

A Crystal Plasticity Model for Ferroelectrics

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A micromechanical constitutive model for a ferroelectric crystal of the perovskite type is developed. The model uses a crystal plasticity formulation in which transformation between crystal variants is treated in the same way as slip on a crystal slip system. The volume fractions of the crystal variants are used as variables which represent the state of the crystal. The dielectric hysteresis and “butterfly” hysteresis of a single crystal under low frequency cyclic electric fields are presented. Estimates of the response of a ferroelectric polycrystal to the same loading are calculated using a self-consistent mean-field homogenization scheme.

Keywords: ferroelectrics; models; micromechanics; self-consistent modelling; polycrystals; hysteresis

INTRODUCTION

Several micromechanical models for the behaviour of ferroelectric single crystals and polycrystals have been developed in recent years^[1-4]. Whilst these models have been able to capture the non-linear hysteresis of ferroelectric polycrystals, they have generally required the presence of one or more non-physical parameters to achieve realistic results. In this paper, a preliminary investigation is made of a new model based on an abstraction of the microstructural features (ferroelectric domains and grains) found in ferroelectric polycrystals. All aspects of this model may be justified on a physical basis. It applies to isothermal loading of ferroelectrics with stresses and electric fields, in low frequency (quasi-static) conditions.

The movement of a ferroelectric domain wall gives rise to changes in remanent strain and electric polarization. These changes arise through

the transformation of material from one crystal variant into another twin crystal variant. The model described here is based on the fundamental observation that the increments in strain and polarization caused by domain wall movement may be treated as if they were caused by slip on crystal slip systems. This observation allows established methods of crystal plasticity theory to be applied to ferroelectrics. The approach relies on two steps: Firstly, constitutive laws governing the behaviour of a single crystal are postulated. Secondly, appropriate crystal slip systems are identified; the volume fractions of each of the crystal variants present are used as variables which represent the current state of the crystal. The instantaneous tangent properties (elastic modulus, piezoelectric coefficients, and dielectric permittivity) of a ferroelectric single crystal can then be calculated.

Using the instantaneous tangent properties of a single crystal, the response of the crystal to any path of loading in electric field and stress space can be simulated. The loading path is broken up into small steps, and at each step, the tangent properties of the crystal are calculated. The tangent properties are then used to relate the small step in electric field and stress to the corresponding changes in strain and electric polarization. These changes include both a linear piezoelectric part, and a part arising from ferroelectric domain wall movement.

The response of a single crystal to cycling electric field and stress loading is predicted. Using a polycrystal homogenization scheme it is possible to estimate the behaviour of a polycrystal consisting of many, randomly oriented crystals. Predictions of polycrystal behaviour using an incremental self-consistent homogenization scheme are presented. This averaging scheme uses a solution to the Eshelby inclusion problem in a ferroelectric medium to relate loads applied at the boundaries of a polycrystal to the local stress and electric field conditions experienced by each crystal. This allows for the effects of residual stress fields and internal bias fields to be accounted for in a consistent way. Details of the self-consistent homogenization scheme are given elsewhere^[5, 6].

Definitions and notation

Notation with subscripted indices is used to represent Cartesian tensors throughout, with summation over repeated indices. The symbol δ_{ij} is used to represent the Kronecker delta which takes on the value unity when $i = j$ and zero otherwise. The stress field and electric field in the single crystal are assumed to be uniform and represented by σ_{ij} and E_i respectively. The total strain and electric displacement in the single crystal, defined relative

to a zero initial state, will be represented by ϵ_{ij} and D_i respectively. The symbols ϵ_{ij}^p and D_i^p refer to the “plastic” or remanent parts of strain and electric displacement in a crystal, which would remain upon removal of stress and electric field. Small increments in all of these quantities are indicated using a dot, such that $\dot{\sigma}_{ij}$ is a small increment in the stress experienced by the single crystal. The analysis leads to a scheme for evaluating *instantaneous, tangent* properties L_{ijkl}^t , a_{kij}^t , b_{ikl}^t and χ_{ik}^t for a single crystal, defined by:

$$\begin{bmatrix} \dot{\sigma}_{ij} \\ \dot{E}_i \end{bmatrix} = \begin{bmatrix} L_{ijkl}^t & a_{kij}^t \\ b_{ikl}^t & \chi_{ik}^t \end{bmatrix} \begin{bmatrix} \dot{\epsilon}_{kl} \\ \dot{D}_k \end{bmatrix} \quad (1)$$

The reciprocity relation $a_{kij}^t \equiv b_{ikl}^t$ is not assumed.

