

HYSTERESIS IN DEFORMABLE FERRO-ELECTROELASTIC MATERIALS

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ABSTRACT. Within the framework of continuum thermodynamics, the paper develops rate-type models of deformable piezo-ferroelectric materials with the purpose of modeling hysteresis and electro-mechanical cross couplings. To this end constitutive functions are assumed to depend on a set of variables that includes their time derivatives. In order to obtain constitutive equations which are form-invariant within the set of Euclidean frames we follow an invariant formulation in which Lagrangian (or material) rather than Eulerian variables are used. While the second law states that the entropy production is non-negative for every admissible thermodynamic process, here the entropy production is viewed as a non-negative constitutive function.

1. Introduction

Piezo-ferroelectricity is a phenomenon involving the coupling among different physical quantities, like temperature, stress, strain, electric field and dielectric polarization (Nowacki 1979). They are thus a challenging subject for mathematical modeling in that they exhibit non-linear behavior and hysteresis (Kamlah 2001; Damjanovic 2006).

Within the framework of continuum thermodynamics, the paper develops rate-type models of deformable piezo-ferroelectric materials. Constitutive functions are assumed to depend on a set of variables that includes their time derivatives. The purpose of modeling hysteresis and electro-mechanical cross couplings by means of rate equations involving independent field rates suggests following an invariant formulation in which Lagrangian (or material) rather than Eulerian variables are used (see also Dorfmann and Ogden 2017). Hence, independent variables and their rates have to be invariant under any change of frame. Moreover, we assume that the constitutive equations are form-invariant within the set of Euclidean frames (see, for instance, Morro and Giorgi 2020).

By symmetry considerations, ferroelectric materials are required to be also piezoelectric and pyroelectric at (absolute) temperatures θ higher than a critical value, θ_c , called the Curie temperature (Damjanovic 2006; Chandra and Littlewood 2007; Bowen *et al.* 2014). In order to account for this, the dielectric polarization is decomposed into a reversible (piezoelectric)

and a residual (irreversible) part which is considered as an independent variable (Kamlah 2001; Giorgi and Vuk 2023).

According to Pao and Hutter (1975), the balance equations of mass density, linear momentum, angular momentum and internal energy are established. All processes which are compatible with the balance equations must satisfy the second law of thermodynamics, which is stated here as a balance equation (see, for instance, Giorgi and Morro 2021b) where both the entropy density and the entropy production rate are viewed as constitutive functions. Moreover, the latter is assumed to be non-negative.

The scheme adopted here has some features in common with that developed by Giorgi and Morro (2020, 2021a). Nevertheless, a significant difference that motivates the originality of this work consists in the use of a representation formula (see Eq. (39) devised by Giorgi and Morro 2021b). By virtue of this tool we are able to built up some general class of electroelastic and hysteretic models describing the behavior of deformable ferroelectrics, including their electromechanical coupling effects.

2. Notations and balance equations

We consider a body occupying a time-dependent region $\Omega \subset \mathcal{E}^3$. Let \mathcal{V} the space of vectors in \mathcal{E}^3 . With respect to a given origin $O \in \mathcal{E}^3$, each point of \mathcal{E}^3 can be identified with its position vector. Moreover, let \cdot and \otimes denote the scalar and vector product in \mathcal{V} , respectively; for any vector $\mathbf{u} \in \mathcal{V}$ we define $|\mathbf{u}|$ as $(\mathbf{u} \cdot \mathbf{u})^{1/2}$.

Let $\mathcal{R} \subset \mathcal{E}^3$ be the region occupied by the body in its reference configuration. The motion is described by means of the function $\boldsymbol{\xi}(\mathbf{X}, t)$, providing the position vector $\mathbf{x} \in \Omega = \boldsymbol{\xi}(\mathcal{R}, t)$. Moreover, $\mathbf{u} = \mathbf{x} - \mathbf{X}$ is named displacement vector. The symbols ∇ and ∇_R denote the gradient operator with respect to $\mathbf{x} \in \Omega$ and $\mathbf{X} \in \mathcal{R}$, respectively. The function $\boldsymbol{\xi}$ is assumed to be differentiable; hence we can define the deformation gradient as $\mathbf{F} = \nabla_R \boldsymbol{\xi}$ or, in suffix notation, $F_{iK} = \partial_{X_K} \xi_i$. The invertibility of $\mathbf{X} \rightarrow \mathbf{x} = \boldsymbol{\xi}(\mathbf{X}, t)$ is guaranteed by letting $J := \det \mathbf{F} > 0$.

A superposed dot denotes time differentiation following the motion of the body, and hence, for any function $f(\mathbf{x}, t)$, we have $\dot{f} = \partial_t f + \mathbf{v} \cdot \nabla f$. In particular, $\mathbf{v}(\mathbf{x}, t) = \dot{\mathbf{u}}(\mathbf{x}, t)$ gives the velocity field on $\Omega \times \mathbb{R}$ and $\mathbf{L} = \nabla \mathbf{v}$ ($L_{ij} = \partial_{x_j} v_i$) denotes its velocity gradient. We recall that

$$\dot{\mathbf{F}} = \mathbf{L}\mathbf{F}.$$

The left Cauchy-Green tensor \mathbf{B} and the right Cauchy-Green tensor \mathbf{C} are defined by

$$\mathbf{B} := \mathbf{F}\mathbf{F}^T, \quad \mathbf{C} := \mathbf{F}^T\mathbf{F}.$$

Moreover \mathbf{D} denotes the stretching tensor, $\mathbf{D} = \text{sym} \mathbf{L}$, and \mathbf{W} the spin tensor, $\mathbf{W} = \text{skw} \mathbf{L}$, so that $\mathbf{L} = \mathbf{D} + \mathbf{W}$.

For later convenience we denote with calligraphic letters the electromagnetic quantities in the frame locally at rest with the body (which corresponds to the current configuration). Let \mathcal{E} and \mathcal{H} denote the electric and magnetic fields. Inside the ferroelectric solid we assume a paramagnetic relation for the magnetization vector¹, $\mathcal{M} = \chi_m \mathcal{H}$, so that the

¹Indeed, very few materials that are both ferromagnetic and ferroelectric in the same phase exist in nature or have been synthesized in the laboratory (Hill 2000).

displacement vector \mathcal{D} and the magnetic induction \mathcal{B} are given by

$$\mathcal{D} = \varepsilon_0 \mathcal{E} + \mathcal{P}, \quad \mathcal{B} = \mu_0(1 + \chi_m) \mathcal{H}. \quad (1)$$

Here \mathcal{P} denotes the polarization vector at the frame locally at rest with the body, whereas ε_0 and μ_0 are positive constants which stand for vacuum permittivity and permeability, respectively. Since we are dealing with conductors, the free electric charge density is neglected, whereas the electric current density is denoted by \mathcal{J} .

A thermodynamically consistent derivation of local balance of force and energy is a delicate issue which has been carefully addressed in remarkable research papers (see for instance, Penfield Jr. and Haus 1967; Pao and Hutter 1975; Eringen and Maugin 1990; Bobbio 2000; Kovetz 2000; Bustamante, Dorfmann, and Ogden 2009; Morro and Giorgi 2023) and, in connection with ferromagnetic media, DeSimone and Guidugli (1996) and Roubíček and Tomassetti (2013). Although the aforementioned derivation does not affect the main focus of the paper, we direct the interested reader to the detailed explanation given by Morro and Giorgi (2023, § 2.16). Accordingly, we assume the balance equations of mass, linear momentum, angular momentum, and energy in the form

$$\begin{aligned} \dot{\rho} + \rho \nabla \cdot \mathbf{v} &= 0, & \rho \dot{\mathbf{v}} &= \nabla \cdot \mathbf{T} + \rho \mathbf{b} + \mathbf{f}_E, \\ \text{skw}(\mathbf{T} + \mathcal{E} \otimes \mathcal{P}) &= \mathbf{0}, \\ \rho \dot{\varepsilon} &= \mathcal{J} \cdot \mathcal{E} + \rho \mathcal{E} \cdot \dot{\mathcal{P}} + \mathbf{T} \cdot \mathbf{L} - \nabla \cdot \mathbf{q} + \rho r, \end{aligned}$$

where ε denotes the specific internal energy, \mathbf{T} the Cauchy stress tensor, \mathbf{q} the heat flux vector, ρ the mass density, r the specific external heat supply, \mathbf{b} the specific mechanical body force and \mathbf{f}_E the specific force of electric character (Morro and Giorgi 2023, p. 151),

$$\mathbf{f}_E = \rho(\mathbf{p} \cdot \nabla) \mathcal{E} - \mu \rho [\dot{\mathcal{P}} \times \mathcal{H} + \mathbf{v} \times (\mathbf{p} \cdot \nabla) \mathcal{H}],$$

where $\mu = \mu_0(1 + \chi_m)$ and $\mathbf{p} = \mathcal{P}/\rho$ denotes the polarization density per unit mass. Both polarization \mathcal{P} and stress \mathbf{T} must be specified by means of suitable constitutive equations (for instance, Eqs. (15)-(16)).

Let η be the entropy density and θ the absolute temperature. The second law of thermodynamics is stated here as a balance equation, namely

$$\rho \dot{\eta} + \nabla \cdot \frac{\mathbf{q}}{\theta} - \frac{\rho r}{\theta} = \rho \zeta \geq 0. \quad (2)$$

The scalar field ζ denotes the entropy production per unit mass; it is non-negative and is viewed as a constitutive function (Giorgi and Morro 2020, 2021a). Hence the thermodynamic process consists of η , \mathbf{q} , ζ and the other functions occurring in the balance equations. All processes which are compatible with the balance equations must satisfy the second law.

Upon substitution of $\nabla \cdot \mathbf{q} - \rho r$ from the energy equation into (2), exploitation of (1) and multiplication by θ we obtain the Clausius-Duhem relation

$$-\rho(\dot{\psi} + \eta \dot{\theta}) + \rho \mathcal{E} \cdot \dot{\mathcal{P}} + \mathcal{J} \cdot \mathcal{E} + \mathbf{T} \cdot \mathbf{L} - \frac{1}{\theta} \mathbf{q} \cdot \nabla \theta = \rho \theta \zeta, \quad \zeta \geq 0, \quad (3)$$

where $\psi = \varepsilon - \theta \eta$ denotes the Helmholtz free energy density. It is a crucial point of electro-elasticity, as well as of magneto-elasticity (Giorgi and Morro 2022), that the stress

tensor \mathbf{T} need not be symmetric. Hence the mechanical power $\mathbf{T} \cdot \mathbf{L}$ need not equal $\mathbf{T} \cdot \mathbf{D}$. In terms of the electric Gibbs free energy density,²

$$\phi = \psi - \mathbf{p} \cdot \mathcal{E} = \psi - \frac{1}{\rho} \mathcal{P} \cdot \mathcal{E},$$

Eq. (3) can be written in the form

$$-\rho(\dot{\phi} + \eta\dot{\theta}) - \mathcal{P} \cdot \dot{\mathcal{E}} + \mathcal{J} \cdot \dot{\mathcal{E}} + \mathbf{T} \cdot \mathbf{L} - \frac{1}{\theta} \mathbf{q} \cdot \nabla \theta = \rho \theta \zeta \geq 0.$$

3. Invariants fields and thermodynamic restrictions

The purpose of constructing constitutive equations involving independent field rates suggests following an invariant formulation in which Lagrangian (or material) rather than Eulerian variables are used. Accordingly, we assume that the constitutive equations obey the principle of material frame-indifference (Noll 1958). Such a principle states that constitutive equations are form-invariant within the set of Euclidean frames (Morro and Giorgi 2020). Namely, they have to be invariant under any change of frame with rotation matrix \mathbf{Q} ,

$$\mathbf{x} \rightarrow \mathbf{x}^*, \quad \mathbf{x}^* = \mathbf{c}(t) + \mathbf{Q}(t)\mathbf{x}, \quad \det \mathbf{Q} = 1.$$

In particular, vectors \mathcal{E} and \mathbf{p} transform according to

$$\mathcal{E} \rightarrow \mathbf{Q}\mathcal{E}, \quad \mathbf{p} \rightarrow \mathbf{Q}\mathbf{p}.$$

Since $\partial_x \mathbf{x}^* = \mathbf{Q}$, the deformation gradient \mathbf{F} is a tensor which transforms as a vector, too.

