

**DRAFT: FREE ENERGIES AND STATE EQUATIONS FOR CHARACTERIZING
NONLINEAR THERMO-ELECTRO-MAGNETO-ELASTIC BEHAVIOR**

Robert L. Lowe*

Department of Mechanical and Aerospace Engineering
The Ohio State University
Columbus, OH 43210
Email: lowe.194@osu.edu

Sushma Santapuri

Stephen E. Bechtel

Marcelo J. Dapino

ABSTRACT

In this paper, we illustrate the development of a catalogue of free energies for characterizing fully coupled thermo-electro-magneto-elastic (TEME) behavior in the nonlinear or finite-deformation regime. In particular, we connect with classical equilibrium thermodynamics and use its formalism as a blueprint for rigorously and transparently introducing free energies that employ any set of intensive or extensive quantities as independent variables. We then use restrictions imposed by the second law of thermodynamics to derive the state equations associated with each of these free energies, and explore the ramifications of invariance and angular momentum. Each free energy in our catalogue characterizes a particular TEME process. Each process, in turn, correlates with a particular experiment, the independent variables being controlled and the dependent variables being the measured responses. The research presented herein will thus serve as a starting point for the development of finite-strain constitutive models for multifunctional materials with fully coupled nonlinear TEME behavior.

1 INTRODUCTION

Multifunctional materials are a broad class of engineered materials that provide structural integrity (e.g., strength, stiffness, rigidity) while simultaneously accomplishing one or more performance-based functions (e.g., sensing, actuation) [1]. The

novel capabilities afforded by multifunctional materials (e.g., self healing, power generation, power storage, crack remediation, and vibration mitigation) promise to (i) improve existing actuating and sensing technologies by increasing efficiency and reducing weight, cost, energy consumption, and size, and (ii) expand the performance space of the next generation of intelligent systems, structures, and devices [2].

Multiferroics are a relatively new class of materials that are promising candidates for a wide variety of multifunctional applications. A distinguishing feature of multiferroics is their simultaneous exhibition of ferroelectricity and ferromagnetism [3–6]. Multiferroic composites, in particular, are engineered to exhibit large magnitudes of magneto-electric coupling [6]. Magneto-electric coupling, or the magneto-electric effect, is the appearance of an electric polarization upon applying a magnetic field, or the appearance of a magnetization upon applying an electric field. Examples of multiferroic composites with magneto-electro coupling include magnetostrictive particles dispersed in an electro-active polymer matrix, magnetostrictive-piezoelectric layered composites, and magnetostrictive and piezoelectric particles dispersed in a polymer matrix [6–14]. To characterize, model, design, and optimize novel multifunctional materials like these (and more!) with complex coupled behavior, we contend that the associated mathematical models must have the breadth to accommodate three-dimensional fully coupled thermo-electro-magneto-elastic (TEME) behavior, finite deformations (geometric nonlinearities), anisotropy, and nonlinear constitutive response (material nonlinearities).

*Address all correspondence to this author.

The foundation for this type of mathematical framework was presented in [15–31]. Theoretical aspects of this seminal work have been revisited in recent years in [32–36], while practical applications to nonlinear electro-elastic solids, nonlinear magneto-elastic solids, and electro-rheological fluids have been pursued in [37–54]. However, certain aspects of constitutive modeling and material characterization, enabled through the principles of thermodynamics, remain unexplored in the literature. For instance:

- (i) The constitutive models do not at the outset explicitly identify the intensive-extensive work conjugates in the second law of thermodynamics, as is customary in classical thermodynamics [55]. As the electromagnetic work conjugates differ from model to model in finite-deformation TME (more on this in Sec. 3.4), this leads to ambiguity in specifying the fundamental energetic relationship [55] (i.e., the functional form of the internal energy). *The fundamental energetic relationship is the starting point for proceeding transparently and rigorously with the formalism of Legendre transformations, which is used to define new free energies* [55]. Often, the fundamental energetic relationship is not explicitly specified, blurring the connection between the characterizing free energy in the constitutive model and the internal energy of the system.
- (ii) The constitutive models are not comprehensive in that they do not consider all possible combinations of independent variables and corresponding thermodynamic potentials. Notably absent from many finite-deformation TME theories are free energies that employ stress as an independent variable. From an experimental perspective, intensive quantities like temperature and stress are often more convenient to control than extensive quantities like entropy, internal energy, and strain. *Hence, it is important to identify all possible free energies to provide the experimentalist with optimal flexibility when characterizing novel multifunctional materials.*

In this paper, we address items (i) and (ii) and, in so doing, take the necessary first steps toward creating a unified, comprehensive, and thermodynamically consistent model for designing and characterizing novel multifunctional materials with nonlinear fully coupled thermo-electro-magneto-elastic response. This paper follows Ref. [56] by the same authors.

2 THE FIRST PRINCIPLES OF THERMO-ELECTRO-MAGNETO-MECHANICS

We adopt the following Eulerian statements of the first principles for a polarizable, magnetizable, deformable thermo-electro-magneto-mechanical (TEMM) continuum [21, 26]:

Gauss's law for magnetism

$$\text{div } \mathbf{b} = 0, \quad (1a)$$

Faraday's law

$$\text{curl } \mathbf{e} = -\dot{\mathbf{b}} - \text{curl}(\mathbf{b} \times \mathbf{v}), \quad (1b)$$

Gauss's law for electricity

$$\text{div } \mathbf{d} = \sigma, \quad (1c)$$

Ampère-Maxwell law

$$\text{curl } \mathbf{h} = \dot{\mathbf{d}} + \text{curl}(\mathbf{d} \times \mathbf{v}) + \sigma \mathbf{v} + \mathbf{j}, \quad (1d)$$

conservation of mass

$$\dot{\rho} + \rho \text{div } \mathbf{v} = 0, \quad (1e)$$

balance of linear momentum

$$\rho \dot{\mathbf{v}} = \rho(\mathbf{f}^m + \mathbf{f}^{em}) + \text{div } \mathbf{T}, \quad (1f)$$

balance of angular momentum

$$\rho \mathbf{G}^{em} + \mathbf{T} - \mathbf{T}^T = \mathbf{0}, \quad (1g)$$

first law of thermodynamics

$$\rho \dot{\varepsilon} = \mathbf{T} \cdot \mathbf{L} + \rho(r^t + r^{em}) - \text{div } \mathbf{q}. \quad (1h)$$

