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A thermodynamic theory of one-dimensional metals

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SUMMARY
One-dimensional metals are crystals exhibiting electrical conduction in a preferred direction while being completely insulating in sheets perpendicular to this direction. A thermodynamic theory is developed for deformable nonlinear 1-D metals. The constitutive theory includes hereditary effects and is kept on a high level of generality.

1. Introduction

In recent years attention has been drawn to the newly discovered so-called *one-dimensional metals*. These are crystals exhibiting electrical conduction in a preferred direction while being almost perfectly insulating in the direction perpendicular to it. Of particular interest thus far has been the mixed square planar complex compounds of Pt, [1]. Measurements on the frequency dependent reflexivity are reported in [2]. Explanations for the behavior of one-dimensional metals have been attempted in the past on the level of the microscopic substructure of the material, [3], [4]. To our knowledge, however macroscopic, phenomenologic theories that would describe one-dimensional metals are lacking. This paper is an attempt to fill this gap. We shall be concerned with the derivation of the theory, leaving applications to a further report. Nevertheless, it will be apparent that the procedure applied here will also be applicable to other types of solids, such as composite materials consisting of alternating electrically insulating or conducting sheets or of a compound of fibres and matrix exhibiting the corresponding electrical properties.

In the purely mechanical theories of the dynamical behavior of deformable bodies an *internal constraint* is characterized by a restriction on the class of possible motions so that the constrained material can only be subjected to a restricted class of deformations. More generally in a field theory, an internal constraint must be characterized as a restriction on the class of possible field responses so that the constrained material can only be subjected to a restricted class of these fields. The usual constraint conditions occur in the purely mechanical theories and appear in the form of incompressibility and inextensibility conditions in the theory of composite materials or the theory of incompressible fluids and solids. The standard reference for treating mechanical constraints is Truesdell and Noll [5]. The procedure presented there has been extended by Green, Naghdi and Trapp [6] to include thermomechanical constraints. This background material, though physically unrelated to 1-D metals is nevertheless helpful, because its mathematical treatment is analogous to the one presented here.

In the axiomatization of the theory of internal constraints Truesdell and Noll [5] assume that the forces maintaining the constraints do not perform work. Together with the constraint condition, they arrive at the result that two inner products formed from three vectors must vanish. Denoting these vectors by $\boldsymbol{A}, \boldsymbol{B}$ and $\boldsymbol{C}$, respectively and the inner product by $\langle \cdot, \cdot \rangle$, the statement is that $\langle \boldsymbol{A}, \boldsymbol{B} \rangle = 0$ for all $\boldsymbol{B}$ such that $\langle \boldsymbol{C}, \boldsymbol{B} \rangle = 0$. In Truesdell and Noll's case of simple mechanical constraints $\boldsymbol{A}$ and $\boldsymbol{C}$ are known to be independent of $\boldsymbol{B}$. It follows that $\boldsymbol{A} = \lambda \boldsymbol{C}$. It is then sufficient to characterize an internal constraint by two inner products among three vectors, two of which are independent of the third.

A different approach starts from the Clausius–Duhem inequality which must be valid for all thermodynamic processes which satisfy the constraint condition. By means of Lagrangian parameters the constraint conditions are incorporated into the entropy inequality. Satisfying this emerging inequality for all thermodynamic processes then leads to the constraint response functions.

It is exactly on this line that Green, Naghdi and Trapp [6] have extended the approach chosen in [5] to include thermo-mechanical constraints. They have found three different generalizations, all of which are based on thermodynamic arguments using the Clausius–Duhem inequality. Basically they arrive at the same answer, namely that two inner products of three vectors vanish, but Green, Naghdi and Trapp *assume* that two of these vectors are independent of the third. This assumption is needed for their conclusions to be valid.

It is thus interesting to note that there exist internal constraint conditions which do not satisfy Green *et al.*'s assumption. The constraint condition for one-dimensional metals is one of those. Although the calculations along the lines of the above inner product argument can be extended to the case of one-dimensional metals, the agreement of its result with that as obtained with the second approach must be considered to be accidental.

In deriving a theory of one-dimensional metals we thus take the position that the constraint conditions must enter the entropy inequality as side conditions. In doing so, the derivation of the constraint response functions becomes a straightforward matter following well known arguments.

The frequency dependence of the reflexivity, [2], suggests that the constitutive theory should account for hereditary effects. The explicit constitutive theory therefore is developed on the basis of Coleman and Noll's theory of fading memory [7]. It turns out that stress, entropy, polarization and magnetization are derivable to within a constraint response from a free energy functional by means of functional differential operations. As usual, heat flux and free current must be given by their own constitutive functionals.

Specially the paper is arranged as follows: In Section 2 we list the balance laws of mechanics and electrodynamics and the constraint conditions. In Section 3, after introducing the thermodynamic principles, the basic constitutive equations of one-dimensional metals are introduced. After some functional analytical preliminaries we finally draw in Section 4 the conclusions from the entropy inequality. In Section 5 the implications from the principle of material frame indifference are derived and it is shown that in doing so the balance law of moment of momentum is satisfied identically. Finally Section 6 summarizes the results.

2. Basic equations

Consider a body $\mathscr{B}$ embedded in Euclidean 3-space. Let $\mathscr{B}_R$ be its reference configuration and $\mathscr{B}_t$ its configuration at time t. The position of a particle with label X in $\mathscr{B}_R$ will be denoted by $\boldsymbol{X}$, while its position in the present configuration is denoted by $\boldsymbol{x}$. A motion of the body is then described by the smooth one-to-one map $\mathscr{B}_R \to \mathscr{B}_t$

$$\boldsymbol{x} = \boldsymbol{\chi}(\boldsymbol{X}, t), \tag{2.1}$$

the functional determinant of which

$$\operatorname{Det} \boldsymbol{F} = \operatorname{Det}(\partial \boldsymbol{x}/\partial \boldsymbol{X}) > 0 \tag{2.2}$$

never vanishes and may be assumed to be positive without loss of generality. The velocity and acceleration

$$\dot{\boldsymbol{x}} = \boldsymbol{v} = \frac{d}{dt}\boldsymbol{\chi}(\boldsymbol{X}, t)\ ; \quad \ddot{\boldsymbol{x}} = \dot{\boldsymbol{v}} = \frac{d^2}{dt^2}\boldsymbol{\chi}(\boldsymbol{X}, t) \tag{2.3}$$

are respectively, the first and second time derivative of the motion $\boldsymbol{\chi}(\cdot)$.

The dynamic behavior of electro-mechanical phenomena is described by the conservation law of mass, the balance laws of linear momentum, moment of momentum, energy and the Maxwell equations. Excluding the balance law of moment of momentum the dynamic equations are

$$\det \boldsymbol{F} = \rho_R/\rho\ ; \tag{2.4}$$

$$\rho \ddot{\boldsymbol{x}} = \operatorname{div} \boldsymbol{T} + \rho \boldsymbol{f} \; ; \tag{2.5}$$

$$\rho \dot{\varepsilon} = \operatorname{tr}(\boldsymbol{TL}) - \operatorname{div} \boldsymbol{q} + \rho r_E + \rho r_T \, , \tag{2.6$\star$}$$

where tr $(\cdot)$ denotes the trace operation and ρ_R the density function in the reference configuration. ρ is the density, $\boldsymbol{T}$ the Cauchy stress tensor, $\boldsymbol{f}$ the body force, $\dot{\varepsilon}$ the internal energy, $\boldsymbol{q}$ the energy flux vector (heat flux) and $\boldsymbol{L} = \operatorname{grad} \boldsymbol{v}$ is the velocity gradient. Dots indicate differentiation with respect to time and ρr_T is the energy supply other than electromagnetic. The remaining term represents the electromagnetic energy supply.

