Contents

1 Introduction **2**

2 Maxwell stress and Toupin's formulation **6**

3 Energy formulation and associated boundary value problems **9**
3.1 Free energy of the system 9
3.2 Euler-Lagrange equations and boundary conditions 11
3.3 Geometric and constitutive linearization: general procedure 16

4 Theories of typical media and applications **17**
4.1 Nonlinear dielectric elastomer 17
4.2 Photoelasticity 19
4.3 Piezoelectricity 20
4.4 Ferroelectrics 21
4.5 Polarization gradient theory 22
4.6 Theory of flexoelectricity 23

5 Generalization to include magnetization **24**
5.1 Free energy and boundary value problems 24
5.2 Nonlinear magnetic elastomers 25
5.3 Magneto-electric materials 26
5.4 Piezo-magneto-electric materials 27

6 Applications **27**
6.1 Stretching of a soft ellipsoid in an external electric field 27
6.2 Magnetoelectric effects of magneto-electric elastomers 30
6.3 Bending of flexoelectric films 34

7 Summary and discussion **36**

8 Appendix **36**
A Higher order variational calculations 36
B Interfacial conditions 37
C Alternative variational formulations of the nongradient theory 38

An energy formulation of continuum magneto-electro-elasticity with applications

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Abstract

We present an energy formulation of continuum electro-elasticity and magneto-electro-elasticity. Based on the principle of minimum free energy, we propose a form of total free energy of the system in three dimensions, and then systematically derive the theories for a hierarchy of materials including dielectric elastomers, piezoelectric ceramics, ferroelectrics, flexoelectric materials, magnetic elastomers, magneto-electric materials, piezo-electric-magnetic materials among others. The effects of mechanical, electrical and magnetic boundary devices, external charges, polarizations and magnetization are taken into account in formulating the free energy. The linear and nonlinear boundary value problems governing these materials are explicitly derived as the Euler-Lagrange equations of the principle of minimum free energy. Finally, we illustrate the applications of the formulations by presenting solutions to a few simple problems and give an outlook of potential applications.

1 Introduction

The classic subjects of electrostatics, magnetostatics and elasticity have been firmly established in the last few centuries. For a continuum deformable body $\Omega \subset \mathbb{R}^3$, the electric, magnetic and elastic state of the body in static equilibrium necessarily satisfy the Maxwell equations and mechanical balance laws:

$$\mathrm{curl}\mathbf{e} = 0, \qquad \mathrm{div}\mathbf{d} = \rho^e, \qquad \mathbf{d} = \epsilon_0\mathbf{e} + \mathbf{p} + \mathbf{p}^e, \tag{1.1a}$$

$$\mathrm{curl}\mathbf{h} = 0, \qquad \mathrm{div}\mathbf{b} = 0, \qquad \mathbf{b} = \mu_0(\mathbf{h} + \mathbf{m} + \mathbf{m}^e), \tag{1.1b}$$

$$\mathbf{F} = \mathrm{Grad}\boldsymbol{\chi}, \qquad \mathrm{div}\boldsymbol{\sigma}_{\mathrm{tot}} = \mathbf{f}^e, \qquad \boldsymbol{\sigma}_{\mathrm{tot}} = \boldsymbol{\sigma}_{\mathrm{tot}}^T. \tag{1.1c}$$

Here, what the symbols stand for is as follows: $\mathbf{e}$ - electric field, $\mathbf{d}$ - electric displacement, $\mathbf{p}$ (resp. $\mathbf{p}^e$) - intrinsic (resp. external) polarization, ρ^e - external charge density; $\mathbf{h}$ - magnetic field, $\mathbf{b}$ - magnetic flux, $\mathbf{m}$ (resp. $\mathbf{m}^e$) - intrinsic (resp. external) magnetization; $\boldsymbol{\chi}$ - deformation, $\mathbf{F}$ - deformation gradient, $\boldsymbol{\sigma}_{\mathrm{tot}}$ -total stress, $\mathbf{f}^e$ - external body force; ϵ_0 (resp. μ_0) - electric permittivity (resp. magnetic permeability) of vacuum. The classic uncoupled theories of electrostatics,

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magnetostatics and elasticity for continuum media are completed by providing constitutive laws such as

$$\mathbf{d} = \mathbf{d}(\mathbf{p}), \qquad \mathbf{b} = \mathbf{b}(\mathbf{m}), \qquad \boldsymbol{\sigma}_{\rm tot} = \boldsymbol{\sigma}_{\rm tot}(\mathbf{F}).$$

In the era of nanotechnology and bioengineering, much of attention has been focused on multifunctional or multiferroic materials where strain, polarization and magnetization are simultaneously coupled. These materials have broad applications ranging from the technologies of actuators, sensors, imaging devices to smart self-adaptive structures, artificial muscles, etc (Fiebig, 2005; Nan et al., 2008). Further, to address novel phenomena observed in complex and heterogeneous systems such as granular media and composites, it is often necessary to consider nonlocal effects that motivate gradient theories. These issues have been addressed in thematic topics of strain gradient theory (Fleck et al., 1994), polarization gradient theory (Mindlin, 1968; Buchanan et al., 1989), flexoelectricity among others (Tagantsev, 1986; Majdoub et al., 2008). The literature, however, lacks of a unified treatment that considers general magneto-electro-elastic materials and their gradient effects.

Since the seminal definitive work of Toupin (1956, 1959) that will be briefly reviewed in § 2, the key ingredients of a theory for electro-elastic materials have been well understood and are worthwhile to mention. First, a somewhat peculiar stress term, namely, the Maxwell stress, emerges from electrostatic field such that the elastic state and electric state of the body are intrinsically coupled. As a nonlinear function of electric field, the Maxwell stress gives rise to substantial difficulty in solving a generic boundary value problem concerning an electro-elastic body. A second important issue, as always, lies in the formulating generic nonlinear constitutive laws that relate elastic, electric and magnetic quantities and guarantees reasonable physical behaviors. Toupin (1956) and many subsequent authors (Eringen, 1963; Mindlin, 1968; McMeeking and Landis, 2005; Suo et al., 2008; Tian, 2007; Tian *et al.*, 2012) propose such constitutive relations by postulating a special form of the stored (or internal) energy function, and then systematically restrict the form of the internal energy function, expand and truncate the internal energy function that will eventually yield simple, possibly linear, constitutive relations. Toupin (1956) in fact began his theory by deriving the basic field equations from the Maxwell equations and mechanical balance laws, and then formulated the constitutive relations by postulating a stored energy function and showed the equivalence between the field equations and the *principle of virtual work*.

Nowadays, as we understand that the *principle of virtual work (or power)* can in general be regarded as the weak form of a variational principle, it is of significant interest to have a variational formulation for magneto-electro-elastic bodies based on the physical free energy. Since our main goal is to identify the static equilibrium of the body at a constant temperature, for clarity we will assume that the body is at a constant temperature, has constant entropy, and hence the equilibrium state of the body is dictated by the *principle of minimum free energy* (Gibbs, 1878, pg. 109; Ericksen, 1991, ch. 1). For a continuum body, the employment of the *principle of minimum free energy* requires two critical hypotheses: (i) the set of thermodynamic variables that completely describe the state of the continuum body and their possible variations, and (ii) the total free energy of the body when the body is interacting with well-defined boundary devices. It is fruitless to justify these hypotheses in the framework of continuum theory, though atomistic models may shed light on the foundation of these hypotheses.

We therefore begin our energy formulation for a magneto-electro-elastic body with these two hypotheses whereas the rest of theory is mathematically deducted. Also, the Maxwell equations (1.1a)-(1.1b) are taken as premises but the concept of stresses including the Maxwell stress are regarded as derived notions. The advantages of such a variational formulation based on the principle of minimum free energy include: (i) embracing the framework of Gibbs, the proposed energy

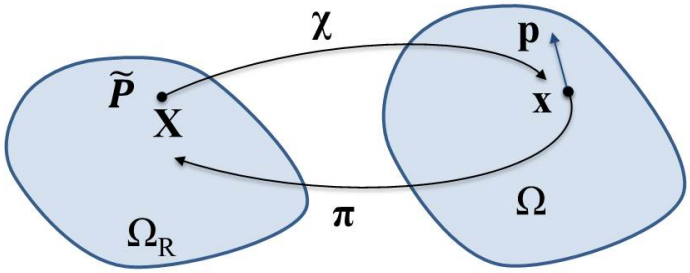
formulation admits clear thermodynamic interpretation and may even be proved by the Second Law in a proper setup (Fosdick and Tang 2007); (ii) we no longer need to forcefully separate the total stress into the *local* mechanical stress and the *nonlocal* Maxwell stress. The Maxwell stress will emerge naturally from the first variation of the total free energy. This calculation also explains the relation between different forms of Maxwell stress in the literature; (iii) novel physical phenomena such as the gradient effects of strain, polarization and magnetization can be treated uniformly without introducing additional constitutive relations. All constitutive assumptions will be lumped into the form of stored/internal energy function of the material; and finally but not the least important, (iv) in regard of recent development of Γ-convergence and homogenization, the proposed energy formulation can be directly applied to rigorously *derive* a hierarchy of theories at different scaling limits (Tian, 2007) and for lower dimensional bodies (§ 6.3), and to obtain bounds on the effective properties of composites (Milton, 2012; Liu, 2013).

In addition, we notice that the proposed energy formulation is closely related with the phenomenological theories of Landau (Landau and Liftshitz, 1935; Landau, 1937; Ginzburg and Landau, 1950) concerning phase transitions of ferromagnetic, ferroelectric and superconducting materials, and the theory of austenite-martensite phase transition (James and Wuttig, 1998). The formulation is also closely related with the Hashin-Shtrikman's variational principle that is important for finding bounds on the effective properties of composites (Hashin-Shtrikman, 1962). As discussed in Liu (2013), the field equations (1.1) in general may enjoy many different variational formulations whose Euler-Lagrange equations are consistent with the field equations (1.1); neither the energy functional nor the independent state variables has to be the same. However, for evolution problems [1] and stability analysis (Chen, 2009; Xu *et al.*, 2010; Dorfmann and Ogden, 2010), we have to identify the physical free energy and possible variations of state variables based on the physical gound. A wrong choice may yield erroneous or even opposite results (Liu, 2013, § 6; Bustamante and Merodio, 2012).

Three solutions of the proposed formulation are presented in § 6. We highlight here a few interesting results and potential applications. In § 6.1 we address the equilibrium shape of a soft nonlinear elastic ellipsoid in an applied electric field. The nonlinear boundary value problem is simplified by assuming uniform deformation gradient and then explicitly solved by utilizing a special property of ellipsoids. The final shape of the body may be regarded as the "best-fitting" ellipsoid of the actual shape. This strategy can be similarly used to address the equilibrium shape and relaxation of vesicles and droplets in an electric field where surface energy becomes predominant (Zhang *et al.*, 2013). In § 6.2 we show a strain-mediated magnetoelectric effects of soft materials. In a simple one-dimensional setting, we solve explicitly the boundary value problem for a magneto-electro-elastic elastomer. Though the elastomer has neither intrinsic magnetoelectric effect nor piezoelectric piezomagnetic effects, magnetoelectricity, i.e., the induction of electric field by magnetic field and vice versa, arises from the Maxwell stress and geometric nonlinear effect. We finally formally derive the flexoelectric theory for a thin plate from the three-dimensional theory of flexoelectricity in § 6.3. This calculation is reminiscent of the classic derivation of Kirchhoff-Love plate theory from elasticity in three dimensions.

The paper is organized as follows. In § 2 we briefly review the classic works of Toupin (1956, 1959) and point out issues associated with the concept of Maxwell stress. In § 3 we postulate a form of the stored/internal energy function, independent state variables for the body, identity the total free energy of the body and derive the Euler-Lagrange equations and boundary conditions for equilibrium states. In this section we assume there is no magnetic coupling for clarity. The

[1] For such evolution problems, a phenomenological kinetic law, e.g., velocity $\propto$ driving force, has to be postulated to close the system, see e.g. Abeyaratne and Knowles (2006, pg. 50).



(a) Reference config. **(b)** Current config.

Figure 1: Reference configuration and current configuration of a deformable body

boundary value problems including the interfacial conditions between different type of materials are explicitly stated in § 3.2. We also outline the general procedure to linearize a general nonlinear theory for small strain and small polarization in § 3.3. In § 4 we systematically expand and truncate the internal energy function conforming to the fundamental restrictions of frame indifference and material symmetries. In particular, we identify the internal energy function and the boundary value problems for elastic elastomers (§ 4.1), isotropic photoelastic materials (§ 4.2), linearized piezoelectric materials (§ 4.3), ferroelectric materials (§ 4.4), polarization gradient theory (§ 4.5) and the strain gradient or flexoelectric theory (§ 6.3). From § 5 we begin to include magnetic coupling, and identify the internal energy function and the boundary value problems for magnetic elastomers (§ 5.2), magnetoelectric materials (§ 5.3) and piezo-magneto-electric materials (§ 5.4). In § 6 we present a few applications of the proposed theory.

Notation. Consider a deformable continuum body as shown in Fig. 1. Let Ω_R be the reference configuration, $\boldsymbol{\chi} : \Omega_R \to \Omega$ be the deformation carrying every material point in the reference configuration to a spatial point in the current configuration, and $\boldsymbol{\pi} : \Omega \to \Omega_R$ be the inverse mapping. As standard in continuum mechanics, a material point in the reference (current) configuration is denoted by Lagrangian coordinate $\mathbf{X}$ (Eulerian coordinate $\mathbf{x}$). The operators $\mathrm{Grad}, \mathrm{Div}, \mathrm{Curl}$ and $\mathrm{grad}, \mathrm{div}, \mathrm{curl}$ are taken with respect to Lagrangian coordinate $\mathbf{X}$ and Eulerian coordinate $\mathbf{x}$, respectively. Let $\mathbf{u}$ be a vector field with components u_p and $\mathbf{A}$ be a tensor field with components A_{pi}. We follow the usual convention that $(\mathrm{Grad}\mathbf{u})_{pi} = u_{p,X_i}$ and $(\mathrm{Div}\mathbf{A})_p = A_{pi,X_i}$, .

Further, let $\mathbf{F} = \mathrm{Grad}\boldsymbol{\chi}$ be the deformation gradient, $\mathbf{C} = \mathbf{F}^T\mathbf{F}$ be the Cauchy-Green strain tensor and $J = \det \mathbf{F}$ be the Jacobian. Quantities in current configuration are denoted by lower cases, e.g., the electric field $\mathbf{e}$, electric displacement $\mathbf{d}$, and polarization $\mathbf{p}$ (per unit volume), mechanical stress $\boldsymbol{\sigma}$, and (electrical) Maxwell stress $\boldsymbol{\sigma}_{\mathrm{MW}}$. The pull-back of these quantities are denoted by upper cases:

$$\mathbf{E} = \mathbf{e} \circ \boldsymbol{\chi}, \quad \mathbf{D} = \mathbf{d} \circ \boldsymbol{\chi}, \quad \mathbf{P} = \mathbf{p} \circ \boldsymbol{\chi}, \quad \boldsymbol{\Sigma} = \boldsymbol{\sigma} \circ \boldsymbol{\chi}, \quad \boldsymbol{\Sigma}_{\mathrm{MW}} = \boldsymbol{\sigma}_{\mathrm{MW}} \circ \boldsymbol{\chi}.$$

Further, in analogy with the familiar concept of Piola-Kirchhoff stress, the differential equations satisfied by electric field, electric displacement and total stress are much simpler and physical meanings are more evident for the so-called *nominal quantities*. For example,

$$\tilde{\mathbf{D}} = J\mathbf{F}^{-1}\mathbf{D} \tag{1.2}$$

is the nominal electric displacement such that the charge enclosed in any subbody $U \subset \Omega$ is given

by

$$Q = \int_{\partial U} \mathbf{d} \cdot \mathbf{n} ds = \int_{\partial U_R} \tilde{\mathbf{D}} \cdot \mathbf{N} dS, \tag{1.3}$$

where $U_R = \boldsymbol{\pi}(U)$ is the subbody in the reference configuration, and $\mathbf{n}$ (resp. $\mathbf{N}$) is the unit outward normal on the interface ∂U (resp. ∂U_R). We also define

$$\tilde{\mathbf{E}} = \mathbf{F}^T \mathbf{E}, \qquad \tilde{\boldsymbol{\Sigma}} = J \boldsymbol{\Sigma} \mathbf{F}^{-T}, \qquad \tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}} = J \boldsymbol{\Sigma}_{\mathrm{MW}} \mathbf{F}^{-T}, \qquad \tilde{\mathbf{P}} = J \mathbf{P}. \tag{1.4}$$

At the presence of external charge density $\rho^e : \Omega \to \mathbb{R}$ and external body force $\mathbf{f}^e : \Omega \to \mathbb{R}^3$, by the Maxwell equations and balance laws (1.1) we have

$$\mathrm{curl} \mathbf{e} = 0, \quad \mathrm{div} \mathbf{d} = \rho^e, \quad \mathbf{d} = \epsilon_0 \mathbf{e} + \mathbf{p}, \quad -\mathrm{div}(\boldsymbol{\sigma} + \boldsymbol{\sigma}_{\mathrm{MW}}) = \mathbf{f}^e \qquad \text{in } \Omega. \tag{1.5}$$

The first of the above equation immediately implies that there exists a scalar potential ξ such that $\mathbf{e} = -\mathrm{grad} \xi$. By (1.3) or the chain rule we immediately find that

$$\mathrm{Curl} \tilde{\mathbf{E}} = 0, \quad \mathrm{Div} \tilde{\mathbf{D}} = \tilde{\rho}^e, \quad \tilde{\mathbf{D}} = \epsilon_0 J \mathbf{C}^{-1} \tilde{\mathbf{E}} + \mathbf{F}^{-1} \tilde{\mathbf{P}}, \quad -\mathrm{Div}(\tilde{\boldsymbol{\Sigma}} + \tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}}) = \tilde{\mathbf{f}}^e \qquad \text{in } \Omega_R, \tag{1.6}$$

where $\tilde{\mathbf{E}} = -\mathrm{Grad} \xi$,

$$\tilde{\rho}^e = J \rho^e \circ \boldsymbol{\chi}, \qquad \tilde{\mathbf{f}}^e = J \mathbf{f}^e \circ \boldsymbol{\chi}. \tag{1.7}$$

As one may notice above, we will mostly use direct notion for brevity and transparency in physical meaning; the Einstein summation over repeated indices is assumed if index notation is in use. The symbol D will be used for differentiating the internal energy function with respect to independent state variables. Scalar product (i.e., dot product) between two matrices $\mathbf{A}, \mathbf{B}$ of the same size are defined as $\mathbf{A} \cdot \mathbf{B} = \mathrm{Tr}(\mathbf{A}\mathbf{B}^T) = A_{ij} B_{ij}$. Domains are assumed to be open with smooth boundaries. The thrust of this work mainly comes from the need of a consistent theory for modeling magneto-electro-elastic materials instead of mathematical analysis. Therefore, the rigor is comprised to some extent that we do not normally state conditions of integrability and differentiability.

2 Maxwell stress and Toupin's formulation

Let $V \subset \mathbb{R}^3$ be a region containing a smooth distribution of charge $\rho : V \to \mathbb{R}$ and polarization $\mathbf{p} : V \to \mathbb{R}^3$. From the very definition of electric field, one may see that electrostatics is coupled with mechanics. From the Maxwell's work, it is well-known that an electrostatic field is given by $\mathbf{e} = -\mathrm{grad} \xi$ for some scalar potential ξ and that the resultant electrostatic force on the charges and dipoles in a fixed *spatial* region $U \subset V$ can be written as

$$\mathbf{f}_U = \int_{\partial U} \boldsymbol{\sigma}_{\mathrm{MW}} \mathbf{n}, \qquad \boldsymbol{\sigma}_{\mathrm{MW}} := \mathbf{e} \otimes \mathbf{d} - \frac{\epsilon_0}{2} |\mathbf{e}|^2 \mathbf{I}. \tag{2.1}$$

A classic derivation of the above formula begins from the assertion that the force on a point charge q and a point dipole $\mathbf{p}$ is given by

$$q\mathbf{e} \qquad \text{and} \qquad (\mathrm{grad} \mathbf{e}) \mathbf{p}, \tag{2.2}$$

respectively, see, e.g., Jackson (1999, (1.1) and (5.69)). Therefore, the resultant force on the charges and dipoles distributed in a subdomain $U \subset V$ is given by

$$\mathbf{f}_U = \int_U \mathbf{f}_{\mathrm{elect}}, \qquad \mathbf{f}_{\mathrm{elect}} := \rho \mathbf{e} + (\mathrm{grad} \mathbf{e}) \mathbf{p}, \tag{2.3}$$

where the vector field $\mathbf{f}_{\text{elect}} : V \to \mathbb{R}^3$ can be interpreted as the body force due to electrostatic interactions.

In continuum mechanics, it is desirable to express balance laws as pointwise differential equations. We are therefore motivated to seek a local differential form of the force formula (2.3), i.e., a tensor field $\boldsymbol{\sigma}_{\text{elect}} : V \to \mathbb{R}^{3\times 3}$, such that

$$\text{div}\boldsymbol{\sigma}_{\text{elect}} = \mathbf{f}_{\text{elect}} \qquad \text{in } V. \tag{2.4}$$

Then by the divergence theory, the force on the subdomain U can be rewritten as

$$\mathbf{f}_U = \int_U \text{div}\, \boldsymbol{\sigma}_{\text{elect}} = \int_{\partial U} \boldsymbol{\sigma}_{\text{elect}}\mathbf{n}, \tag{2.5}$$

where the last equality requires the tensor field $\boldsymbol{\sigma}_{\text{elect}}$ be continuous up to ∂U. A special solution to (2.4) is given by $\boldsymbol{\sigma}_{\text{elect}} = \boldsymbol{\sigma}_{\text{MW}}$ if $(\rho, \mathbf{p})$ and hence ξ are *smooth on* V. To see this, by direct calculations we find that for any $\mathbf{x} \in V$,

$$\begin{cases} \text{div}[(-\text{grad}\xi) \otimes (-\epsilon_0 \text{grad}\xi + \mathbf{p})] = (\text{gradgrad}\xi)(\epsilon_0 \text{grad}\xi - \mathbf{p}) - \rho \text{grad}\xi, \\ \text{grad}(\frac{\epsilon_0}{2}|\text{grad}\xi|^2) = \epsilon_0 (\text{gradgrad}\xi)(\text{grad}\xi). \end{cases} \tag{2.6}$$

Therefore,

$$\text{div}\, \boldsymbol{\sigma}_{\text{MW}} = -\rho \text{grad}\, \xi - (\text{grad}\, \text{grad}\, \xi)\mathbf{p} = \mathbf{f}_{\text{elect}} \qquad \text{in } V.$$

A critical advantage of rewriting the electrostatic force in terms of the Maxwell stress lies in that the force formula (2.5) has less stringent requirement on the differentiability of the electric field. Since non-smooth distributions of charge density and polarization are common in continuum bodies, e.g., a conductor and a heterogeneous dielectric medium with sharp interfaces, the force formula (2.5) enables us to calculate the electrostatic force on singular points, lines and interfaces without ambiguity. On the other hand, replacing the concept of electric body force by Maxwell stress is often criticized for lack of uniqueness [2] and muddy issues in boundary/interfacial conditions (Rinaldi and Brenner, 2002; Bustamante *et al.*, 2009b). Further, for a body with spatially varying permittivity $\epsilon(\mathbf{x})$, a different quantity [3]

$$\boldsymbol{\sigma}'_{\text{MW}} = \mathbf{e} \otimes \mathbf{d} - \frac{\epsilon(\mathbf{x})}{2}|\mathbf{e}|^2\mathbf{I} \tag{2.7}$$

is also referred to as the Maxwell stress. The precise relation between these two expressions, as well as the derivation, necessity, or even definition of Maxwell stress have been discussed in-depth in the literature, including the textbooks of Stratton (1941), Melcher (1981), Jackson (1999) and Kovetz (2000), treatise of Toupin (1956, 1959), Pao (1978), and recent discussions of Rinaldi and Brenner (2002), Steigmann (2009), Bustamante *et al.* (2009). Our position agrees with Toupin (1956, 1959) on the following:

[2] First, we notice that solutions are not unique, even if a boundary condition is supplemented. If $\boldsymbol{\sigma}_{\text{elect}}$ is a solution to (2.4), then $\boldsymbol{\sigma}_{\text{elect}} + \boldsymbol{\sigma}_0$ is also a solution for any tensor field $\boldsymbol{\sigma}_0 : V \to \mathbb{R}^{3\times 3}$ satisfying

$$\begin{cases} \text{div}\boldsymbol{\sigma}_0 = 0 & \text{on } V, \\ \boldsymbol{\sigma}_0\mathbf{n} = 0 & \text{on } \partial V. \end{cases}$$

[3] The associated body force is often written as (Stratton 1941, ch. II; Meltcher ch. 3)

$$\mathbf{f}'_{\text{elect}} = \rho\mathbf{e} - \nabla\epsilon|\mathbf{e}|^2 \qquad \text{in } V.$$

This formula, however, is consistent with (2.7), i.e., $\mathbf{f}'_{\text{elect}} = \text{div}\boldsymbol{\sigma}'_{\text{MW}}$, only if $\epsilon(\mathbf{x})$ is continuous and does not have sharp interfaces.