The thermo-electro-magneto-mechanical fields appearing in Eqs. (1a)-(1h) are Eulerian, i.e., they are functions of the present position \mathbf{x} of a continuum particle and time t . These fields include the present mass density ρ , velocity \mathbf{v} , mechanically and electromagnetically induced specific body forces \mathbf{f}^m and \mathbf{f}^{em} , Cauchy stress \mathbf{T} , electromagnetically induced specific body couple \mathbf{G}^{em} , specific internal energy ε , Eulerian velocity gradient $\mathbf{L} = \text{grad } \mathbf{v} = \partial \mathbf{v} / \partial \mathbf{x}$, thermally and electromagnetically induced specific energy supply rates r^t and r^{em} , and Eulerian heat flux vector \mathbf{q} . Additionally, \mathbf{e} , \mathbf{d} , \mathbf{h} , \mathbf{b} , σ , and \mathbf{j} are the electric field, electric displacement, magnetic field, magnetic induction, free charge density, and conductive current density. Note that \mathbf{e} , \mathbf{d} , \mathbf{h} ,

\mathbf{b} , $\boldsymbol{\sigma}$, and \mathbf{j} are the electromagnetic fields acting on the deforming continuum as seen in its present configuration, measured with respect to a co-moving frame; they are often referred to as the *effective* electromagnetic fields in the literature [25, 29]. In Eqs. (1a)-(1h), $\text{div}(\cdot)$ denotes the Eulerian divergence, $\text{curl}(\cdot)$ denotes the Eulerian curl, \mathbf{T}^T denotes the transpose of \mathbf{T} , and

$$\mathbf{u}' = \frac{\partial \mathbf{u}}{\partial t}, \quad \dot{\mathbf{u}} = \mathbf{u}' + (\mathbf{u} \cdot \text{grad}) \mathbf{u}$$

denote the Eulerian and material time derivatives of an arbitrary vector \mathbf{u} .

The pointwise first principles (1a)-(1h) are supplemented by two additional sets of equations. One set, discussed in Sec. 3, consists of thermodynamic state equations that characterize a general TEMM process by relating the independent and dependent variables in the first-principle equations. A second set quantifies the electromagnetically induced body force \mathbf{f}^{em} , body couple \mathbf{G}^{em} , and energy supply rate r^{em} . The expressions for \mathbf{f}^{em} , \mathbf{G}^{em} , and r^{em} vary from model to model, as each model is based on a different set of principles and postulates [21, 25]. One such model for a polarizable, magnetizable, deformable continuum is coined the Maxwell-Minkowski formulation [21]:

$$\rho \mathbf{f}^{em} = \boldsymbol{\sigma} \mathbf{e} + \mathbf{j} \times \mathbf{b} + (\text{grade})^T \mathbf{p} + \mu_o (\text{grad} \mathbf{h})^T \mathbf{m} + \dot{\mathbf{d}} \times \mathbf{b} + \mathbf{d} \times \dot{\mathbf{b}}, \quad (2a)$$

$$\rho \mathbf{G}^{em} = (\mathbf{e} \otimes \mathbf{p} - \mathbf{p} \otimes \mathbf{e}) + \mu_o (\mathbf{h} \otimes \mathbf{m} - \mathbf{m} \otimes \mathbf{h}), \quad (2b)$$

$$\rho r^{em} = \mathbf{j} \cdot \mathbf{e} + \rho \mathbf{e} \cdot \left(\frac{\dot{\mathbf{p}}}{\rho} \right) + \rho \mu_o \mathbf{h} \cdot \left(\frac{\dot{\mathbf{m}}}{\rho} \right). \quad (2c)$$

In Eqs. (2a)-(2c), $(\cdot) \otimes (\cdot)$ denotes the dyadic product of two vectors,

$$\dot{\mathbf{u}} = \mathbf{u}' + \text{curl}(\mathbf{u} \times \mathbf{v}) + \mathbf{v}(\text{div} \mathbf{u})$$

is a convected rate of an arbitrary vector \mathbf{u} , and

$$\mathbf{p} = \mathbf{d} - \varepsilon_o \mathbf{e}, \quad \mathbf{m} = \frac{1}{\mu_o} \mathbf{b} - \mathbf{h} \quad (3)$$

are the electric polarization and magnetization, with ε_o and μ_o

the electric permittivity and magnetic permeability *in vacuo*. As with the other electromagnetic fields, \mathbf{p} and \mathbf{m} act on the deforming continuum as seen in its present configuration, and are measured with respect to a co-moving frame [25, 29].

3 FREE ENERGIES AND STATE EQUATIONS

In this section, we specialize to elastic materials, and demonstrate how thermodynamic restrictions can be used to develop a catalogue of free energies and thermodynamic state equations for characterizing fully coupled thermo-electro-magneto-elastic (TEME) behavior.