Maxwell's equations will be chosen in the form

$$\frac{1}{\mu_0} \operatorname{curl} \boldsymbol{B} - \frac{\partial \boldsymbol{E}}{\partial t} = \boldsymbol{J} + \frac{\partial \boldsymbol{P}}{\partial t} \operatorname{curl}(\boldsymbol{P} \times \boldsymbol{v}) + \operatorname{curl} \boldsymbol{M} \; ; \tag{2.7}$$

$$\operatorname{curl} \boldsymbol{E} + \frac{\partial \boldsymbol{B}}{\partial t} = 0 \; ; \tag{2.8}$$

$$\varepsilon_0 \operatorname{div} \boldsymbol{E} = -\operatorname{div} \boldsymbol{P} + \sigma \; ; \tag{2.9}$$

$$\operatorname{div} \boldsymbol{B} = 0 \, . \tag{2.10}$$

In this form polarization is modeled by a dipole, while magnetization is modeled by a non-relativistic circuit. It is well known that Eqns. (2.7)–(2.10) contain the conservation of electric charges

$$\frac{\partial \sigma}{\partial t} + \operatorname{div} \boldsymbol{J} \equiv 0 \tag{2.11}$$

and in fact, obvious differentiation procedures produce this equation. Hence satisfying the Maxwell equations implies identical satisfaction of the conservation of charge.

A suggestion has been made in [10] to refer to the above set of equations as the Lorentz formulation. In Eqns. (2.7)–(2.10) we have chosen MKS units. Accordingly, ε_0 and μ_0 represent universal constants such that $\varepsilon_0 \mu_0 = c^{-2}$, c denoting the speed of light in a vacuum. $\boldsymbol{E}$ is called the electric field, $\boldsymbol{B}$ the magnetic induction, $\boldsymbol{J}$ the electric current, σ the charge density and $\boldsymbol{M}$ and $\boldsymbol{P}$ the Lorentzian magnetization and polarization, respectively. All vector and tensor operations used in Eqns. (2.4)–(2.10) are with respect to the present configuration.

In a nonpolar continuum, which restriction we shall use here, the balance law of moment of momentum may be reduced to the form

$$\tfrac{1}{2} \cdot (\boldsymbol{T} - \boldsymbol{T}^T) \equiv -\boldsymbol{N} \, , \tag{2.12}$$

where $\boldsymbol{T}^T$ is the transpose of $\boldsymbol{T}$ and $\boldsymbol{N}$ the dual to the axial vector of body couple, *viz.* $\boldsymbol{N} = \text{dual}\, \boldsymbol{l}$. As usual we shall view Eqns. (2.11) and (2.12) as identities rather than as equations.

The field equations (2.4)–(2.10) are completed by specifying electromagnetic body force, body couple and energy supply. To this end let the rest frame at the particle X be that inertial frame which performs a translatoric motion relative to the frame of the present configuration** coinciding with the velocity of the particle X. Clearly, the Maxwell equations could be written in this frame by performing Lorentz transformations. Denoting the rest frame value of A by $\mathscr{A}$, these equations remain form invariant under Lorentz transformations provided the fields A and $\mathscr{A}$ are related in a certain way. As we are not interested in an exact theory we shall neglect $O(v^2/c^2)$ terms. Then

$$\begin{aligned} \mathscr{E} &= \boldsymbol{E} + \boldsymbol{v} \times \boldsymbol{B} + O(v^2/c^2) \; ; & \mathscr{P} &= \boldsymbol{P} + \frac{\boldsymbol{M} \times \boldsymbol{v}}{c^2} + O(v^2/c^2) \; ; \\ \mathscr{B} &= \boldsymbol{B} - \frac{\boldsymbol{v} \times \boldsymbol{E}}{c^2} + O(v^2/c^2) \; ; & \mathscr{M} &= \boldsymbol{M} + O(v^2/c^2) \; ; \\ \mathscr{J} &= \boldsymbol{J} - \sigma_{\boldsymbol{v}} \; ; & \tilde{\sigma} &= \sigma + O(v^2/c^2) \, . \end{aligned} \tag{2.13}$$

$\star$ $(\operatorname{div} \boldsymbol{T})_i = T_{ji,j}$, $\operatorname{tr}(\boldsymbol{TL}) = T_{ij} L_{ij}$.

** This frame is often called the Laboratory frame.

In accord with the above definitions we shall denote $\mathscr{E}, \mathscr{B}, \mathscr{P}, \mathscr{M}, \mathscr{I}$ and $\tilde{\sigma}$ as rest-frame values of the electric field, magnetic induction, polarization and magnetization, current, and charge density, respectively. Frequently the approximate value of the electric field in the rest frame as given in Eqn. (2.13) is denoted by $\mathfrak{E}$ and is called the electromotive intensity, (see [11]).

With the above definitions we are now able to give explicit expressions for $\boldsymbol{f}$, $\boldsymbol{l}$ and r_E. These expressions have been motivated and derived elsewhere, [10], [12] and are:

$$\rho \boldsymbol{f} = \boldsymbol{P} \cdot \operatorname{grad} \boldsymbol{E} + \boldsymbol{P} \times (\operatorname{curl} \boldsymbol{E}) + \operatorname{grad} \boldsymbol{B} \cdot \boldsymbol{M} - \operatorname{grad} \boldsymbol{B} \cdot (\boldsymbol{P} \times \boldsymbol{v}) + \frac{\partial}{\partial t}(\boldsymbol{P} \times \boldsymbol{B}) + \operatorname{div}(\boldsymbol{v} \otimes (\boldsymbol{P} \times \boldsymbol{B})) + \sigma \boldsymbol{E} + \boldsymbol{J} \times \boldsymbol{B} + \rho \boldsymbol{f}_{\text{ext}} ; \tag{2.14}$$

$$\boldsymbol{l} = \mathscr{P} \times \mathscr{E} + \mathscr{M} \times \mathscr{B} ; \tag{2.15}$$

$$\rho r_E = \mathscr{I} \cdot \mathscr{E} + \rho \mathscr{E} \cdot \dot{\overline{(\mathscr{P}/\rho)}} - \mathscr{M} \cdot \dot{\mathscr{B}} . \tag{2.16}$$

It remains to establish the constraint condition. To this end let $\boldsymbol{D} = \Delta(\boldsymbol{X})$ be a material C^1-vector field over $\mathscr{B}_R$ with $\|\boldsymbol{D}\| = 1$ and $\boldsymbol{\delta}(\cdot)$ its map onto $\mathscr{B}_t$: $\boldsymbol{d} = \boldsymbol{\delta}(\boldsymbol{D}, t)$. Clearly, f: $\Delta(\cdot) \to \delta(\cdot)$ reduces to the unit transformation when the bodies under consideration are immobil. Clearly,

$$\boldsymbol{d} = \boldsymbol{\delta}(\boldsymbol{D}, t) = \boldsymbol{F}\boldsymbol{D} . \tag{2.17}$$

Apart from precisions to be presented later on, I shall *define* a 1-D metal to be a solid satisfying the constraint condition

$$\mathscr{E} \cdot \boldsymbol{F}\boldsymbol{D} = 0 \leftrightarrow \mathscr{E} \cdot \boldsymbol{d} = 0 , \tag{2.18}$$

where $\boldsymbol{D}$ is a prescribed smooth vector field indicating at each point the direction of relatively high electrical conduction. Clearly, because definition (2.18) may be considered to be a constitutive equation, it must (and does in fact) satisfy the principle of material frame indifference, [5].

