A Domain Wall Model for Ferroelectric Hysteresis

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Abstract

This paper summarizes a mathematical model for characterizing hysteresis in ferroelectric materials. The model is based on the quantification of energy required to bend and translate domain walls and is developed in two steps. In the first, the underlying anhysteretic polarization is quantified through constitutive equations derived using Boltzmann statistics. Three anhysteretic models are considered including the Langevin and Ising spin relations as well as a third formulation which combines attributes of the other two. Hysteresis is then incorporated through the consideration of domain wall motion and the quantification of energy losses due to inherent inclusions or pinning sites within the material. This yields a model analogous to that developed by Jiles and Atherton for ferromagnetic materials. The viability of the model is illustrated through comparison with experimental data from a PMN-PT-BT actuator operating at a temperature within the ferroelectric regime.

Keywords: Domain wall model, ferroelectric hysteresis

1. Introduction

In this paper, we address the modeling of dielectric hysteresis in certain ferroelectric and relaxor ferroelectric materials. As motivation, we consider the performance of lead zirconate titanate (PZT) at high applied field levels [1] or relaxor ferroelectrics such as PMN-PT-BT at temperatures well below the Curie point [2]. In both cases, the materials exhibit significant hysteresis in the relation between the applied field and resulting polarization. While the hysteresis can be reduced in some cases through the choice of state variables and operating conditions, accurate, efficient and robust modeling techniques are required to attain the full potential of these materials in high performance actuator design.

The nature of hysteresis models for ferroelectric materials can be roughly categorized as microscopic, semi-macroscopic and macroscopic with derivations based on either quantum mechanical, magnetoelastic or thermodynamic relations, or phenomenological principles. Microscopic theories for ferroelectric materials generally address domain switching processes. The mechanisms for these processes are attributed to the nucleation of domains as well as domain wall motion in response to an applied field. For simple stoichiometries, microscopic models are theoretically plausible (e.g., see [3]). For the crystalline conditions encounted in present actuator designs, however, it is difficult to construct tractable microscopic models which accommodate attributes such as grain boundaries and intergranular stresses as well as the polycrystalline nature of the materials. Moreover, microscopic models are often difficult to employ in control design due to the large number of required states and parameters.

In some cases, these difficulties can be avoided through the application of phenomenological techniques to attain a macroscopic characterization of the materials [4, 5, 6, 7]. In their typical formulation, Preisach models are an example of this approach. Such models are advantageous when the underlying physics is poorly understood or difficult to quantify. Due to their nature, however, it is difficult to employ known physics or physical measurements to directly construct phenomenological models or update them to accommodate changing operating conditions. For many regimes, accurate characterization in this manner also requires a large number of parameters in the model which complicates both model development and control design.

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Semi-macroscopic models are typically derived using a combination of magnetoelastic, thermodynamic and phenomenological principles (e.g., see [8]). The goal in this approach is to employ energy-based relations to the extent possible so that known physics can be used to facilitate model construction and updating. Phenomenological principles contribute to aspects of the constitutive equations and the development of macroscopic averages of microscopic properties such as the intergranular stresses and domain wall pinning site energies. We employ such a semi-macroscopic approach in this paper.

This hysteresis model is based upon the quantification of the domain wall energy in the material. For an ideal material which is free from inclusions or second-phase materials, domain wall movement is unimpeded and the relation between applied stresses, electric fields and temperatures, and the resulting hysteresis-free or anhysteretic polarization is modeled by nonlinear constitutive equations. As detailed in [2], appropriate constitutive relations are derived through thermodynamic theory combined with phenomenological relations such as Hooke's law. In actual materials, however, domain wall movement is impeded by inherent inclusions within the material. The model presented here quantifies the resulting hysteresis through consideration of the energy required to translate domain walls and break pinning sites. An 'average' measure of this energy is employed to obtain a model which requires the identification of only five parameters. Due to the small number and physical nature of certain parameters (e.g., the saturation polarization P_s), the model is easily constructed and updated. We note that this model is the ferroelectric analogue of that developed by Jiles and Atherton for ferromagnetic materials [9]. We also point out that while certain ferroelectric actuator materials provide the motivation, the model is general in nature and can be applied to a variety of ferroelectric materials.

Certain aspects of the constitutive theory of Hom and Shankar are summarized in Section 2. The domain wall theory contributing to both reversible and irreversible changes in polarization is presented in Section 3. In the final section, the viability of the model is illustrated through comparison with experimental data from a PMN-PT-BT actuator.

