning of the nucleation is

$$P_{01} = \left(1 - \frac{\Delta t_s}{\Delta t_l}\right) P_{0b} + \frac{\Delta t_s}{\Delta t_l} P_{0a} \tag{12}$$

As explained before, if the value of Δt_s is much lower than the value of Δt_l, the domain switching process may take many incremental steps to be completed. Due to this fact a number n is introduced for each domain, which has already started the switching process. This number denotes the number of time steps, which has already passed after the beginning of the switching process in the corresponding domain. For the nth time step after nucleation we obtain polarization during domain switching

$$\begin{aligned} P_{0(n)} &= \left(1 - \frac{(n-1)\Delta t_s}{\Delta t_l} - \frac{\Delta t_s}{\Delta t_l}\right) P_{0b} + \frac{(n-1)\Delta t_s}{\Delta t_l} P_{0a} + \frac{\Delta t_s}{\Delta t_l} P_{0a} \\ &= \left(1 - \frac{(n-1)\Delta t_s}{\Delta t_l}\right) P_{0b} + \frac{(n-1)\Delta t_s}{\Delta t_l} P_{0a} + \frac{\Delta t_s}{\Delta t_l}(P_{0a} - P_{0b}) = P_{0(n-1)} + \frac{\Delta t_s}{\Delta t_l}(P_{0a} - P_{0b}) \end{aligned} \tag{13}$$

This is repeated until

$$n\Delta t_s > \Delta t_l \tag{14}$$

where at the end of the switching $P_0 = P_{0a}$ is assumed. Eqs. (11)–(14) are derived in order to get a discretized linear kinetics model for our computer simulation program.

Simulations are performed for electric displacement versus electric field curves with different frequencies (0.01 Hz, 0.1 Hz, and 1 Hz) and amplitudes of the electric field (2 kV/mm, 1.5 kV/mm, and 1 kV/mm). These values are chosen in order to be able to compare the simulation results with experiments that were performed for the same values. The limit time (Δt_l) is taken to be 0.07 s. This means that 0.0018 Hz will be the maximum frequency for a quasi-static loading at an amplitude of 2 kV/mm for the electric field and increments of $\Delta E = 1$ V/mm. PIC 151 is taken as a sample piezoelectric material, which is also used in the experiments (Zhou, 2003). Therefore, the dielectric permittivity (ε) is taken to be 2.124×10^{-8} F/m and is assumed to be isotropic. The spontaneous polarization P_0 and the critical value of the spontaneous polarization (P_c) are chosen to be 0.4 C/m^2. A value of 1 kV/mm is assumed for the coercive electric field level. The piezoelectric constant is considered to behave transversely isotropic. The value of the piezoelectric constant in loading direction is taken as 0.45×10^{-9} (m/V).

For better explanation of the model, we also inserted one additional example for the case of "$\Delta t_s < \Delta t_l$". If domain switching criteria (Eq. (5)) fulfills, new element nucleates and begins to propagate with respect to linear kinetics model. According to the applied electric field with a frequency of 1 Hz, Δt_s is 0.00125. Therefore n equals $\Delta t_l/\Delta t_s = 0.07/0.00125 = 56$. That means 56 steps are required for corresponding element to finish its domain switching. So, polarization change for each step for that particular element is actually the polarization change for quasi-static loading divided by 56. For every step, in order to have simple calculation, we used Eq. (13). The domain switching continues during the range of 56 kV/mm electric field loading range since each electrical loading step (ΔE) is 1 kV/mm.

Simulation results are shown in Figs. 9–11 for the case in which probability for domain switching (Seemann et al., 2004) is not taken into account. It is experimentally found that the coercive electric field is dependent on the frequency of the loading which apparently can also be seen in our simulated curves. The coercive electric field is increasing when the frequency is increased from 0.01 Hz to 1 Hz. This can even be seen in Fig. 11 for an amplitude of 1 kV/mm electric field, which corresponds to the coercive electric field level chosen above. Another important observation for these curves under cyclic loading with a high frequency is that the electric displacement is increasing even for a decreasing electric field during unloading