The definition (2.18) characterizing one-dimensional metals is not the only possible one. In fact $\boldsymbol{d} \cdot \mathscr{I} = 0$, where $\boldsymbol{d}$ now represents the direction of electrical insulation, would be another possibility. We have chosen Eqn. condition (2.18), because it will lead to a simpler theory, being aware however, that only comparison with experiments may decide about its usefulness.

Dependent upon the physical situation the vector field $\boldsymbol{D}$ is either a one or two parameter family of vectors. If e.g. the direction of relatively high electrical insulation is parallel to a preferred axis a, then $\boldsymbol{D}$ represents the planes perpendicular to a*. On the other hand, in a composite material consisting of an electrically insulating matrix and conducting fibers, $\boldsymbol{D}$ coincides with the direction of the fibers.

Sufficient smoothness preassumed, the condition (2.18) may be differentiated to yield

$$\frac{d}{dt}(\mathscr{E} \cdot \boldsymbol{d}) \equiv 0; \quad \operatorname{grad}(\mathscr{E} \cdot \boldsymbol{d}) \equiv 0 \tag{2.19}$$

and even further identities could be constructed, by performing higher derivatives with both sides of condition (2.18), (provided the derivatives do exist).

3. The thermodynamic principle and the constitutive equations

The field equations of section 2 are now completed by a thermodynamic principle and explicit constitutive equations. The first is given by the Clausius–Duhem inequality, the local form of which may be written as

$$\rho \dot{\eta} + \operatorname{div}(\boldsymbol{q}/\vartheta) \geqq \rho r_T/\vartheta , \qquad \vartheta > 0 \tag{3.1}$$

Here, η denotes the entropy and ϑ the temperature. For the entropy flux and entropy supply we have chosen the classical expressions. There are indications that this may be correct (see [10, 13, 14]).

* That is $\boldsymbol{D} = \lambda_1 \boldsymbol{D}_1 + \lambda_2 \boldsymbol{D}_2$ where $\boldsymbol{D}_1$, $\boldsymbol{D}_2$ is a base for the plane perpendicular to a and $-\infty < \lambda_1 < \infty$, $-\infty < \lambda_2 < \infty$.

Introducing the Helmholtz free energy

$$\psi = \varepsilon - \eta\vartheta - \mathscr{E}\cdot(\mathscr{P}/\rho) \tag{3.2}$$

and eliminating r_T between inequality (3.1) and Eqn. (2.6), we obtain

$$\begin{aligned} &-\rho(\dot{\psi}+\eta\dot{\vartheta}) - \frac{\boldsymbol{q}\cdot\operatorname{grad}\vartheta}{\vartheta} + \operatorname{tr}(\boldsymbol{T}\boldsymbol{L}) - \mathscr{M}\cdot\dot{\mathscr{B}} \\ &-\mathscr{P}\cdot\dot{\mathscr{E}} + \mathscr{J}\cdot\mathscr{E} \geqq 0\,, \end{aligned} \tag{3.3}$$

where the expression (2.16) has also been used.

A thermodynamic process $\mathfrak{P}(x)$ at the point X is a collection

$$\begin{aligned} \mathfrak{P}(X) = [&\boldsymbol{E}(\cdot), \boldsymbol{B}(\cdot), \boldsymbol{M}(\cdot), \boldsymbol{P}(\cdot), \boldsymbol{J}(\cdot), \rho(\cdot), \boldsymbol{x}(\cdot), \boldsymbol{T}(\cdot) \\ &\boldsymbol{q}(\cdot), \boldsymbol{g}(\cdot), \vartheta(\cdot), \sigma(\cdot), \varepsilon(\cdot), \psi(\cdot), \eta(\cdot)] \end{aligned} \tag{3.4}$$

compatible with the balance of mass, linear momentum, moment of momentum, energy and the Maxwell equations (2.7)–(2.10) defined for all t $(-\infty, \infty)$.

Let f be a function over the reals. Then

$$f^t = f^t(s) = f(t-s)\,; \qquad s \in [0, \infty) \tag{3.5}$$

and

$$f_d^t = f_d^t(s) = f(t-s) - f(t)\,; \quad s \in [0, \infty) \tag{3.6}$$

are respectively, the *history of f up to time t* and the *difference history of f up to time t.*

We call a material a polarizable and magnetizable 1-D metal if stress tensor $\boldsymbol{T}$, specific magnetization $\mathscr{M}$ and polarization $\mathscr{P}$, energy flux $\boldsymbol{q}$, electric current $\mathscr{J}$, internal energy ε, Helmholtz free energy ψ and the entropy η are, at any given time t and point X determined by the history up to time t of the displacement gradient $\boldsymbol{F}^t$, the rest frame values of the electric field $\mathscr{E}^t$, magnetic induction $\mathscr{B}^t$, the temperature ϑ^t and the present value of the temperature gradient $\boldsymbol{g} = \operatorname{grad}\vartheta$. Knowledge of the history up to time t of a variable is equivalent to knowledge of the difference history and the present value. Thus, adopting the principle of equipresence, [5], the constitutive functionals assume the form