SINGLE CRYSTAL MODEL

This section focuses on a single crystal with the perovskite-like tetragonal structure found in several ferroelectrics, which has six distinct crystal variants. The following form of constitutive law for a ferroelectric single crystal is postulated:

$$\begin{bmatrix} (\epsilon_{ij} - \epsilon_{ij}^p) \\ (D_i - D_i^p) \end{bmatrix} = \begin{bmatrix} M_{ijkl} & d_{kij} \\ d_{ikl} & \kappa_{ik}^\sigma \end{bmatrix} \begin{bmatrix} \sigma_{kl} \\ E_k \end{bmatrix} \quad (2)$$

This relates the linear parts of strain and electric polarization to the applied stress and electric field loading. M_{ijkl} is the current elastic compliance, d_{kij} the current piezoelectric tensor, and κ_{ik}^σ the current dielectric permittivity at constant stress. The following assumptions are made:

1. that the stress σ_{ij} and electric field E_i are uniform throughout the crystal.
2. that the overall strain ϵ_{ij} and electric polarization D_i of the crystal take on the volume average values of the corresponding quantities, as illustrated in figure 1.
3. that the volume-fraction occupied by the domain walls which separate the variants is negligible.

Each variant within a crystal has its own set of linear properties. Let the variants be numbered $I = 1, 6$. The symbols M_{ijkl}^I , d_{ikl}^I and κ_{ik}^I denote the linear properties of the I th variant in the crystal, and are constants.

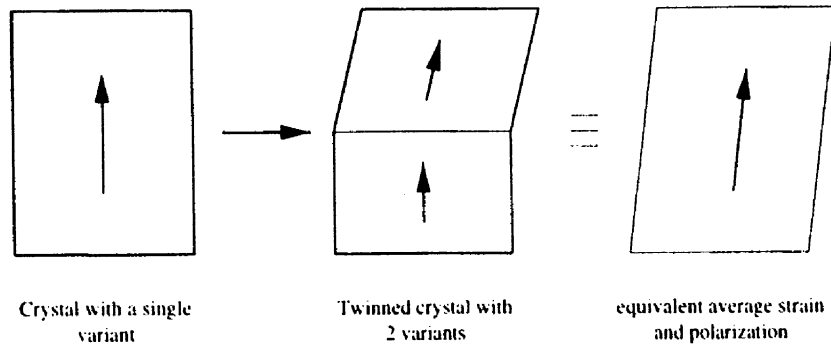


Figure 1: Average strain and polarization assumptions in a crystal

Let the volume-fraction of the I th variant be denoted by v^I , such that $\sum_I v^I = 1$. From assumptions 1 and 2 above, it follows that the overall linear properties of the crystal are given by:

$$M_{ijkl} = \sum_I v^I M_{ijkl}^I \quad (3)$$

$$d_{ikl} = \sum_I v^I d_{ikl}^I \quad (4)$$

$$\kappa_{ik}^\sigma = \sum_I v^I \kappa_{ik}^{I\sigma} \quad (5)$$

Kinematics of twinning transformations

Consider a transformation between different crystal variants. Each of six variants may transform into any of five other variants giving a total of 30 transformations. Each transformation is characterized by a transformation strain and a transformation electrical polarization. Here, transformation kinematics will be analysed as if they were the result of slip on crystal slip systems.

Typical examples of a 90° and a 180° transformation in a tetragonal crystal are shown in figure 2, where the tetragonality is exaggerated for clarity. Concentrating on the 90° transformation in figure 2, the strain associated with complete transformation may be represented as a matrix ϵ in the Cartesian axes 1,2 and 3 of figure 2, given by

$$\epsilon = \epsilon \begin{bmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & 0 \end{bmatrix} \quad (6)$$

