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Free Energies in a General Non-Local Theory of a Material with Memory

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FREE ENERGIES IN A GENERAL NON-LOCAL THEORY OF A MATERIAL WITH MEMORY

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A general theory of non-local materials, with linear constitutive equations and memory effects, is developed within a thermodynamic framework. Several free energy and dissipation functionals are constructed and explored. These include an expression for the minimum free energy and a functional that is a free energy for important categories of memory kernels and is explicitly a functional of the minimal state. The functionals discussed have a similar general form to the corresponding expressions for simple materials. A number of new results are derived for them, most of which apply equally to both types of material. In particular, detailed formulae are given for these quantities in the case of sinusoidal histories.

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1. Introduction

A non-local theory of heat flow in a rigid conductor with memory is developed in Ref. 1, while a similar theory for incompressible fluids is proposed in Ref. 2. Suitable free energy functionals are given for both materials. We refer also to Ref. 3 for a discussion of non-simple materials. The formulation in these references is generalized in the present work to apply to a range of field theories with linear memory constitutive equations. Various quadratic free energy functionals of the most general form and associated rates of dissipation are proposed and discussed. These include an expression for the minimum free energy and a functional that is a free energy for important categories of memory kernels and is explicitly a functional of the minimal state. The functionals discussed have a similar general form to the corresponding expressions for simple materials, with the difference however that new independent field variables are included, which (greatly) extend the vector space on which the functionals are defined. Constitutive relationships derived from thermodynamics, involving these new variables, take a generalized form. A number of new results are derived for the non-simple case, most of which in fact apply equally to simple materials. In particular, detailed formulae are given for the various free energy functionals in the case of sinusoidal histories.

There are two equivalent approaches to developing general equations describing non-local materials.[18] The first is to include terms in the form of a divergence of an extra flux in the first or second laws of thermodynamics. The other is to introduce internal and external mechanical (or electromagnetic or heat) powers and write generalized forms of the first and second laws in terms of these. While the second approach has some theoretical advantages,[18] it is convenient to consider both approaches in parallel. Indeed, it is shown that the two approaches are consistent with a hypothesis on the form of a generalized work function which will be introduced below. This hypothesis is the basis of the general theory presented here.

A number of important references on non-simple materials are given in Ref. 18. Early work on the topic is discussed in, for example, Ref. 14. Regarding minimum and related free energies for simple materials with memory, see, in particular, Refs. 23, 11, 17, 22, 15, 12, 24, 25 and 3. For recent discussions on the topic of modeling viscoelastic behavior, we have, for example, Refs. 31, 13 and 3. On the matter of terminology, we regard simple and local equivalent descriptions of materials. Also non-simple and non-local are interchangeable.

2. Derivation of the Field Equations

Let us begin by stating the first and second laws of thermodynamics. The internal energy per unit mass and the entropy per unit mass at $(\mathbf{x}, t)$, both scalar quantities, are denoted respectively by $e(\mathbf{x}, t)$ and $\eta(\mathbf{x}, t)$. The local absolute temperature is $\theta(\mathbf{x}, t) \in \mathbb{R}^+$. The heat flux vector is denoted by $\mathbf{q}(\mathbf{x}, t) \in \mathbb{R}^3$. We introduce a variable $\kappa(\mathbf{x}, t)$ which is the coldness, given by $1/\theta > 0$. Let us define

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$\mathbf{g}, \mathbf{d} \in \mathbb{R}^3$ as

$$\mathbf{g} = \nabla\theta, \quad \mathbf{d} = \nabla\kappa = -\frac{1}{\theta^2}\mathbf{g}. \tag{2.1}$$

The energy balance equation or first law of thermodynamics has the form

$$p - \rho\dot{e} - \nabla\cdot\mathbf{q} - \nabla\cdot\Phi_e + \rho r = 0, \tag{2.2}$$

where p is a power term related to the case of simple materials and $\Phi_e \in \mathbb{R}^3$ is an extra flux, the *interstitial work flux*, associated with the internal energy and the first law to take account of non-local behavior.[32,14] The quantity ρ is the mass density, r is the external radiation absorbed per unit time, per unit mass at $(\mathbf{x}, t)$.

In the alternative formulation,[18] the first law for thermomechanical systems can be written as

$$\rho\dot{e} = \mathcal{P}_m^i + \mathcal{P}_h^i, \quad \mathcal{P}_h^i = -\nabla\cdot\mathbf{q} + \rho r, \tag{2.3}$$

where $\mathcal{P}_m^i$ is the internal mechanical power which, for simple materials, reduces to p. The correspondence between the two formulations is expressed by $\mathcal{P}_m^i = p - \nabla\cdot\Phi_e$.

The second law of thermodynamics can be written as

$$D = \dot{\eta} + \frac{1}{\rho}\nabla\cdot\left(\frac{\mathbf{q}}{\theta}\right) + \frac{1}{\rho}\nabla\cdot\Phi_\eta - \kappa r \geq 0, \tag{2.4}$$

where the quantity $D(\mathbf{x}, t)$ is the total rate of entropy production per unit mass, while Φ_η is an extra flux of entropy.[33] Such an extra flux is associated with non-local behavior.

The Helmholtz free energy per unit mass is defined by

$$\psi_H = e - \theta\eta. \tag{2.5}$$

In terms of this quantity, we can write (2.2) as

$$\dot{\psi}_H + \theta D = \frac{p}{\rho} - \eta\dot{\theta} - \frac{1}{\rho}\nabla\cdot\Phi_e + \frac{\theta}{\rho}\nabla\cdot\Phi_\eta - \frac{1}{\rho\theta}\mathbf{q}\cdot\mathbf{g}. \tag{2.6}$$

In the alternative formulation, we write (2.4) in the form[18]

$$\dot{\eta} - \frac{1}{\rho}\mathcal{A}_{\mathrm{en}}^i = D \geq 0, \tag{2.7}$$

where $\mathcal{A}_{\mathrm{en}}^i$ is the *internal entropy action* given by

$$\mathcal{A}_{\mathrm{en}}^i = -\nabla\cdot\frac{\mathbf{q}}{\theta} + \rho\frac{r}{\theta} - \nabla\cdot\Phi_\eta = \mathcal{A}_{\mathrm{en}}^e. \tag{2.8}$$

The latter quantity is the *external entropy action*.[18] Instead of (2.6), we have

$$\dot{\psi}_H + \theta D = \frac{1}{\rho}(\mathcal{P}^i - \theta\mathcal{A}_{\mathrm{en}}^i) - \eta\dot{\theta}, \quad \mathcal{P}^i = \mathcal{P}_m^i + \mathcal{P}_h^i, \tag{2.9}$$

where $\mathcal{P}_m^i$ and $\mathcal{P}_h^i$ are introduced in (2.3). For a general framework explaining actions in systems, see Ref. 6.

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Relation (2.6) is unsatisfactory for materials with memory because neither ψ_H nor η are in general uniquely defined. Typically, there are many different functions of state which can be identified as the free energy and entropy, respectively. On the other hand, e is always uniquely defined. It is therefore advantageous to modify the above standard formulation. We introduce the following free energy:

$$\psi = \kappa e - \eta = \kappa \psi_H \tag{2.10}$$

and (2.6) is replaced by

$$\dot{\psi} + D = \kappa \frac{p}{\rho} + e\dot{\kappa} - \frac{\kappa}{\rho} \nabla \cdot \Phi_e + \frac{1}{\rho} \nabla \cdot \Phi_\eta + \frac{1}{\rho} \mathbf{d} \cdot \mathbf{q}. \tag{2.11}$$

It will be assumed that ρ is independent of $\mathbf{x}$, so that the material is homogeneous. Then, (2.11) can be written as

$$\dot{\psi} + D = \kappa \frac{p}{\rho} + e\dot{\kappa} - \nabla \cdot \Phi + \frac{1}{\rho} \mathbf{d} \cdot \mathbf{Q} = \dot{W},$$

$$\Phi = \frac{\kappa}{\rho} \Phi_e - \frac{1}{\rho} \Phi_\eta, \quad \mathbf{Q} = \mathbf{q} + \Phi_e, \tag{2.12}$$

where $\dot{W}$ is the generalized rate of work done on the body.[a] Instead of (2.9), we have

$$\dot{\psi} + D = \frac{\kappa}{\rho}(\mathcal{P}^i - \theta \mathcal{A}^i_{\text{en}}) + e\dot{\kappa} = \dot{W}. \tag{2.13}$$

Observe that, in the formulation based on (2.11), the natural temperature variable is κ rather than θ.

Relation $(2.12)_1$ or (2.13) is the first law, while the second law is the requirement that D be non-negative (see (2.4)).

Alternatively, if (2.13) is adopted, the specification of $\dot{W}$ determines $\mathcal{P}^i - \theta \mathcal{A}^i_{\text{en}}$. We see that it is not necessary to determine $\mathcal{P}^i_m, \mathcal{P}^i_h$ and $\mathcal{A}^i_{\text{en}}$ separately in order to specify the thermodynamic laws and derive consequences from these. It is sufficient to specify the combination $\mathcal{P}^i - \theta \mathcal{A}^i_{\text{en}}$, which is achieved by giving the form of $\dot{W}$.

The choice of whether to formulate the theory in material or spatial coordinates is important in nonlinear mechanics, which is considered in Sec. 2.3. Whichever choice is made does not greatly affect the general formalism.

Let

$$\widehat{\mathbf{d}}(t) = \int_0^t \mathbf{d}(u)du, \tag{2.14}$$

so that

$$\frac{1}{\rho} \mathbf{Q} \cdot \mathbf{d} = \frac{1}{\rho} \mathbf{Q} \cdot \dot{\widehat{\mathbf{d}}}. \tag{2.15}$$

[a]The abstract formulation of thermodynamics in Refs. 16, 17 and 3 for example (based on Refs. 6 and 7) applies to non-simple materials if we identify the work function in those developments with the generalized work $W(t)$ in the present context.

It will be assumed that the free energy is a functional of the history of this quantity, namely $\widehat{\mathbf{d}}^t(s), s > 0$, or specifically a functional of

$$\widehat{\mathbf{d}}(t) - \widehat{\mathbf{d}}^t(s) = \int_{t-s}^{t} \mathbf{d}(u)\,du, \tag{2.16}$$

with no separate dependence on $\widehat{\mathbf{d}}(t)$. This is essentially the basis of the approach in Ref. 29.

We introduce a convenient compact notation. Let the field variables be $\boldsymbol{\Lambda} : \mathbb{R} \mapsto \Gamma, \boldsymbol{\Sigma} : \mathbb{R} \mapsto \Gamma$ where Γ is a given finite-dimensional vector space.[b] We will treat $\boldsymbol{\Lambda}$ as the independent and $\boldsymbol{\Sigma}$ as the dependent field quantity. Putting

$$\boldsymbol{\Lambda} = \{\boldsymbol{\Lambda}_m, \widehat{\mathbf{d}}\}, \quad \boldsymbol{\Sigma} = \left\{\boldsymbol{\Sigma}_m, \frac{\mathbf{Q}}{\rho}\right\}, \quad \kappa \frac{p}{\rho} = \boldsymbol{\Sigma}_m \cdot \dot{\boldsymbol{\Lambda}}_m, \tag{2.17}$$

where $\boldsymbol{\Sigma}_m$ and $\boldsymbol{\Lambda}_m$ are the non-thermal variables, we can write $(2.12)_1$ as[c]

$$\dot{\psi} + D = \dot{W}(t) = e\dot{\kappa} + \boldsymbol{\Sigma} \cdot \dot{\boldsymbol{\Lambda}} - \nabla \cdot \Phi. \tag{2.18}$$

Remark 2.1. It is not helpful to include $\kappa(t)$ in the definition of $\boldsymbol{\Lambda}(t)$ given by $(2.17)_1$, with e included in $\boldsymbol{\Sigma}$. This is because we introduce space gradients of $\boldsymbol{\Lambda}$ below and the space derivatives of $\kappa(t)$ would overlap with $\mathbf{d}$ and its higher derivatives. These in turn are closely related to $\widehat{\mathbf{d}}(t)$ and its derivatives, already included in the definition of $\boldsymbol{\Lambda}(t)$ and its gradients.

The form $(2.17)_3$ is typical of mechanical systems with deformation as the independent variable. In the material representation discussed in Sec. 2.3,

$$p = \mathbf{S} \cdot \dot{\mathbf{F}}, \tag{2.19}$$

where $\mathbf{S}$ is the first Piola–Kirchhoff stress tensor and $\mathbf{F}$ is the material gradient of deformation.

For dielectric materials subject to electromagnetic fields, the form of the power function is somewhat different,[20] given in effect by

$$P_{\mathrm{em}} = \dot{\boldsymbol{\Sigma}}_{\mathrm{em}} \cdot \boldsymbol{\Lambda}_{\mathrm{em}}, \tag{2.20}$$

where $\boldsymbol{\Lambda}_{\mathrm{em}}$ represents the electric and magnetic fields and $\boldsymbol{\Sigma}_{\mathrm{em}}$ the electric and magnetic inductions. However, putting

$$\dot{\boldsymbol{\Sigma}}_{\mathrm{em}} \cdot \boldsymbol{\Lambda}_{\mathrm{em}} = \frac{d}{dt}(\boldsymbol{\Sigma}_{\mathrm{em}} \cdot \boldsymbol{\Lambda}_{\mathrm{em}}) - \boldsymbol{\Sigma}_{\mathrm{em}} \cdot \dot{\boldsymbol{\Lambda}}_{\mathrm{em}} \tag{2.21}$$

and using the free enthalpy $\psi - \boldsymbol{\Sigma}_{\mathrm{em}} \cdot \boldsymbol{\Lambda}_{\mathrm{em}}$ rather than the free energy, we can cast the equations in the same general form as for mechanics, merely changing the sign

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[b]The space dependence of field variables is understood but generally not specified explicitly, even though the space gradients of $\boldsymbol{\Lambda}$ will play a fundamental role in this work.
[c]Note the two quite different uses of the dot product in (2.18). The first is in Γ while the second is in $\mathbb{R}^3$, indicating a divergence.

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of $\boldsymbol{\Sigma}_{\mathrm{em}}$. For the case of electromagnetic conductors, the power can be expressed as in (2.18), using the same device as introduced in (2.14).

Materials for which (2.20) applies will not be specifically considered in what follows. However, this case can be developed in a very similar manner. One difference is pointed out in Sec. 4.

We now seek to develop a non-local theory with linear constitutive equations which generalizes the results of Refs. 1 and 2, so that they apply to a wide range of linear non-simple theories in mechanics, electromagnetism and for heat flow in rigid materials.

The introduction of non-simple behavior can be achieved (see Refs. 35 and 30, for example) by adding n more independent field quantities, which consist of gradient operators acting on $\boldsymbol{\Lambda}$, where n is a positive integer. These would be materials of grade $n + 1$ in the terminology of Ref. 35, for the case of mechanics, which is discussed in Sec. 2.3.

The new independent field variables are $\nabla\boldsymbol{\Lambda}, \nabla^2\boldsymbol{\Lambda}, \nabla^3\boldsymbol{\Lambda}, \ldots, \nabla^n\boldsymbol{\Lambda}$. The operators $\nabla^r, r = 2, 3, \ldots, n$ are completely symmetric. The number of independent quantities among their 3^r components on a given basis is $(r+1)(r+2)/2$ (see p. 39 in Ref. 21). The new histories are denoted by

$$\nabla\boldsymbol{\Lambda}^t(\mathbf{x}, s), \nabla^2\boldsymbol{\Lambda}^t(\mathbf{x}, s), \ldots, \nabla^n\boldsymbol{\Lambda}^t(\mathbf{x}, s), \quad s \in \mathbb{R}^+, \tag{2.22}$$

where the standard notation for the history

$$\boldsymbol{\Lambda}^t(\mathbf{x}, s) = \boldsymbol{\Lambda}(\mathbf{x}, t - s) \tag{2.23}$$

has been used.

2.1. *The fundamental hypotheses*

To completely determine the first and second laws as given by (2.18) and (2.4), one must specify $\dot{W}, \psi$ and D. Indeed, it emerges that if we specify $\dot{W}$ and ψ, then the requirement of consistency with the second law yields the form of D and indeed the detailed form of the constitutive equations for the material. We first give the form of $\dot{W}$ and show how it is consistent in general with (2.18). Also, in Sec. 2.3, the example of nonlinear mechanics is discussed in the context of this assumption on $\dot{W}$ and shown to be consistent with (2.13).

For a wide class of non-simple materials, the generalized rate of work can be put in the form

$$\dot{W} = e\dot{\kappa} + \boldsymbol{\Sigma}_0 \cdot \dot{\boldsymbol{\Lambda}} + \boldsymbol{\Sigma}_1 \cdot \nabla\dot{\boldsymbol{\Lambda}} + \boldsymbol{\Sigma}_2 \cdot \nabla^2\dot{\boldsymbol{\Lambda}} + \cdots + \boldsymbol{\Sigma}_n \cdot \nabla^n\dot{\boldsymbol{\Lambda}}, \tag{2.24}$$

where the tensor functions $\boldsymbol{\Sigma}_r, r = 0, 1, 2, \ldots, n$ are fundamental to the theory. In particular, for simple mechanical materials, $\boldsymbol{\Sigma}_r = \mathbf{0}, r = 1, 2, \ldots, n$, and $\boldsymbol{\Sigma}_0$ is proportional to the stress tensor.[d] The quantity $\boldsymbol{\Sigma}_r$ is a given basis that will have

---

[d]The quantity $\mathbf{0}$ will be used for the zero in any vector space with dimensions greater than unity.

1 the same number of subscripts with the same symmetry properties as $\nabla^r \dot{\mathbf{\Lambda}}$ and each
2 element of this tensor quantity belongs to Γ. The inner product notation $\mathbf{\Sigma}_r \cdot \nabla^r \dot{\mathbf{\Lambda}}$
3 means that each ∇ is connected to a specific dimension of $\mathbf{\Sigma}_r$ in the sense that,
4 on a particular basis, the components of a given ∇ are summed over a particular
5 subscript of $\mathbf{\Sigma}_r$ and each term in the summation is a scalar product $\Gamma \times \Gamma \mapsto \mathbb{R}$.
6 Thus, on a particular basis, $\mathbf{\Sigma}_2$ for example has components which we denote by
7 $\Sigma_{(2)ij} = \Sigma_{(2)ji}, i, j = 1, 2, 3$, and ∇^2 will be represented as $\frac{\partial^2}{\partial x_i \partial x_j} = \partial_i \partial_j$, giving
8 that $\mathbf{\Sigma}_2 \cdot \nabla^2 \dot{\mathbf{\Lambda}} = \Sigma_{(2)ij} \cdot \partial_i \partial_j \dot{\mathbf{\Lambda}} \in \mathbb{R}$. The dot product in this last expression is the
9 scalar product $\Gamma \times \Gamma \mapsto \mathbb{R}$ between $\Sigma_{(2)ij}$ and $\dot{\mathbf{\Lambda}}$.

10 **Proposition 2.1.** *For $\dot{W}$ given by the right-hand side of* $(2.18)_2$, *where $\nabla \cdot \Phi$*
11 *is explicitly a divergence of a vector quantity[e] Φ, then $\mathbf{\Sigma}$ in* (2.18) *is uniquely*
12 *determined to be*

13 $$\mathbf{\Sigma} = \mathbf{\Sigma}_0 - \nabla \cdot \mathbf{\Sigma}_1 + \nabla^2 \cdot \mathbf{\Sigma}_2 + \cdots + (-1)^n \nabla^n \cdot \mathbf{\Sigma}_n, \qquad (2.25)$$

14 *where the notation $\nabla^r \cdot \mathbf{\Sigma}_r \in \Gamma$ means that each ∇ forms a divergence with respect*
15 *to its corresponding tensor index of $\mathbf{\Sigma}_r$ for $r = 1, 2, \ldots, n$.*

16 **Proof.** We seek to extract a scalar term $\mathbf{G} \cdot \dot{\mathbf{\Lambda}}$, where $\mathbf{G} \in \Gamma$ from the quantity
17 $\mathbf{\Sigma}_r \cdot \nabla^r \dot{\mathbf{\Lambda}}$ for example. Given the available differential operators and field quantities,
18 $\mathbf{G}$ must have the form

19 $$\mathbf{G} = \alpha \nabla^r \cdot \mathbf{\Sigma}_r, \qquad (2.26)$$

20 where $\alpha \in \mathbb{R}$ is a constant. Indeed, there is no finite manipulation that could yield
21 an irrational number in this role, so that it can be assumed to be a rational number.
22 The basic tool of transformation of expressions such as (2.24) is the product
23 rule of Calculus[f]

24 $$(\nabla^p \cdot \mathbf{\Sigma}_r) \cdot (\nabla^q \dot{\mathbf{\Lambda}}) = \nabla \cdot [(\nabla^p \cdot \mathbf{\Sigma}_r) \cdot (\nabla^{q-1} \dot{\mathbf{\Lambda}})] - (\nabla^{p+1} \cdot \mathbf{\Sigma}_r) \cdot (\nabla^{q-1} \dot{\mathbf{\Lambda}}), \qquad (2.27)$$

25 or the inverse of this, transferring a ∇ operator from $\mathbf{\Sigma}_r$ to $\dot{\mathbf{\Lambda}}$. The latter transfor-
26 mation is not of interest here. Consider the right-hand side of (2.27). The divergence
27 term is included in the overall divergence term and we can re-apply the operation

---

[e]When we say that $\nabla \cdot \Phi$ is explicitly a divergence of a vector quantity, we mean this in a superficial algebraic sense, in contrast to the deeper (analysis related) statement that it can be expressed as a divergence. This latter property is of course true of many scalar functions. What is meant here is that the ∇'s in the theory can be manipulated so that one of them is operating on vector quantities made up of the $\mathbf{\Sigma}_r$ or their derivatives, multiplying $\dot{\mathbf{\Lambda}}$ or its derivatives.

[f]For the expression in the first bracket, the dot product implies that each ∇ in ∇^p, on a given basis, is summed over its corresponding index in $\mathbf{\Sigma}_r$. The dot product between the two terms in brackets means that q of the remaining dimensions in $\mathbf{\Sigma}_r$ are summed over the indices in ∇^q and a scalar product $\Gamma \times \Gamma \mapsto \mathbb{R}$ occurs for each multiplication of objects in Γ. Thus, for example, using the notation introduced earlier, we have that $(\nabla^2 \cdot \mathbf{\Sigma}_5) \cdot (\nabla^2 \dot{\mathbf{\Lambda}}) \in \mathbb{R}^3$ has components $\partial_i \partial_j \Sigma_{(5)ijklm} \cdot \partial_k \partial_l \dot{\mathbf{\Lambda}}$ in a given basis, where the remaining dot product refers to the scalar product $\Gamma \times \Gamma \mapsto \mathbb{R}$ between $\Sigma_{(5)ijklm}$ and $\dot{\mathbf{\Lambda}}$.