2. Constitutive Theory for Ferroelectric Materials

In this section, we summarize three techniques for modeling the relation between an input electric field and the resulting anhysteretic (hysteresis free) polarization. When combined with mechanical relations for the material, this provides nonlinear constitutive equations which are valid when hysteresis is negligible. Hysteresis effects are then included in the next section through the quantification of energy losses due to pinning sites in the material.

As detailed in [2, 10, 11], the dielectric behavior of ferroelectric materials can be modeled by employing Boltzmann statistics to quantify the probability of dipoles occupying certain energy states. The three models presented here differ only in the assumptions made concerning the dipole orientations and behavior in the material. To describe these models, we consider a ferroelectric crystal as a lattice of cells, where each cell possesses a permanent dipole moment with an associated direction. Collections of neighboring cells having the same polarization then form the domain structure of the crystal.

The first model is obtained under the assumption that the material is isotropic and the orientation of cells can be in any direction. With P_s, T, T_c and E_0 respectively denoting the saturation polarization, temperature, Curie temperature, and a scaling electric field, the relation between the applied field E and the polarization P is modeled by the Langevin relation

$$P = P_s \left[\coth\left(\frac{3T_c E}{E_0 T} \right) - \frac{E_0 T}{3T_c E} \right].$$ (1)

We note that an analogous Langevin formulation is commonly employed in ferromagnetic applications.

The second model which we consider is the Ising spin model which is derived under the assumption that only two dipole orientation are possible in one cell: one in the direction of the applied field and the other in the opposite direction. The resulting anhysteretic relation is

$$P = P_s \tanh\left(\frac{T_c E}{E_0 T} \right).$$ (2)

The final model combined attributes of the Langevin and Ising spin model in that it is based on the assumption that grains within the material are randomly distributed but that dipoles within each cell have only two possible

orientations. The application of Boltzmann statistics in this case yields the relation

$$P = P_s \left\{ \frac{2E_0 T}{3ET_c} \left[\log\left(1 + e^{-3ET_c/E_0 T}\right) + \log\left(1 + e^{3ET_c/E_0 T}\right) \right] \right.$$

$$\left. + \frac{2}{9} \left(\frac{E_0 T}{ET_c}\right)^2 \left[\text{Li}\left(-e^{3ET_c/E_0 T}\right) - \text{Li}\left(-e^{-3ET_c/E_0 T}\right) \right] \right\} \tag{3}$$

where Li represents the dilogarithm function given by

$$\text{Li}(z) = \int_z^0 \frac{\log(1-x)}{x} \, dx \,.$$

Details concerning the derivation of this relation, which we denote the Preferred Orientation model, are provided in [11].

The relative behavior of the three models is illustrated in Figure 1. The Langevin model saturates the slowest since its cells have the most freedom in selecting orientations that are partially aligned with the electric field. The Ising Spin model saturates more quickly since the cells have very little freedom and must completely orient in the direction of the field while the Preferred Orientation model provides a compromise between the other two models. We note that the Langevin and Ising spin models have equivalent first-order terms but differ in the third and higher order terms.

The relations (1), (2) and (3) quantify the anhysteretic polarization in the absence of cell interactions. As detailed in [12, 13], these interactions are often significant, however, and can produce second-order phase transitions as well as domain nucleation and growth. To incorporate the coupling due to neighboring cells, we consider interaction fields E_e and D_e.

Under the assumption of fixed temperature and quasistatic operating conditions, the effective E field is taken to be

$$E_e = E + \alpha P + 2Q_{33} P\sigma \tag{4}$$

where $\alpha \equiv \frac{E_0}{P_s}$ and Q_{33}, σ respectively denote the longitudinal electrostrictive coupling coefficient and the applied stress. The term αP quantifies the field due to interdomain coupling while the term $2Q_{33}P\sigma$ quantifies field contributions due to stress inputs.
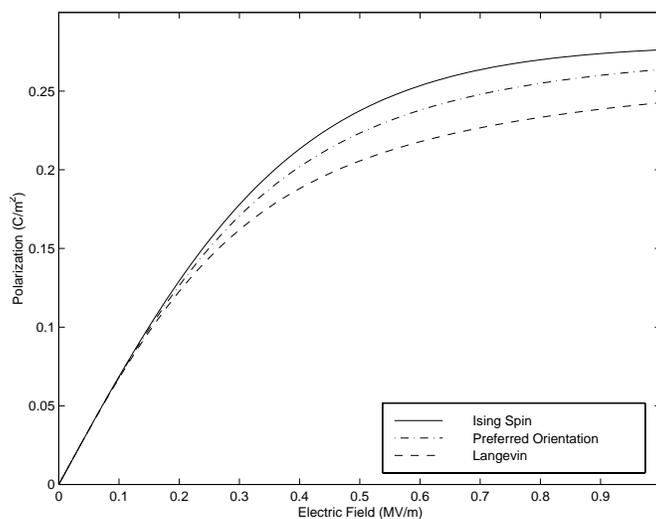
Figure 1. Ising spin, Preferred Orientation and Langevin models for the anhysteretic polarization with no cell interactions.

