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THREE DIMENSIONAL DYNAMIC ELECTROMECHANICAL CONSTITUTIVE RELATIONS FOR FERROELECTRIC MATERIALS

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Abstract—We propose, in this paper, three dimensional constitutive relations suitable for describing the dynamic electromechanical responses of ferroelectric materials. Our approach is phenomenological and is based on the assumptions that the macroscopic electrical properties can be represented by electric dipoles whose magnitude and direction can be altered by external stimuli. The constitutive relations for the stress and the electric displacement take into account the transient and instantaneous responses of the dipole moments and the transient response of domain switching. A specific respesentation of the theory is also derived; this representation is suitable for studying the responses of ferroelectic materials to dynamic mechanical loading and rapidly applied electric field.

1. INTRODUCTION

In an earlier paper Chen and Peercy[1] proposed one dimensional constitutive relations suitable for describing the macroscopic dynamic responses of ferroelectric materials. The constitutive relations of the stress and the electric displacement which they offered are based on the simplest system capable of displaying ferroelectricity; and they take into account the responses of the electric dipole moments and the consequences of domain switching.

The basic premise of their proposition is that the dipole moments respond instantly to external stimuli such as mechanical loading, variation in temperature and applied electric field, but that domain switching seems to exhibit only transient response.† It turns out that such a theory seems quite adequate in describing the mechanical, piezoelectric and dielectric responses of ferroelectric materials to a slowly varying cyclic external electric field, *viz.* in predicting the results corresponding to the butterfly and hysteresis loops as consequences of domain switching.‡ However, when specimens of ferroelectric materials are subjected to dynamic mechanical loading and/or rapidly applied electric field, the dipole moments exhibit both transient and instantaneous responses. These responses of the dipole moments and the transient response of domain switching must be taken into account if we are to model these situations realistically.

Here, we consider a generalization of the one dimensional constitutive relations proposed by Chen and Peercy[1] in that we motivate the three dimensional counterparts of their relations and take into account the transient and instantaneous responses of the dipole moments and the consequences of the transient response of domain switching. We also derive explicit versions of the general relations which are suitable for specific applications.

2. GENERAL PROPERTIES OF DIPOLE MOMENTS AND CONSEQUENCES OF DOMAIN SWITCHING

In stating our three dimensional constitutive relations applicable to ferroelectric materials, our consideration must necessarily be phenomenological because the microscopic behavior of such materials is not totally understood. It is our aim to offer at this time some of the motivations leading to these relations, leaving the examination of the predictions of these relations in comparison with experimental observables to the future. Central to our proposition are certain assumptions concerning the macroscopic properties of these materials. First, we assume that the macroscopic electrical properties can be represented by electric dipoles whose

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† Thus far we have not seen experimental evidence of instantaneous domain switching.

‡ See Chen and Montgomery[2].

magnitude and direction can be altered by external stimuli. Second, we assume that the material is homogeneous on a macroscopic scale.

Let $\boldsymbol{\mu}$ denote the electric dipole moment. We presume that its value and direction depend on the mechanical strain $\mathbf{S}$, the absolute temperature θ and the external electric field $\mathbf{E}$. We further presume that its response to these stimuli may be partitioned into its transient response $\boldsymbol{\mu}_t$ and its instantaneous response $\boldsymbol{\mu}_i$ so that

$$\boldsymbol{\mu} = \boldsymbol{\mu}_t + \boldsymbol{\mu}_i. \tag{2.1}$$

In general, the vectors $\boldsymbol{\mu}(\mathbf{X}, t)$, $\boldsymbol{\mu}_t(\mathbf{X}, t)$ and $\boldsymbol{\mu}_i(\mathbf{X}, t)$ at each material point $\mathbf{X}$ and time t need not be parallel to one another.

A specimen of the material is said to exhibit remanent polarization if there exists a unique unit vector $\mathbf{i}$ such that the vector $\mathbf{N}_r$, defined by the relation

$$\mathbf{N}_r = (\Sigma\boldsymbol{\mu}_r \cdot \mathbf{i})\mathbf{i}, \tag{2.2}$$

is not zero and has maximum magnitude, where $\boldsymbol{\mu}_r$ is the spontaneous dipole moment which we will define later on in this section and where the summation is carried out over each sub-part of the specimen. The direction of the unit vector $\mathbf{i}$ defines the direction of remanent polarization, and the magnitude of $\mathbf{N}_r$ gives the effective number of aligned dipoles in this direction.

