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Free Energies for Materials with Memory in Terms of State Functionals

Murrough Golden

Technological University Dublin, murrough.golden@dit.ie

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2 **Free energies for materials with memory in terms of state**
3 **functionals**

4 **J. M. Golden**

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7 **Abstract** The aim of this work is to determine what free
8 energy functionals are expressible as quadratic forms of
9 the state functional I^t which is discussed in earlier
10 papers. The single integral form is shown to include
11 the functional ψ_F proposed a few years ago, and also a
12 further category of functionals which are easily
13 described but more complicated to construct. These
14 latter examples exist only for certain types of materials.
15 The double integral case is examined in detail, against
16 the background of a new systematic approach developed
17 recently for double integral quadratic forms in terms of
18 strain history, which was used to uncover new free
19 energy functionals. However, while, in principle, the
20 same method should apply to free energies which can be
21 given by quadratic forms in terms of I^t , it emerges that
22 this requirement is very restrictive; indeed, only the
23 minimum free energy can be expressed in such a manner.

24 **Keywords** Thermodynamics · Memory effects
25 · Free energy functional · Minimal state
26 functional · Rate of dissipation

27 **1 Introduction**
29

30 Free energy functionals that are expressible as
31 quadratic forms of the state functional I^t are explored

in the present work. The quantity I^t is discussed in [1, 32
6, 7] and elsewhere. Such free energies have applica- 33
tions in proving results concerning the integro-partial 34
differential equations describing materials with mem- 35
ory. They may also be useful for physical modeling of 36
such materials. However, these applications generally 37
require that the free energy functionals involved have 38
compact, explicit analytic representation. 39

The single integral form is shown to include the 40
functional ψ_F , proposed some years ago [1, 6]. There 41
is also however a further category of functionals of this 42
kind for materials with non-singleton minimal states. 43
These functionals are easily described but more 44
difficult to construct, since basic inequalities relating to 45
thermodynamics must be explicitly imposed; they are 46
therefore not so useful for practical applications. 47

The double integral quadratic form is examined in 48
detail. In this context, a recent paper [10] deals with 49
determining new free energies that are quadratic func- 50
tionals of the history of strain, using a novel approach. 51
This new method is based on a result showing that if a 52
suitable kernel for the rate of dissipation is known, the 53
associated free energy kernel can be determined by a 54
straightforward formula, yielding a non-negative qua- 55
dratic form. It allows us to determine previously 56
unknown free energy functionals by hypothesizing rates 57
of dissipation that are non-negative, and applying the 58
formula. In particular, new free energy functionals 59
related to the minimum free energy are constructed. 60

In principle, the methods developed in [10] apply to 61
quadratic forms in terms of I^t , and should lead to new 62

A1 J. M. Golden (✉)
A2 School of Mathematical Sciences, Dublin Institute of
A3 Technology, Kevin Street, Dublin 8, Ireland
A4 e-mail: murrrough.golden@dit.ie

63 free energies which can be expressed as such quadratic
 64 forms. It emerges however that this is a very restrictive
 65 property; indeed, only the minimum free energy is
 66 expressible as such a functional.

67 Regarding the notational convention for referring to
 68 equations, we adopt the following rule. A group of
 69 relations with a single equation number (***) will be
 70 individually labeled by counting “=” signs or “<”,
 71 “>”, “≤” and “≥”. Thus, (***)₅ refers to the fifth
 72 “=” sign, if all the relations are equalities. Relations
 73 with “∈” are ignored for this purpose.

74 **2 Quadratic models for free energies**

75 As in [10], we discuss the scalar problem, denoting the
 76 independent field variable by $E(t)$, the strain function,
 77 and the dependent variable by $T(t)$, the stress function.
 78 However, it is fairly straightforward to generalize to
 79 tensor fields (for example, [1, 5]) and to certain other
 80 theories such as heat flow in rigid bodies or electro-
 81 magnetic phenomena.

82 Certain basic formulae from [10] and earlier work
 83 are repeated here for convenience. The current value
 84 of the strain function is $E(t)$ while the strain history
 85 and relative history are given by

$$E^t(s) = E(t - s), \quad E_r^t(s) = E^t(s) - E(t), \quad s \in \mathbb{R}^+.$$

(2.1)

87 It is assumed here that

$$\lim_{s \rightarrow \infty} E^t(s) = \lim_{u \rightarrow -\infty} E(u) = 0,$$

(2.2)

89 which simplifies certain formulae. The state of the
 90 material, in the most basic sense, is specified by
 91 $(E^t, E(t))$ or $(E_r^t, E(t))$. Another definition of state will
 92 be introduced in Sect. 5.1.

93 Let $T(t)$ be the stress at time t . Then the constitutive
 94 relations with linear memory terms have the form

$$\begin{aligned} T(t) &= T_e(t) + \int_0^\infty \tilde{G}(u) \dot{E}^t(u) du, \quad \tilde{G}(u) = G(u) - G_\infty, \\ &= T_e(t) + \int_0^\infty G'(u) E_r^t(u) du, \quad \dot{E}^t(u) = \frac{\partial}{\partial t} E^t(u) \\ &= -\frac{\partial}{\partial u} E^t(u) = -\frac{\partial}{\partial u} E_r^t(u), \quad \dot{E}^t(u) = -\frac{\partial}{\partial u} \dot{E}^t(u), \end{aligned}$$

(2.3)

where $T_e(t)$ is the stress function for the equilibrium
 limit, defined by the condition $E^t(s) = E(t) \quad \forall s \in \mathbb{R}^+$,
 and the quantity $G(\cdot) : \mathbb{R}^+ \mapsto \mathbb{R}^+$ is the relaxation
 function of the material. We define

$$\begin{aligned} G'(u) &= \frac{d}{du} G(u), \quad G_\infty = G(\infty), \quad G_0 = G(0), \\ \tilde{G}(0) &= G_0 - G_\infty = \tilde{G}_0. \end{aligned}$$

(2.4)

The assumption is made that

$$\tilde{G}, G' \in L^1(\mathbb{R}^+) \cap L^2(\mathbb{R}^+).$$

(2.5)

Remark 2.1 Various formulae presented here can be
 expressed either in terms of quantities related to $\tilde{G}(u)$
 and $\dot{E}^t(u)$ or $G'(u)$ and $E_r^t(u)$ ([1, 10] and earlier
 references). We shall generally use those related to
 $\tilde{G}(u)$ and $\dot{E}^t(u)$.

Let us denote a particular free energy at time t by
 $\psi(t) = \tilde{\psi}(E^t, E(t))$, where $\tilde{\psi}$ is understood to be a
 functional of E^t and a function of $E(t)$. The Graffi [11]
 conditions obeyed by any free energy are given as
 follows:

P1:

$$\frac{\partial}{\partial E(t)} \tilde{\psi}(E^t, E(t)) = \frac{\partial}{\partial E(t)} \psi(t) = T(t).$$

(2.6)

P2: For any history E^t

$$\tilde{\psi}(E^t, E(t)) \geq \tilde{\phi}(E(t)) \quad \text{or} \quad \psi(t) \geq \phi(t),$$

(2.7)

where $\phi(t)$ is the equilibrium value of the free energy
 $\psi(t)$, defined as

$$\begin{aligned} \tilde{\phi}(E(t)) &= \phi(t) = \tilde{\psi}(E^t, E(t)), \\ \text{where } E^t(s) &= E(t) \quad \forall s \in \mathbb{R}^+. \end{aligned}$$

(2.8)

Thus, equality in (2.7) is achieved for equilibrium
 conditions.

P3: It is assumed that ψ is differentiable. For any
 $(E^t, E(t))$ we have the first law

$$\dot{\psi}(t) + D(t) = T(t) \dot{E}(t),$$

(2.9)

where $D(t) \geq 0$ is the rate of dissipation of energy
 associated with $\psi(t)$.

This non-negativity requirement on $D(t)$ is an expres-
 sion of the second law.

Author Proof

131 Integrating (2.9) over $(-\infty, t]$ yields that

$$\psi(t) + \mathfrak{D}(t) = W(t), \tag{2.10}$$

133 where

$$W(t) = \int_{-\infty}^t T(u)\dot{E}(u)du, \quad \mathfrak{D}(t) = \int_{-\infty}^t D(u)du \geq 0. \tag{2.11}$$

135 We assume that these integrals are finite. The quantity
136 $W(t)$ is the work function, while $\mathfrak{D}(t)$ is the total
137 dissipation resulting from the entire history of deforma-
138 tion of the body.