Because the effective field E_e incorporates the contributions due to interdomain coupling and applied stresses, we will employ it when describing the general models for the anhysteretic polarization and developing constitutive laws for the materials. In the case of constant temperature, the consideration of the effective field in (1) and (2) yields the Langevin and Ising spin relations

$$P = P_s \left[\coth \left(\frac{E_e}{a} \right) - \frac{a}{E_e} \right] \tag{5}$$

and

$$P = P_s \tanh \left(\frac{E_e}{a} \right) \tag{6}$$

where $a = \frac{E_0 T}{3 T_c}$ or $a = \frac{E_0 T}{T_c}$ are constants which must be estimated for a given material or device. The Preferred Orientation model is treated analogously.

As detailed in [11], the slope of the anhysteretic polarization curve is determined by the ratio of the parameters α and a. For most values of α and a, the resulting curves are single-valued which reflects the reversible nature of the anhysteretic polarization. However, for sufficiently small values of a, which correspond to temperatures $T < T_c$ for PMN, one obtains multiple solutions to the equations. While this produces curves which quantify certain aspects of hysteresis, the transition from one saturation region to the other is much steeper than that observed in typical ferroelectric applications. In Section 3, these single and multi-valued anhysteretic curves are incorporated in expressions characterizing energy losses at pinning sites in the material to provide a model which quantifies the hysteresis exhibited by the material.

To specify an appropriate D field, we first recall that for general fields, D is related to E and P through the equation

$$D = \epsilon_0 E + P, \tag{7}$$

where ϵ_0 denotes the free space permittivity, or the relation

$$D = \epsilon E \tag{8}$$

where ϵ is the permittivity of the substance. It should be noted that for ferroelectric materials, the permittivity ϵ is in general both nonlinear and multivalued due to hysteresis. For such materials, the relation (8) has limited value as a constitutive relation until ϵ is characterized. When quantifying hysteresis losses in the material, we employ the interaction field

$$D_e = \epsilon_0 \left(E + \alpha P + 2 Q_{33} P \sigma \right). \tag{9}$$

This definition is analogous to the expression $B_e = \mu_0 (H + \alpha M) = \mu_0 H_e$ employed for ferromagnetic materials (e.g., see [14, page 1266] along with [9, 15, 16]) where H_e and B_e denote the effective magnetic field and induction.

Finally, to obtain a constitutive dielectric equation commensurate with classical mechanical relations, the characterization must be polarization based. This means that the dielectric relations developed in this section must be inverted to obtain the electric field as a function of the polarization. Of the three models, this can be accomplished in closed form only for the Ising spin model. For that model, inversion of the relation (6) and combination with the mechanical relation yields the constitutive equations

$$\sigma = Y(e - Q_{33} P^2)$$
$$E = -2 Q_{33} P \sigma - \frac{E_0}{P_s} P + \frac{E_0 T}{T_c} \text{arctanh} \left(\frac{P}{P_s} \right). \tag{10}$$

We note that these constitutive relations can be modified for piezoelectric materials by replacing the quadratic polarization term in the mechanical relation by a linear term.

3. Domain Wall Theory

The Langevin relation (5) or Ising spin equation (6) with the effective field specified in (4), can be used to quantify the anhysteretic polarization produced by an applied field. For certain ferroelectric application (e.g., relaxor ferroelectric materials employed near their Curie temperature), these relations provide an adequate characterization of the **E-P** curve for the material. Their formulation is based upon the hypothesis of unimpeded motion of dipole

moments, however, and hence the relations are typically inadequate for materials in which pinning sites inhibit domain wall movement. In this section, we quantify the hysteresis observed in certain ferroelectric materials through consideration of the energy required to bend and translate domain walls pinned at inclusions in the material.

3.1. Pinning Energy

All ferroelectric materials exhibit material nonhomogeneities which act as pinning sites for domain walls. In many cases, these inclusions result from the stoichiometries required to attain the large strains and forces associated with the materials and hence are unavoidable in the design process. The formation of domain walls at pinning sites is due to the associated minimization of the magnetostatic energy. As the material is cycled through a full **E-P** loop, it exhibits both reversible changes in the polarization due to local movement within an energy well [8, 17, 18] and irreversible changes when the energy barrier is overcome [8, 18]. These mechanisms respectively correspond to the bending and translation of domain walls and both contribute to the hysteresis exhibited by the material. To incorporate the hysteresis observed under constant temperature, quasistatic operating conditions, we quantify in this section the reversible and irreversible domain wall motion at pinning sites. This theory follows closely that derived by Jiles and Atherton for ferromagnetic materials [9].