$$\begin{aligned}
\boldsymbol{T} &= \mathop{\mathbf{T}}_{s=0}^{\infty} (\boldsymbol{F}_d^t, \mathscr{E}_d^t, \mathscr{B}_d^t, \vartheta_d^t, \boldsymbol{F}, \mathscr{E}, \mathscr{B}, \vartheta, \boldsymbol{g})\,; \\
\mathscr{P} &= \mathop{\boldsymbol{\mathscr{P}}}_{s=0}^{\infty} (\boldsymbol{F}_d^t, \mathscr{E}_d^t, \mathscr{B}_d^t, \vartheta_d^t, \boldsymbol{F}, \mathscr{E}, \mathscr{B}, \vartheta, \boldsymbol{g})\,; \\
\mathscr{M} &= \mathop{\boldsymbol{\mathscr{M}}}_{s=0}^{\infty} (\boldsymbol{F}_d^t, \mathscr{E}_d^t, \mathscr{B}_d^t, \vartheta_d^t, \boldsymbol{F}, \mathscr{E}, \mathscr{B}, \vartheta, \boldsymbol{g})\,; \\
\boldsymbol{q} &= \mathop{\mathbf{q}}_{s=0}^{\infty} (\boldsymbol{F}_d^t, \mathscr{E}_d^t, \mathscr{B}_d^t, \vartheta_d^t, \boldsymbol{F}, \mathscr{E}, \mathscr{B}, \vartheta, \boldsymbol{g})\,; \\
\mathscr{J} &= \mathop{\boldsymbol{\mathscr{J}}}_{s=0}^{\infty} (\boldsymbol{F}_d^t, \mathscr{E}_d^t, \mathscr{B}_d^t, \vartheta_d^t, \boldsymbol{F}, \mathscr{E}, \mathscr{B}, \vartheta, \boldsymbol{g})\,; \\
\varepsilon &= \mathop{\mathcal{E}}_{s=0}^{\infty} (\boldsymbol{F}_d^t, \mathscr{E}_d^t, \mathscr{B}_d^t, \vartheta_d^t, \boldsymbol{F}, \mathscr{E}, \mathscr{B}, \vartheta, \boldsymbol{g})\,; \\
\psi &= \mathop{\boldsymbol{\psi}}_{s=0}^{\infty} (\boldsymbol{F}_d^t, \mathscr{E}_d^t, \mathscr{B}_d^t, \vartheta_d^t, \boldsymbol{F}, \mathscr{E}, \mathscr{B}, \vartheta, \boldsymbol{g})\,; \\
\eta &= \mathop{\boldsymbol{\eta}}_{s=0}^{\infty} (\boldsymbol{F}_d^t, \mathscr{E}_d^t, \mathscr{B}_d^t, \vartheta_d^t, \boldsymbol{F}, \mathscr{E}, \mathscr{B}, \vartheta, \boldsymbol{g})\,;
\end{aligned} \tag{3.7}$$

where quantities on the right-hand side are functionals of the entire histories of the independent variables shown. Clearly, the Clausius–Duhem inequality places restrictions upon the response functionals. Our major goal is the determination of these restrictions. In the sequel we shall use occasionally the following simplified notation:

$$G = (F, \mathscr{E}, \mathscr{B}, \vartheta) \; ; \quad \Gamma = (G, g) \,. \tag{3.8}$$

We assume that the response functionals satisfy the principle of fading memory, [7], due to Coleman and Noll. Accordingly, let $h(s)$ be a positive monotone decreasing continuous function of s on the nonnegative real numbers. It is convenient to regard the ordered quadrupel $G=(F, \mathscr{E}, \mathscr{B}, \vartheta)$ as elements of the vector space $\mathscr{V}_{(16)}=\mathscr{V}_{(9)}\oplus\mathscr{V}_{(3)}\oplus\mathscr{V}_{(3)}\oplus\mathscr{V}_{(1)}$ where $\mathscr{V}_{(n)}$ is a real vector space with dimension n. With the inner product

$$G_1 \cdot G_2 = \operatorname{tr}(F_1 F_2) + \mathscr{E}_1 \cdot \mathscr{E}_2 + \mathscr{B}_1 \cdot \mathscr{B}_2 + \vartheta_1 \vartheta_2 \tag{3.9}$$

$\mathscr{V}_{(16)}$ becomes a Hilbert space, $\mathscr{H}_{(16)}$. Similarly we introduce the vector space $\mathscr{V}_{(19)}$, which with the norm $(\Gamma_1, \Gamma_2)=G_1 \cdot G_2 + g_1 \cdot g_2$ becomes a Hilbert space $\mathscr{H}_{(19)}$. Denoting the norm corresponding to Eqn. (3.9) by $\|\cdot\|$ we introduce a Banach space $\mathscr{A}_{(16)}$ by

$$\mathscr{A}_{(16)} = \left\{ G \in \mathscr{V}_{(16)} \;|\; \|\!|G|\!\|\; = \left[\int_0^\infty [h(s)\|G(s)\|]^2 ds \right]^{\frac{1}{2}} \right\} \tag{3.10}$$

$\|\!|\cdot|\!\|$ is called the recollection or fading memory norm.

In order to be able to draw conclusions from the entropy inequality we restrict our attention to materials, of which the response functionals depend continuously upon the difference histories G_d^t and the present values of Γ. Furthermore, we presume that the free energy depends in a continuously differentiable way on these histories. More precisely, let

$$\mathop{f}_{s=0}^{\infty} (G_d^t, \Gamma)$$

be any scalar valued response functional to be defined for every $G_d^t \in \mathscr{A}_{(16)}$ and $\Gamma \in \mathscr{H}_{(19)}$. Then $f(\cdot)$ is said to be continuously differentiable if for fixed $\Pi \in \mathscr{A}_{(16)}$ and $\pi \in \mathscr{H}_{(19)}$

$$\begin{aligned} \frac{1}{\|\pi\| + \|\!|\Pi|\!\|} \Bigg\{ & \mathop{f}_{s=0}^{\infty} (G_d^t(s) + \Pi, \Gamma + \pi) - \mathop{f}_{s=0}^{\infty} (G_d^t(s), \Gamma) \\ & - \partial_\Gamma \mathop{f}_{s=0}^{\infty} (G_d^t(s), \Gamma) - \nabla_G \mathop{f}_{s=0}^{\infty} (G_d^t(s), \Gamma | \Pi) \Bigg\} \to 0 \end{aligned} \tag{3.11}$$

as $\|\pi\| + \|\!|\Pi|\!\| \to 0$. Here, $\partial_\Gamma f(\cdot)$ is a continuous functional with values in $\mathscr{H}_{(19)}$ and for each fixed G_d^t and Γ, $\nabla_G f_{s=0}^{\infty} (G_d^t(s), \Gamma | \Pi)$ is a functional depending linearly on Π and continuously on G_d^t and Γ. Of course $\partial_\Gamma f(\cdot)$ is the partial derivative of $f_{s=0}^{\infty} (G_d^t(s), \Gamma)$ with respect to Γ holding $G_d^t(s)$ fixed.

For further development we need the following chain rule, first proved by Mizel and Wang [15] and under stronger conditions by Day [16].

Chain rule

Suppose that the functional $f(\cdot)$ is continuously differentiable and that $\Gamma(\cdot)$ and $G_d^t(\cdot)$ are defined on $(-\infty, \infty)$ and satisfy the chain rule condition, that is their values are in $\mathscr{V}_{(19)}$ and $\mathscr{V}_{(16)}$ respectively, they have two continuous derivatives $\dot{\Gamma}(\cdot)$ and $\ddot{\Gamma}(\cdot)$ and for every t G_d^t, $\dot{G}_d^t$ and $\ddot{G}_d^t$ are in $\mathscr{A}_{(16)}$. Then the function $f(t)=f_{s=0}^{\infty} (G_d^t(s), \Gamma)$ is continuously differentiable and its derivative is

$$\dot{f}(t) = \partial_\Gamma \mathop{f}_{s=0}^{\infty} (G_d^t(s), \Gamma) \cdot \dot{\Gamma} + \nabla_G \mathop{f}_{s=0}^{\infty} (G_d^t(s), \Gamma | \dot{G}_d^t) \,. \tag{3.12}$$