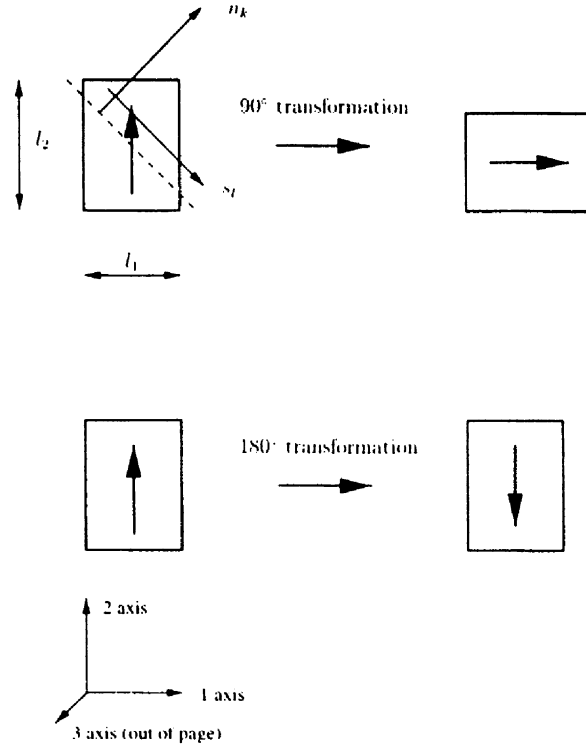


Figure 2: Transformations and slip planes in tetragonal crystals

where $\epsilon = (l_2 - l_1)/l_1$. The corresponding shear strain γ , slip plane normal n_k and slip direction s_l are given by:

$$\gamma = 2\epsilon, \quad \mathbf{n} = \frac{1}{\sqrt{2}} \begin{bmatrix} 1 \\ 1 \\ 0 \end{bmatrix}, \quad \mathbf{s} = \frac{1}{\sqrt{2}} \begin{bmatrix} 1 \\ -1 \\ 0 \end{bmatrix} \quad (7)$$

The transformation strain may be written as $\gamma\mu_{kl}$ where the components of orientation tensor $\mu_{kl} = (n_k s_l + s_k n_l)/2$ are:

$$\boldsymbol{\mu} = \begin{bmatrix} 1/2 & 0 & 0 \\ 0 & -1/2 & 0 \\ 0 & 0 & 0 \end{bmatrix} \quad (8)$$

Note that the same transformation strain could be achieved with $\mathbf{n} = [-1 \ 1 \ 0]^T/\sqrt{2}$ and $\mathbf{s} = [-1 \ -1 \ 0]^T/\sqrt{2}$, but this would give rise to a different final value of polarization. By analogy with the transformation strain, the transformation polarization change may be written as $D_i = Ds_i$, where D is the magnitude of the polarization change.

180° transformations produce no strain change, but a complete reversal of polarization. The 180° transformation shown in figure 2 may be represented formally by $\mathbf{n} = [0 \ 0 \ 0]^T$, $\mathbf{s} = [0 \ -1 \ 0]^T$, giving $\mu_{kl} = 0$

and $D_i = \sqrt{2}Ds_i$. Note that the *magnitude* of the polarization change is a factor $\sqrt{2}$ greater than that of the 90° transformation. The fact that different transformations have different magnitudes of strain and polarization change associated with them can be incorporated into the model in a general way by numbering the different transformations $\alpha = 1, \dots, 30$ and having a set of constants γ^α, D^α which represent the shear strain and polarization change associated with the α th transformation.

Next consider an incremental transformation from one variant to another, with slip system α . Let f^α denote the incremental volume-fraction of material transformed. The corresponding plastic strain increment is $f^\alpha \mu_{ij}^\alpha \gamma^\alpha$. Similarly, there is an increment in plastic polarization $f^\alpha s_i^\alpha D^\alpha$. If several slip systems are active simultaneously, then the plastic strain and polarization increments may be computed by summing over the set of active slip systems:

$$\dot{\epsilon}_{ij}^p = \sum_{\alpha} f^\alpha \mu_{ij}^\alpha \gamma^\alpha \quad (9)$$

$$\dot{D}_i^p = \sum_{\alpha} f^\alpha s_i^\alpha D^\alpha \quad (10)$$

Note that the quantities f^α are a set of kinematic variables which govern the increments in both plastic strain and plastic polarization: $\dot{\epsilon}_{ij}^p$ and \dot{D}_i^p are not independent quantities in ferroelectric crystals.