3.1 The Second Law of Thermodynamics

As our particular statement of the second law of thermodynamics, we adopt the Clausius-Duhem inequality [57, 58]

$$\rho \dot{\eta} - \rho \frac{r^t}{\theta} + \text{div} \left(\frac{\mathbf{q}}{\theta} \right) \geq 0, \quad (4)$$

where θ is the absolute temperature and η is the specific entropy. Inequality (4) is then algebraically combined with the first law of thermodynamics (1h) to produce the reduced Clausius-Duhem inequality

$$\begin{aligned} -\dot{\varepsilon} + \frac{1}{\rho_R} \mathbf{P} \cdot \dot{\mathbf{F}} + \theta \dot{\eta} + \mathbf{e} \cdot \left(\frac{\dot{\mathbf{p}}}{\rho} \right) + \mu_o \mathbf{h} \cdot \left(\frac{\dot{\mathbf{m}}}{\rho} \right) \\ + \frac{1}{\rho} \mathbf{j} \cdot \mathbf{e} - \frac{1}{\rho \theta} \mathbf{q} \cdot \text{grad} \theta \geq 0, \end{aligned} \quad (5)$$

where $\mathbf{P} = J \mathbf{T} \mathbf{F}^{-T}$ is the first Piola-Kirchhoff stress, $J = \rho_R / \rho$ is the determinant of the deformation gradient $\mathbf{F} = \text{Grad} \mathbf{x} = \partial \mathbf{x} / \partial \mathbf{X}$, ρ_R is the reference mass density, and \mathbf{X} is the reference position of a continuum particle. Analogous to classical thermodynamics [55], the *fundamental* statement of the second law (5) consists of contributions from *conjugate pairs* of thermal, electrical, magnetic, and mechanical quantities. Each of these conjugate pairs (or work conjugates) is the product of an *extensive* quantity in rate form (\mathbf{F} , η , \mathbf{p}/ρ , and \mathbf{m}/ρ) and an *intensive* quantity in non-rate form (\mathbf{P} , θ , \mathbf{e} , and \mathbf{h}).

3.2 The Fundamental or All-Extensive Formulation

For the remainder of the paper, we consider material response that is path independent, reversible, and rate insensitive. This implies that the deformation is elastic and fully recoverable, and the material only undergoes non-dissipative processes (e.g., no electrical or magnetic hysteresis). It follows, then, that these

non-dissipative thermo-electro-magneto-elastic processes can be described through the principles of classical equilibrium thermodynamics [55]. Analogous to classical equilibrium thermodynamics, the fundamental energy potential is the internal energy ε , which employs *extensive* quantities as its *independent variables* [55]. Hence, for the fundamental formulation, the natural *independent variables* are the *extensive* quantities \mathbf{F} , η , \mathbf{p}/ρ , and \mathbf{m}/ρ appearing as rates in inequality (5), and the natural *dependent variables* are the conjugate *intensive* quantities \mathbf{P} , θ , \mathbf{e} , and \mathbf{h} . To respect the reversible elastic nature of the processes, we demand that the material response depends on the independent variables \mathbf{F} , η , \mathbf{p}/ρ , and \mathbf{m}/ρ only through their values at the present time t , not their histories, rates, or gradients, i.e.,

$$\begin{aligned}\mathbf{P} &= \breve{\mathbf{P}}\left(\mathbf{F}, \eta, \frac{\mathbf{p}}{\rho}, \frac{\mathbf{m}}{\rho}\right), & \theta &= \breve{\theta}\left(\mathbf{F}, \eta, \frac{\mathbf{p}}{\rho}, \frac{\mathbf{m}}{\rho}\right), \\ \mathbf{e} &= \breve{\mathbf{e}}\left(\mathbf{F}, \eta, \frac{\mathbf{p}}{\rho}, \frac{\mathbf{m}}{\rho}\right), & \mathbf{h} &= \breve{\mathbf{h}}\left(\mathbf{F}, \eta, \frac{\mathbf{p}}{\rho}, \frac{\mathbf{m}}{\rho}\right), \\ \varepsilon &= \breve{\varepsilon}\left(\mathbf{F}, \eta, \frac{\mathbf{p}}{\rho}, \frac{\mathbf{m}}{\rho}\right),\end{aligned}\quad (6)$$

where the superscript breve is used to distinguish a function from its value. Equation (6)₅ is referred to as the *fundamental energetic relationship* [55]. Use of the chain rule on $\varepsilon = \breve{\varepsilon}(\mathbf{F}, \eta, \mathbf{p}/\rho, \mathbf{m}/\rho)$ gives

$$\dot{\varepsilon} = \frac{\partial \breve{\varepsilon}}{\partial \mathbf{F}} \cdot \dot{\mathbf{F}} + \frac{\partial \breve{\varepsilon}}{\partial \eta} \dot{\eta} + \frac{\partial \breve{\varepsilon}}{\partial \left(\frac{\mathbf{p}}{\rho}\right)} \cdot \overline{\left(\frac{\mathbf{p}}{\rho}\right)} + \frac{\partial \breve{\varepsilon}}{\partial \left(\frac{\mathbf{m}}{\rho}\right)} \cdot \overline{\left(\frac{\mathbf{m}}{\rho}\right)}, \quad (7)$$

and substitution of this result into the second law (5) leads to

$$\begin{aligned}& \left(\frac{1}{\rho_R} \mathbf{P} - \frac{\partial \breve{\varepsilon}}{\partial \mathbf{F}}\right) \cdot \dot{\mathbf{F}} + \left(\theta - \frac{\partial \breve{\varepsilon}}{\partial \eta}\right) \dot{\eta} \\ & + \left(\mathbf{e} - \frac{\partial \breve{\varepsilon}}{\partial \left(\frac{\mathbf{p}}{\rho}\right)}\right) \cdot \overline{\left(\frac{\mathbf{p}}{\rho}\right)} + \left(\mu_o \mathbf{h} - \frac{\partial \breve{\varepsilon}}{\partial \left(\frac{\mathbf{m}}{\rho}\right)}\right) \cdot \overline{\left(\frac{\mathbf{m}}{\rho}\right)} \\ & + \frac{1}{\rho} \mathbf{j} \cdot \mathbf{e} - \frac{1}{\rho \theta} \mathbf{q} \cdot \text{grad } \theta \geq 0.\end{aligned}\quad (8)$$