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to the second term, which approaches the desired form (2.26). By systematically proceeding in this manner, we uniquely obtain the relation

$$
\begin{aligned}
\mathbf{\Sigma}_r \cdot \nabla^r \dot{\mathbf{\Lambda}} &= (-1)^r (\nabla^r \cdot \mathbf{\Sigma}_r) \cdot \dot{\mathbf{\Lambda}} + \nabla \cdot \Phi_r, \\
\Phi_r &= \mathbf{\Sigma}_r \cdot (\nabla^{r-1} \dot{\mathbf{\Lambda}}) - (\nabla \cdot \mathbf{\Sigma}_r) \cdot (\nabla^{r-2} \dot{\mathbf{\Lambda}}) \\
&\quad + \cdots + (-1)^s (\nabla^s \cdot \mathbf{\Sigma}_r) \cdot (\nabla^{r-s-1} \dot{\mathbf{\Lambda}}) \\
&\quad + \cdots + (-1)^{r-1} (\nabla^{r-1} \cdot \mathbf{\Sigma}_r) \cdot \dot{\mathbf{\Lambda}}.
\end{aligned}
\tag{2.28}
$$

Thus, we have

$$
\Phi' = \Phi_1 + \Phi_2 + \cdots + \Phi_n,
\tag{2.29}
$$

and (2.18) applies, where $\mathbf{\Sigma}$ is given by (2.25) and $\nabla \cdot \Phi' = \nabla \cdot \Phi$. $\square$

The simplest assumption regarding Φ is to take it equal to Φ' as given by (2.29). For a detailed development of the theory, it is also necessary to make an assumption regarding the form of Φ_e in $(2.12)_3$ and $(2.12)_4$, which will however not be required for our purposes. This issue does not arise if κ is independent of $\mathbf{x}$.

Remark 2.2. The quantity $\mathbf{\Sigma}(t)$ is the dependent variable with an immediate physical interpretation. For example, in mechanics, it is proportional to the physical stress tensor. The quantities $\mathbf{\Sigma}_i(t), i = 0, 1, 2, \ldots$ are also physically measurable quantities and expressions for them will be given below.

The fundamental constitutive assumption, which characterizes the material, is that the free energy ψ depends in a specified way on the independent field variables at the current time t and on their histories. Thus, we write

$$
\begin{aligned}
\psi(t) = \tilde{\psi}(\mathbf{x}, \kappa^t(\mathbf{x}, \cdot), \kappa(\mathbf{x}, t), \mathbf{\Lambda}^t(\mathbf{x}, \cdot), \mathbf{\Lambda}(\mathbf{x}, t), \nabla \mathbf{\Lambda}^t(\mathbf{x}, \cdot), \nabla \mathbf{\Lambda}(\mathbf{x}, t), \\
\nabla^2 \mathbf{\Lambda}^t(\mathbf{x}, \cdot), \nabla^2 \mathbf{\Lambda}(\mathbf{x}, t), \ldots, \nabla^n \mathbf{\Lambda}^t(\mathbf{x}, \cdot), \nabla^n \mathbf{\Lambda}(\mathbf{x}, t)).
\end{aligned}
\tag{2.30}
$$

The dependence on $\mathbf{x}$, over and above that in the independent field variables allows the possibility of spatial inhomogeneity. We will henceforth mainly omit all space variables.

It is assumed that $\tilde{\psi}$ is differentiable with respect to the independent variables at the current time and Fréchet-differentiable with respect to the histories within a suitable Hilbert space $\mathcal{H}$ (Fading Memory Principle[4,5]). The time derivative $\dot{\psi}$ is given by

$$
\dot{\psi} = \partial_t \psi + \partial_h \psi,
\tag{2.31}
$$

where ∂_t indicates differentiation with respect to field variables at the current time t, while $\partial_h \psi$ indicates differentiation with respect to the remainder of the time dependence, which will consist of Fréchet differentials of ψ at the histories of the independent field quantities, in the direction of the time derivatives of these histories.

The quantity $\partial_t \psi$ is given by

$$\partial_t \psi = \frac{\partial \psi}{\partial \kappa(t)} \dot{\kappa}(t) + \frac{\partial \psi}{\partial \mathbf{\Lambda}(t)} \cdot \dot{\mathbf{\Lambda}}(t) + \frac{\partial \psi}{\partial \nabla \mathbf{\Lambda}(t)} \cdot \nabla \dot{\mathbf{\Lambda}}(t)$$

$$+ \frac{\partial \psi}{\partial \nabla^2 \mathbf{\Lambda}(t)} \cdot \nabla^2 \dot{\mathbf{\Lambda}}(t) + \cdots + \frac{\partial \psi}{\partial \nabla^n \mathbf{\Lambda}(t)} \cdot \nabla^n \dot{\mathbf{\Lambda}}(t). \qquad (2.32)$$

The derivatives with respect to field quantities are assumed to be continuous in their arguments. Consider the relation $(2.18)_1$. Using (2.31) with (2.32) to substitute for $\dot{\psi}$ and (2.24) for $\dot{W}(t)$, we generalize a standard argument,[4,5] based on the indeterminacy of the signs of $\dot{\mathbf{\Lambda}}(t), \nabla \dot{\mathbf{\Lambda}}(t), \nabla^2 \dot{\mathbf{\Lambda}}(t), \ldots, \nabla^n \dot{\mathbf{\Lambda}}(t)$. This yields the detailed constitutive relations

$$e(t) = \frac{\partial \psi}{\partial \kappa(t)} \qquad (2.33)$$

and

$$\mathbf{\Sigma}_0(t) = \frac{\partial \psi}{\partial \mathbf{\Lambda}(t)}, \quad \mathbf{\Sigma}_1(t) = \frac{\partial \psi}{\partial \nabla \mathbf{\Lambda}(t)},$$

$$\qquad (2.34)$$

$$\mathbf{\Sigma}_2(t) = \frac{\partial \psi}{\partial \nabla^2 \mathbf{\Lambda}(t)}, \ldots, \mathbf{\Sigma}_n(t) = \frac{\partial \psi}{\partial \nabla^n \mathbf{\Lambda}(t)},$$

together with an expression for the rate of dissipation

$$D = -\partial_h \psi \geq 0, \qquad (2.35)$$

by virtue of (2.4). Thus, the Fréchet differentials, which crucially involve the history dependence of ψ, yield the rate of dissipation of the material. Using (2.33) and (2.34) in (2.32), we see from (2.24) that

$$\partial_t \psi(t) = \dot{W}(t). \qquad (2.36)$$

An immediate consequence of (2.34) and (2.25) is that

$$\mathbf{\Sigma}(t) = \frac{\partial \psi}{\partial \mathbf{\Lambda}(t)} - \nabla \cdot \frac{\partial \psi}{\partial \nabla \mathbf{\Lambda}(t)} + \nabla^2 \cdot \frac{\partial \psi}{\partial \nabla^2 \mathbf{\Lambda}(t)}$$

$$+ \cdots + (-1)^n \nabla^n \cdot \frac{\partial \psi}{\partial \nabla^n \mathbf{\Lambda}(t)} = \frac{\delta \psi}{\delta \mathbf{\Lambda}}, \qquad (2.37)$$

where $\frac{\delta \psi}{\delta \mathbf{\Lambda}}$ is a variational derivative, as in the Calculus of Variations, with respect to the independent field variables at the current time, $\nabla^r \mathbf{\Lambda}(t), r = 0, 1, 2, \ldots, n$.

Let us introduce another compact notation on an encompassing vector space $\mathcal{B}$ containing all the field variables. We define the independent variable

$$\mathbf{C}(t) = (\mathbf{\Lambda}(t), \nabla \mathbf{\Lambda}(t), \nabla^2 \mathbf{\Lambda}(t), \ldots, \nabla^n \mathbf{\Lambda}(t)) \in \mathcal{B} \qquad (2.38)$$

and the dependent variable

$$\mathbf{D}(t) = (\mathbf{\Sigma}_0(t), \mathbf{\Sigma}_1(t), \mathbf{\Sigma}_2(t), \ldots, \mathbf{\Sigma}_n(t)) \in \mathcal{B}. \qquad (2.39)$$

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Relations (2.24) and (2.36) yield that

$$\partial_t \psi = \dot{W}(t) = e\dot{\kappa} + \mathbf{D}(t) \odot \dot{\mathbf{C}}(t), \tag{2.40}$$

where the symbol $\odot$ is the dot product in $\mathcal{B}$, implying a scalar product of the individual components in their respective vector spaces. Let us denote the dimension of $\mathcal{B}$ by m. The sequence of operators $\boldsymbol{\nabla}$ and $\boldsymbol{\nabla}_a$ are defined as

$$\begin{aligned}
\boldsymbol{\nabla} &= (1, \nabla, \nabla^2, \dots, \nabla^n), \\
\boldsymbol{\nabla}_a &= (1, -\nabla, +\nabla^2, \dots, (-1)^n \nabla^n).
\end{aligned} \tag{2.41}$$

Then (2.38) can be written symbolically as

$$\mathbf{C}(t) = \boldsymbol{\nabla} \boldsymbol{\Lambda}(t). \tag{2.42}$$

Also, from (2.25) and (2.37),

$$\boldsymbol{\Sigma}(t) = \frac{\delta \psi}{\delta \boldsymbol{\Lambda}(t)} = \boldsymbol{\nabla}_a \odot \mathbf{D}(t). \tag{2.43}$$

We can write the dependence of ψ on the independent field variables given by (2.30) as

$$\psi = \tilde{\psi}(\kappa^t, \kappa(t), \mathbf{C}^t, \mathbf{C}(t)), \tag{2.44}$$

where $\mathbf{C}^t$ is the past history, given by (see (2.23))

$$\mathbf{C}^t(s) = \mathbf{C}(t - s), \quad s \in \mathbb{R}^{++}. \tag{2.45}$$

The relative history is defined as

$$\mathbf{C}_r^t(s) = \mathbf{C}^t(s) - \mathbf{C}(t). \tag{2.46}$$

It will be assumed that

$$\lim_{s \to \infty} \mathbf{C}^t(s) = \mathbf{0}. \tag{2.47}$$

Note that

$$\begin{aligned}
\frac{\partial}{\partial t} \mathbf{C}^t(s) &= -\frac{\partial}{\partial s} \mathbf{C}^t(s) = -\frac{\partial}{\partial s} \mathbf{C}_r^t(s), \\
\frac{\partial}{\partial t} \mathbf{C}_r^t(s) &= \frac{\partial}{\partial t} \mathbf{C}^t(s) - \dot{\mathbf{C}}(t).
\end{aligned} \tag{2.48}$$

From $(2.18)_1$ and (2.40), we can write the first law as

$$\dot{\psi} + D = e\dot{\kappa} + \mathbf{D} \odot \dot{\mathbf{C}}. \tag{2.49}$$

By virtue of the requirement that D be non-negative (see (2.4)), it will always be true that

$$\dot{\psi} \leq \dot{W} = e\dot{\kappa} + \mathbf{D} \odot \dot{\mathbf{C}}. \tag{2.50}$$

Referring to the abstract formulation of thermodynamics already cited (see Ref. 3, for example), we see that (2.50) is the fundamental requirement for a free energy (if

1 it is differentiable with respect to time). The free energy must also be a non-negative
2 quantity.

3 From (2.49), we see that if $\dot{\mathbf{C}}(s) = \mathbf{0}, s \in [t, \infty)$ (which is true if $\dot{\mathbf{\Lambda}}(s) = \mathbf{0}, s \in$
4 $[t, \infty)$) and $\dot{\kappa}(s) = 0, s \in [t, \infty)$, then $\dot{\psi}(s) \leq 0, \forall\, s \geq t$. Thus, over this interval,
5 $\psi(s)$ is non-increasing if the independent field variables are constant in time. It
6 follows that[4]

$$\tilde{\psi}(\kappa^t, \kappa(t), \mathbf{C}^t, \mathbf{C}(t)) = \psi(t) \geq \phi(t) = \widetilde{\phi}(\kappa(t), \mathbf{C}(t)), \tag{2.51}$$

8 where $\widetilde{\phi}(\kappa(t), \mathbf{C}(t))$ is the equilibrium form of ψ, or this functional for constant
9 histories, specifically those given by $\kappa^t(s) = \kappa(t), \mathbf{C}^t(s) = \mathbf{C}(t), s \in \mathbb{R}^{++}$.

10 Using (2.39), we can write (2.34) in compact form as

$$\frac{\partial}{\partial \mathbf{C}(t)} \tilde{\psi}(\kappa^t, \kappa(t), \mathbf{C}^t, \mathbf{C}(t)) = \tilde{\mathbf{D}}(\kappa^t, \kappa(t), \mathbf{C}^t, \mathbf{C}(t)) = \mathbf{D}(t). \tag{2.52}$$

2.2. *Requirements for a free energy*

13 Let us gather together the properties which must be associated with a functional
14 $\tilde{\psi}$ if it is to be a free energy (see, for example, Refs. 9 and 3 in relation to the
15 corresponding properties for simple materials).

16 (P1) We have

$$e(t) = \frac{\partial \psi}{\partial \kappa(t)}, \quad \mathbf{D}(t) = \frac{\partial \psi}{\partial \mathbf{C}(t)}, \tag{2.53}$$

18 which are (2.33) and (2.52). Then, from (2.37),

$$\frac{\delta \psi}{\delta \mathbf{\Lambda}(t)} = \mathbf{\Sigma}(t). \tag{2.54}$$

20 (P2) Also,

$$\tilde{\psi}(\kappa^t, \kappa(t), \mathbf{C}^t, \mathbf{C}(t)) \geq \widetilde{\phi}(\kappa(t), \mathbf{C}(t)), \tag{2.55}$$

22 which is (2.51).

23 (P3) $\qquad \dfrac{d}{dt} \tilde{\psi}(\kappa^t, \kappa(t), \mathbf{C}^t, \mathbf{C}(t)) \leq e(t)\dot{\kappa}(t) + \mathbf{D}(t) \odot \dot{\mathbf{C}}(t),$ $\qquad$ (2.56)
24 which is the second law. This is (2.50).

25 These may be referred to as the non-local Graffi conditions.

26 A substantial advantage of the compact notation, which emerges below, is that
27 it exposes the fact that the non-local theory is closely analogous to the local tensor
28 case. This applies to all aspects of the theory, including procedures for constructing
29 free energy functionals. Note in particular that the non-local Graffi conditions are
30 identical to the local conditions, except for (2.54) which connects the theory to the
31 quantity $\mathbf{\Sigma}(t)$ (see Remark 2.2).

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1   ### 2.3. *Example*: *Continuum mechanics*

2   It is of interest to explore an important example of the formulae developed in this
3   section, namely the case of nonlinear mechanics under isothermal conditions. Also,
4   we bring the alternative approach based on (2.13) (or (2.9)) into the discussion.

5   Consider (2.13) in the isothermal case. The quantity κ is constant and $\mathcal{A}_{\text{en}}^i$
6   vanishes. Also, $\mathcal{P}_n^i$ in (2.9)$_2$ is zero. We take ρ to be the (time-independent) density
7   in the reference configuration. Let us absorb the constant (in time) factor ρ/κ into
8   ψ and D. This means that ψ is now the Helmholtz free energy (see (2.5)) per unit
9   volume. Equation (2.13) becomes

10
$$\dot{\psi} + D = \mathcal{P}_m^i = \dot{W}(t). \tag{2.57}$$

11   Let

12
$$\boldsymbol{\Lambda}(s) = \mathbf{F}(s), \quad s \in \mathbb{R}, \tag{2.58}$$

13   where $\mathbf{F}(\mathbf{X}, s)$ is the material gradient of deformation (see (2.19)) and $\mathbf{X}$ is the
14   position vector in material coordinates. Thus, in a given basis, the components of
15   $\mathbf{F}$ are

16
$$F_{ij}(t) = \frac{\partial x_i}{\partial X_j} = \frac{\partial \chi_i(\mathbf{X}, t)}{\partial X_i}, \quad i, j = 1, 2, 3, \tag{2.59}$$

17   where

18
$$x_i = \chi_i(\mathbf{X}, t), \quad i = 1, 2, 3, \tag{2.60}$$

are the current position coordinates. Then, (2.30) becomes

$$\psi(t) = \tilde{\psi}(\mathbf{X}, \mathbf{F}^t(\mathbf{X}, \cdot), \mathbf{F}(\mathbf{X}, t), \nabla \mathbf{F}^t(\mathbf{X}, \cdot), \nabla \mathbf{F}(\mathbf{X}, t),$$
$$\nabla^2 \mathbf{F}^t(\mathbf{X}, \cdot), \nabla^2 \mathbf{F}(\mathbf{X}, t), \dots, \nabla^n \mathbf{F}^t(\mathbf{X}, \cdot), \nabla^n \mathbf{F}(\mathbf{X}, t)). \tag{2.61}$$

Also, (2.32) takes the form

$$\partial_t \psi(t) = \frac{\partial \psi(t)}{\partial \mathbf{F}(t)} \cdot \dot{\mathbf{F}}(t) + \frac{\partial \psi(t)}{\partial \nabla \mathbf{F}(t)} \cdot \nabla \dot{\mathbf{F}}(t) + \frac{\partial \psi(t)}{\partial \nabla^2 \mathbf{F}} \cdot \nabla^2 \dot{\mathbf{F}}(t)$$
$$+ \cdots + \frac{\partial \psi(t)}{\partial \nabla^n \mathbf{F}(t)} \cdot \nabla^n \dot{\mathbf{F}}(t) = \dot{W}(t). \tag{2.62}$$

19   The superimposed dot notation indicates the material time derivative. The final
20   relation of (2.62) is simply (2.36) in this context. Relation (2.37) yields

21
$$\mathbf{S}(t) = \frac{\delta \psi(t)}{\delta \mathbf{F}(t)}, \tag{2.63}$$

22   where $\mathbf{S}$ is the first Piola–Kirchhoff stress tensor, introduced in (2.19). The dynam-
23   ical equation of the material has the form

24
$$\rho \dot{\mathbf{v}} = \nabla \cdot \mathbf{S} + \rho \mathbf{b}, \quad \mathbf{v} = \frac{\partial}{\partial t} \mathbf{x}(\mathbf{X}, t), \quad \dot{\mathbf{v}} = \frac{\partial}{\partial t} \mathbf{v}(\mathbf{X}, t), \tag{2.64}$$

where the vector $\mathbf{b}$ represents the body forces per unit mass acting on the material. The quantity $\nabla \cdot \mathbf{S}$ is defined by the relation (see Ref. 30, for example)

$$\mathbf{a}(\nabla \cdot \mathbf{S}) = \nabla \cdot (\mathbf{S}^{\top}\mathbf{a}), \tag{2.65}$$

which holds for any constant vector $\mathbf{a}$. Taking the inner product of $(2.64)_1$ with $\mathbf{v}$, we obtain

$$\frac{d}{dt}T = \mathbf{v} \cdot (\nabla \cdot \mathbf{S}) + \rho\mathbf{b} \cdot \mathbf{v} = \nabla \cdot (\mathbf{S}^{\top}\mathbf{v}) - \mathbf{S} \cdot \dot{\mathbf{F}} + \rho\mathbf{b} \cdot \mathbf{v}, \tag{2.66}$$

where T is the kinetic energy per unit volume. Relation (2.65) has been used and a standard manipulation carried out to obtain the second term of the right-most form of (2.66). One can show, after some detailed algebra entirely analogous to that in the proof of Proposition 2.1, that (see (2.18))

$$\mathbf{S} \cdot \dot{\mathbf{F}} = \dot{W}(t) + \boldsymbol{\nabla} \cdot \Phi', \tag{2.67}$$

where the vector quantity Φ' takes the form (2.29) adapted for the present case. We can therefore write (2.66) as

$$\frac{d}{dt}T + \dot{W}(t) = \nabla \cdot [\mathbf{S}^{\top}\mathbf{v} - \Phi'] + \rho\mathbf{b} \cdot \mathbf{v}. \tag{2.68}$$

For all materials, both simple and non-simple, we have the relation

$$\frac{d}{dt}T + \mathcal{P}_m^i(t) = \mathcal{P}_m^e(t), \tag{2.69}$$

where $\mathcal{P}_m^i$ and $\mathcal{P}_m^e$ are the internal and external mechanical powers, respectively. For simple materials, it is always true that $\mathcal{P}_m^i = \mathbf{S} \cdot \dot{\mathbf{F}}$. Comparing (2.68) and (2.69), the natural identification is

$$\begin{aligned} \mathcal{P}_m^i(t) &= \dot{W}(t) = \partial_t\psi, \\ \mathcal{P}_m^e(t) &= \nabla \cdot [\mathbf{S}^{\top}\mathbf{v} - \Phi'] + \rho\mathbf{b} \cdot \mathbf{v}. \end{aligned} \tag{2.70}$$

The second relation is physically justifiable in that the body forces are clearly external and the divergence term leads to a surface integration, when we carry out spatial integrations over all or part of the material. The first relation is a consequence of this identification.

Thus, $(2.57)_2$ agrees with $(2.70)_1$.

2.4. *Decomposition of the free energy functional*

Returning to the general theory, we additively decompose, for simplicity, the free energy ψ into two parts

$$\tilde{\psi}(\kappa^t, \kappa(t), \mathbf{C}^t, \mathbf{C}(t)) = \tilde{\psi}_1(\kappa^t, \kappa(t)) + \tilde{\psi}_2(\mathbf{C}^t, \mathbf{C}(t)), \tag{2.71}$$

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where

$$\dot{\psi}_1(t) + D_1(t) = e(t)\dot{\kappa}(t), \quad D_1(t) \geq 0, \quad \psi_1(t) = \tilde{\psi}_1(\kappa^t, \kappa(t)) \geq \widetilde{\phi}_1(\kappa(t)), \quad (2.72)$$

the quantity $\widetilde{\phi}_1(\kappa(t))$ being the equilibrium form of $\tilde{\psi}_1(\kappa^t, \kappa(t))$. It follows from (2.49) that

$$\dot{\psi}_2 + D_2 = \mathbf{D} \odot \dot{\mathbf{C}}, \quad D_2 = D - D_1, \quad (2.73)$$

where it is assumed that $D_2 \geq 0$. The quantity $\tilde{\psi}_1$ is in effect a free energy for a simple material and will not be considered further. The properties (P1) and (P3) in Sec. 2.2, with the occurrences of $\kappa(t)$ and $e(t)$ omitted, hold for ψ_2. We also impose (P2) so that

$$\tilde{\psi}_2(\mathbf{C}^t, \mathbf{C}(t)) \geq \widetilde{\phi}_2(\mathbf{C}(t)). \quad (2.74)$$

The subscript 2 will now be dropped. The first law becomes (see (2.49))

$$\dot{\psi} + D = \mathbf{D} \odot \dot{\mathbf{C}}. \quad (2.75)$$

Also, we have

$$\mathbf{D} \odot \dot{\mathbf{C}} = \partial_t \psi = \dot{W}(t), \quad (2.76)$$

where $W(t)$, the generalized rate of work, is redefined to mean that relating only to ψ_2 in (2.71).