We consider first the energy required to translate a domain wall across pinning sites within the material. As detailed in [8], a pinning site is broken and domain wall translation occurs when sufficient energy is provided to overcome a local energy barrier. Consider the domain configuration depicted in Figure 2 in which two domains are separated by a domain wall situated at a pinning site. The dipole moments per unit volume in the two domains are denoted by $\mathbf{p}$ and $\mathbf{p}'$ with $\mathbf{p}$ assumed to be aligned with the effective electric field $\mathbf{E}_e$ (this assumption simplifies the discussion and produces the same final model as that determined with a general configuration). The angle between the moments is denoted by θ.

For ferromagnetic materials, Jiles and Atherton hypothesized that the energy required to overcome the pinning sites, and hence move the domain wall, is proportional to the change in energy required to align the magnetic moments with the field [9]. We make the same assumption for ferroelectric materials.

For a dipole moment $\mathbf{p}$, the energy due to the effective field is

$$\mathcal{E} = -\mathbf{p} \cdot \mathbf{E}_e \tag{11}$$

so that the change in energy required to rotate the moments $\mathbf{p}'$ in the direction of the field is

$$\Delta \mathcal{E} = -\mathbf{p} \cdot \mathbf{E}_e + \mathbf{p}' \cdot \mathbf{E}_e \,.$$

Under the assumption that the energy $\mathcal{E}_{pin}$ required to break pinning sites is proportional to this change, it follows that

$$\mathcal{E}_{pin} = -pE_e c_1 (1 - \cos\theta)$$

where $p = |\mathbf{p}|$, $E_e = |\mathbf{E}_e|$ and c_1 is the constant of proportionality. To eliminate c_1, we let $\mathcal{E}_\pi$ denote the energy for 180° domain walls and note that $\mathcal{E}_\pi = 2pE_e c_1$. This implies that

$$\mathcal{E}_{pin} = -\frac{1}{2}\mathcal{E}_\pi (1 - \cos\theta) \,. \tag{12}$$

The relation (12) quantifies the energy required to break a single pinning site. To obtain a corresponding macroscopic relation which quantifies the behavior of the bulk material, we employ the techniques of Jiles and Atherton [9] and let n denote the average density of pinning sites and $\langle \mathcal{E}_\pi \rangle$ denote the average energy for 180° walls. The energy required to break pinning sites if a domain wall of area A moves a distance x is then

$$\mathcal{E}_{pin}(x) = -\int_0^x \frac{n\langle \mathcal{E}_\pi \rangle}{2}(1 - \cos\theta)A\ dx \,.$$

Because the polarization is the total contribution due to dipole moments in a given volume, the change in polarization due to the reorientation of moments is

$$dP = -p(1 - \cos\theta)Adx \,. \tag{13}$$

With the definition $k = \frac{n\langle \mathcal{E}_\pi \rangle}{2p}$, (12) and (13) can be combined to yield the expression

$$\mathcal{E}_{pin}(P) = -k \int_0^P dP \tag{14}$$

for the energy required to break pinning sites. Note that this relation provides a macroscopic description due to the averaging of domain density and energy in the definition of k.

Completely analogous arguments follow for 90° domain walls as well as materials which exhibit rhombohedral symmetries ($71^o/109^o$ domain walls) with the final expressions for the pinning energy differing only in the values of k. Because the number of pinning sites and average energy are unknown, the parameter k must be estimated for any of these domain configurations through a least squares fit to data.
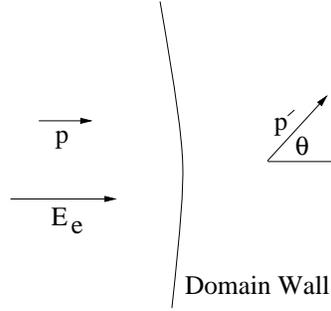
Figure 2. Domain wall and orientation of dipole moments $\mathbf{p}, \mathbf{p}'$ and electric field $\mathbf{E}_e$.

3.2. Work in the Polarization Process and Irreversible Polarization

To quantify the irreversible and reversible polarization processes, we must determine the work required to attain a specified polarization level. This can be formally obtained from classical expressions for the work required to configure a specified charge distribution or derived from first principles of electrostatic field theory.