We also introduce the vector $\mathbf{N}$, defined by the relation

$$\mathbf{N} = (\Sigma\boldsymbol{\mu} \cdot \mathbf{j})\mathbf{j}, \tag{2.3}$$

where $\mathbf{j}$ is a unique unit vector such that $\mathbf{N}$ is not zero and has maximum magnitude, and where the summation is carried out over each sub-part of the specimen. The direction of the unit vector $\mathbf{j}$ defines the direction of polarization and the magnitude of $\mathbf{N}$ gives the effective number of aligned dipoles in this direction.

The preceding definitions of $\mathbf{N}_r$ and $\mathbf{N}$ are sufficiently general to permit the characterization of a virgin and/or thermally depoled specimen. In such situations the domains are random and it follows from (2.2) and (2.3) that

$$\Sigma\boldsymbol{\mu}_r \cdot \mathbf{k} = \Sigma\boldsymbol{\mu} \cdot \mathbf{k} = 0 \tag{2.4}$$

for all unit vectors $\mathbf{k}$, $\boldsymbol{\mu}_r \neq \mathbf{0}$ and $\boldsymbol{\mu} \neq \mathbf{0}$ for all $(\mathbf{X}, t)$, *viz.* the specimen does not exhibit remanent polarization or polarization in any direction. There are, however, other situations for which $\Sigma\boldsymbol{\mu}_r \cdot \mathbf{i} = 0$, but $\Sigma\boldsymbol{\mu} \cdot \mathbf{j} \neq 0$, *viz.* those for which the specimen is initially in a paraelectric or antiferroelectric phase.

We presume that the instantaneous response of the dipole moment $\boldsymbol{\mu}_i$ obeys the constitutive relation

$$\boldsymbol{\mu}_i = \boldsymbol{\mu}_i(\mathbf{S}, \theta, \mathbf{E}); \tag{2.5}$$

whereas the transient response of the dipole moment $\boldsymbol{\mu}_t$ and $\mathbf{N}$, the vector characterizing the effective number of aligned dipoles in the direction of polarization, obey the rate laws

$$\begin{aligned} \dot{\boldsymbol{\mu}}_t &= \mathbf{f}_{\boldsymbol{\mu}_t}(\mathbf{S}, \theta, \mathbf{E}, \boldsymbol{\mu}_t, \mathbf{N}), \\ \dot{\mathbf{N}} &= \mathbf{f}_{\mathbf{N}}(\mathbf{S}, \theta, \mathbf{E}, \boldsymbol{\mu}_t, \mathbf{N}). \end{aligned} \tag{2.6}$$

The implications of the rate law $(2.6)_1$ are straightforward. The rate law $(2.6)_2$ characterizes the consequences of domain switching in a macroscopic context. Microscopic mechanisms of domain switching, which include nucleation of new domains, forward growth and sideway expansion of the domains and coalescence of domains, are not totally understood; see, e.g. Fatuzzo and Merz[3].

For each $\mathbf{E} = \mathbf{E}_e \neq \mathbf{0}$ a material is said to be in an energetic equilibrium state if there exists a pair $(\mathbf{S}_e, \theta_e)$ such that $\dot{\boldsymbol{\mu}}_t$ and $\dot{\mathbf{N}}$ are zero and the equilibrium values $\boldsymbol{\mu}_{te}$ and $\mathbf{N}_e$ of $\boldsymbol{\mu}_t$ and $\mathbf{N}$ are given by the simultaneous solution of

$$\begin{aligned} \mathbf{f}_{\mu_t}(\mathbf{S}_e, \theta_e, \mathbf{E}_e, \boldsymbol{\mu}_{te}, \mathbf{N}_e) &= \mathbf{0}, \\ \mathbf{f}_{\mathbf{N}}(\mathbf{S}_e, \theta_e, \mathbf{E}_e, \boldsymbol{\mu}_{te}, \mathbf{N}_e) &= \mathbf{0}. \end{aligned} \tag{2.7}$$

Clearly, a multitude of these states may exist. We expect that at such an equilibrium state the value $\boldsymbol{\mu}_e$ of $\boldsymbol{\mu}$ is given by

$$\boldsymbol{\mu}_e = \boldsymbol{\mu}_{te} + \boldsymbol{\mu}_i, \tag{2.8}$$

and that these vectors are parallel to one another.