There is no strong physical motivation for this decomposition. Indeed, heat flow variables are retained in $\mathbf{C}$ and $\mathbf{D}$, as we see from (2.17). It is done for pragmatic reasons of simplicity, to allow subsequent developments to be phrased only in terms of $\mathbf{C}$ and $\mathbf{D}$, rather than continuing to include κ and e. For materials undisturbed in the distant past, we can integrate (2.75) to obtain

$$\psi(t) + \mathcal{D}(t) = W(t) = \int_{-\infty}^{t} \mathbf{D}(u) \odot \dot{\mathbf{C}}(u) \, du, \quad (2.77)$$

where $\mathcal{D}(t)$ is the total dissipation over the history of the motion.

3. A Quadratic Model for Free Energies

The simplest choice of ψ is obtained by means of a functional Taylor expansion of $\tilde{\psi}(\mathbf{C}^t, \mathbf{C}(t))$, stopping at the quadratic term.[25,3] This in fact yields a constitutive equation with linear memory terms, as presented in (3.11) below.[g] We put

$$\psi(t) = \widetilde{\phi}(\mathbf{C}(t)) + \frac{1}{2} \int_0^\infty \int_0^\infty \mathbf{C}_r^t(s) \odot \mathbb{K}(s, u) \mathbf{C}_r^t(u) \, ds \, du. \quad (3.1)$$

The operator $\mathbb{K} \in \text{Lin}(\mathcal{B})$ is at least positive semi-definite in the sense that the function $\mathbb{K}(\cdot, \cdot)$ is such that the integral is non-negative. This must be true in view

---

[g]For the case of a purely mechanical simple material, see, for example, Ref. 11.

1   of (2.74). It should be noted that (2.74) also excludes linear functionals in (3.1). It
2   is assumed that $\mathbb{K}(\cdot, \cdot) \in L^1(\mathbb{R}^+ \times \mathbb{R}^+)$ and, in particular,

$$
\lim_{s \to \infty} \mathbb{K}(s, u) = \mathbf{0}, \quad u \in \mathbb{R}^+. \tag{3.2}
$$

4   A similar relation holds for the limit of large u at fixed s. There is no loss of
5   generality in taking

$$
\mathbb{K}^\top(s, u) = \mathbb{K}(u, s), \tag{3.3}
$$

7   where the transpose refers to $\mathrm{Lin}(\mathcal{B})$. Any space dependence in $\mathbb{K}$ is neglected, so
8   that we are dealing with a homogeneous material.
9   Let us define $\mathbb{L} \in \mathrm{Lin}(\mathcal{B})$ by

$$
\mathbb{L}(s, u) = \mathbb{L}_\infty + \int_s^\infty \int_u^\infty \mathbb{K}(t_1, t_2) dt_1 \, dt_2, \quad \mathbb{L}_\infty = \mathbb{L}_\infty^\top, \tag{3.4}
$$

11   which gives

$$
\mathbb{L}_{12}(s, u) = \frac{\partial^2}{\partial s \partial u} \mathbb{L}(s, u) = \mathbb{K}(s, u), \quad \mathbb{L}^\top(s, u) = \mathbb{L}(u, s) \tag{3.5}
$$

13   and

$$
\lim_{s \to \infty} \mathbb{L}(s, u) = \mathbb{L}_\infty, \quad \lim_{s \to \infty} \frac{\partial}{\partial u} \mathbb{L}(s, u) = \mathbf{0}, \quad u \in \mathbb{R}^+, \tag{3.6}
$$

15   with similar limits at large u holding for fixed s. Also, from (3.2) and (3.4),

$$
\lim_{u \to \infty} \frac{\partial}{\partial u} \mathbb{L}(s, u) = \mathbf{0}, \ s \in \mathbb{R}^+, \quad \lim_{s \to \infty} \frac{\partial}{\partial s} \mathbb{L}(s, u) = \mathbf{0}, \ u \in \mathbb{R}^+. \tag{3.7}
$$

17   We write (3.1) as

$$
\psi(t) = \widetilde{\phi}(\mathbf{C}(t)) + \frac{1}{2} \int_0^\infty \int_0^\infty \mathbf{C}_r^t(s) \odot \mathbb{L}_{12}(s, u) \mathbf{C}_r^t(u) ds du. \tag{3.8}
$$

19   It is possible to develop the theory without restriction, for a general equilibrium
20   term ϕ. However, in order to achieve simplifications, we specialize here to the case
21   of a fully linear theory by taking

$$
\widetilde{\phi}(\mathbf{C}(t)) = \phi(t) = \frac{1}{2} \mathbf{C}(t) \odot \mathbb{L}_\infty \mathbf{C}(t) \geq 0. \tag{3.9}
$$

By partial integration, one can put (3.8) in the form

$$
\psi(t) = \frac{1}{2} \int_0^\infty \int_0^\infty \dot{\mathbf{C}}^t(s) \odot \mathbb{L}(s, u) \dot{\mathbf{C}}^t(u) ds du,
$$

$$
\dot{\mathbf{C}}^t(u) = \frac{\partial}{\partial t} \mathbf{C}^t(u) = -\frac{\partial}{\partial u} \mathbf{C}^t(u) = -\frac{\partial}{\partial u} \mathbf{C}_r^t(u). \tag{3.10}
$$

23   It follows that $\mathbb{L}$ must be a non-negative operator in the same sense as $\mathbb{K}$. Also,
24   from (3.9), we have $\mathbb{L}_\infty \geq \mathbf{0}$.

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3.1. *Constitutive relations*

Applying (2.52) to (3.8) yields, with the aid of (3.3),

$$\mathbf{D}(t) = \mathbb{L}_\infty \mathbf{C}(t) + \int_0^\infty \mathbb{L}'(u)\mathbf{C}_r^t(u)\,du$$

$$= \mathbb{L}_0 \mathbf{C}(t) + \int_0^\infty \mathbb{L}'(u)\mathbf{C}^t(u)\,du$$

$$= \int_0^\infty \mathbb{L}(u)\dot{\mathbf{C}}^t(u)\,du, \tag{3.11}$$

where $\mathbf{C}^t(0) = \mathbf{C}(t)$ and

$$\mathbb{L}(u) = \mathbb{L}(0, u), \quad \mathbb{L}_0 = \mathbb{L}(0), \quad \mathbb{L}(\infty) = \mathbb{L}_\infty, \quad \mathbb{L}'(\infty) = 0, \tag{3.12}$$

and the prime indicates differentiation with respect to the argument. Relations (3.6) have been used. It will be assumed that

$$\mathbb{L}(u) = \mathbb{L}^\top(u), \quad u \in \mathbb{R}^+. \tag{3.13}$$

From (2.42), (2.43) and (3.11)$_1$, we have the constitutive relation for $\boldsymbol{\Sigma}(t)$,

$$\boldsymbol{\Sigma}(t) = \boldsymbol{\nabla}_a \odot \mathbf{D}(t) = \boldsymbol{\nabla}_a \odot \mathbb{L}_\infty \mathbf{C}(t) + \int_0^\infty \boldsymbol{\nabla}_a \odot \mathbb{L}'(u)\mathbf{C}_r^t(u)\,du$$

$$= \boldsymbol{\nabla}_a \odot \mathbb{L}_\infty(\boldsymbol{\nabla}\boldsymbol{\Lambda}(t)) + \int_0^\infty \boldsymbol{\nabla}_a \odot \mathbb{L}'(u)(\boldsymbol{\nabla}\boldsymbol{\Lambda}_r^t(u))\,du. \tag{3.14}$$

Similar relations are derivable from (3.11)$_{2,3}$.

Consider the case where (2.38) and (2.39) reduce to $\mathbf{C}(t) = (\boldsymbol{\Lambda}(t), \boldsymbol{\nabla}\boldsymbol{\Lambda}(t))$ and $\mathbf{D}(t) = (\boldsymbol{\Sigma}_0(t), \boldsymbol{\Sigma}_1(t))$. More explicitly, we write

$$\mathbf{C}(t) = \boldsymbol{\nabla}\boldsymbol{\Lambda}(t) = \begin{pmatrix} \boldsymbol{\Lambda}(t) \\ \partial_1\boldsymbol{\Lambda}(t) \\ \partial_2\boldsymbol{\Lambda}(t) \\ \partial_3\boldsymbol{\Lambda}(t) \end{pmatrix}, \quad \mathbf{D}(t) = \begin{pmatrix} \boldsymbol{\Sigma}_0(t) \\ \boldsymbol{\Sigma}_{(1)1}(t) \\ \boldsymbol{\Sigma}_{(1)2}(t) \\ \boldsymbol{\Sigma}_{(1)3}(t) \end{pmatrix}, \tag{3.15}$$

where

$$\partial_i = \frac{\partial}{\partial x_i}, \quad i = 1, 2, 3. \tag{3.16}$$

Also

$$\boldsymbol{\nabla}_a\odot = (1 \quad -\partial_1 \quad -\partial_2 \quad -\partial_3), \tag{3.17}$$

yielding in particular (see (2.43))

$$\boldsymbol{\Sigma}(t) = \boldsymbol{\nabla}_a \odot \mathbf{D}(t) = \boldsymbol{\Sigma}_0(t) - \partial_1\boldsymbol{\Sigma}_{(1)1}(t) - \partial_2\boldsymbol{\Sigma}_{(1)2}(t) - \partial_3\boldsymbol{\Sigma}_{(1)3}(t). \tag{3.18}$$

The tensor $\mathbb{L}$ is representable as a 4×4 matrix of transformations in $\mathrm{Lin}(\Gamma)$. Assume that the individual components are scalars. In the case where it is diagonal, with

AQ: Please check the sentence "The tensor..." for sense.

scalar components $L_{kk}, k = 0, 1, 2, 3$, the integrand in the first line of $(3.14)_2$ is given by

$$L'_{00}(u)\mathbf{\Lambda}_r^t(u) - \sum_{i=1}^{3} L'_{ii}(u)\partial_i^2 \mathbf{\Lambda}_r^t(u). \tag{3.19}$$

If $L'_{11}(u) = L'_{22}(u) = L'_{33}(u)$, we see that this agrees with the form of the constitutive equation in Refs. 1–3. For those materials, the first term on the right of (3.14) vanishes.

3.2. *Alternative form of the free energy*

With the aid of (2.46), relation (3.8) can be written as

$$\psi(t) = S(t) + \frac{1}{2}\int_0^\infty \int_0^\infty \mathbf{C}^t(s) \odot \mathbb{L}_{12}(s, u)\mathbf{C}^t(u)\,ds\,du,$$
$$S(t) = \mathbf{D}(t) \odot \mathbf{C}(t) - \frac{1}{2}\mathbf{C}(t) \odot \mathbb{L}_0\mathbf{C}(t), \tag{3.20}$$

where $\mathbb{L}_0$ is defined by $(3.12)_2$. One can show that

$$\frac{\partial}{\partial \mathbf{C}(t)}S(t) = \mathbf{D}(t), \tag{3.21}$$

which demonstrates consistency with (2.52). A disadvantage of using the form (3.20) is that $S(t)$ is not a non-negative quantity.

3.3. *Dissipation rate*

The rate of dissipation D is given by (2.35). However, we can avoid functional derivatives by observing from (2.75) that D emerges from the differentiation of $\psi(t)$ with respect to t, excluding terms with $\dot{\mathbf{C}}(t)$. By differentiating $(3.10)_1$ and carrying out partial integrations, one obtains, with the aid of $(3.10)_2$–$(3.10)_4$,

$$D(t) = -\frac{1}{2}\int_0^\infty \int_0^\infty \dot{\mathbf{C}}^t(s) \odot [\mathbb{L}_1(s, u) + \mathbb{L}_2(s, u)]\dot{\mathbf{C}}^t(u)\,ds\,du, \tag{3.22}$$

when the terms proportional to $\dot{\mathbf{C}}(t)$ are omitted. With further partial integrations, we can also write D in the form

$$D(t) = -\frac{1}{2}\int_0^\infty \int_0^\infty \mathbf{C}_r^t(s) \odot [\mathbb{L}_{121}(s, u) + \mathbb{L}_{122}(s, u)]\mathbf{C}_r^t(u)\,ds\,du, \tag{3.23}$$

where $(3.10)_2$–$(3.10)_4$ have been used again.

Remark 3.1. Many of the results in this and later sections are entirely similar in a formal sense to corresponding formulae for simple materials, given in earlier papers by the authors and others, in particular Refs. 19, 11, 15, 25 and 3. For this reason, some detailed derivations are omitted. However, we seek to give, as complete and self-contained, a description as possible of the theory in this new context, without relying excessively on references to other work, so a certain amount of repetition is unavoidable.

AQ: The citation $(3.10)_2$-$(3.10)_4$ is okay. Please clarify.

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4. Constitutive Equations in the Frequency Domain

Let us now consider the frequency domain representation of (3.11). We write the Fourier transform of the causal function $\mathbb{L}'$ in (3.11) as

$$\mathbb{L}'_+(\omega) = \int_0^\infty \mathbb{L}'(s) e^{-i\omega s} ds = \mathbb{L}'_c(\omega) - i\mathbb{L}'_s(\omega). \tag{4.1}$$

Unless indicated otherwise, the frequency may be taken to be real. We have

$$\mathbb{L}'_c(\omega) = \frac{1}{2}(\mathbb{L}'_+(\omega) + \overline{\mathbb{L}'_+}(\omega)), \quad \mathbb{L}'_s(\omega) = -\frac{1}{2i}(\mathbb{L}'_+(\omega) - \overline{\mathbb{L}'_+}(\omega)) \tag{4.2}$$

and

$$\overline{\mathbb{L}'_+}(\omega) = \mathbb{L}'_+(-\omega), \quad \mathbb{L}'_c(-\omega) = \mathbb{L}'_c(\omega), \quad \mathbb{L}'_s(-\omega) = -\mathbb{L}'_s(\omega). \tag{4.3}$$

It is assumed that $\mathbb{L}' \in L^1(\mathbb{R}^+) \cap L^2(\mathbb{R}^+)$, giving $\mathbb{L}'_+ \in L^2(\mathbb{R})$. From (4.1), it follows that

$$\int_{-\infty}^\infty \frac{d}{ds}\mathbb{L}(|s|) e^{-i\omega s} ds = -2i\mathbb{L}'_s(\omega), \tag{4.4}$$

a result which will prove useful later.

If $\mathbf{\Lambda}^t(s)$ has sinusoidal behavior, then it is easily shown from (2.38) that $\mathbf{C}^t(s)$ will also have such behavior. Consider (2.75) for oscillatory histories of period T, where transient effects have died away. The functional $\tilde{\psi}$ will consist of oscillatory terms (see Sec. 10). Integrating (2.75) over a cycle, we obtain the following form of the second law

$$\int_t^{t+T} \mathbf{D}(s) \odot \dot{\mathbf{C}}(s) ds = \int_t^{t+T} D(s) ds \geq 0. \tag{4.5}$$

Using such histories, it can be demonstrated (see Ref. 19, for example) that (4.5) yields

$$\mathbb{L}_0 = \mathbb{L}_0^\top, \quad \mathbb{L}_\infty = \mathbb{L}_\infty^\top, \tag{4.6}$$

which are special cases of (3.13). Indeed, the second relation was assumed earlier in (3.4). It can also be shown that

$$\mathbb{L}'_s(\omega_0) < \mathbf{0}, \quad 0 < \omega_0 < \infty. \tag{4.7}$$

Relation (4.7) is in effect proved by (10.23) below, if reversible processes, for which the rate of dissipation vanishes, are excluded.

Remark 4.1. For materials with (2.20) as the power function, the inequality in (4.7) is reversed. This leads to a kernel function which tends to increase (similarly to the creep function in viscoelasticity) rather than decrease, as emerges from (4.13) below for the inequality (4.7). This is what follows from the form $\mathbf{\Sigma} \odot \dot{\mathbf{\Lambda}}$ in (2.18).

Note that

$$\mathbb{L}'_s(\omega) = \int_0^\infty \mathbb{L}'(s) \sin \omega s\, ds < \mathbf{0}, \tag{4.8}$$

by virtue of (4.7). The sine inversion formula gives

$$\mathbb{L}'(s) = \frac{2}{\pi} \int_0^\infty \mathbb{L}'_s(\omega) \sin \omega s\, d\omega. \tag{4.9}$$

Integration yields

$$\mathbb{L}(s) - \mathbb{L}_0 = \frac{2}{\pi} \int_0^\infty \frac{1 - \cos \omega s}{\omega} \mathbb{L}'_s(\omega)\, d\omega \le \mathbf{0}, \tag{4.10}$$

so that

$$\mathbb{L}(s) \le \mathbb{L}_0. \tag{4.11}$$

We divide by s and take the limit $s \to 0$ which is assumed to exist. This yields $\mathbb{L}'(0) \le \mathbf{0}$. The stronger relation

$$\mathbb{L}'(0) < \mathbf{0} \tag{4.12}$$

will be assumed. Taking the limit $s \to \infty$ in (4.10) gives with the aid of the Riemann–Lebesgue lemma

$$\mathbb{L}_0 - \mathbb{L}_\infty = -\frac{2}{\pi} \int_0^\infty \frac{\mathbb{L}'_s(\omega)}{\omega}\, d\omega > \mathbf{0}. \tag{4.13}$$

We shall further assume that

$$\mathbf{0} < \left| \int_0^\infty \widetilde{\mathbb{L}}(s)\, ds \right| < \infty, \quad \widetilde{\mathbb{L}}(s) = \mathbb{L}(s) - \mathbb{L}_\infty. \tag{4.14}$$

From (3.9) and (4.13), it follows that

$$\mathbb{L}_0 > \mathbf{0}, \quad \mathbb{L}_\infty > \mathbf{0}. \tag{4.15}$$

Near the origin in the frequency domain,

$$\mathbb{L}'_+(\omega) = \int_0^\infty \mathbb{L}'(s)\, ds - i\omega \int_0^\infty s\mathbb{L}'(s)\, ds + O(\omega^2)$$

$$= -\mathbb{L}_0 + \mathbb{L}_\infty + i\omega \int_0^\infty \widetilde{\mathbb{L}}(s)\, ds + O(\omega^2). \tag{4.16}$$

Thus,

$$\mathbb{L}'_c(0) = \mathbb{L}_\infty - \mathbb{L}_0, \quad \frac{\mathbb{L}'_s(\omega)}{\omega} \xrightarrow[\omega \to 0]{} -\int_0^\infty \widetilde{\mathbb{L}}(s)\, ds. \tag{4.17}$$

We will be considering frequency space quantities, defined by analytic continuation from integral definitions, as functions on the complex ω-plane, denoted by Ω,

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where

$$\Omega^+ = \{\omega \in \Omega \,|\, \mathrm{Im}\,\omega \in \mathbb{R}^+\},$$
$$\Omega^{(+)} = \{\omega \in \Omega \,|\, \mathrm{Im}\,\omega \in \mathbb{R}^{++}\}. \tag{4.18}$$

Similarly, Ω^- and $\Omega^{(-)}$ are the lower half-planes including and excluding the real axis, respectively. The quantity $\mathbb{L}'_+$ is analytic on $\Omega^{(-)}$. This property is extended, by assumption, to an open set containing Ω^-. It is further assumed for simplicity to be analytic at infinity.

The function $\mathbb{L}'_s$ has singularities in both $\Omega^{(+)}$ and $\Omega^{(-)}$ that are symmetric under reflection in the origin, by $(4.3)_3$. They are also symmetric under reflection in the real axis, by virtue of $(4.2)_2$ so that they are mirror images of each other also with respect to the imaginary axis. The function $\mathbb{L}'_s$ goes to zero at the origin and must also be analytic on the real axis, since $\mathbb{L}'_+$ has this property. It vanishes linearly at the origin by (4.14) and $(4.17)_2$.

A quantity central to our discussions, analogous to that originally considered for the mechanical case in Refs. 23 and 11, is defined by

$$\mathbb{H}(\omega) = -\omega \mathbb{L}'_s(\omega) \geq \mathbf{0}, \quad \omega \in \mathbb{R}. \tag{4.19}$$

It is a non-negative, even tensor function of the frequency, vanishing quadratically at the origin. It is a symmetric tensor in $\mathrm{Lin}(\mathcal{B})$. From the behavior of Fourier transforms at large ω, one finds that

$$i \lim_{\omega \to \infty} \omega \mathbb{L}'_+(\omega) = \lim_{\omega \to \infty} \omega \mathbb{L}'_s(\omega) = \mathbb{L}'(0) \tag{4.20}$$

yields

$$\mathbb{L}'(0) = -\mathbb{H}(\infty) \equiv -\mathbb{H}_\infty. \tag{4.21}$$

The symmetric quantity $\mathbb{H}_\infty$ is positive definite by virtue of (4.12).