As detailed in [19], if ρ denotes a charge density, then the work necessary to produce a change $\delta\rho$ is given by

$$\delta W = \int_V \phi \delta\rho \, dV$$

where ϕ is the associated scalar potential. The application of Maxwell's equations then yields

$$\delta W = \int_V \mathbf{E} \cdot \delta\mathbf{D} \, dV$$

so that the change in energy density is $\mathbf{E} \cdot \delta\mathbf{D}$. This yields the classical expression

$$w = \int_0^{\mathbf{D}} \mathbf{E} \cdot d\mathbf{D} \tag{15}$$

for the energy required to create the field at a point.

For this analysis, we are interested in the work required to attain a specified polarization level through the application of the interaction field $\mathbf{D}_e$. Such a relation can be formally obtained by employing the relation (7) in (15) to obtain

$$w = \frac{1}{\epsilon_0} \int_0^{\mathbf{D}} \mathbf{D} \cdot d\mathbf{D} - \frac{1}{\epsilon_0} \int_0^{\mathbf{D}} \mathbf{P} \cdot d\mathbf{D} \,.$$

Over a full cycle, the first term on the right hand side is zero so that the losses are given by

$$w = -\frac{1}{\epsilon_0} \oint \mathbf{P} \cdot d\mathbf{D} \,.$$

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Employing the interaction field $\mathbf{D}_e$, one is then left with the energy functional

$$w = \frac{1}{\epsilon_0} \int_0^{\mathbf{D}_e} \mathbf{P} \cdot d\mathbf{D}_e \tag{16}$$

for specifying hysteresis losses over a portion of the polarization cycle. We point out that the energy functional (16) is analogous to that employed by Jiles and Atherton [9] in their magnetization model. Further details regarding the derivation of this energy functional are provided in [11].

To characterize the irreversible polarization, we note that the polarization energy for a given effective field level can be expressed as that observed in the ideal (anhysteretic) case minus losses necessary to overcome local energy barriers and hence break pinning sites. Employing the relation (16) for the energy and (14) for the losses at pinning sites, this yields

$$\frac{1}{\epsilon_0} \int_0^{D_e} P_{irr}(E_e)\, dD_e = \frac{1}{\epsilon_0} \int_0^{D_e} P_{an}(E_e)\, dD_e - \int_0^{D_e} k \frac{dP_{irr}}{dD_e}\, dD_e$$

where P_{irr} denotes the irreversible polarization and P_{an} is the anhysteretic polarization given by (5) or (6) for the effective field $E_e = E + \alpha P_{irr}$. Differentiation then yields

$$P_{irr} = P_{an} - \delta \epsilon_0 k \frac{dP_{irr}}{dD_e} \tag{17}$$

where the parameter $\delta = \mathrm{sign}(dE)$ ensures that the energy required to break pinning sites always opposes changes in the polarization. To obtain an expression which facilitates numerical implementation, we reformulate (17) to obtain

$$
\begin{aligned}
P_{an} - P_{irr} &= \delta \epsilon_0 k \frac{dP_{irr}}{dD_e} \\
&= \delta k \frac{dP_{irr}}{dE} \left(\frac{1}{1 + \alpha \frac{dP_{irr}}{dE}} \right)
\end{aligned}
$$

from which it follows that

$$\frac{dP_{irr}}{dE} = \frac{P_{an} - P_{irr}}{\delta k - \alpha(P_{an} - P_{irr})}. \tag{18}$$

We note that for certain materials and operating regimes, the indiscriminate use of the differential equation (18) can lead to nonphysical solutions when the field is reversed in the saturation region of the hysteresis loop (near the tip in the first and third quadrants). Specifically, for very wide hysteresis loops of the type encountered in many ferroelectric applications, direct solution of (18) yields a negative differential electric susceptibility $\frac{dP}{dE}$ when E is reversed and the magnitude of P is less than that of the global anhysteretic. This phenomenon is not observed experimentally and the model is modified slightly for these situations. As noted in [15] where the analogous ferromagnetic phenomenon is analyzed, domain walls remain pinned when the field is initially reversed and the primary change in polarization is due to the reversible relaxation of bulged domain walls. Hence we enforce the condition $\frac{dP_{irr}}{dE} = 0$ until the polarization crosses the anhysteretic curve. This produces the more physically realistic expression

$$\frac{dP_{irr}}{dE} = \widetilde{\delta} \frac{P_{an} - P_{irr}}{k\delta - \alpha \left(P_{an} - P_{irr} \right)} \tag{19}$$

where

$$\widetilde{\delta} = \begin{cases} 0 , & \{dE > 0 \text{ and } P > P_{an}\} \text{ or } \{dE < 0 \text{ and } P < P_{an}\} \\ 1 , & \text{otherwise}. \end{cases} \tag{20}$$

We point out that this additional constrain is unnecessary for most sigmoid hysteresis curves and is required only for materials which exhibit large hysteresis losses and are driven to full saturation.