1. The independent variables for describing the local state of an electro-elastic body are the deformation relative to some natural state and the polarization (Toupin, 1956, § 2).

2. Different expressions of the Maxwell stress arise from different decompositions of the total energy into internal energy and electric field energy. As remarked by Toupin (1959): "*Any division of energy, momentum, stress and energy flux into electromagnetic and mechanical components is bound to be somewhat arbitrary, and it is fruitless to attempt an independent theory of either component.*"

3. A particular decomposition of the total stress into mechanical stress and electromagnetic stress is for "*the intuitive guides in the construction of admissible constitutive equations (for the body) but are otherwise irrelevant*" (Toupin, 1956). Toupin has chosen the expression (2.1) as the electric stress since "*it has the same form in all materials*" and is "*not a state function*" (of deformation gradient and polarization).

Toupin (1956) began his formulation from stress hypothesis, and hence the total stress is a primitive notion and may be decomposed into

$$\boldsymbol{\sigma}_{\rm tot} = \boldsymbol{\sigma} + \boldsymbol{\sigma}_{\rm MW}.$$

As mentioned above, it was then argued that the Maxwell stress $\boldsymbol{\sigma}_{\rm MW}$ is not a state function whereas the "mechanical stress" $\boldsymbol{\sigma}$ is a state function:

$$\boldsymbol{\sigma} = \boldsymbol{\sigma}(\mathbf{F}, \mathbf{p}),$$

where $(\mathbf{F}, \mathbf{p})$ are the deformation gradient and polarization describing the local state of the body. The above constitutive postulation, together with the balance of linear and angular momenta and the Maxwell equations, form a boundary value problem that may be solved to determine all physical quantities of interest and completes the theory.

Toupin (1956, 1959) also proposed a variational principle for his theory of electro-elastic materials based on the principle of virtual work. To include the Maxwell equation as a consequence of the principle of virtual work, the electrostatic potential, in addition to the deformation gradient and polarization, is regarded as one of the *independent variables*, which, from the standpoint of thermodynamics, somewhat contradicts his prior statement that the local state of the body is described only by the deformation gradient and polarization (Toupin, 1956, § 2). A more severe conceptual difficulty lies in that the proposed energy functional has no clear thermodynamic interpretation and the variational problem turns out to be a min-max problem instead of the usual minimization problem of the total free energy. These issues with Toupin's variational formulation have been recognized by many authors (Brown, 1966) and addressed by Ericksen (2007) using vector potential associated with electric displacement. A number of alternative variational formulations have been proposed in the literature. Undoubtedly, the field equations of all formulations are consistent with each other, consisting of the Maxwell equations and mechanical balance laws, and are equivalent to the extent of determining the local fields. However, for stability analysis and evolution problems among others it is necessary to have a variational formulation in terms of the physical free energy such that *upon minimizing the total free energy over the admissible states, all of the field equations and boundary conditions follow as necessary conditions for a minimizer of the free energy (i.e., Euler-Lagrange equations)*. It is in this spirit that we propose the following energy formulation.

3 Energy formulation and associated boundary value problems

3.1 Free energy of the system

For clarity we begin with electro-elastics; the theory including magnetic couplings will be developed in § 5. Consider a deformable body Ω_R in an ambient medium of permittivity ϵ_0. Assume that the ambient medium is elastically *trivial*, meaning that it has zero elastic stiffness. The body, together with the ambient medium of permittivity ϵ_0, occupies domain V_R. The thermodynamic state of the body is described by deformation $\boldsymbol{\chi} : \Omega_R \to \Omega$ and polarization $\tilde{\mathbf{P}} : \Omega_R \to \mathbb{R}^3$:

$$\mathbf{x} = \boldsymbol{\chi}(\mathbf{X}), \qquad \tilde{\mathbf{P}} = \tilde{\mathbf{P}}(\mathbf{X}).$$

The current configuration $\Omega = \boldsymbol{\chi}(\Omega_R)$ depends on the deformation $\boldsymbol{\chi}$, and for ease of notation, polarization $\tilde{\mathbf{P}}$ is extended by zero to domain V_R and deformation $\boldsymbol{\chi}$ is extended continuously to domain V_R. The precise extension $\boldsymbol{\chi}$ to $V_R \setminus \Omega_R$ has no physical consequence, we may choose such that $\boldsymbol{\chi} \to \mathbf{X}$ quickly away from $\partial\Omega_R$. Below we refer to the body Ω_R as *the system* described by state variables $(\boldsymbol{\chi}, \tilde{\mathbf{P}})$.

By the Maxwell equation $\mathrm{curl}\mathbf{e} = 0$, there exists a scalar potential ξ such that the electric field $\mathbf{e} = -\mathrm{grad}\xi$. Further, we assume that there exist external distributions of charge $\tilde{\rho}^e : \Omega_R \to \mathbb{R}$ and dipoles $\tilde{\mathbf{P}}^e : \Omega_R \to \mathbb{R}^3$ "attached to" *material* points, and hence under deformation $\boldsymbol{\chi} : \Omega_R \to \Omega$ the external charge and dipole distributions in the *current configuration* are given by

$$\rho^e = \frac{1}{J}\tilde{\rho}^e \circ \boldsymbol{\pi}, \qquad \mathbf{p}^e = \frac{1}{J}\tilde{\mathbf{P}}^e \circ \boldsymbol{\pi}, \tag{3.1}$$

respectively, where $\boldsymbol{\pi} : \Omega \to \Omega_R$ is the inverse mapping of $\boldsymbol{\chi}$ (cf., Fig. 1). Therefore, by the Maxwell equation $\mathrm{div}\mathbf{d} = \rho^e$ and (1.3) we have

$$\mathrm{Div}\tilde{\mathbf{D}} = \tilde{\rho}^e \qquad \text{in } V_R, \qquad \tilde{\mathbf{D}} = -\epsilon_0 J\mathbf{C}^{-1}\mathrm{Grad}\xi + \mathbf{F}^{-1}(\tilde{\mathbf{P}} + \tilde{\mathbf{P}}^e). \tag{3.2}$$

Further, we impose some electrostatic boundary conditions on ∂V_R and some mechanical boundary conditions on $\partial\Omega_R$. As illustrated in Fig. 2, let Γ_D and Γ_R be a subdivision of ∂V_R and S_D and S_N be a subdivision of $\partial\Omega_R$. The *electrical* Dirichlet boundary on Γ_D and Robin boundary condition on Γ_R are prescribed as

$$\xi = \xi_b \qquad \text{on } \Gamma_D, \qquad\qquad k(\xi - \xi_b) - \mathbf{n}\cdot\tilde{\mathbf{D}} - \sigma_0 = 0 \qquad \text{on } \Gamma_R, \tag{3.3}$$

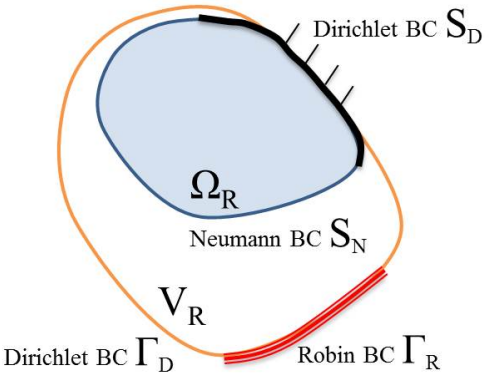
where $\xi_b : \partial V_R \to \mathbb{R}$, $k : \Gamma_R \to [0, +\infty)$ and $\sigma_0 : \Gamma_R \to \mathbb{R}$ are given boundary data. In addition, we apply a mechanical body force $\tilde{\mathbf{f}}^e : \Omega_R \to \mathbb{R}^3$, surface traction $\tilde{\mathbf{t}}^e$ on S_N, and prescribe the deformation $\boldsymbol{\chi}$ on S_D to be $\boldsymbol{\chi}_b$:

$$\begin{cases} \boldsymbol{\chi} = \boldsymbol{\chi}_b & \text{on } S_D, \\ \text{applied traction (dead load)} = \tilde{\mathbf{t}}^e & \text{on } S_N. \end{cases} \tag{3.4}$$

The boundary ∂V_R may overlap with part or all of $\partial\Omega_R$ which, however, does not require a separate consideration.

For simplicity, we assume that the external charges and dipoles $(\tilde{\rho}^e, \tilde{\mathbf{P}}^e)$, applied body force $\tilde{\mathbf{f}}^e$ and boundary data $\xi_b, k, \sigma_0, \tilde{\mathbf{t}}^e$ are defined in the *reference configuration* and will be *independent* of the deformation and polarization $(\boldsymbol{\chi}, \tilde{\mathbf{P}})$. Mechanical loads $\tilde{\mathbf{f}}^e$ and $\tilde{\mathbf{t}}^e$ of this kind are referred to as "dead" loads.

To formulate our theory of electro-elastic materials, we begin with the hypothesis that the internal/stored energy of the body is additive, admits an internal energy density function Ψ :



Mechanical BC on $\partial\,\Omega_R$

$\chi = \chi_b$ on S_D

applied dead load $= \tilde{\mathbf{t}}^e$ on S_N

Electrostatic BC on $\partial\,V_R$

$\xi = \xi_b$ on Γ_D

$k(\xi - \xi_b) - \mathbf{n}\cdot\tilde{\mathbf{D}} - \sigma_0 = 0$ on Γ_R

Figure 2: Mechanical and electrical boundary conditions are imposed on the reference configuration: the deformation $\boldsymbol{\chi} = \boldsymbol{\chi}_0$ on S_D and a traction of $\tilde{\mathbf{t}}^e$ is applied on S_N; the electric potential on Γ_D is prescribed as $\xi = \xi_b$ and a Robin boundary condition is applied on Γ_R. Here, S_D and S_N is a subdivision of $\partial\Omega_R$ while Γ_D and Γ_R is a subdivision of ∂V_R.

$\mathbb{R}^3 \times \mathbb{R}^{3\times3} \times \mathbb{R}^{3\times3\times3} \times \mathbb{R}^3 \times \mathbb{R}^{3\times3} \to \mathbb{R}$ at the reference configuration, and hence the internal energy of the body Ω_R with state variables $(\boldsymbol{\chi}, \tilde{\mathbf{P}})$ is given by

(**H1**)
$$U[\boldsymbol{\chi}, \tilde{\mathbf{P}}] = \int_{\Omega_R} \Psi(\mathbf{X}; \mathbf{F},\, \mathbf{G},\, \tilde{\mathbf{P}},\, \boldsymbol{\Pi}), \tag{3.5}$$

where the explicit $\mathbf{X}$-dependence of Ψ reflects that the body may be heterogeneous, and

$$\mathbf{F} = \mathrm{Grad}\boldsymbol{\chi}, \quad \mathbf{G} = \mathrm{GradGrad}\boldsymbol{\chi}, \quad \boldsymbol{\Pi} = \mathrm{Grad}\tilde{\mathbf{P}}.$$

For brevity, we sometimes omit the explicit $\mathbf{X}$-dependence of Ψ in notation. Let $So(3) \subset \mathbb{R}^{3\times3}$ be the group consisting of all rigid rotations and $\mathcal{G} \subset \mathbb{R}^{3\times3}$ be the symmetry group of the underlying crystals/materials. Then the principle of frame indifference implies

$$\Psi(\mathbf{F}, \mathbf{G}, \tilde{\mathbf{P}}, \boldsymbol{\Pi}) = \Psi(\mathbf{RF}, \mathbf{RG}, \mathbf{R}\tilde{\mathbf{P}}, \mathbf{R}\boldsymbol{\Pi}) \qquad \forall\, \mathbf{R} \in So(3); \tag{3.6}$$

material symmetries imply

$$\Psi(\mathbf{F}, \mathbf{G}, \tilde{\mathbf{P}}, \boldsymbol{\Pi}) = \Psi(\mathbf{FQ}, \mathbf{G}_{\mathbf{Q}}, \tilde{\mathbf{P}}, \boldsymbol{\Pi}\mathbf{Q}) \qquad \forall\, \mathbf{Q} \in \mathcal{G}, \tag{3.7}$$

where the components of the third-order tensor $\mathbf{G}_{\mathbf{Q}}$ are given by $(\mathbf{G}_{\mathbf{Q}})_{pij} = (\mathbf{G})_{pkl}(\mathbf{Q})_{ki}(\mathbf{Q})_{lj}$.

The polarization on the body inevitably induces an electric field penetrating into the ambient medium in $V \setminus \Omega$ and interacting with boundary devices. To include the nonlocal field energy, we need to solve for the electric field which in the *current configuration* is determined by the Maxwell equation (1.1a). By (1.1a), (1.2), (1.4) and (3.3), in the *reference configuration* the electrostatic boundary value problem can be written as

$$\mathrm{Div}\tilde{\mathbf{D}} = \tilde{\rho}^e, \quad \tilde{\mathbf{D}} = -\epsilon_0 J\mathbf{C}^{-1}\mathrm{Grad}\xi + \mathbf{F}^{-1}(\tilde{\mathbf{P}} + \tilde{\mathbf{P}}^e) \quad \text{in } V_R, \tag{3.8}$$

with boundary conditions (3.3).

Following the discussions in Liu (2013), we identify the total free energy of the system as

(**H1′**)
$$F[\boldsymbol{\chi}, \tilde{\mathbf{P}}] = U[\boldsymbol{\chi}, \tilde{\mathbf{P}}] + \mathcal{E}^{\mathrm{elect}}[\boldsymbol{\chi}, \tilde{\mathbf{P}}] + P^{\mathrm{mech}}[\boldsymbol{\chi}], \tag{3.9}$$

where $U[\boldsymbol{\chi}, \tilde{\mathbf{P}}]$, given by (3.5), is the internal energy,

$$\begin{aligned}\mathcal{E}^{\text{elect}}[\boldsymbol{\chi}, \tilde{\mathbf{P}}] &= \frac{\epsilon_0}{2}\int_V |\mathbf{e}|^2 + \int_{\Gamma_D} \xi_b \mathbf{N}\cdot\tilde{\mathbf{D}} + \int_{\Gamma_R}\frac{k}{2}(\xi)^2 \\ &= \frac{\epsilon_0}{2}\int_{V_R} J|\mathbf{F}^{-T}\text{Grad}\xi|^2 + \int_{\Gamma_D} \xi_b \mathbf{N}\cdot\tilde{\mathbf{D}} + \int_{\Gamma_R}\frac{k}{2}(\xi)^2 \end{aligned} \tag{3.10}$$

is the total electric energy associated with the electric field (the first term) and the boundary electrical devices (the second and third terms), and

$$P^{\text{mech}}[\boldsymbol{\chi}] = -\int_{S_N} \tilde{\mathbf{t}}^e\cdot\boldsymbol{\chi} - \int_{\Omega_R}\tilde{\mathbf{f}}^e\cdot\boldsymbol{\chi} \tag{3.11}$$

is the familiar potential energy of mechanical loadings.

3.2 Euler-Lagrange equations and boundary conditions

Our interest is to identify the equilibrium state of the body and the effects of external mechanical and electrical loads. To this end, we employ the *principle of minimum free energy*, i.e., the equilibrium state $(\boldsymbol{\chi}, \tilde{\mathbf{P}})$, by definition, is such that the free energy is minimized among "all possible variations" of the state variables:

$$\min_{(\boldsymbol{\chi},\tilde{\mathbf{P}})\in\mathcal{S}} F[\boldsymbol{\chi}, \tilde{\mathbf{P}}]. \tag{3.12}$$

As remarked by Ericksen (1991, p7), we have to "exercise some judgement in deciding what these (all possible variations) might be". Also, it is unlikely that "possible variations" are unique and universal for all materials. Our judgement concerning the admissible space for state variables and their possible *independent* variations constitutes the second hypothesis of our theory.
(**H2**) Denote the admissible space of state variables by

$$(\boldsymbol{\chi}, \tilde{\mathbf{P}}) \in \mathcal{S}, \tag{3.13}$$

which will be specified later for typical materials. For a given state $(\boldsymbol{\chi}, \tilde{\mathbf{P}}) \in \mathcal{S}$, possible independent infinitesimal variations include ($\delta \in \mathbb{R}$ and $|\delta| << 1$):

1. Variation of polarization:

$$\boldsymbol{\chi} \to \boldsymbol{\chi}_\delta = \boldsymbol{\chi}, \qquad \tilde{\mathbf{P}} \to \tilde{\mathbf{P}}_\delta = \tilde{\mathbf{P}} + \delta\tilde{\mathbf{P}}_1; \tag{3.14}$$

2. Variation of deformation:

$$\boldsymbol{\chi} \to \boldsymbol{\chi}_\delta = \boldsymbol{\chi} + \delta\boldsymbol{\chi}_1, \qquad \tilde{\mathbf{P}} \to \tilde{\mathbf{P}}_\delta = \tilde{\mathbf{P}}. \tag{3.15}$$

Variations of polarization

We now derive the Euler-Lagrange equations and boundary conditions associated with the variational principle (3.12). For variations (3.14), it is clear that

$$\frac{d}{d\delta}U[\boldsymbol{\chi}_\delta, \tilde{\mathbf{P}}_\delta]\Big|_{\delta=0} = \int_{\Omega_R}[D_{\tilde{\mathbf{P}}}\Psi\cdot\tilde{\mathbf{P}}_1 + D_{\boldsymbol{\Pi}}\Psi\cdot\text{Grad}\tilde{\mathbf{P}}_1]. \tag{3.16}$$

To find the change of electric field to the leading order, we assume that

$$\xi_\delta = \xi + \delta\xi_1 + o(\delta), \qquad \tilde{\mathbf{D}}_\delta = \tilde{\mathbf{D}} + \delta\tilde{\mathbf{D}}_1 + o(\delta). \tag{3.17}$$

Then by (3.10) we have

$$\mathcal{E}^{\text{elect}}[\boldsymbol{\chi}, \mathbf{P}_\delta] = \mathcal{E}^{\text{elect}}[\boldsymbol{\chi}, \mathbf{P}] + \delta T_1 + o(\delta)$$

where

$$T_1 = \int_{V_R} \epsilon_0 J(\mathbf{F}^{-T}\text{Grad}\xi)\cdot(\mathbf{F}^{-T}\text{Grad}\xi_1) + \int_{\Gamma_D} \xi_b \mathbf{N}\cdot\tilde{\mathbf{D}}_1 + \int_{\Gamma_R} k\xi\xi_1. \tag{3.18}$$

By (3.8), (3.3) and (3.17), ξ_1 and $\tilde{\mathbf{D}}_1$ satisfy

$$\begin{cases} \text{Div}\tilde{\mathbf{D}}_1 = 0, \quad \tilde{\mathbf{D}}_1 = -\epsilon_0 J\mathbf{C}^{-1}\text{Grad}\xi_1 + \mathbf{F}^{-1}\tilde{\mathbf{P}}_1 & \text{in } V_R, \\ \xi_1 = 0 & \text{on } \Gamma_D, \\ k\xi_1 - \mathbf{N}\cdot\tilde{\mathbf{D}}_1 = 0 & \text{on } \Gamma_R. \end{cases} \tag{3.19}$$

By the divergence theorem we have

$$\int_{\partial V_R} (\xi\tilde{\mathbf{D}}_1)\cdot\mathbf{N} = \int_{V_R} \text{Div}(\xi\tilde{\mathbf{D}}_1) = \int_{V_R} \text{Grad}\xi\cdot\tilde{\mathbf{D}}_1 + \xi\text{Div}\tilde{\mathbf{D}}_1 = \int_{V_R} \text{Grad}\xi\cdot\tilde{\mathbf{D}}_1,$$

where the last equality follows from $(3.19)_1$. Inserting the above equation into (3.18) we obtain

$$\begin{aligned} T_1 &= \int_{V_R} \text{Grad}\xi\cdot\mathbf{F}^{-1}\tilde{\mathbf{P}}_1 - \int_{\partial V_R} (\xi\tilde{\mathbf{D}}_1)\cdot\mathbf{N} + \int_{\Gamma_D} \xi_b\mathbf{N}\cdot\tilde{\mathbf{D}}_1 + \int_{\Gamma_R} k\xi\xi_1 \\ &= \int_{\Omega_R} \tilde{\mathbf{P}}_1\cdot\mathbf{F}^{-T}\text{Grad}\xi, \end{aligned} \tag{3.20}$$

where the last equality follows from the boundary conditions (3.3) and $(3.19)_3$. Therefore, the first variation of the free energy associated with (3.14) is given by

$$\frac{d}{d\delta}F[\boldsymbol{\chi}, \tilde{\mathbf{P}}_\delta]\Big|_{\delta=0} = \int_{\Omega_R} \left[(\mathbf{F}^{-T}\text{Grad}\xi + D_{\tilde{\mathbf{P}}}\Psi - \text{Div}D_{\boldsymbol{\Pi}}\Psi)\cdot\tilde{\mathbf{P}}_1\right] + \int_{\partial\Omega_R} \tilde{\mathbf{P}}_1\cdot(D_{\boldsymbol{\Pi}}\Psi)\mathbf{N}. \tag{3.21}$$

Since the above quantity vanishes for arbitrary $\tilde{\mathbf{P}}_1$, an equilibrium state $(\boldsymbol{\chi}, \tilde{\mathbf{P}})$ necessarily satisfies

$$\begin{cases} \mathbf{F}^{-T}\text{Grad}\xi + D_{\tilde{\mathbf{P}}}\Psi - \text{Div}D_{\boldsymbol{\Pi}}\Psi = 0 & \text{in } \Omega_R, \\ (\text{Div}D_{\boldsymbol{\Pi}}\Psi)\mathbf{N} = 0 & \text{on } \partial\Omega_R. \end{cases} \tag{3.22}$$