4.1. *Frequency domain representation of the history*

Let $\mathbf{C}^t \in L^1(\mathbb{R}^+) \cap L^2(\mathbb{R}^+)$ and recall the assumption (2.47). The Fourier transform of $\mathbf{C}^t$ is given by

$$\mathbf{C}^t_+(\omega) = \int_0^\infty \mathbf{C}^t(u) e^{-i\omega u}\, du, \tag{4.22}$$

which is analytic on $\Omega^{(-)}$, since the integral exists and is uniquely differentiable in this region. The derivative of $\mathbf{C}^t_+$ with respect to t will be required. Assuming that $\mathbf{C}^t \in C^1(\mathbb{R}^+)$ we find, with the aid of (2.45) and a partial integration, that

$$\frac{d}{dt}\mathbf{C}^t_+(\omega) = -i\omega \mathbf{C}^t_+(\omega) + \mathbf{C}(t). \tag{4.23}$$

In this context, the property (4.20) of Fourier transforms yields

$$i \lim_{\omega \to \infty} \omega \mathbf{C}^t_+(\omega) = \lim_{\omega \to \infty} \omega \mathbf{C}^t_s(\omega) = \mathbf{C}(t). \tag{4.24}$$

The Fourier transform of $\mathbf{C}_r^t(s) = \mathbf{C}^t(s) - \mathbf{C}(t), s \in \mathbb{R}$ is given by[h]

$$\mathbf{C}_{r+}^t(\omega) = \mathbf{C}_+^t(\omega) - \frac{\mathbf{C}(t)}{i\omega^-}, \tag{4.25}$$

which, on noting (4.24), can be seen to behave as ω^{-2} at large frequencies. Also, with the aid of (4.23), we see that

$$\frac{d}{dt}\mathbf{C}_+^t(\omega) = -i\omega\mathbf{C}_{r+}^t(\omega), \quad \frac{d}{dt}\mathbf{C}_{r+}^t(\omega) = -i\omega\mathbf{C}_{r+}^t(\omega) - \frac{\dot{\mathbf{C}}(t)}{i\omega^-}. \tag{4.26}$$

It is assumed that $\mathbf{C}_+^t(\omega)$ has no singularities on the real axis.

A continuation of the history $\mathbf{C}^t(u), u \in \mathbb{R}^+$ is $\mathbf{C}^t(u), u \in \mathbb{R}^{--}$. Its Fourier transform is given by

$$\mathbf{C}_-^t(\omega) = \int_{-\infty}^0 \mathbf{C}^t(u)e^{-i\omega u}du \tag{4.27}$$

and, instead of (4.24), we have

$$i \lim_{\omega \to \infty} \omega\mathbf{C}_-^t(\omega) = -\mathbf{C}(t^+), \tag{4.28}$$

which differs from $\mathbf{C}(t)$ if there is a discontinuity between the history and continuation (see (8.45)). Corresponding to (4.25), we have

$$\mathbf{C}_{r-}^t(\omega) = \mathbf{C}_-^t(\omega) + \frac{\mathbf{C}(t)}{i\omega^+} \tag{4.29}$$

and, for $\omega \to \infty$,

$$\mathbf{C}_{r-}^t(\omega) \approx \frac{\mathbf{C}(t) - \mathbf{C}(t^+)}{i\omega^+}. \tag{4.30}$$

Thus, if there is a discontinuity between the history and continuation, $\mathbf{C}_{r-}^t(\omega)$ behaves as ω^{-1} at large frequencies. If there is no such discontinuity, then $\mathbf{C}_{r-}^t(\omega)$ behaves as ω^{-2} at large ω.

The Fourier transform over $\mathbb{R}$ is

$$\mathbf{C}_F^t(\omega) = \int_{-\infty}^\infty \mathbf{C}^t(u)e^{-i\omega u}du = \mathbf{C}_+^t(\omega) + \mathbf{C}_-^t(\omega) \tag{4.31}$$

and

$$\mathbf{C}_{rF}^t(\omega) = \mathbf{C}_{r+}^t(\omega) + \mathbf{C}_{r-}^t(\omega) = \mathbf{C}_F^t(\omega) + i\mathbf{C}(t)\left\{\frac{1}{\omega^-} - \frac{1}{\omega^+}\right\}, \tag{4.32}$$

if there is no discontinuity between history and continuation. The last term on the right can be shown to be proportional to the singular function $\delta(\omega)$.

---

[h]These relations are discussed more fully in Refs. 24, 25, 3 and earlier work. In particular, we note here that ω^- can be thought of as $\omega - i\alpha$, where α is a small real positive quantity. Similarly ω^+ corresponds to $\omega + i\alpha$. The limit $\alpha \to 0$ is taken only after any integrations on Ω have been carried out. This is a device which avoids the use of distributions in various contexts. When $\omega^\pm$ is in the numerator of an expression, the superscript is irrelevant and may be dropped. The choice of $\omega^\mp$ in (4.25), (4.29) and (4.33) ensures that the singularity is in the correct half-plane.

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For small frequencies, we have

$$\mathbf{C}_{r\pm}^t(\omega) \approx \pm \frac{\mathbf{C}_r^t(\pm\infty)}{i\omega^{\mp}}, \tag{4.33}$$

which can be shown by considering Fourier transforms of functions that do not vanish at infinity and therefore do not belong to $L^1(\mathbb{R}^+) \cap L^2(\mathbb{R}^+)$.

4.2. *Constitutive equations in terms of frequency domain quantities*

Applying Parseval's formula to $(3.11)_1$ and $(3.11)_2$ by extending the range of integration to $\mathbb{R}$, one obtains

$$\mathbf{D}(t) = \mathbb{L}_\infty \mathbf{C}(t) + \frac{1}{2\pi}\int_{-\infty}^{\infty}\overline{\mathbb{L}_+'}(\omega)\mathbf{C}_{r+}^t(\omega)d\omega$$

$$= \mathbb{L}_0 \mathbf{C}(t) + \frac{1}{2\pi}\int_{-\infty}^{\infty}\overline{\mathbb{L}_+'}(\omega)\mathbf{C}_+^t(\omega)d\omega. \tag{4.34}$$

If we replace $\overline{\mathbb{L}_+'}(\omega)$ by $[\overline{\mathbb{L}_+'}(\omega) + \mathbb{F}(\omega)]$ where $\mathbb{F}(\omega)$ is analytic on Ω^- and is a constant or zero at infinity, $(4.34)_1$ still holds. This follows by a simple application of Cauchy's theorem over Ω^-, using the behavior of $\mathbf{C}_{r+}^t$ at large ω. In particular, we have

$$\mathbf{D}(t) = \mathbb{L}_\infty \mathbf{C}(t) + \frac{1}{2\pi}\int_{-\infty}^{\infty}[\overline{\mathbb{L}_+'}(\omega) + \lambda\mathbb{L}_+'(\omega)]\mathbf{C}_{r+}^t(\omega)d\omega, \tag{4.35}$$

where λ is any complex constant. Choosing $\lambda = -1$ yields

$$\mathbf{D}(t) = \mathbb{L}_\infty \mathbf{C}(t) + \frac{i}{\pi}\int_{-\infty}^{\infty}\mathbb{L}_s'(\omega)\mathbf{C}_{r+}^t(\omega)d\omega$$

$$= \mathbb{L}_\infty \mathbf{C}(t) - \frac{i}{\pi}\int_{-\infty}^{\infty}\frac{\mathbb{H}(\omega)}{\omega}\mathbf{C}_{r+}^t(\omega)d\omega, \tag{4.36}$$

where (4.19) has been used. Similarly, we find that

$$\mathbf{D}(t) = \mathbb{L}_0 \mathbf{C}(t) - \frac{i}{\pi}\int_{-\infty}^{\infty}\frac{\mathbb{H}(\omega)}{\omega}\mathbf{C}_+^t(\omega)d\omega, \tag{4.37}$$

with the aid of relation (4.13) rewritten so that the integration is over $\mathbb{R}$. Relation (4.37) was first given for viscoelastic materials in Ref. 16.

5. Minimal States

In the classical approach to materials with memory, the state is identified with the history of the independent variables. Noll's characterization of state,[34] discussed for the case of linear viscoelasticity in Ref. 9, is an interesting alternative. He takes the material response as the basis for the definition of state: if an arbitrary process, acting on different given histories up to time t, leads to the same response

1 of the material after time t, then the given histories are equivalent and the state
2 is represented as the class of all these equivalent histories. We shall refer to it as
3 the *minimal state*.[17] A minimal state is in effect an equivalence class under this
4 definition. The idea has been applied to linear materials in Refs. 28, 8, 9, 11, 17,
5 24, 25 and other more recent work, in particular Ref. 3.

Two states $(\mathbf{C}_1^t, \mathbf{C}_1(t))$, $(\mathbf{C}_2^t, \mathbf{C}_2(t))$ are equivalent, or determine the same minimal state if

$$
\begin{aligned}
\mathbf{D}_1(t+s) &= \mathbf{D}_2(t+s), \\
\mathbf{C}_1(t+s) &= \mathbf{C}_2(t+s) + \mathbf{C}_c, \quad s \geq 0,
\end{aligned}
\tag{5.1}
$$

where $\mathbf{D}_1, \mathbf{D}_2$ are the dependent variables corresponding to the states $(\mathbf{C}_1^t, \mathbf{C}_1(t)), (\mathbf{C}_2^t, \mathbf{C}_2(t))$. In the second relation of (5.1), $\mathbf{C}_c$ is a constant (in time) member of $\mathcal{B}$. This is the condition that the process $\dot{\mathbf{C}}_i(t+s), i = 1, 2, s \in \mathbb{R}^+$ acting on each is the same. Note that it follows from $(5.1)_1$ that $\mathbf{\Sigma}^{(1)} = \mathbf{\Sigma}^{(2)}$ where $\mathbf{\Sigma}^{(1)}$ and $\mathbf{\Sigma}^{(2)}$ are the dependent quantities (for example, stress tensors in mechanics) defined by (2.25). We have used superscripts to avoid confusion with $\mathbf{\Sigma}_1$ and $\mathbf{\Sigma}_2$ etc. in (2.24). Also, referring to $(3.11)_2$, we see that $(5.1)_1$ is true if for $s \geq 0$,

$$
\mathbb{L}_0 \mathbf{C}_1(t+s) + \int_0^\infty \mathbb{L}'(u) \mathbf{C}_1^{t+s}(u)\, du = \mathbb{L}_0 \mathbf{C}_2(t+s) + \int_0^\infty \mathbb{L}'(u) \mathbf{C}_2^{t+s}(u)\, du.
\tag{5.2}
$$

6 Let

7
$$
\mathbf{C}_d(t) = \mathbf{C}_1(t) - \mathbf{C}_2(t), \quad \mathbf{C}_d^t(s) = \mathbf{C}_1^t(s) - \mathbf{C}_2^t(s).
\tag{5.3}
$$

8 Then (5.2) with $(5.1)_2$ yield

9
$$
\mathbb{L}(s)\mathbf{C}_c + \mathbf{I}(s, \mathbf{C}_d^t) = \mathbf{0}, \quad \mathbf{I}(s, \mathbf{C}_d^t) = \int_0^\infty \mathbb{L}'(u+s) \mathbf{C}_d^t(u)\, du.
\tag{5.4}
$$

10 By virtue of $(3.12)_4$, we have

11
$$
\lim_{s \to \infty} \mathbf{I}(s, \mathbf{C}_d^t) = \mathbf{0}.
\tag{5.5}
$$

12 Thus, assuming that $\mathbb{L}(\infty)$ is non-zero and invertible, then $\mathbf{C}_c$ must vanish and the
13 conditions of equivalence can be stated as

14
$$
\mathbf{C}_d(t+s) = \mathbf{0}, \quad \mathbf{I}(s, \mathbf{C}_d^t) = \mathbf{0}, \quad s \in \mathbb{R}^+.
\tag{5.6}
$$

15 Observe that, in the theory under consideration, with linear constitutive rela-
16 tions, the states $(\mathbf{C}_1^t, \mathbf{C}_1(t))$ and $(\mathbf{C}_2^t, \mathbf{C}_2(t))$ are equivalent if and only if $(\mathbf{C}_d^t, \mathbf{C}_d(t))$
17 is equivalent to the zero state, consisting of a zero history and zero current value.

18 A functional of $(\mathbf{C}^t, \mathbf{C}(t))$ or $(\mathbf{C}_r^t, \mathbf{C}(t))$ which yields the same value for all
19 members of the same minimal state will be referred to as a functional of the minimal
20 state or as a minimal state variable. Note that, by virtue of (5.6), $\mathbf{I}(s, \mathbf{C}^t)$ has this
21 property.

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Let us introduce the notation

$$\langle \mathbf{C}_1^t, \mathbf{C}_2^t \rangle = \int_0^\infty \int_0^\infty \mathbf{C}_1^t(s_1) \odot \mathbb{L}_{12}(s_1, s_2) \mathbf{C}_2^t(s_2) \, ds_1 \, ds_2. \tag{5.7}$$

We have

$$\langle \mathbf{C}_1^t, \mathbf{C}_2^t \rangle = \langle \mathbf{C}_2^t, \mathbf{C}_1^t \rangle \tag{5.8}$$

by virtue of (3.3).

Let $(\mathbf{C}_1^t, \mathbf{C}(t))$ and $(\mathbf{C}_2^t, \mathbf{C}(t))$ be any equivalent states. Then, a free energy is a functional of the minimal state if

$$\psi(\mathbf{C}_1^t, \mathbf{C}(t)) = \psi(\mathbf{C}_2^t, \mathbf{C}(t)). \tag{5.9}$$

It is not necessary that a free energy should have this property, though it holds for some of the free energies discussed in later sections. Notably, this is true for the minimum free energy, as can be shown in great generality, but also in other cases. It follows from (3.20) and (5.9) that

$$\langle \mathbf{C}_1^t, \mathbf{C}_1^t \rangle = \langle \mathbf{C}_2^t, \mathbf{C}_2^t \rangle. \tag{5.10}$$

Note that if $\mathbf{C}_d^t$ is equivalent to the zero history, then, by (5.9),

$$\psi(\mathbf{C}_d^t, \mathbf{0}) = 0, \tag{5.11}$$

or

$$\langle \mathbf{C}_d^t, \mathbf{C}_d^t \rangle = 0. \tag{5.12}$$

If the material has minimal states that are non-singleton, then (5.12) implies that the quadratic function with kernel $\mathbb{L}_{12}$ is positive semi-definite. If the material has singleton minimal states, then this quadratic functional is positive definite.

Both (5.10) and (5.12) must hold if the two states are equivalent and if the free energy is a functional of the minimal state. This implies that

$$\langle \mathbf{C}_1^t, \mathbf{C}_1^t \rangle = \langle \mathbf{C}_2^t, \mathbf{C}_2^t \rangle = \langle \mathbf{C}_1^t, \mathbf{C}_2^t \rangle, \tag{5.13}$$

which conversely implies (5.9).

6. The Generalized Work Function

Relation $(3.11)_3$ can be put in the form

$$\mathbf{D}(t) = \int_{-\infty}^t \mathbb{L}(t - s) \dot{\mathbf{C}}(s) \, ds, \tag{6.1}$$

by a change of variable. Substituting (6.1) into $(2.77)_2$ gives

$$W(t) = \int_{-\infty}^t du \int_{-\infty}^u ds \, \mathbb{L}(u - s) \dot{\mathbf{C}}(s) \odot \dot{\mathbf{C}}(u). \tag{6.2}$$

Consider the identities

$$\int_{-\infty}^{t} du \int_{-\infty}^{u} ds f(u,s) = \int_{-\infty}^{t} ds \int_{s}^{t} du f(u,s) = \int_{-\infty}^{t} du \int_{u}^{t} ds f(s,u). \qquad (6.3)$$

The first follows from the geometry of integration in $\mathbb{R}^2$, the second by renaming integration variables. It follows that

$$\int_{-\infty}^{t} du \int_{-\infty}^{u} ds f(u,s) = \frac{1}{2} \int_{-\infty}^{t} du \int_{-\infty}^{t} ds f_S(s,u)$$

$$f_S(u,s) = \begin{cases} f(u,s), & u \geq s, \\ f(s,u), & s > u. \end{cases} \qquad (6.4)$$

Applying this general identity to (6.2) and recalling (3.13), we obtain

$$W(t) = \frac{1}{2} \int_{-\infty}^{t} \int_{-\infty}^{t} \mathbb{L}(|u-s|)\dot{\mathbf{C}}(s) \odot \dot{\mathbf{C}}(u)\,ds\,du$$

$$= \frac{1}{2} \int_{-\infty}^{t} \int_{-\infty}^{t} \mathbb{L}(|u-s|)\frac{\partial}{\partial s}[\mathbf{C}(s) - \mathbf{C}(t)] \odot \frac{\partial}{\partial u}[\mathbf{C}(u) - \mathbf{C}(t)]\,ds\,du. \qquad (6.5)$$

Two partial integrations give

$$W(t) = \phi(t) + \frac{1}{2}\int_{-\infty}^{t}\int_{-\infty}^{t} \mathbb{L}_{12}(|u-s|)[\mathbf{C}(u) - \mathbf{C}(t)] \odot [\mathbf{C}(s) - \mathbf{C}(t)]\,ds\,du$$

$$= \phi(t) + \frac{1}{2}\int_{0}^{\infty}\int_{0}^{\infty} \mathbb{L}_{12}(|u-s|)\mathbf{C}_r^t(u) \odot \mathbf{C}_r^t(s)\,ds\,du$$

$$= \phi(t) + \frac{1}{2}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} \mathbb{L}_{12}(|u-s|)\mathbf{C}_r^t(u) \odot \mathbf{C}_r^t(s)\,ds\,du, \qquad (6.6)$$

where $\phi(t)$ is defined by (3.9) and where the last relation presumes that $\mathbf{C}_r^t(u)$ vanishes for $u < 0$. Let

$$\mathbf{V}^{(t)}(s) = \int_{-\infty}^{\infty} \frac{\partial}{\partial s}\mathbb{L}(|s-u|)\mathbf{C}_r^t(u)\,du$$

$$= \int_{-\infty}^{\infty} \mathbb{L}_1(|s-u|)\mathbf{C}_r^t(u)\,du = -\int_{-\infty}^{\infty} \mathbb{L}_2(|s-u|)\mathbf{C}_r^t(u)\,du, \qquad (6.7)$$

so that

$$\frac{\partial}{\partial s}\mathbf{V}^{(t)}(s) = -\int_{-\infty}^{\infty} \mathbb{L}_{12}(|s-u|)\mathbf{C}_r^t(u)\,du. \qquad (6.8)$$

The Fourier transform of $\mathbf{V}^{(t)}(s)$ is, by virtue of the convolution theorem and (4.4),

$$\mathbf{V}_F^{(t)}(\omega) = -2i\mathbb{L}_s'(\omega)\mathbf{C}_{r+}^t(\omega), \qquad (6.9)$$

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which yields

$$\frac{1}{2}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\mathbb{L}_{12}(|s-u|)\mathbf{C}_r^t(u)\odot\mathbf{C}_r^t(s)\,du\,ds$$

$$=-\frac{1}{2}\int_{-\infty}^{\infty}\frac{\partial}{\partial s}\mathbf{V}^{(t)}(s)\odot\mathbf{C}_r^t(s)\,ds$$

$$=-\frac{1}{2\pi}\int_{-\infty}^{\infty}\omega\mathbb{L}_s'(\omega)\mathbf{C}_{r+}^t(\omega)\odot\overline{\mathbf{C}_{r+}^t}(\omega)\,d\omega, \tag{6.10}$$

1   where the last form is obtained using Parseval's formula. Thus, we obtain

2   $$W(t)=\phi(t)+\frac{1}{2\pi}\int_{-\infty}^{\infty}\overline{\mathbf{C}_{r+}^t}(\omega)\odot\mathbb{H}(\omega)\mathbf{C}_{r+}^t(\omega)\,d\omega, \tag{6.11}$$

3   where (4.19) has been used. The right-hand side is manifestly non-negative.

4   We will also need $W(\infty)$ below. It is assumed that $\mathbf{C}(s)$ vanishes as $s\to\pm\infty$,

5   which is stronger than (2.47). Let us write $(6.5)_1$ for $t=\infty$ as

6   $$W(\infty)=\frac{1}{2}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\mathbb{L}(|u-s|)\dot{\mathbf{C}}(s)\odot\dot{\mathbf{C}}(u)\,ds\,du \tag{6.12}$$

7   and write $(6.5)_2$ with the integrations over $\mathbb{R}$ and where the time in $\mathbf{C}(t)$ is arbitrary.

8   The partial integrations to yield (6.6) can be carried out, where the terms at $\pm\infty$

9   cancel. Thus, $(6.6)_3$ takes the form

10   $$W(\infty)=\frac{1}{2}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\mathbb{L}_{12}(|u-s|)\mathbf{C}_r^t(u)\odot\mathbf{C}_r^t(s)\,ds\,du, \tag{6.13}$$

11   where the variable t can take any value. In later applications, it will be taken to be

12   the current time. The quantities $\mathbf{C}_r^t(u)$ and $\mathbf{C}_r^t(s)$ are not now assumed to vanish

13   for $u,s<0$. Applying Parseval's formula, as in (6.10), we obtain

14   $$W(\infty)=\frac{1}{2\pi}\int_{-\infty}^{\infty}\mathbf{C}_{rF}^t(\omega)\odot\mathbb{H}(\omega)\overline{\mathbf{C}_{rF}^t}(\omega)\,d\omega, \tag{6.14}$$

15   where $\mathbf{C}_{rF}^t$ is defined by (4.32).

Equations (6.6) and (6.11) are expressed in terms of relative histories. We can write them in terms of histories, as follows:

$$W(t)=S(t)+\frac{1}{2}\int_0^{\infty}\int_0^{\infty}\mathbb{L}_{12}(|s-u|)\mathbf{C}^t(u)\odot\mathbf{C}^t(s)\,du\,ds$$

$$=S(t)+\frac{1}{2}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\mathbb{L}_{12}(|s-u|)\mathbf{C}^t(u)\odot\mathbf{C}^t(s)\,du\,ds$$

$$=S(t)+\frac{1}{2\pi}\int_{-\infty}^{\infty}\overline{\mathbf{C}_+^t}(\omega)\odot\mathbb{H}(\omega)\mathbf{C}_+^t(\omega)\,d\omega, \tag{6.15}$$

16   where in $(6.15)_2$ and $(6.15)_3$ it is assumed that $\mathbf{C}^t(u)$ vanishes for $u<0$. Rela-

17   tion $(6.15)_1$ corresponds to $(3.20)_1$ and can be demonstrated in a similar manner.

Relation $(6.15)_3$ follows from (4.25), (6.11) and (4.37), with the aid of (4.13), or by applying Parseval's formula.

We see from $(6.6)_2$ that $W(t)$ can be cast in the form (3.8) by putting

$$\mathbb{L}_{12}(s, u) = \mathbb{L}_{12}(|s - u|). \tag{6.16}$$

Thus, $W(t)$ can be regarded as a free energy, but with zero dissipation, which is clear from (3.22) or indeed (2.76). Because of the vanishing dissipation, it must be the maximum free energy associated with the material or greater than this quantity, an observation which follows by integrating (2.75) over $(-\infty, t]$. Both of these situations can occur, depending on whether the minimal state is a singleton or not.[24,10,25,3] Clearly, of course, zero dissipation is non-physical for a material with memory, so the main interest of this quantity, in the context of free energies, is that it provides an upper bound for the physical free energy.