According to this definition, it is easy to check that the right Cauchy-Green tensor \mathbf{C} is an invariant tensor, as well as the left Cauchy-Green tensor \mathbf{B} , whereas $J = \det \mathbf{F}$ is an invariant scalar. Moreover, $\mathbf{F}^T \mathcal{E}$ and $\mathbf{F}^T \mathbf{p}$ are invariant vectors. Other invariant vectors are $\mathbf{F}^{-1} \mathcal{E}$ and $\mathbf{F}^{-1} \mathbf{p}$ (see, for instance, Giorgi and Morro 2021a). Hereafter, using the identity $\rho_R = J\rho$ we let

$$\mathbf{e} := \mathbf{F}^T \mathcal{E}, \quad \mathfrak{p} := J\mathbf{F}^{-1} \mathbf{p} = \rho_R \mathbf{F}^{-1} \mathbf{p}, \quad (4)$$

also denoting the electric and polarization fields in the reference configuration, respectively (Kovetz 2000, § 69). Since they are Lagrangian (or material) fields, their time derivatives,

$$\dot{\mathbf{e}}(\mathbf{X}, t) = \partial_t \mathbf{e}(\mathbf{X}, t), \quad \dot{\mathfrak{p}}(\mathbf{X}, t) = \partial_t \mathfrak{p}(\mathbf{X}, t),$$

are invariant vectors, too. On the contrary, the standard time derivative of a vector field in the Eulerian (or spatial) description is not objective, since the velocity field depends on the choice of the Euclidean frame.

To express the second law inequality (3) in term of the invariant fields \mathbf{e} and \mathfrak{p} , we first consider the power term $\rho \mathcal{E} \cdot \dot{\mathbf{p}}$. Since $\dot{\mathbf{p}} = \mathbf{F}\dot{\mathfrak{p}}/\rho_R + \mathbf{L}\mathbf{p}$ we obtain

$$\rho \mathcal{E} \cdot \dot{\mathbf{p}} = \frac{\rho}{\rho_R} (\mathbf{F}^T \mathcal{E}) \cdot \dot{\mathfrak{p}} + \rho (\mathcal{E} \otimes \mathbf{p}) \cdot \mathbf{L} = J^{-1} \mathbf{e} \cdot \dot{\mathfrak{p}} + (\mathcal{E} \otimes \mathbf{p}) \cdot \mathbf{L}. \quad (5)$$

Applying this result to (3) and letting for simplicity

$$\mathcal{I} = \mathbf{T} + \mathcal{E} \otimes \mathcal{P},$$

²We view the electric Gibbs free energy $\rho\phi$ as a Legendre transform of the Helmholtz free energy $\rho\psi$ with respect to the pair of vector fields \mathcal{E}, \mathcal{P} .

it follows

$$-\rho(\dot{\psi} + \eta\dot{\theta}) + \mathcal{J} \cdot \mathcal{E} + J^{-1} \mathbf{e} \cdot \dot{\mathfrak{P}} + \mathcal{I} \cdot \mathbf{L} - \frac{1}{\theta} \mathbf{q} \cdot \nabla \theta = \rho \theta \zeta, \quad \zeta \geq 0. \quad (6)$$

Since \mathcal{I} need not be symmetric, the electro-mechanical power $\mathcal{I} \cdot \mathbf{L}$ need not equal $\mathcal{I} \cdot \mathbf{D}$ and moreover

$$\mathcal{I} \cdot \mathbf{L} = J^{-1} (\mathbf{F} \mathcal{I}_{RR} \mathbf{F}^T) \cdot \mathbf{L} = J^{-1} \mathcal{I}_{RR} \cdot (\mathbf{F}^T \mathbf{D} \mathbf{F}) + J^{-1} \mathcal{I}_{RR} \cdot (\mathbf{F}^T \mathbf{W} \mathbf{F}),$$

where

$$\mathcal{I}_{RR} := J \mathbf{F}^{-1} \mathcal{I} \mathbf{F}^{-T} = \mathbf{T}_{RR} + J \mathbf{F}^{-1} \mathcal{E} \otimes \mathcal{P} \mathbf{F}^{-T} = \mathbf{T}_{RR} + \mathbf{C}^{-1} \mathbf{e} \otimes \mathfrak{P},$$

\mathbf{T}_{RR} being the second Piola-Kirchhoff stress tensor³. Since $\dot{\mathbf{C}} = 2\mathbf{F}^T \mathbf{D} \mathbf{F}$, it turns out to be independent of \mathbf{W} and moreover

$$\mathcal{I} \cdot \mathbf{L} = J^{-1} \mathcal{I}_{RR} \cdot \frac{1}{2} \dot{\mathbf{C}} + \mathcal{I} \cdot \mathbf{W}. \quad (7)$$

Then multiplying (6) by J and using (7), we obtain the Clausius-Duhem relation in the material description,

$$-\rho_R(\dot{\psi} + \eta\dot{\theta}) + \mathbf{e} \cdot \dot{\mathfrak{P}} + \frac{1}{2} \mathcal{I}_{RR} \cdot \dot{\mathbf{C}} + J \mathcal{I} \cdot \mathbf{W} + \mathcal{I}_R \cdot \mathbf{e} - \frac{1}{\theta} \mathbf{q}_R \cdot \nabla_R \theta = \rho_R \theta \zeta, \quad (8)$$

where $\zeta \geq 0$ and

$$\mathcal{I}_R = J \mathbf{F}^{-1} \mathcal{I}, \quad \mathbf{q}_R = J \mathbf{F}^{-1} \mathbf{q}, \quad \nabla_R = \mathbf{F}^T \nabla.$$

In force of the Second Law of thermodynamics, the Clausius-Duhem relation (8) must be valid for any thermodynamic process. As a consequence, the functional dependence of the constitutive relations must be subject to thermodynamic restrictions. To determine them, it is necessary to define in advance the set of independent variables, henceforth denoted by Σ . Thermodynamic restrictions are then derived from (8) by letting ψ , η and $\zeta \geq 0$ to be continuous functions of Σ .

For further convenience, we observe that in the material description the electric Gibbs free energy density ϕ takes the form

$$\phi = \psi - \frac{1}{\rho_R} \mathfrak{P} \cdot \mathbf{e}. \quad (9)$$

Accordingly, (8) can be rewritten in the form

$$-\rho_R(\dot{\phi} + \eta\dot{\theta}) - \mathfrak{P} \cdot \dot{\mathbf{e}} + \frac{1}{2} \mathcal{I}_{RR} \cdot \dot{\mathbf{C}} + J \mathcal{I} \cdot \mathbf{W} + \mathcal{I}_R \cdot \mathbf{e} - \frac{1}{\theta} \mathbf{q}_R \cdot \nabla_R \theta = \rho_R \theta \zeta. \quad (10)$$

³We observe that \mathbf{T}_{RR} and \mathcal{I}_{RR} are invariants.

4. Electroelastic constitutive relations

Let $\Sigma = (\theta, \nabla_R \theta, \mathbf{C}, \mathfrak{P})$ and $\psi = \psi(\Sigma)$, $\eta = \eta(\Sigma)$, $\zeta = \zeta(\Sigma) \geq 0$. Moreover, we assume $\mathbf{E} = \mathbf{E}(\Sigma)$ as the constitutive relation of the electric field in the material (Lagrangian) description⁴. Upon evaluation of $\dot{\psi}$ and substitution into (8) we obtain

$$\begin{aligned}
 & -\rho_R(\partial_\theta \psi + \eta)\dot{\theta} - \rho_R \partial_{\nabla_R \theta} \psi \cdot \nabla_R \dot{\theta} - (\rho_R \partial_{\mathbf{C}} \psi - \frac{1}{2} \mathcal{T}_{RR}) \cdot \dot{\mathbf{C}} \\
 & -(\rho_R \partial_{\mathfrak{P}} \psi - \mathbf{E}) \cdot \dot{\mathfrak{P}} + J \mathcal{T} \cdot \mathbf{W} + \mathcal{J}_R \cdot \mathbf{E} - \frac{1}{\theta} \mathbf{q}_R \cdot \nabla_R \theta = \rho_R \theta \zeta.
 \end{aligned}$$

The arbitrariness and linearity of $\dot{\theta}, \nabla_R \dot{\theta}, \dot{\mathfrak{P}}, \dot{\mathbf{C}}$ and \mathbf{W} imply

$$\psi = \psi(\theta, \mathbf{C}, \mathfrak{P}), \quad \eta = -\partial_\theta \psi, \quad \mathbf{E} = \rho_R \partial_{\mathfrak{P}} \psi, \tag{11}$$

$$\text{skw } \mathcal{T} := \text{skw}(\mathbf{T} + \mathcal{E} \otimes \mathcal{P}) = \mathbf{0}, \quad \mathcal{T}_{RR} = 2\rho_R \partial_{\mathbf{C}} \psi, \tag{12}$$

from which it follows

$$\mathbf{T}_{RR} = 2\rho_R \partial_{\mathbf{C}} \psi - \mathbf{C}^{-1} \mathbf{E} \otimes \mathfrak{P} = 2\rho_R \partial_{\mathbf{C}} \psi - \rho_R \mathbf{C}^{-1} \partial_{\mathfrak{P}} \psi \otimes \mathfrak{P}. \tag{13}$$

Observe that the symmetry of \mathbf{C} implies $\mathcal{T}_{RR} = \mathcal{T}_{RR}^T$, whereas \mathbf{T}_{RR} in general is not symmetrical. Using (11)-(13), we can rewrite the Clausius-Duhem inequality in the reduced form

$$\mathcal{J}_R \cdot \mathbf{E} - \frac{1}{\theta} \mathbf{q}_R \cdot \nabla_R \theta = \rho_R \theta \zeta \geq 0.$$

After applying Fourier’s and Ohm’s laws as constitutive relations for \mathbf{q}_R and \mathcal{J}_R , namely

$$\mathbf{q}_R = -\boldsymbol{\kappa} \nabla_R \theta, \quad \mathcal{J}_R = \boldsymbol{\sigma} \mathbf{E}, \tag{14}$$

$\boldsymbol{\kappa}$ and $\boldsymbol{\sigma}$ being (symmetric) positive semidefinite second-order tensors, we obtain

$$\rho_R \theta \zeta(\Sigma) = \frac{1}{\theta} \nabla_R \theta \cdot \boldsymbol{\kappa} \nabla_R \theta + \mathbf{E} \cdot \boldsymbol{\sigma} \mathbf{E} \geq 0.$$

Analogously, using the electric Gibbs (or conjugate) free energy function ϕ defined by (9) we perform a Legendre transformation from \mathfrak{P} to \mathbf{E} . Hence, $\Sigma = (\theta, \nabla_R \theta, \mathbf{C}, \mathbf{E})$ and ϕ, η, ζ and \mathfrak{P} are assumed to depend on Σ . Upon evaluation of $\dot{\phi}$ and substitution into (10) we obtain

$$\phi = \phi(\theta, \mathbf{C}, \mathbf{E}), \quad \eta = -\partial_\theta \phi, \quad \mathfrak{P} = -\rho_R \partial_{\mathbf{E}} \phi, \tag{15}$$

$$\text{skw } \mathcal{T} := \text{skw}(\mathbf{T} + \mathcal{E} \otimes \mathcal{P}) = \mathbf{0}, \quad \mathcal{T}_{RR} = 2\rho_R \partial_{\mathbf{C}} \phi, \tag{16}$$

from which it follows

$$\mathbf{T}_{RR} = 2\rho_R \partial_{\mathbf{C}} \phi + \rho_R \mathbf{C}^{-1} \mathbf{E} \otimes \partial_{\mathbf{E}} \phi. \tag{17}$$

Depending on the choice of the free energy expression, models for different materials are obtained.

⁴A more general model is considered by Dorfmann and Ogden (2017).