Similarly, increments in the *linear* coefficients $\dot{M}_{ijkl}, \dot{d}_{kij}$ and $\dot{\kappa}_{ik}^\sigma$ may be computed in terms of the kinematic variables f^α . If slip system α depletes a variant I and produces a variant J then it produces an increment \dot{M}_{ijkl}^α of the form $f^\alpha (M_{ijkl}^J - M_{ijkl}^I)$. A "connectivity matrix" $A^{I\alpha}$ may be defined such that $A^{I\alpha} = 1$ if slip system α produces variant I , $A^{I\alpha} = -1$ if slip system α depletes variant I , and $A^{I\alpha} = 0$ in all other cases. Then the increment \dot{M}_{ijkl}^α due to slip on system α is given by $\sum_{I=1,6} A^{I\alpha} f^\alpha M_{ijkl}^I$. Similar relations are valid for \dot{d}_{kij} and $\dot{\kappa}_{ik}^\sigma$. Summing over all active slip systems gives:

$$\dot{M}_{ijkl} = \sum_{I=1}^6 \sum_{\alpha} A^{I\alpha} f^\alpha M_{ijkl}^I \quad (11)$$

$$\dot{d}_{kij} = \sum_{I=1}^6 \sum_{\alpha} A^{I\alpha} f^\alpha d_{kij}^I \quad (12)$$

$$\dot{\kappa}_{ik}^\sigma = \sum_{I=1}^6 \sum_{\alpha} A^{I\alpha} f^\alpha \kappa_{ik}^I \quad (13)$$

These equations could equally have been derived by differentiating equations 3, 4 and 5, noting that $\dot{v}^I = \sum_{\alpha} A^{I\alpha} f^\alpha$.

Yield criterion

A yield criterion is required to define the combinations of stress σ_{ij} and electric field E_i which can give rise to transformation on a particular slip system α . The following yield criterion is proposed:

$$\sigma_{ij}\mu_{ij}^{\alpha}\dot{\gamma}^{\alpha} + E_i s_i^{\alpha} \dot{D}^{\alpha} = G^{\alpha} \quad (14)$$

Here G^{α} is a dissipated energy per unit volume; it is the magnitude of the plastic dissipation for full switching on slip system α . Addition of the electrical and mechanical terms on the left hand side of equation 14 may be justified in the following way. Suppose that yield occurs at some combination of stress and electric field σ_{ij} , E_i , and that the entire volume of the crystal transforms from variant 1 to variant 2 by slip system α . The work w done per unit volume by the combined stress and electric field during transformation is:

$$w = \sigma_{ij}\mu_{ij}^{\alpha}\dot{\gamma}^{\alpha} + E_i s_i^{\alpha} \dot{D}^{\alpha} \quad (15)$$

which is the left hand side of the yield criterion. Equation 14 states that the work done in transforming the crystal from variant 1 to variant 2 is independent of the type of loading which causes the transformation. For present purposes, equation 14 is insufficient to specify whether or not a slip system is active. If the l th variant is depleted by a transformation, and has volume-fraction $v^l = 0$, then the transformation cannot occur. Consequently,

$$v^l > 0 \quad (16)$$

is also required for yield. A slip system which satisfies both equations 16 and 14 will be described as "potentially-active". For an active slip system:

$$\dot{f}^{\alpha} > 0 \quad (17)$$

Hardening

Hardening behaviour is included in this model to stabilize numerical computations of the increments $\dot{\sigma}_{ij}$, $\dot{\epsilon}_{ij}$, \dot{E}_i and \dot{D}_i . The hardening rate is chosen to be small enough to give several cycles of hysteresis without a noticeable increase in G^{α} . For simplicity, the hardening law adopted is independent hardening, with a constant, positive hardening rate h such that:

$$\dot{G}^{\alpha} = h\dot{f}^{\alpha} \quad (18)$$

Following Hill ^[7] a hardening matrix $h^{\alpha\beta}$ (or \mathbf{h}) is defined so that equation 18 may be rewritten as

$$\dot{G}^\alpha = \sum_\beta h^{\alpha\beta} \dot{f}^\beta \quad (19)$$

The chosen hardening law has $\mathbf{h} = h\mathbf{I}$ where \mathbf{I} is the identity matrix. Note that, since $\dot{f}^\beta \geq 0$ the hardening rate is never negative.

The specification of equations 2 to 5 and equations 9 to 19 suffices to define the tangent properties of equation 1 for a crystal, given a set of potentially active systems. The method is analogous to that used by Hutchinson ^[8] to find the tangent properties of metal crystals; details are given separately ^[6].