Variations of deformation

For variation (3.15), we have

$$\mathbf{F}(\mathbf{X}) \to \mathbf{F}_\delta(\mathbf{X}) = \mathbf{F} + \delta\mathbf{F}_1, \qquad \mathbf{G}(\mathbf{X}) \to \mathbf{G}_\delta(\mathbf{X}) = \mathbf{G}(\mathbf{X}) + \delta\text{GradGrad}\boldsymbol{\chi}_1,$$

where $\mathbf{F}_1 = \text{Grad}\boldsymbol{\chi}_1$. Algebraic calculations show that

$$\begin{aligned} &\mathbf{F}_\delta^{-1} = \mathbf{F}^{-1} - \delta\mathbf{F}^{-1}\mathbf{F}_1\mathbf{F}^{-1} + o(\delta), \qquad J_\delta = J[1 + \delta\text{Tr}(\mathbf{F}^{-1}\mathbf{F}_1)] + o(\delta), \\ &\mathbf{C}_\delta^{-1} = \mathbf{C}^{-1} - \delta(\mathbf{F}^{-1}\mathbf{F}_1\mathbf{C}^{-1} + \mathbf{C}^{-1}\mathbf{F}_1^T\mathbf{F}^{-T}) + o(\delta). \end{aligned} \tag{3.23}$$

Further, by (3.8) and (3.3) we see that the electric potential ξ_δ satisfies

$$\begin{cases} \mathrm{Div}[-\epsilon_0 J_\delta \mathbf{C}_\delta^{-1}\mathrm{Grad}\xi_\delta + \mathbf{F}_\delta^{-1}(\tilde{\mathbf{P}} + \tilde{\mathbf{P}}^e)] = \tilde{\rho}^e & \text{in } V_R, \\ \xi_\delta - \xi_b = 0 & \text{on } \Gamma_D, \\ k(\xi - \xi_b) - \tilde{\mathbf{D}}_\delta \cdot \mathbf{N} = 0 & \text{on } \Gamma_R. \end{cases} \tag{3.24}$$

Clearly, the actual electric field and hence the electric energy depend on the deformation. To find the change of electric field to the leading order, we once again assume (3.17) and, by (3.8), find that

$$\begin{aligned} \tilde{\mathbf{D}}_1 = &-\epsilon_0 J\mathbf{C}^{-1}\mathrm{Grad}\xi_1 - \epsilon_0 J\mathrm{Tr}(\mathbf{F}^{-1}\mathbf{F}_1)\mathbf{C}^{-1}\mathrm{Grad}\xi \\ &+\epsilon_0 J(\mathbf{F}^{-1}\mathbf{F}_1\mathbf{C}^{-1} + \mathbf{C}^{-1}\mathbf{F}_1^T\mathbf{F}^{-T})\mathrm{Grad}\xi - \mathbf{F}^{-1}\mathbf{F}_1\mathbf{F}^{-1}\tilde{\mathbf{P}}. \end{aligned} \tag{3.25}$$

Inserting the above equation into (3.24) and keeping the terms of order δ, we obtain

$$\begin{cases} \mathrm{Div}\tilde{\mathbf{D}}_1 = 0 & \text{in } V_R, \\ \xi_1 = 0 & \text{on } \Gamma_D, \\ k\xi_1 - \tilde{\mathbf{D}}_1 \cdot \mathbf{N} = 0 & \text{on } \Gamma_R. \end{cases}$$

Multiplying the first of the above equations by ξ, by the divergence theorem we find that

$$\int_{\partial V_R} \xi\tilde{\mathbf{D}}_1 \cdot \mathbf{N} - \int_{V_R} \mathrm{Grad}\xi \cdot \tilde{\mathbf{D}}_1 = 0. \tag{3.26}$$

By (3.25) we rewrite the above equation as

$$\begin{aligned} \int_{V_R} \epsilon_0 J\mathrm{Grad}\xi \cdot \mathbf{C}^{-1}\mathrm{Grad}\xi_1 = &-\int_{\partial V_R} \xi\tilde{\mathbf{D}}_1 \cdot \mathbf{N} + \int_{V_R} \mathrm{Grad}\xi \cdot \Big[-\epsilon_0 J\mathrm{Tr}(\mathbf{F}^{-1}\mathbf{F}_1)\mathbf{C}^{-1}\mathrm{Grad}\xi \\ &+\epsilon_0 J(\mathbf{F}^{-1}\mathbf{F}_1\mathbf{C}^{-1} + \mathbf{C}^{-1}\mathbf{F}_1^T\mathbf{F}^{-T})\mathrm{Grad}\xi - \mathbf{F}^{-1}\mathbf{F}_1\mathbf{F}^{-1}\tilde{\mathbf{P}}\Big]. \end{aligned} \tag{3.27}$$

Further, by (3.23) we find that

$$\mathcal{E}^{\mathrm{elect}}[\boldsymbol{\chi}_\delta, \tilde{\mathbf{P}}] = \mathcal{E}^{\mathrm{elect}}[\boldsymbol{\chi}, \tilde{\mathbf{P}}] + \delta\mathrm{Var1} + o(\delta), \tag{3.28}$$

where

$$\begin{aligned} \mathrm{Var1} = \frac{\epsilon_0}{2}\int_{V_R} \Big[&J\mathrm{Tr}(\mathbf{F}^{-1}\mathbf{F}_1)|\mathbf{F}^{-T}\mathrm{Grad}\xi|^2 - J\mathrm{Grad}\xi \cdot (\mathbf{F}^{-1}\mathbf{F}_1\mathbf{C}^{-1} + \mathbf{C}^{-1}\mathbf{F}_1^T\mathbf{F}^{-T})\mathrm{Grad}\xi \\ &+2J\mathrm{Grad}\xi \cdot \mathbf{C}^{-1}\mathrm{Grad}\xi_1\Big] + \int_{\Gamma_R} k\xi\xi_1 + \int_{\Gamma_D} \xi_b\tilde{\mathbf{D}}_1 \cdot \mathbf{N}. \end{aligned}$$

Inserting (3.27) into the above equation we obtain

$$\begin{aligned} \mathrm{Var1} &= \int_{V_R} \Big[-\frac{\epsilon_0}{2} J\mathrm{Tr}(\mathbf{F}^{-1}\mathbf{F}_1)|\mathbf{E}|^2 + \frac{\epsilon_0}{2} J\mathbf{E} \cdot (\mathbf{F}_1\mathbf{F}^{-1} + \mathbf{F}^{-T}\mathbf{F}_1^T)\mathbf{E} + \mathbf{E} \cdot \mathbf{F}_1\mathbf{F}^{-1}\tilde{\mathbf{P}}\Big] \\ &= \int_{V_R} \mathbf{F}_1 \cdot \Big[-\frac{\epsilon_0}{2} J|\mathbf{E}|^2\mathbf{F}^{-T} + \mathbf{E} \otimes \tilde{\mathbf{D}}\Big] = \int_{V_R} \mathbf{F}_1 \cdot \tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}}, \end{aligned} \tag{3.29}$$

where

$$\tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}} = -\frac{\epsilon_0}{2} J|\mathbf{E}|^2\mathbf{F}^{-T} + \mathbf{E} \otimes \tilde{\mathbf{D}}. \tag{3.30}$$

By (1.4), we recoganize

$$\boldsymbol{\sigma}_{\rm MW} = \frac{1}{J}\tilde{\boldsymbol{\Sigma}}_{\rm MW}\mathbf{F}^T = \mathbf{e}\otimes\mathbf{d} - \frac{\epsilon_0}{2}|\mathbf{e}|^2\mathbf{I}$$

as the familiar expression of Maxwell stress in the current configuration (cf., (2.1)). Subsequently, we refer to $\tilde{\boldsymbol{\Sigma}}_{\rm MW}$ as the *Piola-Maxwell* stress. We remark that the above derivation of the Maxwell stress has been presented in Tian (2007).

Moreover, the first variation of the internal energy is given by

$$U[\boldsymbol{\chi}_\delta, \tilde{\mathbf{P}}] = U[\boldsymbol{\chi}, \tilde{\mathbf{P}}] + \int_{\Omega_R} D_{\mathbf{F}}\Psi\cdot{\rm Grad}\boldsymbol{\chi}_1 + \int_{\Omega_R} D_{\mathbf{G}}\Psi\cdot{\rm GradGrad}\boldsymbol{\chi}_1 + o(\delta). \tag{3.31}$$

Integrating by parts, we rewrite the above integrals on the right hand side as

$$\int_{\Omega_R} D_{\mathbf{F}}\Psi\cdot{\rm Grad}\boldsymbol{\chi}_1 = \int_{\partial\Omega_R}\boldsymbol{\chi}_1\cdot(D_{\mathbf{F}}\Psi)\mathbf{N} - \int_{\Omega_R}\boldsymbol{\chi}_1\cdot{\rm Div}(D_{\mathbf{F}}\Psi), \tag{3.32}$$

and (see Appendix A for details)

$$\begin{aligned}\int_{\Omega_R} D_{\mathbf{G}}\Psi\cdot{\rm GradGrad}\boldsymbol{\chi}_1 = \int_{\Omega_R}\boldsymbol{\chi}_1\cdot{\rm DivDiv}(D_{\mathbf{G}}\Psi) + \int_{\partial\Omega_R}[(D_{\mathbf{G}}\Psi)\mathbf{N}\otimes\mathbf{N}]\cdot({\rm Grad}\boldsymbol{\chi}_1)\mathbf{N}\\ - \int_{\partial\Omega_R}\boldsymbol{\chi}_1\cdot\big\{\boldsymbol{\tau} + ({\rm Div}D_{\mathbf{G}}\Psi)\mathbf{N}\big\}.\end{aligned} \tag{3.33}$$

where the components of the vector field $\boldsymbol{\tau}:\partial\Omega_R\to\mathbb{R}^3$ are given by

$$(\boldsymbol{\tau})_p = [\tilde{\Lambda}_{pij}N_j(\delta_{ik}-N_iN_k)]_{,k} - [\tilde{\Lambda}_{pij}N_j(\delta_{ik}-N_iN_k)]_{,m}N_mN_k, \tag{3.34}$$

and $\tilde{\Lambda}_{pij}$ are the components of $(D_{\mathbf{G}}\Psi)_{pij}$. Further, the components of the symbols on the right hand side of (3.33) are given by

$$[(D_{\mathbf{G}}\Psi)\mathbf{N}\otimes\mathbf{N}]_p = \tilde{\Lambda}_{pij}N_jN_i,\quad [{\rm DivDiv}(D_{\mathbf{G}}\Psi)]_p = \tilde{\Lambda}_{pij,ij},\quad [({\rm Div}D_{\mathbf{G}}\Psi)\mathbf{N}]_p = \tilde{\Lambda}_{pij,j}N_i.$$

In other words, in index form equation (3.33) can be written as

$$\int_{\Omega_R}\tilde{\Lambda}_{pij}u_{p,ij} = \int_{\partial\Omega_R}\tilde{\Lambda}_{pij}N_jN_iu_{p,k}N_k - \int_{\partial\Omega_R}(\tau_p+\tilde{\Lambda}_{pij,j}N_i)u_p + \int_{\Omega_R}\tilde{\Lambda}_{pij,ij}u_p, \tag{3.35}$$

where u_p are the components of $\boldsymbol{\chi}_1$.

Henceforth, by (3.28), (3.29), (3.31), (3.32) and (3.33), associated with the variation (3.15) the first variation of the free energy (3.9) of the system is given by

$$\begin{aligned}\frac{d}{d\delta}F[\boldsymbol{\chi}_\delta,\tilde{\mathbf{P}}]\Big|_{\delta=0} &= \int_V\boldsymbol{\chi}_1\cdot\Big\{\chi_{\Omega_R}\Big[{\rm DivDiv}(D_{\mathbf{G}}\Psi) - {\rm Div}D_{\mathbf{F}}\Psi - \tilde{\mathbf{f}}^e\Big] - {\rm Div}\tilde{\boldsymbol{\Sigma}}_{\rm MW}\Big\}\\ &\quad + \int_{S_N}\boldsymbol{\chi}_1\cdot\Big[(D_{\mathbf{F}}\Psi - [\![\tilde{\boldsymbol{\Sigma}}_{\rm MW}]\!])\mathbf{N} - \boldsymbol{\tau} - ({\rm Div}D_{\mathbf{G}}\Psi)\mathbf{N} - \tilde{\mathbf{t}}^e\Big]\\ &\quad + \int_{\partial\Omega_R}[(D_{\mathbf{G}}\Psi)\mathbf{N}\otimes\mathbf{N}]\cdot({\rm Grad}\boldsymbol{\chi}_1)\mathbf{N},\end{aligned} \tag{3.36}$$

where $[\![\ ]\!] = (\ )_+ - (\ )_-$ denotes the jump from the exterior (+ side) to the interior of Ω_R (− side). If S_N overlaps with ∂V_R, the exterior value of $\tilde{\boldsymbol{\Sigma}}_{\rm MW}$ depends on the details of the electrical loading

devices. Since the above quantity vanishes for arbitrary $\boldsymbol{\chi}_1$ satisfying $\boldsymbol{\chi}_1 = 0$ on Γ_D, an equilibrium state $(\boldsymbol{\chi}, \tilde{\mathbf{P}})$ necessarily satisfies

$$\begin{cases} \mathrm{Div}\mathrm{Div} D_{\mathbf{G}}\Psi - \mathrm{Div} D_{\mathbf{F}}\Psi - \mathrm{Div}\tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}} - \tilde{\mathbf{f}}^e = 0 & \text{in } \Omega_R, \\ \mathrm{Div}\tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}} = 0 & \text{in } V_R \setminus \Omega_R, \\ (D_{\mathbf{F}}\Psi)\mathbf{N} - (\mathrm{Div} D_{\mathbf{G}}\Psi)\mathbf{N} - [\![\tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}}]\!]\mathbf{N} - \boldsymbol{\tau} - \tilde{\mathbf{t}}^e = 0 & \text{on } S_N, \\ (D_{\mathbf{G}}\Psi)\mathbf{N} \otimes \mathbf{N} = 0 & \text{on } \partial\Omega_R. \end{cases} \tag{3.37}$$

The boundary value problem

In summary, by (3.8), (3.22), (3.21) and (3.36) we conclude that a solution $(\boldsymbol{\chi}, \tilde{\mathbf{P}})$ to the variational problem (3.12) necessarily satisfies the following differential equations:

$$\begin{cases} \mathbf{F}^{-T}\mathrm{Grad}\xi + D_{\tilde{\mathbf{P}}}\Psi - \mathrm{Div} D_{\boldsymbol{\Pi}}\Psi = 0 & \text{in } \Omega_R, \\ \mathrm{Div}\tilde{\mathbf{D}} = \tilde{\rho}^e, \quad \tilde{\mathbf{D}} = -\epsilon_0 J\mathbf{C}^{-1}\mathrm{Grad}\xi + \mathbf{F}^{-1}(\tilde{\mathbf{P}} + \tilde{\mathbf{P}}^e) & \text{in } V_R, \\ \mathrm{Div}\mathrm{Div} D_{\mathbf{G}}\Psi - \mathrm{Div} D_{\mathbf{F}}\Psi - \mathrm{Div}\tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}} - \tilde{\mathbf{f}}^e = 0 & \text{in } \Omega_R, \end{cases} \tag{3.38}$$

and boundary conditions (3.3), $(3.4)_1$ and

$$\begin{cases} (D_{\boldsymbol{\Pi}}\Psi)\mathbf{N} = 0 & \text{on } \partial\Omega_R, \\ (D_{\mathbf{F}}\Psi)\mathbf{N} - (\mathrm{Div} D_{\mathbf{G}}\Psi)\mathbf{N} - [\![\tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}}]\!]\mathbf{N} - \boldsymbol{\tau} - \tilde{\mathbf{t}}^e = 0 & \text{on } S_N, \\ (D_{\mathbf{G}}\Psi)\mathbf{N} \otimes \mathbf{N} = 0 & \text{on } \partial\Omega_R. \end{cases} \tag{3.39}$$

We remark that if the second of (3.38) holds, the second of (3.37) is identically satisfied, and henceforth, not repeated in (3.38) and subsequent equations. Further, the boundary value problems (3.38)-(3.39) follow as the Euler-Lagrangian equations for internal energy density function that depends on polarization, deformation gradient, and polarization gradient and strain gradient, i.e., $(\tilde{\mathbf{P}}, \mathrm{Grad}\tilde{\mathbf{P}}, \mathrm{Grad}\boldsymbol{\chi}, \mathrm{GradGrad}\boldsymbol{\chi})$. For many materials of interest, it may be sufficient to use a simpler form of internal energy which will be discussed subsequently.

3.2.1 Nongradient theory of elastic dielectrics

Since Toupin (1956) a frequently-used model for elastic dielectrics assumes that the internal energy of the body is given by

$$U[\boldsymbol{\chi}, \tilde{\mathbf{P}}] = \int_{\Omega_R} \Psi(\mathbf{X}; \mathrm{Grad}\boldsymbol{\chi}, \tilde{\mathbf{P}}), \tag{3.40}$$

where the internal energy density Ψ is independent of polarization gradient $\mathrm{Grad}\tilde{\mathbf{P}}$ and strain gradient $\mathrm{GradGrad}\boldsymbol{\chi}$. Then the boundary value problem (3.38)-(3.39) can be specialized as

$$\begin{cases} \mathbf{F}^{-T}\mathrm{Grad}\xi + D_{\tilde{\mathbf{P}}}\Psi = 0 & \text{in } \Omega_R, \\ \mathrm{Div}\tilde{\mathbf{D}} = \tilde{\rho}^e, \quad \tilde{\mathbf{D}} = -\epsilon_0 J\mathbf{C}^{-1}\mathrm{Grad}\xi + \mathbf{F}^{-1}(\tilde{\mathbf{P}} + \tilde{\mathbf{P}}^e) & \text{in } V_R, \\ \mathrm{Div} D_{\mathbf{F}}\Psi + \mathrm{Div}\tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}} + \tilde{\mathbf{f}}^e = 0 & \text{in } \Omega_R, \end{cases} \tag{3.41}$$

with boundary conditions (3.3), $(3.4)_1$ and

$$(D_{\mathbf{F}}\Psi)\mathbf{N} - [\![\tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}}]\!]\mathbf{N} - \tilde{\mathbf{t}}^e = 0 \quad \text{on } S_N. \tag{3.42}$$

We remark that the first of (3.41) is often interpreted as a constitutive law between the polarization and electric field.

3.2.2 Polarization gradient theory

As pioneered by Mindlin (1968), the polarization gradient theory for elastic dielectrics assumes the following form of internal energy:

$$U[\boldsymbol{\chi}, \tilde{\mathbf{P}}] = \int_{\Omega_R} \Psi(\mathbf{X}; \mathrm{Grad}\boldsymbol{\chi},\ \tilde{\mathbf{P}},\ \mathrm{Grad}\tilde{\mathbf{P}}). \tag{3.43}$$

Then the boundary value problem (3.38)-(3.39) can be rewritten as

$$\begin{cases} \mathbf{F}^{-T}\mathrm{Grad}\xi + D_{\tilde{\mathbf{P}}}\Psi - \mathrm{Div}D_{\mathbf{\Pi}}\Psi = 0 & \text{in } \Omega_R, \\ \mathrm{Div}\tilde{\mathbf{D}} = \tilde{\rho}^e, \quad \tilde{\mathbf{D}} = -\epsilon_0 J\mathbf{C}^{-1}\mathrm{Grad}\xi + \mathbf{F}^{-1}(\tilde{\mathbf{P}} + \tilde{\mathbf{P}}^e) & \text{in } V_R, \\ \mathrm{Div}D_{\mathbf{F}}\Psi + \mathrm{Div}\tilde{\mathbf{\Sigma}}_{\mathrm{MW}} + \tilde{\mathbf{f}}^e = 0 & \text{in } \Omega_R, \end{cases} \tag{3.44}$$

with boundary conditions (3.3), $(3.4)_1$, (3.42) and $(3.39)_1$.

3.2.3 Strain gradient (flexoelectric) theory

The internal energy is given by

$$U[\boldsymbol{\chi}, \tilde{\mathbf{P}}] = \int_{\Omega_R} \Psi(\mathbf{X}; \mathrm{Grad}\boldsymbol{\chi},\ \mathrm{GradGrad}\boldsymbol{\chi},\ \tilde{\mathbf{P}}). \tag{3.45}$$

Then by (3.38) we obtain

$$\begin{cases} \mathbf{F}^{-T}\mathrm{Grad}\xi + D_{\tilde{\mathbf{P}}}\Psi = 0 & \text{in } \Omega_R, \\ \mathrm{Div}\tilde{\mathbf{D}} = \tilde{\rho}^e, \quad \tilde{\mathbf{D}} = -\epsilon_0 J\mathbf{C}^{-1}\mathrm{Grad}\xi + \mathbf{F}^{-1}(\tilde{\mathbf{P}} + \tilde{\mathbf{P}}^e) & \text{in } V_R, \\ \mathrm{DivDiv}D_{\mathbf{G}}\Psi - \mathrm{Div}D_{\mathbf{F}}\Psi - \mathrm{Div}\tilde{\mathbf{\Sigma}}_{\mathrm{MW}} - \tilde{\mathbf{f}}^e = 0 & \text{in } \Omega_R, \end{cases} \tag{3.46}$$

with boundary conditions (3.3), $(3.4)_1$ and $(3.39)_{2,3}$.

3.2.4 Interfacial conditions

The body Ω_R could be inhomogeneous and in particular, there could be a sudden change of material properties. In terms of the internal energy density function, this sudden change of material properties can be recognized as interfaces across which the function $\mathbf{X} \mapsto \Psi(\mathbf{X}; \cdots)$ is discontinuous. Moreover, there could be different *types* of materials on the two sides of the interfaces. For example, the material on one side is sufficiently described by the nongradient theory while the material on the other side requires a more sophisticated theory such as the polarization gradient theory or the strain gradient theory. Across these interfaces, the differential equations such as (3.38) shall be interpreted as *interfacial conditions*. Although the interfacial conditions can be directly seen from the differential equations and the kinematic requirements ensuring that the internal energy is bounded (or integrable), to be clear we list in Appendix B the interfacial conditions across interfaces between different types of materials.

3.3 Geometric and constitutive linearization: general procedure

The presented general theory is not amenable to solution since (i) the governing electrostatic equation is nonlinear in the reference configuration, and (ii) the frame indifference implies that any physically reasonable internal energy function Ψ has to be a nonlinear function of state variables $(\boldsymbol{\chi}, \tilde{\mathbf{P}})$ and hence the mechanical equilibrium equation is nonlinear as well. We are therefore

motivated to linearize the theory and obtain explicit solutions to make predictions. The general procedure of linearization is outlined below whereas specific examples are presented in § 4-5.