139 The function $T_e(t)$ in (2.3) is given by

$$T_e(t) = \frac{\partial \phi(t)}{\partial E(t)}. \tag{2.12}$$

141 It follows that

$$\dot{\phi}(t) = T_e(t)\dot{E}(t). \tag{2.13}$$

143 For a scalar theory with a linear memory constitu-
144 tive relation defining stress, the most general form of a
145 free energy is

$$\psi(t) = \phi(t) + \frac{1}{2} \int_0^\infty \int_0^\infty \dot{E}^t(s)\tilde{G}(s, u)\dot{E}^t(u)dsdu,$$

$$\tilde{G}(s, u) = G(s, u) - G_\infty. \tag{2.14}$$

147 There is no loss of generality in taking

$$\tilde{G}(s, u) = \tilde{G}(u, s). \tag{2.15}$$

149 The Grafti condition P2, given by (2.7), requires that the
150 kernel \tilde{G} must be such that the integral term in (2.14) is
151 non-negative. Various properties of $\tilde{G}(s, u)$ are given
152 in [10] and earlier references. The relaxation function
153 $G(u)$ introduced in (2.3) is related to $G(s, u)$ by

$$G(u) = G(0, u) = G(u, 0) \quad \forall u \in \mathbb{R}^+. \tag{2.16}$$

155 Note that, with the aid of (2.4), we have

$$G(0) = G(0, 0) = G_0. \tag{2.17}$$

157 The rate of dissipation can be deduced from (2.9) and
158 (2.3) to be

$$D(t) = -\frac{1}{2} \int_0^\infty \int_0^\infty \dot{E}^t(s)K(s, u)\dot{E}^t(u)dsdu, \tag{2.18}$$

where

$$K(s, u) = G_1(s, u) + G_2(s, u). \tag{2.19}$$

The subscripts 1, 2 indicate differentiation with respect
to the first and second arguments. The quantity G must
be such that the integral in (2.18) is non-positive, as
required by P3 of the Grafti conditions. The quantity K
can also be taken to be symmetric in its arguments, *i.e.*

$$K(s, u) = K(u, s). \tag{2.20}$$

Seeking to express $\mathfrak{D}(t)$, given by (2.11)₂, as a general
quadratic functional form similar to those in (2.14) or
(2.18), we put

$$\mathfrak{D}(t) = \frac{1}{2} \int_0^\infty \int_0^\infty \dot{E}^t(s)Q(s, u)\dot{E}^t(u)dsdu. \tag{2.21}$$

2.1 The work function

This quantity, given by (2.11)₁, can be put in the form
([1, 10], p 153 and earlier references cited therein):

$$W(t) = \phi(t) + \frac{1}{2} \int_0^\infty \int_0^\infty \dot{E}^t(s)\tilde{G}(|s - u|)\dot{E}^t(u)duds. \tag{2.22}$$

We see that it has the form (2.14) where

$$\tilde{G}(s, u) = \tilde{G}(|s - u|). \tag{2.23}$$

Remark 2.2 The quantity $W(t)$ can be regarded as a
free energy, but with zero total dissipation, which is
clear from (2.10). Because of the vanishing dissipa-
tion, it must be the maximum free energy associated
with the material or greater than this quantity, an
observation which follows from (2.10).

Thus, we have in general the requirement that

$$\psi(t) \leq W(t). \tag{2.24}$$

It follows from (2.10) that $Q(s, u)$ in (2.21) is given by

$$Q(s, u) = \tilde{G}(|s - u|) - \tilde{G}(s, u), \tag{2.25}$$

so that

$$Q(s, 0) = Q(0, u) = 0, \quad \forall s, u \in \mathbb{R}^+. \tag{2.26}$$

Remark 2.3 The integral term in (2.14) and (2.21) are
in general positive-definite quadratic forms, in the

Author Proof

192 sense that they vanish only if $\dot{E}^t(u) = 0$, $u \in \mathbb{R}^+$,
 193 while $D(t)$, given by (2.18), may be positive semi-
 194 definite, so that it can vanish for non-zero histories.

195 **3 Frequency domain quantities**

196 Let Ω be the complex ω plane and

$$\Omega^+ = \{\omega \in \Omega \mid \text{Im}(\omega) \in \mathbb{R}^+\},$$

$$\Omega^{(+)} = \{\omega \in \Omega \mid \text{Im}(\omega) \in \mathbb{R}^{++}\}.$$

198 These define the upper half-plane including and
 199 excluding the real axis, respectively. Similarly, Ω^- ,
 200 $\Omega^{(-)}$ are the lower half-planes including and excluding
 201 the real axis, respectively.

202 *Remark 3.1* Throughout this work, a subscript “+”
 203 attached to any quantity defined on Ω will imply that it
 204 is analytic on Ω^- , with all its singularities in $\Omega^{(+)}$.
 205 Similarly, a subscript “-” will indicate that it is
 206 analytic on Ω^+ , with all its singularities in $\Omega^{(-)}$.

207 The notation for and properties of Fourier trans-
 208 formed quantities is specified in [1, 10] and earlier
 209 references. It is assumed that all frequency domain
 210 quantities of interest are analytic on an open set
 211 including the real axis. The functions and relations

$$\tilde{G}_+(\omega) = \int_0^\infty \tilde{G}(s)e^{-i\omega s} ds = \tilde{G}_c(\omega) - i\tilde{G}_s(\omega),$$

$$G'_+(\omega) = \int_0^\infty G'(s)e^{-i\omega s} ds = G'_c(\omega) - iG'_s(\omega)$$

$$= -\tilde{G}_0 + i\omega\tilde{G}_+(\omega) \tag{3.2}$$

213 will be required, where the quantities $\tilde{G}_c(\omega)$, $G'_c(\omega)$
 214 and $\tilde{G}_s(\omega)$, $G'_s(\omega)$ are the cosine and sine transforms
 215 of $\tilde{G}(s)$, $G'(s)$, respectively; the former quantities are
 216 even functions of ω while the latter are odd functions.
 217 It follows from (2.5) that $\tilde{G}_+(\omega), G'_+(\omega) \in L^2(\mathbb{R})$.
 218 The quantities $\tilde{G}_+(\omega)$ and $G'_+(\omega)$ are analytic in Ω^- .
 219 Because \tilde{G} is real, we have

$$\overline{\tilde{G}_+(\omega)} = \tilde{G}_+(-\bar{\omega}). \tag{3.3}$$

221 This constraint means that the singularities are sym-
 222 metric under reflection in the positive imaginary axis.

A similar relation applies to $G'_+(\omega)$. Also, we have 223

$$G''_+(\omega) = \int_0^\infty G''(s)e^{-i\omega s} ds = -G'(0) + i\omega G'_+(\omega). \tag{3.4}$$

A function of significant interest, particularly in the 225
 context of the minimum and related free energies, is 226

$$(\omega) = \omega^2 \tilde{G}_c(\omega) = -\omega G'_s(\omega) = -G''_c(\omega)$$

$$- G'(0) \geq 0, \quad \omega \in \mathbb{R}, \tag{3.5}$$

where the inequality is an expression of the second law 228
 ([1], p 159 and earlier references). The quantity $H(\omega)$ 229
 goes to zero quadratically at the origin since $H(\omega)/\omega^2$ 230
 tends to a finite, non-zero quantity $\tilde{G}_c(0)$, as ω tends to 231
 zero. One can show that 232

$$H_\infty = \lim_{\omega \rightarrow \infty} H(\omega) = -G'(0) \geq 0. \tag{3.6}$$

We assume for present purposes that $G'(0)$ is non-zero 234
 so that H_∞ is a finite, positive number. Then 235
 $H(\omega) \in \mathbb{R}^{++} \forall \omega \in \mathbb{R}, \omega \neq 0$. 236

If $G(s)$, $s \in \mathbb{R}^+$, is extended to the even function 237
 $G(|s|)$ on \mathbb{R} , then $dG(|s|)/ds$ is an odd function with 238
 Fourier transform ([1], p 144) 239

$$G'_F(\omega) = -2iG'_s(\omega) = \frac{2i}{\omega}H(\omega). \tag{3.7}$$

The non-negative quantity $H(\omega)$ can always be 241
 expressed as the product of two factors [8] 242

$$H(\omega) = H_+(\omega)H_-(\omega), \tag{3.8}$$

where $H_+(\omega)$ has no singularities or zeros in $\Omega^{(-)}$ and 244
 is thus analytic in Ω^- . Similarly, $H_-(\omega)$ is analytic in 245
 Ω^+ with no zeros in $\Omega^{(+)}$. We put [1, 8] 246

$$H_\pm(\omega) = H_\mp(-\omega) = \overline{H_\mp(\omega)},$$

$$H(\omega) = |H_\pm(\omega)|^2, \quad \omega \in \mathbb{R}. \tag{3.9}$$

The factorization (3.8) is the one relevant to the 248
 minimum free energy. For materials with only isolated 249
 singularities, we shall require a much broader class of 250
 factorizations, where the property that the zeros of 251
 $H_\pm(\omega)$ are in $\Omega^{(\pm)}$ respectively need not be true. These 252
 generate a range of free energies related to the 253
 minimum free energy [1, 7, 9], as discussed briefly 254
 in Sect. 4. 255

Author Proof

256 The Fourier transform of $E^t(s)$, $E_r^t(s)$, given by
 257 (2.1) for $s \in \mathbb{R}^+$, are defined for example in [1, 10] and
 258 denoted by $E_+^t(\omega)$, $E_{r+}^t(\omega)$. These have the same
 259 analyticity properties as $\tilde{G}_+(\omega)$. However, $E_r^t(s)$ does
 260 not have the property (2.5), so that $E_{r+}^t(\omega)$ must be
 261 defined with care. For a constant history, $E^t(s) = E(t)$,
 262 $s \in \mathbb{R}^+$, we have ([1], p 551)

$$E_+^t(\omega) = \frac{E(t)}{i\omega^-}, \quad (3.10)$$

264 where the notation ω^- (and ω^+) is defined in [1, 10]
 265 and earlier work. Briefly, $x^\pm = x \pm i\alpha$, respectively,
 266 where $\alpha \rightarrow 0^+$ after integrations are carried out. Thus,
 267 we have

$$E_{r+}^t(\omega) = E_+^t(\omega) - \frac{E(t)}{i\omega^-}. \quad (3.11)$$

269 Also ([1], p 145),

$$\frac{d}{dt}E_+^t(\omega) = \dot{E}_+^t(\omega) = -i\omega E_+^t(\omega) + E(t) = -i\omega E_{r+}^t(\omega), \quad (3.12)$$

271 and

$$\frac{d}{dt}\dot{E}_+^t(\omega) = -i\omega\dot{E}_+^t(\omega) + \dot{E}(t),$$

$$\frac{d}{dt}E_{r+}^t(\omega) = \dot{E}_{r+}^t(\omega) = -i\omega E_{r+}^t(\omega) - \frac{\dot{E}(t)}{i\omega^-}.$$

273 For large ω ,

$$E_+^t(\omega) \sim \frac{E(t)}{i\omega}, \quad E_{r+}^t(\omega) \sim \frac{A(t)}{\omega^2}, \quad (3.14)$$

275 where $A(t)$ is independent of ω . Also, from (3.12),

$$\dot{E}_+^t(\omega) \sim \frac{A(t)}{i\omega}, \quad (3.15)$$

277 for large ω . Relation (3.12) is convenient for convert-
 278 ing formulae from those in terms of $E_{r+}^t(\omega)$ to
 279 equivalent expressions in terms of $\dot{E}_+^t(\omega)$ or vice
 280 versa.