As is customary, we demand that the second law hold for all processes [57]. Since the coefficients of the rates ($\dot{\mathbf{F}}$, $\dot{\eta}$, etc.) in inequality (8) are independent of the rates themselves, and the rates may be varied independently and are arbitrary, it follows

that the coefficients vanish, i.e.,

$$\begin{aligned}\mathbf{P} &= \rho_R \frac{\partial \breve{\varepsilon}}{\partial \mathbf{F}}, & \theta &= \frac{\partial \breve{\varepsilon}}{\partial \eta}, \\ \mathbf{e} &= \frac{\partial \breve{\varepsilon}}{\partial \left(\frac{\mathbf{p}}{\rho}\right)}, & \mathbf{h} &= \frac{1}{\mu_o} \frac{\partial \breve{\varepsilon}}{\partial \left(\frac{\mathbf{m}}{\rho}\right)}.\end{aligned}\quad (9)$$

What remains of inequality (8), i.e.,

$$\mathbf{j} \cdot \mathbf{e} - \frac{1}{\theta} \mathbf{q} \cdot \text{grad } \theta \geq 0,$$

is called the residual dissipation inequality, which, in this case, quantifies losses due to Joule heating and heat conduction, both transport processes. We collectively coin the set of extensive independent variables $\{\mathbf{F}, \eta, \mathbf{p}/\rho, \mathbf{m}/\rho\}$, the thermodynamic energy potential $\varepsilon = \breve{\varepsilon}(\mathbf{F}, \eta, \mathbf{p}/\rho, \mathbf{m}/\rho)$, and the state equations (9) the *fundamental formulation* (or, alternatively, the *internal energy formulation* or the *all-extensive formulation*).

3.2.1 Polarization and Magnetization as Independent Variables. From an experimental point of view, it is more practical to control the electric polarization \mathbf{p} and magnetization \mathbf{m} than the polarization per unit mass \mathbf{p}/ρ and magnetization per unit mass \mathbf{m}/ρ . Hence, we modify the all-extensive formulation presented in Sec. 3.2 to accommodate the use of \mathbf{p} and \mathbf{m} as the electromagnetic independent variables.

We proceed by using the chain rule

$$\begin{aligned}\overline{\left(\frac{\mathbf{p}}{\rho}\right)} &= \frac{1}{\rho} \dot{\mathbf{p}} + \frac{1}{\rho} (\mathbf{F}^{-T} \cdot \dot{\mathbf{F}}) \mathbf{p}, \\ \overline{\left(\frac{\mathbf{m}}{\rho}\right)} &= \frac{1}{\rho} \dot{\mathbf{m}} + \frac{1}{\rho} (\mathbf{F}^{-T} \cdot \dot{\mathbf{F}}) \mathbf{m}\end{aligned}\quad (10)$$

to rewrite the fundamental form (5) of the second law:

$$\begin{aligned}-\dot{\varepsilon} &+ \left[\frac{1}{\rho_R} \mathbf{P} + \frac{1}{\rho} (\mathbf{e} \cdot \mathbf{p} + \mu_o \mathbf{h} \cdot \mathbf{m}) \mathbf{F}^{-T}\right] \cdot \dot{\mathbf{F}} + \theta \dot{\eta} \\ &+ \frac{1}{\rho} \mathbf{e} \cdot \dot{\mathbf{p}} + \frac{\mu_o}{\rho} \mathbf{h} \cdot \dot{\mathbf{m}} + \frac{1}{\rho} \mathbf{j} \cdot \mathbf{e} - \frac{1}{\rho \theta} \mathbf{q} \cdot \text{grad } \theta \geq 0,\end{aligned}\quad (11)$$

where we have used

$$\overline{\left(\frac{1}{\rho}\right)} = \frac{1}{\rho} \operatorname{div} \mathbf{v}, \quad \operatorname{div} \mathbf{v} = \operatorname{tr} \mathbf{L} = \operatorname{tr}(\dot{\mathbf{F}} \mathbf{F}^{-1}) = \mathbf{F}^{-T} \cdot \dot{\mathbf{F}}, \quad (12)$$

and \mathbf{F}^{-1} denotes the inverse of \mathbf{F} . In the modified form (11) of the second law, polarization \mathbf{p} and magnetization \mathbf{m} appear as rates (i.e., natural independent variables). Accordingly, the thermodynamic potential ε is a function of \mathbf{F} , η , \mathbf{p} , and \mathbf{m} , i.e., $\varepsilon = \bar{\varepsilon}(\mathbf{F}, \eta, \mathbf{p}, \mathbf{m})$; a superscript bar is used instead of a superscript breve (cf. $\varepsilon = \breve{\varepsilon}(\mathbf{F}, \eta, \mathbf{p}/\rho, \mathbf{m}/\rho)$ and $\varepsilon = \bar{\varepsilon}(\mathbf{F}, \eta, \mathbf{p}, \mathbf{m})$) to signify a different internal energy *function* with the same *value*. Use of the chain rule gives

$$\dot{\varepsilon} = \frac{\partial \bar{\varepsilon}}{\partial \mathbf{F}} \cdot \dot{\mathbf{F}} + \frac{\partial \bar{\varepsilon}}{\partial \eta} \dot{\eta} + \frac{\partial \bar{\varepsilon}}{\partial \mathbf{p}} \cdot \dot{\mathbf{p}} + \frac{\partial \bar{\varepsilon}}{\partial \mathbf{m}} \cdot \dot{\mathbf{m}}, \quad (13)$$

and substitution of this result into (11), coupled with the use of standard arguments (refer to Sec. 3.2), leads to

$$\begin{aligned} \mathbf{P} &= \rho_R \frac{\partial \bar{\varepsilon}}{\partial \mathbf{F}} - J(\mathbf{e} \cdot \mathbf{p} + \mu_o \mathbf{h} \cdot \mathbf{m}) \mathbf{F}^{-T}, & \theta &= \frac{\partial \bar{\varepsilon}}{\partial \eta}, \\ \mathbf{e} &= \rho \frac{\partial \bar{\varepsilon}}{\partial \mathbf{p}}, & \mathbf{h} &= \frac{\rho}{\mu_o} \frac{\partial \bar{\varepsilon}}{\partial \mathbf{m}}. \end{aligned} \quad (14)$$

We collectively coin the set of independent variables $\{\mathbf{F}, \eta, \mathbf{p}, \mathbf{m}\}$, the thermodynamic energy potential $\varepsilon = \bar{\varepsilon}(\mathbf{F}, \eta, \mathbf{p}, \mathbf{m})$, and the state equations (14) the *deformation-entropy-polarization-magnetization formulation*.