For many materials and regimes of interest, the strain and the polarization are small:

$$\mathbf{S} := \mathrm{Grad}\mathbf{u} = \mathbf{F} - \mathbf{I} \sim \varepsilon, \qquad \tilde{\mathbf{P}} \sim \delta, \tag{3.47}$$

where $\mathbf{u}(\mathbf{X}) = \boldsymbol{\chi}(\mathbf{X}) - \mathbf{X}$ is the displacement, and ϵ, δ are two small numbers.[4] For materials without a spontaneous strain and polarization, we may impose that in the absence of external electrical and mechanical loadings, the trivial state $(\boldsymbol{\chi}, \tilde{\mathbf{P}}) = (\mathbf{X}, 0)$ is an equilibrium state that minimizes the total free energy, i.e.,

$$\frac{\partial \Psi}{\partial \mathbf{F}}\Big|_{(\mathbf{I},0,0,0)} = \frac{\partial \Psi}{\partial \mathbf{G}}\Big|_{(\mathbf{I},0,0,0)} = \frac{\partial \Psi}{\partial \tilde{\mathbf{P}}}\Big|_{(\mathbf{I},0,0,0)} = \frac{\partial \Psi}{\partial \boldsymbol{\Pi}}\Big|_{(\mathbf{I},0,0,0)} = 0, \tag{3.48}$$

and that the second derivatives of Ψ are semi-positive-definite:

$$D^2\Psi\Big|_{(\mathbf{I},0,0,0)} \geq 0. \tag{3.49}$$

To arrive at geometric linear theories we may expand and truncate the internal energy density function Ψ as

$$\begin{aligned}\Psi(\mathbf{F}, \mathbf{G}, \tilde{\mathbf{P}}, \boldsymbol{\Pi}) = \Psi(\mathbf{I}, 0, 0, 0) &+ \varepsilon^2 \mathcal{B}_1(\mathbf{S}_0, \mathbf{S}_0) + \varepsilon^2 \mathcal{B}_2(\mathbf{G}_0, \mathbf{G}_0) + \delta^2 \mathcal{B}_3(\tilde{\mathbf{P}}_0, \tilde{\mathbf{P}}_0) + \delta^2 \mathcal{B}_4(\boldsymbol{\Pi}_0, \boldsymbol{\Pi}_0) \\ &+ \varepsilon^2 \mathcal{B}_5(\mathbf{S}_0, \mathbf{G}_0) + \varepsilon\delta \mathcal{B}_6(\mathbf{S}_0, \tilde{\mathbf{P}}_0) + \varepsilon\delta \mathcal{B}_7(\mathbf{S}_0, \boldsymbol{\Pi}_0) + \varepsilon\delta \mathcal{B}_8(\mathbf{G}_0, \tilde{\mathbf{P}}_0) \\ &+ \varepsilon\delta \mathcal{B}_9(\mathbf{G}_0, \boldsymbol{\Pi}_0) + \delta^2 \mathcal{B}_{10}(\tilde{\mathbf{P}}_0, \boldsymbol{\Pi}_0) + \varepsilon\delta^2 \mathcal{B}_{11}(\mathbf{S}_0, \tilde{\mathbf{P}}_0 \otimes \tilde{\mathbf{P}}_0) + \cdots,\end{aligned}$$

where $\mathcal{B}_i$ $(i = 1, \cdots)$ are bilinear forms of their arguments, and $\mathbf{S}_0 = \mathbf{S}/\varepsilon$, $\tilde{\mathbf{P}}_0 = \tilde{\mathbf{P}}/\delta$, $\mathbf{G}_0 = \mathbf{G}/\varepsilon$, $\boldsymbol{\Pi}_0 = \boldsymbol{\Pi}/\delta$ are of order one. Inserting the above truncated energy function into (3.38)-(3.39) and keeping only the leading order terms for each equations, we will obtain a hierarchy of theories for different regimes of $a := \log\delta/\log\varepsilon$ that would describe the asymptotic behaviors of the body for "small strain" and "small polarization". The interested reader is referred to Tian (2007) for rigorous derivations of dielectric elastomers and piezoelectric materials in the framework of Γ-convergence. Below we present only the formal calculations of these theories in § 4-5 instead of rigorous Γ-convergence argument.

4 Theories of typical media and applications

4.1 Nonlinear dielectric elastomer

From the above analysis, we observe that elasticity and electricity are *intrinsically* coupled through the Maxwell stress. Dielectric materials can sustain an electric field before the bound electrons become free, which is referred to as the *dielectric strength*. The dielectric strength of materials is typically at the order $10^6 - 10^8 V/m$, and hence the maximum Maxwell stress ($\sim \epsilon|\mathbf{e}|^2$) before electric breakdown is roughly $10^6 Pa$. Therefore, the effect of Maxwell stress is only significant for soft rubber-like materials with Young's modulus at the order of $0.1 Gpa$ (1% strain) or less. For such a soft elastic materials, it is often necessary to use the nonlinear finite deformation theory of elasticity.

[4] Dimensionless criterion for δ may be established for "small polarization" by comparing with typical/saturation polarization.

To conform with the principle of frame indifference (3.6), the internal energy density Ψ can be rewritten as

$$\Psi(\mathbf{F},\tilde{\mathbf{P}}) = \psi(\mathbf{U},\mathbf{R}^T\tilde{\mathbf{P}}) \qquad (\text{recall that } \mathbf{U}=(\mathbf{F}^T\mathbf{F})^{1/2},\ \ \mathbf{R}=\mathbf{F}\mathbf{U}^{-1}). \tag{4.1}$$

Further, based on experimental measurements we postulate that the material is a *linear* dielectrics and the dielectric tensor is *independent of* strain $\mathbf{U}$. In other words, if the dielectric tensor of the body is given by $\boldsymbol{\epsilon} \in \mathbb{R}^{3\times 3}_{\text{sym}}$ with respect to a fixed Cartesian frame $\{\mathbf{e}_1,\mathbf{e}_2,\mathbf{e}_3\}$, then under any homogeneous deformation $\boldsymbol{\chi}(\mathbf{X})=\mathbf{F}\mathbf{X}$, the electric field-polarization relation of the deformed body in the *current configuration* is given by

$$\mathbf{e} = \mathbf{R}\mathbf{A}\mathbf{R}^T\mathbf{p}, \qquad \mathbf{A}=(\boldsymbol{\epsilon}-\epsilon_0\mathbf{I})^{-1}. \tag{4.2}$$

From the above postulation, by $(3.41)_1$ and (4.1) we infer that

$$\mathbf{e} = D_{\tilde{\mathbf{P}}}\Psi = \frac{1}{J}\mathbf{R}\mathbf{A}\mathbf{R}^T\tilde{\mathbf{P}} \qquad \forall\, \mathbf{U}\in\mathbb{R}^{3\times 3}_{\text{sym}}\ \&\ \tilde{\mathbf{P}}\in\mathbb{R}^3.$$

Therefore,

$$\psi(\mathbf{U},\mathbf{R}^T\tilde{\mathbf{P}}) = W_{\text{elast}}(\mathbf{U}) + \frac{1}{2J}(\mathbf{R}^T\tilde{\mathbf{P}})\cdot\mathbf{A}(\mathbf{R}^T\tilde{\mathbf{P}}). \tag{4.3}$$

It will be convenient to introduce the *modified Maxwell stress* for linear dielectrics:

$$\tilde{\boldsymbol{\Sigma}}'_{\text{MW}} = \tilde{\boldsymbol{\Sigma}}_{\text{MW}} - \frac{1}{2J}(\mathbf{R}^T\tilde{\mathbf{P}})\cdot\mathbf{A}(\mathbf{R}^T\tilde{\mathbf{P}})\mathbf{F}^{-T} \qquad \text{in } V_R,$$

which, by (1.4) and (4.2), in the *current configuration* can be expressed as

$$\boldsymbol{\sigma}'_{\text{MW}} = \boldsymbol{\sigma}_{\text{MW}} - \frac{1}{2}(\mathbf{R}^T\mathbf{p})\cdot\mathbf{A}(\mathbf{R}^T\mathbf{p})\mathbf{I} = \mathbf{e}\otimes\mathbf{d} - \frac{\mathbf{d}\cdot\mathbf{e}}{2}\mathbf{I} \qquad \text{in } V. \tag{4.4}$$

We remark that the above expression of Maxwell stress is particularly popular among authors in fluid mechanics (Stratton, 1941; Melcher, 1981). Inserting the above equation into (3.41) and noticing the identity $D_{\mathbf{F}}J^{-1} = -J^{-1}\mathbf{F}^{-T}$, we obtain the following equations in the *current configuration*:

$$\begin{cases} \text{div}\mathbf{d} = \rho^e, & \mathbf{d} = -(\epsilon_0\mathbf{I}+\mathbf{R}\mathbf{A}^{-1}\mathbf{R}^T\chi_\Omega)\text{grad}\xi + \mathbf{p} + \mathbf{p}^e \quad \text{in } V,\\ -\text{div}(\boldsymbol{\sigma}_{\text{mech}}+\boldsymbol{\sigma}'_{\text{MW}}) = \mathbf{f}^e, & \boldsymbol{\sigma}_{\text{mech}} = [\frac{1}{J}D_{\mathbf{F}}W_{\text{elast}}(\mathbf{U})\mathbf{F}^T]\circ\boldsymbol{\pi} \quad \text{in } \Omega, \end{cases} \tag{4.5}$$

where $\boldsymbol{\sigma}_{\text{mech}}$ can be identified as the mechanical part of the total stress that is independent of the polarization of the body.

If, in addition, the deformation is small in the sense that the strain

$$\mathbf{S} := \text{Grad}\mathbf{u} = \mathbf{F}-\mathbf{I} \qquad \big(\mathbf{u}(\mathbf{X}) := \boldsymbol{\chi}(\mathbf{X})-\mathbf{X}\big) \tag{4.6}$$

is at the order of ε, $\varepsilon << 1$, we can expand and truncate the internal energy function in a neighborhood of $\mathbf{F}=\mathbf{I}$ as

$$W_{\text{elast}}(\mathbf{U}) = \frac{1}{2}\mathbf{S}\cdot\mathbf{C}\mathbf{S} + o(\varepsilon^2), \qquad \mathbf{C} = \frac{\partial^2 W_{\text{elast}}(\mathbf{U})}{\partial\mathbf{F}\partial\mathbf{F}}\Big|_{\mathbf{F}=\mathbf{I}},$$

where the forth-order tensor $\mathbf{C}:\mathbb{R}^{3\times 3}_{\text{sym}}\to\mathbb{R}^{3\times 3}_{\text{sym}}$ is recognized as the familiar *elastic stiffness* tensor. Inserting the above equation into (3.41)-(3.42) and keeping only the leading-order terms in each of (4.5), we obtain the simplified boundary value problems for dielectric elastomers:

$$\begin{cases} \text{div}[-(\epsilon_0\mathbf{I}+\mathbf{A}^{-1}\chi_\Omega)\nabla\xi + \mathbf{P}^e] = \rho^e & \text{in } V_R,\\ -\text{div}(\mathbf{C}\nabla\mathbf{u}+\boldsymbol{\sigma}'_{\text{MW}}) = \mathbf{f}^e & \text{in } \Omega_R, \end{cases} \tag{4.7}$$

with electrical boundary conditions

$$\xi - \xi_b = 0 \quad \text{on } \Gamma_D, \qquad k(\xi - \xi_b) - \mathbf{n}\cdot\mathbf{D} - \sigma_0 = 0 \quad \text{on } \Gamma_R, \tag{4.8}$$

and mechanical boundary conditions

$$\begin{cases} \mathbf{u} = \mathbf{u}_0 & \text{on } S_D, \\ (\mathbf{C}\nabla\mathbf{u})\mathbf{n} - [\![\boldsymbol{\sigma}'_{\mathrm{MW}}]\!]\mathbf{n} - \mathbf{t}^e = 0 & \text{on } S_N. \end{cases} \tag{4.9}$$

In the above equations (4.7)-(4.9), as usual, for small strains we no longer differentiate between the reference and current configurations.

4.2 Photoelasticity

The *stress-optic* law of photoelasticity can be formulated by setting

$$\psi(\mathbf{U}, \mathbf{R}^T\tilde{\mathbf{P}}) = W_{\mathrm{elast}}(\mathbf{U}) + \frac{1}{2J}(\mathbf{R}^T\tilde{\mathbf{P}})\cdot\mathbf{A}(\mathbf{U})(\mathbf{R}^T\tilde{\mathbf{P}}). \tag{4.10}$$

To see this, by $(3.41)_1$ the above internal energy function implies

$$\mathbf{e} = \mathbf{R}\mathbf{A}(\mathbf{U})\mathbf{R}^T\mathbf{p}, \quad \text{i.e.,} \quad \mathbf{d} = \mathbf{R}[\epsilon_0\mathbf{I} + \mathbf{A}(\mathbf{U})^{-1}]\mathbf{R}^T\mathbf{e}.$$

In other words, the permittivity tensor of the material depends on the strain.

For an *isotropic* photoelastic materials, i.e., the symmetry group of the material includes all rigid rotations $\mathcal{G} = So(3)$ (cf. (3.7)), the functional dependence of tensor $\mathbf{A}$ on strain $\mathbf{U}$ is strongly restricted. To see this, we first notice that for $\mathbf{Q} \in So(3)$, the transformation $\mathbf{F} = \mathbf{R}\mathbf{U} \to \mathbf{F}' = \mathbf{F}\mathbf{Q} = \mathbf{R}\mathbf{U}\mathbf{Q} =: \mathbf{R}'\mathbf{U}'$ implies that

$$\mathbf{U}' = \mathbf{Q}^T\mathbf{U}\mathbf{Q}, \qquad \mathbf{R}' = \mathbf{R}\mathbf{Q} \tag{4.11}$$

Therefore, by (3.7) we have that for any $\mathbf{Q} \in \mathcal{G} = So(3)$,

$$W_{\mathrm{elast}}(\mathbf{U}) + \frac{1}{2J}\tilde{\mathbf{P}}\cdot\mathbf{R}\mathbf{A}(\mathbf{U})\mathbf{R}^T\tilde{\mathbf{P}} = W_{\mathrm{elast}}(\mathbf{Q}^T\mathbf{U}\mathbf{Q}) + \frac{1}{2J}\tilde{\mathbf{P}}\cdot\mathbf{R}\mathbf{Q}\mathbf{A}(\mathbf{Q}^T\mathbf{U}\mathbf{Q})\mathbf{Q}^T\mathbf{R}^T\tilde{\mathbf{P}}.$$

Since $(\mathbf{U}, \tilde{\mathbf{P}})$ can be arbitrarily chosen, the above equation implies that for all $\mathbf{Q} \in So(3)$,

$$\begin{cases} W_{\mathrm{elast}}(\mathbf{U}) = W_{\mathrm{elast}}(\mathbf{Q}^T\mathbf{U}\mathbf{Q}), \\ \mathbf{A}(\mathbf{U}) = \mathbf{Q}\mathbf{A}(\mathbf{Q}^T\mathbf{U}\mathbf{Q})\mathbf{Q}^T. \end{cases} \tag{4.12}$$

Clearly, the first of (4.12) is the usual requirement for the strain energy function of isotropic materials whereas the second of (4.12) implies that $\mathbf{A}(\mathbf{I}) = \frac{1}{\epsilon-\epsilon_0}\mathbf{I}$. ($\epsilon$ - the permittivity of the undeformed material) Further, for small strain $\mathbf{U} - \mathbf{I} \approx \frac{1}{2}(\mathbf{S} + \mathbf{S}^T) \sim \eta << 1$ we can expand and truncate the matrix function $\mathbf{F} \mapsto \mathbf{A}(\mathbf{U})$ in a neighborhood of $\mathbf{F} = \mathbf{I}$:

$$\mathbf{A}(\mathbf{U}) = \frac{1}{\epsilon - \epsilon_0}\mathbf{I} + \mathbf{O}(\mathbf{U} - \mathbf{I}) + o(\eta),$$

where the forth-order tensor $\mathbf{O} : \mathbb{R}^{3\times3}_{\mathrm{sym}} \to \mathbb{R}^{3\times3}_{\mathrm{sym}}$, by the second of (4.12), necessarily satisfies that for any $\mathbf{Q} \in \mathcal{G} = So(3)$,

$$O_{ijkl} = Q_{ii'}Q_{jj'}Q_{kk'}Q_{ll'}S_{i'j'k'l'}.$$

We immediately recognize that the above equation implies the tensor $\mathbf{O}$ has to be the same form as an isotropic elasticity tensor, and hence there are two material constants a_μ and a_λ such that

$$\mathbf{O}(\mathbf{U}-\mathbf{I}) = 2a_\mu(\mathbf{U}-\mathbf{I}) + a_\lambda \mathrm{Tr}(\mathbf{U}-\mathbf{I})\mathbf{I}.$$

In conclusion, the permittivity tensor of the *strained* material is given by

$$\begin{aligned}\boldsymbol{\epsilon}(\mathbf{U}) &= \epsilon_0\mathbf{I} + \mathbf{A}(\mathbf{U})^{-1} = \epsilon_0\mathbf{I} + \Big[\frac{1}{\epsilon-\epsilon_0}\mathbf{I} + 2a_\mu(\mathbf{U}-\mathbf{I}) + a_\lambda\mathrm{Tr}(\mathbf{U}-\mathbf{I})\mathbf{I}\Big]^{-1}\\ &= \epsilon\mathbf{I} - (\epsilon-\epsilon_0)^2\Big[2a_\mu(\mathbf{U}-\mathbf{I}) + a_\lambda\mathrm{Tr}(\mathbf{U}-\mathbf{I})\mathbf{I}\Big] + o(\eta).\end{aligned}$$

Recall that the optical birefringence $\mathbf{n}'$ is the deviatoric part of the refractive index matrix $\mathbf{n} = (\mu_0\boldsymbol{\epsilon})^{1/2}$. (Here, the magnetic permeability is assumed to be independent of strain and given by μ_0.) For small strain and linearized elasticity, it is easy to see that the above equation implies the linear *stress-optic* law for an *isotropic* photoelastic materials:

$$\mathbf{n}' = C_B\boldsymbol{\sigma}', \tag{4.13}$$

where $\boldsymbol{\sigma}'$ is the deviatoric stress and C_B is referred to as the *Brewsters* constant. From the above calculation, we see that the above widely used stress-optic law of birefringence/anisotropy arises precisely from the *isotropy* of the photoelastic materials. Similar calculations can be carried out and yield restrictions on the *stress-optic* law for general anisotropic photoelastic crystals.

4.3 Piezoelectricity

The theory of piezoelectricity proposed by Voight (1910) can be recovered by adding a *coupling* term $\mathcal{C}_{\mathrm{pze}}(\mathbf{U}, \mathbf{R}^T\tilde{\mathbf{P}})$ between strain and polarization:

$$\psi(\mathbf{U}, \mathbf{R}^T\tilde{\mathbf{P}}) = W_{\mathrm{elast}}(\mathbf{U}) + \mathcal{C}_{\mathrm{pze}}(\mathbf{U}, \mathbf{R}^T\tilde{\mathbf{P}}) + \frac{1}{2J}(\mathbf{R}^T\tilde{\mathbf{P}})\cdot\mathbf{A}(\mathbf{R}^T\tilde{\mathbf{P}}). \tag{4.14}$$

Naturally, we impose the restrictions (3.48) and (3.49). To conform with (3.48), a simple and linear coupling term may be postulated as

$$\mathcal{C}_{\mathrm{pze}}(\mathbf{U}, \mathbf{R}^T\tilde{\mathbf{P}}) = (\mathbf{R}^T\tilde{\mathbf{P}})\cdot\mathbf{B}(\mathbf{U}-\mathbf{I}), \tag{4.15}$$

where $\mathbf{B} : \mathbb{R}^{3\times3}_{\mathrm{sym}} \to \mathbb{R}^3$ is a third-order tensor characterizing the piezoelectric effects of the body. For materials with symmetry gourp $\mathcal{G}$, by (3.7) and (4.11) we have that for any $\mathbf{Q} \in \mathcal{G}$,

$$B_{ijk} = Q_{ii'}Q_{jj'}Q_{kk'}B_{i'j'k'}$$

For small strains, upon expansion and truncation we can rewrite the internal energy function as (recall that $\mathbf{S} = \mathbf{F} - \mathbf{I} \sim \varepsilon << 1$)

$$\Psi(\mathbf{F}, \mathbf{P}) = \frac{1}{2}\mathbf{S}\cdot\mathbf{C}\mathbf{S} + \mathbf{P}\cdot\mathbf{B}\mathbf{S} + \frac{1}{2}\mathbf{P}\cdot\mathbf{A}\mathbf{P}. \tag{4.16}$$

Then by (3.41)-(3.42) we obtain the simplified boundary value problems for piezoelectric materials:

$$\begin{cases}\nabla\xi + \mathbf{B}\nabla\mathbf{u} + \mathbf{A}\mathbf{P} = 0 & \text{in } \Omega,\\ -\mathrm{div}(\mathbf{C}\nabla\mathbf{u} + \mathbf{B}^T\mathbf{P} + \boldsymbol{\sigma}'_{\mathrm{MW}}) = \mathbf{f}^e & \text{in } \Omega,\\ \mathrm{div}(-\epsilon_0\nabla\xi + \mathbf{P} + \mathbf{P}^e) = \rho^e & \text{in } V,\end{cases} \tag{4.17}$$

with boundary conditions (4.8) and (4.9). In addition, piezoelectric materials are typically hard ceramics with Young's modulus at the order of 10-100 *Gpa* (Yang, 2009). Therefore, we can safely neglect the Maxwell stress $\boldsymbol{\sigma}'_{\mathrm{MW}}$ in $(4.17)_2$ and the boundary condition $(4.9)_2$ since it is at most at the order of 10^6 Pa before electric breakdown.

The theory of piezoelectricity is typically formulated by postulating constitutive laws between stress-electric displacement $(\boldsymbol{\sigma}, \mathbf{D})$ and strain-electric field $(\mathbf{S}, \mathbf{E})$, see e.g. Yang (2009). To recover these constitutive relations in our formulation, we notice that the first and second of (4.17) can be interpreted as the constitutive relations

$$\begin{cases} \boldsymbol{\sigma} = D_{\mathbf{S}}\Psi, \\ \mathbf{E} = D_{\mathbf{P}}\Psi, \end{cases} \quad \Rightarrow \quad \begin{cases} \sigma_{ij} = C_{ijkl}S_{kl} + B_{kij}P_k, \\ E_i = B_{ikl}S_{kl} + A_{ij}P_j. \end{cases}$$

By algebraic calculations we can show that the above relations can be rewritten as

$$\begin{cases} \sigma_{ij} = C'_{ijkl}S_{kl} + B'_{kij}D_k, \\ E_i = B'_{ikl}S_{kl} + A'_{ij}D_j, \end{cases} \quad \text{or} \quad \begin{cases} \sigma_{ij} = C''_{ijkl}S_{kl} - B''_{kij}E_k, \\ D_i = B''_{ikl}S_{kl} + (A''_{ij} + \epsilon_0\delta_{ij})E_j, \end{cases}$$

where

$$\begin{cases} A'_{ij} = [(\mathbf{A}^{-1} + \epsilon_0\mathbf{I})^{-1}]_{ij}, \\ B'_{kij} = [(\mathbf{I} + \epsilon_0\mathbf{A})^{-1}]_{km}B_{mij}, \\ C'_{ijkl} = C_{ijkl} - \epsilon_0 B_{mij}[(\mathbf{I} + \epsilon_0\mathbf{A})^{-1}]_{mn}B_{nkl}, \end{cases} \quad \begin{cases} A''_{ij} = (\mathbf{A}^{-1})_{ij}, \\ B''_{kij} = -(\mathbf{A}^{-1})_{km}B_{mij}, \\ C''_{ijkl} = C_{ijkl} - B_{mij}A_{mn}B_{nkl}. \end{cases}$$

Direct calculations yield

$$\begin{aligned} \Psi'(\mathbf{S}, \mathbf{D}) &= \frac{1}{2}(\boldsymbol{\sigma} \cdot \mathbf{S} + \mathbf{E} \cdot \mathbf{D}) = \frac{1}{2}\mathbf{S} \cdot \mathbf{C}'\mathbf{S} + \mathbf{D} \cdot \mathbf{B}'\mathbf{S} + \frac{1}{2}\mathbf{D} \cdot \mathbf{A}'\mathbf{D} \\ &= \frac{1}{2}[\boldsymbol{\sigma} \cdot \mathbf{S} + \mathbf{E} \cdot (\mathbf{P} + \epsilon_0\mathbf{E})] = \Psi + \frac{\epsilon_0}{2}|\mathbf{E}|^2. \end{aligned}$$

Therefore, the stability requirement (3.49) in our formulation, i.e., $\Psi \geq 0$ for all $(\mathbf{S}, \mathbf{P}) \in \mathbb{R}^{3\times3}_{\mathrm{sym}} \times \mathbb{R}^3$, is stronger than the conventional requirement that $\Psi' \geq 0$ for all $(\mathbf{S}, \mathbf{D}) \in \mathbb{R}^{3\times3}_{\mathrm{sym}} \times \mathbb{R}^3$; the polarization $\mathbf{P}$ being the independent variable implies stronger stability conditions than the electric displacement being the independent variable (Landau and Lifshitz, 1995, § 14 vs. § 18).