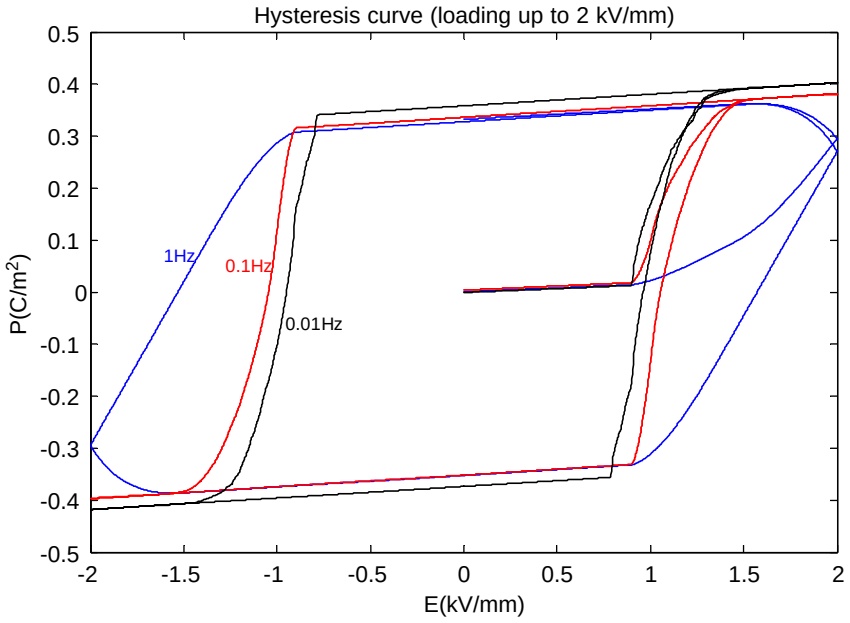

Fig. 9. Hysteresis curves without probabilistic approach, $\hat{E} = 2$ kV/mm.

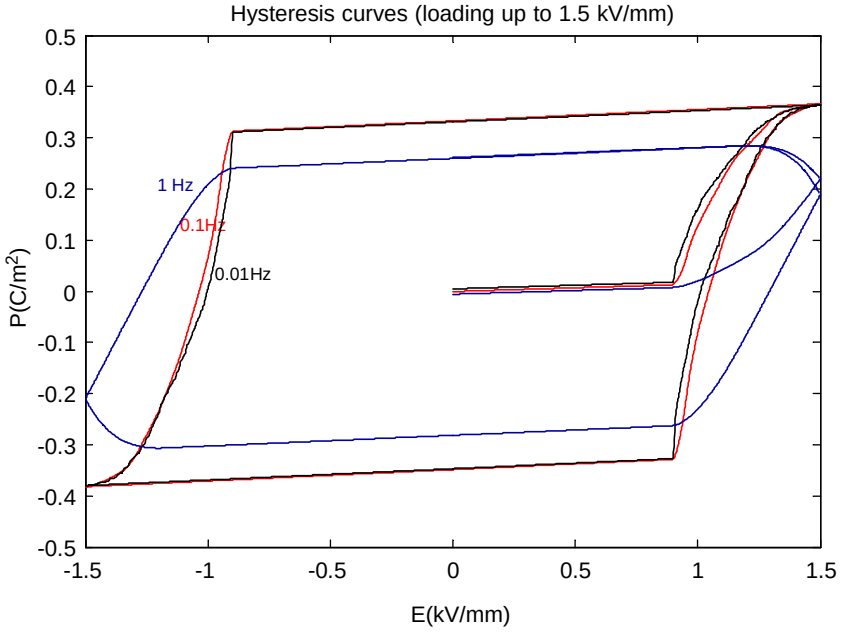

Fig. 10. Hysteresis curves without probabilistic approach, $\hat{E} = 1.5$ kV/mm.

provided that the actual electric field is larger than the coercive field. For a cyclic loading with a frequency of 1 Hz and an amplitude of the electric field of 2 and 1.5 kV/mm, the electric displacement cannot reach