3.3 Other Energetic Formulations

From an experimental perspective, process characterization is most straightforwardly accomplished when the independent and dependent variables synchronize with those one wishes to control and measure, respectively. In experiments, it is often more practical to control intensive quantities than extensive quantities; for example, temperature is easier to control than entropy or internal energy. To change an independent variable in the thermodynamic energy potential from extensive to intensive, a new free energy is defined through a Legendre transformation [55], i.e.,

$$\text{new free energy} = \text{internal energy} - (\text{intensive})(\text{extensive}).$$

Following this blueprint, we present in Tabs. 1-4 a catalogue of the fifteen possible Legendre transformations of the internal

energy. These Legendre transformations are divided into four families, each employing a common set of thermomechanical independent variables: Family 1, deformation and entropy (both extensive); Family 2, deformation (extensive) and temperature (intensive); Family 3, stress (intensive) and entropy (extensive); and Family 4, stress and temperature (both intensive). For the sake of brevity, we employ the compact notation $E^{(a)(b)(c)(d)}$ for the Legendre-transformed energy potentials, where the superscript letters (a), (b), (c), and (d) are placeholders for an appropriate mechanical, thermal, electrical, and magnetic independent variable, respectively. This compact notation denotes that the Legendre-transformed energy potential $E^{F\theta pm}$, for instance, is a function of deformation \mathbf{F} , temperature θ , electric polarization per unit mass \mathbf{p}/ρ , and magnetization per unit mass \mathbf{m}/ρ . Note that the Legendre transformations in Tabs. 1-4 provide explicit connections between the new free energies $E^{(a)(b)(c)(d)}$ and the primitive internal energy ε of the system, whose evolution is governed by the first law of thermodynamics (1h).

To derive the state equations associated with the fifteen free energies shown in Tabs. 1-4, we apply the standard Coleman-Noll procedure [57], as was done in Sec. 3.2.¹ As an example, we consider the *all-intensive formulation*.

3.3.1 The All-Intensive Formulation. The independent variables for this formulation are the intensive quantities \mathbf{P} , θ , \mathbf{e} , and \mathbf{h} . The corresponding thermodynamic energy potential $E^{P\theta eh}$ is defined as the Legendre transformation of internal energy $\varepsilon = \breve{\varepsilon}(\mathbf{F}, \eta, \mathbf{p}/\rho, \mathbf{m}/\rho)$ with respect to the mechanical, thermal, electrical, and magnetic variables, from \mathbf{F} to \mathbf{P} , η to θ , \mathbf{p}/ρ to \mathbf{e} , and \mathbf{m}/ρ to \mathbf{h} ,

$$E^{P\theta eh} = \varepsilon - \frac{1}{\rho_R} \mathbf{P} \cdot \mathbf{F} - \theta \eta - \mathbf{e} \cdot \frac{\mathbf{p}}{\rho} - \mu_o \mathbf{h} \cdot \frac{\mathbf{m}}{\rho}. \quad (15)$$

Taking the rate of (15) gives

$$\begin{aligned} \dot{E}^{P\theta eh} &= \dot{\varepsilon} - \frac{1}{\rho_R} \mathbf{P} \cdot \dot{\mathbf{F}} - \frac{1}{\rho_R} \mathbf{F} \cdot \dot{\mathbf{P}} - \theta \dot{\eta} - \eta \dot{\theta} - \mathbf{e} \cdot \overline{\left(\frac{\mathbf{p}}{\rho}\right)} \\ &\quad - \frac{\mathbf{p}}{\rho} \cdot \dot{\mathbf{e}} - \mu_o \mathbf{h} \cdot \overline{\left(\frac{\mathbf{m}}{\rho}\right)} - \mu_o \frac{\mathbf{m}}{\rho} \cdot \dot{\mathbf{h}}, \end{aligned} \quad (16)$$

¹For formulations that employ either the electric polarization per unit mass \mathbf{p}/ρ or the magnetization per unit mass \mathbf{m}/ρ as an independent variable, as was the case with the all-extensive formulation in Sec. 3.2, \mathbf{p} or \mathbf{m} can *post facto* be introduced as the independent variable using the procedure described in Sec. 3.2.1.

TABLE 1. ENERGY FAMILY 1

IVs	Energy	Legendre Transformation
F, η, \mathbf{p}/ρ, \mathbf{m}/ρ	$\check{\epsilon}$	N/A
F, η, \mathbf{e}, \mathbf{m}/ρ	$E^{F\eta em}$	$E^{F\eta em} = \check{\epsilon} - \mathbf{e} \cdot \frac{\mathbf{p}}{\rho}$
F, η, \mathbf{p}/ρ, \mathbf{h}	$E^{F\eta ph}$	$E^{F\eta ph} = \check{\epsilon} - \mu_o \mathbf{h} \cdot \frac{\mathbf{m}}{\rho}$
F, η, \mathbf{e}, \mathbf{h}	$E^{F\eta eh}$	$E^{F\eta eh} = \check{\epsilon} - \mathbf{e} \cdot \frac{\mathbf{p}}{\rho} - \mu_o \mathbf{h} \cdot \frac{\mathbf{m}}{\rho}$

TABLE 2. ENERGY FAMILY 2

IVs	Energy	Legendre Transformation
F, θ, \mathbf{p}/ρ, \mathbf{m}/ρ	$E^{F\theta pm}$	$E^{F\theta pm} = \check{\epsilon} - \theta \eta$
F, θ, \mathbf{e}, \mathbf{m}/ρ	$E^{F\theta em}$	$E^{F\theta em} = \check{\epsilon} - \theta \eta - \mathbf{e} \cdot \frac{\mathbf{p}}{\rho}$
F, θ, \mathbf{p}/ρ, \mathbf{h}	$E^{F\theta ph}$	$E^{F\theta ph} = \check{\epsilon} - \theta \eta - \mu_o \mathbf{h} \cdot \frac{\mathbf{m}}{\rho}$
F, θ, \mathbf{e}, \mathbf{h}	$E^{F\theta eh}$	$E^{F\theta eh} = \check{\epsilon} - \theta \eta - \mathbf{e} \cdot \frac{\mathbf{p}}{\rho} - \mu_o \mathbf{h} \cdot \frac{\mathbf{m}}{\rho}$