281 Applying Parseval's formula to (2.3)₁, we obtain

$$T(t) = T_e(t) + \frac{1}{2\pi} \int_{-\infty}^{\infty} \overline{\tilde{G}_+(\omega)} \dot{E}_+^t(\omega) d\omega. \quad (3.16)$$

283 There is a non-uniqueness in this form allowing us to
 284 write it as [1, 10]

$$T(t) = T_e(t) + \frac{1}{\pi} \int_{-\infty}^{\infty} \frac{H(\omega)}{\omega^2} \dot{E}_+^t(\omega) d\omega. \quad (3.17)$$

More detail is included on this argument in (5.38)– 286
 (5.40) below. 287

We shall be using the Plemelj formulae on the real 288
 axis ([1], p 542) several times in this work, in relation 289
 to frequency dependent quantities. These are given as 290
 follows. Let 291

$$F(z) = \frac{1}{2\pi i} \int_{-\infty}^{\infty} \frac{f(u)}{u-z} du, \quad z \in \Omega \setminus \mathbb{R}, \quad (3.18)$$

where $f(u)$ is any Hölder continuous function. For 293
 $z \in \Omega^{(+)}$, the function $F(z)$ is analytic in $\Omega^{(+)}$, while 294
 for $z \in \Omega^{(-)}$, it is analytic in $\Omega^{(-)}$. Let $z = x + i\alpha$, 295
 $\alpha > 0$ where α approaches zero. Then, we write (3.18) 296
 as (recall Remark 3.1) 297

$$F_-(x) = \frac{1}{2\pi i} \int_{-\infty}^{\infty} \frac{f(u)}{u-x^+} du = \frac{1}{2}f(x) + \frac{1}{2\pi i} P \int_{-\infty}^{\infty} \frac{f(u)}{u-x} du, \quad (3.19)$$

where the symbol “P” indicates a principal value 299
 integral. Similarly, 300

$$F_+(x) = \frac{1}{2\pi i} \int_{-\infty}^{\infty} \frac{f(u)}{u-x^-} du = -\frac{1}{2}f(x) + \frac{1}{2\pi i} P \int_{-\infty}^{\infty} \frac{f(u)}{u-x} du. \quad (3.20)$$

4 The minimum and related free energies 302

It is shown in [7, 9] that, for materials with only 303
 isolated singularities, the quantity $H(\omega)$ is a rational 304
 function and has many factorizations other than (3.8), 305
 denoted by 306

$$H(\omega) = H_+^f(\omega)H_-^f(\omega), \quad (4.1)$$

$$H_{\pm}^f(\omega) = H_{\mp}^f(-\omega) = \overline{H_{\mp}^f(\omega)},$$

where f is an identification label distinguishing a 308
 particular factorization. These are obtained by 309

Author Proof

310 exchanging the zeros of $H_+(\omega)$ and $H_-(\omega)$, leaving
 311 the singularities unchanged.
 312 Each factorization yields a (usually) different free
 313 energy of the form

$$\psi_f(t) = \phi(t) + \frac{1}{2\pi} \int_{-\infty}^{\infty} |p_-^f(\omega)|^2 d\omega, \quad (4.2)$$

315 where, recalling (3.12),

$$\begin{aligned} p_-^f(\omega) &= i \frac{H_-^f(\omega)}{\omega} \dot{E}_+^t(\omega) = H_-^f(\omega) E_{r+}^t(\omega) \\ &= p_-^f(\omega) - p_+^f(\omega), \\ p_{\pm}^f(\omega) &= \frac{1}{2\pi i} \int_{-\infty}^{\infty} \frac{P^f(\omega')}{\omega' - \omega^{\mp}} d\omega'. \end{aligned} \quad (4.3)$$

317 The quantity p_-^f is analytic on Ω^+ while p_+^f is analytic
 318 on Ω^- [1]. Note that (4.3) involves the use of the
 319 Plemelj formulae, as given by (3.19) and (3.20). The
 320 total dissipation is given by

$$\mathfrak{D}_f(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} |p_+^f(\omega)|^2 d\omega. \quad (4.4)$$

322 Defining

$$\begin{aligned} K_f(t) &= -\frac{1}{2\pi i} \int_{-\infty}^{\infty} \frac{H_-^f(\omega)}{\omega} \dot{E}_+^t(\omega) d\omega \\ &= \lim_{\omega \rightarrow \infty} [-i\omega p_-^f(\omega)], \end{aligned} \quad (4.5)$$

324 we can write the associated rate of dissipation in the
 325 form

$$D_f(t) = |K_f(t)|^2. \quad (4.6)$$

326 These formulae apply in particular to the case
 328 where no exchange of zeros takes place, which is
 329 denoted by $f = 1$. In this case, the formulae in fact
 330 apply to all materials, not just those characterized by
 331 isolated singularities.

332 We can write $\psi_f(t)$ in the form [1, 8–10]

$$\begin{aligned} \psi_f(t) &= \phi(t) + \frac{i}{4\pi^2} \\ &\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{\overline{\dot{E}_+^t(\omega_1)} H_+^f(\omega_1) H_-^f(\omega_2) \dot{E}_+^t(\omega_2)}{\omega_1 \omega_2 (\omega_1^+ - \omega_2^-)} d\omega_1 d\omega_2. \end{aligned} \quad (4.7)$$

The notation in the denominator [1, 10] indicates that
 if, for example, the ω_1 integration is carried out first,
 then $\omega_1^+ - \omega_2^-$ becomes $\omega_1 - \omega_2^-$. Also, the total
 dissipation (see (4.4)) can be shown, by similar
 manipulations, to have the form

$$\begin{aligned} \mathfrak{D}_f(t) &= -\frac{i}{4\pi^2} \\ &\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{\overline{\dot{E}_+^t(\omega_1)} H_+^f(\omega_1) H_-^f(\omega_2) \dot{E}_+^t(\omega_2)}{\omega_1 \omega_2 (\omega_1^- - \omega_2^+)} d\omega_1 d\omega_2, \end{aligned} \quad (4.8)$$

while $D_f(t)$, given by (4.6), can be expressed as

$$D_f(t) = \frac{1}{4\pi^2} \int_{-\infty}^{\infty} \frac{\overline{\dot{E}_+^t(\omega_1)} H_+^f(\omega_1) H_-^f(\omega_2) \dot{E}_+^t(\omega_2)}{\omega_1 \omega_2} d\omega_1 d\omega_2. \quad (4.9)$$

The factorization $f = 1$, given by (3.8), yields the
 minimum free energy $\psi_m(t)$. Each exchange of zeros,
 starting from these factors, yields a free energy which
 is greater than or equal to the previous quantity. The
 maximum free energy, denoted by $\psi_M(t)$, is obtained
 by interchanging all the zeros, which produces a
 factorization labeled $f = N$. The quantity $\psi_M(t)$ is
 less than the work function [1, 10].

The most general free energy and rate of dissipation
 arising from these factorizations is given by

$$\begin{aligned} \psi(t) &= \sum_{f=1}^N \lambda_f \psi_f(t), \quad D(t) = \sum_{f=1}^N \lambda_f D_f(t), \\ \sum_{f=1}^N \lambda_f &= 1, \quad \lambda_f \geq 0. \end{aligned} \quad (4.10)$$

A particular case of this linear form is the physical free
 energy, proposed in [9].

4.1 Discrete spectrum materials

Consider a material with relaxation function of the
 form

$$\tilde{G}(s) = \sum_{i=1}^n G_i e^{-\alpha_i s}, \quad (4.11)$$

where n is a positive integer. The inverse decay times
 $\alpha_i \in \mathbb{R}^+$, $i = 1, 2, \dots, n$ and the coefficients G_i are
 assumed to be positive. We arrange that

Author Proof

362 $\alpha_1 < \alpha_2 < \alpha_3 \dots$. These are discrete spectrum materials
 363 which will be used in later discussions.