4.1. Linear dielectrics. Let us consider the case in which the free energy expression does not contain terms that couple strain and polarization field.

Let χ , possibly dependent on θ , be the electric susceptibility per unit volume in the current configuration. Then we assume that the free energy density is the sum of a thermoelastic strain contribution and a quadratic (isotropic) term due to polarization, namely

$$\rho\psi(\theta, \mathbf{B}, \mathcal{P}) = \rho\Psi(\theta, \mathbf{B}) + \frac{1}{2}\varepsilon_0^{-1}\chi^{-1}(\theta)|\mathcal{P}|^2. \quad (18)$$

By virtue of (4), a multiplication by J and simple manipulations give the free energy density per unit volume in the reference configuration

$$\rho_R\psi(\theta, \mathbf{C}, \mathfrak{P}) = \rho_R\Psi(\theta, \mathbf{C}) + \frac{1}{2}[\varepsilon_0\chi J]^{-1}\mathbf{C}\mathfrak{P} \cdot \mathfrak{P}.$$

Such a functional is thus expressed as a function of invariant quantities. Hence, from (11) we have

$$\mathfrak{E} = [\varepsilon_0\chi J]^{-1}\mathbf{C}\mathfrak{P}, \quad \mathfrak{P} = \varepsilon_0\chi J\mathbf{C}^{-1}\mathfrak{E}. \quad (19)$$

This in turn implies $\mathcal{P} = \varepsilon_0\chi\mathcal{E}$, which represents the usual expression of the polarization in a linear dielectric material. Moreover, the substitution of (15) and (19)₂ into (9) yields

$$\rho_R\phi(\theta, \mathbf{C}, \mathfrak{E}) = \rho_R\Psi(\theta, \mathbf{C}) - \frac{1}{2}\varepsilon_0\chi J\mathbf{C}^{-1}\mathfrak{E} \cdot \mathfrak{E}, \quad (20)$$

or, in the spatial description,

$$\rho\phi(\theta, \mathbf{B}, \mathcal{E}) = \rho\Psi(\theta, \mathbf{B}) - \frac{1}{2}\varepsilon_0\chi|\mathcal{E}|^2.$$

Otherwise, assuming that $\mathfrak{P} = \tilde{\mathfrak{P}}(\theta, \mathbf{C}, \mathfrak{E})$ is given by (19)₂, the electric Gibbs free energy (20) can also be written as

$$\rho_R\phi = \rho_R\Psi(\theta, \mathbf{C}) - \frac{1}{2}[\varepsilon_0\chi J]^{-1}\mathbf{C}\tilde{\mathfrak{P}} \cdot \tilde{\mathfrak{P}}.$$

Taking into account that $J = \sqrt{\det \mathbf{C}} = \sqrt{I_3(\mathbf{C})}$ and

$$\partial_{\mathbf{C}}J^{-1} = \partial_{\mathbf{C}}I_3^{-1/2}(\mathbf{C}) = -\frac{1}{2}I_3^{-3/2}(\mathbf{C})\partial_{\mathbf{C}}I_3(\mathbf{C}) = -\frac{1}{2}J^{-1}\mathbf{C}^{-1},$$

the substitution of (11) and (19)₁ into (13) yields

$$\begin{aligned} \mathbf{T}_{RR} &= 2[\rho_R\partial_{\mathbf{C}}\Psi + \frac{1}{2}[\varepsilon_0\chi J]^{-1}\mathfrak{P} \otimes \mathfrak{P} - \frac{1}{4}[\varepsilon_0\chi J]^{-1}(\mathbf{C}\mathfrak{P} \cdot \mathfrak{P})\mathbf{C}^{-1}] - [\varepsilon_0\chi J]^{-1}\mathfrak{P} \otimes \mathfrak{P} \\ &= 2\rho_R\partial_{\mathbf{C}}\Psi - \frac{1}{2}[\varepsilon_0\chi J]^{-1}(\mathbf{C}\mathfrak{P} \cdot \mathfrak{P})\mathbf{C}^{-1}, \end{aligned}$$

which turns out to be a symmetric tensor. So, introducing the electrostriction tensor⁵

$$\mathbf{Q} = \frac{1}{2}[\varepsilon_0\chi J]^{-1}\mathbf{C}^{-1} \otimes \mathbf{C}, \quad (21)$$

we obtain the synthetic representation

$$\mathbf{T}_{RR} = 2\rho_R\partial_{\mathbf{C}}\Psi - \mathbf{Q}\mathfrak{P} \otimes \mathfrak{P}.$$

⁵ \mathbf{Q} is a fourth-order tensor; indeed, in indicial notation $(\mathbf{C}^{-1} \otimes \mathbf{C})_{ijkl} = \mathbf{C}_{ij}^{-1}\mathbf{C}_{hk}$.

Finally, since

$$\partial_{\mathbf{F}}\Psi = \partial_{\mathbf{C}}\Psi\partial_{\mathbf{F}}\mathbf{C} = 2\mathbf{F}\partial_{\mathbf{C}}\Psi$$

(in indicial notation $\partial_{F_{iK}}C_{JH} = \partial_{F_{iK}}[F_{pJ}F_{pH}] = \delta_{KJ}F_{iH} + F_{iJ}\delta_{HK}$) the corresponding Cauchy stress becomes

$$\mathbf{T} = \rho\partial_{\mathbf{F}}\Psi\mathbf{F}^T - \frac{1}{2}[\varepsilon_0\chi]^{-1}|\mathcal{P}|^2\mathbf{1},$$

where $\mathbf{1}$ is the second-order identity tensor. The the last addendum accounts for the electrostriction pressure⁶ acting on the dielectric.

Of course, the same results follow from (17) and (20) by applying the identities⁷

$$\partial_{\mathbf{C}}J = \frac{1}{2}J\mathbf{C}^{-1}, \quad \partial_{\mathbf{C}}\mathbf{C}^{-1} = -\mathbf{C}^{-1} \boxtimes \mathbf{C}^{-1}.$$

Indeed,

$$\begin{aligned} \mathbf{T}_{RR} &= 2[\rho_R\partial_{\mathbf{C}}\Psi + \frac{1}{2}\varepsilon_0\chi J\mathbf{C}^{-1}\boldsymbol{\epsilon} \otimes \mathbf{C}^{-1}\boldsymbol{\epsilon} - \frac{1}{4}\varepsilon_0\chi J(\mathbf{C}^{-1}\boldsymbol{\epsilon} \cdot \boldsymbol{\epsilon})\mathbf{C}^{-1}] - \varepsilon_0\chi J\mathbf{C}^{-1}\boldsymbol{\epsilon} \otimes \mathbf{C}^{-1}\boldsymbol{\epsilon} \\ &= 2\rho_R\partial_{\mathbf{C}}\Psi - \frac{1}{2}\varepsilon_0\chi J(\mathbf{C}^{-1}\boldsymbol{\epsilon} \cdot \boldsymbol{\epsilon})\mathbf{C}^{-1} = 2\rho_R\partial_{\mathbf{C}}\Psi - \mathbf{Q}^*\boldsymbol{\epsilon} \otimes \boldsymbol{\epsilon}, \end{aligned}$$

$\mathbf{Q}^* = \frac{1}{2}\varepsilon_0\chi J\mathbf{C}^{-1} \otimes \mathbf{C}^{-1}$ denoting the fully-symmetric fourth-order electrostriction tensor, and the corresponding Cauchy tensor reads

$$\mathbf{T} = \rho\partial_{\mathbf{F}}\Psi\mathbf{F}^T - \frac{1}{2}\varepsilon_0\chi|\mathcal{E}|^2\mathbf{1}.$$

Maxwell stress tensor. The Maxwell stress tensor is a symmetric second-order tensor used in classical electromagnetism to represent the interaction between electromagnetic forces and mechanical momentum. If only an electric field is applied in vacuum, it takes the form

$$\mathbf{T}^M = \varepsilon_0[\mathbf{E} \otimes \mathbf{E} - \frac{1}{2}|\mathcal{E}|^2\mathbf{1}].$$

In the present approach the Maxwell stress tensor is represented by the component of the stress \mathcal{T}_{RR} which depends on the electric field. Indeed, starting from (20) and applying (16)₂ we have

$$\mathcal{T}_{RR} = 2\rho_R\partial_{\mathbf{C}}\phi = 2\rho_R\partial_{\mathbf{C}}\Psi(\theta, \mathbf{C}) + \varepsilon_0\chi J\mathbf{C}^{-1}\boldsymbol{\epsilon} \otimes \mathbf{C}^{-1}\boldsymbol{\epsilon} - \frac{1}{2}\varepsilon_0\chi J[\mathbf{C}^{-1}\boldsymbol{\epsilon} \cdot \boldsymbol{\epsilon}]\mathbf{C}^{-1},$$

so that in the spatial description

$$\mathcal{T} = \rho\partial_{\mathbf{F}}\Psi\mathbf{F}^T + \varepsilon_0\chi[\mathbf{E} \otimes \mathbf{E} - \frac{1}{2}|\mathcal{E}|^2\mathbf{1}] = \rho\partial_{\mathbf{F}}\Psi\mathbf{F}^T + \chi\mathbf{T}^M.$$

It is worth noting that in the material description the Maxwell stress comes from the (Maxwell) potential energy $\rho_R\psi^M = \frac{1}{2}\varepsilon_0\chi J\mathbf{C}^{-1}\boldsymbol{\epsilon} \cdot \boldsymbol{\epsilon}$. Indeed

$$\mathbf{T}_{RR}^M = \varepsilon_0\chi J\left[\mathbf{C}^{-1}\boldsymbol{\epsilon} \otimes \mathbf{C}^{-1}\boldsymbol{\epsilon} - \frac{1}{2}(\mathbf{C}^{-1}\boldsymbol{\epsilon} \cdot \boldsymbol{\epsilon})\mathbf{C}^{-1}\right] = 2\rho_R\partial_{\mathbf{C}}\psi^M,$$

while in the spatial description the Maxwell potential energy takes the form $\rho\psi^M = \frac{1}{2}\varepsilon_0\chi|\mathbf{E}|^2$.

⁶Electrostriction is a quadratic effect that applies to all crystal symmetries.

⁷In indicial notation, $(\mathbf{C}^{-1} \boxtimes \mathbf{C}^{-1})_{ijkh} = \mathbf{C}_{ih}^{-1}\mathbf{C}_{jk}^{-1}$.