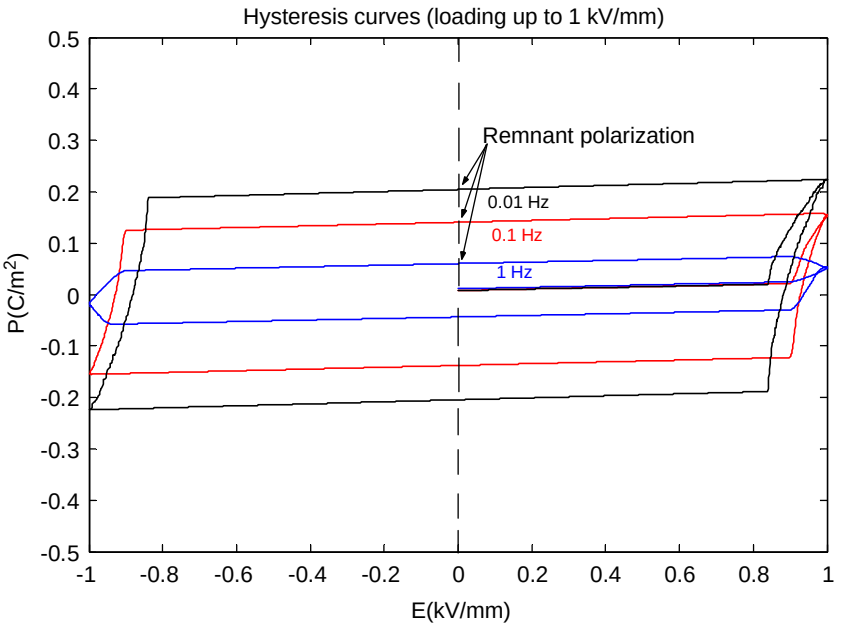

Fig. 11. Hysteresis curves without probabilistic approach, $\hat{E} = 1$ kV/mm, leading to different remnant polarizations for different loading frequencies.

the saturation level which corresponds to a quasi-static loading. Instead, the electric displacement is increasing for a decreasing electric field as far as the applied electric field is larger than the coercive field. The remnant polarization, which is the polarization at zero electric field, is not changing too much for frequencies of 0.01 Hz and 0.1 Hz and amplitudes of the electric field of 2 and 1.5 kV/mm. Because all possible domain switchings are approximately completed for such amplitudes and frequencies of the applied loading. On the other hand, as shown in Fig. 11 the saturation polarization cannot be reached and the frequency of the loading has a strong influence on the remnant polarization, if the amplitude of the applied electric field is near the coercive field. The remnant polarization is decreasing if the frequency of the cyclic loading is increasing, see Fig. 11.

As explained before, in piezoceramic materials it is observed that there occur some domain switchings below the coercive field. In previous publications this was modeled by a probability criterion (Delibas et al., 2004). Such a probability was also implemented for the simulation of the rate dependent characteristics of PIC 151 using a micromechanical model, for which the nucleation starts with a certain probability depending on the energy difference during switching. Figs. 12–14 show electric displacement versus electric field curves again for a cyclic loading with different amplitudes and frequencies. For the simulations a fourth order probability function is implemented. As it can be seen in the figures, the curves are smoother, especially in the range near the coercive field level compared to the ones that were simulated without adding the probability criterion. Therefore, these curves correspond better to the experimental ones for electric field values near the coercive field. All other important properties already explained for the simulation without probability criterion are observed as well: decreasing of the electric field level for zero overall polarization with decreasing frequency of loading. Other phenomena are the increase of the remnant polarization for smaller amplitudes of the loading and decreasing frequencies or continuing further domain switchings during the early unloading stage as far as the cyclic electric field is above the coercive field.

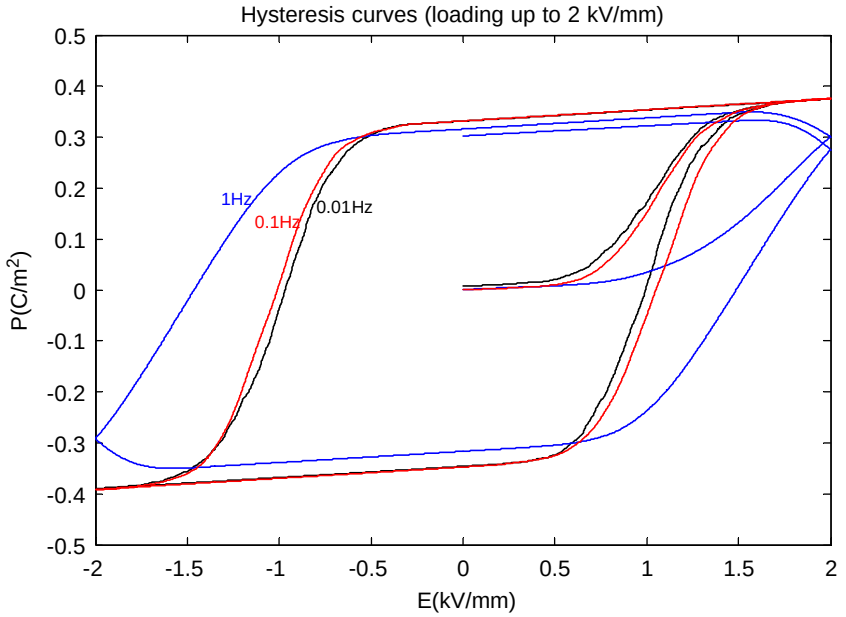

Fig. 12. Hysteresis curves with fourth order polynomial for the probability function, $\hat{E} = 2$ kV/mm.