SINGLE CRYSTAL AND POLYCRYSTAL RESPONSES

Consider a tetragonal ferroelectric material in which the elastic modulus, piezoelectric tensor and dielectric permittivity of each crystal variant are given by:

$$L_{ijkl} = \lambda\delta_{ij}\delta_{kl} + \mu(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) \quad (20)$$

$$d_{kij} = p_k(3p_i p_j - \delta_{ij})d/2 \quad (21)$$

$$\kappa_{ik}^\sigma = \kappa\delta_{ik} \quad (22)$$

Here λ and μ are the Lamé elastic parameters (μ being the shear modulus of the crystal), d is the axial piezoelectric coefficient and κ is the dielectric permittivity; p_k represents a unit vector parallel to the remanent polarization vector of the crystal variant. This material has isotropic elastic and dielectric behaviour, and a simple form of piezoelectricity. Let the starting state for the crystal be that in which there is an equal volume fraction of each of the six crystal variants:

$$v^I = 1/6 \quad (I = 1\dots 6) \quad (23)$$

The orientation of the crystal is chosen such that its crystallographic axes lie parallel to the +1, +2 and +3 axes of the coordinate system in which the calculations are done. For computation, it is convenient to normalize all physical quantities in such a way that their values are of order unity. This can be achieved by choosing four normalizing quantities — a shear strain γ^0 to normalize all strains, a shear stress τ^0 to normalize stresses, an electric field E^0 and an electric polarization D^0 . Here, a normalization

is chosen such that in a plane strain, pure shear test in the 1-2 plane, yield occurs when $\sigma_{12}/\tau^0 = 1$ and $2\epsilon_{12}/\gamma^0 = 1$. In pure electric field loading along the 1 axis, yield is chosen to occur at $E_1/E^0 = 1$ and $D_1/D^0 = 1$. The model is chosen to have "lock-up" strains and polarizations about 5 times the values at yield, so that an extensive plastic regime exists. Consequently, the values of γ^α and D^α on various slip systems α are as follows:

$$\begin{aligned} 180^\circ \text{ slip systems: } & \gamma^\alpha = 0 & D^\alpha &= 5D^0 \\ 90^\circ \text{ slip systems: } & \gamma^\alpha = 5\gamma^0 & D^\alpha &= 5D^0/\sqrt{2} \end{aligned}$$

Note that the kinematics of the slip systems demands that for a 180° slip system $\gamma^\alpha = 0$, and that $D^\alpha(180^\circ) = \sqrt{2}D^\alpha(90^\circ)$.

Now consider loading this crystal with slowly cycling electric field E_1 as shown in figure 3. A normalized graph of D_1 against E_1 in pure electric field loading is shown in figure 4. The corresponding graph of strain ϵ_{11} against electric field is shown in figure 5. In both figures the loading is one and a half full cycles of electric field E_1 of amplitude $4E^0$, with the loading initially proceeding in the positive 1 direction. The initial transient response is labelled A-B-C-D in figure 4.

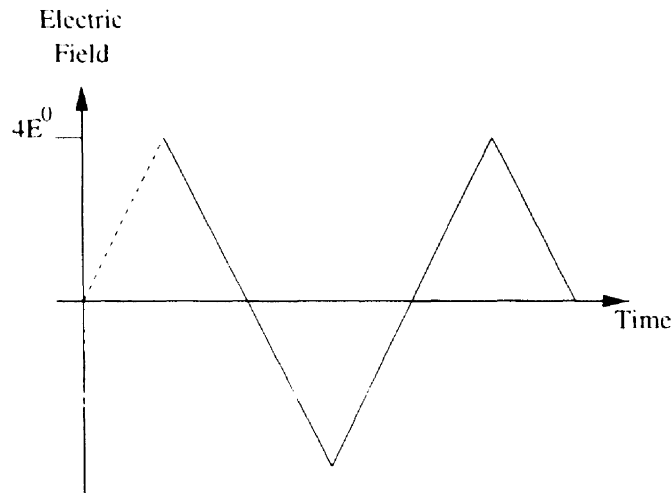


Figure 3: Electric field loading path

The model predicts a simple form of dielectric hysteresis and butterfly hysteresis in the single crystal. After one half cycle of loading, the single crystal settles into a stable cyclic response. Ferroelectric switching occurs at a constant magnitude of electric field, giving rise to sudden "switching" behaviour in the crystal, and sharp corners on the hysteresis curves.