4.2. Piezoelectric materials. Electrostriction applies to all crystal symmetries, while the piezoelectric effect⁸ only applies to the 20 piezoelectric point groups. Piezoelectricity, unlike electrostriction, is a linear effect. Piezoelectric materials change their electric polarization properties when compressed or stretched. To describe this effect when large deformations occur we are led to add a coupling term in the expression of the Helmholtz free energy. For example, let

$$\rho\psi(\theta, \mathbf{B}, \mathcal{P}) = \rho\Psi(\theta, \mathbf{B}) + \frac{1}{2}\varepsilon_0^{-1}\chi^{-1}(\theta)|\mathcal{P}|^2 - \varepsilon_0^{-1}\chi^{-1}(\theta)\mathcal{P} \cdot \mathbf{A}(\theta)\mathbf{S}, \quad (22)$$

where $\mathbf{S} := \frac{1}{2}(\mathbf{1} - \mathbf{B}^{-1})$ is the Eulerian-Almansi strain tensor and $\mathbf{A} : \text{Sym}(\mathcal{V}) \rightarrow \mathcal{V}$ denotes a third-order tensor such that

$$\Lambda_{ijk} = \Lambda_{ikj}, \quad \Lambda_{ijk}^T = \Lambda_{jki}.$$

As a consequence, we observe that

$$2\mathbf{A}\mathbf{S} = \Lambda_{ijk}(\delta_{jk} - F_{jl}^{-T}F_{lk}^{-1}) = F_{nk}^{-1}\Lambda_{ijk}F_{jm}^{-T}(F_{mp}^T F_{pn} - \delta_{mn}) = 2\widehat{\mathbf{A}}\mathbf{E},$$

where $\mathbf{E} := \frac{1}{2}(\mathbf{C} - \mathbf{1})$ is the Green-St. Venant (Lagrangian) strain tensor. Since $\widehat{\mathbf{A}}$ transforms as a vector, namely $\widehat{\mathbf{A}}^* = \mathbf{Q}\widehat{\mathbf{A}}$, then the quantity $\mathfrak{L} = \mathbf{F}^{-1}\widehat{\mathbf{A}}$ is an objective third-order tensor⁹. After multiplying (22) by J we get

$$\rho_R\psi(\theta, \mathbf{C}, \mathfrak{P}) = \rho_R\Psi(\theta, \mathbf{C}) + \frac{1}{2}(\varepsilon_0\chi J)^{-1}\mathbf{C}\mathfrak{P} \cdot \mathfrak{P} - (\varepsilon_0\chi)^{-1}\mathbf{C}\mathfrak{P} \cdot \mathfrak{L}\mathbf{E}.$$

From thermodynamic restrictions (11) it then follows

$$\mathfrak{E} = [\varepsilon_0\chi J]^{-1}\mathbf{C}[\mathfrak{P} - J\mathfrak{L}\mathbf{E}], \quad \mathfrak{P} = \varepsilon_0\chi J\mathbf{C}^{-1}\mathfrak{E} + J\mathfrak{L}\mathbf{E}. \quad (23)$$

Accordingly, in the spatial description we obtain the usual linear form

$$\mathcal{P} = \varepsilon_0\chi\mathcal{E} + \widehat{\mathbf{A}}\mathbf{E} = \varepsilon_0\chi\mathcal{E} + \mathbf{A}\mathbf{S}, \quad (24)$$

where the tensor \mathbf{A} accounts for the *direct piezoelectric effect*. Moreover, assuming that $\mathfrak{P} = \mathfrak{P}(\theta, \mathbf{C}, \mathfrak{E})$ is given by (23)₂, the free energy can be rewritten as

$$\rho_R\psi = \rho_R\Psi(\theta, \mathbf{C}) + \frac{1}{2}\varepsilon_0\chi J\mathbf{C}^{-1}\mathfrak{E} \cdot \mathfrak{E} - \frac{1}{2}(\varepsilon_0\chi)^{-1}J\mathbf{C}\mathfrak{L}\mathbf{E} \cdot \mathfrak{L}\mathbf{E} \quad (25)$$

and the electric Gibbs free energy takes the form

$$\begin{aligned} \rho_R\phi(\theta, \mathbf{C}, \mathfrak{E}) &= \rho_R\Psi(\theta, \mathbf{C}) - \frac{1}{2}\varepsilon_0\chi J\mathbf{C}^{-1}\mathfrak{E} \cdot \mathfrak{E} - J\mathfrak{L}\mathbf{E} \cdot \mathfrak{E} - \frac{1}{2}(\varepsilon_0\chi)^{-1}J\mathbf{C}\mathfrak{L}\mathbf{E} \cdot \mathfrak{L}\mathbf{E} \\ &= \rho_R\Psi(\theta, \mathbf{C}) - \frac{1}{2}(\varepsilon_0\chi J)^{-1}\mathbf{C}\mathfrak{P}(\theta, \mathbf{C}, \mathfrak{E}) \cdot \mathfrak{P}(\theta, \mathbf{C}, \mathfrak{E}). \end{aligned}$$

⁸The piezoelectric effect occurs only in a few dielectric materials that can be polarized, in addition to an electric field, also by applying a mechanical stress.

⁹ $\mathfrak{L}^* = (\mathbf{F}^{-1}\widehat{\mathbf{A}})^* = (\mathbf{Q}\mathbf{F})^{-1}\mathbf{Q}\widehat{\mathbf{A}} = \mathbf{F}^{-1}\mathbf{Q}^{-1}\mathbf{Q}\widehat{\mathbf{A}} = \mathbf{F}^{-1}\widehat{\mathbf{A}} = \mathfrak{L}$.

In order to obtain the expression of the piezoelectric stress tensor, we observe that $\mathbf{C}\mathfrak{P} \cdot \boldsymbol{\varepsilon} \mathbf{E} = \boldsymbol{\varepsilon}^T(\mathbf{C}\mathfrak{P}) \cdot \mathbf{E}$ and

$$\begin{aligned} \rho_R \partial_C \Psi &= \rho_R \partial_C \Psi - \frac{1}{4}(\varepsilon_0 \chi J)^{-1} [\mathbf{C}\mathfrak{P} \cdot \mathfrak{P}] \mathbf{C}^{-1} + \frac{1}{2}(\varepsilon_0 \chi J)^{-1} \mathfrak{P} \otimes \mathfrak{P} \\ &\quad - (\varepsilon_0 \chi)^{-1} \boldsymbol{\varepsilon} \mathbf{E} \otimes \mathfrak{P} - \frac{1}{2}(\varepsilon_0 \chi)^{-1} \boldsymbol{\varepsilon}^T(\mathbf{C}\mathfrak{P}). \end{aligned}$$

Accordingly, after replacing (23) into (13) we have

$$\mathbf{T}_{RR} = \rho_R \partial_E \Psi - (\varepsilon_0 \chi)^{-1} \boldsymbol{\varepsilon}^T(\mathbf{C}\mathfrak{P}) - (\varepsilon_0 \chi)^{-1} \boldsymbol{\varepsilon} \mathbf{E} \otimes \mathfrak{P} - \mathbf{Q}\mathfrak{P} \otimes \mathfrak{P},$$

in which we used $2\partial_C = \partial_E$ and (21). Note that

$$\boldsymbol{\varepsilon}^T(\mathbf{C}\mathfrak{P}) = \hat{\boldsymbol{\Lambda}}^T \mathbf{F}^{-T} (\mathbf{F}^T \mathbf{F} \mathbf{J} \mathbf{F}^{-1} \mathcal{P}) = J \hat{\boldsymbol{\Lambda}}^T \mathcal{P}, \quad J^{-1} \mathbf{F} (J \hat{\boldsymbol{\Lambda}}^T \mathcal{P}) \mathbf{F}^T = \boldsymbol{\Lambda}^T \mathcal{P}.$$

Consequently, we obtain

$$\mathbf{T} = \rho \partial_F \Psi \mathbf{F}^T - (\varepsilon_0 \chi)^{-1} \boldsymbol{\Lambda}^T \mathcal{P} - (\varepsilon_0 \chi)^{-1} \boldsymbol{\Lambda} \mathbf{S} \otimes \mathcal{P} - \frac{1}{2}(\varepsilon_0 \chi)^{-1} |\mathcal{P}|^2 \mathbf{1}.$$

Applying the "small deformations" approximation we have $J \approx 1$ and

$$\mathbf{S}, \mathbf{E} \approx \boldsymbol{\varepsilon} = \frac{1}{2}(\nabla \mathbf{u} + \nabla \mathbf{u}^T),$$

so that (24) takes the form

$$\mathcal{P} = \varepsilon_0 \chi \boldsymbol{\varepsilon} + \boldsymbol{\Lambda} \boldsymbol{\varepsilon}. \quad (26)$$

Then letting

$$\rho_R \Psi(\boldsymbol{\theta}, \mathbf{C}) \approx \frac{1}{2} \boldsymbol{\varepsilon} \cdot \mathbf{C} \boldsymbol{\varepsilon}, \quad (27)$$

where \mathbf{C} is the totally-symmetric fourth-order elasticity tensor, it follows

$$\mathbf{T}_{RR} \approx \mathbf{T} \approx \mathbf{C} \boldsymbol{\varepsilon} - (\varepsilon_0 \chi)^{-1} [\boldsymbol{\Lambda}^T \mathcal{P} + \boldsymbol{\Lambda} \boldsymbol{\varepsilon} \otimes \mathcal{P} + |\mathcal{P}|^2 \mathbf{1}]. \quad (28)$$

Note that the occurrence of the dyadic term makes the piezoelectric stress non-symmetrical even when small deformations are considered. Otherwise, if we neglect nonlinear terms in $\boldsymbol{\varepsilon}$ and \mathcal{P} , the stress reduces to the symmetrical (approximate) form

$$\mathbf{T} = \mathbf{C} \boldsymbol{\varepsilon} - (\varepsilon_0 \chi)^{-1} \boldsymbol{\Lambda}^T \mathcal{P}.$$

Finally, after replacing \mathcal{P} from (26) and letting $\mathbf{K} = \mathbf{C} - (\varepsilon_0 \chi)^{-1} \boldsymbol{\Lambda}^T \boldsymbol{\Lambda}$ we obtain

$$\mathbf{T} = \mathbf{K} \boldsymbol{\varepsilon} - \boldsymbol{\Lambda}^T \boldsymbol{\varepsilon}, \quad (29)$$

where the tensor $\boldsymbol{\Lambda}^T$ accounts for the *converse piezoelectric effect*. Note that \mathbf{K} is a totally symmetric fourth-order tensor, as well as \mathbf{C} . The linear system of coupled equations (26)-(29) is usually referred to as the *stress-charge form* of piezoelectricity.

5. Ferro-electro-elastic constitutive relations

Unlike dielectrics and piezoelectrics, in ferroelectrics the polarization vector \mathcal{P} cannot be assigned as a constitutive function. According to Ogden and Steigmann (2011), it is only partially determined through a constitutive function. Taking into account that all the ferroelectric materials exhibit piezoelectric effects at temperatures above the Curie point, say θ_c , but show a hysteretic behaviour below this point, Giorgi and Vuk (2023) assumed

$$\mathcal{P} = \mathcal{G}(\theta, \mathbf{S}, \mathcal{E}) + h(\theta)\mathcal{R}, \quad h(\theta) = \max\{0, 1 - \theta/\theta_c\},$$

where \mathcal{R} is an independent variable, while \mathcal{G} is a differentiable function which vanishes when $\mathbf{S} = \mathbf{0}$ (no deformation) and $\mathcal{E} = \mathbf{0}$ (no applied electric field). For instance, as in (24), we let

$$\mathcal{G} = \varepsilon_0\chi\mathcal{E} + \Lambda\mathbf{S}.$$

The last term, $h(\theta)\mathcal{R}$, denotes the *remanent polarization density*. The adjective ‘‘remanent’’ is justified because, in stationary conditions without deformation, it represents the so-called *spontaneous polarization*, that is the polarization left behind in a ferroelectric material (such as iron) after an external electric field is removed. Due to the factor h in front of \mathcal{R} , the remanent polarization depends on temperature and disappears when $\theta \geq \theta_c$. Accordingly, in the material description the polarization vector \mathfrak{P} takes the form

$$\mathfrak{P} = \mathfrak{E}(\theta, \mathbf{E}, \mathbf{e}) + h(\theta)\mathbf{\Pi}, \quad (30)$$

where $\mathfrak{E} = J\mathbf{F}^{-1}\mathcal{G}$ and $\mathbf{\Pi} = J\mathbf{F}^{-1}\mathcal{R}$. Note that both \mathfrak{E} and $\mathbf{\Pi}$ are invariant fields, as well as \mathfrak{P} . Since \mathfrak{E} is expected to vanish when $\mathbf{E} = \mathbf{e} = \mathbf{0}$, we can choose, as in (23),

$$\mathfrak{E} = \varepsilon_0\chi J\mathbf{C}^{-1}\mathbf{e} + J\mathcal{L}\mathbf{E}.$$

As apparent, when a ferro-electroelastic material is in the para-electric phase, $\theta \geq \theta_c$, its constitutive equations and thermodynamic restrictions are the same as those already studied in section 4.2. Instead, when $\theta < \theta_c$ it is necessary to add a further independent variable to the set Σ with the purpose of modeling the hysteresis. This variable can be \mathcal{R} , as assumed by Giorgi and Vuk (2023), or the whole polarization \mathcal{P} , as linked to \mathcal{R} by a linear relationship. In this section we opt for the first choice. However, invariance requirements demand that the free energy be a function of Euclidean invariants and the dependence on the derivatives occurs in an objective way, hence we let

$$\Sigma := (\theta, \nabla_R\theta, \mathbf{C}, \mathbf{e}, \mathbf{\Pi}, \dot{\mathbf{e}})$$

be the set of variables involved in any thermodynamic process.