Let $\Omega \in \mathscr{V}_{(16)}$ be an element in the neighborhood of zero. The function $\Omega^+ = \Omega i(s)$, where $i(s)=1$, $\forall s \in [0, \infty)$ is a constant function in $\mathscr{A}_{(16)}$. The functional $\nabla_G f_{s=0}^{\infty} (\cdot | \Omega^+)$ is therefore a linear function in Ω. Thus we can define a new functional $\Delta_G f_{s=0}^{\infty} (\cdot)$ such that

$$\nabla_G \mathop{f}_{s=0}^{\infty} (\cdot | \Omega^+) = \Delta_G \mathop{f}_{s=0}^{\infty} (\cdot) \; \Omega \,. \tag{3.13}$$

With this definition and because of

$$\dot{G}_d^t(s) = -\frac{\mathrm{d}}{\mathrm{d}s} G_d^t(s) - \dot{G}\, i(s)\,, \tag{3.14}$$

we may write instead of Eqn. (3.12)

$$\begin{aligned} \dot{f}(t) = {} & D_G \mathop{f}_{s=0}^{\infty} (G_d^t(s), \Gamma) \cdot \dot{G} + \partial_g \mathop{f}_{s=0}^{\infty} (G_d^t(s), \Gamma) \cdot \dot{g} \\ & - \nabla_G \mathop{f}_{s=0}^{\infty} \left(G_d^t(s), \Gamma \,\Big|\, \frac{d}{ds} G_d^t(s) \right) \end{aligned} \tag{3.15}$$

where we have defined

$$D_G \mathop{f}_{s=0}^{\infty} (G_d^t(s), \Gamma) = \partial_G \mathop{f}_{s=0}^{\infty} (G_d^t(s), \Gamma) - \Delta_G \mathop{f}_{s=0}^{\infty} (G_d^t(s), \Gamma) \tag{3.16}$$

Provided G_d^t and Γ satisfy the chain rule condition we may thus write

$$\begin{aligned} \dot{\psi} = {} & D_G \mathop{\psi}_{s=0}^{\infty} (G_d^t(s), \Gamma) \cdot \dot{G} + \partial_g \mathop{\psi}_{s=0}^{\infty} (G_d^t(s), \Gamma) \cdot \dot{g} \\ & - \nabla_G \mathop{\psi}_{s=0}^{\infty} \left(G_d^t(s), \Gamma \,\Big|\, \frac{d}{ds} G^t(s) \right) \\ = {} & \mathrm{tr} \left\{ D_F \mathop{\psi}_{s=0}^{\infty} (G_d^t(s), \Gamma) \cdot \dot{F} \right\} \\ & + D_{\mathscr{E}} \mathop{\psi}_{s=0}^{\infty} (G_d^t(s), \Gamma) \cdot \dot{\mathscr{E}} \\ & + D_{\mathscr{B}} \mathop{\psi}_{s=0}^{\infty} (G_d^t(s), \Gamma) \cdot \dot{\mathscr{B}} \\ & + D_{\vartheta} \mathop{\psi}_{s=0}^{\infty} (G_d^t(s), \Gamma) \cdot \dot{\vartheta} \\ & + \partial_g \mathop{\psi}_{s=0}^{\infty} (G_d^t(s), \Gamma) \cdot \dot{g} - \nabla_G \mathop{\psi}_{s=0}^{\infty} \left(G_d^t(s), \Gamma \,\Big|\, \frac{d}{ds} G^t(s) \right). \end{aligned} \tag{3.17}$$

With this expression we are in a position to draw the conclusions from the entropy inequality (3.3). This inequality must be satisfied for all admissible thermodynamic processes. "Admissible" hereby means that they satisfy the balance laws of electrodynamics and mechanics and the constraint conditions as well. At this point, there arises the question as to which and how many of the constraint conditions (2.18) and (2.19) must be taken into account in satisfying the entropy inequality (3.3). A clue in this direction is obtained by observing that in order the balance laws, Eqns. (2.4)–(2.11), with the source terms, Eqns. (2.14)–(2.16), to be satisfied it is necessary and sufficient that the fields $\boldsymbol{x}$, $\mathscr{E}$, $\mathscr{B}$ and ϑ be twice continuously differentiable. Thus the following constraint conditions must be met:

$$\begin{aligned} & \mathscr{E} \cdot \boldsymbol{FD} \equiv 0\ ; \\ & \dot{\mathscr{E}} \cdot \boldsymbol{FD} + \mathscr{E} \cdot \dot{\boldsymbol{F}}\boldsymbol{D} \equiv 0\ ; \\ & \mathscr{E} \cdot \mathscr{L} \cdot \boldsymbol{D} + \mathrm{grad}\, \mathscr{E} \cdot \boldsymbol{FD} \equiv K \equiv 0\ ; \end{aligned} \tag{3.18}$$

where

$$\mathscr{L}_{ij\alpha} \equiv \boldsymbol{F}_{i\alpha,j}\,. \tag{3.19}$$

Higher derivatives of the constraint condition (2.18) must not be taken into account if the field $\boldsymbol{x}$ is at most twice continuously differentiable.

With the Lagrangian parameters λ, μ and ν which are functions of $\boldsymbol{x}$ and t only, using Eqns. (3.17) and (3.18) inequality (3.3) becomes

$$-\rho[D_\vartheta\psi(\cdot)-\eta]\dot{\vartheta}+\mathrm{tr}[(\rho D_{\boldsymbol{F}}\psi(\cdot)-\boldsymbol{F}^{-1}\boldsymbol{T}+\mu\mathscr{E}\otimes\boldsymbol{D})\dot{\boldsymbol{F}}]$$
$$-[\mathscr{P}+\rho D_{\mathscr{E}}\psi(\cdot)-\mu\boldsymbol{F}\boldsymbol{D}]\cdot\dot{\mathscr{E}}-[\mathscr{M}+\rho D_{\mathscr{B}}\psi(\cdot)]\cdot\dot{\mathscr{B}}-\rho D_{\boldsymbol{g}}\psi(\cdot)$$
$$+[\mathscr{I}+\lambda\boldsymbol{D}\boldsymbol{F}^T]\cdot\mathscr{E}+\boldsymbol{v}\cdot\boldsymbol{K}-\frac{\boldsymbol{q}\cdot\boldsymbol{g}}{\vartheta}-\nabla_{\boldsymbol{G}}\psi(\cdot|\cdot)\geqq 0\,. \tag{3.20}$$

4. The implications from inequality (3.20)

To draw the conclusions from the above inequality we need some information with regard to which variables may have arbitrarily assigned values. We assert that there exists an admissible thermodynamic process at the particle X at time t, for which $\{\boldsymbol{F}, \mathscr{E}, \mathscr{B}, \vartheta, \boldsymbol{g}\}$, the material time derivatives $\{\boldsymbol{F}, \mathscr{E}, \vartheta, \boldsymbol{g}\}$, grad $\mathscr{E}$ and $\mathscr{L}$ may have any arbitrary assigned values. In fact, since $\boldsymbol{f}_{\mathrm{ext}}$, r_T and σ are sources the balance laws of linear momentum (2.5), energy (2.6) and the Maxwell equation (2.9) can always be satisfied, irrespective of what values the aforementioned variables are assigned to. From the remaining Maxwell equations (2.7), (2.8) and (2.10) it then follows that the values of $d\mathscr{B}/dt$ and grad $\mathscr{B}$ are no longer freely assignable. Of the inequality (3.20), the fourth term thus needs special care. To facilitate future calculations, consider a Lorentz transformation from the Laboratory-frame to the rest-frame of particle X.