364 From (3.2)_{1,2}, we have

$$\begin{aligned} \tilde{G}_+(\omega) &= \sum_{i=1}^n \frac{G_i}{\alpha_i + i\omega}, & \tilde{G}_c(\omega) &= \sum_{i=1}^n \frac{\alpha_i G_i}{\alpha_i^2 + \omega^2}, \\ \tilde{G}_s(\omega) &= \omega \sum_{i=1}^n \frac{G_i}{\alpha_i^2 + \omega^2}, \end{aligned} \tag{4.12}$$

366 so that $\tilde{G}_+(\omega)$ consists of a sum of simple pole terms
 367 on the positive imaginary axis. From (2.3)₁ and (4.11),
 368 we have that

$$T(t) = T_e(t) + \sum_{i=1}^n G_i \dot{E}_+^t(-i\alpha_i). \tag{4.13}$$

370 Relations (3.5) and (4.12)₂ give

$$\begin{aligned} H(\omega) &= \omega^2 \sum_{i=1}^n \frac{\alpha_i G_i}{\alpha_i^2 + \omega^2} = H_\infty - \sum_{i=1}^n \frac{\alpha_i^3 G_i}{\alpha_i^2 + \omega^2} \geq 0, \\ H_\infty &= \sum_{i=1}^n \alpha_i G_i. \end{aligned} \tag{4.14}$$

372 This quantity can be expressed in the form [8]

$$H(\omega) = H_\infty \prod_{i=1}^n \left\{ \frac{\gamma_i^2 + \omega^2}{\alpha_i^2 + \omega^2} \right\}, \tag{4.15}$$

374 where the γ_i^2 are the zeros of $f(z) = H(\omega)$, $z = -\omega^2$,
 375 and obey the relations

$$\gamma_1 = 0, \quad \alpha_1^2 < \gamma_2^2 < \alpha_2^2 < \gamma_3^2 \dots \tag{4.16}$$

377 Observe that

$$\begin{aligned} G_i &= \frac{2i}{\alpha_i^2} \lim_{\omega \rightarrow -i\alpha_i} (\omega + i\alpha_i) H(\omega) \\ &= -\frac{2i}{\alpha_i^2} \lim_{\omega \rightarrow i\alpha_i} (\omega - i\alpha_i) H(\omega). \end{aligned} \tag{4.17}$$

379 To obtain the minimum free energy for discrete
 380 spectrum materials, one chooses the factorization of
 381 (4.15) given by

$$\begin{aligned} H_+(\omega) &= h_\infty \prod_{i=1}^n \left\{ \frac{\omega - i\gamma_i}{\omega - i\alpha_i} \right\}, & h_\infty &= [H_\infty]^{1/2}, \\ H_-(\omega) &= h_\infty \prod_{i=1}^n \left\{ \frac{\omega + i\gamma_i}{\omega + i\alpha_i} \right\} = \overline{H_+}(\omega). \end{aligned} \tag{4.18}$$

383 Equations (4.18) can be written as [1, 2]

$$\begin{aligned} H_-(\omega) &= h_\infty \left[1 + i \sum_{i=1}^n \frac{U_i}{\omega + i\alpha_i} \right] = -h_\infty \omega \sum_{i=1}^n \frac{U_i}{\alpha_i(\omega + i\alpha_i)}, \\ U_i &= (\gamma_i - \alpha_i) \prod_{\substack{j=1 \\ j \neq i}}^n \left\{ \frac{\gamma_j - \alpha_i}{\alpha_j - \alpha_i} \right\}, & \sum_{i=1}^n \frac{U_i}{\alpha_i} &= -1. \end{aligned} \tag{4.19}$$

For discrete spectrum materials, the interchange of
 385 zeros referred to after (4.1) means switching a given γ_i
 386 to $-\gamma_i$ in both $H_+(\omega)$ and $H_-(\omega)$. Let us introduce an
 387 n -dimensional vector with components ϵ_i^f , $i =$
 388 $1, 2, \dots, n$ where each ϵ_i^f can take values ± 1 . We
 389 define $\rho_i^f = \epsilon_i^f \gamma_i$, and write
 390

$$H_+^f(\omega) = h_\infty \prod_{i=1}^n \left\{ \frac{\omega - i\rho_i^f}{\omega - i\alpha_i} \right\}, \quad H_-^f(\omega) = h_\infty \prod_{i=1}^n \left\{ \frac{\omega + i\rho_i^f}{\omega + i\alpha_i} \right\}. \tag{4.20}$$

The case where all the zeros are interchanged [1, 6, 7,
 392 9] is labeled $f = N$. The resulting factors are given
 393 by
 394

$$H_+^N(\omega) = h_\infty \prod_{i=1}^n \left\{ \frac{\omega + i\gamma_i}{\omega - i\alpha_i} \right\}, \quad H_-^N(\omega) = h_\infty \prod_{i=1}^n \left\{ \frac{\omega - i\gamma_i}{\omega + i\alpha_i} \right\}. \tag{4.21}$$

5 The functional I^t

5.1 Minimal states

As noted after (2.2), a viscoelastic state is defined in
 398 general by the history and current value of strain
 399 ($E^t, E(t)$). The concept of a minimal state, defined in
 400 [7] and based on the work of Noll [13] (see also for
 401 example [1, 3–5, 12]), can be expressed as follows:
 402 two viscoelastic states ($E_1^t, E_1(t)$), ($E_2^t, E_2(t)$) are
 403 equivalent or in the same equivalence class or minimal
 404 state if
 405

$$\begin{aligned} E_1(t) &= E_2(t), \int_0^\infty G^t(s + \tau) [E_1^t(s) - E_2^t(s)] ds \\ &= I^t(\tau, E_1^t) - I^t(\tau, E_2^t) = 0 \quad \forall \tau \geq 0, \\ I^t(\tau, E^t) &= \int_0^\infty G^t(s + \tau) E_r^t(s) ds = \int_0^\infty \tilde{G}(s + \tau) \dot{E}^t(s) ds \\ &= I^t(\tau). \end{aligned} \tag{5.1}$$

Author Proof

407 The abbreviated notation $I^t(\tau)$ will be used henceforth.
 408 Note the property

$$\lim_{\tau \rightarrow \infty} I^t(\tau) = 0. \tag{5.2}$$

410 It follows from (2.3)₁ and (5.1) that

$$I^t(0) = T(t) - T_e(t). \tag{5.3}$$

412 A functional of $(E^t, E(t))$ which yields the same value
 413 for all members of the same minimal state is referred
 414 to as a FMS or functional of the minimal state, or a
 415 minimal state variable. The quantity $I^t(\tau)$ is a FMS, in
 416 fact, the defining example of a FMS.

417 *Remark 5.1* A distinction between materials [1] is
 418 that for certain relaxation functions, namely those
 419 with only isolated singularities (in the frequency
 420 domain), the minimal states are non-singleton,
 421 while if some branch cuts are present in the
 422 relaxation function, the material has only singleton
 423 minimal states. For relaxation functions with only
 424 isolated singularities, there is a maximum free
 425 energy that is less than the work function $W(t)$ and
 426 also a range of related intermediate free energies, as
 427 noted in Sect. 4.

428 On the other hand, if branch cuts are present, the
 429 maximum free energy is $W(t)$ and there are no
 430 intermediate free energies of type $\psi_f(t)$.

431 *Remark 5.2* There will be some later contexts where
 432 we confine the discussion to materials with only
 433 isolated singularities, for reasons connected with the
 434 properties noted in Remark 5.1. Treating the general
 435 case of such materials is algebraically complicated [1,
 436 9], because any given singularity or zero may be of
 437 higher order. We simplify the treatment, while main-
 438 taining the essential content, by separating higher order
 439 poles or zeros into simple poles or zeros. A further
 440 simplification will be made, which also retains most
 441 essential properties,¹ by taking all the singularities and
 442 zeros on the imaginary axis. This means, in effect, that
 443 the material is a discrete spectrum material, as defined
 444 in Sect. 4.1.

Thus, we will use discrete spectrum materials as
 simple but realistic proxies for more general materials
 with only isolated singularities.

The quantities $p_-^t(\omega)$, defined by (4.3), are FMSs; in
 particular, $p_-^t(\omega)$ corresponding to the minimum free
 energy for general materials ([1], p 253). The func-
 tionals $p_+^t(\omega)$ do not have this property, by virtue of
 (4.3)₂.

Let us characterize minimal states for discrete
 spectrum materials in the following simple manner.
 Consider two states $(E_1^t, E_1(t))$ and $(E_2^t, E_2(t))$ obey-
 ing conditions (5.1), so that they are equivalent. We
 define the difference between these states as
 $(E_d^t, E_d(t))$ where

$$\begin{aligned} E_d^t(s) &= E_1^t(s) - E_2^t(s) \quad \forall s \in R^+, \\ E_d(t) &= E_1(t) - E_2(t). \end{aligned} \tag{5.4}$$

The conditions (5.1) holds for all $\tau \geq 0$ if and only if

$$\begin{aligned} E_d(t) &= 0, \quad \int_0^\infty e^{-\alpha_i s} E_d^t(s) ds = E_{d+}^t(-i\alpha_i) = 0, \\ & i = 1, 2, \dots, n. \end{aligned}$$

Remark 5.3 Therefore, for a given discrete spectrum
 material, the property that two histories are equivalent,
 or in the same minimal state, is determined by (5.5)₁
 and by the values of those histories in the frequency
 domain, at $\omega = -i\alpha_i, i = 1, 2, \dots, n$. This is a special
 case of the general requirement given in [1], p 359.

Thus, if a quantity depends on the strain history only
 through the values $E_+^t(-i\alpha_i)$ or $E_{r+}^t(-i\alpha_i)$ or (see
 (3.12)) $\dot{E}_+^t(-i\alpha_i)$, for $i = 1, 2, \dots, n$, this quantity is a
 FMS.

For discrete spectrum materials,

$$I^t(\tau) = \sum_{i=1}^n G_i \dot{E}_+^t(-i\alpha_i) e^{-\alpha_i \tau}, \tag{5.6}$$

which is an example of the property described in
 Remark 5.3. The property that $p_-^t(\omega)$ is a FMS can be
 perceived for discrete spectrum materials by complet-
 ing the contour in (4.3)₄ on $\Omega^{(-)}$.

We now present a more general characterization of
 minimal states, which leads to results consistent with
 (5.5). The condition that minimal states are non-
 singleton is that the integral equation

¹ There is a noteworthy difference between the general case
 where singularities may be off the imaginary axis and discrete
 spectrum materials, namely that in the latter case, the relaxation
 function decays monotonically, while in the former case, the
 possibility exists of oscillatory decay.