There are physical processes for which the time scale associated with the transient response of the dipole moments differs markedly from that associated with the transient response of domain switching; for instance, those processes associated with the mechanical and electrical responses to a slowly varying cyclic electric field during the determination of the results associated with the butterfly and hysteresis loops. In such instances $\dot{\boldsymbol{\mu}}_t = \mathbf{0}$ so that $(2.6)_1$ yields the quasi-equilibrium value $\boldsymbol{\mu}_{tQ}$ of $\boldsymbol{\mu}_t$

$$\boldsymbol{\mu}_{tQ} = \boldsymbol{\mu}_{tQ}(\mathbf{S}, \theta, \mathbf{E}, \mathbf{N}), \tag{2.9}$$

and its substitution into $(2.6)_2$ gives

$$\dot{\mathbf{N}} = \mathbf{g}_{\mathbf{N}}(\mathbf{S}, \theta, \mathbf{E}, \mathbf{N}). \tag{2.10}$$

Formula (2.10) is the three dimensional counterpart of the rate law characterizing the consequences of domain switching proposed by Chen and Peercy[1].

The rate laws (2.6) are quite general and difficult to implement in the construction of an explicit theory. It seems natural that we may seek certain simplifications when the need arises and at the same time not destroy the essence of our present approach.

A characteristic feature of ferroelectric materials is that they possess spontaneous dipole moments within certain temperature ranges. This feature may be rendered explicit in our present development, *viz.* there exist states $(\mathbf{S}_r, \theta_r)$ and $\mathbf{E} = \mathbf{0}$ such that $\dot{\boldsymbol{\mu}}_t = \mathbf{0}$, $\dot{\mathbf{N}} = \mathbf{0}$ and $\mathbf{N} = \mathbf{N}_r$. Simultaneous solution of (2.6) yields the values $\boldsymbol{\mu}_{tr}$ of $\boldsymbol{\mu}_t$ and $\mathbf{N}_r$ in terms of $(\mathbf{S}_r, \theta_r, \mathbf{0})$, i.e.

$$\begin{aligned} \boldsymbol{\mu}_{tr} &= \boldsymbol{\mu}_{tr}(\mathbf{S}_r, \theta_r, \mathbf{0}), \\ \mathbf{N}_r &= \mathbf{N}_r(\mathbf{S}_r, \theta_r, \mathbf{0}). \end{aligned} \tag{2.11}$$

Formulae (2.1), (2.5) and $(2.11)_1$ imply that the value $\boldsymbol{\mu}_r$ of $\boldsymbol{\mu}$ is now given by

$$\begin{aligned} \boldsymbol{\mu}_r &= \boldsymbol{\mu}_r(\mathbf{S}_r, \theta_r, \mathbf{0}) \\ &= \boldsymbol{\mu}_{tr}(\mathbf{S}_r, \theta_r, \mathbf{0}) + \boldsymbol{\mu}_i(\mathbf{S}_r, \theta_r, \mathbf{0}). \end{aligned} \tag{2.12}$$

$\boldsymbol{\mu}_r$ may be identified as the spontaneous dipole moment at the state $(\mathbf{S}_r, \theta_r, \mathbf{0})$. It is implicit in the definition (2.12) that $\boldsymbol{\mu}_r \neq \mathbf{0}$ only within certain temperature ranges.

In the application of our theory to ferroelectric ceramics we note that these materials are polycrystalline with a great many domains. For a virgin specimen the domains are in complete disorder, i.e. for any given direction there is an equal number of domains in and opposite to this direction and this number is the same for all directions. Hence for any direction of an orthogonal triad 1/3 of the domains have a preferred orientation in and opposite to this direction and 2/3 of the domains have a preferred orientation in the plane perpendicular to this direction. The former are called parallel domains and the latter are called perpendicular domains. Once the specimen has been thoroughly poled and returned electrically to a state exhibiting no net polarization there is now some order in its domains, *viz.* even though there is an equal number of domains in and opposite to any given direction, this number is no longer the same for all directions. We presume that now essentially all the domains are parallel domains with respect to the poling direction. Each of these classes of domains may also be separated into two subclasses consisting of permanently switchable and non-permanently switchable domains. This is motivated by the observation that the value of the polarization after some momentary delay is somewhat lower than its value immediately following the removal of the electric field during the experimental determination of the hysteresis loop, indicative of the fact that many domains remain switched, but that there is some degree of reorientation occurring on a somewhat slower time scale. We may presume that the switching behavior of the non-permanently switchable perpendicular domains is always reversible.