It can be proved[11,3] that the non-negative tensor $\mathbb{H}(\omega)$ can always be expressed as the product of two factors

$$\mathbb{H}(\omega) = \mathbb{H}_+(\omega)\mathbb{H}_-(\omega), \quad \mathbb{H}_\pm(\omega) = \mathbb{H}_\mp^*(\omega), \quad \omega \in \mathbb{R}, \tag{6.17}$$

where all the singularities of $\mathbb{H}_\pm(\omega)$ and all the zeros of $\det \mathbb{H}_\pm(\omega)$ are in $\Omega^{(\pm)}$, respectively. This factorization is fundamental to the determination of the minimum free energy in Sec. 8.

7. Special Forms of Free Energy

We present in this section functionals that are free energies provided certain assumptions on the relaxation function are valid.

In Secs. 7.1 and 7.2, we consider quadratic functionals which are free energies only for a sub-category (though an important one) of materials, namely those with the property

$$\mathbb{L}'(s) \le \mathbf{0}, \ \mathbb{L}''(s) \ge \mathbf{0}, \quad \forall\, s \in \mathbb{R}^+. \tag{7.1}$$

Remark 7.1. Note that the assumption $\mathbb{L}''(u) \ge \mathbf{0}, u \ge s$ implies $\mathbb{L}'(s) \le \mathbf{0}$. It implies $\mathbb{L}'(s) < \mathbf{0}$ if $\mathbb{L}''$ is non-zero on a set of finite measure with elements $u > s$. This in particular implies that $\mathbb{L}'(0) < \mathbf{0}$ except in trivial cases, as assumed in (4.12).

In Sec. 7.3, a stronger assumption must be made.

7.1. *The Graffi–Volterra free energy*

Let us first present the Graffi–Volterra functional[26,27,9,8,3]

$$\psi_G(t) = \phi(t) - \frac{1}{2}\int_0^\infty \mathbb{L}'(s)\mathbf{C}_r^t(s) \odot \mathbf{C}_r^t(s)\,ds$$

$$= S(t) - \frac{1}{2}\int_0^\infty \mathbb{L}'(s)\mathbf{C}^t(s) \odot \mathbf{C}^t(s)\,ds, \tag{7.2}$$

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where $\phi(t)$ and $S(t)$ are given by (3.9) and (3.20)$_2$, respectively. This is a free energy if (7.1)$_1$ holds. The rate of dissipation associated with ψ_G has the form

$$D_G(t) = \frac{1}{2} \int_0^\infty \mathbb{L}''(s) \mathbf{C}_r^t(s) \odot \mathbf{C}_r^t(s) ds, \tag{7.3}$$

which is non-negative under assumption (7.1)$_2$. This relation can be derived using (2.75) and (2.48). The properties in Sec. 2.2 are easily confirmed for ψ_G.

A special case of ψ_G is discussed in Refs. 1–3, where $\mathbb{L}(s)$ is a diagonal tensor. This also applies to the free energies introduced in Secs. 7.2, 7.3 and 8.

The functional ψ_G can be expressed in the form (3.8), with

$$\mathbb{L}(s,u) = \frac{1}{2}[\mathbb{L}(s)I_{[0,s)}(u) + \mathbb{L}(u)I_{[0,u)}(s)], \tag{7.4}$$

where I_S is the characteristic function of the set S, defined by

$$I_{[0,s)}(u) = \begin{cases} 1, & u \in [0,s), \\ 0, & u \notin [0,s). \end{cases} \tag{7.5}$$

We observe that $\mathbb{L}_{12}$ is unbounded.

The Graffi–Volterra free energy is a functional of the minimal state only if the material is such that the minimal states are singletons, in other words, if the minimal state is simply $(\mathbf{C}^t, \mathbf{C}(t))$. Indeed, if we have a history $\mathbf{C}_d^t \neq \mathbf{0}$, equivalent to the zero history, as given by (5.3), then (5.11) does not hold, because, from (7.1), we have

$$\int_0^\infty \mathbb{L}'(s) \mathbf{C}_d^t(s) \odot \mathbf{C}_d^t(s) ds \geq 0, \tag{7.6}$$

where equality occurs only in the case where $\mathbb{L}'(s)$ vanishes for $s \in \mathbb{R}^+$.

We have the following result.

Proposition 7.1. *If $\mathbb{L}_{12}(s_1, s_2)$ is a bounded, non-negative (positive semi-definite) tensor for all $s_1, s_2 \in \mathbb{R}^+$, then*

$$\psi(t) \leq \psi_G(t), \quad t \in \mathbb{R}, \tag{7.7}$$

where ψ is any functional of the form (3.20) and ψ_G is the Graffi–Volterra functional (7.2).

Proof. Consider the identity

$$\frac{1}{2} \int_0^\infty \int_0^\infty [\mathbf{C}^t(s_1) - \mathbf{C}^t(s_2)] \odot \mathbb{L}_{12}(s_1, s_2)[\mathbf{C}^t(s_1) - \mathbf{C}^t(s_2)] ds_1 ds_2$$

$$= \frac{1}{2} \int_0^\infty \int_0^\infty \mathbf{C}^t(s_1) \odot \mathbb{L}_{12}(s_1, s_2)\mathbf{C}^t(s_1) ds_1 ds_2$$

$$+ \frac{1}{2} \int_0^\infty \int_0^\infty \mathbf{C}^t(s_2) \odot \mathbb{L}_{12}(s_1, s_2) \mathbf{C}^t(s_2) ds_1 ds_2$$

$$- \int_0^\infty \int_0^\infty \mathbf{C}^t(s_1) \odot \mathbb{L}_{12}(s_1, s_2) \mathbf{C}^t(s_2) ds_1 ds_2. \tag{7.8}$$

The left-hand side is non-negative by virtue of the assumption on $\mathbb{L}_{12}(s_1, s_2)$. The first two terms on the right yield, after integration, the integral term in (7.2) and the last term is the memory integral in $(3.20)_1$, multiplied in both cases by a factor 2. Relation (7.7) follows immediately. □

Remark 7.2. It must be emphasized that the non-negativity we have assumed for $\mathbb{L}_{12}(s_1, s_2)$ is not necessary in general for ψ, given by (3.8), to be a free energy, though $\mathbb{L}_{12}$ must be a non-negative operator in the sense that the integral term in (3.8) must be non-negative.

7.2. *Single integral quadratic functionals of* $\mathbf{I}^t$

We now introduce a functional which is a free energy for materials with the property (7.1) and is a functional of the minimal state. These results were first reported in Ref. 10 and are also discussed in Ref. 3.

Consider the functional

$$\tilde{\psi}_F(\mathbf{I}^t) = \psi_F(t) = \phi(t) - \frac{1}{2} \int_0^\infty \mathbb{L}'^{-1}(\tau) \dot{\mathbf{I}}^t(\tau) \odot \dot{\mathbf{I}}^t(\tau) d\tau,$$

$$\dot{\mathbf{I}}^t(\tau) = \frac{\partial}{\partial \tau} \mathbf{I}^t(\tau), \quad \mathbf{I}^t(\tau) = \mathbf{I}(\tau, \mathbf{C}_r^t), \tag{7.9}$$

where $\mathbb{L}'^{-1}(\tau)$ is the inverse of the tensor $\mathbb{L}'(\tau)$ in the algebraic sense (treating them as matrices) and $\mathbf{I}(\cdot, \mathbf{C}_r^t)$ is defined by (5.4) with $\mathbf{C}_r^t$ replacing $\mathbf{C}_d^t$. The integral term in (7.9) is non-negative by virtue of (7.1). The tensor $(\mathbb{L}')^{-1}$ becomes singular at large τ, but it is clear from the representation (7.12) below that the integral exists.

The domain of definition of the functional $\tilde{\psi}_F$ will be denoted by

$$\mathcal{H}_F^*(\mathbb{R}^+) = \left\{ \mathbf{I}^t; \left| \int_0^\infty \mathbb{L}'^{-1}(\tau) \dot{\mathbf{I}}^t(\tau) \odot \dot{\mathbf{I}}^t(\tau) d\tau \right| < \infty \right\}. \tag{7.10}$$

This space is very much larger than the domain of definition of the Graffi–Volterra free energy, as we see for a kernel given by an exponential or a sum of exponentials. One can choose exponentially diverging histories such that the integral in (7.2) will diverge — because of the quadratic dependence on $\mathbf{C}_r^t$ — but for which $\mathbf{I}^t$ exists.

Note that the function $\tilde{\psi}_F$ can also be written in the form

$$\tilde{\psi}_F(\mathbf{I}^t) = S(t) - \frac{1}{2} \int_0^\infty \mathbb{L}'^{-1}(\tau) \dot{\mathbf{I}}(\tau, \mathbf{C}^t) \odot \dot{\mathbf{I}}(\tau, \mathbf{C}^t) d\tau,$$

$$\dot{\mathbf{I}}(\tau, \mathbf{C}^t) = \frac{\partial}{\partial \tau} \mathbf{I}(\tau, \mathbf{C}^t), \tag{7.11}$$

which is a special case of (3.20).

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We can write ψ_F, given by (7.9) explicitly, in terms of the relative history $\mathbf{C}_r^t$ as follows:

$$\psi_F(t) = \phi(t) + \frac{1}{2} \int_0^\infty \int_0^\infty \mathbb{L}_{12}(s_1, s_2) \mathbf{C}_r^t(s_1) \odot \mathbf{C}_r^t(s_2) ds_1 ds_2, \tag{7.12}$$

where

$$\mathbb{L}(s_1, s_2) = -\int_0^\infty \mathbb{L}'(\tau + s_2) \mathbb{L}'^{-1}(\tau) \mathbb{L}'(\tau + s_1) d\tau = \mathbb{L}(s_2, s_1). \tag{7.13}$$

One can derive an expression for the rate of dissipation using $(3.22)^1$ and, from the fact that it is non-negative, deduce that ψ_F is a free energy. A more direct demonstration of the fact that ψ_F is a free energy can also be given. From (2.46), (2.48), (5.4) and (7.9) we have that

$$\frac{d}{dt}\dot{\mathbf{I}}^t(\tau) = \mathbb{L}'(\tau)\dot{\mathbf{C}}(t) + \ddot{\mathbf{I}}^t(\tau), \tag{7.14}$$

so that

$$\frac{d}{dt}\psi_F(t) = \mathbf{D}(t) \odot \dot{\mathbf{C}}(t) - \int_0^\infty \mathbb{L}'^{-1}(\tau)\ddot{\mathbf{I}}^t(\tau) \odot \dot{\mathbf{I}}^t(\tau) d\tau$$

$$= \mathbf{D}(t) \odot \dot{\mathbf{C}}(t) + \frac{1}{2}\mathbb{L}'^{-1}(0)\dot{\mathbf{I}}^t(0) \odot \dot{\mathbf{I}}^t(0)$$

$$+ \frac{1}{2}\int_0^\infty \left[\frac{d}{d\tau}\mathbb{L}'^{-1}(\tau)\right] \dot{\mathbf{I}}^t(\tau) \odot \dot{\mathbf{I}}^t(\tau) d\tau, \tag{7.15}$$

using the fact that $\mathbf{I}^t(0)$ is the integral term in $(3.11)_1$. Thus, we have, from (2.75),

$$D_F(t) = -\frac{1}{2}\mathbb{L}'^{-1}(0)\dot{\mathbf{I}}^t(0) \odot \dot{\mathbf{I}}^t(0) - \frac{1}{2}\int_0^\infty \left[\frac{d}{d\tau}\mathbb{L}'^{-1}(\tau)\right] \dot{\mathbf{I}}^t(\tau) \odot \dot{\mathbf{I}}^t(\tau) d\tau \geq 0, \tag{7.16}$$

since

$$\frac{d}{d\tau}\mathbb{L}'^{-1}(\tau) = -\mathbb{L}'^{-1}(\tau)\mathbb{L}''(\tau)\mathbb{L}'^{-1}(\tau) \leq \mathbf{0}, \tag{7.17}$$

by virtue of (7.1). Note that

$$D_F(t) \geq -\frac{1}{2}\int_0^\infty \left[\frac{d}{d\tau}\mathbb{L}'^{-1}(\tau)\right] \dot{\mathbf{I}}^t(\tau) \odot \dot{\mathbf{I}}^t(\tau) d\tau \geq 0. \tag{7.18}$$

It is straightforward to confirm that ψ_F has the properties of a free energy listed in Sec. 2.2.

Let us assume further that there exists a non-negative $\alpha_1 \in \mathbb{R}^{++}$ such that

$$\mathbb{L}''(\tau) + \alpha_1 \mathbb{L}'(\tau) \geq \mathbf{0}, \quad \forall \tau \in \mathbb{R}^+. \tag{7.19}$$

This yields

$$\frac{d}{d\tau}\mathbb{L}'^{-1}(\tau) \leq \alpha_1 \mathbb{L}'^{-1}(\tau) \leq \mathbf{0}, \tag{7.20}$$

1   using (7.17) and, from (7.18),

$$D_F(t) \geq \alpha_1[\psi_F(t) - \phi(t)]. \tag{7.21}$$

3   One can write down a family of similar free energies using a simple generalization
4   of the above procedure.[10,3]

5   ### 7.3. *The Dill free energy*

The functional

$$\psi_{\text{Dill}}(t) = \phi(t) + \frac{1}{2} \int_0^\infty \int_0^\infty \mathbb{L}''(s_1 + s_2)\mathbf{C}_r^t(s_2) \odot \mathbf{C}_r^t(s_1)ds_1 ds_2$$

$$= S(t) + \frac{1}{2} \int_0^\infty \int_0^\infty \mathbb{L}''(s_1 + s_2)\mathbf{C}^t(s_2) \odot \mathbf{C}^t(s_1)ds_1 ds_2 \tag{7.22}$$

is a free energy with rate of dissipation given by

$$D_{\text{Dill}}(t) = - \int_0^\infty \int_0^\infty \mathbb{L}'''(s_1 + s_2)\mathbf{C}_r^t(s_2) \odot \mathbf{C}_r^t(s_1)ds_1 ds_2$$

$$= - \int_0^\infty \int_0^\infty \mathbb{L}'(s_1 + s_2)\dot{\mathbf{C}}^t(s_2) \odot \dot{\mathbf{C}}^t(s_1)ds_1 ds_2, \tag{7.23}$$

6   if and only if $\mathbb{L}$ is completely monotonic,[8] in other words, if it is differentiable to
7   any order and

$$(-1)^n\mathbb{L}^{(n)}(s) \geq \mathbf{0}, \quad s \in \mathbb{R}^+, \; n = 0, 1, 2, \ldots, \tag{7.24}$$

9   where the superscript indicates the nth derivative. Relations (7.22) and (7.23)
10   are special cases of (3.8), (3.20), (3.22) and (3.23). Note that in this case, $(3.5)_2$
11   implies (3.13), namely the symmetry of $\mathbb{L}$. This functional is also referred to as the
12   Staverman–Schwarzl free energy.[8]

13   The required properties listed in Sec. 2.2 are easily checked for ψ_{Dill}.
14   Because of the non-negativity of the tensor $\mathbb{L}''$, we have, from Proposition 7.1,
15   that

$$\psi_{\text{Dill}}(t) \leq \psi_G(t), \quad t \in \mathbb{R}. \tag{7.25}$$

17   The quantity ψ_{Dill} is a functional of the minimal state. Indeed, it can be shown
18   to obey (5.13).
19   Relation (7.24) will be true if and only if[8]

$$\mathbb{L}(s) = \int_0^\infty e^{-\alpha s}d\mathbb{K}(\alpha), \tag{7.26}$$

21   where $\mathbb{K} : [0, \infty) \mapsto \text{Lin}(\mathcal{B})$ is symmetric, bounded and non-decreasing.[i] Let us
assume that $\mathbb{K}$ is sectionally smooth, with a finite number of discontinuities, so

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[i]This quantity has no connection with the $\mathbb{K}$ in (3.1).

that

$$\mathbb{L}(s) = \int_0^\infty \mathbb{K}'(\alpha)e^{-\alpha s}d\alpha + \sum_{1=0}^n \mathbb{K}_i e^{-\alpha_i s}, \qquad (7.27)$$

where

$$\mathbb{K}_0 = \mathbb{K}(0), \quad \alpha_0 = 0, \qquad (7.28)$$

and $\alpha_i, i = 1, 2, \ldots, n$ are the points of discontinuity so that

$$\mathbb{K}_i = \mathbb{K}(\alpha_i^+) - \mathbb{K}(\alpha_i^-) > 0, \quad i = 1, 2, \ldots, n. \qquad (7.29)$$

The origin is in effect also a point of discontinuity if $\mathbb{K}(0) = \mathbb{K}(0^+)$ is non-zero, in that $\mathbb{K}(0^-)$ vanishes. Note that $\mathbb{K}_0 = \mathbb{L}(\infty)$.

Using (7.27) in $(7.22)_1$ and $(7.23)_1$ yields

$$\psi_{\text{Dill}}(t) = \phi(t) + \frac{1}{2}\int_0^\infty \alpha^2 \mathbf{C}_{rL}^t(\alpha) \odot \mathbb{K}'(\alpha)\mathbf{C}_{rL}^t(\alpha)d\alpha$$

$$+ \frac{1}{2}\sum_{i=1}^n \alpha_i^2 \mathbf{C}_{rL}^t(\alpha_i) \odot \mathbb{K}_i \mathbf{C}_{rL}^t(\alpha_i), \qquad (7.30)$$

where $\mathbf{C}_{rL}^t$ is the Laplace transform

$$\mathbf{C}_{rL}^t(\alpha) = \int_0^\infty e^{-\alpha s}\mathbf{C}_r^t(\alpha)d\alpha \qquad (7.31)$$

and

$$D_{\text{Dill}}(t) = \int_0^\infty \alpha^3 \mathbf{C}_{rL}^t(\alpha) \odot \mathbb{K}'(\alpha)\mathbf{C}_{rL}^t(\alpha)d\alpha + \sum_{i=1}^n \alpha_i^3 \mathbf{C}_{rL}^t(\alpha_i) \odot \mathbb{K}_i \mathbf{C}_{rL}^t(\alpha_i).$$

$$(7.32)$$

For this restricted class of materials, ψ_{Dill} is clearly a free energy in that it obeys all the criteria. The converse, proved in Ref. 8, is more difficult.

Note that

$$\mathbb{L}'_+(\omega) = -\int_0^\infty \frac{\alpha\mathbb{K}'(\alpha)}{\alpha + i\omega}d\alpha - \sum_{i=1}^n \frac{\alpha_i\mathbb{K}_i}{\alpha_i + i\omega},$$

$$\mathbb{L}''_+(\omega) = \int_0^\infty \frac{\alpha^2\mathbb{K}'(\alpha)}{\alpha + i\omega}d\alpha + \sum_{i=1}^n \frac{\alpha_i^2\mathbb{K}_i}{\alpha_i + i\omega} \qquad (7.33)$$

$$= i\omega\mathbb{L}'_+(\omega) + \int_0^\infty \alpha\mathbb{K}'(\alpha)d\alpha + \sum_{i=1}^n \alpha_i\mathbb{K}_i.$$

Also, using the inverse Fourier transform, we have

$$\mathbb{L}''(s) = \frac{1}{2\pi}\int_{-\infty}^\infty \mathbb{L}''_+(\omega)e^{i\omega s}d\omega. \qquad (7.34)$$

Substituting (7.34) for $\mathbb{L}''$ into (7.22), we obtain

$$\psi_{\text{Dill}}(t) = \phi(t) + \frac{1}{4\pi} \int_{-\infty}^{\infty} \overline{\mathbf{C}_{r+}^t}(\omega) \odot \mathbb{L}_+''(\omega) \overline{\mathbf{C}_{r+}^t}(\omega) d\omega$$

$$= \phi(t) + \frac{1}{4\pi} \int_{-\infty}^{\infty} \mathbf{C}_{r+}^t(\omega) \odot \overline{\mathbb{L}_+''}(\omega) \mathbf{C}_{r+}^t(\omega) d\omega$$

$$= \phi(t) + \frac{1}{4\pi} \int_{-\infty}^{\infty} \text{Re}\{\mathbf{C}_{r+}^t(\omega) \odot \overline{\mathbb{L}_+''}(\omega) \mathbf{C}_{r+}^t(\omega)\} d\omega, \qquad (7.35)$$

where the reality of ψ_{Dill} has been invoked in writing the second and third forms.

8. Derivation of the Form of the Minimum Free Energy

It can be shown within an abstract formulation of thermodynamics referred to above (see Refs. 16, 17 and 3 for example) that the minimum free energy is equal to the maximum generalized recoverable work, which from the state at time t is given by

$$W_R(t) = -\int_t^{\infty} \mathbf{D}(u) \odot \dot{\mathbf{C}}(u) du = W(t) - W(\infty). \qquad (8.1)$$

As in Sec. 6, we consider the set of continuations $\mathbf{C}^t(s), s \in \mathbb{R}^{--}$ that go to zero at large $|s|$. This is based on results in Refs. 11 and 22, showing for a linear model that the results of the minimization are insensitive to the final value of the continuation, which is related to $\phi(\infty)$. We obtain from (6.14) and (4.32) that

$$W(\infty) = \frac{1}{2\pi} \int_{-\infty}^{\infty} [\overline{\mathbf{C}_{r+}^t}(\omega) + \overline{\mathbf{C}_{r-}^t}(\omega)] \odot \mathbb{H}(\omega)[\mathbf{C}_{r+}^t(\omega) + \mathbf{C}_{r-}^t(\omega)] d\omega. \qquad (8.2)$$

Note that, by virtue of (4.32), we could replace $\mathbf{C}_{rF}^t$ by $\mathbf{C}_F^t$ in the above relation, since $\mathbb{H}(\omega)$ vanishes at $\omega = 0$.

We seek to minimize $W(\infty)$, given by (8.2), since $W(t)$ is specified.

The optimization procedure was carried out in past work by two different but equivalent techniques: a direct variational method[23,11] or by reducing the problem to the solution of a Weiner–Hopf integral equation.[17] A new and simple method, first introduced in Ref. 3, is outlined below.