Author Proof

$$\int_0^\infty G'(s + \tau)E_d^t(s)ds = 0, \quad \tau \in \mathbb{R}^+, \quad (5.7)$$

483 for $E_d^t(s) = E_1^t(s) - E_2^t(s)$ in (5.1), has non-zero
 484 solutions. The other requirement (5.1)₁ will be
 485 enforced below by (5.17). Putting $E_d^t(s) = 0, s \in \mathbb{R}^-$
 486 and $\tau = -u$, we can write (5.7) as ([1], p 341)

$$\int_{-\infty}^\infty \frac{\partial}{\partial u} G(|u - s|)E_d^t(s)ds = 0, \quad u \in \mathbb{R}^-. \quad (5.8)$$

488 This is a Wiener–Hopf equation, which can be solved
 489 by a standard technique. We put

$$\int_{-\infty}^\infty \frac{\partial}{\partial u} G(|u - s|)E_d^t(s)ds = \begin{cases} J(u), & u \in \mathbb{R}^{++} \\ 0, & u \in \mathbb{R}^- \end{cases}, \quad (5.9)$$

491 where $J(u)$ is a quantity to be determined. Taking the
 492 Fourier transform of both sides, we obtain, with the aid
 493 of the convolution theorem and (3.7),

$$\frac{2i}{\omega} H(\omega)E_{d+}^t(\omega) = J_+(\omega). \quad (5.10)$$

495 Using (4.1) and (4.3), we can write (5.10) in the form

$$\frac{2i}{\omega} \left\{ H_+^f(\omega) \left[p_{d-}^{ft}(\omega) - p_{d+}^{ft}(\omega) \right] \right\} = J_+(\omega), \quad (5.11)$$

497 where the subscript d implies that E_{d+}^t is used in (4.3).
 498 The value of the superscript f will be assigned below.
 499 Because $p_{d-}^{ft}(\omega)$ is a FMS, we have

$$p_{d-}^{ft}(\omega) = 0. \quad (5.12)$$

501 It then follows from (5.11) that

$$p_{d+}^{ft}(\omega) = -\frac{\omega J_+(\omega)}{2i H_+^f(\omega)}. \quad (5.13)$$

503 Using (5.13) in (5.10), we obtain

$$H(\omega)E_{d+}^t(\omega) = -H_+^f(\omega)p_{d+}^{ft}(\omega), \quad (5.14)$$

505 or

$$E_{d+}^t(\omega) = -\frac{p_{d+}^{ft}(\omega)}{H_-^f(\omega)}. \quad (5.15)$$

This quantity must be analytic on Ω^- , so that all the
 507 zeros of $H_\pm(\omega)$ must have been interchanged. This is
 508 the case where $f = N$ and the resulting factors are
 509 those given by (4.21), which yield the maximum free
 510 energy $\psi_M(t)$, introduced after (4.9).
 511

Thus, if we can find a quantity $E_{d+}^t(\omega)$ which
 512 satisfies (5.12), it satisfies (5.14) and (5.15) by virtue
 513 of (4.3)₃, applied to this history difference. Relation
 514 (5.14) is equivalent to (5.10), with $J_+(\omega)$
 515 determined by (5.13). Therefore, a solution to (5.9)
 516 or (5.8) is provided by any choice of $E_d^t(s)$ where the
 517 corresponding $E_{d+}^t(\omega)$ satisfies (5.12). Now, from
 518 (4.3)₄,
 519

$$p_{d-}^{Nt}(\omega) = \frac{1}{2\pi i} \int_{-\infty}^\infty \frac{H_-^N(\omega')E_{d+}^t(\omega')}{\omega' - \omega^+} d\omega' = 0. \quad (5.16)$$

If there are non-isolated singularities in the material,
 520 we know (remark 5.1) that the only solution is
 521 the trivial one, $E_{d+}^t(\omega) = 0$. Thus, we can focus on
 522 the case of a material with only isolated singularities.
 523 The simplifying assumptions of Remark 5.2 will
 524 be adopted so that we are dealing with discrete
 525 spectrum materials. Then, $H_\pm^f(\omega)$ are given by
 526 (4.20).
 527

The simplifying assumption will now be made that
 529 $E_{d+}^t(\omega)$ is a rational function. More generally, it could
 530 also have branch cuts in $\Omega^{(+)}$.
 531

At large ω , we must have
 532

$$E_{d+}^t(\omega) \sim \frac{1}{\omega^2}, \quad (5.17)$$

by virtue of (3.14) and (5.1)₁. If the zeros of $E_{d+}^t(\omega)$
 534 cancel the poles in $H_-^N(\omega)$, given by (4.21), then, by
 535 taking the contour around $\Omega^{(-)}$, we see that (5.16) is
 536 obeyed. Thus, non-trivial solutions to (5.8) or (5.10)
 537 are given by
 538

$$E_{d+}^t(\omega) = \frac{E_0(t)}{\omega - i\chi_0} \prod_{j=1}^n \left\{ \frac{\omega + i\alpha_j}{\omega - i\chi_j} \right\} \frac{1}{\omega - i\chi_{n+1}}, \quad (5.18)$$

where the constants $\chi_i, i = 0, 1, \dots, n + 1$ indicate
 540 the positions of singularities on the imaginary
 541 axis in $\Omega^{(+)}$. These are arbitrary positive quantities.
 542 The factor $E_0(t)$, which determines the time dependence
 543 of $E_{d+}^t(\omega)$, is also arbitrary. Note that
 544

Author Proof

545 (5.18) obeys the constraints (5.5). We can write it in
546 the form

$$E_{d+}^t(\omega) = -iE_0(t) \sum_{i=0}^{n+1} \frac{A_i}{\omega - i\chi_i},$$

$$A_i = \frac{\chi_i + \alpha_i}{\chi_i - \chi_0} \prod_{\substack{j=1 \\ j \neq i}}^n \left\{ \frac{\chi_i + \alpha_j}{\chi_i - \chi_j} \right\} \frac{1}{\chi_i - \chi_{n+1}},$$

$i = 1, 2, \dots, n,$

$$A_0 = \prod_{j=1}^n \left\{ \frac{\chi_0 + \alpha_j}{\chi_0 - \chi_j} \right\} \frac{1}{\chi_0 - \chi_{n+1}},$$

$$A_{n+1} = \frac{1}{\chi_{n+1} - \chi_0} \prod_{j=1}^n \left\{ \frac{\chi_{n+1} + \alpha_j}{\chi_{n+1} - \chi_j} \right\}, \quad (5.19)$$

548 where, to satisfy (5.17), we must have

$$\sum_{i=0}^{n+1} A_i = 0. \quad (5.20)$$

550 Taking the inverse transform of (5.19)₁, we obtain
551 that

$$E_d^t(s) = E_0(t) \sum_{i=0}^{n+1} A_i e^{-\chi_i s}$$

$$= E_d^t(\chi_j, j = 0, 1, \dots, n + 1; s). \quad (5.21)$$

553 A given history $E_1^t(s)$ belongs to the minimal state
554 with members

$$E^t(\chi_j, j = 0, 1, \dots, n + 1; s) = E_1^t(s)$$

$$+ E_d^t(\chi_j, j = 0, 1, \dots, n + 1; s), \quad (5.22)$$

556 where the parameters χ_j may take any positive value.

557 If (5.7) is true for \tilde{G} given by (4.11), we must have

$$\sum_{j=0}^{n+1} \frac{A_j}{\chi_j + \alpha_i} = 0, \quad i = 1, 2, \dots, n, \quad (5.23)$$

559 which is simply a statement that $E_{d+}^t(\omega)$, given by
560 (5.19)₁, vanishes at ω equal to each $-i\chi_i$.

561 If $E_0(t)$ in (5.18) were replaced by $E_0(\omega, t)$, where
562 $\lim_{\omega \rightarrow \infty} E_0(\omega, t)$ is a non-zero finite constant, and the
563 singularities of this quantity consists of branch cuts in
564 $\Omega^{(+)}$, then the resulting $E_{d+}^t(\omega)$ would be equally
565 satisfactory, except that the simple relation (5.21)
566 would not hold.

5.2 Free energies that are FMSs, as quadratic
forms of histories for discrete spectrum
materials

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We now briefly describe a general form of free
energies that are FMSs for discrete spectrum materials
([1] and references therein). Let us define a vector \mathbf{e} in
 \mathbb{R}^n with components

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$$e_i(t) = E(t) - \alpha_i E_+^t(-i\alpha_i) = \frac{d}{dt} E_+^t(-i\alpha_i)$$

$$= \dot{E}_+^t(-i\alpha_i) = -\alpha_i E_{r+}^t(-i\alpha_i), \quad i = 1, 2, \dots, n, \quad (5.24)$$

where (3.12) has been used². As we see from (5.5), the
quantities $E_+^t(-i\alpha_i)$ are real. Consider the function

575
576

$$\psi(t) = \phi(t) + \frac{1}{2} \mathbf{e}^\top \mathbf{C} \mathbf{e} = \phi(t) + \frac{1}{2} \mathbf{e} \cdot \mathbf{C} \mathbf{e}, \quad (5.25)$$

where $\phi(t)$ is the equilibrium free energy and \mathbf{C} is a
symmetric, positive definite matrix with components
 C_{ij} , $i, j = 1, 2, \dots, n$. It is clear that $\psi(t)$ has property
P2 of a free energy, given by (2.7). For a stationary
history $E^t(s) = E(t)$, $s \in \mathbb{R}^+$, we have, from (3.10),
that $E_+^t(-i\alpha_i) = E(t)/\alpha_i$, so that $e_i(t) = 0$, $i = 1,$
 $2, \dots, n$. Relations (2.6) and (4.13) yield the condition

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$$\sum_{j=1}^n C_{ij} = G_i, \quad i = 1, 2, \dots, n. \quad (5.26)$$

From (3.13)₁ or (5.24), we have

586

$$\dot{e}_i(t) = \dot{E}(t) - \alpha_i e_i(t), \quad i = 1, 2, \dots, n, \quad (5.27)$$

so that, using (5.26), we obtain

588

$$\dot{\psi}(t) + D(t) = T(t) \dot{E}(t),$$

$$D(t) = \frac{1}{2} \mathbf{e}^\top \Gamma \mathbf{e}, \quad \Gamma_{ij} = (\alpha_i + \alpha_j) C_{ij}, \quad (5.28)$$

where Γ_{ij} are the elements of the matrix Γ . Condition
P3 (see (2.9)) requires that Γ must be at least positive
semidefinite.