The preceding discussion suggests that the rate law $(2.6)_2$ governing the effective number of aligned dipoles in the direction of polarization needs to be partitioned. However, this depends on the physical situations we have in mind. For instance, in the consideration of the mechanical and electrical responses of virgin ceramics to large electrical stimuli, both parallel and perpendicular domains are present and both permanently switchable and non-permanently switchable domains are present. However, in the consideration of the dynamic mechanical depoling of power supply devices,† it seems sufficient to take into account only the permanently switchable domains and the number of such domains in the direction of remanent polarization. Further, we have only parallel domains. Therefore, it seems that we should reconsider this question for each particular physical situation. The fact that the particulars concerning the nature of the domains are implicit in the rate law $(2.6)_2$ need not detract from our presentation at this time. They may be rendered explicity whenever the need arises.

3. THREE DIMENSIONAL CONSTITUTIVE RELATIONS

Our discussions concerning the properties of dipole moments and the consequences of domain switching given in the previous section constitute the basis of our development of a three dimensional theory of dynamic ferroelectricity. It is clear that the discussions are phenomenological and the proposed macroscopic relations describe in rather general terms the transient and instantaneous responses associated with the action of external stimuli on dipole moments and domain switching. No specific attempt is made to establish a definitive connection with the developments in microscopic physics because these matters are not totally understood.

The formulation of a theory of three dimensional ferroelectricity requires that we specify the mechanical and the dielectric responses. The dielectric response of ferroelectric materials to large electrical stimuli is known in the study of ferroelectrics. For example, in the experimental determination of the hysteresis loop, the changing electric displacement is attributed essentially to domain switching. The mechanical response of ferroelectric materials is pretty much an open subject except for small amplitude peizoelectric responses and, perhaps, knowledge of the existence of the butterfly loop. This loop is essentially another manifestation of domain switching corresponding to an external cyclic electric field resulting in the mechanical straining of the ferroelectrics. There are very few experimental measurements of the butterfly loop. It is only quite recently that a minimal theory has been proposed which has sufficient structure to predict the existence of the butterfly and hysteresis loops.‡Our theory must include these effects as well as other effects which may be present.

To begin with, we presume that the electric displacement depends on the dipole moment and the effective number of aligned dipoles, *viz.* $(\boldsymbol{\mu}, \mathbf{N})$. This rather simple postulate seems quite sufficient for describing the dielectric response. Indeed, it follows from (2.1) and (2.5) that the electric displacement is given by

$$\mathbf{D} = \hat{\mathbf{D}}(\mathbf{S}, \theta, \mathbf{E}, \boldsymbol{\mu}_l, \mathbf{N}). \tag{3.1}$$

We presume that the dependences of the response function $\hat{\mathbf{D}}$ on its arguments are non-trivial,

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†See, e.g. Besancon, David and Vedel[4]. In this regard, also refer to Lysne and Percival[5].

‡See Chen and Montgomery[2].

and, in particular, $\hat{\mathbf{D}}$ has the property

$$\nabla_{\mathbf{S}}\hat{\mathbf{D}}(\mathbf{S}, \theta, \mathbf{0}, \boldsymbol{\mu}_t, \mathbf{N}) \neq \mathbf{0} \tag{3.2}$$

for all $(\mathbf{S}, \theta, \mathbf{0}, \boldsymbol{\mu}_t, \mathbf{N})$ if and only if $\mathbf{N} \neq \mathbf{0}$. The condition (3.2) states that the material exhibits piezoelectric effects only when $\mathbf{N} \neq \mathbf{0}$.

A state of remanent polarization is an equilibrium state defined by the pair $(\mathbf{S}_r, \theta_r)$ and $\mathbf{E} = \mathbf{0}$ such that $\dot{\boldsymbol{\mu}}_t = \mathbf{0}$, $\dot{\mathbf{N}} = \mathbf{0}$, and

$$\mathbf{D}_r = \hat{\mathbf{D}}(\mathbf{S}_r, \theta_r, \mathbf{0}, \boldsymbol{\mu}_{tr}, \mathbf{N}_r), \tag{3.3}$$

where $\mathbf{D}_r$ denotes the remanent polarization and $\boldsymbol{\mu}_{tr}$ and $\mathbf{N}_r$ follow from the simultaneous solution of (2.6) and are given by (2.11). Notice that the remanent polarization $\mathbf{D}_r$ depends, in particular, on the effective number and orientation of aligned dipoles $\mathbf{N}_r$ so that innumerable remanent states exist.