TABLE 3. ENERGY FAMILY 3

IVs	Energy	Legendre Transformation
P, η, \mathbf{p}/ρ, \mathbf{m}/ρ	$E^{P\eta pm}$	$E^{P\eta pm} = \check{\epsilon} - \frac{1}{\rho_R} \mathbf{P} \cdot \mathbf{F}$
P, η, \mathbf{e}, \mathbf{m}/ρ	$E^{P\eta em}$	$E^{P\eta em} = \check{\epsilon} - \frac{1}{\rho_R} \mathbf{P} \cdot \mathbf{F} - \mathbf{e} \cdot \frac{\mathbf{p}}{\rho}$
P, η, \mathbf{p}/ρ, \mathbf{h}	$E^{P\eta ph}$	$E^{P\eta ph} = \check{\epsilon} - \frac{1}{\rho_R} \mathbf{P} \cdot \mathbf{F} - \mu_o \mathbf{h} \cdot \frac{\mathbf{m}}{\rho}$
P, η, \mathbf{e}, \mathbf{h}	$E^{P\eta eh}$	$E^{P\eta eh} = \check{\epsilon} - \frac{1}{\rho_R} \mathbf{P} \cdot \mathbf{F} - \mathbf{e} \cdot \frac{\mathbf{p}}{\rho} - \mu_o \mathbf{h} \cdot \frac{\mathbf{m}}{\rho}$

and substitution of this result into (5) yields

$$\begin{aligned}
 & -\dot{E}^{P\theta eh} - \frac{1}{\rho_R} \mathbf{F} \cdot \dot{\mathbf{P}} - \eta \dot{\theta} - \frac{\mathbf{p}}{\rho} \cdot \dot{\mathbf{e}} - \mu_o \frac{\mathbf{m}}{\rho} \cdot \dot{\mathbf{h}} \quad (17) \\
 & + \frac{1}{\rho} \mathbf{j} \cdot \mathbf{e} - \frac{1}{\rho \theta} \mathbf{q} \cdot \text{grad} \theta \geq 0,
 \end{aligned}$$

a statement of the second law for this formulation. Use of standard arguments (refer to Sec. 3.2) leads to the state equations

TABLE 4. ENERGY FAMILY 4

IVs	Energy	Legendre Transformation
$\mathbf{P}, \theta, \mathbf{p}/\rho, \mathbf{m}/\rho$	$E^{P\theta pm}$	$E^{P\theta pm} = \check{\varepsilon} - \frac{1}{\rho_R} \mathbf{P} \cdot \mathbf{F} - \theta \eta$
$\mathbf{P}, \theta, \mathbf{e}, \mathbf{m}/\rho$	$E^{P\theta em}$	$E^{P\theta em} = \check{\varepsilon} - \frac{1}{\rho_R} \mathbf{P} \cdot \mathbf{F} - \theta \eta - \mathbf{e} \cdot \frac{\mathbf{p}}{\rho}$
$\mathbf{P}, \theta, \mathbf{p}/\rho, \mathbf{h}$	$E^{P\theta ph}$	$E^{P\theta ph} = \check{\varepsilon} - \frac{1}{\rho_R} \mathbf{P} \cdot \mathbf{F} - \theta \eta - \mu_o \mathbf{h} \cdot \frac{\mathbf{m}}{\rho}$
$\mathbf{P}, \theta, \mathbf{e}, \mathbf{h}$	$E^{P\theta eh}$	$E^{P\theta eh} = \check{\varepsilon} - \frac{1}{\rho_R} \mathbf{P} \cdot \mathbf{F} - \theta \eta - \mathbf{e} \cdot \frac{\mathbf{p}}{\rho} - \mu_o \mathbf{h} \cdot \frac{\mathbf{m}}{\rho}$

$$\begin{aligned} \mathbf{F} &= -\rho_R \frac{\partial E^{P\theta eh}}{\partial \mathbf{P}}, & \eta &= -\frac{\partial E^{P\theta eh}}{\partial \theta}, \\ \mathbf{p} &= -\rho \frac{\partial E^{P\theta eh}}{\partial \mathbf{e}}, & \mathbf{m} &= -\frac{\rho}{\mu_o} \frac{\partial E^{P\theta eh}}{\partial \mathbf{h}}. \end{aligned} \quad (18)$$

We collectively coin the set of intensive independent variables $\{\mathbf{P}, \theta, \mathbf{e}, \mathbf{h}\}$, the thermodynamic energy potential $E^{P\theta eh}$, and the state equations (18) the *all-intensive formulation*.

3.4 Secondary Electromagnetic Quantities as Independent Variables

Recall that the internal energy $\varepsilon = \check{\varepsilon}(\mathbf{F}, \eta, \mathbf{p}/\rho, \mathbf{m}/\rho)$ is a function of the *extensive* quantities \mathbf{F} , η , \mathbf{p}/ρ , and \mathbf{m}/ρ . Thermodynamic energy potentials employing one or more of the *intensive* quantities \mathbf{P} , θ , \mathbf{e} , and \mathbf{h} as independent variables were introduced using Legendre transformations of ε (refer to Tabs. 1-4). These *intensive-extensive conjugate pairs* or *work conjugates* (i.e., \mathbf{P} and \mathbf{F} , θ and η , \mathbf{e} and \mathbf{p}/ρ , \mathbf{h} and \mathbf{m}/ρ) appearing in the *fundamental form* of the second law of thermodynamics (5) are dictated by the choice (2c) of the electromagnetic energy r^{em} . It follows, then, that different choices of r^{em} (of which there are many options available in the literature; see, for instance, [21, 26, 29, 32]) lead to different sets of electromagnetic conjugate pairs in finite-deformation TEMM. For instance, the electric displacement \mathbf{d} and magnetic induction \mathbf{b} frequently appear as parts of a conjugate pair (see, for example, [30, 32, 38, 43, 47]).