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5.3 Properties of I' in the frequency domain

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Let us revert now to discussing general materials but
returning periodically to the discrete spectrum case as
an illustrative example. Some results presented here

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² Note that analytic continuation into Ω^- is straightforward
since E_+^t is analytic in this half-plane.

2FL01
2FL02

Author Proof

597 are the same as or equivalent to certain formulae given
598 previously in [1, 6]. Let

$$I'_k(\tau) = \frac{d^k}{d\tau^k} I'(\tau), \quad k = 1, 2, \dots, \quad (5.29)$$

600 so that

$$\begin{aligned} I'_1(\tau) &= \int_0^\infty G'(\tau + u) \dot{E}^t(u) du, \\ I'_2(\tau) &= \int_0^\infty G''(\tau + u) \dot{E}^t(u) du. \end{aligned} \quad (5.30)$$

602 Also,

$$\begin{aligned} \frac{\partial}{\partial t} I'_1(s) &= G'(s) \dot{E}^t(t) + I'_2(s), \\ \frac{\partial}{\partial t} I'_2(s) &= G''(s) \dot{E}^t(t) + I'_3(s). \end{aligned} \quad (5.31)$$

604 Just as in (5.2), we have

$$\lim_{\tau \rightarrow \infty} I'_k(\tau) = 0, \quad k = 1, 2, 3, \dots \quad (5.32)$$

606 The quantity $I'(s)$, $s \in \mathbb{R}$, will be required. This can be
607 defined in a number of ways. We choose the following
608 formula. Let

$$I'(s) = \int_0^\infty \tilde{G}(|s + u|) \dot{E}^t(u) du, \quad s \in \mathbb{R}. \quad (5.33)$$

610 Then

$$\begin{aligned} I'_2(s) &= \int_0^\infty \frac{\partial^2}{\partial s^2} G(|s + u|) \dot{E}^t(u) du, \\ \frac{\partial}{\partial t} I'_2(s) &= \frac{\partial^2}{\partial s^2} G(|s|) \dot{E}^t(t) + I'_3(s), \quad s \in \mathbb{R}. \end{aligned} \quad (5.34)$$

612 Note that

$$\lim_{|s| \rightarrow \infty} I'_k(s) = 0, \quad k = 1, 2, 3, \dots \quad (5.35)$$

614 We now seek to express I' in terms of frequency
615 domain quantities. Let us put

$$\tilde{G}(u) = 0, \quad \dot{E}^t(u) = 0, \quad u \in \mathbb{R}^{--}. \quad (5.36)$$

617 Then

$$\begin{aligned} \int_{-\infty}^\infty \tilde{G}(u + \tau) e^{-i\omega u} du &= \int_0^\infty \tilde{G}(v) e^{-i\omega v} dv e^{i\omega\tau} \\ &= \tilde{G}_+(\omega) e^{i\omega\tau}. \end{aligned} \quad (5.37)$$

Parseval's formula, applied to (5.1)₅, gives

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$$I'(\tau) = \frac{1}{2\pi} \int_{-\infty}^\infty \overline{\tilde{G}_+(\omega)} \dot{E}_+^t(\omega) e^{-i\omega\tau} d\omega, \quad \tau \geq 0. \quad (5.38)$$

We have

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$$I'(\tau) = \frac{1}{2\pi} \int_{-\infty}^\infty [\overline{\tilde{G}_+(\omega)} + \lambda \tilde{G}_+(\omega)] \dot{E}_+^t(\omega) e^{-i\omega\tau} d\omega, \quad (5.39)$$

for arbitrary complex values of λ , since the added term
gives zero. This can be seen by integrating over a
contour around $\Omega^{(-)}$, noting that the exponential goes
to zero as $Im\omega \rightarrow -\infty$ and using (3.15). Let us choose
 $\lambda = 1$. Then, recalling (3.5)₁, we find that

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$$\begin{aligned} I'(\tau) &= \frac{1}{\pi} \int_{-\infty}^\infty \frac{H(\omega)}{\omega^2} \dot{E}_+^t(\omega) e^{-i\omega\tau} d\omega \\ &= \frac{1}{\pi} \int_{-\infty}^\infty \frac{H(\omega)}{\omega^2} \overline{\dot{E}_+^t(\omega)} e^{i\omega\tau} d\omega, \end{aligned} \quad (5.40)$$

for $\tau \geq 0$, where the reality of I' has been used. This
relation generalizes (3.17). It follows that

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$$\begin{aligned} I'_+(\omega) &= \int_0^\infty I'(\tau) e^{-i\omega\tau} d\tau \\ &= -\frac{1}{\pi i} \int_{-\infty}^\infty \frac{H(\omega') \overline{\dot{E}_+^t(\omega')}}{(\omega')^2 (\omega' - \omega^-)} d\omega'. \end{aligned} \quad (5.41)$$

We must choose ω^- so that the integration over the
exponential converges. From (5.1)₃, it follows that
 $I'_+(\omega)$ is a FMS. Similarly, the derivatives of $I'(s)$,
given by (5.29), for $s \in \mathbb{R}^+$ are also FMSs, in
particular $I'_{1+}(\omega)$ and $I'_{2+}(\omega)$.

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For the discrete spectrum case, it follows from (5.6)
that

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$$I'_+(\omega) = -i \sum_{i=1}^n \frac{G_i \dot{E}_+^t(-i\alpha_i)}{\omega - i\alpha_i}. \quad (5.42)$$

By virtue of remark 5.3, equation (5.42) implies that
 $I'_+(\omega)$ is a FMS, which confirms for such materials the
general property stated after (5.41).

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Author Proof

643 Similarly, let I' be defined by (5.39) for $\tau < 0$. In this
 644 case, we cannot close the contour in $\Omega^{(-)}$ because the
 645 exponential diverges on this half-plane. It follows that
 646 $I'(\tau)$ depends on λ for $\tau < 0$. Let us take $\lambda = 1$ so that it
 647 is given by (5.40) for $\tau < 0$. This is equivalent to the
 648 choice given by (5.33), as may be seen by transforming
 649 the integration variable in (5.33) from u to $-u$ and using
 650 (3.7) together with the convolution theorem. Also,

$$I'_-(\omega) = \int_{-\infty}^0 I'(\tau)e^{-i\omega\tau}d\tau = \frac{1}{i\pi} \int_{-\infty}^{\infty} \frac{H(\omega')\overline{\dot{E}'_+(\omega')}}{(\omega')^2(\omega' - \omega^+)} d\omega', \quad (5.43)$$

652 and

$$I'_F(\omega) = I'_-(\omega) + I'_+(\omega) = \int_{-\infty}^{\infty} I'(\tau)e^{-i\omega\tau}d\tau = \frac{2H(\omega)\overline{\dot{E}'_+(\omega)}}{\omega^2}, \quad (5.44)$$

654 by virtue of the Plemelj formulae (3.19) and (3.20). It
 655 follows from (5.44) that I'_- is not a FMS. Also, one can
 656 deduce from (3.13)₁ and (5.44) that

$$I'_F(\omega) = i\omega I'_F(\omega) + 2 \frac{H(\omega)}{\omega^2} \dot{E}(t). \quad (5.45)$$

658 We see, using (3.6) and (3.15), that

$$I'_F(\omega) \sim \omega^{-3}, \quad (5.46)$$

660 at large ω .

661 Note that (5.44) allows us to write (3.17) in the form

$$T(t) = T_e(t) + \frac{1}{2\pi} \int_{-\infty}^{\infty} \overline{I'_F(\omega)} d\omega = T_e(t) + \frac{1}{2\pi} \int_{-\infty}^{\infty} I'_F(\omega) d\omega. \quad (5.47)$$

663 For the discrete spectrum case, we have from (4.14)₁,
 664 (5.42) and (5.44) that

$$I'_-(\omega) = I'_F(\omega) - I'_+(\omega) = i \sum_{i=1}^n \frac{G_i[\dot{E}'_+(-i\alpha_i) - \overline{\dot{E}'_+(\omega)}]}{\omega - i\alpha_i} + i \sum_{i=1}^n \frac{G_i\overline{\dot{E}'_+(\omega)}}{\omega + i\alpha_i}, \quad (5.48)$$

which is analytic on $\Omega^{(+)}$. Returning to general
 materials, we see from (5.40)₂ that

$$I'_1(\tau) = -\frac{1}{i\pi} \int_{-\infty}^{\infty} \frac{H(\omega)\overline{\dot{E}'_+(\omega)}}{\omega} e^{i\omega\tau} d\omega, \quad I'_2(\tau) = -\frac{1}{\pi} \int_{-\infty}^{\infty} H(\omega)\overline{\dot{E}'_+(\omega)} e^{i\omega\tau} d\omega, \quad \tau \geq 0. \quad (5.49)$$

Thus

$$I'_{1\pm}(\omega) = \mp \frac{1}{\pi} \int_{-\infty}^{\infty} \frac{H(\omega')\overline{\dot{E}'_+(\omega')}}{\omega'(\omega' - \omega^{\mp})} d\omega', \quad I'_{2\pm}(\omega) = \pm \frac{1}{\pi i} \int_{-\infty}^{\infty} \frac{H(\omega')\overline{\dot{E}'_+(\omega')}}{\omega' - \omega^{\mp}} d\omega', \quad (5.50)$$

$$I'_{1F}(\omega) = i\omega I'_F(\omega), \quad I'_{2F}(\omega) = -\omega^2 I'_F(\omega).$$

We have

$$I'_{2F}(\omega) = -2H(\omega)\overline{\dot{E}'_+(\omega)} = I'_{2+}(\omega) + I'_{2-}(\omega), \quad (5.51)$$

673 by virtue of (5.44) and the Plemelj formulae (3.19) and
 674 (3.20). The quantities I'_{1+} , I'_{1+} and I'_{2+} are analytic in Ω^-
 675 while I'_{1-} , I'_{1-} and I'_{2-} are analytic in Ω^+ . For the
 676 complex conjugate of these quantities, the opposite is
 677 true.