We presume that the mechanical stress $\mathbf{T}$ depends not only on the strain and the temperature, but also on the dipole moment and the effective number of aligned dipoles, *viz.* $(\mathbf{S}, \theta, \boldsymbol{\mu}, \mathbf{N})$. Hence, formulae (2.1) and (2.5) imply that the stress is given by

$$\mathbf{T} = \hat{\mathbf{T}}(\mathbf{S}, \theta, \mathbf{E}, \boldsymbol{\mu}_t, \mathbf{N}). \tag{3.4}$$

The dependences of $\hat{\mathbf{T}}$ on its arguments are, of course, non-trivial and, in particular,

$$\nabla_{\mathbf{E}}\hat{\mathbf{T}}(\mathbf{S}, \theta, \mathbf{0}, \boldsymbol{\mu}_t, \mathbf{N}) \neq \mathbf{0} \tag{3.5}$$

for all $(\mathbf{S}, \theta, \mathbf{0}, \boldsymbol{\mu}_t, \mathbf{N})$ if and only if $\mathbf{N} \neq \mathbf{0}$. Here again, formula (3.5) states that the material exhibits piezoelectric effects only when $\mathbf{N} \neq \mathbf{0}$.

States of remanent polarization are equilibrium states for which $\mathbf{T} = \mathbf{0}$, and it follows from (3.4) that at such a state

$$\hat{\mathbf{T}}(\mathbf{S}_r, \theta, \mathbf{0}, \boldsymbol{\mu}_{tr}, \mathbf{N}_r) = \mathbf{0}. \tag{3.6}$$

Formula (3.6) should not be viewed as additional conditions defining a state of remanent polarization. It yields the values of $\boldsymbol{\mu}_{tr}$ and $\mathbf{N}_r$ which must be consistent with those given by (2.11).

The constitutive relations for the stress and the electric displacement

$$\begin{aligned} \mathbf{T} &= \hat{\mathbf{T}}(\mathbf{S}, \theta, \mathbf{E}, \boldsymbol{\mu}_t, \mathbf{N}), \\ \mathbf{D} &= \hat{\mathbf{D}}(\mathbf{S}, \theta, \mathbf{E}, \boldsymbol{\mu}_t, \mathbf{N}) \end{aligned} \tag{3.7}$$

together with the rate laws

$$\begin{aligned} \dot{\boldsymbol{\mu}}_t &= \mathbf{f}_{\mu_t}(\mathbf{S}, \theta, \mathbf{E}, \boldsymbol{\mu}_t, \mathbf{N}), \\ \dot{\mathbf{N}} &= \mathbf{f}_{\mathbf{N}}(\mathbf{S}, \theta, \mathbf{E}, \boldsymbol{\mu}_t, \mathbf{N}) \end{aligned} \tag{3.8}$$

constitute a fully coupled three dimensional theory of dynamic ferroelectricity. The basis of our formulation is entirely phenomenolgocal. It is based on the notion that the mechanical properties of the ceramic are thermoelastic and the piezoelectric and the dielectric properties of the material are due to the transient and instantaneous responses of the dipole moments and the transient response of domain switching to external influences of the strain, the absolute temperature and the external electric field. It should be a rather straightforward exercise to include a more comprehensive treatment of the thermomechanical behavior if the need arises. On the other hand, the piezoelectric and dielectric properties which we have included in (3.7) and (3.8) seem sufficient for the purpose of describing the dynamic responses of ferroelectric materials.

Actually, our initial postulates of the constitutive relations for the stress and the electric displacement, *viz.*

$$\mathrm{T} = \bar{\mathrm{T}}(\mathrm{S}, \theta, \boldsymbol{\mu}, \mathrm{N}),$$
$$\mathrm{D} = \bar{\mathrm{D}}(\boldsymbol{\mu}, \mathrm{N}), \tag{3.9}$$

are more fundamental than (3.7). The existence of (3.7) follows from (3.9), (2.1) and (2.5) and as such we have suppressed the dependences of the stress and the electric displacement on the direction of the instantaneous response of the dipole moments. These dependences need not be empty, but their implications are beyond the scope of this paper. In any case, the constitutive relations (3.7) are more practical in physical applications.

The theory as expressed in (3.7) and (3.8) is quite general. There is, of course, no *a priori* reason to expect that there are symmetry conditions which may be imposed except under specific conditions. The theory is fully anisotropic and its implementation as it stands need not be straightforward. In the following section, we consider a specific representation of the theory which is applicable for certain particular physical processes.

4. A SPECIFIC REPRESENTATION

The principal difficulty in the construction of a specific representation of the general theory is the vector N which defines the direction of polarization and the effective number of aligned dipoles. The mechanical, piezoelectric and dielectric properties depend on both its direction and magnitude so that implementation of a representation containing both these dependences can be quite cumbersome, especially since N varies both spatially and termporally. Hence, it seems reasonable that we could restrict the properties of N in some reasonable way so as not to destroy the essence of our present development.