The formalism of Legendre transformations presented in Sec. 3.3, however, does not enable us to use \mathbf{d} and \mathbf{b} as the electromagnetic independent variables in a free energy since they are not part of the electromagnetic work conjugates employed in this paper (i.e., \mathbf{e} and \mathbf{p}/ρ , \mathbf{h} and \mathbf{m}/ρ). We thus coin \mathbf{d} and \mathbf{b} *sec-*

ondary or *auxiliary quantities*. To overcome this limitation of conventional Legendre transformations and use \mathbf{d} and \mathbf{b} as independent variables, we posit a *Legendre-type transformation* of $\varepsilon = \check{\varepsilon}(\mathbf{F}, \eta, \mathbf{p}, \mathbf{m})$, i.e.,

$$E^{F\eta db} = \varepsilon + \frac{\varepsilon_o}{2\rho} \mathbf{e} \cdot \mathbf{e} + \frac{\mu_o}{2\rho} \mathbf{h} \cdot \mathbf{h}, \quad (19)$$

whose rate form is

$$\begin{aligned} \dot{E}^{F\eta db} &= \dot{\varepsilon} + \frac{1}{2\rho} (\varepsilon_o \mathbf{e} \cdot \mathbf{e} + \mu_o \mathbf{h} \cdot \mathbf{h}) \mathbf{F}^{-T} \cdot \dot{\mathbf{F}} \\ &+ \frac{1}{\rho} (\varepsilon_o \mathbf{e} \cdot \dot{\mathbf{e}} + \mu_o \mathbf{h} \cdot \dot{\mathbf{h}}), \end{aligned} \quad (20)$$

where we have used (12). Note that the last two terms on the right-hand side of the Legendre-type transformation (19) represent electrical and magnetic energies, respectively, *in vacuo*. Substitution of (20) into the modified second law (11), and subsequent use of the algebraic relationships in (3), leads to

$$\begin{aligned} & -\dot{E}^{F\eta db} + \left[\frac{1}{\rho_R} \mathbf{P} + \frac{1}{\rho} \left(\mathbf{e} \cdot \mathbf{d} + \mathbf{h} \cdot \mathbf{b} - \frac{1}{2} \varepsilon_o \mathbf{e} \cdot \mathbf{e} \right. \right. \\ & \left. \left. - \frac{1}{2} \mu_o \mathbf{h} \cdot \mathbf{h} \right) \mathbf{F}^{-T} \right] \cdot \dot{\mathbf{F}} + \theta \dot{\eta} + \frac{1}{\rho} \mathbf{e} \cdot \dot{\mathbf{d}} + \frac{1}{\rho} \mathbf{h} \cdot \dot{\mathbf{b}} \\ & + \frac{1}{\rho} \mathbf{j} \cdot \mathbf{e} - \frac{1}{\rho \theta} \mathbf{q} \cdot \text{grad } \theta \geq 0, \end{aligned} \quad (21)$$

the second law statement for this formulation. Note that \mathbf{d} and \mathbf{b} appear as rates in inequality (21), i.e., as natural independent variables. Use of standard arguments (refer to Sec. 3.2) leads to

TABLE 5. ENERGY FAMILY 5

IVs	Energy	Legendre-Type Transformation
F, η, d, m	$E^{F\eta dm}$	$E^{F\eta dm} = \bar{\varepsilon} + \frac{\varepsilon_o}{2\rho} \mathbf{e} \cdot \mathbf{e}$
F, η, d, h	$E^{F\eta dh}$	$E^{F\eta dh} = \bar{\varepsilon} + \frac{\varepsilon_o}{2\rho} \mathbf{e} \cdot \mathbf{e} - \frac{\mu_o}{\rho} \mathbf{h} \cdot \mathbf{m}$
F, η, p, b	$E^{F\eta pb}$	$E^{F\eta pb} = \bar{\varepsilon} + \frac{\mu_o}{2\rho} \mathbf{h} \cdot \mathbf{h}$
F, η, e, b	$E^{F\eta eb}$	$E^{F\eta eb} = \bar{\varepsilon} - \frac{1}{\rho} \mathbf{e} \cdot \mathbf{p} + \frac{\mu_o}{2\rho} \mathbf{h} \cdot \mathbf{h}$
F, η, d, b	$E^{F\eta db}$	$E^{F\eta db} = \bar{\varepsilon} + \frac{\varepsilon_o}{2\rho} \mathbf{e} \cdot \mathbf{e} + \frac{\mu_o}{2\rho} \mathbf{h} \cdot \mathbf{h}$

TABLE 6. ENERGY FAMILY 6

IVs	Energy	Legendre-Type Transformation
F, θ, d, m	$E^{F\theta dm}$	$E^{F\theta dm} = \bar{\varepsilon} - \theta\eta + \frac{\varepsilon_o}{2\rho} \mathbf{e} \cdot \mathbf{e}$
F, θ, d, h	$E^{F\theta dh}$	$E^{F\theta dh} = \bar{\varepsilon} - \theta\eta + \frac{\varepsilon_o}{2\rho} \mathbf{e} \cdot \mathbf{e} - \frac{\mu_o}{\rho} \mathbf{h} \cdot \mathbf{m}$
F, θ, p, b	$E^{F\theta pb}$	$E^{F\theta pb} = \bar{\varepsilon} - \theta\eta + \frac{\mu_o}{2\rho} \mathbf{h} \cdot \mathbf{h}$
F, θ, e, b	$E^{F\theta eb}$	$E^{F\theta eb} = \bar{\varepsilon} - \theta\eta - \frac{1}{\rho} \mathbf{e} \cdot \mathbf{p} + \frac{\mu_o}{2\rho} \mathbf{h} \cdot \mathbf{h}$
F, θ, d, b	$E^{F\theta db}$	$E^{F\theta db} = \bar{\varepsilon} - \theta\eta + \frac{\varepsilon_o}{2\rho} \mathbf{e} \cdot \mathbf{e} + \frac{\mu_o}{2\rho} \mathbf{h} \cdot \mathbf{h}$