Equation (2.8) then becomes

$$\mathrm{curl}'\,\mathscr{E}+\partial\mathscr{B}/\partial t'=0\,, \tag{4.1}$$

where primes indicate differentiation with respect to the rest-frame coordinates. With the transformation (see [12]),

$$\left.\begin{aligned}\partial/\partial t' &= \gamma\frac{d}{dt}\\ \mathrm{grad}' &= \mathrm{grad}_\perp+\gamma\left(\mathrm{grad}_\parallel+\frac{\boldsymbol{v}}{c^2}\otimes\frac{\partial}{\partial t}\right)\end{aligned}\right\}\quad \gamma=(1-v^2/c^2)^{-\frac{1}{2}}\,; \tag{4.2}$$

where $\mathrm{grad}_\perp$ and $\mathrm{grad}_\parallel$ denote vector differentiation perpendicular and parallel to $\boldsymbol{v}$, respectively. We thus obtain from Eqns. (4.1) and (4.2)

$$\begin{aligned}-\frac{d}{dt}=\frac{1}{2\gamma}(\mathrm{grad}\,\mathscr{E}-(\mathrm{grad}\,\mathscr{E})^T) &= \frac{1}{2\gamma}(\mathrm{grad}_\perp\mathscr{E}-(\mathrm{grad}_\perp\mathscr{E})^T)\\ &\quad+\tfrac{1}{2}(\mathrm{grad}_\parallel\mathscr{E}-(\mathrm{grad}_\parallel\mathscr{E})^T)\\ &\quad+\tfrac{1}{2}\left\{\left[\frac{\boldsymbol{v}}{c^2}\otimes\left(\frac{\partial\mathscr{E}}{\partial t}\right)\right]-\left[\frac{\boldsymbol{v}}{c^2}\otimes\left(\frac{\partial\mathscr{E}}{\partial t}\right)\right]^T\right\}.\end{aligned} \tag{4.3}$$

Substituting Eqns. (4.3) and (3.19) into Eqn. (3.20) it is readily seen that the emerging inequality is linear in $\mathscr{L}$ which may have any value. Thus its coefficient must vanish, implying that

$$v=0\,. \tag{4.4}$$

Since the remaining inequality is then linear in $\dot{\vartheta}$, $\dot{\boldsymbol{F}}$, $\dot{\mathscr{E}}$, $\dot{\boldsymbol{g}}$ and $\dot{\mathscr{B}}$ one obtains

$$\begin{aligned} &D_{\boldsymbol{g}}\underset{s=0}{\overset{\infty}{\psi}}(\cdot)=0\rightarrow\psi=\underset{s=0}{\overset{\infty}{\psi}}(G_d^t(s),G)\,;\\ &\boldsymbol{T}=-\rho D_{\boldsymbol{F}}\underset{s=0}{\overset{\infty}{\psi}}(G_d^t(s),\boldsymbol{G})\boldsymbol{F}^T+\mu\mathscr{E}\otimes(\boldsymbol{F}\boldsymbol{D})\,;\\ &\mathscr{P}=-\rho D_{\mathscr{E}}\underset{s=0}{\overset{\infty}{\psi}}(G_d^t(s),\boldsymbol{G})-\mu\boldsymbol{F}\boldsymbol{D}\,;\\ &\mathscr{M}=-\rho D_{\mathscr{B}}\underset{s=0}{\overset{\infty}{\psi}}(G_d^t(s),\boldsymbol{G})\,;\\ &\eta=\quad D_\theta\underset{s=0}{\overset{\infty}{\psi}}(G_d^t(s),\boldsymbol{G})\,;\end{aligned} \tag{4.5}$$

and

$$\nabla_G \underset{s=0}{\overset{\infty}{\psi}} \left(G_d^t, G \left| \frac{d}{ds} G^t(s) \right.\right) - \underset{s=0}{\overset{\infty}{\boldsymbol{q}}} (G_d^t(s), \Gamma) \cdot \boldsymbol{g}/\vartheta$$

$$+ \left[\underset{s=0}{\overset{\infty}{\mathscr{J}}} (G_d^t(s), \Gamma) + \lambda \boldsymbol{D} \boldsymbol{F}^T \right] \cdot \mathscr{E} \geqq 0 . \tag{4.6}$$

Thus, the stress and polarization possess a constraint response. Note that heat flux and free current are not restricted by the entropy inequality. It proves to be advantageous to introduce the representation

$$\mathscr{J} = \underset{s=0}{\overset{\infty}{\mathscr{J}}} (G_d^t, \Gamma) - \lambda \boldsymbol{F} \boldsymbol{D} \tag{4.7}$$

where λ is the Lagrangian parameter associated with Eqn. $(3.18)_1$. With this choice the dissipation inequality (4.6) becomes

$$\nabla_G \underset{s=0}{\overset{\infty}{\psi}} \left(G_d^t(s), G \left| \frac{d}{ds} G^t(s) \right.\right) + \underset{s=0}{\overset{\infty}{\mathscr{J}}} (G_d^t(s), \Gamma) \cdot \mathscr{E} - \underset{s=0}{\overset{\infty}{\boldsymbol{q}}} (G_d^t(s), \Gamma) \cdot \boldsymbol{g}/\vartheta \geqq 0 \tag{4.8}$$

Note that this implies

$$\nabla_G \underset{s=0}{\overset{\infty}{\psi}} \left(G_d^t(s), G \left| \frac{d}{ds} G^t(s) \right.\right) + \underset{s=0}{\overset{\infty}{\mathscr{J}}} (G_d^t(s), \Gamma) \cdot \mathscr{E} \geqq 0 \tag{4.9}$$

but generally neither

$$\underset{s=0}{\overset{\infty}{\boldsymbol{q}}} (G_d^t(s), \Gamma) \cdot \boldsymbol{g} \leqq 0 , \tag{4.10}$$

nor

$$\underset{s=0}{\overset{\infty}{\mathscr{J}}} (G_d^t(s), \Gamma) \cdot \mathscr{E} \geqq 0 , \tag{4.11}$$

nor

$$\nabla_G \underset{s=0}{\overset{\infty}{\psi}} \left(G_d^t(s), G \left| \frac{d}{ds} G(s) \right.\right) \geqq 0 . \tag{4.12}$$