4.4 Ferroelectrics

Ferroelectric materials may be phenomenologically described by postulating the internal energy density:

$$\Psi(\mathbf{F}, \tilde{\mathbf{P}}, \boldsymbol{\Pi}) = \frac{1}{2}\boldsymbol{\Pi} \cdot \mathbf{C}_{\mathrm{pg}}\boldsymbol{\Pi} + \psi(\mathbf{U}, \mathbf{R}^T\tilde{\mathbf{P}}), \tag{4.18}$$

where $\mathbf{C}_{\mathrm{pg}} : \mathbb{R}^{3\times3} \to \mathbb{R}^{3\times3}$ is a fourth-order positive tensor, and $\psi : \mathbb{R}^{3\times3}_{\mathrm{sym}} \times \mathbb{R}^3 \to \mathbb{R}$ is the part of internal energy density depending on the local strain and polarization. The term $\frac{1}{2}\boldsymbol{\Pi} \cdot \mathbf{C}_{\mathrm{pg}}\boldsymbol{\Pi}$ is included to reflect the nonlocal exchange effects of polarization.

Below the phase transition temperature (Curie temperature), a ferroelectric body has a spontaneous polarization and associated strain which can be modelled by assuming ψ is minimized at, e.g.,

$(\mathbf{U}_1, \mathbf{p}_1)$. By material symmetries (3.7) and (4.11), we immediately infer that ψ is also minimized at $(\mathbf{Q}^T\mathbf{U}_1\mathbf{Q}, \mathbf{Q}^T\mathbf{p}_1)$ for all $\mathbf{Q} \in \mathcal{G}$. By (3.44), the boundary value problem for $\xi, \boldsymbol{\chi}, \tilde{\mathbf{P}}$ is as follows:

$$\begin{cases} \mathbf{F}^{-T}\mathrm{Grad}\xi + D_{\tilde{\mathbf{P}}}\psi - \mathrm{Div}(\mathbf{C}_{\mathrm{pg}}\mathrm{Grad}\tilde{\mathbf{P}}) = 0 & \text{in } \Omega_R, \\ \mathrm{Div}\tilde{\mathbf{D}} = \tilde{\rho}^e, \quad \tilde{\mathbf{D}} = -\epsilon_0 J\mathbf{C}^{-1}\mathrm{Grad}\xi + \mathbf{F}^{-1}(\tilde{\mathbf{P}} + \tilde{\mathbf{P}}^e) & \text{in } V_R, \\ \mathrm{Div}D_{\mathbf{F}}\psi + \mathrm{Div}\tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}} + \tilde{\mathbf{f}}^e = 0 & \text{in } \Omega_R, \end{cases} \tag{4.19}$$

with boundary conditions (3.3), $(3.4)_1$ and

$$\begin{cases} (\mathbf{C}_{\mathrm{pg}}\mathrm{Grad}\tilde{\mathbf{P}})\mathbf{N} = 0 & \text{on } \partial\Omega_R, \\ (D_{\mathbf{F}}\psi)\mathbf{N} - [\![\tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}}]\!]\mathbf{N} - \tilde{\mathbf{t}}^e = 0 & \text{on } S_N. \end{cases} \tag{4.20}$$

We remark that the variational principle (3.12), together with the free energy functional (3.9) and internal energy density (4.18), is referred to as the Landau-Ginzburg-Devonshire theory of ferroelectrics (Landau, 1937; Ginzburg and Landau, 1950; Devonshire, 1949; 1954; Cao 2008).

4.5 Polarization gradient theory

Introduced by Mindlin (1968), the polarization gradient theory assumes that

$$\Psi(\mathbf{F}, \tilde{\mathbf{P}}, \boldsymbol{\Pi}) = \psi(\mathbf{U}, \mathbf{R}^T\tilde{\mathbf{P}}, \mathbf{R}^T\boldsymbol{\Pi}).$$

As a generalization of nongradient theories, for simplicity we further assume that

$$\begin{aligned} \psi(\mathbf{U}, \mathbf{R}^T\tilde{\mathbf{P}}, \mathbf{R}^T\boldsymbol{\Pi}) = W_{\mathrm{elast}}(\mathbf{U}) + \mathcal{C}_{\mathrm{pze}}(\mathbf{U}, \mathbf{R}^T\tilde{\mathbf{P}}) + \mathcal{C}_{\mathrm{pg}}(\mathbf{U}, \mathbf{R}^T\boldsymbol{\Pi}) \\ + \frac{1}{2}\boldsymbol{\Pi}\cdot\mathbf{C}_{\mathrm{pg}}\boldsymbol{\Pi} + \frac{1}{2J}(\mathbf{R}^T\tilde{\mathbf{P}})\cdot\mathbf{A}(\mathbf{R}^T\tilde{\mathbf{P}}), \end{aligned}$$

where $\mathcal{C}_{\mathrm{pg}}(\mathbf{U}, \mathbf{R}^T\boldsymbol{\Pi})$ is the coupling term between deformation gradient and polarization gradient. For small strains, by expansion and truncation we rewrite the internal energy function as (recall that $\mathbf{S} = \mathbf{F} - \mathbf{I}$):

$$\Psi(\mathbf{F}, \mathbf{P}) = \frac{1}{2}\mathbf{S}\cdot\mathbf{C}\mathbf{S} + \boldsymbol{\Pi}\cdot\mathbf{B}^{\mathrm{pg}}\mathbf{S} + \mathbf{P}\cdot\mathbf{B}^{\mathrm{pze}}\mathbf{S} + \frac{1}{2}\boldsymbol{\Pi}\cdot\mathbf{C}^{\mathrm{pg}}\boldsymbol{\Pi} + \frac{1}{2}\mathbf{P}\cdot\mathbf{A}\mathbf{P}, \tag{4.21}$$

where by frame indifference (3.6) the tensors $\mathbf{C}^{\mathrm{pg}}$ and $\mathbf{B}^{\mathrm{pg}}$ shall enjoy the major and minor symmetries as the elastic stiffness tensor $\mathbf{C}$. Further, for isotropic media, by material symmetry (3.7) we infer that $\mathbf{B}^{\mathrm{pze}} = 0$, and $\mathbf{C}^{\mathrm{pg}}$ and $\mathbf{B}_{\mathrm{pg}}$ shall be of the following form:

$$(\mathbf{B}^{\mathrm{pg}})_{piqj} = b_\mu(\delta_{pq}\delta_{ij} + \delta_{pj}\delta_{iq}) + b_\lambda\delta_{pi}\delta_{qj}, \qquad (\mathbf{C}^{\mathrm{pg}})_{piqj} = c_\mu(\delta_{pq}\delta_{ij} + \delta_{pj}\delta_{iq}) + c_\lambda\delta_{pi}\delta_{qj},$$

where b_μ, b_λ and c_μ, c_λ are constant in analogy with the Lamé constants μ, λ in the isotropic elastic tensor. Therefore, by (3.44) we have the boundary value problem:

$$\begin{cases} \nabla\xi + \frac{1}{\epsilon-\epsilon_0}\mathbf{P} - \mathrm{div}(\mathbf{B}^{\mathrm{pg}}\nabla\mathbf{u}) - \mathrm{div}(\mathbf{C}^{\mathrm{pg}}\nabla\mathbf{P}) = 0 & \text{in } \Omega, \\ \mathrm{div}\mathbf{D} = \rho^e, \qquad \mathbf{D} = -\epsilon_0\nabla\xi + \mathbf{P} + \mathbf{P}^e & \text{in } V, \\ -\mathrm{div}(\mathbf{C}\nabla\mathbf{u} + \mathbf{B}^{\mathrm{pg}}\nabla\mathbf{P}) - \mathrm{div}\boldsymbol{\sigma}'_{\mathrm{MW}} - \mathbf{f}^e = 0 & \text{in } \Omega, \end{cases} \tag{4.22}$$

with boundary conditions (4.8), $(4.9)_1$ and

$$\begin{cases} (\mathbf{C}\nabla\mathbf{u} + \mathbf{B}_{\mathrm{pg}}\nabla\mathbf{P})\mathbf{n} - [\![\boldsymbol{\sigma}'_{\mathrm{MW}}]\!]\mathbf{n} - \mathbf{t}^e = 0 & \text{on } S_N, \\ (\mathbf{B}^{\mathrm{pg}}\nabla\mathbf{u} + \mathbf{C}^{\mathrm{pg}}\nabla\mathbf{P})\mathbf{n} = 0 & \text{on } \partial\Omega. \end{cases} \tag{4.23}$$

We remark that upon neglecting the effect of Maxwell stress, the above linearized polarization gradient has been addressed in Buchaman *et al.* (1989).

4.6 Theory of flexoelectricity

Flexoelectricity refers to the coupling between strain gradient and polarization and can be modelled by the internal energy density function of the following form:

$$\Psi(\mathbf{F},\mathbf{G},\tilde{\mathbf{P}}) = W_{\text{elast}}(\mathbf{U}) + \mathcal{C}_{\text{flexo}}(\mathbf{R}^T\mathbf{G},\mathbf{R}^T\tilde{\mathbf{P}}) + \frac{1}{2J}(\mathbf{R}^T\tilde{\mathbf{P}})\cdot\mathbf{A}(\mathbf{R}^T\tilde{\mathbf{P}}), \tag{4.24}$$

where $\mathcal{C}_{\text{flexo}}(\mathbf{R}^T\mathbf{G},\mathbf{R}^T\tilde{\mathbf{P}})$ reflects the coupling between strain gradient and polarization. To conform with the restrictions (3.48) and (3.49), a simple flexoelectric coupling term may be postulated as

$$\mathcal{C}_{\text{flexo}}(\mathbf{R}^T\mathbf{G},\mathbf{R}^T\tilde{\mathbf{P}}) = \mathcal{B}_g(\mathbf{R}^T\mathbf{G},\mathbf{R}^T\mathbf{G}) + \mathcal{B}_f(\mathbf{R}^T\mathbf{G},\mathbf{R}^T\tilde{\mathbf{P}}),$$

where $\mathcal{B}_g$ and $\mathcal{B}_f$ are bilinear forms of their arguments. We remark that an additional term B_g is necessary for the stability of the trivial solution $(\mathbf{F},\mathbf{G},\tilde{\mathbf{P}}) = (\mathbf{I},0,0)$. By material symmetry (3.7), these bilinear form shall satisfy that

$$\mathcal{B}_g(\mathbf{R}^T\mathbf{G}_{\mathbf{Q}},\mathbf{R}^T\mathbf{G}_{\mathbf{Q}}) = \mathcal{B}_g(\mathbf{R}^T\mathbf{G},\mathbf{R}^T\mathbf{G}), \qquad \mathcal{B}_f(\mathbf{R}^T\mathbf{G}_{\mathbf{Q}},\mathbf{R}^T\tilde{\mathbf{P}}\mathbf{Q}) = \mathcal{B}_f(\mathbf{R}^T\mathbf{G},\mathbf{R}^T\tilde{\mathbf{P}}),$$

for any $\mathbf{Q}\in\mathcal{G}$, where $(\mathbf{G}_{\mathbf{Q}})_{pij} = (\mathbf{G})_{pkl}(\mathbf{Q})_{ki}(\mathbf{Q})_{lj}$.

For isotropic media, i.e., $\mathcal{G} = So(3)$, the most general bilinear forms of $\mathcal{B}_g$ and $\mathcal{B}_f$ satisfying the above identity can be systematically derived, see, e.g., Jaric et al. (2008). For simplicity we assume a special form of B_g and B_f as follows:

$$\mathcal{B}_g(\mathbf{R}^T\mathbf{G},\mathbf{R}^T\mathbf{G}) = \frac{g}{2}(\mathbf{G})_{ikk}(\mathbf{G})_{ill}, \qquad \mathcal{B}_f(\mathbf{R}^T\mathbf{G},\mathbf{R}^T\tilde{\mathbf{P}}) = f(\tilde{\mathbf{P}})_i(\mathbf{G})_{ikk}. \tag{4.25}$$

Then the boundary value problems (3.46) can be simplified as

$$\begin{cases} \mathbf{F}^{-T}\text{Grad}\xi + \frac{1}{J(\epsilon-\epsilon_0)}\tilde{\mathbf{P}} + f\text{Lap}\,\boldsymbol{\chi} = 0 & \text{in } \Omega_R,\\ \text{Div}\tilde{\mathbf{D}} = \tilde{\rho}^e, \quad \tilde{\mathbf{D}} = -\epsilon_0 J\mathbf{C}^{-1}\text{Grad}\xi + \mathbf{F}^{-1}(\tilde{\mathbf{P}}+\tilde{\mathbf{P}}^e) & \text{in } V_R,\\ -\text{Div}D_{\mathbf{F}}W_{\text{elast}} + \text{Lap}(g\text{Lap}\boldsymbol{\chi} + f\tilde{\mathbf{P}}) - \text{Div}\tilde{\boldsymbol{\Sigma}}'_{\text{MW}} - \tilde{\mathbf{f}}^e = 0 & \text{in } \Omega_R, \end{cases} \tag{4.26}$$

with boundary conditions (3.3), $(3.4)_1$ and

$$\begin{cases} (D_{\mathbf{F}}W_{\text{elast}})\mathbf{N} - [\text{Grad}(g\text{Lap}\boldsymbol{\chi} + f\tilde{\mathbf{P}})]\mathbf{N} - [\![\tilde{\boldsymbol{\Sigma}}'_{\text{MW}}]\!]\mathbf{N} - \tilde{\mathbf{t}}^e = 0 & \text{on } S_N,\\ g\text{Lap}\boldsymbol{\chi} + f\tilde{\mathbf{P}} = 0 & \text{on } \partial\Omega_R, \end{cases} \tag{4.27}$$

where Lap = DivGrad denotes the Laplace operator with respect to Lagrangian coordinates.

To arrive at a linear theory for small strain, keeping only the leading order terms the internal energy can be written as (recall that $\mathbf{u} = \boldsymbol{\chi} - \mathbf{X}$)

$$U[\boldsymbol{\chi},\mathbf{P}] = \int_\Omega \Big[\frac{g}{2}|\Delta\mathbf{u}|^2 + f\mathbf{P}\cdot\Delta\mathbf{u} + \frac{1}{2}\nabla\mathbf{u}\cdot\mathbf{C}\nabla\mathbf{u} + \frac{1}{2(\epsilon-\epsilon_0)}|\mathbf{P}|^2\Big], \tag{4.28}$$

and the boundary value problems (4.26)-(4.27) can be rewritten as

$$\begin{cases} -\nabla\xi = \frac{1}{\epsilon-\epsilon_0}\mathbf{P} + f\Delta\mathbf{u} & \text{in } \Omega,\\ \text{div}(-\epsilon_0\nabla\xi + \mathbf{P} + \mathbf{P}^e) = \rho^e & \text{in } V,\\ -\text{div}(\mathbf{C}\nabla\mathbf{u}) + \Delta(g\Delta\mathbf{u} + f\mathbf{P}) - \text{div}\boldsymbol{\sigma}'_{\text{MW}} - \mathbf{f}^e = 0 & \text{in } \Omega, \end{cases} \tag{4.29}$$

with boundary conditions (4.8), $(4.9)_1$, and

$$\begin{cases} (\mathbf{C}\nabla\mathbf{u})\mathbf{n} - [\nabla(g\Delta\mathbf{u} + f\mathbf{P})]\mathbf{n} - [\![\boldsymbol{\sigma}'_{\text{MW}}]\!]\mathbf{n} - \mathbf{t}^e = 0 & \text{on } S_N,\\ g\Delta\mathbf{u} + f\mathbf{P} = 0 & \text{on } \partial\Omega. \end{cases} \tag{4.30}$$

Here, again, we no longer differentiate the reference and current configurations for small strains.

5 Generalization to include magnetization

5.1 Free energy and boundary value problems

The energy formulation concerning electrical-mechanical couplings can be generalized to include another important physical phenomena: magnetics. The mutual couplings between strain, polarization and magnetization can be modelled by postulating the following form of internal energy

$$U[\boldsymbol{\chi}, \tilde{\mathbf{P}}, \tilde{\mathbf{M}}] = \int_{\Omega_R} \Psi(\mathrm{Grad}\boldsymbol{\chi}, \mathrm{GradGrad}\boldsymbol{\chi}, \tilde{\mathbf{P}}, \mathrm{Grad}\tilde{\mathbf{P}}, \tilde{\mathbf{M}}, \mathrm{Grad}\tilde{\mathbf{M}}),$$

where $\tilde{\mathbf{M}} : \Omega_R \to \mathbb{R}^3$ is the intrinsic magnetization of the body,

$$\Psi = \Psi(\mathbf{F}, \mathbf{G}, \tilde{\mathbf{P}}, \boldsymbol{\Pi}, \tilde{\mathbf{M}}, \boldsymbol{\Lambda})$$

is the internal energy density function that depends on deformation gradient, polarization, magnetization and their gradients.

To fix the magnetic energy of the system, we need to prescribe the external magnetic sources and "boundary devices" that interact with the body. For simplicity, we will assume that there is a distribution of external magnetic moments $\tilde{\mathbf{M}}^e : \Omega_R \to \mathbb{R}^3$ on the reference body. For ease of notation, the intrinsic and external magnetizations are extended by zero to the entire space $\mathbb{R}^3$. Further, upon removing the body and external magnetization an external field $\mathbf{h}^e : \mathbb{R}^3 \to \mathbb{R}^3$ permeates into the entire space. In other words, this external field $\mathbf{h}^e$ arises from magnetization or electric currents at the infinity. Then by the Maxwell equations (1.1b), the total magnetic field $-\nabla\zeta$ shall satisfy

$$\begin{cases} \mathrm{div}(-\mathrm{grad}\zeta + \mathbf{m} + \mathbf{m}^e) = 0 & \text{in } \mathbb{R}^3, \\ -\mathrm{grad}\zeta \to \mathbf{h}^e & \text{as } |\mathbf{x}| \to +\infty, \end{cases} \tag{5.1}$$

where (cf., Fig. 1, (1.7) and (3.1))

$$\mathbf{m}^e(\mathbf{x}) = \frac{1}{J}\tilde{\mathbf{M}}^e \circ \boldsymbol{\pi}(\mathbf{x}), \qquad \mathbf{m}(\mathbf{x}) = \frac{1}{J}\tilde{\mathbf{M}} \circ \boldsymbol{\pi}(\mathbf{x}).$$

In the reference configuration, the above equation (5.1) can be rewritten as

$$\begin{cases} \mathrm{Div}[-J\mathbf{C}^{-1}\mathrm{Grad}\zeta + \mathbf{F}^{-1}(\tilde{\mathbf{M}} + \tilde{\mathbf{M}}^e)] = 0 & \text{in } \mathbb{R}^3, \\ -\mathrm{Grad}\zeta \to \mathbf{H}^e & \text{as } |\mathbf{x}| \to +\infty, \end{cases} \tag{5.2}$$

where $\mathbf{H}^e = \mathbf{h}^e \circ \boldsymbol{\chi}$. Then the total free energy can be identified as

$$F[\boldsymbol{\chi}, \tilde{\mathbf{P}}, \tilde{\mathbf{M}}] = U[\boldsymbol{\chi}, \tilde{\mathbf{P}}, \tilde{\mathbf{M}}] + \mathcal{E}^{\mathrm{elect}}[\boldsymbol{\chi}, \tilde{\mathbf{P}}] + \mathcal{E}^{\mathrm{mag}}[\boldsymbol{\chi}, \tilde{\mathbf{M}}] + P^{\mathrm{mech}}[\boldsymbol{\chi}], \tag{5.3}$$

where the magnetic energy is the field energy given by

$$\mathcal{E}^{\mathrm{mag}}[\boldsymbol{\chi}, \tilde{\mathbf{M}}] = \int_{\mathbb{R}^3} \frac{\mu_0}{2}|\mathrm{grad}\zeta|^2 = \int_{\mathbb{R}^3} \frac{\mu_0}{2} J|\mathbf{F}^{-T}\mathrm{Grad}\zeta|^2.$$

By similar calculations as in Liu (2013, Eq. 2.14-2.17), we can show that within a constant independent of the state variables $(\boldsymbol{\chi}, \tilde{\mathbf{P}}, \tilde{\mathbf{M}})$, the magnetic energy can be rewritten as

$$\mathcal{E}^{\mathrm{mag}}[\boldsymbol{\chi}, \tilde{\mathbf{M}}] = \int_{\mathbb{R}^3} \frac{\mu_0}{2} J|\mathbf{F}^{-T}\mathrm{Grad}\zeta'|^2 - \mu_0 \int_{\Omega_R} \mathbf{H}^e \cdot (\tilde{\mathbf{M}} + \tilde{\mathbf{M}}^e), \tag{5.4}$$

where ζ' satisfies

$$\begin{cases} \mathrm{Div}[-J\mathbf{C}^{-1}\mathrm{Grad}\zeta' + \mathbf{F}^{-1}(\tilde{\mathbf{M}} + \tilde{\mathbf{M}}^e)] = 0 & \text{in } \mathbb{R}^3, \\ -\mathrm{Grad}\zeta' \to 0 & \text{as } |\mathbf{x}| \to +\infty. \end{cases} \tag{5.5}$$

In addition to variations of polarization and deformation, possible independent variations of the state of the body include variation of magnetization:

$$\tilde{\mathbf{M}} \to \tilde{\mathbf{M}}_\delta = \tilde{\mathbf{M}} + \delta\tilde{\mathbf{M}}_1.$$

By similar calculations as in § 3.2, we find the Euler-Lagrange equations and boundary conditions for a minimizer of the total free energy:

$$\begin{cases} \mathbf{F}^{-T}\mathrm{Grad}\xi + D_{\tilde{\mathbf{P}}}\Psi - \mathrm{Div}D_{\mathbf{\Pi}}\Psi = 0 & \text{in } \Omega_R, \\ \mu_0\mathbf{F}^{-T}\mathrm{Grad}\zeta + D_{\tilde{\mathbf{M}}}\Psi - \mathrm{Div}D_{\mathbf{\Lambda}}\Psi = 0 & \text{in } \Omega_R, \\ \mathrm{Div}\tilde{\mathbf{D}} = \tilde{\rho}^e, \quad \tilde{\mathbf{D}} = -\epsilon_0 J\mathbf{C}^{-1}\mathrm{Grad}\xi + \mathbf{F}^{-1}(\tilde{\mathbf{P}} + \tilde{\mathbf{P}}^e) & \text{in } V_R, \\ \mathrm{Div}[-J\mathbf{C}^{-1}\mathrm{Grad}\zeta + \mathbf{F}^{-1}(\tilde{\mathbf{M}} + \tilde{\mathbf{M}}^e)] = 0 & \text{in } \mathbb{R}^3, \\ \mathrm{Div}\mathrm{Div}D_{\mathbf{G}}\Psi - \mathrm{Div}D_{\mathbf{F}}\Psi - \mathrm{Div}\tilde{\mathbf{\Sigma}}_{\mathrm{MW}} - \tilde{\mathbf{f}}^e = 0 & \text{in } \Omega_R, \end{cases} \tag{5.6}$$

with the boundary conditions (3.3), $(3.4)_1$, $(5.1)_2$, and

$$\begin{cases} (D_{\mathbf{\Pi}}\Psi)\mathbf{N} = 0, \quad (D_{\mathbf{\Lambda}}\Psi)\mathbf{N} = 0 & \text{on } \partial\Omega_R, \\ (D_{\mathbf{F}}\Psi)\mathbf{N} - (\mathrm{Div}D_{\mathbf{G}}\Psi)\mathbf{N} - [\![\tilde{\mathbf{\Sigma}}_{\mathrm{MW}}]\!]\mathbf{N} - \boldsymbol{\tau} - \tilde{\mathbf{t}}^e = 0 & \text{on } S_N, \\ (D_{\mathbf{G}}\Psi)\mathbf{N} \otimes \mathbf{N} = 0 & \text{on } \partial\Omega_R. \end{cases} \tag{5.7}$$

Compared with (3.38)-(3.39), a key difference of the above boundary value problem for magneto-electro-elastic materials lies in that the *Piola-Maxwell* stress $\tilde{\mathbf{\Sigma}}_{\mathrm{MW}}$ shall now include the magnetic contribution and is given by

$$\tilde{\mathbf{\Sigma}}_{\mathrm{MW}} = \mathbf{E} \otimes \tilde{\mathbf{D}} + \mathbf{H} \otimes \tilde{\mathbf{B}} - J(\frac{\epsilon_0}{2}|\mathbf{E}|^2 + \frac{\mu_0}{2}|\mathbf{H}|^2)\mathbf{F}^{-T},$$

where

$$\mathbf{H} = -\mathrm{grad}\zeta \circ \boldsymbol{\chi}, \qquad \tilde{\mathbf{B}} = \mu_0[J\mathbf{F}^{-1}\mathbf{H} + \mathbf{F}^{-1}(\tilde{\mathbf{M}} + \tilde{\mathbf{M}}^e].$$

We remark that the above boundary value problem (5.6)-(5.7) for electric potential, magnetic potential, deformation, polarization and magnetization may be solved to determine any physical quantities of interest. (11 equations in (5.6) for 11 unknowns: ξ, ζ, $\boldsymbol{\chi}$, $\tilde{\mathbf{P}}, \tilde{\mathbf{M}}$.) The constitutive relations are prescribed by the internal energy density function Ψ. For many materials of interest, it may be sufficient to use some simple internal energy which will be discussed below. For brevity, below we only consider nongradient theories with the internal energy density independent of gradients of strain, polarization and magnetization.