3.3. Reversible Polarization

As noted previously, the irreversible polarization is accompanied by a reversible polarization component which is due to domain wall bending before pinning sites are broken [8, 17, 18]. While the reversible polarization is typically smaller in magnitude than the irreversible polarization, the effects are still significant and must be incorporated to obtain an accurate model. The development of the reversible polarization follows closely the theory provided by Jiles and Atherton [9] for ferromagnetic materials.

To quantify the reversible polarization P_{rev}, we first consider the energy required to change the polarization from $P_{an}(E_e)$ to $P(E_e)$, where again $E_e = E + \alpha P$, at a fixed field level E. From (11), it follows that the change in energy per unit volume required to attain this change is

$$\Delta \mathcal{E}_V = -[\mathbf{P} \cdot \mathbf{E}_e - \mathbf{P}_{an} \cdot \mathbf{E}_e] .$$

Under the assumption that $|\mathbf{P}| = |\mathbf{P}_{an}| = P$ and employing the law of cosines, this yields

$$\begin{aligned} \Delta \mathcal{E}_V &= PE_e(1 - \cos \theta) \\ &= \frac{E_e}{2P}(P - P_{an})^2 \end{aligned}$$

where $(P - P_{an}) = |\mathbf{P} - \mathbf{P}_{an}|$. For fixed field values, the force on the domain walls is then

$$F = \frac{\alpha[\epsilon_0 - \epsilon(E)] - 1}{\epsilon(E) - \epsilon_0}(P - P_{an})$$

since $P = [\epsilon(E) - \epsilon_0]E$. Neglecting higher order terms and denoting the multiplicative constant by k_1 then yields the expression

$$\mathcal{P} = k_1(P - P_{an}),$$

for the pressure on the domain wall.

To obtain the reversible polarization due to wall movement in response to this pressure, we need to approximate the displaced volume. To specify the geometry, we consider a domain wall between two pinning sites separated by a distance of $2y$. The wall is assumed to bow an amount x in response to the applied pressure $\mathcal{P}$ with a resulting radius of curvature r (see Figure 3).

If we let $\mathcal{E}_S$ denote the surface energy of the domain wall, then the pressure can be expressed as

$$\mathcal{P} = \frac{2\mathcal{E}_S}{r} .$$

Using techniques analogous to those employed in Jiles and Atherton [9], it then follows that

$$\begin{aligned} x &= r - \sqrt{r^2 - y^2} \\ &\approx \frac{y^2 k_1}{4\mathcal{E}_S}(P_{an} - P) \end{aligned}$$

(for details, see [11]). Under the assumption that the wall displaces a spherical solid angle, the change in volume is given by $\Delta V = \frac{\pi}{6}x(3y^2 + x^2)$. For the case in which separated domains are parallel and antiparallel to the applied field, the reversible polarization is $P_{rev} = 2\Delta V p$ from which it follows that

$$P_{rev} = \frac{p\pi y^4 k_1}{4\mathcal{E}_S}(P_{an} - P) + \frac{p\pi}{3}\left(\frac{y^2 k_1}{4\mathcal{E}_S}\right)^3 (P_{an} - P)^3 .$$

Consideration of first-order terms then yields the expression

$$P_{rev} = c_1(P_{an} - P) \tag{21}$$

for the reversible polarization. The constant $c_1 = \frac{p\pi y^4 k_1}{4\mathcal{E}_S}$ must be estimated through a least squares fit to data since the components p, y, k_1 and $\mathcal{E}_S$ are unknown.

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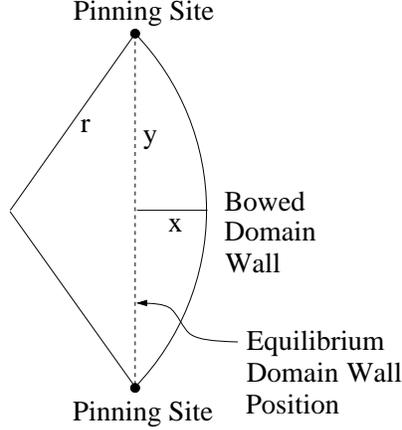
Figure 3. Geometry of the domain wall with reversible bowing (from Jiles and Atherton [9]).