Our construction of a specific representation of the general theory necessitates the adoption of the following assumptions:

(i) It seems convenient to regard the state of a virgin specimen as the natural reference state and in this state the material properties of the specimen are isotropic.

(ii) As is customary, we take the polar direction as the X_3-direction. We assume that for all the physical processes to which the specimen is subjected the vector N is always parallel to the X_3-axis.

Assumption (i) is quite straightforward. However, assumption (ii) overcomes the principal difficulty alluded to earlier. Its net effect is that the directions of polarization and of remanent polarization coincide and this common direction does not alter, but that the effective number of aligned dipoles with respect to either the polarization or the remanent polarization may change. In particular, the components of N and N_r are given by

$$\mathrm{N} = (0, 0, N_3), \quad \mathrm{N}_r = (0, 0, N_{r3}). \tag{4.1}$$

Very simply, our aim is to construct a specific representation such that in a virgin state the material is isotropic exhibiting no piezoelectric effect, and that whenever $N_3 \neq 0$ the material is transversely isotropic with respect to the X_3-axis exhibiting piezoelectric effects which depend on N_3. For convenience, we restrict our consideration to situations involving isothermal processes so that the temperature remains constant.

We now introduce certain simplifications concerning the rate laws (3.8). We assume that the transient response of the dipole moments to the influences of strain and electric field may be taken into account separately. Hence, instead of $(3.8)_1$, we have

$$\dot{\boldsymbol{\mu}}_{t\mathrm{S}} = \mathbf{f}_{\boldsymbol{\mu}_{t\mathrm{S}}}(\mathrm{S}, N_3, \boldsymbol{\mu}_{t\mathrm{S}}),$$
$$\dot{\boldsymbol{\mu}}_{t\mathrm{E}} = \mathbf{f}_{\boldsymbol{\mu}_{\mathrm{E}}}(\mathbf{E}, \mathbf{N}_3, \boldsymbol{\mu}_{t\mathrm{E}}), \tag{4.2}$$

where we have introduced the obvious suggestive notation for the separate dependences, and $\boldsymbol{\mu}_t$ is now the pair $(\boldsymbol{\mu}_{t\mathrm{S}}, \boldsymbol{\mu}_{t\mathrm{E}})$. We also assume that the transient response of domain switching is independent of the transient response of the dipole moments so that $(3.8)_2$ results in the rate law

$$\dot{N}_3 = f_{N_3}(\mathbf{S}, \mathbf{E}, N_3), \tag{4.3}$$

for the effective number of aligned dipoles N_3.

Since the construction of the specific representation involves the expansion of the constitutive relations for the stress $(3.7)_1$ and the electric displacement $(3.7)_2$ with the retention of the appropriate terms and the choice of specific expressions for the rate laws (4.2) and (4.3), we will not repeat the rather tedious though straightforward process here but will simply list the results. Indeed, it can be shown that the stress and the electric displacement are given by[†]

$$\begin{aligned} T_{ij} = {} & (C^{I}_{ijkl} + a^{T}_{ijkl3}N_3)S_{kl} + (b_{ijk} + d_{ijk3}N_3)\mu_{t\mathbf{S}_k} \\ & - e_{kij3}N_3E_k - g_{kij3}N_3\mu_{t\mathbf{E}_k} + h_{3ij}N_3, \end{aligned} \tag{4.4}$$

$$\begin{aligned} D_i = {} & e_{ijk3}N_3S_{jk} + j_{ij3}N_3\mu_{t\mathbf{S}_j} \\ & + (\epsilon^{I}\delta_{ij} + \epsilon_{ij3}N_3)E_j + (\delta^{I}\delta_{ij} + \delta_{ij3}N_3)\mu_{t\mathbf{E}_j} + k_{i3}N_3, \end{aligned} \tag{4.5}$$

together with the rate laws

$$b_{ijk}\dot{\mu}_{t\mathbf{S}_k} + \alpha(N_3)b_{ijk}\mu_{t\mathbf{S}_k} = \beta_{\mathbf{b}ijlm}(N_3)S_{lm}, \tag{4.6}$$

$$d_{ijk3}\dot{\mu}_{t\mathbf{S}_k} = \alpha(N_3)d_{ijk3}\mu_{t\mathbf{S}_k} = \beta^{T}_{\mathbf{d}ijlm3}(N_3)S_{lm}, \tag{4.7}$$