Remark 8.1. The only restriction on the relaxation function required in this section is the analyticity of certain quantities on the real axis in the frequency domain. For these constraints to be satisfied, it is sufficient that $\mathbb{L}'(\omega)$ has no singularity on the real axis, a condition which was imposed after (4.18). This implies that $\mathbb{L}'(s)$ decays exponentially at large s. However, the rate of decay can be chosen to be very slow, if required.

8.1. *A new optimization procedure*

Let $\mathbf{C}_o^t$ be the optimal future continuation (so that $\dot{\mathbf{C}}_o$ is the optimal process) and

$$\mathbf{C}_{ro}^t(s) = \mathbf{C}_o^t(s) - \mathbf{C}(t), \quad s \in \mathbb{R}^-. \tag{8.3}$$

Let $\mathbf{C}_m^t$ denote the Fourier transform of $\mathbf{C}_{ro}^t$ so that $\mathbf{C}_m^t(\omega) = \mathbf{C}_{ro-}^t(\omega)$. Recalling (6.17), let us consider now the quantity $\mathbb{H}_-(\omega)\mathbf{C}_{r+}^t(\omega)$, the components of which are continuous, indeed analytic on $\mathbb{R}$ (strictly, an open set containing $\mathbb{R}$), by virtue of the analyticity properties of $\mathbb{H}_-(\omega)$ and $\mathbf{C}_+^t(\omega)$. The Plemelj formulae give that

$$\mathbf{P}^t(\omega) = \mathbb{H}_-(\omega)\mathbf{C}_{r+}^t(\omega) = \mathbf{p}_-^t(\omega) - \mathbf{p}_+^t(\omega), \tag{8.4}$$

where

$$\mathbf{p}^t(z) = \frac{1}{2\pi i} \int_{-\infty}^{\infty} \frac{\mathbf{P}^t(\omega')}{\omega' - z}d\omega', \quad \mathbf{p}_\pm^t(\omega) = \lim_{\alpha \to 0^\mp} \mathbf{p}^t(\omega + i\alpha). \tag{8.5}$$

Moreover, $\mathbf{p}^t(z) = \mathbf{p}_+^t(z)$ is analytic in $z \in \Omega^{(-)}$ and $\mathbf{p}^t(z) = \mathbf{p}_-^t(z)$ is analytic in $z \in \Omega^{(+)}$. Both are analytic on the real axis. Referring to the footnote on (4.25), we write them in the form

$$\mathbf{p}_\pm^t(\omega) = \frac{1}{2\pi i} \int_{-\infty}^{\infty} \frac{\mathbf{P}^t(\omega')}{\omega' - \omega^\mp}d\omega'. \tag{8.6}$$

If we can determine explicit formulae for $\mathbf{p}_\pm^t(\omega), \omega \in \mathbb{R}$ (or $\omega \in \Omega^{(\mp)}$), then they can be analytically continued into $\Omega^{(\pm)}$ respectively, defined everywhere except at singularities. Let us write (8.2) as

$$W(\infty) = \frac{1}{2\pi} \int_{-\infty}^{\infty} |\mathbf{p}_-^t(\omega) - \mathbf{p}_+^t(\omega) + \mathbb{H}_-(\omega)\mathbf{C}_{r-}^t(\omega)|^2 d\omega, \tag{8.7}$$

using (6.17) and (8.4). The notation $|\mathbf{A}|^2$, where $\mathbf{A} \in \mathcal{B}$, denotes according to standard convention $\mathbf{A} \odot \mathbf{A}$. Putting

$$\mathbf{p}_{1-}^t(\omega) = \mathbf{p}_-^t(\omega) + \mathbb{H}_-(\omega)\mathbf{C}_{r-}^t(\omega), \tag{8.8}$$

where $\mathbf{p}_{1-}^t$ is analytic on $\Omega^{(+)}$, we have

$$W(\infty) = \frac{1}{2\pi} \int_{-\infty}^{\infty} |(\mathbf{p}_{1-}^t(\omega) - \mathbf{p}_+^t(\omega)|^2 d\omega$$

$$= \frac{1}{2\pi} \int_{-\infty}^{\infty} [|\mathbf{p}_{1-}^t(\omega)|^2 + |\mathbf{p}_+^t(\omega)|^2]d\omega, \tag{8.9}$$

since the cross-terms can be shown to vanish by Cauchy's theorem. Only $\mathbf{p}_{1-}^t$ depends on $\mathbf{C}_{r-}^t$ so that the minimum can only be given by the condition

$$\mathbf{p}_{1-}^t(\omega) = \mathbf{0}, \tag{8.10}$$

or

$$\mathbf{p}_-^t(\omega) + \mathbb{H}_-(\omega)\mathbf{C}_m^t(\omega) = \mathbf{0}, \quad \forall \omega \in \mathbb{R}, \tag{8.11}$$

yielding

$$\mathbf{C}_m^t(\omega) = -(\mathbb{H}_-(\omega))^{-1}\,\mathbf{p}_-^t(\omega). \tag{8.12}$$

Also, the optimal value of $W(\infty)$, given by (8.9), is

$$W_{\mathrm{opt}}(\infty) = \frac{1}{2\pi}\int_{-\infty}^{\infty}|\mathbf{p}_+^t(\omega)|^2 d\omega. \tag{8.13}$$

Note that, from (6.11) and (8.4)

$$W(t) = \phi(t) + \frac{1}{2\pi}\int_{-\infty}^{\infty}|\mathbf{p}_-^t(\omega)-\mathbf{p}_+^t(\omega)|^2 d\omega$$

$$= \phi(t) + \frac{1}{2\pi}\int_{-\infty}^{\infty}[|\mathbf{p}_-^t(\omega)|^2 + |\mathbf{p}_+^t(\omega)|^2]d\omega \tag{8.14}$$

since the cross-terms again vanish. Thus, from (8.1) and (8.13), we have that the maximum recoverable work or the minimum free energy is given by

$$\psi_m(t) = \phi(t) + \frac{1}{2\pi}\int_{-\infty}^{\infty}|\mathbf{p}_-^t(\omega)|^2 d\omega. \tag{8.15}$$

Remark 8.2. Using the method given in Ref. 11 (see also Ref. 3), it can be shown that $\mathbf{p}_-^t$ is a function of the minimum state (see Sec. 5), so that it follows immediately from (8.15) that $\psi_m(t)$ has this property.

With the aid of (8.12), we can write (8.15) as

$$\psi_m(t) = \phi(t) + \frac{1}{2\pi}\int_{-\infty}^{\infty}\overline{\mathbf{C}_m^t}(\omega)\odot\mathbb{H}(\omega)\mathbf{C}_m^t(\omega)d\omega$$

$$= \phi(t) + \frac{1}{2}\int_{-\infty}^{0}\int_{-\infty}^{0}\mathbb{L}_{12}(|s-u|)\mathbf{C}_{ro}^t(u)\odot\mathbf{C}_{ro}^t(s)\,du\,ds$$

$$= \frac{1}{2}\int_{-\infty}^{0}\int_{-\infty}^{0}\mathbb{L}(|s-u|)\dot{\mathbf{C}}_o^t(u)\odot\dot{\mathbf{C}}_o^t(s)\,du\,ds, \tag{8.16}$$

where $\mathbf{C}_{ro}^t$, given by (8.3), is the inverse Fourier transform of $\mathbf{C}_m^t(\omega)$. The first form of (8.16) follows from the second form by close analogy with the steps from $(6.6)_2$ to (6.11). In both cases, the steps can be reversed. The third relation follows by partial integration.

From (2.75), we have

$$\dot{\psi}_m(t) + D_m(t) = \mathbf{D}(t)\odot\dot{\mathbf{C}}(t), \tag{8.17}$$

where D_m is the rate of dissipation corresponding to the minimum free energy, and must be non-negative by the second law. Recalling that the material is undisturbed

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in the distant past, we integrate (8.17) up to time t to obtain (see (2.77))

$$\psi_m(t) + \mathcal{D}_m(t) = W(t), \tag{8.18}$$

where

$$\mathcal{D}_m(t) = \int_{-\infty}^{t} D_m(s)\,ds \tag{8.19}$$

is the total dissipation up to time t, corresponding to the minimum free energy. Since ψ_m is less than or equal to any other free energy, it follows from (8.18) that $\mathcal{D}_m(t)$ is the largest estimate of dissipation in the material element. We have, from (8.14) and (8.15),

$$\mathcal{D}_m(t) = W(t) - \psi_m(t) = W_{\mathrm{opt}}(\infty) = \frac{1}{2\pi} \int_{-\infty}^{\infty} |\mathbf{p}_+^t(\omega)|^2 d\omega \geq 0. \tag{8.20}$$

Also,

$$D_m(t) = \dot{\mathcal{D}}_m(t). \tag{8.21}$$

In order to give an explicit expression for D_m, we note certain properties of $\mathbf{p}_\pm^t$. From (8.6) and (4.26)$_2$, it follows that

$$\frac{d}{dt}\mathbf{p}_+^t(\omega) = -i\omega\mathbf{p}_+^t(\omega) - \mathbf{K}(t),$$

$$\frac{d}{dt}\mathbf{p}_-^t(\omega) = -i\omega\mathbf{p}_-^t(\omega) - \mathbf{K}(t) - \frac{\mathbb{H}_-(\omega)\dot{\mathbf{C}}(t)}{i\omega}, \tag{8.22}$$

$$\mathbf{K}(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \mathbb{H}_-(\omega)\mathbf{C}_{r+}^t(\omega)\,d\omega.$$

The relation

$$\frac{1}{2\pi i} \int_{-\infty}^{\infty} \frac{\mathbb{H}_-(\omega')}{i\omega'^-(\omega' - \omega^+)} d\omega' = \frac{\mathbb{H}_-(\omega)}{i\omega} \tag{8.23}$$

has been used. This follows by remembering that $\mathbb{H}_-(\omega)$ vanishes linearly at the origin and by closing the contour on $\Omega^{(+)}$, where $\mathbb{H}_-$ is analytic. If $(\omega' - \omega^-)$ occurs in the denominator, the integral vanishes. Furthermore

$$\lim_{|\omega| \to \infty} \omega\mathbf{p}_\pm^t(\omega) = i\mathbf{K}(t),$$

$$\frac{1}{2\pi} \int_{-\infty}^{\infty} \mathbf{p}_\pm^t(\omega)\,d\omega = \mp\frac{1}{2}\mathbf{K}(t). \tag{8.24}$$

The first relation follows from (8.6), while the second emerges by closing the contour on $\Omega^{(\mp)}$, remembering the analyticity properties of $\mathbf{p}_\pm^t$. Differentiating (8.20) with respect to t, we find the explicitly non-negative form for the rate of dissipation:

$$D_m(t) = |\mathbf{K}(t)|^2, \tag{8.25}$$

where $\mathbf{K}$ is given by (8.22)$_3$. The analogous expression for simple viscoelastic materials was derived in Refs. 23 and 11.

1   ### 8.2. *Histories rather than relative histories*

2   In early work on the minimum and other free energies,[23,11,17] histories, rather than
3   relative histories, were used. By almost identical arguments to those above, one can
4   show the following: let

$$\mathbf{Q}^t(\omega) = \mathbb{H}_-(\omega)\mathbf{C}_+^t(\omega) = \mathbf{q}_-^t(\omega) - \mathbf{q}_+^t(\omega), \tag{8.26}$$

6   where

$$\mathbf{q}_\pm^t(\omega) = \frac{1}{2\pi i}\int_{-\infty}^{\infty}\frac{\mathbf{Q}^t(\omega')}{\omega' - \omega^\mp}d\omega'. \tag{8.27}$$

8   Then

$$\psi(t) = S(t) + \frac{1}{2\pi}\int_{-\infty}^{\infty}|\mathbf{q}_-^t(\omega)|^2 d\omega, \tag{8.28}$$

10   where $S(t)$ is given by $(3.20)_2$. The various free energies in Sec. 7 can also be
11   expressed as functionals of histories, rather than relative histories, in entirely anal-
12   ogous ways, as indicated in $(7.2)_2$, (7.11) and $(7.22)_2$. There are two disadvantages
13   to this approach, one noted earlier, namely that $S(t)$ is not a non-negative quantity;
14   the other is that $\mathbf{C}_+^t(\omega)$ behaves as ω^{-1} at large ω while $\mathbf{C}_{r+}^t(\omega)$ behaves as ω^{-2}.
15   It should be noted that $\mathbf{q}$ and $\mathbf{Q}$ in this subsection have quite different meanings
16   to the quantities introduced and used in Sec. 2.

17   ### 8.3. *Confirmation that ψ_m is a free energy*

18   We now ascertain that ψ_m has the characteristic properties of a free energy, as
19   summarized in Sec. 2.2.

20   **Proposition 8.1.** *The functional $\psi_m(t)$, given by (8.15), obeys the non-local Graffi*
21   *conditions, given by* (P1)–(P3), *in Sec.* 2.2, *omitting the temperature variables.*

22   **Proof.** Property (P2) is immediately apparent. Property (P3) follows from (8.17)
23   and the fact that D_m, given by (8.25), is manifestly non-negative. Finally, property
24   (P1), namely $(2.53)_2$, can be proved with the aid of

$$\frac{\partial\mathbf{p}_-^t(\omega)}{\partial\mathbf{C}(t)} = -\frac{\mathbb{H}_-(\omega)}{i\omega}, \tag{8.29}$$

26   which follows from (8.23) and, recalling $(6.17)_2$,

$$\frac{\partial\overline{\mathbf{p}_-^t(\omega)}}{\partial\mathbf{C}(t)} = \frac{\mathbb{H}_+(\omega)^\top}{i\omega}, \tag{8.30}$$

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giving

$$\frac{\partial}{\partial \mathbf{C}(t)} \frac{1}{2\pi} \int_{-\infty}^{\infty} \overline{\mathbf{p}_-^t(\omega)} \odot \mathbf{p}_-^t(\omega) d\omega$$

$$= \frac{\partial}{\partial \mathbf{C}(t)} \frac{1}{2\pi} \int_{-\infty}^{\infty} [\overline{\mathbf{p}_-^t(\omega)}]^\top \mathbf{p}_-^t(\omega) d\omega$$

$$= \frac{1}{2\pi i} \int_{-\infty}^{\infty} \frac{\mathbb{H}_+(\omega)}{\omega} \mathbf{p}_-^t(\omega) d\omega - \frac{1}{2\pi i} \int_{-\infty}^{\infty} \overline{\frac{\mathbb{H}_+(\omega)}{\omega} \mathbf{p}_-^t(\omega)} d\omega$$

$$= \mathrm{Re} \left\{ \frac{1}{\pi i} \int_{-\infty}^{\infty} \frac{\mathbb{H}_+(\omega)}{\omega} \mathbf{p}_-^t(\omega) d\omega \right\}. \tag{8.31}$$

Recall that $\mathbb{H}_\pm$ vanish linearly at the origin. Also, using (8.4), the frequency integral in $(4.36)_2$, which must be real, can be written as

$$-\frac{i}{\pi} \int_{-\infty}^{\infty} \frac{\mathbb{H}(\omega)}{\omega} \mathbf{C}_{r+}^t(\omega) d\omega = -\frac{i}{\pi} \int_{-\infty}^{\infty} \frac{\mathbb{H}_+(\omega)}{\omega} [\mathbf{p}_-^t(\omega) - \mathbf{p}_+^t(\omega)] d\omega,$$

$$= -\frac{i}{\pi} \int_{-\infty}^{\infty} \frac{\mathbb{H}_+(\omega)}{\omega} \mathbf{p}_-^t(\omega) d\omega, \tag{8.32}$$

1 because the term involving $\mathbf{p}_+^t$ vanishes by Cauchy's theorem. Since the last quan-
2 tity is real, we have, from $(4.36)_2$, (8.31) and (3.9), that

3
$$\frac{\partial \psi_m(t)}{\partial \mathbf{C}(t)} = \mathbf{D}(t), \tag{8.33}$$

4 which is $(2.53)_2$.   □

5 **8.4. *Double frequency integral form***

By using (8.6) to carry out the frequency integration in (8.15), noting that $\overline{\omega^+} = \omega^-$ and $\omega' - \omega^+$ can be replaced by $(\omega')^- - \omega$, we can write $\psi_m(t)$ in the form

$$\psi_m(t) = \phi(t) + \frac{i}{4\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{\overline{\mathbf{C}_{r+}^t(\omega_1)} \odot \mathbb{M}_m(\omega_1, \omega_2) \mathbf{C}_{r+}^t(\omega_2)}{\omega_1^+ - \omega_2^-} d\omega_1 d\omega_2, \tag{8.34}$$

$$\mathbb{M}_m(\omega_1, \omega_2) = \mathbb{H}_+(\omega_1) \mathbb{H}_-(\omega_2),$$

6 by closing the contour on either $\Omega^{(+)}$ or $\Omega^{(-)}$. The notation in the denominator of
7 the integral in $(8.34)_1$ means that if we integrate first over ω_1, it becomes $(\omega_1 - \omega_2^-)$
8 or if ω_2 first then it is $(\omega_1^+ - \omega_2)$. Also, using $(8.22)_3$, $D_m(t)$, given by (8.25), can
9 be expressed as

10
$$D_m(t) = \frac{1}{4\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \overline{\mathbf{C}_{r+}^t(\omega_1)} \odot \mathbb{M}_m(\omega_1, \omega_2) \mathbf{C}_{r+}^t(\omega_2) d\omega_1 d\omega_2. \tag{8.35}$$

Let us write the double integral in (8.34) in the form

$$P_-(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} |\mathbf{p}_-^t(\omega)|^2 d\omega = \frac{i}{4\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{A^t(\omega_1, \omega_2)}{\omega_1^+ - \omega_2^-} d\omega_1 d\omega_2,$$

$$A^t(\omega_1, \omega_2) = \overline{\mathbf{C}_{r+}^t}(\omega_1) \odot \mathbb{M}_m(\omega_1, \omega_2) \mathbf{C}_{r+}^t(\omega_2). \tag{8.36}$$

Also, in the same way, we obtain

$$P_+(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} |\mathbf{p}_+^t(\omega)|^2 d\omega = -\frac{i}{4\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{A^t(\omega_1, \omega_2)}{\omega_1^- - \omega_2^+} d\omega_1 d\omega_2. \tag{8.37}$$

From (8.20), we see that this is the total dissipation up to time t. One can show that

$$R_-(t) = \frac{i}{4\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{B^t(\omega_1, \omega_2)}{\omega_1^+ - \omega_2^-} d\omega_1 d\omega_2 = 0,$$

$$B^t(\omega_1, \omega_2) = \overline{\mathbf{C}_{r+}^t}(\omega_1) \odot \mathbb{M}_m(\omega_2, \omega_1) \mathbf{C}_{r+}^t(\omega_2) \tag{8.38}$$

by integrating over ω_2 for example and closing the contour on $\Omega^{(-)}$, since $\mathbb{H}_+$ and $\mathbf{C}_{r+}^t$ have no singularity in the lower half-plane. Furthermore, using the same procedure, one obtains

$$R_+(t) = -\frac{i}{4\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{B^t(\omega_1, \omega_2)}{\omega_1^- - \omega_2^+} d\omega_1 d\omega_2$$

$$= \frac{1}{2\pi} \int_{-\infty}^{\infty} \overline{\mathbf{C}_{r+}^t}(\omega) \odot \mathbb{H}(\omega) \mathbf{C}_{r+}^t(\omega) d\omega$$

$$= P_-(t) + P_+(t) \tag{8.39}$$

by virtue of (6.11) and (8.14). Relation (8.38) allows us to write (8.36) in the explicitly convergent form

$$P_-(t) = \frac{i}{4\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{A^t(\omega_1, \omega_2) - B^t(\omega_1, \omega_2)}{\omega_1 - \omega_2} d\omega_1 d\omega_2, \tag{8.40}$$

which is convenient for numerical evaluation. We can replace the $\omega_1 - \omega_2$ in the denominator by $\omega_1^+ - \omega_2^-$ which gives (8.36), or by $\omega_1^- - \omega_2^+$ which gives the same result by way of (8.39) and (8.37). Relation (8.40) implies that the quantity

$$\mathbb{D}(\omega_1, \omega_2) = i \frac{[\mathbb{H}_+(\omega_1)\mathbb{H}_-(\omega_2) - \mathbb{H}_+(\omega_2)\mathbb{H}_-(\omega_1)]}{\omega_1 - \omega_2} \tag{8.41}$$

is a non-negative operator in the same sense as $\mathbb{K}$ in (3.1). By using very localized choices of $\mathbf{C}_+^t(\omega)$, we deduce that the "diagonal elements" of $\mathbb{D}(\omega_1, \omega_2)$ are non-negative. Using a prime to denote differentiation, these can be written as

$$\mathbb{D}(\omega) = i[\mathbb{H}_+'(\omega)\mathbb{H}_-(\omega) - \mathbb{H}_+(\omega)\mathbb{H}_-'(\omega)] \geq \mathbf{0}, \quad \omega \in \mathbb{R}, \tag{8.42}$$

a quantity which will appear in another context towards the end of Sec. 10.

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8.5. *Properties of the optimal future continuation*

We draw attention to certain properties of the optimal future continuation. From (8.12) and (8.24)$_2$, it follows that

$$\mathbf{C}^t_m(\omega) \xrightarrow[\omega\to\infty]{} \frac{\mathbb{H}_{\frac{1}{2}}^{-1}\mathbf{K}(t)}{i\omega}, \quad \mathbb{H}_{\frac{1}{2}}^{-1} = \lim_{\omega\to\infty}\mathbb{H}_{\pm}(\omega) = [\mathbb{H}(\infty)]^{1/2}, \tag{8.43}$$

so that the relative optimal continuation, given by (8.3), has the form at $s = 0$ (cf. (4.30))

$$\mathbf{C}^t_{ro}(0) = -\mathbb{H}_{\frac{1}{2}}^{-1}\mathbf{K}(t) \tag{8.44}$$

and the optimal continuation $\mathbf{C}^t_o(0)$ is given by

$$\mathbf{C}^t_o(0) = \mathbf{C}(t) - \mathbb{H}_{\frac{1}{2}}^{-1}\mathbf{K}(t). \tag{8.45}$$

Thus, the optimal continuation involves a sudden discontinuity at time t, the magnitude of which is related to the rate of dissipation, as we see from (8.25).