Thermodynamic restrictions are scrutinized by letting

$$\phi = \phi(\Sigma) := \psi(\Sigma) - \frac{1}{\rho_R}\mathfrak{P} \cdot \mathbf{e}, \quad \eta = \eta(\Sigma), \quad \zeta = \zeta(\Sigma) \geq 0,$$

and assuming ψ to be a differentiable function of all its arguments. Upon evaluation of $\dot{\phi}$ and substitution into (10) we obtain

$$\begin{aligned} & -\rho_R(\partial_\theta\phi + \eta)\dot{\theta} - \rho_R\partial_{\nabla_R\theta}\phi \cdot \nabla_R\dot{\theta} - \left(\rho_R\partial_{\mathbf{C}}\phi - \frac{1}{2}\mathcal{I}_{RR}\right) \cdot \dot{\mathbf{C}} - (\rho_R\partial_{\mathbf{e}}\phi + \mathfrak{E} + h\mathbf{\Pi}) \cdot \dot{\mathbf{e}} \\ & - \rho_R\partial_{\mathbf{\Pi}}\phi \cdot \dot{\mathbf{\Pi}} - \rho_R\partial_{\dot{\mathbf{e}}}\phi \cdot \dot{\dot{\mathbf{e}}} + J\mathcal{I} \cdot \mathbf{W} + \mathcal{I}_R \cdot \mathbf{e} - \frac{1}{\theta}\mathbf{q}_R \cdot \nabla_R\theta = \rho_R\theta\zeta \geq 0. \end{aligned} \quad (31)$$

The arbitrariness and linearity of $\dot{\theta}$, $\nabla_R \dot{\theta}$, $\dot{\mathbf{C}}$, $\dot{\mathbf{e}}$ and \mathbf{W} imply

$$\phi = \phi(\theta, \mathbf{C}, \mathbf{e}, \mathbf{\Pi}), \quad \eta = -\partial_\theta \phi, \tag{32}$$

$$\text{skw } \mathcal{J} := \text{skw}(\mathbf{T} + \mathcal{E} \otimes \mathcal{P}) = \mathbf{0}, \tag{33}$$

$$\mathcal{J}_{RR} = 2\rho_R \partial_{\mathbf{C}} \phi(\theta, \mathbf{C}, \mathbf{e}, \mathbf{\Pi}), \tag{34}$$

from which it follows

$$\mathbf{T}_{RR} = 2\rho_R \partial_{\mathbf{C}} \phi - \mathbf{C}^{-1} \mathbf{e} \otimes [\mathbf{\Xi} + h\mathbf{\Pi}]. \tag{35}$$

Observe that the symmetry of \mathbf{C} implies $\mathcal{J}_{RR} = \mathcal{J}_{RR}^T$, whereas \mathbf{T}_{RR} in general is not symmetrical. Using (32)-(34) we can rewrite the Clausius-Duhem inequality (31) in the following reduced form

$$-(\rho_R \partial_{\mathbf{e}} \phi + \mathbf{\Xi} + h\mathbf{\Pi}) \cdot \dot{\mathbf{e}} - \rho_R \partial_{\mathbf{\Pi}} \phi \cdot \dot{\mathbf{\Pi}} + \mathcal{J}_R \cdot \mathbf{e} - \frac{1}{\theta} \mathbf{q}_R \cdot \nabla_R \theta = \rho_R \theta \zeta \geq 0.$$

After applying Fourier's and Ohm's laws (14) we obtain

$$(\rho_R \partial_{\mathbf{e}} \phi + \mathbf{\Xi} + h\mathbf{\Pi}) \cdot \dot{\mathbf{e}} + \rho_R \partial_{\mathbf{\Pi}} \phi \cdot \dot{\mathbf{\Pi}} = -\rho_R \theta \tilde{\zeta}, \tag{36}$$

where

$$\rho_R \theta \tilde{\zeta} = \rho_R \theta \zeta - \frac{1}{\theta} \nabla_R \theta \cdot \boldsymbol{\kappa} \nabla_R \theta - \mathbf{e} \cdot \boldsymbol{\sigma} \mathbf{e}.$$

Indeed, in this framework we cannot state that $\dot{\mathbf{e}}$ and $\dot{\mathbf{\Pi}}$ are mutually independent. Furthermore, assuming that $\tilde{\zeta}$ is independent of \mathbf{e} and $\nabla_R \theta$, namely

$$\tilde{\zeta} = \tilde{\zeta}(\theta, \mathbf{C}, \mathbf{\Pi}, \dot{\mathbf{e}}),$$

it follows that $\tilde{\zeta} = \zeta|_{\mathbf{e}=\nabla_R \theta=0}$ and then $\tilde{\zeta}$ is nonnegative along any thermodynamic process as well as ζ .

In order to study in detail which models are compatible with (36), let us recall a useful representation formula (Giorgi and Morro 2021b).

A representation formula. Our thermodynamic analysis has led us to a relation of the following form

$$\mathbf{A}_1 \cdot \dot{\mathcal{X}} + \mathbf{A}_2 \cdot \dot{\mathcal{Y}} = f, \tag{37}$$

where $\mathbf{A}_1, \mathbf{A}_2, \dot{\mathcal{X}}, \dot{\mathcal{Y}}$ are vectors and f is a scalar. If $\dot{\mathcal{X}}$ and $\dot{\mathcal{Y}}$ are arbitrary and independent then it follows that $\mathbf{A}_1 = \mathbf{A}_2 = \mathbf{0}$ (and $f = 0$). If, instead, $\dot{\mathcal{X}}$ and $\dot{\mathcal{Y}}$ are not independent (and this is the case given by (36)) then we can determine a relation between $\dot{\mathcal{Y}}$ and $\dot{\mathcal{X}}$ through a representation formula. A similar approach occurs in nonholonomic system described by a finite set of independent parameters subject to differential constraints that make their rates mutually dependent.

Let \mathbf{N} be a unit vector, $|\mathbf{N}| = 1$. Then

$$\dot{\mathcal{Y}} = (\dot{\mathcal{Y}} \cdot \mathbf{N})\mathbf{N} + \dot{\mathcal{Y}}_\perp, \quad \dot{\mathcal{Y}}_\perp \cdot \mathbf{N} = 0.$$

It happens that $\dot{\mathcal{Y}} \cdot \mathbf{N}$ is known, say $\dot{\mathcal{Y}} \cdot \mathbf{N} = g$, whereas $\dot{\mathcal{Y}}_\perp$ is unknown and then $\dot{\mathcal{Y}}_\perp$ can be expressed by

$$\dot{\mathcal{Y}}_\perp = (\mathbf{1} - \mathbf{N} \otimes \mathbf{N})\mathbf{G} = \mathbf{G} - (\mathbf{G} \cdot \mathbf{N})\mathbf{N},$$

where \mathbf{G} is an arbitrary vector. Once $\dot{\mathcal{Y}} \cdot \mathbf{N} = g$ is given we can write the representation formula

$$\dot{\mathcal{Y}} = g\mathbf{N} + (\mathbf{1} - \mathbf{N} \otimes \mathbf{N})\mathbf{G}.$$

Back to (37), if $\mathbf{A}_2 \neq \mathbf{0}$ we let $\mathbf{N} = \mathbf{A}_2/|\mathbf{A}_2|$ and $g = (f - \mathbf{A} \cdot \dot{\mathcal{X}})/|\mathbf{A}_2|$ to have

$$\dot{\mathcal{Y}} = \frac{f - \mathbf{A}_1 \cdot \dot{\mathcal{X}}}{|\mathbf{A}_2|^2} \mathbf{A}_2 + \left(\mathbf{1} - \frac{\mathbf{A}_2}{|\mathbf{A}_2|} \otimes \frac{\mathbf{A}_2}{|\mathbf{A}_2|} \right) \mathbf{G}. \quad (38)$$

Since $(\mathbf{A}_2 \otimes \mathbf{A}_2)\mathbf{G} = (\mathbf{A}_2 \cdot \mathbf{G})\mathbf{A}_2$, it follows

$$\dot{\mathcal{Y}} = \mathbf{G} + \frac{f - \mathbf{A}_1 \cdot \dot{\mathcal{X}} - \mathbf{A}_2 \cdot \mathbf{G}}{|\mathbf{A}_2|^2} \mathbf{A}_2,$$

so that by choosing $\mathbf{G} = \mathbf{\Gamma} \dot{\mathcal{X}}$ we obtain

$$\dot{\mathcal{Y}} = \left[\mathbf{\Gamma} - \frac{\mathbf{A}_2 \otimes (\mathbf{A}_1 + \mathbf{\Gamma}^T \mathbf{A}_2)}{|\mathbf{A}_2|^2} \right] \dot{\mathcal{X}} + \frac{f}{|\mathbf{A}_2|^2} \mathbf{A}_2, \quad (39)$$

where $\mathbf{\Gamma}$ is an arbitrary second-order tensor.

5.1. Hypo-electroelastic materials. Let $\phi_R := \rho_R \phi(\theta, \mathbf{C}, \mathbf{\epsilon}, \mathbf{\Pi})$ and $\mathbf{\Xi} = \mathbf{\Xi}(\theta, \mathbf{C}, \mathbf{\epsilon})$. Choosing $\zeta = 0$, (36) reduces to

$$(\partial_{\mathbf{\epsilon}} \phi_R + \mathbf{\Xi} + h\mathbf{\Pi}) \cdot \dot{\mathbf{\epsilon}} + \partial_{\mathbf{\Pi}} \phi_R \cdot \dot{\mathbf{\Pi}} = 0.$$

We first observe that $\dot{\mathbf{\Pi}}$ and $\dot{\mathbf{\epsilon}}$ are not independent fields when $\theta \in (0, \theta_C)$. In fact, if instead they were independent then their arbitrariness would imply

$$\partial_{\mathbf{\Pi}} \phi_R = \mathbf{0}, \quad \partial_{\mathbf{\epsilon}} \phi_R = -\mathbf{\Xi} - h\mathbf{\Pi}.$$

Since $\mathbf{\Xi}$ is independent of $\mathbf{\Pi}$ by assumption, a further differentiation gives

$$\partial_{\mathbf{\Pi}, \mathbf{\epsilon}}^2 \phi_R = \mathbf{0}, \quad \partial_{\mathbf{\epsilon}, \mathbf{\Pi}}^2 \phi_R = -h\mathbf{1},$$

a result that contradicts the assumed regularity of the function ϕ_R when $h(\theta) \neq 0$. So, in the range $\theta \in (0, \theta_C)$ the representation formula (38) can be applied by letting

$$\mathcal{Y} = \mathbf{\Pi}, \quad \mathcal{X} = \mathbf{\epsilon}, \quad f = 0, \quad \mathbf{A}_2 = \partial_{\mathbf{\Pi}} \phi_R, \quad \mathbf{A}_1 = \partial_{\mathbf{\epsilon}} \phi_R + \mathbf{\Xi} + h\mathbf{\Pi}.$$

It follows

$$\dot{\mathbf{\Pi}} = - \frac{(\mathbf{\Xi} + h\mathbf{\Pi} + \partial_{\mathbf{\epsilon}} \phi_R) \cdot \dot{\mathbf{\epsilon}}}{|\partial_{\mathbf{\Pi}} \phi_R|^2} \partial_{\mathbf{\Pi}} \phi_R + \left(\mathbf{1} - \frac{\partial_{\mathbf{\Pi}} \phi_R}{|\partial_{\mathbf{\Pi}} \phi_R|} \otimes \frac{\partial_{\mathbf{\Pi}} \phi_R}{|\partial_{\mathbf{\Pi}} \phi_R|} \right) \mathbf{G}.$$

We only need to assume $\partial_{\mathbf{\Pi}} \phi_R \neq \mathbf{0}$. Otherwise, by applying (39) we get

$$\dot{\mathbf{\Pi}} = \boldsymbol{\chi} \dot{\mathbf{\epsilon}},$$

where

$$\boldsymbol{\chi}(\theta, \mathbf{C}, \mathbf{\epsilon}, \mathbf{\Pi}) = \mathbf{\Gamma} - \frac{1}{|\partial_{\mathbf{\Pi}} \phi_R|^2} \partial_{\mathbf{\Pi}} \phi_R \otimes [\mathbf{\Xi} + h\mathbf{\Pi} + \partial_{\mathbf{\epsilon}} \phi_R + \mathbf{\Gamma}^T \partial_{\mathbf{\Pi}} \phi_R]$$

denotes a family of (possibly non symmetric) second-order tensor-valued functions parametrized by $\mathbf{\Gamma}$. Accordingly, at a fixed temperature $\theta = \bar{\theta} \in (0, \theta_C)$

$$\dot{\mathbf{\Phi}} := \dot{\mathbf{\Xi}} + h(\bar{\theta}) \dot{\mathbf{\Pi}} = \partial_{\mathbf{C}} \mathbf{\Xi} \dot{\mathbf{C}} + [h(\bar{\theta}) \boldsymbol{\chi} + \partial_{\mathbf{\epsilon}} \mathbf{\Xi}] \dot{\mathbf{\epsilon}}.$$

Since this differential equation recalls the constitutive equation of hypoelastic media, we can say that it characterizes hypo-electroelastic materials.