5.2 Nonlinear magnetic elastomers

The magnetic field (**H**-field) of most applications reaches up to the order of $10^6 A/m$, i.e., the magnetic flux (**B**-field) is at the order of 1Tesla. Therefore, the Maxwell stress contributed by magnetic fields can reach roughly the order of 10^6Pa (1% strain for materials of Young's modulus of 0.1Gpa), which is as large as the Maxwell stress contributed by electric fields (cf., § 4.1). Moreover,

since there exists no magnetic monopole, magnetization in materials is more stable than polarization and may be more favorable for some applications. From the theoretical viewpoint the model for magnetic elastomers is exactly parallel to that for dielectric elastomer discussed in § 4.1. To establish the boundary value problem for magnetic elastomers, we start by postulating the internal energy density function depends only on the deformation gradient $\mathbf{F}$ and magnetization $\tilde{\mathbf{M}}$ and is given by

$$\Psi = \Psi(\mathbf{F}, \tilde{\mathbf{M}}) = W_{\text{elast}}(\mathbf{U}) + \frac{\mu_0}{2J}(\mathbf{R}^T\tilde{\mathbf{M}}) \cdot \mathbf{A}^{\text{mag}}(\mathbf{R}^T\tilde{\mathbf{M}}),$$

where we have assumed that the magnetic permeability tensor $\boldsymbol{\mu} \in \mathbb{R}^{3\times 3}_{\text{sym}}$ is independent of deformation gradient and the tensor $\mathbf{A}^{\text{mag}} = (\boldsymbol{\mu}/\mu_0 - \mathbf{I})^{-1}$. Upon neglecting all electric effects, then in the *current configuration* equations (5.6) can be rewritten as (cf., (4.5))

$$\begin{cases} \text{div}\mathbf{b} = 0, & \mathbf{b} = \mu_0[-(\mathbf{I} + \mathbf{R}(\mathbf{A}^{\text{mag}})^{-1}\mathbf{R}^T\chi_\Omega)\text{grad}\zeta + \mathbf{m} + \mathbf{m}^e] \quad \text{in } \mathbb{R}^3, \\ -\text{div}(\boldsymbol{\sigma}_{\text{mech}} + \boldsymbol{\sigma}'_{\text{MW}}) = \mathbf{f}^e, & \boldsymbol{\sigma}_{\text{mech}} = [\frac{1}{J}D_{\mathbf{F}}W_{\text{elast}}(\mathbf{U})\mathbf{F}^T] \circ \boldsymbol{\pi} \quad \text{in } \Omega, \end{cases} \tag{5.8}$$

where the *modified Maxwell stress* in current configuration is given by

$$\boldsymbol{\sigma}'_{\text{MW}} = \mathbf{h} \otimes \mathbf{b} - \frac{\mathbf{b}\cdot\mathbf{h}}{2}\mathbf{I}, \qquad \mathbf{h} = -\text{grad}\zeta. \tag{5.9}$$

For small strains, upon linearization the boundary value problem can be formulated as (cf., (4.7))

$$\begin{cases} \text{div}[-(\mathbf{I} + (\mathbf{A}^{\text{mag}})^{-1}\chi_\Omega)\nabla\zeta + \mathbf{M}^e] = 0 & \text{in } \mathbb{R}^3, \\ -\text{div}(\mathbf{C}\nabla\mathbf{u} + \boldsymbol{\sigma}'_{\text{MW}}) = \mathbf{f}^e & \text{in } \Omega, \end{cases} \tag{5.10}$$

with mechanical boundary conditions (4.9) and magnetic boundary condition $(5.1)_2$.

5.3 Magneto-electric materials

For the potential applications in wireless energy transfer, spintronics, and multiple-sate memory bits among others (Velev et al., 2011; Scott, 2007; Pyatakov and Zvezdin 2012), the magnetoelectric effects has recently attracted a lot of interest. For direct coupling between magnetization and polarization, we may model it by postulating the internal energy density function is given by

$$\Psi(\mathbf{F}, \tilde{\mathbf{P}}, \tilde{\mathbf{M}}) = W_{\text{elast}}(\mathbf{U}) + \mathcal{C}_{\text{me}}(\tilde{\mathbf{P}}, \tilde{\mathbf{M}}) + \frac{1}{2J}(\mathbf{R}^T\tilde{\mathbf{P}}) \cdot \mathbf{A}(\mathbf{R}^T\tilde{\mathbf{P}}) + \frac{\mu_0}{2J}(\mathbf{R}^T\tilde{\mathbf{M}}) \cdot \mathbf{A}^{\text{mag}}(\mathbf{R}^T\tilde{\mathbf{M}}),$$

where $\mathcal{C}_{\text{me}}(\tilde{\mathbf{P}}, \tilde{\mathbf{M}})$ reflects the coupling between polarization and magnetization. For small strains, upon expansion and truncation we can rewrite the internal energy function as (recall that $\mathbf{S} = \mathbf{F} - \mathbf{I}$):

$$\Psi(\mathbf{F}, \mathbf{P}, \mathbf{M}) = \frac{1}{2}\mathbf{S} \cdot \mathbf{C}\mathbf{S} + \mathbf{P} \cdot \mathbf{B}^{\text{me}}\mathbf{M} + \frac{1}{2}\mathbf{P} \cdot \mathbf{A}\mathbf{P} + \frac{\mu_0}{2}\mathbf{M} \cdot \mathbf{A}^{\text{mag}}\mathbf{M}, \tag{5.11}$$

where $\mathbf{B}^{\text{me}} \in \mathbb{R}^{3\times 3}_{\text{sym}}$ is the coupling tensor between $\mathbf{P}$ and $\mathbf{M}$. If the material is isotropic, by (3.7) we infer that the tensor $\mathbf{B}^{\text{me}}$ can be written as $\mathbf{B}^{\text{me}} = b_{\text{me}}\mathbf{I}$. At the absence of external polarization, magnetization, traction, body force, electric and magnetic fields, it is natural to assume that the trivial state $(\mathbf{F}, \mathbf{P}, \mathbf{M}) = (\mathbf{I}, 0, 0)$ is the equilibrium state such that the total free energy is minimized. Therefore, the constant b_{me} shall satisfy that (recall that $\mathbf{A} = \frac{1}{\epsilon_0(\hat{\epsilon}_r - 1)}\mathbf{I}$ and $\mathbf{A}^{\text{mag}} = \frac{1}{\hat{\mu}_r - 1}\mathbf{I}$)

$$b^2_{\text{me}} < \frac{\mu_0}{\epsilon_0(\hat{\epsilon}_r - 1)(\hat{\mu}_r - 1)},$$

where $\hat{\epsilon}_r, \hat{\mu}_r$ are the relative electric permittivity and magnetic permeability of the material, respectively. The above upper bound on the direct magnetization and polarization coupling coefficient is equivalent to the bound derived by Brown *et al.* (1968).[5] Finally, by (5.6), (5.7) and (5.11), the linearized boundary value problem for magneto-electric materials can be written as

$$\begin{cases} \nabla\xi + \mathbf{A}\mathbf{P} + \mathbf{B}^{\mathrm{me}}\mathbf{M} = 0, & \text{in } \Omega, \\ \mu_0\nabla\zeta + \mathbf{B}^{\mathrm{me}}\mathbf{P} + \mu_0\mathbf{A}^{\mathrm{mag}}\mathbf{M} = 0 & \text{in } \Omega, \\ \mathrm{div}[-\epsilon_0\nabla\xi + \mathbf{P} + \mathbf{P}^e] = \rho^e & \text{in } V, \\ \mathrm{div}[-\nabla\zeta + \mathbf{M} + \mathbf{M}^e] = 0 & \text{in } \mathbb{R}^3, \\ -\mathrm{div}(\mathbf{C}\nabla\mathbf{u} + \boldsymbol{\sigma}'_{\mathrm{MW}}) = \mathbf{f}^e & \text{in } \Omega, \end{cases} \tag{5.12}$$

with electrical boundary conditions (4.8), mechanical boundary conditions (4.9) and magnetic boundary condition $(5.1)_2$. In the above equations, we have neglected the differences between the reference and current configurations and the (*modified*) Maxwell stress is now given by (cf., (4.4) and (5.9))

$$\boldsymbol{\sigma}'_{\mathrm{MW}} = \mathbf{h}\otimes\mathbf{b} + \mathbf{e}\otimes\mathbf{d} - \frac{\mathbf{b}\cdot\mathbf{h} + \mathbf{d}\cdot\mathbf{e}}{2}\mathbf{I}. \tag{5.13}$$

5.4 Piezo-magneto-electric materials

By Piezo-magneto-electric effects, we refer to materials that have couplings between strain and polarization as well as magnetization. For small strain, upon expansion and truncation we can write the internal energy density as (recall that $\mathbf{S} = \mathbf{F} - \mathbf{I}$)

$$\Psi(\mathbf{F}, \mathbf{P}, \mathbf{M}) = \frac{1}{2}\mathbf{S}\cdot\mathbf{C}\mathbf{S} + \mathbf{P}\cdot\mathbf{B}\mathbf{S} + \mathbf{M}\cdot\mathbf{B}^{\mathrm{pzm}}\mathbf{S} + \mathbf{P}\cdot\mathbf{B}^{\mathrm{me}}\mathbf{M} + \frac{1}{2}\mathbf{P}\cdot\mathbf{A}\mathbf{P} + \frac{\mu_0}{2}\mathbf{M}\cdot\mathbf{A}^{\mathrm{mag}}\mathbf{M}, \tag{5.14}$$

Then by (5.6), (5.7) and (5.11), the linearized boundary value problem for piezo-magneto-electric materials can be written as

$$\begin{cases} \nabla\xi + \mathbf{B}\nabla\mathbf{u} + \mathbf{A}\mathbf{P} + \mathbf{B}^{\mathrm{me}}\mathbf{P} = 0, & \text{in } \Omega, \\ \mu_0\nabla\zeta + \mathbf{B}^{\mathrm{pzm}}\nabla\mathbf{u} + \mathbf{B}^{\mathrm{me}}\mathbf{P} + \mu_0\mathbf{A}^{\mathrm{mag}}\mathbf{M} = 0 & \text{in } \Omega, \\ \mathrm{div}[-\epsilon_0\nabla\xi + \mathbf{P} + \mathbf{P}^e] = \rho^e & \text{in } V, \\ \mathrm{div}[-\nabla\zeta + \mathbf{M} + \mathbf{M}^e] = 0 & \text{in } \mathbb{R}^3, \\ -\mathrm{div}[\mathbf{C}\nabla\mathbf{u} + \mathbf{B}^T\mathbf{P} + (\mathbf{B}^{\mathrm{pzm}})\mathbf{M} + \boldsymbol{\sigma}'_{\mathrm{MW}}] = \mathbf{f}^e & \text{in } \Omega, \end{cases} \tag{5.15}$$

with electrical boundary conditions (4.8), mechanical boundary conditions $(4.9)_1$, magnetic boundary condition $(5.1)_2$ and

$$[\mathbf{C}\nabla\mathbf{u} + \mathbf{B}^T\mathbf{P} + (\mathbf{B}^{\mathrm{pzm}})\mathbf{M}]\mathbf{n} - [\![\boldsymbol{\sigma}'_{\mathrm{MW}}]\!]\mathbf{n} - \mathbf{t}^e = 0 \quad \text{on } S_N. \tag{5.16}$$

Also, the (*modified*) Maxwell stress is given by (5.13).

6 Applications

6.1 Stretching of a soft ellipsoid in an external electric field

Consider a soft axis-symmetric ellipsoid Ω_R with semi-axis lengths a_0, b_0 and of relative permittivity $\hat{\epsilon}_r$ in a uniform external electric field as shown in Fig. 3. The ambient medium is assumed to be

[5] The coupling coefficient α defined in Brown *et al.* (1968) is given by $\alpha = \epsilon_0(\hat{\mu}_r - 1)(\hat{\epsilon}_r - 1)b_{\mathrm{me}}$ in SI units.

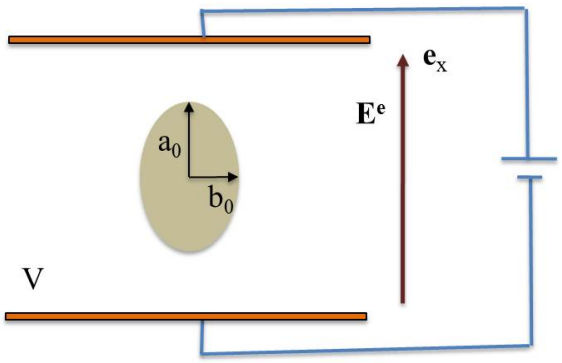


Figure 3: Deformation of a soft ellipsoid in an external electric field.

air or vacuum with permittivity ϵ_0; the body Ω_R consists of dielectric elastomer described by the Neo-Hookean hyperelastic model with $W_{\text{elast}}(\boldsymbol{\lambda})$ given by (μ - shear modulus, κ - bulk modulus)

$$W_{\text{elast}}(\mathbf{U}) = \frac{\mu}{2}\Big[J^{-2/3}(\lambda_1^2+\lambda_2^2+\lambda_3^2)-3\Big] + \frac{\kappa}{2}(J-1)^2, \tag{6.1}$$

where $(\lambda_1, \lambda_2, \lambda_3)$ are the eigenvalues of the strain tensor $\mathbf{U}$. We remark that the following solution procedure can be applied to more general nonlinear elastic models or models that include surface energy.

The application of an external electric field will deform the ellipsoid due to the Maxwell stress. The Maxwell stress of a homogeneous ellipsoid has been explicitly calculated in Toupin (1956, §12). From Toupin's expression (1956, Eq. 12.30) we see that the Maxwell stress can be regarded as a nonuniform outward normal traction and that the ellipsoid cannot remain as an ellipsoid after deformation. Nevertheless, by symmetry we see that the deformed shape shall be axis-symmetric with some aspect ratio a/b and volume $|\Omega|$. To calculate the aspect ratio and volume that are easy to measure in experiments, for simplicity we may *a priori assume* the deformation is homogeneous (i.e., Grad$\boldsymbol{\chi}$ is constant on Ω_R) and hence the deformed shape remains as an axis-symmetric ellipsoid with semi-axis lengths a, b. In other words, what we will predict is the "best-fitting" ellipsoid of the actual shape of the deformed body.

By (3.9), the total free energy of the body can be written as

$$F[a,b,\tilde{\mathbf{P}}] = \int_{\Omega_R}\Big[W_{\text{elast}}(\mathbf{U}) + \frac{|\tilde{\mathbf{P}}|^2}{2J(\epsilon-\epsilon_0)}\Big] + \frac{\epsilon_0}{2}\int_V |\text{grad}\xi|^2 + \int_{\partial V}\xi_b \mathbf{d}\cdot\mathbf{n}, \tag{6.2}$$

where in the current configuration the electric field $-\text{grad}\xi$ satisfies ($\mathbf{p} = \tilde{\mathbf{P}}/J$)

$$\begin{cases} \text{div}\mathbf{d} = 0, \qquad \mathbf{d} = -\epsilon_0 \text{grad}\xi + \mathbf{p}\chi_\Omega & \text{in } V, \\ \xi = \xi_b = -\mathbf{E}^e\cdot\mathbf{x} & \text{on } \partial V, \end{cases}$$

where $\mathbf{E}^e \in \mathbb{R}^3$ is an uniform external electric field and assumed to be along x-direction, and χ_Ω, equal to one on Ω and zero otherwise, is the characteristic function of domain Ω. By the calculations in Liu (2013, Eq. 2.14-2.17), within a constant independent of $a, b, \tilde{\mathbf{P}}$, the free energy can be rewritten as

$$F[a,b,\tilde{\mathbf{P}}] = \int_{\Omega_R}\Big[W_{\text{elast}}(\mathbf{U}) + \frac{|\tilde{\mathbf{P}}|^2}{2J(\epsilon-\epsilon_0)} - \mathbf{E}^e\cdot\tilde{\mathbf{P}}\Big] + \frac{\epsilon_0}{2}\int_V |\text{grad}\xi^{\text{self}}|^2, \tag{6.3}$$

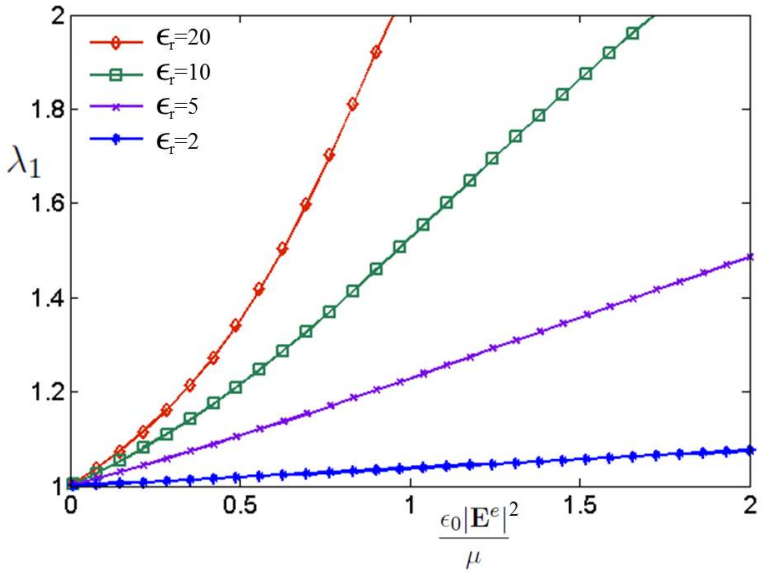


Figure 4: The stretching λ_1 in x-direction of a sphere in an external electric field.

where $-\text{grad}\xi^{\text{self}}$ is the "self-field" arising from the polarized body Ω:

$$\begin{cases} \text{div}(-\epsilon_0 \text{grad}\xi^{\text{self}} + \mathbf{p}\chi_\Omega) = 0 & \text{in } V, \\ \xi^{\text{self}} = 0 & \text{on } \partial V. \end{cases}$$

If the body Ω is much smaller than V and far away from the boundary ∂V, we can replace V by $\mathbb{R}^3$ and the solution of this problem for a uniform polarization $\mathbf{p} \in \mathbb{R}^3$ is given by

$$\text{grad}\xi^{\text{self}}(\mathbf{x}) = \frac{1}{\epsilon_0}\mathbf{Q}\mathbf{p} \qquad \forall\, \mathbf{x} \in \Omega,$$

where the symmetric positive matrix $\mathbf{Q}$, depending only on the aspect ratio, is called the *depolarization matrix* and given by

$$\mathbf{Q} = \int_{S^2} \frac{\hat{\mathbf{k}} \otimes \hat{\mathbf{k}} d\hat{\mathbf{k}}}{(a\hat{k}_1^2 + 2b\hat{k}_2^2)^{1/2}} = \text{diag}[I_1, I_2, I_3],$$
$$I_2 = I_3 = \frac{ab^2}{2}\int_0^{+\infty} \frac{du}{(b^2+u)^2\sqrt{(a^2+u)}}, \qquad I_1 = 1 - 2I_2.$$

In this case, by the divergence theorem we have

$$\frac{\epsilon_0}{2}\int_V |\text{grad}\xi^{\text{self}}|^2 = \frac{1}{2}\int_V \text{grad}\xi^{\text{self}} \cdot \mathbf{p}\chi_\Omega = |\Omega|\frac{1}{2\epsilon_0}\mathbf{p}\cdot\mathbf{Q}\mathbf{p}.$$

We now consider the problem of minimizing the free energy (6.3) over all possible $a, b, \tilde{\mathbf{P}}$. First, it can be shown that the minimizing polarization $\tilde{\mathbf{P}}$ has to be uniform on the body Ω_R, i.e., the polarization $\mathbf{p}$ has to be uniform on Ω (Desimone and James, 2002; Liu *et al.,* 2006). Therefore,

the free energy (6.3) can be rewritten as

$$\frac{F[a,b,\tilde{\mathbf{P}}]}{\pi a_0 b_0^2} = \frac{\mu}{2}\Big[J^{-2/3}((\frac{a}{a_0})^2 + 2(\frac{b}{b_0})^2 - 3)\Big] + \frac{\kappa}{2}(J-1)^2 + \frac{|\tilde{\mathbf{P}}|^2}{2J(\epsilon-\epsilon_0)} - \mathbf{E}^e\cdot\tilde{\mathbf{P}} + \frac{1}{2\epsilon_0 J}\tilde{\mathbf{P}}\cdot\mathbf{Q}\tilde{\mathbf{P}}.$$

The equilibrium or "best-fitting" ellipsoid and polarization are determined by

$$\frac{\partial F[a,b,\tilde{\mathbf{P}}]}{\partial a} = 0, \quad \frac{\partial F[a,b,\tilde{\mathbf{P}}]}{\partial b} = 0, \quad \frac{\partial F[a,b,\tilde{\mathbf{P}}]}{\partial \tilde{\mathbf{P}}} = 0. \tag{6.4}$$

The above set of algebraic equations can be explicitly solved; the general solutions and their physical implications will not be discussed here. Below we consider the special case of *incompressible* materials with $J \equiv 1$. Then by symmetry and the last of (6.4) we have

$$\lambda_2 = \frac{b}{b_0} = \lambda_1^{-1/2} = (\frac{a}{a_0})^{-1/2}, \qquad \tilde{\mathbf{P}} = (\frac{\mathbf{I}}{\epsilon-\epsilon_0} + \frac{\mathbf{Q}}{\epsilon_0})^{-1}\mathbf{E}^e = \frac{\hat{\epsilon}_r - 1}{1 + I_1(\hat{\epsilon}_r - 1)}\epsilon_0|\mathbf{E}^e|^2\mathbf{e}_x,$$

and hence

$$\frac{2F[a,b,\tilde{\mathbf{P}}]}{\pi\mu a_0 b_0^2} = \lambda_1^2 + \frac{2}{\lambda_1} - 3 - \frac{\hat{\epsilon}_r - 1}{1 + I_1(\hat{\epsilon}_r - 1)}\frac{\epsilon_0|\mathbf{E}^e|^2}{\mu}. \tag{6.5}$$

For given $\mathbf{E}^e$ and $\hat{\epsilon}_r$, the equilibrium stretching λ_1 can be easily calculated by minimizing the free energy (6.5) over λ_1. The results are shown in Fig. 4 for a spherical Ω_R. From Fig. 4 we observe that the Maxwell stress stretches the sphere and the stretching λ_1 depends linearly on $|\mathbf{E}^e|^2$ for small $|\mathbf{E}^e|$ whereas significant nonlinearity emerges when $\hat{\epsilon}_r$ is large or $|\mathbf{E}^e|$ is large.