Thus the Plank inequality is not satisfied except in an electric insulator or if $\mathscr{E}=0$. Following methods of Coleman it can then be shown that, if equilibrium is defined by $\mathscr{E}=\boldsymbol{g}=0$ and vanishing time derivatives of the various independent fields that the equilibrium response functions

$$f^\infty = \underset{s=0}{\overset{\infty}{f}} (G_d^t(s) = 0, \boldsymbol{F}, 0, \mathscr{B}, \vartheta, 0)$$

are given

$$\boldsymbol{T}^\infty = \frac{\partial \psi^\infty}{\partial \boldsymbol{F}} \boldsymbol{F}^T + \mathring{\boldsymbol{T}} \; ; \quad \mathscr{M}^\infty = -\rho \frac{\partial \psi^\infty}{\partial \mathscr{B}} \; ;$$

$$\mathscr{P}^\infty = -\rho \frac{\partial \psi^\infty}{\partial \mathscr{E}} + \mathring{\mathscr{P}} \; ; \quad \eta^\infty = -\frac{\partial \psi^\infty}{\partial \vartheta} , \tag{4.13}$$

while

$$\boldsymbol{q}^\infty = 0 \; ; \quad \mathscr{J}^\infty = \mathring{\mathscr{J}} \; ; \quad \nabla_G \psi^\infty = 0 , \tag{4.14}$$

where

$$\mathring{\boldsymbol{T}} = \mu \mathscr{E} \otimes (\boldsymbol{F}\boldsymbol{D}) \; ; \quad \mathring{\mathscr{P}} = -\mu \boldsymbol{F}\boldsymbol{D} \; ; \quad \mathring{\mathscr{J}} = -\lambda \boldsymbol{F}\boldsymbol{D} . \tag{4.15}$$

Moreover, *in a theory neglecting Joule* heat it can be shown by a standard approach (see Coleman [8]) that $\dot{\psi} \leqq 0$, provided the deformation gradient, temperature, electric field and magnetic induction are held fixed. In particular the equilibrium Helmholtz free energy can never increase.

Let $G_{t_0}^c$ be the constant continuation of the history $G(s)$, that is, let $G_{t_0}^c(s) = G(s)$ for $s = t_0$ and

$G^c_{t_0}(s) = G(t_0)$ when $s = t_0$. Then from a proof similar to that of Coleman [8] it follows

$$\underset{s=0}{\overset{\infty}{\psi}}\left((G^{ct}_{t_0}(s)_d, G\right) \to \psi^{\infty}(G)$$

as $t \to \infty$. That is to say, under constant continuation the free energy relaxes to the equilibrium value. This property together with $\dot{\psi} \leqq 0$ (under constant continuation) implies

$$\underset{s=0}{\overset{\infty}{\psi}}\left((G^{ct}_{t_0}(s)_d, G^c_{t_0}\right) \geqq \psi^{\infty}(G), \tag{4.14}$$

asserting that among all histories ending with given values of the deformation gradient the electric field, magnetic induction and the temperature, the constant history yields the least energy.

These results, which are only correct when Joule heat is neglected, are valid in particular in a linearized theory.

5. Consequences of material frame indifference

The principle of material frame indifference, [5], which states that an admissible process must remain admissible after a change of frame of reference of the form

$$x^*(t) = c(t) + Q(t)x(t) \; ; \quad t^* = t - a, \tag{5.1}$$

where $c(t)$ is any time dependent vector, $Q(t)$ any proper orthogonal transformation and a a constant, imposes certain restrictions on the constitutive functionals. The scalars ψ, η, ε and ϑ are unaffected by the change of frame, but $F, \mathscr{E}, \mathscr{B}, \mathscr{P}, \mathscr{M}, \mathscr{I}, q, g$ and T are assumed to transform as follows:

$$\begin{aligned} &F \to QF \; ; \quad \mathscr{E} \to Q\mathscr{E} \; ; \quad \mathscr{P} \to Q\mathscr{P} \; ; \\ &q \to Qq \; ; \quad \mathscr{B} \to Q\mathscr{B} \; ; \quad \mathscr{M} \to Q\mathscr{M} \; ; \\ &g \to Qg \; ; \quad \mathscr{I} \to Q\mathscr{I} \; ; \quad T \to QTQ^T . \end{aligned} \tag{5.2}$$

Defining

$$\begin{aligned} &C^t(s) = [F^t(s)]^T F^t(s) \; ; \\ &\mathsf{E}^t(s) = [F^t(s)]^T \mathscr{E}^t(s) \; ; \qquad \mathrm{Grad}\, \vartheta = Fg \; ; \\ &\mathsf{B}^t(s) = [F^t(s)]^T \mathscr{B}^t(s) , \end{aligned} \tag{5.3}$$

and the Piola stress T^P by

$$T^P = F^{-1}(T - \mathring{T})(F^{-1})^T , \tag{5.4}$$

it can be shown using methods introduced by Noll [17] and Coleman [8] that

$$\begin{aligned} \psi &= \underset{s=0}{\overset{\infty}{\psi}}\left(C^t_d, \mathsf{E}^t_d, \mathsf{B}^t_d, \vartheta^t_d ; C, \mathsf{E}, \mathsf{B}, \vartheta\right) ; \\ \varepsilon &= \underset{s=0}{\overset{\infty}{\varepsilon}}\left(C^t_d, \mathsf{E}^t_d, \mathsf{B}^t_d, \vartheta^t_d ; C, \mathsf{E}, \mathsf{B}, \vartheta\right) ; \\ \eta &= \underset{s=0}{\overset{\infty}{\eta}}\left(C^t_d, \mathsf{E}^t_d, \mathsf{B}^t_d, \vartheta^t_d ; C, \mathsf{E}, \mathsf{B}, \vartheta\right) ; \\ T &= F \underset{s=0}{\overset{\infty}{T}}\left(C^t_d, \mathsf{E}^t_d, \mathsf{B}^t_d, \vartheta^t_d ; C, \mathsf{E}, \mathsf{B}, \vartheta\right) F^T + \mathring{T} \; ; \\ \mathscr{P} &= F \underset{s=0}{\overset{\infty}{\mathscr{P}}}\left(C^t_d, \mathsf{E}^t_d, \mathsf{B}^t_d, \vartheta^t_d ; C, \mathsf{E}, \mathsf{B}, \vartheta\right) + \mathring{\mathscr{P}} \; ; \\ \mathscr{M} &= F \underset{s=0}{\overset{\infty}{\mathscr{M}}}\left(C^t_d, \mathsf{E}^t_d, \mathsf{B}^t_d, \vartheta^t_d ; C, \mathsf{E}, \mathsf{B}, \vartheta\right) ; \end{aligned}$$

and

$$\mathscr{I} = \boldsymbol{F} \underset{s=0}{\overset{\infty}{\mathscr{J}}} (C_d^t, \mathsf{E}_d^t, \mathsf{B}_d^t, \vartheta_d^t; C, \mathsf{E}, \mathsf{B}, \vartheta, \text{Grad}\,\vartheta) + ;$$

$$\boldsymbol{q} = \boldsymbol{F} \underset{s=0}{\overset{\infty}{\boldsymbol{q}}} (C_d^t, \mathsf{E}_d^t, \mathsf{B}_d^t, \vartheta_d^t; C, \mathsf{E}, \mathsf{B}, \vartheta, \text{Grad}\,\vartheta),$$

where $C = C(t)$ and $C_d^t(s) = C(t-s) - C(s)$ and similarly for the other variables.