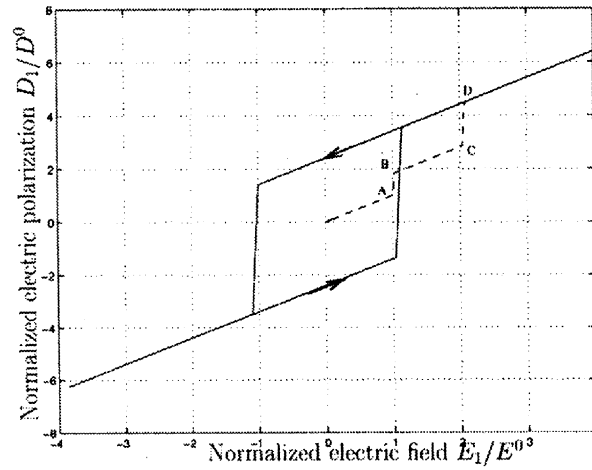


Figure 4: Simulated electric polarization D_1/D^0 versus electric field E_1/E^0 for a single crystal

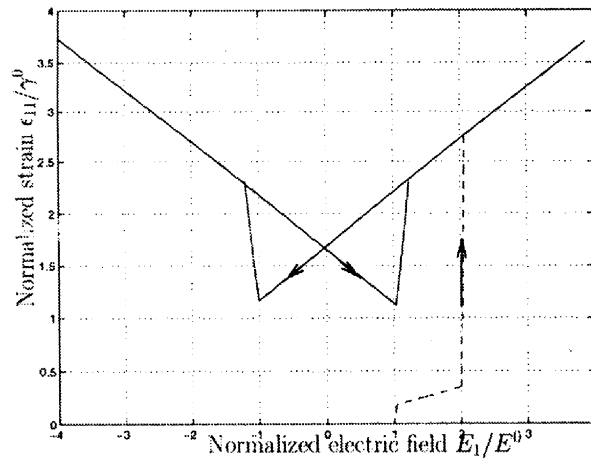


Figure 5: Simulated strain ϵ_{11}/γ^0 versus electric field E_1/E^0 for a single crystal with simple piezoelectricity

Ferroelectric polycrystal

Using a polycrystal homogenization scheme it is possible to estimate the response of a ferroelectric polycrystal to the electric field loading shown in figure 3. The homogenization scheme used here is the incremental self-consistent scheme. Each crystal in the polycrystal is modelled as a spherical inclusion embedded in an infinite matrix of surrounding material which has the current tangent properties of the whole polycrystal. Solutions of the Eshelby inclusion problem for a ferroelectric medium are needed to relate the local stress and electric field conditions in each crystal to the remote applied loads on the polycrystal. Expressions derived by Deeg and others^[9, 10] allow for numerical solution of the constrained inclusion problem. The incremental self-consistent scheme is described in detail elsewhere^[5, 6]. This paper gives some results calculated using the self-consistent scheme to illustrate the differences between the response of the single crystal and that of the polycrystal.

Let the remote stress and electric fields in the polycrystal be $\bar{\sigma}_{ij}$ and \bar{E}_i respectively. Similarly, let the total strain and electric displacement of the polycrystal be $\bar{\epsilon}_{ij}$ and \bar{D}_i . Note that $\bar{\sigma}_{ij}$, \bar{E}_i , $\bar{\epsilon}_{ij}$, and \bar{D}_i may be written as the volume averages of the corresponding quantities σ_{ij} , E_i , ϵ_{ij} and D_i over the set of crystals which make up the polycrystal. The self consistent scheme produces an estimate of the overall tangent properties L_{ijkl}^0 , a_{kij}^0 , b_{ikl}^0 and χ_{ik}^0 defined by:

$$\begin{bmatrix} \dot{\bar{\sigma}}_{ij} \\ \bar{E}_i \end{bmatrix} = \begin{bmatrix} L_{ijkl}^0 & a_{kij}^0 \\ b_{ikl}^0 & \chi_{ik}^0 \end{bmatrix} \begin{bmatrix} \dot{\bar{\epsilon}}_{kl} \\ \bar{D}_k \end{bmatrix} \quad (24)$$

Using the overall tangent properties, the response to any path of loading applied to the polycrystal can be simulated by breaking up the path into a series of short steps. At each step, the overall tangent properties of the polycrystal are estimated, and the increments $\bar{\epsilon}_{ij}$ and \bar{D}_i are calculated by inverting equation 24.