$$j_{ij3}\dot{\mu}_{t\mathbf{S}_j} + \alpha(N_3)j_{ij3}\mu_{t\mathbf{S}_j} = \beta_{\mathbf{j}ikl3}(N_3)S_{kl}, \tag{4.8}$$

$$\dot{\mu}_{t\mathbf{E}_i} + \alpha_{\mu_{t\mathbf{E}_i}}(N_3)\mu_{t\mathbf{E}_i} = \beta_{\mu_{t\mathbf{E}_i}}(N_3)E_i \quad \text{(no sum)}, \tag{4.9}$$

$$\dot{N}_3 + (\alpha_{\mathbf{S}}(S_{ij}) + \alpha_{\mathbf{E}}(E_3))N_3 = \beta_{\mathbf{S}}(S_{ij}) + \beta_{\mathbf{E}}(E_3). \tag{4.10}$$

In formulae (4.4) and (4.5), C^{I}_{ijkl} and ϵ^{I} are the instantaneous isotropic elastic constants and the instantaneous dielectric constant of the virgin specimen. a^{T}_{ijkl3} give the transient changes to C^{I}_{ijkl} due to domain switching and are transversely isotropic. $b_{ijk}\mu_{t\mathbf{S}_k}$ together with the rate law (4.6) give the transient changes to $C^{I}_{ijkl} + a^{T}_{ijkl3}N_3$ due to the transient response of the dipole moments, whereas $d_{ijk3}\mu_{t\mathbf{S}_k}$ together with the rate law (4.7) give the transient change to a^{T}_{ijkl3} due to the transient responses of the dipole moments and domain switching. $\beta_{\mathbf{b}ijkl}$ are isotropic when $N_3 = 0$, otherwise $\beta_{\mathbf{b}ijkl}$ and $\beta^{T}_{\mathbf{d}ijkl3}$ are transversely isotropic. e_{ijk3} are the transient piezoelectric constants due to domain switching, whereas g_{ijk3} and $j_{ij3}\mu_{t\mathbf{S}_j}$ together with the rate laws (4.8) and (4.9) give the transient changes to e_{ijk3} due to the transient responses of the dipole moment and domain switching. All the three quantities e_{ijk3}, g_{ijk3} and $\beta_{\mathbf{j}ijk3}$ are transversely isotropic. In addition, we require that

$$\begin{gathered} \frac{\beta_{\mu_{t\mathbf{E}_i}}}{\alpha_{\mu_{t\mathbf{E}_i}}} g_{ijk3} = \frac{\beta_{\mathbf{j}ijk3}}{\alpha} \quad \text{(no sum)}, \\ \alpha_{\mu_{t\mathbf{E}_i}} = \alpha. \end{gathered} \tag{4.11}$$

These conditions ensure the compatibility of the piezoelectric coupling terms for all time. h_{3ij} give the additional stress due to domain switching and are transversely isotropic. δ^{I} together with the rate law (4.9) gives the transient change to ϵ^{I} due to the transient response of the dipole moments of the virgin specimen. ϵ_{ij3} are the transient changes to ϵ^{I} due to domain switching, whereas δ_{ij3} together with the rate law (4.9) give the transient changes to ϵ_{ij3} due to

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[†] It is a simple matter to establish the connection of (4.5) to the classical relation $\mathbf{D} = \mathbf{P} + \epsilon_0\mathbf{E}$, where $\mathbf{P}$ is the polarization. In this regard, refer to Chen and Peercy[1].

the transient responses of the dipole moments and domain switching. Both ϵ_{ij3} and δ_{ij3} are transversely isotropic. Finally, k_{i3} give the additional electric displacement due to domain switching.

Notice that the rate laws (4.6), (4.7) and (4.8) which arise because of the transient response of the dipole moments to the influence of strain are tensorial in character. They may be readily derived with the following representation of $(4.2)_1$:

$$\dot{\mu}_{t\mathrm{S}_i} + \alpha(N_3)\mu_{t\mathrm{S}_i} = \beta_{ijk}(N_3)S_{jk}. \tag{4.12}$$

Hence

$$\beta_{\mathrm{b}ijkl} = b_{ijm}\beta_{mkl}, \quad \beta^T_{\mathrm{d}ijkl3} = d_{ijm3}\beta_{mkl}, \quad \beta_{\mathrm{j}ijk3} = j_{im3}\beta_{mjk}. \tag{4.13}$$

In the applications of (4.6), (4.7) and (4.8) we need only to specify the values of $\alpha(N_3)$, $\beta_{\mathrm{b}ijkl}$, $\beta^T_{\mathrm{d}ijkl3}$ and $\beta_{\mathrm{j}ijk3}$. Since b_{ijk}, d_{ijk3} and j_{ij3} are arbitrary, these rate laws will ensure the compatible solution of $\mu_{t\mathrm{S}_i}$ as long as b_{ijk}, d_{ijk3} are j_{ij3} are invertible. The fact that any of the parameters b_{ijk}, d_{ijk3} and j_{ij3} may be zero simply implies that the corresponding rate law is trivial.