Also, putting

$$\mathbb{H}_1(\omega) = \frac{1}{\omega}\mathbb{H}_-(\omega) \tag{8.46}$$

we have

$$\mathbf{C}^t_m(\omega) \approx -\frac{[\mathbb{H}_1(0)]^{-1}}{2\pi i\omega^+} \int_{-\infty}^{\infty} \mathbb{H}_1(\omega')\mathbf{C}^t_{r+}(\omega')d\omega' \tag{8.47}$$

as $\omega \to 0$, which, with the aid of (4.33), gives

$$\mathbf{C}^t_{ro}(-\infty) = \frac{[\mathbb{H}_1(0)]^{-1}}{2\pi} \int_{-\infty}^{\infty} \mathbb{H}_1(\omega')\mathbf{C}^t_{r+}(\omega')d\omega' \tag{8.48}$$

and

$$\mathbf{C}^t_o(-\infty) = \mathbf{C}^t_{ro}(-\infty) + \mathbf{C}(t). \tag{8.49}$$

This quantity is in general non-zero.

9. Factorization: Time-Independent Eigenspaces

The functionals introduced in Secs. 7.1–7.3 are valid free energies under the specified monotonicity restrictions on the kernel $\mathbb{L}$. For the minimum free energy, there is no restriction of this sort on $\mathbb{L}$. However, it is not possible in general to determine explicitly the factors in (6.17). To do this, we must introduce another constraint on the tensor relaxation function, namely that the eigenspaces of $\mathbb{L}(t)$ do not depend on t. Then, an extension to the tensorial case of the method for determining factors used in Refs. 23, 24 and 3 for a scalar constitutive relation yields a simple direct construction of $\mathbb{H}_+(\omega)$ and $\mathbb{H}_-(\omega)$. Also the factorization is commutative (i.e. where $\mathbb{H}_+(\omega)$ and $\mathbb{H}_-(\omega)$ commute). Without this constraint, it is not generally practical to explicitly determine the factors $\mathbb{H}_\pm$.

Under the requirement that the eigenspaces of $\mathbb{L}(t)$ do not depend on time, we can write

$$\mathbb{L}(t) = \sum_{k=1}^{m} L_k(t)\mathbb{B}^k$$

$$\mathbb{H}(\omega) = -\omega\mathbb{L}'_s(\omega) = \sum_{k=1}^{m} H_k(\omega)\mathbb{B}^k, \tag{9.1}$$

$$H_k(\omega) > 0; \quad \omega \in \mathbb{R}\backslash\{0\}, \quad k = 1, \ldots, m,$$

where $\mathbb{B}^k = \mathbf{B}^k \otimes \mathbf{B}^k, k = 1, \ldots, m$ are the orthonormal projectors on the m (time-independent) eigenspaces of $\mathbb{L}$ and $\mathbb{H}$; while $\{\mathbf{B}^k\}$ are their normalized eigenvectors, which constitute an orthonormal basis of $\mathcal{B}$. Recall that m, introduced after (2.40), is the dimension of the vector space $\mathcal{B}$.

From (3.13) it follows that, on a particular basis, we can represent $\mathbf{B}^k, \mathbb{B}^k$ in terms of a time-independent rotation $\mathbf{R}$ on $\mathbb{R}^m$, where $\mathbf{B}^k$ is given by

$$B_i^k = R_{ik}, \quad i = 1, 2, \ldots, m, \tag{9.2}$$

and $\mathbb{B}^k$ by (no summation)

$$B_{ij}^k = R_{ik}R_{jk}, \quad i, j = 1, 2, \ldots, m, \tag{9.3}$$

so that, for example, the most general form of $\mathbb{L}(t)$, in a given basis, allowed under the hypothesis of time-independent eigenspaces, is

$$(\mathbb{L}(t))_{ij} = \sum_{k=1}^{m} L_k(t)R_{ik}R_{jk}, \quad i, j = 1, 2, \ldots, m, \tag{9.4}$$

where R_{ik} are constants, restricted so that $\mathbf{R}$ is a rotation matrix on $\mathbb{R}^m$.

Since $\mathbb{L}(t)$ is a symmetric tensor, it has $m(m+1)/2$ independent components and these must be expressible in terms of the m quantities $L_k, k = 1, 2, \ldots, m$. There must therefore be $m(m-1)/2$ further relations among the components of $\mathbb{L}(t)$, over and above those required by symmetry. These may be written down for example by partially solving (9.4) to express $L_k, k = 1, 2, \ldots, m$ in terms of the diagonal components of $\mathbb{L}(t)$. These can then be substituted back into (9.4).

One can derive explicit forms for the factors of a scalar function H equal to any one of the H_k in $(9.1)_2$ by a procedure outlined in Refs. 24 and 3, for example. Thus, the quantities H_k can be factorized into H_{k+}, H_{k-} where

$$H_{k\pm}(\omega) = H_{k\mp}(-\omega) = \overline{H_{k\mp}}(\omega); \quad H_k(\omega) = |H_{k\pm}(\omega)|^2. \tag{9.5}$$

We put

$$H_k(\infty) = H_{k\infty}, \quad k = 1, 2, \ldots, m. \tag{9.6}$$

Since the $\{\mathbb{B}^k\}$ are orthonormal projectors, it follows that

$$\mathbb{H}(\omega) = \sum_{k=1}^{m} H_{k+}(\omega)H_{k-}(\omega)\mathbb{B}^k = \mathbb{H}_+(\omega)\mathbb{H}_-(\omega), \tag{9.7}$$

where

$$\mathbb{H}_\pm(\omega) = \sum_{k=1}^{m} H_{k\pm}(\omega)\mathbb{B}^k = \mathbb{H}_\pm^\top(\omega). \tag{9.8}$$

The last relation follows from the symmetry of the $\mathbb{B}^k$. It is clear that the factors $\mathbb{H}_+(\omega)$ and $\mathbb{H}_-(\omega)$ commute. From (9.5) we have

$$\mathbb{H}_\pm(\omega) = \mathbb{H}_\mp(-\omega) = \overline{\mathbb{H}_\mp}(\omega). \tag{9.9}$$

These relations are consistent with but more restrictive than $(6.17)_2$.

Note that in the light of (9.9), the quantity $\mathbf{K}$ given by $(8.22)_3$ can be shown to be real by taking its complex conjugate and changing the sign of the integration variable.

In the basis $\{\mathbf{B}^k\}$ and $\{\mathbb{B}^k\}$, the individual components of each of the relevant quantities obey the relationships that hold in the scalar case, as developed in Ref. 24 and earlier work. We can expand any member of $\mathcal{B}$ in this basis; in particular

$$\mathbf{C}(t) = \sum_{k=1}^{m} C_k(t)\mathbf{B}^k, \qquad \mathbf{C}^t(s) = \sum_{k=1}^{m} C_k^t(s)\mathbf{B}^k,$$
$$\mathbf{C}_r^t(s) = \sum_{k=1}^{m} C_{kr}^t(s)\mathbf{B}^k, \quad \mathbf{C}_{r\pm}^t(\omega) = \sum_{k=1}^{m} C_{kr\pm}^t(\omega)\mathbf{B}^k. \tag{9.10}$$

Scalar quantities, such as a free energy or a rate of dissipation, are given by the sum of contributions to this quantity from each eigenspace.

Remark 9.1. If $\mathbb{L}$ is a diagonal tensor, then the rotation $\mathbf{R}$ in the above formalism is unity. This leads to a diagonal tensor $\mathbb{H}$, the scalar components of which can be factorized as in Refs. 23, 24 and 3, yielding explicit factors $\mathbb{H}_\pm(\omega)$. This was the assumption which yielded explicit formulae for the minimum free energies in Refs. 1–3.

9.1. *A simple example with non-diagonal* $\mathbb{L}$

We seek an illustration of the above formalism where $\mathbf{R}$ is not unity, which is sufficiently simple so that algebraic manipulations can be carried out without difficulty. Let us consider a scalar, non-local linear viscoelastic model, where $(E^t(s), E(t))$ is the infinitesimal strain history and current strain, while $T(t)$ is the stress. The gradients of $(E^t(s), E(t))$ will also be included. Without further assumptions, this would involve dealing with 4×4 matrices, as outlined in Sec. 3.1. Therefore, we restrict the model to be one-dimensional, where $(E^t(s), E(t))$ depends on x but not on y, z. Thus, we have

$$\mathbf{C}^t(s) = (E^t(s), E_x^t(s)), \quad E_x^t = \frac{\partial}{\partial x} E^t = \partial_x E^t. \tag{9.11}$$

1   Equations (3.15) and (3.18) give

2
$$T(t) = D_1 - \partial_x D_2, \tag{9.12}$$

where from $(3.11)_2$ (using subscripts $1, 2$ rather than $0, 1$) we have

$$
\begin{aligned}
D_1(t) &= G_{11}(0)E(t) + G_{12}(0)E_x(t) + \int_0^\infty G_{11}'(u)E^t(u)\,du \\
&\quad + \int_0^\infty G_{12}'(u)E_x^t(u)\,du, \\
D_2(t) &= G_{21}(0)E(t) + G_{22}(0)E_x(t) + \int_0^\infty G_{21}'(u)E^t(u)\,du \\
&\quad + \int_0^\infty G_{22}'(u)E_x^t(u)\,du.
\end{aligned}
\tag{9.13}
$$

3   The quantities $G_{11}', G_{12}', G_{21}'$ and G_{22}' are scalar functions and $G_{11}(0), G_{12}(0),$
4    $G_{21}(0), G_{22}(0)$ are constants. Also, from (3.13),

5
$$G_{12}(0) = G_{21}(0), \quad G_{12}'(u) = G_{21}'(u). \tag{9.14}$$

6   By virtue of $(4.15)_1$, we have that

7
$$
\begin{pmatrix} G_{11}(0) & G_{12}(0) \\ G_{12}(0) & G_{22}(0) \end{pmatrix} > \mathbf{0}
\tag{9.15}
$$

8   so that $G_{11}(0) > 0, G_{22}(0) > 0$ and $G_{11}(0)G_{22}(0) > [G_{12}(0)]^2$.

We need to be able to diagonalize the above system of equations with a rotation that is independent of time. Let there exist a constant rotation $\mathbf{R}$ such that

$$
\begin{aligned}
\mathbf{R} \begin{pmatrix} G_{11}(0) & G_{12}(0) \\ G_{12}(0) & G_{22}(0) \end{pmatrix} \mathbf{R}^\top &= \begin{pmatrix} G_1(0) & 0 \\ 0 & G_2(0) \end{pmatrix}, \\
\mathbf{R} \begin{pmatrix} G_{11}'(u) & G_{12}'(u) \\ G_{12}'(u) & G_{22}'(u) \end{pmatrix} \mathbf{R}^\top &= \begin{pmatrix} G_1'(u) & 0 \\ 0 & G_2'(u) \end{pmatrix}, \\
\mathbf{R} = \begin{pmatrix} c & s \\ -s & c \end{pmatrix}, &\quad c^2 + s^2 = 1,
\end{aligned}
\tag{9.16}
$$

where $G_1(0), G_2(0), G_1', G_2'$ are the required eigenvalues. It follows that (cf. (9.4))

$$
\begin{aligned}
\begin{pmatrix} G_{11}(0) & G_{12}(0) \\ G_{12}(0) & G_{22}(0) \end{pmatrix} &= G_1(0) \begin{pmatrix} c^2 & cs \\ cs & s^2 \end{pmatrix} + G_2(0) \begin{pmatrix} s^2 & -cs \\ -cs & c^2 \end{pmatrix}, \\
\begin{pmatrix} G_{11}'(u) & G_{12}'(u) \\ G_{12}'(u) & G_{22}'(u) \end{pmatrix} &= G_1'(u) \begin{pmatrix} c^2 & cs \\ cs & s^2 \end{pmatrix} + G_2'(u) \begin{pmatrix} s^2 & -cs \\ -cs & c^2 \end{pmatrix}.
\end{aligned}
\tag{9.17}
$$

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These relations yield that

$$
(c^2 - s^2)G_{12}(0) = cs[G_{11}(0) - G_{22}(0)],
$$
$$
(c^2 - s^2)G'_{12}(u) = cs[G'_{11}(u) - G'_{22}(u)],
$$

(9.18)

1   by solving (9.17) for $(G_1(0), G_2(0)), (G'_1(u), G'_2(u))$ in terms of $(G_{11}(0), G_{22}(0))$,
2   (G'_{11}, G'_{22}) respectively and then imposing the non-diagonal relation. Thus (9.18)
3   gives the constraints imposed by the requirement of time-independent eigenspaces.
4   The first constraint determines the angle of rotation. Using this in the second
5   gives G'_{12} uniquely in terms of $G'_{11} - G'_{22}$, for all values of $u \in \mathbb{R}^+$. This is not a
6   physical requirement but a mathematical constraint which yields time-independent
7   eigenspaces.

Solving for the eigenvalues gives

$$
G_1(0) = \frac{1}{2}[G_{11}(0) + G_{22}(0) + \sqrt{[G_{11}(0) - G_{22}(0)]^2 + 4[G_{12}(0)]^2}],
$$
$$
G_2(0) = \frac{1}{2}[G_{11}(0) + G_{22}(0) - \sqrt{[G_{11}(0) - G_{22}(0)]^2 + 4[G_{12}(0)]^2}],
$$

(9.19)

and

$$
G'_1(u) = \frac{1}{2}[G'_{11}(u) + G'_{22}(u) + \sqrt{[G'_{11}(u) - G'_{22}(u)]^2 + 4[G'_{12}(u)]^2}],
$$
$$
G'_2(u) = \frac{1}{2}[G'_{11}(u) + G'_{22}(u) - \sqrt{[G'_{11}(u) - G'_{22}(u)]^2 + 4[G'_{12}(u)]^2}].
$$

(9.20)

8   Let us for definiteness take the case where $G_{11}(0) > G_{22}(0), G'_{11}(u) > G'_{22}(u)$ and
9    $c^2 > s^2$. Then, with the aid of (9.18), these can be written as

10    $$
G_1(0) = aG_{11}(0) + bG_{22}(0), \quad G_2(0) = bG_{11}(0) + aG_{22}(0)
$$ (9.21)

11   and

12    $$
G'_1(u) = aG'_{11}(u) + bG'_{22}(u), \quad G'_2(u) = bG'_{11}(u) + aG'_{22}(u),
$$ (9.22)

13   where

14    $$
a = \frac{c^2}{c^2 - s^2}, \quad b = 1 - a.
$$ (9.23)

For different inequalities, these formulae are somewhat altered. The case where
$s = 0$ corresponds to where the equations were diagonal to begin with. Relations
(9.13) can be written in the form

$$
X_1(t) = G_1(0)Y_1(t) + \int_0^\infty G'_1(u)Y_1^t(u)\,du,
$$
$$
X_2(t) = G_2(0)Y_2(t) + \int_0^\infty G'_2(u)Y_2^t(u)\,du,
$$

(9.24)

where

$$\begin{pmatrix} X_1(t) \\ X_2(t) \end{pmatrix} = \mathbf{R} \begin{pmatrix} D_1(t) \\ D_2(t) \end{pmatrix},$$

$$\begin{pmatrix} Y_1(t) \\ Y_2(t) \end{pmatrix} = \mathbf{R} \begin{pmatrix} E(t) \\ E_x(t) \end{pmatrix}, \qquad (9.25)$$

$$\begin{pmatrix} Y_1^t(u) \\ Y_2^t(u) \end{pmatrix} = \mathbf{R} \begin{pmatrix} E^t(u) \\ E_x^t(u) \end{pmatrix}.$$

1 These relationships can of course be inverted and we can write (9.24) in the
2 form (9.13), though where $G_{ij}(0), G_{ij}'(u), i, j = 1, 2$, are expressed in terms of the
3 eigenvalues $G_i(0), G_i'(u), i = 1, 2$. Then, in turn, we can express $T(t)$, given by
4 (9.12), in terms of these eigenvalues.

5 Relation (9.17) corresponds to (9.4) or $(9.1)_1$ where G_1' and G_2' play the role of
6 $L_k', k = 1, 2$. We can then determine explicitly the scalar functions $H_k(\omega), k = 1, 2$,
7 in $(9.1)_2$ which can be factorized by a standard technique.[23,3] Thus, we can write
8 down (9.8) explicitly, which yields a formula for the minimum free energy.

9 Indeed, because of the orthonormal character of (9.1) and subsequent relations,
10 we can write the minimum free energy in the form

11 $$\psi_m(t) = \psi_{1m}(t) + \psi_{2m}(t), \qquad (9.26)$$

12 where $\psi_{1m}(t), \psi_{2m}(t)$ are the minimum free energies corresponding to the scalar
13 problems with kernel function derivatives G_1', G_2', respectively and where the inde-
14 pendent variables are the quantities on the left of $(9.25)_{2,3}$.

15 **Remark 9.2.** The maximum free energy for materials that have non-singleton
16 minimal states (those with kernels $\mathbb{L}'(\omega)$ that have only isolated singularities in the
17 frequency domain) and a family of intermediate free energies can be derived as in
18 Refs. 17, 25 and 3.

19 **10. Free Energies for Sinusoidal Histories**

20 Consider a current value and history $(\mathbf{C}^t, \mathbf{C}(t))$ defined by

21 $$\mathbf{C}(t) = \mathbf{C}_0 e^{i\omega_- t} + \overline{\mathbf{C}_0} e^{-i\omega_+ t}, \quad \mathbf{C}^t(s) = \mathbf{C}(t - s), \qquad (10.1)$$

22 where $\mathbf{C}_0 \in \mathcal{B}$ is an amplitude and $\overline{\mathbf{C}_0}$ its complex conjugate. Furthermore,

23 $$\omega_- = \omega_0 - i\eta, \quad \omega_+ = \overline{\omega}_-, \quad \omega_0 \in \mathbb{R}, \quad \eta \in \mathbb{R}^{++}. \qquad (10.2)$$

24 The quantity η is introduced to maintain (2.47) and to ensure finite results in
25 certain quantities. The quantity $\mathbf{C}_+^t$ has the form

26 $$\mathbf{C}_+^t(\omega) = \mathbf{C}_0 \frac{e^{i\omega_- t}}{i(\omega + \omega_-)} + \overline{\mathbf{C}_0} \frac{e^{-i\omega_+ t}}{i(\omega - \omega_+)}, \qquad (10.3)$$

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and the Fourier transform of the relative history $\mathbf{C}_r^t(s) = \mathbf{C}^t(s) - \mathbf{C}(t)$, namely $\mathbf{C}_{r+}^t(\omega)$, is given by

$$\mathbf{C}_{r+}^t(\omega) = \mathbf{C}_+^t(\omega) - \frac{\mathbf{C}(t)}{i\omega^-} = -\mathbf{C}_0 \frac{\omega_-}{\omega^-} \frac{e^{i\omega_- t}}{i(\omega + \omega_-)} + \overline{\mathbf{C}_0} \frac{\omega_+}{\omega^-} \frac{e^{-i\omega_+ t}}{i(\omega - \omega_+)}. \qquad (10.4)$$

Recall that $\mathbb{L}', \mathbb{L}_0$ and $\mathbb{L}_\infty$ are symmetric tensors, a property that will be used frequently below. From $(3.11)_2$, we have

$$\mathbf{D}(t) = \mathbb{L}_0 \mathbf{C}(t) + \int_0^\infty \mathbb{L}'(s) \mathbf{C}^t(s) \, ds$$

$$= [\mathbb{L}_0 + \mathbb{L}'_+(\omega_-)] \mathbf{C}_0 e^{i\omega_- t} + [\mathbb{L}_0 + \mathbb{L}'_+(-\omega_+)] \overline{\mathbf{C}_0} e^{-i\omega_+ t}. \qquad (10.5)$$

Any real algebraic quadratic form in $\mathbf{C}(t)$ or real functional quadratic form in $\mathbf{C}^t(s)$ can be written, in the limit $\eta \to 0$, as

$$Q = \mathbf{C}_0 \odot \mathbb{M} \mathbf{C}_0 e^{2i\omega_0 t} + \overline{\mathbf{C}_0} \odot \overline{\mathbb{M}} \, \overline{\mathbf{C}_0} e^{-2i\omega_0 t} + \overline{\mathbf{C}_0} \odot \mathbb{N} \mathbf{C}_0, \qquad (10.6)$$

where $\mathbb{M}, \overline{\mathbb{M}}, \mathbb{N} \in \text{Lin}(\mathcal{B})$. The quantities $\mathbb{M}$ and $\overline{\mathbb{M}}$ can be taken to be symmetric, while $\mathbb{N}$ can be taken to be hermitian. Indeed, since we will be dealing with symmetric quantities, $\mathbb{N}$ must be real. Let us introduce the abbreviated notation

$$Q = \{\mathbb{M}, \mathbb{N}\}. \qquad (10.7)$$

The generalized work $W(t)$ done on the material to achieve the state $(\mathbf{C}^t, \mathbf{C}(t))$ is obtained by integrating the form

$$\mathbf{D}(t) \odot \dot{\mathbf{C}}(t) = i\omega_- \mathbf{C}_0 \odot [\mathbb{L}_0 + \mathbb{L}'_+(\omega_-)] \mathbf{C}_0 e^{2i\omega_- t} - i\omega_+$$

$$\times \overline{\mathbf{C}_0} \odot [\mathbb{L}_0 + \mathbb{L}'_+(-\omega_+)] \overline{\mathbf{C}_0} e^{-2i\omega_+ t} + i\overline{\mathbf{C}_0} \odot [(\omega_- - \omega_+)\mathbb{L}_0$$

$$+ \omega_- \mathbb{L}'_+(-\omega_+) - \omega_+ \mathbb{L}'_+(\omega_-)] \mathbf{C}_0 e^{i(\omega_- - \omega_+)t}. \qquad (10.8)$$

In the limit $\eta \to 0$, this converges to a finite result of the form

$$\dot{W}(t) = \mathbf{D}(t) \odot \dot{\mathbf{C}}(t) = \{i\omega_0[\mathbb{L}_0 + \mathbb{L}'_+(\omega_0)], -2\omega_0 \mathbb{L}'_s(\omega_0)\} \qquad (10.9)$$

in the notation of (10.7). Carrying out the integration of (10.8) gives

$$W(t) = \frac{1}{2}\{\mathbf{C}_0 \odot [\mathbb{L}_0 + \mathbb{L}'_+(\omega_-)] \mathbf{C}_0 e^{2i\omega_- t} + \overline{\mathbf{C}_0} \odot [\mathbb{L}_0 + \mathbb{L}'_+(-\omega_+)] \overline{\mathbf{C}_0} e^{-2i\omega_+ t}\}$$

$$+ \overline{\mathbf{C}_0} \odot [(\omega_- - \omega_+)\mathbb{L}_0 + \omega_- \mathbb{L}'_+(-\omega_+) - \omega_+ \mathbb{L}'_+(\omega_-)] \mathbf{C}_0 \frac{e^{i(\omega_- - \omega_+)t}}{(\omega_- - \omega_+)}. \qquad (10.10)$$