3.4. Total Polarization

The total polarization is then specified in the following manner. For a given field E, polarization P and constant stress σ, the effective field and anhysteretic polarization are given by (4) and (5) or (6), respectively. The differential equation (19) is then integrated to compute the irreversible polarization P_{irr}. Finally, the total polarization P is given by the sum

$$P = P_{irr} + P_{rev}. \tag{22}$$

The reversible polarization P_{rev} can be specified either by (21) or by the expression

$$P_{rev} = c(P_{an} - P_{irr}) \tag{23}$$

where $c = \frac{c_1}{1+c_1}$. For the remainder of this development, we will employ the expression (23) since it permits the separation of the reversible and irreversible polarizations. The full time-dependent algorithm specifying the polarization $P(t)$ generated by an input field $E(t)$ is summarized in Algorithm 1. Details regarding the implementation of this algorithm are provided in [11].

(i) $\quad E_e(t) = E(t) + \alpha P_{irr}(t)$

(iia) $\quad P_{an}(t) = P_s \left[\coth\left(\frac{E_e(t)}{a} \right) - \left(\frac{a}{E_e(t)} \right) \right] \qquad$ (Langevin Model)

(iib) $\quad P_{an}(t) = P_s \tanh\left(\frac{E_e(t)}{a} \right) \qquad$ (Ising Spin Model)

(iii) $\quad \dfrac{dP_{irr}}{dt}(t) = \dfrac{dE}{dt} \cdot \dfrac{\widetilde{\delta}\,[P_{an}(t) - P_{irr}(t)]}{k\delta - \alpha[P_{an}(t) - P_{irr}(t)]}$

(iv) $\quad P_{rev}(t) = c[P_{an}(t) - P_{irr}(t)]$

(v) $\quad P(t) = P_{rev}(t) + P_{irr}(t)$

**Algorithm 1.** Time-dependent model quantifying the output polarization $P(t)$.

4. Model Validation

The model summarized in Section 3.4 provides a relationship between input electric fields and the resulting polarization which incorporates the nonlinear constitutive behavior and hysteresis due to impeded domain wall movement. In this section, we provide an example illustrating the model fit to experimental data from a PMN-PT-BT actuator employed in its ferroelectric range. The material was composed of 12% PT and 2% BT in solid solution and had a Curie temperature of $T_c = 45^o$ C. The data was collected from a stress-free sample ($\sigma = 0$ in (10)) at $T = -20^o$ C. To maintain quasistatic conditions, the frequency for the input electric field was taken to be $\omega = 1$ Hz for a complete steady-state cycle.

The parameters P_s, a, α, k and c must then be determined for the given material. For many materials, the saturation polarization P_s is well known and values can be specified directly. The parameters a, α, k, c are less easily specified, however, since they are defined in terms of material properties which are either unknown or not easily measured. Furthermore, the values of a, α, k, c depend to some extent on the choice of anhysteretic model since they depend upon the shape of the modeled anhysteretic and energy differences between the anhysteretic and measured data. In applications, a, α, k, c and in many cases P_s, are estimated through a least squares fit to data.

The modeled polarization obtained with the Langevin relation and the parameter values $a = 0.70 \times 10^5 \, C/m^2$, $\alpha = 1.53 \times 10^6 \, V m/C, k = 4.5 \times 10^5 \, C/m^2$, $c = .80, P_s = .296 \, C/m^2$ is compared with the experimental data in Figure 4. We reiterate that the data was collected under quasistatic conditions with one complete steady-state cycle plotted in the figure. The measured electric field data was used as input to the model for three complete cycles. While the initial polarization curve for the model is depicted here, one would typically bring the model to a polarized state before employing it for physical transducer characterization or control design. In this regime, the model very accurately quantifies the nonlinear and hysteretic behavior of the material.