In the case of discrete spectrum materials,
 678 we have, from (5.6),
 679

$$I'_1(\tau) = -\sum_{i=1}^n \alpha_i G_i \dot{E}'_+(-i\alpha_i) e^{-\alpha_i\tau}, \quad I'_2(\tau) = \sum_{i=1}^n \alpha_i^2 G_i \dot{E}'_+(-i\alpha_i) e^{-\alpha_i\tau}, \quad (5.52)$$

and

$$I'_{1+}(\omega) = i \sum_{i=1}^n \frac{\alpha_i G_i}{\omega - i\alpha_i} \dot{E}'_+(-i\alpha_i), \quad I'_{2+}(\omega) = -i \sum_{i=1}^n \frac{\alpha_i^2 G_i}{\omega - i\alpha_i} \dot{E}'_+(-i\alpha_i). \quad (5.53)$$

Author Proof

683 The corresponding quantities $I_{1-}(\omega)$ and $I_{2-}(\omega)$ can
 684 be given in the same way as (5.48).

685 5.4 Frequency domain representation of the work
 686 function

687 The frequency domain version of (2.22) is [1, 10]

$$\begin{aligned} W(t) &= \phi(t) + \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{H(\omega)}{\omega^2} |\dot{E}_+^t(\omega)|^2 d\omega \\ &= \phi(t) + \frac{1}{8\pi} \int_{-\infty}^{\infty} \frac{\omega^2}{H(\omega)} |I_F^t(\omega)|^2 d\omega \quad (5.54) \\ &= \phi(t) + \frac{1}{8\pi} \int_{-\infty}^{\infty} \frac{|I_{2F}^t(\omega)|^2}{\omega^2 H(\omega)} d\omega, \end{aligned}$$

689 by virtue of (5.44) and (5.50)₄.

690 **6 Single integral quadratic forms in terms of I'**
 691 **derivatives**

692 Consider the functional

$$\psi(t) = \phi(t) + \frac{1}{2} \int_0^{\infty} L(\tau) [I_1^t(\tau)]^2 d\tau, \quad (6.1)$$

694 in terms of $I_1(\tau)$, defined by (5.30)₁. This quantity is
 695 assumed to be a free energy. We now explore the
 696 constraints on $L(\tau)$ imposed by this requirement.

697 The relation (2.9) must hold. Using (2.13), (5.31)₁
 698 and (5.32), we deduce that

$$\begin{aligned} \dot{\psi}(t) &= \dot{E}(t) \left[T_e(t) + \int_0^{\infty} G'(\tau)L(\tau)I_1^t(\tau)d\tau \right] \\ &+ \int_0^{\infty} I_2^t(\tau)L(\tau)I_1^t(\tau)d\tau = T(t)\dot{E}(t) \\ &- \frac{1}{2}L(0)[I_1^t(0)]^2 - \frac{1}{2} \int_0^{\infty} L'(\tau)[I_1^t(\tau)]^2 d\tau, \quad (6.2) \end{aligned}$$

700 provided that the condition

$$\int_0^{\infty} G'(\tau)L(\tau)I_1^t(\tau)d\tau = T(t) - T_e(t) \quad (6.3)$$

702 holds. With the help of (2.3), (5.3) and (5.30)₁, this can
 703 be written as

$$\begin{aligned} &\int_0^{\infty} [G'(\tau)L(\tau) + 1]I_1^t(\tau)d\tau \\ &= \int_0^{\infty} \int_0^{\infty} [G'(\tau)L(\tau) + 1]G'(\tau + u)\dot{E}^t(u)d\tau du = 0, \end{aligned} \quad (6.4)$$

705 which must be true for arbitrary histories. Let us write
 706 the resulting condition as an integral equation of the
 707 form

$$\begin{aligned} &\int_0^{\infty} G'(\tau + u)f(\tau)d\tau = 0 \quad \forall u \in \mathbb{R}^+, \\ f(\tau) &= G'(\tau)L(\tau) + 1. \end{aligned} \quad (6.5)$$

An alternative pathway to (6.5) is to express (6.1) in
 709 the form (2.14) with
 710

$$\tilde{G}(s, u) = \int_0^{\infty} G'(\tau + s)L(\tau)G'(\tau + u)d\tau, \quad (6.6)$$

and to impose the constraint (2.16), written in terms of
 712 $\tilde{G}(u)$. Condition (6.5) has the same form as (5.7),
 713 leading to
 714

$$\frac{2i}{\omega}H(\omega)f_+(\omega) = J_+(\omega), \quad (6.7)$$

716 where $J_+(\omega)$ is an unknown function, analytic in $\Omega^{(-)}$.
 717 This corresponds to (5.10).

718 If the material has only isolated singularities, taken
 719 here to be the discrete spectrum type, in accordance
 720 with remark 5.2, we see that there are many non-trivial
 721 solutions of (6.5) given by a form similar to (5.18).
 722 However, in this case, there is no reason for $f(0)$ to be
 723 zero, so that, at large ω ,

$$f_+(\omega) \sim \frac{f(0)}{i\omega}. \quad (6.8)$$

725 which differs from (5.17). Thus, we put

$$f_+(\omega) = -\frac{if_0}{\omega - i\chi_0} \prod_{j=1}^n \left\{ \frac{\omega + i\alpha_j}{\omega - i\chi_j} \right\}, \quad f_0 = f(0), \quad (6.9)$$

727 where the constants χ_i , $i = 0, 1, \dots, n$ are arbi-
 728 trary positive quantities. Also, f_0 may be chosen
 729 arbitrarily.

Author Proof

730 *Remark 6.1* The observations before (5.17) and at
 731 the end of subsection 5.1 on more general choices of
 732 $E_{d+}(\omega)$ do not apply to $f_+(\omega)$. This is because for $f(\tau)$,
 733 given by (6.5)₂, a material with only isolated singu-
 734 larities cannot have branch cuts in the Fourier
 735 transform of the quantities $G'(\tau)$ and $L(\tau)$. Thus,
 736 (6.9) is the most general form of $f_+(\omega)$ for discrete
 737 spectrum materials.

738 Note that if we choose $\chi_i = \gamma_i, i = 1, 2, \dots, n$ then

$$f_+(\omega) = -\frac{if_0 h_\infty}{(\omega - i\chi_0)H_-^N(\omega)}, \quad (6.10)$$

740 where $H_-^N(\omega)$ is given by (4.21) and χ_0 is an arbitrary
 741 non-negative quantity.

742 The quantity $f(\tau)$ is the inverse transform of $f_+(\omega)$.
 743 It follows from (6.5)₂ that

$$L(\tau) = -\frac{1}{G'(\tau)} + \frac{f(\tau)}{G'(\tau)}, \quad \tau \in \mathbb{R}^+. \quad (6.11)$$

745 We deduce from (2.9) and (6.2) that the rate of
 746 dissipation is given by

$$D(t) = \frac{1}{2}L(0)[I_1'(0)]^2 + \frac{1}{2} \int_0^\infty L'(\tau)[I_1'(\tau)]^2 d\tau. \quad (6.12)$$

748 In order that $\psi(t) - \phi(t)$ and $D(t)$ be non-negative, we
 749 must have

$$L(s) \geq 0, \quad L'(s) \geq 0, \quad \forall s \in \mathbb{R}^+. \quad (6.13)$$

751 Note that, from (4.11), the relaxation function of the
 752 material obeys the constraints

$$G'(s) \leq 0, \quad G''(s) \geq 0, \quad \forall s \in \mathbb{R}^+. \quad (6.14)$$

754 The quantity $L(\tau)$, given by (6.11), obeys (6.13) if

$$f(s) \leq 1, \quad \frac{f'(s)}{f(s) - 1} \geq \frac{G''(s)}{G'(s)}, \quad \forall s \in \mathbb{R}^+. \quad (6.15)$$

756 If the free energies of the form (6.1) are to exist, based
 757 on (6.5)₂ with $f(s)$ non-zero, we must show that the set
 758 of functions $f(\cdot)$, obeying the conditions (6.15), is not
 759 empty. We can write (6.9) in the form

$$f_+(\omega) = -if_0 \sum_{i=0}^n \frac{B_i}{\omega - i\chi_i},$$

$$B_i = \frac{\chi_i + \alpha_i}{\chi_i - \chi_0} \prod_{\substack{j=1 \\ j \neq i}}^n \left\{ \frac{\chi_i + \alpha_j}{\chi_i - \chi_j} \right\}, \quad i = 1, 2, \dots, n,$$

$$B_0 = \prod_{j=1}^n \left\{ \frac{\chi_0 + \alpha_j}{\chi_0 - \chi_j} \right\}, \quad \sum_{i=0}^n B_i = 1, \quad (6.16)$$

where the last relation follows from (6.8). Taking the
 inverse Fourier transform of (6.16)₁, we obtain that

$$f(s) = f_0 \sum_{i=0}^n B_i e^{-\chi_i s}, \quad s \in \mathbb{R}^+. \quad (6.17)$$

It may be confirmed from (6.16) that a relation similar
 to (5.23) holds. The coefficients B_i alternate in sign, so
 that $f(s)$ and $f'(s)$ may take both positive and negative
 values. However, by taking $|f_0|$ to be sufficiently small,
 we can ensure that (6.15)₁ holds, as may be seen by the
 following argument. Let

$$f(s) = f_0 [T_1(s) - T_2(s)],$$

$$T_1(s) = \sum_{B_i > 0} B_i e^{-\chi_i s}, \quad T_2(s) = -\sum_{B_i < 0} B_i e^{-\chi_i s}. \quad (6.18)$$