Two questions remain left to be answered. First, how are the operators $D_F[\cdot]$, $D_{\mathscr{E}}[\cdot]$, $D_{\mathscr{B}}[\cdot]$ and $D_C[\cdot]$, $D_{\mathsf{E}}[\cdot]$ and $D_{\mathsf{B}}[\cdot]$ connected, and second, is angular momentum satisfied identically? Both questions can be answered by following an argument due to Coleman, [8], involving jump continuations. As the calculations are straightforward generalizations of the ones of Coleman we simply give the result. To this end, let

$$\begin{aligned} \psi &= \underset{s=0}{\overset{\infty}{\psi}} (F_d^t(s), \ \mathscr{E}_d^t(s), \ \mathscr{B}_d^t(s), \ \vartheta_d^t(s), \ C, \mathscr{E}, \mathscr{B}, \vartheta) \\ &= \underset{s=0}{\overset{\infty}{\tilde{\psi}}} (C_d^t(s), \ \mathsf{E}_d^t(s), \ \mathsf{B}_d^t(s), \ \vartheta_d^t(s), \ C, \mathsf{E}, \mathsf{B}, \vartheta) \end{aligned} \tag{5.6}$$

be the functionals for the Helmholtz free energy ψ. We then show that

$$\begin{aligned} D_F \underset{s=0}{\overset{\infty}{\psi}} (\cdot) &= 2FD_C \underset{s=0}{\overset{\infty}{\tilde{\psi}}} (\cdot) + \left(D_{\mathsf{E}} \underset{s=0}{\overset{\infty}{\tilde{\psi}}} (\cdot)\right) \otimes \mathscr{E} + \left(D_{\mathsf{B}} \underset{s=0}{\overset{\infty}{\tilde{\psi}}} (\cdot)\right) \otimes \mathscr{B} \\ D_{\mathscr{E}} \underset{s=0}{\overset{\infty}{\psi}} (\cdot) &= FD_{\mathsf{E}} \underset{s=0}{\overset{\infty}{\tilde{\psi}}} (\cdot); \quad D_{\mathscr{B}} \underset{s=0}{\overset{\infty}{\psi}} (\cdot) = FD_{\mathsf{B}} \underset{s=0}{\overset{\infty}{\tilde{\psi}}} (\cdot). \end{aligned} \tag{5.7}$$

Using these representations we may write for stress, polarization and magnetization

$$\begin{aligned} T &= \mathring{T} + 2\rho FD_C \underset{s=0}{\overset{\infty}{\tilde{\psi}}} (\cdot)F^T + \left(\rho FD_{\mathsf{E}} \underset{s=0}{\overset{\infty}{\tilde{\psi}}} (\cdot)\right) \otimes \mathscr{E} + \left(\rho FD_{\mathsf{B}} \underset{s=0}{\overset{\infty}{\tilde{\psi}}} (\cdot)\right) \otimes \mathscr{B} \\ \mathscr{P} &= -\rho FD_{\mathsf{E}} \underset{s=0}{\overset{\infty}{\tilde{\psi}}} (\cdot) + \mathring{\mathscr{P}}; \\ \mathscr{M} &= -\rho FD_{\mathsf{E}} \underset{s=0}{\overset{\infty}{\tilde{\psi}}} (\cdot), \end{aligned} \tag{5.8}$$

from which one easily deduces

$$1/2(T = T^T) = -1/2[\mathscr{E} \otimes \mathscr{P} - \mathscr{P} \otimes \mathscr{E} + \mathscr{B} \otimes \mathscr{M} - \mathscr{M} \otimes \mathscr{B}] \tag{5.9}$$

which is equal to dual $\boldsymbol{l}$. Conservation of moment of momentum is therefore identically satisfied once objectivity has been applied to the constitutive functionals.

6. Alternative approaches. Discussion

In the preceding derivation of a theory of deformable one-dimensional metals the constraint conditions were introduced into the entropy inequality by means of Lagrangian parameters, which are assumed to be functions of place and time only. Once, this step was taken, the subsequent derivations turned out to be more or less straightforward.

A more subtle question arose when it was decided how many constraint conditions had to be taken into account when drawing the conclusions from the entropy inequality. In principle, by successive differentiation as many constraint conditions as we please can be derived. Of those only identities were considered, which did not require higher smoothness properties of the basic field variables than the balance laws of mechanics and electrodynamics. Obviously this leaves us with the open question that other results might emerge when a different point of view is taken. In the extreme case of analytic solutions, all fields are infinitely differentiable. Our point of view then would request, that an infinite number of constraint conditions be considered.

By induction it is easily shown that for one-dimensional metals the same results emerge as the ones which were derived in the preceding section.

As stated in the introduction there is an alternative approach to derive a theory of one-dimensional metals, which goes back to Green, Naghdi and Trapp [6]. One starts by assuming that the constitutive functionals are determined to within a constraint response:

$$f = \underset{s=0}{\overset{\infty}{\mathfrak{F}}} \left(\boldsymbol{G}_d^t(s), \boldsymbol{G}, \boldsymbol{g}\right) + \mu \mathring{f} \tag{6.1}$$

where μ is an undetermined parameter which is only a function of position and time.

These representations are then substituted into the entropy inequality, which in our situation gives

$$-\rho\dot{\mu}\mathring{\psi} + \mu\left(-\rho(\dot{\overline{\mathring{\psi}}} + \mathring{\eta}\vartheta) - \frac{\mathring{\boldsymbol{q}}\cdot\boldsymbol{g}}{\vartheta} + \operatorname{tr}(\mathring{\boldsymbol{T}}\boldsymbol{L}) - \mathring{\mathscr{P}}\cdot\dot{\mathscr{E}} - \mathring{\mathscr{M}}\cdot\dot{\mathscr{B}} + \mathring{\mathscr{J}}\cdot\mathscr{E}\right) + \Xi \geqq 0\,. \tag{6.2}$$

Here Ξ is a term not involving μ and $\dot{\mu}$. Since there exists a thermodynamic process where the last two quantities may have arbitrarily assigned values and since Ξ is not a function of μ and $\dot{\mu}$ one concludes that

$$\mathring{\psi} = 0\,;$$

$$-\rho(\dot{\overline{\mathring{\psi}}} + \mathring{\eta}\vartheta) - \frac{\boldsymbol{q}\cdot\boldsymbol{g}}{\vartheta} + \operatorname{tr}(\mathring{\boldsymbol{T}}\boldsymbol{L}) - \mathring{\mathscr{P}}\cdot\mathscr{E} - \mathscr{M}\cdot\dot{\mathscr{B}} + \mathring{\mathscr{J}}\cdot\mathscr{E} = 0 \tag{6.3}$$

and

$$\Xi \geqq 0 \tag{6.4}$$

It is now observed that Eqn. $(6.3)_2$ has the form of an inner product, say $\langle \boldsymbol{A}, \boldsymbol{D}\rangle$. Similarly the constraint conditions can be summarized by $\langle \boldsymbol{C}, \boldsymbol{D}\rangle = 0$. Two difficulties now arise. First no unique definition of $\boldsymbol{C}$ is possible, since some of its components depend upon those of $\boldsymbol{D}$. Second, $\boldsymbol{D}$ must not have any arbitrarily assigned value, but be chosen compatibly with the balance laws of electrodynamics (and mechanics). There is no unique way of introducing these side conditions. Should those be introduced in Eqn. (6.2) or in one of the inner products $\langle \boldsymbol{A}, \boldsymbol{D}\rangle$ and $\langle \boldsymbol{C}, \boldsymbol{D}\rangle$ or both? The values of the constraint response thus depends on the choice of these possibilities. Hence we must reject this approach as a means for determining the constraint responses.

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