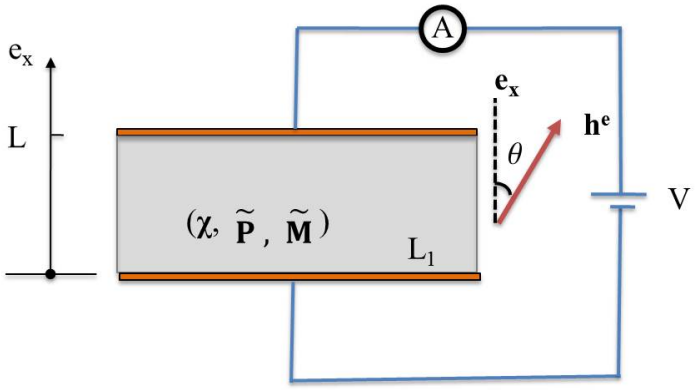


Figure 5: Magnetoelectric effects arising from the Maxwell stress and nonlinear elasticity of finite deformation.

6.2 Magnetoelectric effects of magneto-electric elastomers

A generic boundary value problem concerning magneto-electro-elastic bodies are not amenable to explicit solution. In analogy with the classic theories for lower dimensional bodies in elasticity, we now develop a one-dimensional theory for magneto-electro-elastic films as shown in Fig. 5. Based

on the geometric and loading features, we *a priori assume* that the deformation is of the following form:

$$x = X + u(X), \quad y = Y(1 + \alpha(X)), \quad z = Z(1 + \beta(X)), \tag{6.6}$$

where $\mathbf{X} = (X, Y, Z)$ are the Lagrange coordinates, $\mathbf{x} = (x, y, z)$ are the Euler coordinates and $u, \alpha, \beta : (0, L) \to \mathbb{R}$ are scalar functions describing the deformed state of the body. Further, we assume that there are external charges $\tilde{\rho}^e(X)$, external electric dipoles $\tilde{P}^e(X)$ (along X-direction), and external magnetization $\tilde{\mathbf{M}}^e(X)$ distributed on the film. The conducting electrodes maintain constant electrostatic potentials on both the top and bottom faces but are assumed to be mechanically trivial. In addition, there is a constant external magnetic field $\mathbf{h}^e \in \mathbb{R}^3$ applied on the film. We are interested in the magneto-electro-elastic state of the film.

Compared with the analogous theory in elasticity, we point out that the body is a thin film (the thickness L is much smaller than the width L_1 in the other two directions) instead of being a slender body (St. Venant problems) since it is desirable to neglect the electric field in the ambient medium (i.e., the fringe fields). Further, it is reasonable to assume that the intrinsic polarization and magnetization are independent of in-plane positions Y, Z:

$$\tilde{\mathbf{P}} = \tilde{P}(X)\mathbf{e}_X, \qquad \tilde{\mathbf{M}} = \tilde{\mathbf{M}}(X). \tag{6.7}$$

It is clear that the response of the film depends on the form of internal energy density function Ψ, i.e., the constitutive relations. The most general nonlinear, strain/polarization/magnetization gradient theory will not be explored here, though the problem is essentially one-dimensional for the "kinematic" assumptions (6.6) and (6.7). Below we present a solution concerning nonlinear magneto-electric elastomers with the internal energy density function given by

$$\Psi(\mathbf{F}, \mathbf{P}, \mathbf{M}) = W_{\text{elast}}(\mathbf{U}) + \frac{1}{2\epsilon_0(\hat{\epsilon}_r - 1)J}|\tilde{\mathbf{P}}|^2 + \frac{\mu_0}{2(\hat{\mu}_r - 1)J}|\tilde{\mathbf{M}}|^2, \tag{6.8}$$

where $\hat{\mu}_r$ (resp. $\hat{\epsilon}_r$) is the relative magnetic permeability (resp. electric permittivity) of the film. We remark that there is no direct coupling between magnetization and polarization, i.e., the direct magnetoelectric coupling tensor $\mathbf{B}^{\text{me}} = 0$ in (5.11). Nevertheless, *effective* magnetoelectric coupling arises from the Maxwell stress and geometric nonlinearity, as one will see shortly.

Without loss of generality assume that $x(0) = 0, x(L) = l$. For simplicity assume that the external charges, polarization and magnetization $\tilde{\rho}^e = 0$, $\tilde{P}^e = 0$ and $\tilde{\mathbf{M}}^e = 0$. It will be convenient to write the electrostatic and magnetostatic problem in the *current configuration* whereas the mechanical balance equations in the *reference configuration*. By (1.1), we infer the boundary value problems for electric and magnetic fields are as follows:

$$\begin{cases} (-\epsilon_0\xi_{,x} + p)_{,x} = 0 \quad \text{on } (0, l), \\ p = -(\epsilon - \epsilon_0)\xi_{,x} \quad \text{on } (0, l), \\ \xi(0) = 0, \quad \xi(l) = V \end{cases} \qquad \begin{cases} \operatorname{div}[-\operatorname{grad}\zeta + \mathbf{m}\chi_\Omega] = 0 & \text{on } \mathbb{R}^3, \\ \mathbf{m} = -(\hat{\mu}_r - 1)\operatorname{grad}\zeta & \text{on } \Omega, \\ -\operatorname{grad}\zeta \to \mathbf{h}^e & \text{as } |\mathbf{x}| \to \infty. \end{cases} \tag{6.9}$$

For ease of exposition, we assume that the material is elastically isotropic–a reasonable assumption for soft polymers and that the elastic properties are described by the *compressible* Neo-Hookean hyperelastic model with $W_{\text{elast}}(\mathbf{U})$ given by (6.1). By symmetry we can conclude that $\lambda_2 = \lambda_3 = (J/\lambda_1)^{1/2}$. For simplicity we assume that λ_1, λ_2 are independent of X. Then we immediately find that the electric field $-\operatorname{grad}\xi$ and polarization for $x \in (0, l)$ are given by

$$-\operatorname{grad}\xi = -\frac{V}{l}\mathbf{e}_x, \qquad \mathbf{p} = -\epsilon_0(\hat{\epsilon}_r - 1)\frac{V}{l}\mathbf{e}_x. \tag{6.10}$$

Also, if the external magnetic field is out-of-plane: $\mathbf{h}^e = h_x^e \mathbf{e}_x$, the self magnetic field $-\text{grad}\zeta^{\text{self}} := -\text{grad}\zeta - \mathbf{h}^e$ and magnetization for $x \in (0, l)$ are given by

$$-\text{grad}\zeta^{\text{self}} = -\frac{\mu_r - 1}{\mu_r} h_x^e \mathbf{e}_x, \qquad \mathbf{m} = \frac{\mu_r - 1}{\mu_r} h_x^e \mathbf{e}_x. \tag{6.11}$$

whereas if the external magnetic field is in-plane: $\mathbf{h}^e = h_y^e \mathbf{e}_y$, the self magnetic field $-\text{grad}\zeta^{\text{self}}$ and magnetization is given by

$$-\text{grad}\zeta^{\text{self}} = 0, \qquad \mathbf{m} = (\mu_r - 1) h_y^e \mathbf{e}_y. \tag{6.12}$$

Note that by (3.1),

$$\tilde{P} = pJ, \qquad \tilde{\mathbf{M}} = \mathbf{m}J. \tag{6.13}$$

Therefore, by (5.3), (5.4), (3.10), (6.8) and (6.10) we find the total free energy as

$$\frac{F(\lambda_1, J)}{\mu L L_1^2} = \hat{W}_{\text{elast}}(\lambda_1, J) - \frac{J}{2\lambda_1^2} \hat{f}^{\text{elect}} - \frac{J}{2} \hat{f}^{\text{mgf}} \tag{6.14}$$

where the following dimensionless quantities are introduced for clarity:

$$\hat{W}_{\text{elast}}(\lambda_1, J) = \frac{1}{2}\Big[J^{-2/3}(\lambda_1^2 + \frac{2J}{\lambda_1}) - 3\Big] + \frac{\hat{\kappa}}{2}(J-1)^2,$$
$$\hat{\kappa} = \frac{\kappa}{\mu}, \qquad \hat{f}^{\text{elect}} = \frac{\epsilon_r \epsilon_0 V^2}{\mu L^2}, \qquad \hat{f}^{\text{mgf}} = \frac{\mu_0}{\mu}\Big[\frac{\mu_r - 1}{\mu_r}(h_x^e)^2 + (\mu_r - 1)(h_y^e)^2\Big].$$

Upon minimizing $F(\lambda_1, J)$ over (λ_1, J) we obtain the necessary conditions $\frac{\partial F}{\partial \lambda_1} = \frac{\partial F}{\partial J} = 0$, i.e.,

$$\begin{cases} J^{-2/3}\lambda_1 - J^{1/3}\lambda_1^{-2} + J\lambda_1^{-3}\hat{f}^{\text{elect}} = 0, \\ -\frac{1}{3}(J^{-5/3}\lambda_1^2 - J^{-2/3}\lambda_1^{-1}) + \hat{\kappa}(J-1) - \frac{1}{2}\lambda_1^{-2}\hat{f}^{\text{elect}} - \frac{1}{2}\hat{f}^{\text{mgf}} = 0. \end{cases} \tag{6.15}$$

From (6.15) we observe that the stretching λ_1 and hence the polarization along $\mathbf{e}_x$-direction $p = -\epsilon_0(\hat{\epsilon}_r - 1)\xi_{,x} = -\epsilon_0(\hat{\epsilon}_r - 1)\frac{V}{L\lambda_1}$ depend on the magnitude and direction of the external magnetic field $\mathbf{h}^e$. From this viewpoint, the magnetoelectric coupling is *effectively* created from the Maxwell stress and geometric nonlinearity. The change of polarization at the presence of external magnetic field is appropriate for evaluating the strength of this magnetoelectric coupling which is given by

$$\triangle p = -\epsilon_0(\hat{\epsilon}_r - 1)\frac{V}{L}(\lambda_1^{-1}\Big|_{\mathbf{h}^e \neq 0} - \lambda_1^{-1}\Big|_{\mathbf{h}^e = 0}). \tag{6.16}$$

We can numerically solve (6.15) and determine the polarization $\triangle p$ induced by the external magnetic field; the dependence of $\triangle p$ on $|\mathbf{h}^e|^2$ ($\mathbf{h}^e$ is parallel to y-axis) is shown in Fig. 6 for a fixed nominal electric field $\tilde{E}_0 := V/L = 10^7\ V/m$ ($\epsilon_0|\tilde{E}_0|^2 = 885.4Pa$) . The material properties are chosen as $\hat{\epsilon}_r = 20$, $\hat{\mu}_r = 5$, $\hat{\kappa} = 10$, $\mu = 1,\ 0.9,\ 0.8,\ 0.7,\ 0.6,\ 0.5$ MPa. At the magnetic field of $|\mathbf{h}^e| = 10^6\ A/m$ (i.e., the $\mathbf{B}$-field is $1.26T$ in vacuum), the induced polarization is at the order of $100\mu C/m^2$ that may be compared with the magnetoelectricity of crystal $TbMnO_3$ that has induced polarization at the order of $200 - 600\ \mu C/m^2$ in a magnetic flux of $2 - 9$T at a temperature below 30K (Kimura *et al.*, 2003). From Fig. 6 we also observe that the induced polarization by the external magnetic field is roughly proportional to $|\mathbf{h}^e|^2$ and the slope of the curves increases as the shear modulus μ or bulk modulus κ decrease. To calculate the slope explicitly, we assume small

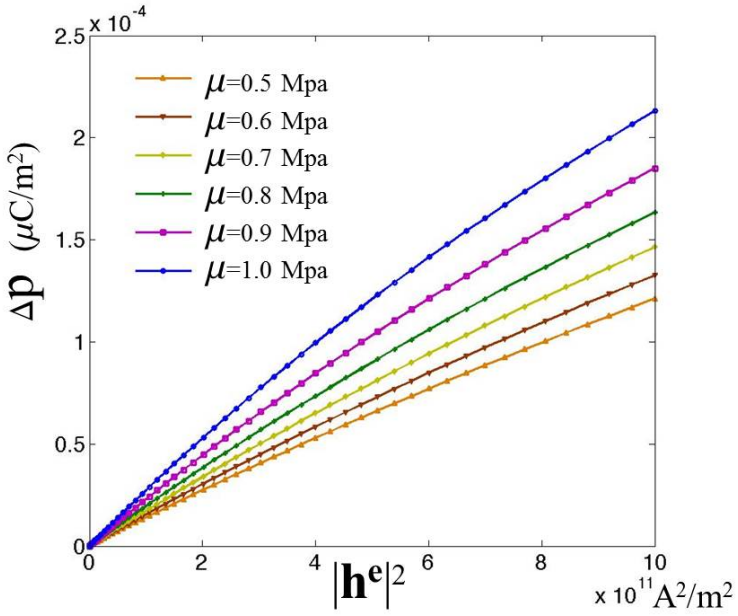


Figure 6: Magnetoeletric effects arising from the Maxwell stress and geometric nonlinearity. The curves show the dependence of $\triangle p$ (change of polarization) on $|\mathbf{h}^e|^2$ ($\mathbf{h}^e$-external magnetic field, parallel to y-axis) for a fixed nominal electric field $\tilde{E}_0 := V/L = 10^7\ V/m$. The material properties are chosen as $\hat{\epsilon}_r = 20$, $\hat{\mu}_r = 5$, $\hat{\kappa} = 10$, $\mu = 1$, 0.9, 0.8, 0.7, 0.6, 0.5 MPa.

strain $\triangle\lambda := \lambda_1 - 1 \sim \triangle J := J - 1 \sim \eta << 1$, and then expand and truncate the free energy (6.14) up to the order of η^2 as

$$\frac{F(\triangle\lambda, \triangle J)}{\mu L L_1^2} = \frac{1}{2}\Big[3\triangle\lambda^2 - 2\triangle J\triangle\lambda + \frac{\triangle J^2}{3}\Big] + \frac{\hat{\kappa}}{2}(\triangle J)^2 \\ -(\triangle J - 2\triangle\lambda - 2\triangle\lambda\triangle J + 3\triangle\lambda^2)\frac{1}{2}\hat{f}^{\text{elect}} - \triangle J\frac{1}{2}\hat{f}^{\text{mgf}} + o(\eta^2). \tag{6.17}$$

where an immaterial (λ_1, J)-independent constant has been neglected. Then the necessary conditions $\partial F/\partial\triangle\lambda = \partial F/\partial\triangle J = 0$ for the equilibrium $(\triangle\lambda, \triangle J)$ imply

$$\begin{cases} 3(1-\hat{f}^{\text{elect}})\triangle\lambda - (1-\hat{f}^{\text{elect}})\triangle J + \hat{f}^{\text{elect}} = 0, \\ -(1-\hat{f}^{\text{elect}})\triangle\lambda + (\frac{1}{3} + \hat{\kappa})\triangle J - \frac{1}{2}\hat{f}^{\text{elect}} - \frac{1}{2}\hat{f}^{\text{mgf}} = 0. \end{cases} \tag{6.18}$$

Solving the above equations we obtain that $\triangle J = \frac{\frac{3}{2}\hat{f}^{\text{mgf}} + \frac{1}{2}\hat{f}^{\text{elect}}}{3\hat{\kappa} + \hat{f}^{\text{elect}}}$ and

$$\triangle\lambda = \frac{\hat{f}^{\text{elect}}(\frac{1}{6} - \hat{\kappa} - \frac{1}{2}\hat{f}^{\text{elect}}) + \frac{1}{2}\hat{f}^{\text{mgf}}(1-\hat{f}^{\text{elect}})}{(1-\hat{f}^{\text{elect}})(3\hat{\kappa} + \hat{f}^{\text{elect}})}.$$

Therefore, the induced polarization defined by (6.16) is given by

$$\triangle p = \frac{\mu\hat{f}^{\text{mgf}}}{2(\frac{3\kappa}{\epsilon_0(\hat{\epsilon}_r - 1)\tilde{E}_0} + \frac{\hat{\epsilon}_r\tilde{E}_0}{(\hat{\epsilon}_r - 1)})}. \tag{6.19}$$

From the above expression we see that there exists an optimal applied nominal field such that the proportional constant between $\triangle p$ and $\mu \hat{f}^{\text{mgf}}$ is maximized:

$$(\triangle p)^{\max} = \frac{(\hat{\epsilon}_r - 1)\sqrt{\epsilon_0}}{4\sqrt{3\hat{\epsilon}_r \kappa}} \mu \hat{f}^{\text{mgf}}, \qquad \tilde{E}_0^{opt} = \sqrt{\frac{3\kappa}{\epsilon_0 \hat{\epsilon}_r}}.$$

As noticed earlier (Xu *et al.*, 2010), soft elastomers may lose mechanical stability if the applied electric field exceeds some critical value. Similar phenomenon occurs for elastomer and the critical electric field and external magnetization can be calculated by solving the algebraic equation $\det(D^2 F(\lambda_1, J)) = 0$, where D^2 represents the Hessian of $F(\lambda_1, J)$ in the regime of finite strain. In the regime of small strain, by (6.17) we immediately see the stability condition is given by $\hat{f}^{\text{elect}} < 1$. This stability condition shall be satisfied in practical applications.

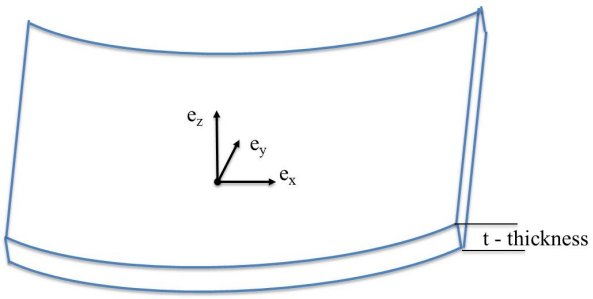


Figure 7: A flexoelectric plate under bending.

6.3 Bending of flexoelectric films

Flexoelectricity, unlike piezoelectricity, is not limited to crystalline solids and exist in all materials including soft polymers and biological membranes. As a coupling between strain gradient and polarization, a natural manifestation of flexoelectricity lie in the bending of a thin plate or membrane. We have proposed a theory for flexoelectric membranes in Mohammadi et al. (2013) and Liu and Sharma (2013). Below we formally derive this theory from the three dimensional theory of flexoelectricity described in § 6.3.

Consider a thin-plate $\Omega = U \times (-\frac{h}{2}, \frac{h}{2})$ as shown in Fig. 7, where $U \subset \mathbb{R}^2$ is a regular domain occupied by the mid-plane of the plate in the reference configuration. For small strain, the internal energy density function describing flexoelectricity is given by (4.28), i.e.,

$$U[\mathbf{u}, \mathbf{P}] = \int_\Omega \Big[\frac{1}{2} \nabla \mathbf{u} \cdot \mathbf{C} \nabla \mathbf{u} + \frac{g}{2} |\Delta \mathbf{u}|^2 + f \mathbf{P} \cdot \Delta \mathbf{u} + \frac{1}{2(\epsilon - \epsilon_0)} |\mathbf{P}|^2 \Big]. \tag{6.20}$$

Since $h << 1$, we anticipate the plate is prone to bend. As in the classic Kirchhoff-Love plate theory, kinematically we postulate that (H1) the in-plane displacements depend linearly on x_3 and the out-of-plane displacement is independent of x_3, (H2) the in-plane displacements of the mid-plane vanish, i.e., the mid-plane is not stretched, (H3) the mid-plane undergoes an infinitesimal rigid rotation, and (H4) the overall plate is nearly in the state of plane stress, i.e., $\sigma_{13} \approx \sigma_{23} \approx \sigma_{33} \approx 0$. By (H1) and (H2) we have

$$u_1 = x_3 \beta_1(x_1, x_2), \qquad u_2 = x_3 \beta_2(x_1, x_2), \qquad u_3 = w(x_1, x_2), \tag{6.21}$$

and hence on the mid-plane $x_3 = 0$,

$$\nabla \mathbf{u} = \begin{bmatrix} 0 & 0 & \beta_1 \\ 0 & 0 & \beta_2 \\ w_{,1} & w_{,2} & 0 \end{bmatrix},$$

By (H3) we have

$$\beta_1 = -w_{,1}, \qquad \beta_2 = -w_{,2}.$$

Therefore, the in-plane strain is completely described by the out-of-plane displacement w on the mid-plane and given by

$$(\nabla \mathbf{u})_{\text{in-plane}} = -\begin{bmatrix} x_3 w_{,11} & x_3 w_{,12} \\ x_3 w_{,21} & x_3 w_{,22} \end{bmatrix} = -x_3 \nabla\nabla w \qquad \forall\, (x_1, x_2, x_3) \in \Omega \times (-\frac{h}{2}, \frac{h}{2}).$$

By (H4), the overall plate is in the state of plane stress and hence

$$(\boldsymbol{\sigma})_{pi} = (\mathbf{L})_{piqj} (\nabla \mathbf{u})_{qj} \qquad (p, i, q, j = 1, 2), \tag{6.22}$$

where $\mathbf{L} : \mathbb{R}^{2\times 2}_{\text{sym}} \to \mathbb{R}^{2\times 2}_{\text{sym}}$ is the stiffness tensor for plane stress. Therefore, the elastic energy stored in the plate is given by $(p, i, q, j = 1, 2)$

$$\frac{1}{2} \int_U \int_{-\frac{h}{2}}^{\frac{h}{2}} (\boldsymbol{\sigma})_{pi} (\nabla \mathbf{u})_{pi} = \frac{1}{2} \int_U (\mathbf{L}')_{piqj} w_{,pi} w_{,qj}, \qquad (\mathbf{L}')_{piqj} = \frac{h^3}{12} (\mathbf{L})_{piqj}. \tag{6.23}$$

In addition, by (6.21) we have

$$\Delta \mathbf{u} \approx [x_3 \Delta' \beta_1, x_3 \Delta' \beta_2, \Delta' w] = [-x_3 (\Delta' w)_{,1}\,,\ -x_3 (\Delta' w)_{,2}\,,\ \Delta' w],$$

where $\Delta' = \frac{\partial^2}{\partial x_1^2} + \frac{\partial^2}{\partial x_2^2}$ denotes the in-plane Laplacian. Further, since $h << 1$, we may introduce a new kinematic assumption concerning the polarization $\mathbf{P}$: (H5) the polarization depends only on (x_1, x_2), i.e., $\mathbf{P} = \mathbf{P}(x_1, x_2)$. Therefore, to the leading-order the rest of terms in (6.20) are given by

$$\begin{aligned} \int_\Omega \frac{g}{2} |\Delta \mathbf{u}|^2 &= \int_{-\frac{h}{2}}^{\frac{h}{2}} \int_U \frac{g}{2} |\Delta \mathbf{u}|^2 = \int_U \frac{g'}{2} (\Delta' w)^2 + O(h^3), \\ \int_\Omega f \mathbf{P} \cdot \Delta \mathbf{u} &= \int_{-\frac{h}{2}}^{\frac{h}{2}} \int_U f \mathbf{P} \cdot \Delta \mathbf{u} = \int_U f' P_z \Delta' w + O(h^2), \\ \int_\Omega \frac{a}{2} |\mathbf{P}|^2 &= \int_{-\frac{h}{2}}^{\frac{h}{2}} \int_U \frac{a}{2} |\mathbf{P}|^2 = \int_U \frac{a'}{2} |\mathbf{P}|^2 + O(h^2). \end{aligned} \tag{6.24}$$

where $g' = hg$, $f' = hf$, and $a' = ha$. Keeping only the leading order terms in (6.20), by (6.23) and (6.24) we can rewrite the internal energy of the plate as

$$U[w, \mathbf{P}] \approx \int_U \Big[\frac{g'}{2} (\Delta' w)^2 + f' P_z \Delta' w + \frac{1}{2} \nabla\nabla w \cdot \mathbf{L}' \nabla\nabla w + \frac{a'}{2} |\mathbf{P}|^2 \Big],$$

which is precisely the internal energy used in Mohammadi *et al.* (2013) and Liu and Sharma (2013).