A simple model. A special but significant class of hypo-electroelastic models is obtained assuming the free energy ψ be independent of $\mathbf{\Pi}$, as well as $\mathbf{\Xi}$. In this case $\partial_{\mathbf{\Pi}}\phi_R = -\mathbf{e}$ and

$$\boldsymbol{\chi}(\theta, \mathbf{C}, \mathbf{e}) = \boldsymbol{\Gamma} + \frac{1}{|\mathbf{e}|^2} \mathbf{e} \otimes [\mathbf{\Xi} + h\mathbf{\Pi} + \partial_{\mathbf{e}}\phi_R - \boldsymbol{\Gamma}^T \mathbf{e}]. \quad (40)$$

In the special case $\boldsymbol{\Gamma} = \mathbf{0}$ it follows

$$\boldsymbol{\chi}(\theta, \mathbf{C}, \mathbf{e}) = \frac{1}{|\mathbf{e}|^2} \mathbf{e} \otimes (\mathbf{\Xi} + h\mathbf{\Pi} + \partial_{\mathbf{e}}\phi_R).$$

Otherwise, if $\boldsymbol{\Gamma} \neq \mathbf{0}$ we can choose $\boldsymbol{\Gamma} = \hat{\boldsymbol{\Gamma}}(\theta, \mathbf{C}, \mathbf{e})$ such that

$$\mathbf{\Xi} + h\mathbf{\Pi} + \partial_{\mathbf{e}}\phi_R - \hat{\boldsymbol{\Gamma}}^T \mathbf{e} = \mathbf{0} \quad (41)$$

holds as an identity for any value of $\theta, \mathbf{C}, \mathbf{e}$. From (40) it follows $\boldsymbol{\chi} = \hat{\boldsymbol{\Gamma}}$, thus eliminating the dyadic term, and then

$$\dot{\mathbf{\Pi}} = \hat{\boldsymbol{\Gamma}}(\theta, \mathbf{C}, \mathbf{e}) \dot{\mathbf{e}}. \quad (42)$$

For definiteness, we exhibit a simple example assuming the quadratic free energy

$$\psi_R = \Psi_R(\theta, \mathbf{C}) + \frac{1}{2} \mathbf{e} \cdot \boldsymbol{\Upsilon}_{\mathbf{e}}(\theta, \mathbf{C}) \mathbf{e}, \quad \boldsymbol{\Upsilon}_{\mathbf{e}} = \boldsymbol{\Upsilon}_{\mathbf{e}}^T.$$

Since

$$\partial_{\mathbf{e}}\phi_R = \partial_{\mathbf{e}}[\psi_R - \mathbf{e} \cdot (\mathbf{\Xi} + h\mathbf{\Pi})] = \partial_{\mathbf{e}}\psi_R - \mathbf{\Xi} - \mathbf{e} \partial_{\mathbf{e}}\mathbf{\Xi} - h\mathbf{\Pi} = \boldsymbol{\Upsilon}_{\mathbf{e}} \mathbf{e} - \mathbf{\Xi} - \mathbf{e} \partial_{\mathbf{e}}\mathbf{\Xi} - h\mathbf{\Pi}$$

condition (41) reads

$$\boldsymbol{\Upsilon}_{\mathbf{e}} \mathbf{e} - \mathbf{e} \partial_{\mathbf{e}}\mathbf{\Xi} - \hat{\boldsymbol{\Gamma}}^T \mathbf{e} = \mathbf{0}$$

and we conclude that $\hat{\boldsymbol{\Gamma}}$ must solve the identity

$$\mathbf{e} [\partial_{\mathbf{e}}\mathbf{\Xi} - \boldsymbol{\Upsilon}_{\mathbf{e}} + \hat{\boldsymbol{\Gamma}}] = \mathbf{0}.$$

The arbitrariness of \mathbf{e} finally implies $\hat{\boldsymbol{\Gamma}} = \boldsymbol{\Upsilon}_{\mathbf{e}} - \partial_{\mathbf{e}}\mathbf{\Xi}$, so that (42) becomes

$$\dot{\mathbf{\Pi}} = (\boldsymbol{\Upsilon}_{\mathbf{e}} - \partial_{\mathbf{e}}\mathbf{\Xi}) \dot{\mathbf{e}}.$$

In isothermal conditions this result yields the simple constitutive differential equation

$$\dot{\mathfrak{P}} = \partial_{\mathbf{C}}\mathbf{\Xi} \dot{\mathbf{C}} + [(1-h)\partial_{\mathbf{e}}\mathbf{\Xi} + h\boldsymbol{\Upsilon}_{\mathbf{e}}] \dot{\mathbf{e}}.$$

Note that if $h = 0$ we have $\dot{\mathfrak{P}} = \dot{\mathbf{\Xi}}$, so that by choosing

$$\mathbf{\Xi} = \varepsilon_0 \chi(\theta) J \mathbf{C}^{-1} \mathbf{e}, \quad (43)$$

we recover the time derivative of the constitutive equation of a linear dielectric (19)₂.

Otherwise, if we choose $\partial_{\mathbf{e}}\mathbf{\Xi} = \boldsymbol{\Upsilon}_{\mathbf{e}}$, then it follows $\dot{\mathbf{\Pi}} = \mathbf{0}$. Assuming $\mathbf{\Pi}(0) = \mathbf{\Pi}_0$ we have $\mathbf{\Pi}(t) = \mathbf{\Pi}_0$ for all t and

$$\mathcal{P} = \mathcal{G}(\theta, \mathbf{S}, \mathcal{E}) + hJ^{-1} \mathbf{F} \mathbf{\Pi}_0, \quad \partial_{\mathcal{E}} \mathcal{G} = J^{-1} \mathbf{F} \boldsymbol{\Upsilon}_{\mathbf{e}} \mathbf{F}^T.$$

The special choice (43) leads to $\boldsymbol{\Upsilon}_{\mathbf{e}} = \varepsilon_0 \chi(\theta) J \mathbf{C}^{-1}$ and $\partial_{\mathcal{E}} \mathcal{G} = \varepsilon_0 \chi \mathbf{1}$. This condition allows us, for example, to assume that

$$\mathcal{G} = \varepsilon_0 \chi \mathcal{E} + \boldsymbol{\Lambda} \mathbf{S},$$

which therefore implies

$$\mathcal{P} = \varepsilon_0 \chi \mathcal{E} + \boldsymbol{\Lambda} \mathbf{S} + hJ^{-1} \mathbf{F} \mathbf{\Pi}_0.$$

For temperatures above the Curie value we have $h = 0$ and therefore we recover the piezoelectric constitutive equation. At lower temperatures, however, the behavior of the material is ruled by

$$\mathcal{P} = \varepsilon_0 \chi \mathcal{E} + \Lambda \mathbf{S} + h \mathcal{R}_0,$$

where $\mathcal{R}_0 = J^{-1} \mathbf{F} \mathbf{\Pi}_0$ denotes the initial remanent polarization.

5.2. Ferro-electroelastic materials. Now, in order to describe hysteretic phenomena, we analyze the relation (36) where $\tilde{\zeta}$ depends on $|\dot{\mathbf{e}}|$. In particular we can choose a linear dependence on $|\dot{\mathbf{e}}|$, namely

$$\tilde{\zeta} = \frac{1}{\theta \rho_R} \tilde{\zeta}_{\mathbf{e}}(\theta, \mathbf{C}, \mathbf{\Pi}) |\dot{\mathbf{e}}|, \quad \tilde{\zeta}_{\mathbf{e}} \geq 0.$$

Accordingly, (36) gives

$$(\partial_{\mathbf{e}} \phi_R + \mathbf{\Xi} + h \mathbf{\Pi}) \cdot \dot{\mathbf{e}} + \partial_{\mathbf{\Pi}} \phi_R \cdot \dot{\mathbf{\Pi}} = -\tilde{\zeta}_{\mathbf{e}} |\dot{\mathbf{e}}|.$$

The resulting differential equation yields a Duhem-like model that characterizes materials undergoing ferroelectric hysteresis and elastic deformations (keep in mind that here both ϕ and $\mathbf{\Xi}$ also depends on \mathbf{C}).

The representation formula (39) can be applied by letting

$$\mathcal{Y} = \mathbf{\Pi}, \quad \mathcal{X} = \mathbf{e}, \quad f = -\tilde{\zeta}_{\mathbf{e}} |\dot{\mathbf{e}}|, \quad \mathbf{A}_2 = \partial_{\mathbf{\Pi}} \phi_R, \quad \mathbf{A}_1 = \partial_{\mathbf{e}} \phi_R + \mathbf{\Xi} + h \mathbf{\Pi},$$

and assuming $\partial_{\mathbf{\Pi}} \phi_R \neq \mathbf{0}$. Hence it follows

$$\dot{\mathbf{\Pi}} = \left[\mathbf{\Gamma} - \frac{\mathbf{N} \otimes (\mathbf{\Xi} + h \mathbf{\Pi} + \partial_{\mathbf{e}} \phi_R + \mathbf{\Gamma}^T \partial_{\mathbf{\Pi}} \phi_R)}{|\partial_{\mathbf{\Pi}} \phi_R|} \right] \dot{\mathbf{e}} - \frac{\tilde{\zeta}_{\mathbf{e}} |\dot{\mathbf{e}}|}{|\partial_{\mathbf{\Pi}} \phi_R|} \mathbf{N}, \quad (44)$$

where $\mathbf{N} = \partial_{\mathbf{\Pi}} \phi_R / |\partial_{\mathbf{\Pi}} \phi_R|$. Now let $\tilde{\mathbf{\Gamma}}$ such that

$$\mathbf{\Xi} + h \mathbf{\Pi} + \partial_{\mathbf{e}} \phi_R = -\tilde{\mathbf{\Gamma}}^T \partial_{\mathbf{\Pi}} \phi_R. \quad (45)$$

Choosing $\mathbf{\Gamma} = \tilde{\mathbf{\Gamma}}$ we can write (44) in the simpler form not involving the dyadic product,

$$\dot{\mathbf{\Pi}} = \tilde{\mathbf{\Gamma}} \dot{\mathbf{e}} - \frac{\tilde{\zeta}_{\mathbf{e}} |\dot{\mathbf{e}}|}{|\partial_{\mathbf{\Pi}} \phi_R|^2} \partial_{\mathbf{\Pi}} \phi_R.$$