The corresponding model employing the Ising spin relation is compared with the data in Figure 5. In this case, the model was computed with the parameters $a = 4.2 \times 10^5 \, C/m^2, \alpha = 2 \times 10^6 \, V m/C$, $k = 3.7 \times 10^5 \, C/m^2$, $c = .65, P_s = .28 \, C/m^2$. As expected, less than 5.5% change is observed in the saturation polarization P_s. The other four parameters differ from those obtained with the Langevin relation to accommodate the different qualitative behavior of the curves. A comparison of Figures 4 and 5 indicates that the use of the Langevin relation for the anhysteretic polarization provides a somewhat better fit than the Ising spin-based model. This indicates that for this material, the Ising spin model may saturate too quickly due to the imposed requirement that cells are restricted to only two possible orientations.
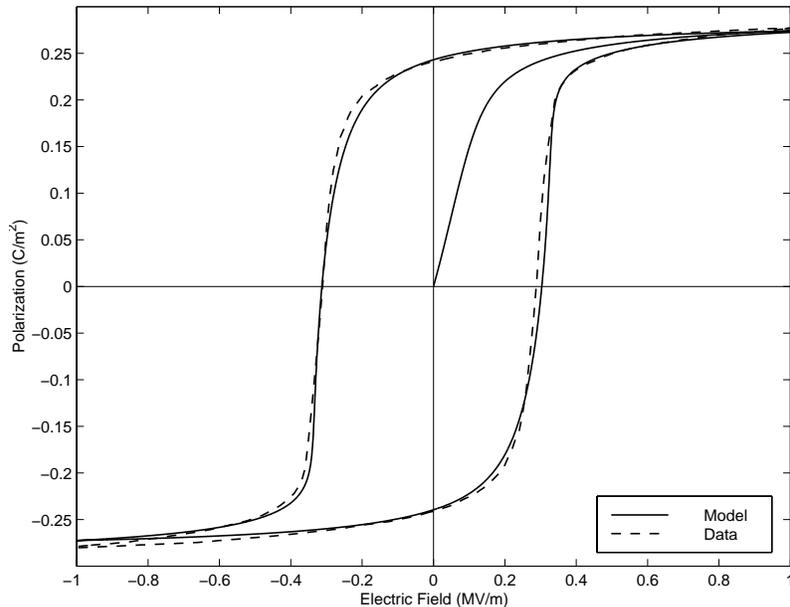
Figure 4. Experimental PMN-PT-BT data and model fit with the Langevin anhysteresis model.
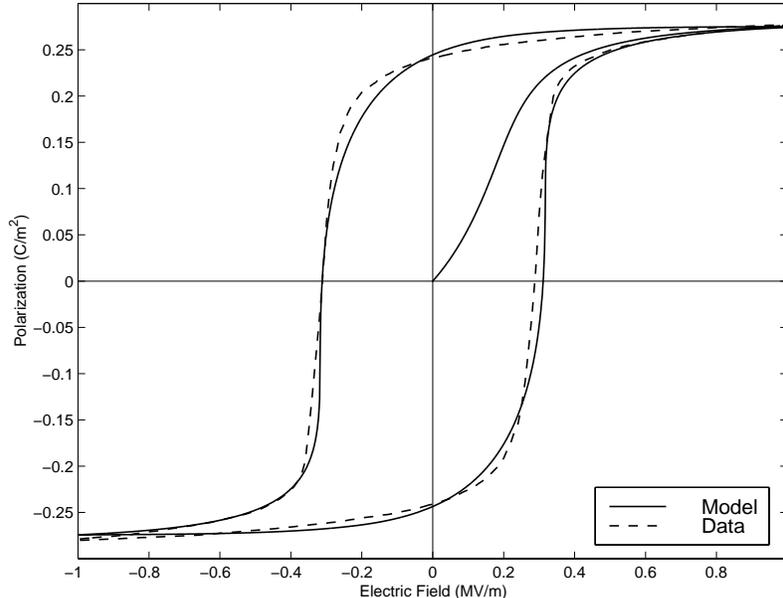
Figure 5. Experimental PMN-PT-BT data and model fit with the Ising spin anhysteresis model.

5. Concluding Remarks

The model presented here addresses the characterization of hysteresis in ferroelectric materials as well as relaxor ferroelectrics employed at temperatures well below the Curie temperature. The model is based on the quantification of energy differences between the minimum energy anhysteretic polarization and the polarization observed in applications. The anhysteretic E-P curve is derived using the nonlinear constitutive relations developed by Hom and Shankar. Hysteresis effects are quantified through the computation of the average energy required to move domain walls across inclusions or pinning sites inherent to the material. The resulting quasistatic ferroelectric model is analogous to that developed by Jiles and Atherton for ferromagnetic materials.

To illustrate the capabilities of the model, the characterization of data collected from a PMN-PT-BT actuator at a temperature 65° below the Curie temperature was considered. In this regime, the model provided a highly accurate characterization of the hysteresis exhibited by the material. While illustrated in the context of relaxor ferroelectrics at low temperatures, the electrostatic basis for the model is sufficiently general to permit its application to a variety of ferroelectric and relaxor ferroelectric materials.

A significant advantage of this modeling approach is the small number of required parameters (5) and the direct physical nature of certain parameters (P_s can often be directly measured). This facilitates the determination of parameter values when modeling the ferroelectric as well as the updating of parameters to accommodate changing operating conditions and the design of control methods for systems which employ ferroelectric transducers.

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