7 Summary and discussion

We have developed systematically an energy formulation for magneto-electro-elastic bodies based on the principle of minimum free energy. In this framework, the free energy, state variables and possible variations of state variables, and the Maxwell equations governing electrostatic and magnetostatic fields are regarded as primitive notions or constraints while the concepts of stresses are regarded as derived quantities conjugate to deformation gradient. In this way, we untangle some of the issues concerning the Maxwell stress. Also, the theoretical framework is particularly convenient for complex materials with significant gradient effects, analysis of stability and fully coupled magneto-electro-elastic materials.

As emphasized in Liu (2013), a set of field equations can enjoy infinitely many variational formulations; neither the independent variables nor the functional has to be the same (Dorfmann and Ogden, 2005; Bustamante *et al.*, 2009a). In the Appendix C, we show how the nongradient theory can be reformulated using the electric displacement or electric field (instead of polarization) as the independent variables. Upon specifying the physical behaviors of the materials, we eventually solve for local fields by the *same* boundary value problems that consist of the Maxwell equations (1.1a)-(1.1b) and mechanical balance laws (1.1c). Nevertheless, different formulations may have their respective pros and cons. It is undoubtedly useful to have a clear distinction and exploration of the relationship between different formulations.

8 Appendix

A Higher order variational calculations

The variation of energy associated with strain gradient is given by (3.33) or (3.35). Below we present the detailed derivation of (3.35). Recall that $\tilde{\Lambda}_{pij}$ (resp. u_p) denote the components of $D_{\mathbf{G}}\Psi$ (resp. $\boldsymbol{\chi}_1$). In index notation, integrating by parts we have

$$\int_{\Omega_R} \tilde{\Lambda}_{pij} u_{p,ij} = \int_{\partial\Omega_R} (\tilde{\Lambda}_{pij} N_j) u_{p,i} - \int_{\partial\Omega_R} (\tilde{\Lambda}_{pij,j} N_i) u_p + \int_{\Omega_R} \tilde{\Lambda}_{pij,ij} u_p. \tag{A.1}$$

The tangential components of $u_{p,i}$ and u_p, however, are not independent of each other on the surface $\partial\Omega_R$. We therefore rewrite the first integral on the right hand side as

$$\int_{\partial\Omega_R} (\tilde{\Lambda}_{pij} N_j) u_{p,k} N_k N_i + \int_{\partial\Omega_R} (\tilde{\Lambda}_{pij} N_j) u_{p,k} (\delta_{ki} - N_k N_i). \tag{A.2}$$

Since $\partial\Omega_R$ is a closed surface without boundary, for any differentiable tangential vector field $\mathbf{v} : \partial\Omega_R \to \mathbb{R}^3$ (i.e., $\mathbf{v}\cdot\mathbf{N} = 0$) we have

$$\int_{\partial\Omega_R} \mathrm{div}_s \mathbf{v} := \int_{\partial\Omega_R} v_{k,i}(\delta_{ik} - N_i N_k) = 0. \tag{A.3}$$

Let

$$v_k = (\tilde{\Lambda}_{pij} N_j)(\delta_{ki} - N_k N_i) u_p.$$

It is clear that $v_k N_k = 0$. Therefore, by (A.3) and the product rule we have

$$\begin{aligned} 0 &= \int_{\partial\Omega_R} [(\tilde{\Lambda}_{pmj} N_j)(\delta_{km} - N_k N_m) u_p]_{,i} (\delta_{ik} - N_i N_k) \\ &= \int_{\partial\Omega_R} \tau_p u_p + \int_{\partial\Omega_R} (\tilde{\Lambda}_{pmj} N_j) u_{p,i} (\delta_{im} - N_i N_m). \end{aligned} \tag{A.4}$$

Mat. B, − / Mat. A, +	NG	PG	SG	PGSG
NG	NG/NG	NG/PG	NG/SG	NG/PGSG
PG		PG/PG	PG/SG	PG/PGSG
SG			SG/SG	SG/PGSG
PGSG				PGSG/PGSG

Table 1: Interfacial conditions for an interface between materials described by different theories: NG - nongradient, PG - polarization gradient, SG - strain gradient, PGSG - polarization and strain gradient.

where the last integral on the right hand side of the above equation can be identified as the last integral in (A.2), and

$$\tau_p = [(\tilde{\Lambda}_{pmj}N_j)(\delta_{km} - N_kN_m)]_{,i}(\delta_{ik} - N_iN_k).$$

Inserting (A.2)-(A.4) into (A.1) we obtain (3.33).

B Interfacial conditions

As illustrated in Fig. 8 we consider the interface Υ in the reference body Ω_R and the two sides of Υ consist of material A (Mat. A) and material B (Mat. B), respectively. Then the kinematic continuity conditions and the differential equations in (3.37) require the following interfacial conditions on interfaces between different types of materials:

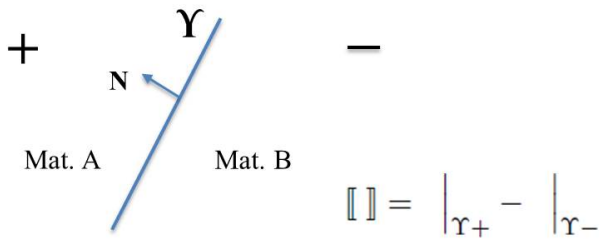


Figure 8: An interface between material A and material B.

NG/NG: $$[\![\boldsymbol{\chi}]\!] = 0, \qquad [\![(D_{\mathbf{F}}\Psi) + \tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}}]\!]\mathbf{N} = 0 \quad \text{on } \Upsilon;$$

NG/PG: $$\begin{cases} [\![\boldsymbol{\chi}]\!] = 0, \qquad [\![(D_{\mathbf{F}}\Psi) + \tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}}]\!]\mathbf{N} = 0 & \text{on } \Upsilon, \\ (D_{\boldsymbol{\Pi}}\Psi)\mathbf{N} = 0 & \text{on } \Upsilon-; \end{cases}$$

NG/SG: $$\begin{cases} [\![\boldsymbol{\chi}]\!] = 0 & \text{on } \Upsilon, \\ [\![(D_{\mathbf{F}}\Psi) + \tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}}]\!]\mathbf{N} + (\mathrm{Div}D_{\mathbf{G}}\Psi)\mathbf{N} + \boldsymbol{\tau} = 0 & \text{on } \Upsilon-, \\ (D_{\mathbf{G}}\Psi)\mathbf{N}\otimes\mathbf{N} = 0 & \text{on } \Upsilon-; \end{cases}$$

NG/PGSG:
$$\begin{cases} [\![\boldsymbol{\chi}]\!] = 0 & \text{on } \Upsilon, \\ [\![(D_{\mathbf{F}}\Psi) + \tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}}]\!]\mathbf{N} + (\mathrm{Div} D_{\mathbf{G}}\Psi)\mathbf{N} + \boldsymbol{\tau} = 0 & \text{on } \Upsilon-, \\ (D_{\boldsymbol{\Pi}}\Psi)\mathbf{N} = 0, \qquad (D_{\mathbf{G}}\Psi)\mathbf{N}\otimes\mathbf{N} = 0 & \text{on } \Upsilon-; \end{cases}$$

PG/PG:
$$\begin{cases} [\![\boldsymbol{\chi}]\!] = 0, \qquad [\![\tilde{\mathbf{P}}]\!] = 0 & \text{on } \Upsilon, \\ [\![D_{\boldsymbol{\Pi}}\Psi]\!]\mathbf{N} = 0, \quad [\![(D_{\mathbf{F}}\Psi) + \tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}}]\!]\mathbf{N} = 0 & \text{on } \Upsilon; \end{cases}$$

PG/SG:
$$\begin{cases} [\![\boldsymbol{\chi}]\!] = 0, & \text{on } \Upsilon, \\ (D_{\boldsymbol{\Pi}}\Psi)\mathbf{N} = 0 & \text{on } \Upsilon+, \\ [\![(D_{\mathbf{F}}\Psi) + \tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}}]\!]\mathbf{N} + (\mathrm{Div} D_{\mathbf{G}}\Psi)\mathbf{N} + \boldsymbol{\tau} = 0 & \text{on } \Upsilon-, \\ (D_{\mathbf{G}}\Psi)\mathbf{N}\otimes\mathbf{N} = 0 & \text{on } \Upsilon-; \end{cases}$$

PG/PGSG:
$$\begin{cases} [\![\boldsymbol{\chi}]\!] = 0, \qquad [\![D_{\boldsymbol{\Pi}}\Psi]\!]\mathbf{N} = 0 & \text{on } \Upsilon, \\ [\![(D_{\mathbf{F}}\Psi) + \tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}}]\!]\mathbf{N} + (\mathrm{Div} D_{\mathbf{G}}\Psi)\mathbf{N} + \boldsymbol{\tau} = 0 & \text{on } \Upsilon-, \\ (D_{\mathbf{G}}\Psi)\mathbf{N}\otimes\mathbf{N} = 0 & \text{on } \Upsilon-; \end{cases}$$

SG/SG:
$$\begin{cases} [\![\boldsymbol{\chi}]\!] = 0, \qquad [\![\mathrm{Grad}\boldsymbol{\chi}]\!] = 0 & \text{on } \Upsilon, \\ [\![(\mathrm{Div} D_{\mathbf{G}}\Psi) + (D_{\mathbf{F}}\Psi) + \tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}}]\!]\mathbf{N} + [\![\boldsymbol{\tau}]\!] = 0 & \text{on } \Upsilon, \\ [\![(D_{\mathbf{G}}\Psi)\mathbf{N}\otimes\mathbf{N}]\!] = 0 & \text{on } \Upsilon; \end{cases} \tag{B.5}$$

SG/PGSG:
$$\begin{cases} [\![\boldsymbol{\chi}]\!] = 0, \qquad [\![\mathrm{Grad}\boldsymbol{\chi}]\!] = 0 & \text{on } \Upsilon, \\ (D_{\boldsymbol{\Pi}}\Psi)\mathbf{N} = 0 & \text{on } \Upsilon-, \\ [\![(\mathrm{Div} D_{\mathbf{G}}\Psi) + (D_{\mathbf{F}}\Psi) + \tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}}]\!]\mathbf{N} + [\![\boldsymbol{\tau}]\!] = 0 & \text{on } \Upsilon, \\ [\![(D_{\mathbf{G}}\Psi)\mathbf{N}\otimes\mathbf{N}]\!] = 0 & \text{on } \Upsilon; \end{cases}$$

PGSG/PGSG:
$$\begin{cases} [\![\boldsymbol{\chi}]\!] = 0, \qquad [\![\mathrm{Grad}\boldsymbol{\chi}]\!] = 0, \qquad [\![D_{\boldsymbol{\Pi}}\Psi]\!]\mathbf{N} = 0 & \text{on } \Upsilon, \\ [\![(\mathrm{Div} D_{\mathbf{G}}\Psi) + (D_{\mathbf{F}}\Psi) + \tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}}]\!]\mathbf{N} + [\![\boldsymbol{\tau}]\!] = 0 & \text{on } \Upsilon, \\ [\![(D_{\mathbf{G}}\Psi)\mathbf{N}\otimes\mathbf{N}]\!] = 0 & \text{on } \Upsilon. \end{cases}$$

C Alternative variational formulations of the nongradient theory

We can alternatively formulate the nongradient continuum theory for a magneto-electro-elastic body by choosing different independent variables. For simplicity, below we restrict ourselves to electro-elastic materials and remark that similar calculations apply to magneto-elastic or magneto-electro-elastic materials. In addition, for simplicity we assume that there exists no external charges and polarization, i.e., $\tilde{\rho}^e = \tilde{\mathbf{P}}^e = 0$.

Formulation in terms of nominal electric displacement $\tilde{\mathbf{D}}$

To use nominal electric displacement $\tilde{\mathbf{D}}$ as the independent variable for electrostatics, we necessarily assume that the electrostatic boundary condition is entirely of Neumann type:

$$\mathbf{N}\cdot\tilde{\mathbf{D}}=\sigma_0 \qquad \text{on } \partial V_R, \tag{C.6}$$

whereas the mechanical loading conditions remain the same as (3.4). Then we postulate that the total free energy of the system as a functional of $(\boldsymbol{\chi},\tilde{\mathbf{D}})$ is given by

$$F'[\boldsymbol{\chi},\tilde{\mathbf{D}}]=\int_{V_R}\Phi(\mathbf{X};\mathbf{F},\tilde{\mathbf{D}})-\int_{S_N}\tilde{\mathbf{t}}^e\cdot\boldsymbol{\chi}-\int_{\Omega_R}\tilde{\mathbf{f}}^e\cdot\boldsymbol{\chi},$$

where Φ is the new stored energy density function of the system. Note that the system now includes the ambient medium $V_R\setminus\Omega_R$ instead of being restricted to the body Ω_R. We claim that the equilibrium state is governed by the variational principle

$$\min_{(\boldsymbol{\chi},\tilde{\mathbf{D}})} F'[\boldsymbol{\chi},\tilde{\mathbf{D}}], \tag{C.7}$$

where all admissible vector field $\tilde{\mathbf{D}}$ shall satisfy the boundary condition (C.6) and

$$\mathrm{Div}\tilde{\mathbf{D}}=0 \qquad \text{in } V_R. \tag{C.8}$$

By the standard calculation of first variations, we find the associated Euler-Lagrange equations are given by

$$\begin{cases}\mathrm{Curl}(D_{\tilde{\mathbf{D}}}\Phi)=0 & \text{in } V_R,\\ \mathrm{Div}D_{\mathbf{F}}\Phi+\tilde{\mathbf{f}}^e=0 & \text{in } V_R.\end{cases} \tag{C.9}$$

The first of the above equation implies that there exists a scalar potential $\xi:V_R\to\mathbb{R}$ such that

$$D_{\tilde{\mathbf{D}}}\Phi(\mathbf{F},\tilde{\mathbf{D}})=-\mathrm{Grad}\xi=:\tilde{\mathbf{E}} \qquad \text{in } V_R.$$

The above quantity is identified as the nominal electric field. Comparing the above equation with (3.41), for the same material we shall have

$$\begin{cases}\tilde{\mathbf{E}}=D_{\tilde{\mathbf{D}}}\Phi(\mathbf{F},\tilde{\mathbf{D}})=\mathbf{F}^TD_{\tilde{\mathbf{P}}}\Psi(\mathbf{F},\tilde{\mathbf{P}}),\\ D_{\mathbf{F}}\Phi(\mathbf{F},\tilde{\mathbf{D}})=D_{\mathbf{F}}\Psi(\mathbf{F},\tilde{\mathbf{P}})+\tilde{\boldsymbol{\Sigma}}_{\mathrm{MW}},\end{cases} \tag{C.10}$$

where, by (1.6), $\tilde{\mathbf{D}}=\epsilon_0J\mathbf{C}^{-1}\tilde{\mathbf{E}}+\mathbf{F}^{-1}\tilde{\mathbf{P}}$. Therefore, upon specifying the energy function $\Psi(\mathbf{F},\tilde{\mathbf{P}})$ for a material, we can solve for the energy function $\Phi(\mathbf{F},\tilde{\mathbf{D}})$ and vice versa if (i) the mapping $\tilde{\mathbf{P}}\mapsto\tilde{\mathbf{E}}=\mathbf{F}^TD_{\tilde{\mathbf{P}}}\Psi(\mathbf{F},\tilde{\mathbf{P}})$ is invertible (the inversion is denoted by $\tilde{\mathbf{P}}=\hat{\mathbf{P}}(\mathbf{F},\tilde{\mathbf{E}})$), and (ii) the mapping $\tilde{\mathbf{E}}\mapsto\tilde{\mathbf{D}}=\epsilon_0J\mathbf{C}^{-1}\tilde{\mathbf{E}}+\mathbf{F}^{-1}\hat{\mathbf{P}}(\mathbf{F},\tilde{\mathbf{E}})$ is invertible. For example, the nonlinear *isotropic* elastomer discussed in § 4.1 has the energy function $\Psi(\mathbf{F},\tilde{\mathbf{P}})$ given by (cf., (4.3))

$$\Psi(\mathbf{F},\tilde{\mathbf{P}})=W_{\mathrm{elast}}(\mathbf{U})+\frac{|\tilde{\mathbf{P}}|^2}{2(\epsilon-\epsilon_0)J}, \tag{C.11}$$

where ϵ is the deformation-independent permittivity of the elastomer. Then by straightforward calculation we can rewrite equation (C.10) as

$$\begin{cases}D_{\tilde{\mathbf{D}}}\Phi(\mathbf{F},\tilde{\mathbf{D}})=\frac{1}{\epsilon J}\mathbf{C}\tilde{\mathbf{D}},\\ D_{\mathbf{F}}\Phi(\mathbf{F},\tilde{\mathbf{D}})=D_{\mathbf{F}}W_{\mathrm{elast}}(\mathbf{U})+D_{\mathbf{F}}[\frac{1}{2\epsilon J}\tilde{\mathbf{D}}\cdot\mathbf{C}\tilde{\mathbf{D}}],\end{cases} \tag{C.12}$$

Therefore, the energy function $\Phi(\mathbf{F}, \tilde{\mathbf{D}})$ of the elastomer, within an immaterial constant, is necessarily given by

$$\Phi(\mathbf{F}, \tilde{\mathbf{D}}) = W_{\text{elast}}(\mathbf{U}) + \frac{1}{2\epsilon J}\tilde{\mathbf{D}} \cdot \mathbf{C}\tilde{\mathbf{D}}.$$

Indeed, the above energy function $\Phi(\boldsymbol{\chi}, \tilde{\mathbf{D}})$ has been formulated and used by, e.g., Suo *et al.* (2008) and Li and Landis (2012).

Formulation in terms of nominal electric field $\tilde{\mathbf{E}}$

We can also use $(\mathbf{F}, \tilde{\mathbf{E}})$ as the independent variables to formulate the theory. By the Maxwell equation (1.6) we rewrite $\tilde{\mathbf{E}} = -\text{Grad}\xi$ for a scalar potential. We define an energy functional as

$$F''[\boldsymbol{\chi}, \xi] = \int_{V_R} W(\mathbf{X}; \mathbf{F}, \tilde{\mathbf{E}}) - \int_{\Gamma_R} [\frac{1}{2}k\xi^2 - \xi(k\xi_b + \sigma_0)] - \int_{S_N} \tilde{\mathbf{t}}^e \cdot \boldsymbol{\chi} - \int_{\Omega_R} \tilde{\mathbf{f}}^e \cdot \boldsymbol{\chi}$$

and claim that the equilibrium state is determined by the variational principle

$$\min_{\boldsymbol{\chi}} \max_{\xi} F''[\boldsymbol{\chi}, \xi], \tag{C.13}$$

where the admissible potential ξ satisfies $\xi = \xi_b$ on Γ_D. By the standard calculation of first variations, we find the associated Euler-Lagrange equations are given by

$$\begin{cases} \text{Div}(D_{\tilde{\mathbf{E}}} W) = 0 & \text{in } V_R, \\ \text{Div} D_{\mathbf{F}} W + \tilde{\mathbf{f}}^e = 0 & \text{in } V_R. \end{cases} \tag{C.14}$$

Comparing the above equation with (3.41), for the same material we shall have

$$\begin{cases} \tilde{\mathbf{D}} = -D_{\tilde{\mathbf{E}}} W(\mathbf{F}, \tilde{\mathbf{E}}) = \mathbf{F}^{-1}(\epsilon_0 J \mathbf{F}^{-T}\tilde{\mathbf{E}} + \tilde{\mathbf{P}}), \qquad \tilde{\mathbf{E}} = \mathbf{F}^T D_{\tilde{\mathbf{P}}} \Psi(\mathbf{F}, \tilde{\mathbf{P}}), \\ D_{\mathbf{F}} W(\mathbf{F}, \tilde{\mathbf{E}}) = D_{\mathbf{F}} \Psi(\mathbf{F}, \tilde{\mathbf{P}}) + \tilde{\boldsymbol{\Sigma}}_{\text{MW}}. \end{cases} \tag{C.15}$$

Therefore, if the energy function $\Psi(\mathbf{F}, \tilde{\mathbf{P}})$ is specified, we may solve for the energy function $W(\mathbf{F}, \tilde{\mathbf{E}})$ and vice versa if the mapping $\tilde{\mathbf{P}} \mapsto \tilde{\mathbf{E}} = \mathbf{F}^T D_{\tilde{\mathbf{P}}} \Psi(\mathbf{F}, \tilde{\mathbf{P}})$ is invertible. As an example, we consider the isotropic nonlinear elastomer with the energy function $\Psi(\mathbf{F}, \tilde{\mathbf{P}})$ given by (C.11). Then by straightforward calculation we can rewrite equation (C.15) as

$$\begin{cases} D_{\tilde{\mathbf{E}}} W(\mathbf{F}, \tilde{\mathbf{E}}) = -\epsilon J \mathbf{C}^{-1}\tilde{\mathbf{E}}, \\ D_{\mathbf{F}} W(\mathbf{F}, \tilde{\mathbf{E}}) = D_{\mathbf{F}} W_{\text{elast}}(\mathbf{U}) + D_{\mathbf{F}}[-\frac{\epsilon J}{2}\tilde{\mathbf{E}} \cdot \mathbf{C}^{-1}\tilde{\mathbf{E}}], \end{cases} \tag{C.16}$$

Therefore, the energy function $W(\mathbf{F}, \tilde{\mathbf{E}})$ of the elastomer, within an immaterial constant, shall be given by

$$W(\mathbf{F}, \tilde{\mathbf{E}}) = W_{\text{elast}}(\mathbf{U}) - \frac{\epsilon J}{2}\tilde{\mathbf{E}} \cdot \mathbf{C}^{-1}\tilde{\mathbf{E}}.$$

A few remarks are in order here regarding the alternative variational formulations. First, formulations in terms of $\tilde{\mathbf{D}}$ and $\tilde{\mathbf{E}}$ have to include the ambient medium $V_R \setminus \Omega_R$ as part of the system; the total energy is a local functional of the independent variables. Therefore, an important advantage of the formulations with $\tilde{\mathbf{D}}$ or $\tilde{\mathbf{E}}$ being the independent variables for electrostatics lies in the Euler-Lagrange equations (C.9) and (C.14) are exceptionally simple and easy to derive. In

addition, the formulation in terms of $\tilde{\mathbf{E}}$ (i.e., the scalar potential ξ) implies a natural and simple numerical methods for solving the associated boundary value problems (Yang and Dayal 2011; Li and Landis, 2012). However, the trade-off is that (i) the variational principle (C.7) cannot address Dirichlet-type and Robin-type boundary conditions, and the independent variable $\tilde{\mathbf{D}}$ is *a priori* required to satisfy a differential constraint (C.8), (ii) the variational principle (C.13) is a min-max problem and doesn't admit a clear thermodynamic interpretation, (iii) if the ambient medium is vacuum, it is somewhat ungrounded to interpret $\tilde{\mathbf{D}}$ or $\tilde{\mathbf{E}}$ as the state of vacuum that tends to minimize or maximize the free energy of the system in a thermodynamic process until the equilibrium, and (iv) it is more natural to use $\tilde{\mathbf{P}}$ instead of $\tilde{\mathbf{D}}$ or $\tilde{\mathbf{E}}$ for gradient theories.

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