This quantity diverges as $\eta \to 0$, as would be expected on physical grounds. Taking the limit $\eta \to 0$ in the terms which are convergent, we can write this in the form

$$W(t) = \left\{ \frac{1}{2}[\mathbb{L}_0 + \mathbb{L}'_+(\omega_0)], \mathbb{L}_0 + \mathbb{L}'_c(\omega_0) - \omega_0 \frac{\partial}{\partial \omega_0} \mathbb{L}'_c(\omega_0) \right.$$

$$\left. - 2\omega_0 t \mathbb{L}'_s(\omega_0) - \mathbb{L}'_s(\omega_0) \frac{\omega_0}{\eta} \right\}. \qquad (10.11)$$

The quantity $S(t)$, given by $(3.20)_2$, is finite in the limit $\eta \to 0$. It reduces to

$$S(t) = \left\{ \frac{1}{2}\mathbb{L}_0 + \mathbb{L}'_+(\omega_0), \mathbb{L}_0 + 2\mathbb{L}'_c(\omega_0) \right\}. \tag{10.12}$$

The general form (3.20) can be expressed as

$$\psi(t) = \left\{ \frac{1}{2}\mathbb{L}_0 + \mathbb{L}'_+(\omega_0) + \frac{1}{2}\mathbb{L}_{12+}(\omega_0, \omega_0), \mathbb{L}_0 + 2\mathbb{L}'_c(\omega_0) \right.$$
$$\left. + \frac{1}{2}[\mathbb{L}_{12+}(\omega_0, -\omega_0) + \mathbb{L}_{12+}(-\omega_0, \omega_0)] \right\}, \tag{10.13}$$

where

$$\mathbb{L}_{12+}(\omega_1, \omega_2) = \int_0^\infty \int_0^\infty \mathbb{L}_{12}(s, u)e^{-i\omega_1 s - i\omega_2 u}$$
$$= -\mathbb{L}'_+(\omega_2) + i\omega_1 \mathbb{L}_{2+}(\omega_1, \omega_2)$$
$$= -\mathbb{L}'_+(\omega_1) + i\omega_2 \mathbb{L}_{1+}(\omega_1, \omega_2)$$
$$= \frac{1}{2}[-\mathbb{L}'_+(\omega_1) - \mathbb{L}'_+(\omega_2) + i\omega_1 \mathbb{L}_{2+}(\omega_1, \omega_2)$$
$$+ i\omega_2 \mathbb{L}_{1+}(\omega_1, \omega_2)]. \tag{10.14}$$

Also,

$$\mathbb{L}_{1+}(\omega_1, \omega_2) = \int_0^\infty \int_0^\infty \mathbb{L}_1(s, u)e^{-i\omega_1 s - i\omega_2 u} ds du, \tag{10.15}$$

and similarly for $\mathbb{L}_{2+}$. Partial integrations have been used to obtain the second and third forms of (10.14). Note that

$$\overline{\mathbb{L}_{12+}}(\omega_1, \omega_2) = \mathbb{L}_{12+}(-\omega_1, -\omega_2) \tag{10.16}$$

and, from $(3.5)_2$,

$$\mathbb{L}_{12+}^\top(\omega_1, \omega_2) = \mathbb{L}_{12+}(\omega_2, \omega_1). \tag{10.17}$$

Therefore, $\mathbb{L}_{12+}(\omega_0, \omega_0)$ is symmetric and the last term in square brackets in (10.13) is symmetric and real. Also,

$$\overline{\mathbb{L}_{1+}}(\omega_1, \omega_2) = \mathbb{L}_{1+}(-\omega_1, -\omega_2) \tag{10.18}$$

and

$$\mathbb{L}_{1+}^\top(\omega_1, \omega_2) = \mathbb{L}_{1+}(\omega_2, \omega_1), \tag{10.19}$$

with similar relations holding for $\mathbb{L}_{2+}$. Using $(10.14)_4$, we can write (10.13) in the form

$$\psi(t) = \left\{ \frac{1}{2}[\mathbb{L}_0 + \mathbb{L}'_+(\omega_0)] + \frac{i\omega_0}{4}[\mathbb{L}_{1+}(\omega_0, \omega_0) + \mathbb{L}_{2+}(\omega_0, \omega_0)], \right.$$
$$\left. \mathbb{L}_0 + \mathbb{L}'_c(\omega_0) + \frac{\omega_0}{2}\text{Im}[\mathbb{L}_{1+}(\omega_0, -\omega_0) - \mathbb{L}_{2+}(\omega_0, -\omega_0)] \right\}, \tag{10.20}$$

The most interesting part of this expression, from a physical point of view, is the second term which yields the time average over a period,

$$\langle \psi(t) \rangle_{\mathrm{av}} = \overline{\mathbf{C}_0} \odot \left\{ \mathbb{L}_0 + \mathbb{L}'_c(\omega_0) + \frac{\omega_0}{2} \mathrm{Im}[\mathbb{L}_{1+}(\omega_0, -\omega_0) - \mathbb{L}_{2+}(\omega_0, -\omega_0)] \right\} \mathbf{C}_0. \tag{10.21}$$

Remark 10.1. The quantity in braces must be a non-negative operator in $\mathrm{Lin}(\mathcal{B})$ for all ω_0. A corresponding observation must apply to each of the free energy functionals below, if the assumptions that make the relevant functional a free energy are true.

The rate of dissipation, given by (3.22), becomes

$$D(t) = \left\{ \frac{\omega_0^2}{2}[\mathbb{L}_{1+}(\omega_0, \omega_0) + \mathbb{L}_{2+}(\omega_0, \omega_0)], -\frac{\omega_0^2}{2}[\mathbb{L}_{1+}(\omega_0, -\omega_0) \right.$$

$$\left. + \mathbb{L}_{1+}(-\omega_0, \omega_0) + \mathbb{L}_{2+}(\omega_0, -\omega_0) + \mathbb{L}_{2+}(-\omega_0, \omega_0)] \right\}$$

$$= \left\{ \frac{\omega_0^2}{2}[\mathbb{L}_{1+}(\omega_0, \omega_0) + \mathbb{L}_{2+}(\omega_0, \omega_0)], -2\omega_0 \mathbb{L}'_s(\omega_0) \right\}, \tag{10.22}$$

where $(10.14)_2$ and $(10.14)_3$ have been invoked.

Remark 10.2. The time average of the rate of dissipation is

$$\langle D(t) \rangle_{\mathrm{av}} = \overline{\mathbf{C}_0} \odot (-2\omega_0 \mathbb{L}'_s(\omega_0)) \mathbf{C}_0 = \langle \dot{W}(t) \rangle_{\mathrm{av}} \geq 0, \tag{10.23}$$

from (10.9), which is a special case of (4.5). Note that (10.23) implies (4.7) (if we exclude reversible processes).

Relation (10.23) is true for the rate of dissipation associated with any free energy, since (3.22) is entirely general for a material with linear constitutive equations.

It is noteworthy that, in classical viscoelasticity, $-\mathbb{L}'_s(\omega) = \mathrm{Im}\,\mathbb{L}'_+(\omega)$ is referred to as the loss modulus.

Observe that

$$2\mathbb{H}(\omega_0) = -2\omega_0 \mathbb{L}'_s(\omega_0), \tag{10.24}$$

in the notation of (4.19). This notation has been and will be particularly used in the context of the minimum free energy (see (10.51) below).

We have, from (10.20),

$$\dot{\psi}(t) = \left\{ i\omega_0[\mathbb{L}_0 + \mathbb{L}'_+(\omega_0)] - \frac{\omega_0^2}{2}[\mathbb{L}_{1+}(\omega_0, \omega_0) + \mathbb{L}_{2+}(\omega_0, \omega_0)], 0 \right\}. \tag{10.25}$$

Then, from (2.75) and (10.9), we can also obtain (10.22). The fact that $\langle \dot{\psi}(t) \rangle_{\mathrm{av}}$ vanishes is also an entirely general feature, closely connected with (10.23), by virtue of (2.75).

1   The Graffi–Volterra free energy, given by $(7.2)_2$, has the form

$$\psi_G(t) = \left\{ \frac{1}{2}\mathbb{L}_0 + \mathbb{L}'_+(\omega_0) - \frac{1}{2}\mathbb{L}'_+(2\omega_0), 2\mathbb{L}_0 - \mathbb{L}_\infty + 2\mathbb{L}'_c(\omega_0) \right\} \qquad (10.26)$$

3   and

$$\langle \psi_G(t) \rangle = \overline{\mathbf{C}_0} \odot [2\mathbb{L}_0 - \mathbb{L}_\infty + 2\mathbb{L}'_c(\omega_0)]\mathbf{C}_0. \qquad (10.27)$$

5   By invoking Remark 10.1, we see that the quantity $2\mathbb{L}_0 - \mathbb{L}_\infty + 2\mathbb{L}'_c(\omega_0)$ must be a
6   non-negative quantity if (7.1) applies. The time derivative of (10.26) gives

$$\dot{\psi}_G(t) = 2i\omega_0 \left\{ \frac{1}{2}\mathbb{L}_0 + \mathbb{L}'_+(\omega_0) - \frac{1}{2}\mathbb{L}'_+(2\omega_0), 0 \right\}. \qquad (10.28)$$

8   Using (2.75) and (10.9), we obtain

$$D_G(t) = \{ -i\omega_0[\mathbb{L}'_+(\omega_0) - \mathbb{L}'_+(2\omega_0)], -2\omega_0\mathbb{L}'_s(\omega_0) \}, \qquad (10.29)$$

10   which can also be derived from (7.3).

We next consider the expression $(7.22)_2$ for the Dill free energy. The function $S(t)$ is given by (10.12). We now consider the integral term. Note that, by a change of variables,

$$\frac{1}{2} \int_0^\infty \int_0^\infty \mathbb{L}''(s_1 + s_2)e^{-i\omega_0(s_1 + s_2)} \, ds_1 ds_2$$

$$= \frac{1}{4} \int_0^\infty ds \int_{-s}^s du \mathbb{L}''(s)e^{-i\omega_0 s}$$

$$= \frac{1}{2} \int_0^\infty s\mathbb{L}''(s)e^{-i\omega_0 s} \, ds$$

$$= -\frac{1}{2} \int_0^\infty \mathbb{L}'(s)e^{-i\omega_0 s} \, ds + \frac{i\omega_0}{2} \int_0^\infty s\mathbb{L}'(s)e^{-i\omega_0 s} \, ds$$

$$= -\frac{1}{2} \left[\mathbb{L}'_+(\omega_0) + \omega_0 \frac{\partial}{\partial \omega_0} \mathbb{L}'_+(\omega_0) \right], \qquad (10.30)$$

where a partial integration was used to obtain the third line. Also

$$\frac{1}{2} \int_0^\infty \int_0^\infty \mathbb{L}''(s_1 + s_2)e^{-i\omega_0(s_1 - s_2)} \, ds_1 ds_2$$

$$= \frac{1}{4} \int_0^\infty ds \int_{-s}^s du \mathbb{L}''(s)e^{-i\omega_0 u} = \frac{1}{2\omega_0}\mathbb{L}''_s(\omega_0) = -\frac{1}{2}\mathbb{L}'_c(\omega_0). \qquad (10.31)$$

11   The term where s_1 and s_2 are interchanged contributes an equal result and we
12   obtain

$$\psi_{\text{Dill}}(t) = \left\{ \frac{1}{2}[\mathbb{L}_0 + \mathbb{L}'_+(\omega_0) - \omega_0 \frac{\partial}{\partial \omega_0}\mathbb{L}'_+(\omega_0)], \mathbb{L}_0 + \mathbb{L}'_c(\omega_0) \right\}. \qquad (10.32)$$

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This also follows from (10.20) and the relation

$$\int_0^\infty \int_0^\infty \mathbb{L}'(s_1 + s_2)e^{-i\omega_0(s_1 + s_2)}\, ds_1\, ds_2 = i\frac{\partial}{\partial\omega_0}\mathbb{L}'_+(\omega_0), \tag{10.33}$$

which can be shown by the technique outlined in (10.30). Note that

$$\langle\psi_{\mathrm{Dill}}(t)\rangle_{\mathrm{av}} = \overline{\mathbf{C}_0} \odot [\mathbb{L}_0 + \mathbb{L}'_c(\omega_0)\mathbf{C}_0], \tag{10.34}$$

where the quantity $\mathbb{L}_0 + \mathbb{L}'_c(\omega_0) = \mathrm{Re}(\mathbb{L}_0 + \mathbb{L}'_+(\omega_0))$ is known in the context of classical viscoelasticity as the storage modulus. In fact, this label is sometimes applied to $\mathbb{L}'_c(\omega_0)$. By virtue of Remark 10.1, the quantity $\mathbb{L}_0 + \mathbb{L}'_c(\omega_0)$ must be non-negative if (7.24) is valid.

The inequality (7.25) for time-averaged quantities takes the form $\langle\psi_{\mathrm{Dill}}(t)\rangle_{\mathrm{av}} \leq \langle\psi_G(t)\rangle_{\mathrm{av}}$ or, from (10.27),

$$\overline{\mathbf{C}_0} \odot (\mathbb{L}_0 + \mathbb{L}'_c(\omega_0)\mathbf{C}_0 \geq \overline{\mathbf{C}_0} \odot \mathbb{L}_\infty \mathbf{C}_0, \tag{10.35}$$

which is in fact the very general requirement (2.74) for free energies, applied to the form $(7.22)_1$, with ϕ given by (3.9). Also,

$$\dot{\psi}_{\mathrm{Dill}}(t) = i\omega_0 \left\{ \mathbb{L}_0 + \mathbb{L}'_+(\omega_0) - \omega_0\frac{\partial}{\partial\omega_0}\mathbb{L}'_+(\omega_0), 0 \right\}. \tag{10.36}$$

Then, from (2.75) and (10.9),

$$D_{\mathrm{Dill}}(t) = \left\{ i\omega_0^2\frac{\partial}{\partial\omega_0}\mathbb{L}'_+(\omega_0), -2\omega_0\mathbb{L}'_s(\omega_0) \right\}, \tag{10.37}$$

which can also be shown using (7.23) and (10.33).

Next, let us consider ψ_F, given by (7.11). We have, from $(5.4)_2$,

$$\mathbf{I}(s, \mathbf{C}^t) = \mathbf{F}(\omega_0, s)\mathbf{C}_0 e^{i\omega_0 t} + \overline{\mathbf{F}}(\omega_0, s)\overline{\mathbf{C}_0}e^{-i\omega_0 t},$$

$$\mathbf{F}(\omega_0, s) = \int_0^\infty \mathbb{L}'(u + s)e^{-i\omega_0 u}\, du = e^{i\omega_0 s}\int_s^\infty \mathbb{L}'(u)e^{-i\omega_0 u}\, du, \tag{10.38}$$

$$\mathbf{F}^\top(\omega_0, s) = \mathbf{F}(\omega_0, s), \quad \overline{\mathbf{F}}(\omega_0, s) = \mathbf{F}(-\omega_0, s),$$

where $\mathbf{F} \in \mathrm{Lin}(\mathcal{B})$. The functional ψ_F has the form given by (10.13), where (see (7.13))

$$\mathbb{L}_{12+}(\omega_0, \omega_0) = \int_0^\infty \mathbf{F}_s(\omega_0, s)\mathbb{L}'^{-1}(s)\mathbf{F}_s(\omega_0, s)\, ds,$$
$$\mathbb{L}_{12+}(\omega_0, -\omega_0) = \int_0^\infty \mathbf{F}_s(\omega_0, s)\mathbb{L}'^{-1}(s)\mathbf{F}_s(-\omega_0, s)\, ds, \tag{10.39}$$

with

$$\mathbf{F}_s(\omega_0, s) = \frac{\partial}{\partial s}\mathbf{F}(\omega_0, s) = \int_0^\infty \mathbb{L}''(u + s)e^{-i\omega_0 u}\, du. \tag{10.40}$$

Also,

$$\mathbb{L}_{1+}(\omega_0, \pm\omega_0) = \int_0^\infty \mathbf{F}_s(\omega_0, s)\mathbb{L}'^{-1}(s)\mathbf{F}(\pm\omega_0, s)ds,$$

$$\mathbb{L}_{2+}(\omega_0, \pm\omega_0) = \int_0^\infty \mathbf{F}(\omega_0, s)\mathbb{L}'^{-1}(s)\mathbf{F}_s(\pm\omega_0, s)ds. \tag{10.41}$$

1   The rate of dissipation is given by (10.22), with the aid of (10.41).

2   Finally, the expression for the minimum free energy will be explored. We shall
3   require the relation

$$\overline{\mathbb{H}_\pm(\omega)} = \mathbb{H}_\pm(-\overline{\omega}), \quad \omega \in \Omega, \tag{10.42}$$

5   which generalizes $(9.9)_2$ to the complex plane. The minimum free energy $\psi_m(t)$ is
6   given by (8.15). Using (10.4), we evaluate the integral in (8.6) by closing the contour
7   on $\Omega^{(+)}$ to obtain

$$\mathbf{p}_+^t(\omega) = -\left[\frac{e^{i\omega_- t}}{i(\omega + \omega_-)}\mathbb{H}_-(-\omega_-)\mathbf{C}_0 + \frac{e^{-i\omega_+ t}}{i(\omega - \omega_+)}\mathbb{H}_-(\omega_+)\overline{\mathbf{C}_0}\right] \tag{10.43}$$

9   and

$$\mathbf{p}_-^t(\omega) = \mathbb{H}_-(\omega)\mathbf{C}_{r+}^t(\omega) + \mathbf{p}_+^t(\omega).$$

From (10.43) we obtain

$$\frac{1}{2\pi}\int_{-\infty}^\infty |\mathbf{p}_+^t(\omega)|^2 d\omega = -\frac{ie^{2i\omega_- t}}{2\omega_-}\mathbf{C}_0 \odot \mathbb{H}_-^2(-\omega_-)\mathbf{C}_0$$

$$+ \frac{ie^{-2i\omega_+ t}}{2\omega_+}\overline{\mathbf{C}_0} \odot \mathbb{H}_-^2(\omega_+)\overline{\mathbf{C}_0}$$

$$- \frac{2ie^{i(\omega_- - \omega_+)t}}{(\omega_- - \omega_+)}\overline{\mathbf{C}_0} \odot \mathbb{H}_-(\omega_+)\mathbb{H}_-(-\omega_-)\mathbf{C}_0, \tag{10.44}$$

where (10.42) has been used. It will be observed that the last term diverges in the
limit $\eta \to 0$. Recalling (8.20), we see that the quantity given by (10.44) in the limit
$\eta \to 0$ is the total dissipation over history so this divergence is an expression of a
physically obvious fact. Its derivative is the rate of dissipation and is finite. Taking
the limit $\eta \to 0$ in the convergent terms, we obtain

$$\frac{1}{2\pi}\int_{-\infty}^\infty |\mathbf{p}_+^t(\omega)|^2 d\omega = -\frac{ie^{2i\omega_0 t}}{2\omega_0}\mathbf{C}_0 \odot \mathbb{H}_-^2(-\omega_0)\mathbf{C}_0$$

$$+ \frac{ie^{-2i\omega_0 t}}{2\omega_0}\overline{\mathbf{C}_0} \odot \mathbb{H}_-^2(\omega_0)\overline{\mathbf{C}_0}$$

$$+ \overline{\mathbf{C}_0} \odot \left[2t\mathbb{H}(\omega_0) - \mathbb{D}(\omega_0) + \frac{1}{\eta}\mathbb{H}(\omega_0)\right]\mathbf{C}_0, \tag{10.45}$$

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where $\mathbb{D}(\omega)$, given by (8.42), is real and non-negative for $\omega \in \mathbb{R}$. From (10.11), (10.45), (8.18) and (8.20), taking the limit $\eta \to 0$, we obtain

$$\psi_m(t) = \left\{ \frac{1}{2}\mathbb{L}_0 + \mathbb{B}_1(\omega_0), \mathbb{L}_0 + \mathbb{B}_2(\omega_0) \right\} \tag{10.46}$$

where

$$\mathbb{B}_1(\omega_0) = \frac{1}{2}\left[\mathbb{L}'_+(\omega_0) + \frac{i}{\omega_0}\mathbb{H}^2_-(-\omega_0) \right],$$
$$\mathbb{B}_2(\omega_0) = \mathbb{L}'_c(\omega_0) - \omega_0\frac{d}{d\omega_0}\mathbb{L}'_c(\omega_0) + \mathbb{D}(\omega_0). \tag{10.47}$$

The divergent terms and those proportional to t cancel. The average over a time cycle is given by (see (10.21))

$$\langle \psi_m \rangle_{\mathrm{av}} = \overline{\mathbf{C}_0} \odot [\mathbb{L}_0 + \mathbb{B}_2(\omega_0)]\mathbf{C}_0. \tag{10.48}$$

Note that $\mathbb{L}_0 + \mathbb{B}_2$ must be a non-negative quantity for all $\omega \in \mathbb{R}$, by virtue of Remark 10.1. We have

$$\dot{\psi}_m(t) = i\omega_0\left\{ \mathbb{L}_0 + \mathbb{L}'_+(\omega_0) + \frac{i}{\omega_0}\mathbb{H}^2_-(-\omega_0), 0 \right\}. \tag{10.49}$$

The rate of dissipation is given by (8.25) and (8.22)$_3$. Using (10.4) and closing on Ω^+, we find that

$$\mathbf{K}(t) = \mathbb{H}_-(-\omega_0)\mathbf{C}_0 e^{i\omega_0 t} + \mathbb{H}_-(\omega_0)\overline{\mathbf{C}_0}e^{-i\omega_0 t} \tag{10.50}$$

on taking $\eta \to 0$. Therefore

$$D_m(t) = \{\mathbb{H}^2_-(-\omega_0), 2\mathbb{H}(\omega_0)\}. \tag{10.51}$$

One may check that (8.17) holds, using (10.9), (10.49) and (10.51). The time average $\langle D_m(t) \rangle_{\mathrm{av}}$ is equal to $\overline{\mathbf{C}_0}2\mathbb{H}(\omega_0)\mathbf{C}_0$, which is consistent with Remark 10.2, by virtue of (10.24).

The formulae derived here for the minimum free energy apply with little change to the other free energies referred to in Remark 9.2.

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