the state equations

$$\begin{aligned} \mathbf{P} &= \rho_R \frac{\partial E^{F\eta db}}{\partial \mathbf{F}} - J \left(\mathbf{e} \cdot \mathbf{d} + \mathbf{h} \cdot \mathbf{b} - \frac{1}{2} \varepsilon_o \mathbf{e} \cdot \mathbf{e} - \frac{1}{2} \mu_o \mathbf{h} \cdot \mathbf{h} \right) \mathbf{F}^{-T}, \\ \theta &= \frac{\partial E^{F\eta db}}{\partial \eta}, \quad \mathbf{e} = \rho \frac{\partial E^{F\eta db}}{\partial \mathbf{d}}, \\ \mathbf{h} &= \rho \frac{\partial E^{F\eta db}}{\partial \mathbf{b}}. \end{aligned} \quad (22)$$

We collectively coin the set of independent variables $\{\mathbf{F}, \eta, \mathbf{d}, \mathbf{b}\}$, the thermodynamic energy potential $E^{F\eta db}$,

and the state equations (22) the *deformation-entropy-electric displacement-magnetic induction formulation*.

Other Legendre-type transformations of the internal energy are shown in Tables 5 and 6.

4 RAMIFICATIONS OF INVARIANCE AND ANGULAR MOMENTUM

In addition to satisfying the restrictions imposed by the second law of thermodynamics (refer to Sec. 3), the constitutive equations for a particular material must satisfy invariance requirements. In this paper, we emphasize a particular notion of invariance, known as invariance under superposed rigid body mo-

tions (SRBMs). Invariance under SRBMs demands that if two motions of a body composed of the same material differ only by a SRBM (i.e., translation and rotation), the constitutive response generated in the two motions must be the same, apart from orientation. In this section, we demonstrate the procedure for imposing invariance and angular momentum restrictions on the state equations corresponding to a particular potential $E^{F\theta eh}$. The state equations corresponding to this potential are derived in [56]:

$$\begin{aligned} \mathbf{P} &= \rho_R \frac{\partial E^{F\theta eh}}{\partial \mathbf{F}}, & \eta &= -\frac{\partial E^{F\theta eh}}{\partial \theta}, \\ \mathbf{p} &= -\rho \frac{\partial E^{F\theta eh}}{\partial \mathbf{e}}, & \mathbf{m} &= -\frac{\rho}{\mu_o} \frac{\partial E^{F\theta eh}}{\partial \mathbf{h}}. \end{aligned} \quad (23)$$

In order to satisfy invariance under SRBMs, the free energy $E^{F\theta eh}$ must depend on \mathbf{F} , \mathbf{e} , and \mathbf{h} as follows:

$$E^{F\theta eh}(\mathbf{F}, \theta, \mathbf{e}, \mathbf{h}) = \tilde{E}^{F\theta eh}(\mathbf{C}, \theta, \mathbf{e}_e, \mathbf{h}_e), \quad (24)$$

where

$$\mathbf{C} = \mathbf{F}^T \mathbf{F}, \quad \mathbf{e}_e = \mathbf{F}^T \mathbf{e}, \quad \mathbf{h}_e = \mathbf{F}^T \mathbf{h} \quad (25)$$

are the right Cauchy-Green deformation tensor, referential electric field, and referential magnetic field, respectively. It follows that

$$\begin{aligned} \mathbf{T} &= 2\rho \mathbf{F} \frac{\partial \tilde{E}^{F\theta eh}}{\partial \mathbf{C}} \mathbf{F}^T - \mathbf{p} \otimes \mathbf{e} - \mu_o \mathbf{m} \otimes \mathbf{h}, & \eta &= -\frac{\partial \tilde{E}^{F\theta eh}}{\partial \theta}, \\ \mathbf{p} &= -\rho \mathbf{F} \frac{\partial \tilde{E}^{F\theta eh}}{\partial \mathbf{e}_e}, & \mathbf{m} &= -\frac{\rho}{\mu_o} \mathbf{F} \frac{\partial \tilde{E}^{F\theta eh}}{\partial \mathbf{h}_e}. \end{aligned} \quad (26)$$

Equations (26) are the frame-invariant forms of the state equations (23). It can be verified that the Cauchy stress (26)₁ satisfies balance of angular momentum (1g).

5 CONCLUSION

In this paper, we presented a catalogue of free energies for characterizing nonlinear thermo-electro-magneto-elastic (TEME) behavior. To ensure a transparent development of this catalogue, the fundamental energetic relationship (i.e., internal energy as a function of extensive variables) was identified at the outset, as is customary in classical thermodynamics. From this thermodynamically consistent starting point, the formalism of

Legendre transformations was employed to derive free energies that employed various sets of intensive quantities as the independent variables. Parallel to this, novel Legendre-type transformations were introduced that allowed previously inaccessible secondary electromagnetic quantities to be used as independent variables in a free energy. Restrictions imposed by the second law of thermodynamics were used to derive the state equations associated with each of the free energies in our catalogue, and the ramifications of invariance and angular momentum were explored.

Each free energy in our catalogue characterizes a particular TEME process. Each process, in turn, correlates with a particular experiment, the independent variables being controlled and the dependent variables being the measured responses. Hence, our framework provides a promising platform for the future development of finite-strain constitutive models for multifunctional materials with fully coupled nonlinear TEME behavior. We also envision the research in this paper potentially leading to the development of a new inverse design paradigm, whereby targeted macroscopic-scale performance properties are converted into conditions on the free energy through the state equations, thereby providing a “recipe” for microscopic-scale material design and fabrication.

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