The response of a polycrystal consisting of 100 randomly oriented crystals with the properties defined by equations 20, 21 and 22 was estimated using the incremental self-consistent scheme. The electrical loading was as shown in figure 3, except that five full cycles were applied. The resulting hysteresis in electric polarization versus electric field is shown in figure 6. The corresponding butterfly hysteresis is shown in figure 7.

The dielectric hysteresis and butterfly hysteresis for the polycrystal show a smooth transition from linear behaviour (without ferroelectric transformation) to non-linear behaviour (with ferroelectric transformation). This contrasts with the single crystal simulations which show sharp corners at

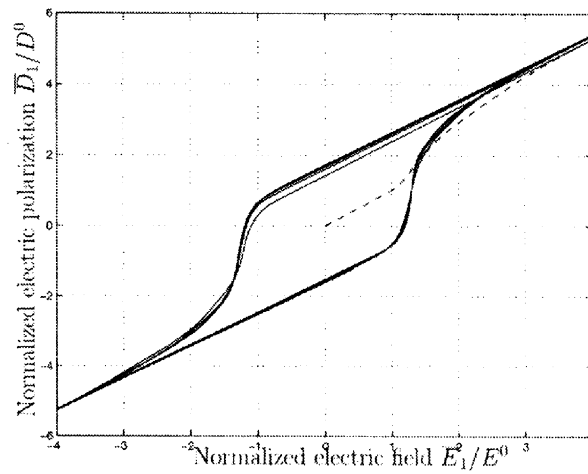


Figure 6: Simulated electric polarization \bar{D}_1/D^0 versus electric field \bar{E}_1/E^0 for a polycrystal with piezoelectricity

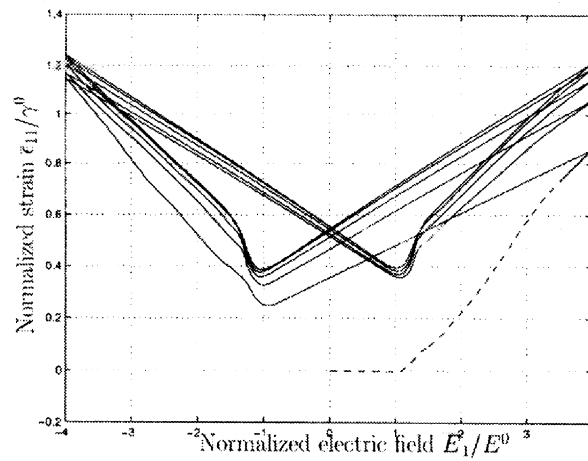


Figure 7: Simulated strain $\bar{\epsilon}_{11}/\gamma^0$ versus electric field \bar{E}_1/E^0 for a polycrystal with piezoelectricity

the onset of non-linearity. In the polycrystal, each crystal transforms gradually, over a range of values of the remotely applied load \bar{E}_i . When a crystal begins to transform, it produces remanent strain and polarization states which are different from its surroundings. This gives rise to local residual stresses and electric fields which oppose the transformation. The result is that transformation is spread over a range of values of \bar{E}_i ; domain walls sweep gradually through the crystal as the remote loading is increased.

The presence of residual stresses and electric fields gives rise to a memory effect which means that the response of the polycrystal is strongly dependent on its loading history. The butterfly hysteresis curve settles into a stable shape only after several full cycles of applied loading. The stable shapes of the hysteresis curves are qualitatively similar to those found in ferroelectric polycrystals.

CONCLUSION

A crystal plasticity approach to modelling ferroelectric materials has been developed. The basis of this approach is the observation that the remanent strains and polarizations produced by ferroelectric switching may be treated as if they were slip increments on crystal slip systems. Expressions for the instantaneous tangent properties of a ferroelectric crystal following a simple constitutive law have been derived. The incremental self-consistent homogenization scheme has been used to estimate the response of a polycrystal composed of randomly oriented single crystals following this constitutive model. The results show smooth dielectric and butterfly hysteresis loops, similar to those observed in ferroelectric polycrystals of tetragonal perovskite structure. The constitutive model may thus have applications in modelling the isothermal behaviour of ferroelectrics under low frequency loading with stresses and electric fields.

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