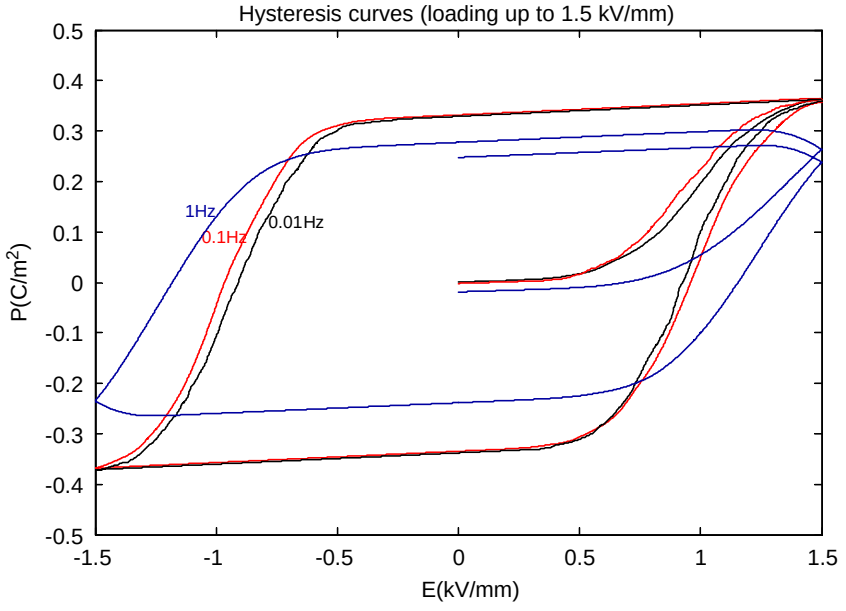

Fig. 13. Hysteresis curves with fourth order polynomial for the probability function, $\hat{E} = 1.5$ kV/mm.

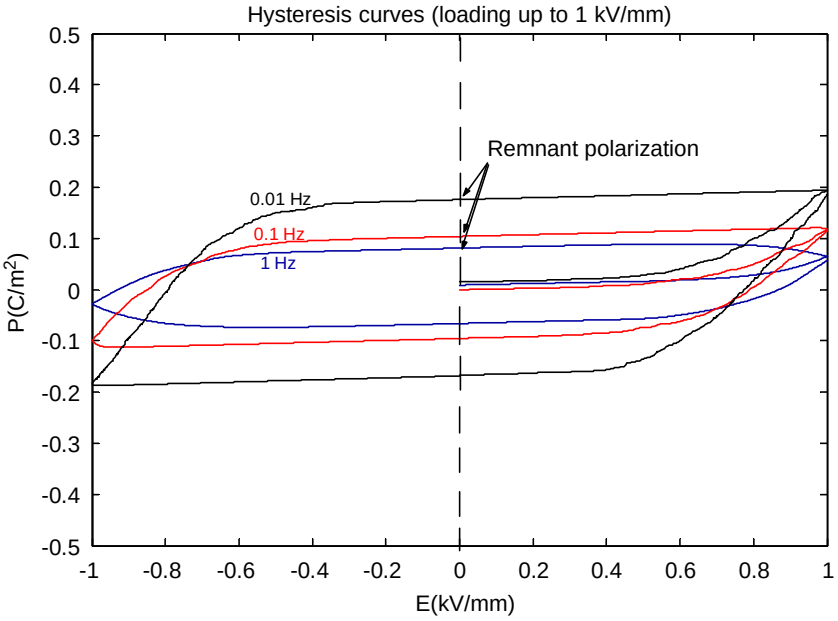

Fig. 14. Hysteresis curves with fourth order polynomial for the probability function, $\hat{E} = 1$ kV/mm.

5. Concluding remarks

Nonlinear properties of perovskite type tetragonal ferroelectric and piezoelectric materials under uni-axial, high, cyclic electric loading with various loading frequencies and amplitudes are simulated by using a micromechanical model. For the simulation material parameters of PIC 151 PZT ceramic are used. A piezoelectric linear constitutive model and a nonlinear domain switching with probability functions have been applied in the model. A linear kinetics model is used for the propagation of the new phase during the domain switching process. The hysteresis curves simulated with a probabilistic approach do better match to the experimental hysteresis curves than simulation results in which probability functions have not been used. The model follows the basic characteristics of piezoelectric materials for different loading rates and amplitudes such as the change of the coercive field and the change of the remnant polarization.

Acknowledgment

The authors would like to acknowledge the financial support of Deutsche Forschungsgemeinschaft (DFG) for the research.

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Further reading

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