6. A ferro-electroelastic model with quadratic free energy

A special class of ferro-electroelastic materials is obtained assuming the free energy is a quadratic function of \mathbf{e} and $\mathbf{\Pi}$, namely

$$\psi_R = \Phi_R(\theta, \mathbf{C}) + \frac{1}{2} \mathbf{e} \cdot \mathbf{Y}_{\mathbf{e}}(\theta, \mathbf{C}) \mathbf{e} + \frac{1}{2} \mathbf{\Pi} \cdot h \mathbf{Y}_{\mathbf{\Pi}}(\theta, \mathbf{C}) \mathbf{\Pi} + \mathbf{e} \cdot h \mathbf{Y}(\theta, \mathbf{C}) \mathbf{\Pi},$$

where $\mathbf{Y}_{\mathbf{e}}$, $\mathbf{Y}_{\mathbf{\Pi}}$ and \mathbf{Y} are given second-order tensor. Moreover, $\mathbf{Y}_{\mathbf{e}}$ and $\mathbf{Y}_{\mathbf{\Pi}}$ are symmetric and $\mathbf{Y}_{\mathbf{\Pi}}$ is positive-definite. It follows

$$\phi_R = \Phi_R(\theta, \mathbf{C}) + \frac{1}{2} \mathbf{e} \cdot \mathbf{Y}_{\mathbf{e}}(\theta, \mathbf{C}) \mathbf{e} + \frac{1}{2} \mathbf{\Pi} \cdot h \mathbf{Y}_{\mathbf{\Pi}}(\theta, \mathbf{C}) \mathbf{\Pi} + \mathbf{e} \cdot h \mathbf{Y}(\theta, \mathbf{C}) \mathbf{\Pi} - \mathbf{e} \cdot (\mathbf{\Xi} + h \mathbf{\Pi})$$

and then

$$\partial_{\mathbf{e}} \phi_R = \partial_{\mathbf{e}} [\psi_R - \mathbf{e} \cdot (\mathbf{\Xi} + h \mathbf{\Pi})] = \mathbf{Y}_{\mathbf{e}} \mathbf{e} + h \mathbf{Y} \mathbf{\Pi} - \mathbf{\Xi} - \mathbf{e} \partial_{\mathbf{e}} \mathbf{\Xi} - h \mathbf{\Pi},$$

$$\partial_{\Pi} \phi_R = \partial_{\Pi} [\psi_R - \mathbf{e} \cdot (\mathbf{\Xi} + h\Pi)] = h[\mathbf{\Upsilon}_{\Pi}\Pi + \mathbf{\Upsilon}^T \mathbf{e} - \mathbf{e}].$$

Now we look for $\tilde{\mathbf{\Gamma}}$ satisfying (45), namely

$$\mathbf{\Upsilon}_{\mathbf{e}} \mathbf{e} - \mathbf{e} \partial_{\mathbf{e}} \mathbf{\Xi} + h\mathbf{\Upsilon}\Pi = -h\tilde{\mathbf{\Gamma}}^T (\mathbf{\Upsilon}_{\Pi}\Pi + \mathbf{\Upsilon}^T \mathbf{e} - \mathbf{e}).$$

Since $\mathbf{\Xi}$ is independent of Π , the arbitrariness of Π implies

$$h[\mathbf{\Upsilon} + \tilde{\mathbf{\Gamma}}^T \mathbf{\Upsilon}_{\Pi}] = \mathbf{0}, \quad \mathbf{\Upsilon}_{\mathbf{e}} \mathbf{e} - \mathbf{e} \partial_{\mathbf{e}} \mathbf{\Xi} + h\tilde{\mathbf{\Gamma}}^T (\mathbf{\Upsilon}^T - \mathbf{1}) \mathbf{e} = \mathbf{0}.$$

These conditions allow us to find either $h = 0$ and $\partial_{\mathbf{e}} \mathbf{\Xi} = \mathbf{\Upsilon}_{\mathbf{e}}$, or

$$\tilde{\mathbf{\Gamma}}^T = -\mathbf{\Upsilon}\mathbf{\Upsilon}_{\Pi}^{-1}, \quad \partial_{\mathbf{e}} \mathbf{\Xi} = \mathbf{\Upsilon}_{\mathbf{e}} - h(\mathbf{\Upsilon} - \mathbf{1})\mathbf{\Upsilon}_{\Pi}^{-1}\mathbf{\Upsilon}^T.$$

Note that the former occurrence leads to a para-magnetoelastic regime. In the latter case, instead, $\tilde{\mathbf{\Gamma}}$ need not to be symmetric, as well as $\mathbf{\Upsilon}$. Moreover it follows

$$\mathbf{\Xi}(\theta, \mathbf{C}, \mathbf{e}) = [\mathbf{\Upsilon}_{\mathbf{e}} + h(\mathbf{1} - \mathbf{\Upsilon})\mathbf{\Upsilon}_{\Pi}^{-1}\mathbf{\Upsilon}^T] \mathbf{e} + \mathbf{\Xi}_0(\theta, \mathbf{C}),$$

and choosing $\mathbf{\Gamma} = \tilde{\mathbf{\Gamma}}$ (44) becomes

$$\dot{\Pi} = -\mathbf{\Upsilon}_{\Pi}^{-1}\mathbf{\Upsilon}^T \dot{\mathbf{e}} - \frac{\tilde{\zeta}_{\mathbf{e}} |\dot{\mathbf{e}}|}{h|\mathbf{\Upsilon}_{\Pi}\Pi + (\mathbf{\Upsilon}^T - \mathbf{1})\mathbf{e}|^2} (\mathbf{\Upsilon}_{\Pi}\Pi + (\mathbf{\Upsilon}^T - \mathbf{1})\mathbf{e}).$$

For definiteness this expression simplifies by letting

$$\mathbf{\Upsilon}_{\mathbf{e}} = \varepsilon_0 \chi(\theta) \mathbf{J} \mathbf{C}^{-1}, \quad \mathbf{\Upsilon}_{\Pi} = \mathbf{\Upsilon}_{\mathbf{e}}^{-1}, \quad \mathbf{\Upsilon} = \beta \mathbf{1}, \quad \mathbf{\Xi}_0 = \mathbf{J} \mathbf{\mathcal{L}}(\theta) \mathbf{E},$$

with $\beta \in [0, 1]$, so that the evolution of the remanent polarization is ruled by

$$\dot{\Pi} = -\varepsilon_0 \beta \chi(\theta) \mathbf{J} \mathbf{C}^{-1} \dot{\mathbf{e}} - \frac{\tilde{\zeta}_{\mathbf{e}} |\dot{\mathbf{e}}|}{h|\mathbf{\Upsilon}_{\mathbf{e}}^{-1}\Pi + (\beta - 1)\mathbf{e}|^2} [\mathbf{\Upsilon}_{\mathbf{e}}^{-1}\Pi + (\beta - 1)\mathbf{e}]. \tag{46}$$

Moreover, denoting $\chi^* = [1 + h\beta(1 - \beta)]\chi$, we obtain

$$\mathbf{\Xi} = \varepsilon_0 \chi^* \mathbf{J} \mathbf{C}^{-1} \mathbf{e} + \mathbf{J} \mathbf{\mathcal{L}} \mathbf{E}, \quad \mathcal{G} = \varepsilon_0 \chi^* \mathcal{E} + \mathbf{A} \mathbf{S}.$$

Since $\chi^* = \chi$ at the extreme values $\beta = 0, 1$, these relations are consistent with (23)₂ and (24) which represent the direct piezoelectric effect in the material and spatial description, respectively.

Summarizing previous assumptions we can write

$$\psi_R = \Phi_R(\theta, \mathbf{C}) + \frac{1}{2} \varepsilon_0 \chi(\theta) \mathbf{J} \mathbf{C}^{-1} \mathbf{e} \cdot \mathbf{e} + \frac{1}{2} h [\varepsilon_0 \chi(\theta) \mathbf{J}]^{-1} \mathbf{C} \Pi \cdot \Pi + \beta h \mathbf{e} \cdot \Pi, \tag{47}$$

and the corresponding electric Gibbs free energy reads

$$\begin{aligned} \phi_R &= \psi_R - \mathbf{e} \cdot \mathfrak{P} = \Phi_R(\theta, \mathbf{C}) + \frac{1}{2} \varepsilon_0 (\chi - 2\chi^*) \mathbf{J} \mathbf{C}^{-1} \mathbf{e} \cdot \mathbf{e} \\ &\quad + \frac{1}{2} h [\varepsilon_0 \chi(\theta) \mathbf{J}]^{-1} \mathbf{C} \Pi \cdot \Pi + (\beta - 1) h \mathbf{e} \cdot \Pi - \mathbf{J} \mathbf{e} \cdot \mathbf{\mathcal{L}} \mathbf{E}. \end{aligned}$$

When $h = 0$ it is worth noting that $\chi^* = \chi$ and the piezoelectric free energy (25) is recovered from (47) by letting

$$\Phi_R(\theta, \mathbf{C}) = \Psi_R(\theta, \mathbf{C}) - \frac{1}{2} (\varepsilon_0 \chi)^{-1} \mathbf{J} \mathbf{C} \mathbf{\mathcal{L}} \mathbf{E} \cdot \mathbf{\mathcal{L}} \mathbf{E}.$$

According to (35), the corresponding ferro-electroelastic stress takes the form

$$\mathbf{T}_{RR} = 2\partial_{\mathbf{C}}\Psi_R(\theta, \mathbf{C}) + \mathfrak{T}_0(\theta, \mathbf{C}) + \mathfrak{T}_{\mathbf{e}}(\theta, \mathbf{C}, \mathbf{e}) + h\mathfrak{T}_{\Pi}(\theta, \mathbf{C}, \Pi) - h\mathbf{C}^{-1}\mathbf{e} \otimes \Pi, \quad (48)$$

where

$$\begin{aligned} \mathfrak{T}_0(\theta, \mathbf{C}) &= -\frac{1}{2}(\varepsilon_0\chi)^{-1}J(\mathbf{C}\mathbf{L}\mathbf{E} \cdot \mathbf{L}\mathbf{E})\mathbf{C}^{-1} - (\varepsilon_0\chi)^{-1}J\mathbf{L}\mathbf{E} \otimes \mathbf{L}\mathbf{E} - 2(\varepsilon_0\chi)^{-1}J\mathbf{L}^T(\mathbf{C}\mathbf{L}\mathbf{E}), \\ \mathfrak{T}_{\mathbf{e}}(\theta, \mathbf{C}, \mathbf{e}) &= \frac{1}{2}\varepsilon_0(\chi - 2\chi^*)J(\mathbf{C}^{-1}\mathbf{e} \cdot \mathbf{e})\mathbf{C}^{-1} - \varepsilon_0(\chi - \chi^*)J\mathbf{C}^{-1}\mathbf{e} \otimes \mathbf{C}^{-1}\mathbf{e} - J\mathbf{C}^{-1}\mathbf{e} \otimes \mathbf{L}\mathbf{E} \\ &\quad - J(\mathbf{e} \cdot \mathbf{L}\mathbf{E})\mathbf{C}^{-1} - J\mathbf{L}^T\mathbf{e}, \\ \mathfrak{T}_{\Pi}(\theta, \mathbf{C}, \Pi) &= -\frac{1}{2}[\varepsilon_0\chi J]^{-1}(\mathbf{C}\Pi \cdot \Pi)\mathbf{C}^{-1} + [\varepsilon_0\chi J]^{-1}\Pi \otimes \Pi. \end{aligned}$$

When $\beta = 0, 1$ it is worth noting that $\mathfrak{T}_{\mathbf{e}}$ reduces to

$$\mathfrak{T}_{\mathbf{e}} = -\frac{1}{2}\varepsilon_0\chi J(\mathbf{C}^{-1}\mathbf{e} \cdot \mathbf{e})\mathbf{C}^{-1} - J\mathbf{C}^{-1}\mathbf{e} \otimes \mathbf{L}\mathbf{E} - J(\mathbf{e} \cdot \mathbf{L}\mathbf{E})\mathbf{C}^{-1} - J\mathbf{L}^T\mathbf{e}.$$

6.1. One-dimensional ferro-electroelastic hysteresis. Finally, we restrict attention to small deformations at a constant temperature $\theta \in (0, \theta_c)$, so that $h \in (0, 1)$. Moreover, for simplicity we assume $\beta = 0$. Accordingly, $J \approx 1$, $\mathbf{C}, \mathbf{B} \approx \mathbf{1}$ and $\mathbf{E}, \mathbf{S} \approx \mathbf{e}$ from which it follows $\mathbf{L}\mathbf{E} \approx \mathbf{\Lambda}\mathbf{e}$ and

$$\mathcal{P} = \varepsilon_0\chi\mathcal{E} + \mathbf{\Lambda}\mathbf{e} + h\mathcal{R}.$$