The rate law (4.10) characterizing the consequences of domain switching also merits some comments. It yields the special case

$$\dot{N}_3 + \alpha_{\mathrm{S}}(S_{ij})N_3 = \beta_{\mathrm{S}}(S_{ij}) \tag{4.14}$$

for domain switching due to strain alone, and the special case

$$\dot{N}_3 + \alpha_{\mathrm{E}}(E_3)N_3 = \beta_{\mathrm{E}}(E_3) \tag{4.15}$$

for domain switching due to electric field alone. First, domain switching due to strain depends non-linearly on the strain. Whether this dependence is too general or not is open to further investigations. However, we know that it depends at least on S_{ii}, because the imposition of hydrostatic pressure causes domain switching.† In any case, we expect that extensive domain switching due to strain occurs only if the strain is compressive.‡ For compressive strain, we expect that α_S is a monotonically increasing function of $|S_{ii}|$, and that $|\beta_{\mathrm{S}}/\alpha_{\mathrm{S}}|$ is a monotonically decreasing function of $|S_{ii}|$ such that $\operatorname{sgn}\beta_{\mathrm{S}}/\alpha_{\mathrm{S}} = \operatorname{sgn}N_3$. For expansive strain, we expect that α_{S} tends to zero and $\beta_{\mathrm{S}}/\alpha_{\mathrm{S}}$ is pretty much a constant function of $|S_{ii}|$ such that $\operatorname{sgn}\beta_{\mathrm{S}}/\alpha_{\mathrm{S}} = \operatorname{sgn}N_3$. Second, domain switching due to electric field depends only on its E_3-component. This is because of the assumptions which we have adopted. We expect that α_{E} is an even function of E_3 and increases monotonically with $|E_3|$, and that $\beta_{\mathrm{E}}/\alpha_{\mathrm{E}}$ is an odd function of E_3 such that $\operatorname{sgn}\beta_{\mathrm{E}}/\alpha_{\mathrm{E}} = \operatorname{sgn}E_3$ and whose magnitude increases monotonically with $|E_3|$.

There are also certain restrictions which must be imposed concerning the rate laws (4.14) and (4.15).§ Formula (4.14) holds if and only if

$$|N_3| > |\beta_{\mathrm{S}}/\alpha_{\mathrm{S}}|, \tag{4.16}$$

and formula (4.15) holds if and only if

$$\operatorname{sgn}N_3 \neq \operatorname{sgn}\beta_{\mathrm{E}}/\alpha_{\mathrm{E}} \tag{4.17}$$

or

$$\operatorname{sgn}N_3 = \operatorname{sgn}\beta_{\mathrm{E}}/\alpha_{\mathrm{E}} \text{ and } |N_3| \leq |\beta_{\mathrm{E}}/\alpha_{\mathrm{E}}|. \tag{4.18}$$

Otherwise, there is no domain switching. It is a simple matter to infer the implications of the conditions (4.16), (4.17) and (4.18) concerning the rate law (4.10).

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†See, e.g Jaffe, Cook and Jaffe ([6], p. 180).

‡Thus far we have not seen experimental evidence of extensive domain switching due to expansive strain.

§Strictly speaking, these restriction apply only to the permanently switchable domains.

The constitutive relations (4.4) and (4.5) are also generalizations of those which appear in the open literature concerning ferroelectric materials.† If we suppress the explicit dependences on N_3 and assume that the time scale is such that all the transient processes have reached equilibrium, then (4.4) and (4.5) may be condensed so that they are similar to those in the open literature except for the terms $h_{3ij}N_3$ and $k_{i3}N_3$. These terms account for the additional stress and electric displacement due to domain switching.

It should also be stated that the theory as stated in (4.4) through (4.10) contains that which Chen and Montgomery[2] used to establish the results pertaining to the butterfly and hysteresis loops. Essentially, they assume that the transient response associated with the dipole moments has reached equilibrium and there is no domain switching associated with strain, and they make use of the reduced forms of (4.4) and (4.5) together with the rate law (4.15).

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†See, e.g. Tiersten[7].