Then, the usual assumption (27) allows (47) to be approximated as follows

$$\psi_R \approx \frac{1}{2}\mathbf{e} \cdot \mathbf{C}\mathbf{e} - \frac{1}{2}(\varepsilon_0\chi)^{-1}|\mathbf{\Lambda}\mathbf{e}|^2 + \frac{1}{2}\varepsilon_0\chi|\mathcal{E}|^2 + \frac{1}{2}h(\varepsilon_0\chi)^{-1}|\mathcal{R}|^2.$$

Since $|\mathbf{\Lambda}\mathbf{e}|^2 = \mathbf{e} \cdot \mathbf{\Lambda}^T\mathbf{\Lambda}\mathbf{e}$, letting $\mathbf{K} = \mathbf{C} - (\varepsilon_0\chi)^{-1}\mathbf{\Lambda}^T\mathbf{\Lambda}$ as in (29), the (approximate) stress representation corresponding to (48) becomes

$$\begin{aligned} \mathbf{T}_{RR} &\approx \mathbf{K}\mathbf{e} - \frac{1}{2}\varepsilon_0\chi|\mathcal{E}|^2\mathbf{1} - \mathcal{E} \otimes \mathbf{\Lambda}\mathbf{e} - (\mathcal{E} \cdot \mathbf{\Lambda}\mathbf{e})\mathbf{1} - \mathbf{\Lambda}^T\mathcal{E} \\ &\quad - \frac{1}{2}h(\varepsilon_0\chi)^{-1}|\mathcal{R}|^2\mathbf{1} + h(\varepsilon_0\chi)^{-1}\mathcal{R} \otimes \mathcal{R} - h\mathcal{E} \otimes \mathcal{R}. \end{aligned} \quad (49)$$

For definiteness, let consider a transversely isotropic piezoelectric solid (e.g. PZT-4 piezoceramic, barium titanate) with symmetry axis (easy axis) parallel to the unit vector \mathbf{e}_1 . When uniaxial anisotropy occurs, we are allowed to assume the electric field \mathcal{E} and the residual polarization \mathcal{R} are in the direction of easy axis \mathbf{e}_1 (Schröder and Romanowski 2005). Hence we put

$$\mathcal{E} = \mathcal{E}\mathbf{e}_1, \quad \mathcal{E} = |\mathcal{E}|, \quad \mathcal{R} = \mathcal{R}\mathbf{e}_1, \quad \mathcal{R} = |\mathcal{R}|.$$

After taking the orthonormal basis $(\mathbf{e}_1, \mathbf{e}_2, \mathbf{e}_3)$, we represent the deformation gradient in the form

$$\mathbf{F} = \text{diag}(1 + \gamma, 1 - \delta, 1 - \delta),$$

where γ and δ are the longitudinal and transverse strains, respectively, and $|\gamma|, |\delta| \ll 1$. Consequently,

$$\mathbf{E}, \mathbf{S} \approx \mathbf{e} = \text{diag}(\gamma, -\delta, -\delta), \quad \mathbf{e} = \mathbf{F}^T\mathcal{E} \approx \mathcal{E}\mathbf{e}_1, \quad \Pi = J\mathbf{F}^{-1}\mathcal{R} \approx \mathcal{R}\mathbf{e}_1.$$

The resulting scheme allows us to look at a one-dimensional setting. Letting $\beta = 0$ and applying previous assumptions, Eq. (46) becomes

$$\dot{\mathcal{R}} = -\frac{\varepsilon_0 \chi \zeta_{\mathcal{E}} |\dot{\mathcal{E}}|}{h[\mathcal{R} - \varepsilon_0 \chi \mathcal{E}]} \tag{50}$$

This differential relation is rate independent and rules the evolution of the remanent polarization \mathcal{R} depending on the choice of the dissipation function $\zeta_{\mathcal{E}}$. In order to scrutinize the evolution of the total polarization \mathcal{P} we have to take into account the form of $\mathbf{\Lambda}$ for a transversely isotropic body. Since

$$\mathbf{\Lambda} = \begin{bmatrix} \Lambda_{11} & \Lambda_{12} & \Lambda_{12} & 0 & 0 & 0 \\ 0 & 0 & 0 & \Lambda_{24} & 0 & 0 \\ 0 & 0 & 0 & 0 & \Lambda_{24} & 0 \end{bmatrix}, \quad \mathbf{\Lambda}^T = \begin{bmatrix} \Lambda_{11} & 0 & 0 \\ \Lambda_{12} & 0 & 0 \\ \Lambda_{12} & 0 & 0 \\ 0 & \Lambda_{24} & 0 \\ 0 & 0 & \Lambda_{24} \\ 0 & 0 & 0 \end{bmatrix}$$

we obtain $\mathcal{P} = \mathcal{P} \mathbf{e}_1$ and

$$\mathcal{P} = \varepsilon_0 \chi \mathcal{E} + \Lambda_{11} \gamma - 2\Lambda_{12} \delta + h\mathcal{R}. \tag{51}$$

Finally, to express the relation between γ , δ and \mathcal{E} , \mathcal{R} we consider the thickness mode (both stress and electric field are along the easy direction \mathbf{e}_1), so that

$$\mathbf{T} = \text{diag}(\tau, 0, 0),$$

and assume the tension τ constant over time. Using the Mandel-Voigt notation for symmetric tensors, we have

$$\boldsymbol{\varepsilon} = (\gamma, -\delta, -\delta, 0, 0, 0)^T, \quad \mathbf{T} = (\tau, 0, 0, 0, 0, 0)^T.$$

Moreover, for a transversely isotropic body

$$\mathbf{C} = \begin{bmatrix} C_{11} & C_{12} & C_{12} & 0 & 0 & 0 \\ C_{12} & C_{22} & C_{23} & 0 & 0 & 0 \\ C_{12} & C_{23} & C_{22} & 0 & 0 & 0 \\ 0 & 0 & 0 & C_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & C_{44} & 0 \\ 0 & 0 & 0 & 0 & 0 & \frac{1}{2}(C_{23} - C_{22}) \end{bmatrix}$$

so that it follows

$$\mathbf{K} = \mathbf{C} - (\varepsilon_0 \chi)^{-1} \mathbf{\Lambda}^T \mathbf{\Lambda} = \begin{bmatrix} K_{11} & K_{12} & K_{12} & 0 & 0 & 0 \\ K_{12} & K_{22} & K_{23} & 0 & 0 & 0 \\ K_{12} & K_{23} & K_{22} & 0 & 0 & 0 \\ 0 & 0 & 0 & K_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & K_{44} & 0 \\ 0 & 0 & 0 & 0 & 0 & K_{66} \end{bmatrix}$$

where

$$\begin{aligned} K_{11} &= C_{11} - (\varepsilon_0 \chi)^{-1} \Lambda_{11}^2, & K_{12} &= C_{12} - (\varepsilon_0 \chi)^{-1} \Lambda_{11} \Lambda_{12}, \\ K_{22} &= C_{22} - (\varepsilon_0 \chi)^{-1} \Lambda_{12}^2, & K_{23} &= C_{23} - (\varepsilon_0 \chi)^{-1} \Lambda_{12}^2, \\ K_{44} &= C_{44} - (\varepsilon_0 \chi)^{-1} \Lambda_{24}^2, & K_{66} &= \frac{1}{2}(C_{23} - C_{22}). \end{aligned}$$

Consequently, from (49) we obtain

$$\begin{aligned} [K_{11} - 2\Lambda_{11}\mathcal{E}] \gamma - 2[K_{12} - 2\Lambda_{12}\mathcal{E}] \delta &= \Lambda_{11}\mathcal{E} + \frac{1}{2}\varepsilon_0\chi|\mathcal{E}|^2 - \frac{h}{2}(\varepsilon_0\chi)^{-1}|\mathcal{R}|^2 + h\mathcal{E}\mathcal{R} + \tau, \\ [K_{12} - \Lambda_{11}\mathcal{E}] \gamma - [K_{22} + K_{23} - 2\Lambda_{12}\mathcal{E}] \delta &= \Lambda_{12}\mathcal{E} + \frac{1}{2}\varepsilon_0\chi|\mathcal{E}|^2 + \frac{h}{2}(\varepsilon_0\chi)^{-1}|\mathcal{R}|^2. \end{aligned} \quad (52)$$

This system admits one and only one solution γ, δ provided that

$$\Delta = [2K_{12}^2 - K_{11}(K_{22} + K_{23})] + 2[\Lambda_{12}(K_{11} - 2K_{12}) + \Lambda_{11}(K_{22} + K_{23} - K_{12})]\mathcal{E} \neq 0.$$

This condition must hold for all \mathcal{E} (including $\mathcal{E} = 0$) and then in turns implies that

$$2K_{12}^2 - K_{11}(K_{22} + K_{23}) \neq 0.$$

If, in addition,

$$\Lambda_{12}(K_{11} - 2K_{12}) + \Lambda_{11}(K_{22} + K_{23} - K_{12}) \neq 0$$

then $\Delta \neq 0$ provides $\mathcal{E} \neq \tilde{\mathcal{E}}$, where

$$\tilde{\mathcal{E}} = \frac{K_{11}(K_{22} + K_{23}) - 2K_{12}^2}{2[\Lambda_{12}(K_{11} - 2K_{12}) + \Lambda_{11}(K_{22} + K_{23} - K_{12})]}.$$

When a given tension τ is kept constant and the electric field oscillates with amplitude \mathcal{E}_0 (namely, $\mathcal{E}(t) = \mathcal{E}_0 \cos \omega t$) the evolution of the electric polarization is ruled by (51), (52) and the solution of differential system

$$\begin{cases} \dot{\mathcal{R}} = -\frac{\varepsilon_0\chi\tilde{\zeta}\mathbf{e}|\omega\mathcal{E}_0 \sin \omega t|}{h[\mathcal{R} - \varepsilon_0\chi\mathcal{E}_0 \cos \omega t]} \\ \dot{\mathcal{E}} = -\omega\mathcal{E}_0 \sin \omega t. \end{cases}$$

for a given initial value $\mathcal{R}(0) = \mathcal{R}_0$.

7. Conclusions

In this paper the principle of objectivity is satisfied by using invariant fields as independent variables in constitutive functions. By virtue of this, the standard balance of torques (16), which states that the skew part of the stress equals the skew part of the dyadic product between \mathcal{P} and \mathcal{E} , is a consequence of thermodynamic restrictions. Moreover, due to the form (5) of the power, the pair $\mathfrak{P}, \mathfrak{E}$ defined in (4) seems to be the most convenient pair of invariant fields to describe the electric behaviour in deformable bodies.

Dielectric and piezoelectric constitutive equations are derived by suitable choices of the free energy potential, (18) and (22) respectively. Compatibility with thermodynamics is implemented following to the Coleman-Noll procedure. It is worth noting that the expression of the free energy potentials in the material description naturally involves the right Cauchy-Green tensor, thus leading to the electrostriction effect (21) as a consequence of thermodynamic restrictions. We note that in piezoelectric materials undergoing small deformation, the stress (28) gets a contribution linear in the electric field in addition to the dependence on the squared electric field which is involved in normal electrostrictive materials. However, if we neglect nonlinear terms, the stress reduces to the usual symmetrical (approximate) form (29).

In order to model ferro-electroelastic hysteresis the ferroelectric polarization vector \mathcal{P} is decomposed into a reversible (piezoelectric) and a residual (remanent) part. All the constitutive functions are assumed to depend on the (invariant) residual polarization $\mathbf{\Pi}$, as well as on the electric field, temperature and deformation of the material. By means of the representation formulas (38) and (39) we derive the explicit rate-type evolution equation for the polarization vector for hypo-electroelastic and ferro-electroelastic materials.

The coupling between electric field and mechanical strain is described under the small deformation assumption. Cauchy stress (48) and inelastic deformation are directly related to the ferroelectric polarization and implemented in this model by a suitable quadratic free energy potential (47). Finally, one-dimensional ferro-electroelastic hysteresis at constant stress is fully described in a transversely isotropic body.

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