Both $T_1(s)$ and $T_2(s)$ are positive quantities, decaying
 monotonically to zero at large s . Let $f_0 > 0$ ($f_0 < 0$).
 Then, if we choose

$$f_0 \leq \frac{1}{T_1(0)} \left(|f_0| \leq \frac{1}{T_2(0)} \right), \quad (6.19)$$

condition (6.15)₁ holds. We choose f_0 so that $f(s) < 1$,
 $s \in \mathbb{R}^+$ by choosing the inequalities in (6.19) to be
 strict. It follows that

$$M_1 = \min_{s \in \mathbb{R}^+} |f_0 [T_1(s) - T_2(s)] - 1| > 0. \quad (6.20)$$

Now, from (4.11), we have

$$-\frac{G''(s)}{G'(s)} \in [a, b] \quad \forall s \in \mathbb{R}^+, \quad (6.21)$$

where a, b are positive quantities, obeying $a < b$. Let
 $f_0 > 0$. We put

$$f'(s) = f_0 [-T_3(s) + T_4(s)],$$

$$T_3(s) = \sum_{B_i > 0} B_i \chi_i e^{-\chi_i s} \geq 0, \quad T_4(s) = -\sum_{B_i < 0} B_i \chi_i e^{-\chi_i s} \geq 0. \quad (6.22)$$

784 Then (6.15)₂ is satisfied if

$$\frac{f_0[T_3(s) - T_4(s)]}{|f_0[T_1(s) - T_2(s)] - 1|} > -a, \tag{6.23}$$

786 or

$$f_0[T_3(s) - T_4(s)] > -a|f_0[T_1(s) - T_2(s)] - 1|. \tag{6.24}$$

788 This will be true if

$$f_0[T_3(s) - T_4(s)] > -aM_1. \tag{6.25}$$

790 where M_1 is defined by (6.20). Let

$$M_2 = \min_{s \in \mathbb{R}^+} [T_3(s) - T_4(s)]. \tag{6.26}$$

792 If $M_2 \geq 0$, then (6.24) holds. If $M_2 < 0$, we choose

$$f_0 < a \frac{M_1}{|M_2|}, \tag{6.27}$$

794 to ensure that (6.15)₂ holds. If $f_0 < 0$, we define

$$M_2 = \min_{s \in \mathbb{R}^+} [T_4(s) - T_3(s)]. \tag{6.28}$$

796 and (6.27) is replaced by

$$|f_0| < a \frac{M_1}{|M_2|}. \tag{6.29}$$

798 For materials where $n = 1$, all free energies which are
 799 FMSs reduce to the same form [2]. It can be shown
 800 easily that for $L(\tau)$ given by (6.31) below, the
 801 functional defined in (6.1) has this form, so that the
 802 extra quadratic form involving $f(\tau)$ cannot contribute.
 803 We see that (6.17) is given by

$$\begin{aligned} f(s) &= f_0[B_0 e^{-\chi_0 s} + B_1 e^{-\chi_1 s}], \\ B_0 &= -\frac{\chi_0 + \alpha}{\chi_1 - \chi_0}, \quad B_1 = \frac{\chi_1 + \alpha}{\chi_1 - \chi_0}, \\ B_0 &= 1 - B_1, \quad B_1 > 1, \end{aligned} \tag{6.30}$$

805 for $n = 1$. Using (5.52)₁, it is straightforward to show
 806 that the resulting contribution to (6.1) indeed vanishes.

807 If the material has branch cut singularities, then
 808 $f(\tau) = 0$, $\tau \in \mathbb{R}^+$ is the only solution of (6.5), so that

$$L(\tau) = -\frac{1}{G'(\tau)}, \quad \tau \in \mathbb{R}^+, \tag{6.31}$$

810 and the only possibility for a free energy given by a
 811 single integral quadratic form is the quantity ψ_F ,
 812 introduced in [6]. This functional and the associated
 813 rate of dissipation have the forms

$$\psi_F(t) = \phi(t) - \frac{1}{2} \int_0^\infty \frac{[I'_1(\tau)]^2}{G'(\tau)} d\tau, \tag{6.32}$$

and

$$\begin{aligned} D_F(t) &= -\frac{1}{2} \frac{[I'_1(0)]^2}{G'(0)} - \frac{1}{2} \int_0^\infty \left[\frac{d}{d\tau} \frac{1}{G'(\tau)} \right] [I'_1(\tau)]^2 d\tau \\ &= -\frac{1}{2} \frac{[I'_1(0)]^2}{G'(0)} + \frac{1}{2} \int_0^\infty G''(\tau) \left[\frac{I'_1(\tau)}{G'(\tau)} \right]^2 d\tau. \end{aligned} \tag{6.33}$$

These quantities are non-negative and $\psi_F(t)$ is a valid
 free energy if conditions (6.14) hold, not only for
 materials with branch point singularities, but for all
 materials. It is a relatively simple functional, conven-
 nient for applications.

For materials with only isolated singularities, a more
 general choice of $L(s)$, given by (6.11), also produces
 valid free energy functionals, provided that the
 inequalities (6.15) are enforced. This can be done by
 ensuring that f_0 obeys (6.19) and (6.27) or (6.29), for
 any given choices of the quantities χ_i , $i = 0, 1, \dots, n$.
 The necessity to enforce such conditions renders these
 choices less convenient for practical applications.

7 Double integral quadratic forms in terms of I' derivatives: time domain representations

We now discuss double integral quadratic forms for
 free energies and rates of dissipation. The time domain
 formulation is explored in this section, while the
 corresponding frequency domain relations are pre-
 sented in the next.

Consider the form

$$\psi(t) = \phi(t) + \frac{1}{2} \int_0^\infty \int_0^\infty I'_2(s)L(s,u)I'_2(u)dsdu, \tag{7.1}$$

There is no loss of generality in putting

$$L(s,u) = L(u,s). \tag{7.2}$$

The assumptions

$$\begin{aligned} L(\cdot, \cdot) &\in L^1(\mathbb{R}^+ \times \mathbb{R}^+) \cap L^2(\mathbb{R}^+ \times \mathbb{R}^+), \\ \lim_{s \rightarrow \infty} L(s,u) &= \lim_{s \rightarrow \infty} L(u,s) = 0 \end{aligned} \tag{7.3}$$

Author Proof

843 will be adopted. It is understood that $L(s, u)$ vanishes
 844 for negative values of s and u . We have from (2.13)
 845 and (5.31)₂ that

$$\begin{aligned} \dot{\psi}(t) = \dot{E}(t) & \left[T_e(t) + \frac{1}{2} \int_0^\infty \int_0^\infty G''(s)L(s, u)I_2^t(u)dsdu \right. \\ & \left. + \frac{1}{2} \int_0^\infty \int_0^\infty I_2^t(s)L(s, u)G''(u)dsdu \right] \\ & + \frac{1}{2} \int_0^\infty \int_0^\infty I_3^t(s)L(s, u)I_2^t(u)dsdu \\ & + \frac{1}{2} \int_0^\infty \int_0^\infty I_2^t(s)L(s, u)I_3^t(u)dsdu. \end{aligned} \tag{7.4}$$

847 It is assumed that

$$L(0, u) = L(s, 0) = 0. \tag{7.5}$$

849 This property greatly simplifies the next step of the
 850 argument, making possible an analogy with the history
 851 based formalism presented in [10].

852 The two integrals in brackets in (7.4) can be shown
 853 to be equal by interchanging integration variables.
 854 Applying partial integrations and using (5.32), we
 855 obtain

$$\begin{aligned} \dot{\psi}(t) = \dot{E}(t) & \left[T_e(t) + \int_0^\infty \int_0^\infty G''(s)L(s, u)I_2^t(u)dsdu \right] \\ & - \frac{1}{2} \int_0^\infty \int_0^\infty I_2^t(s)[L_1(s, u) + L_2(s, u)]I_2^t(u)dsdu. \end{aligned} \tag{7.6}$$

857 It is assumed in general that

$$\int_0^\infty \int_0^\infty G''(s)L(s, u)I_2^t(u)dsdu = \int_0^\infty \tilde{G}(s)\dot{E}^t(s)ds, \tag{7.7}$$

859 for arbitrary choices of histories. Using (5.30)₂, this
 860 leads to the condition

$$\int_0^\infty \int_0^\infty G''(s)L(s, u)G''(u+v)dsdu = \tilde{G}(v). \tag{7.8}$$

This can also be derived in an alternative manner. We
 observe from (2.14), (5.30)₂ and (7.1) that

$$\tilde{G}(s, u) = \int_0^\infty \int_0^\infty G''(s+s_1)L(s_1, u_1)G''(u_1+u)ds_1du_1. \tag{7.9}$$

This relation corresponds to (6.6). Applying (2.16)
 gives (7.8). Let

$$m(u) = \int_0^\infty G''(s)L(s, u)ds, \tag{7.10}$$

noting that $m(0) = 0$, by virtue of (7.5). Then, with the
 aid of a partial integration, (7.8) can be expressed as

$$\begin{aligned} \int_0^\infty G'(s+u)f(u)du & = 0, \quad \forall s \in \mathbb{R}^+, \\ f(u) = 1 - m'(u) & = 1 - \int_0^\infty G''(s)L_2(s, u)ds \\ & = 1 + \int_0^\infty G'(s)L_{12}(s, u)ds, \end{aligned} \tag{7.11}$$

which corresponds to (6.5). Note that Remark 6.1 also
 applies here. Referring to (2.3)₁ and (2.9), equation
 (7.6) can be written as

$$\begin{aligned} \dot{\psi}(t) + D(t) & = T(t)\dot{E}(t), \\ D(t) & = \frac{1}{2} \int_0^\infty \int_0^\infty I_2^t(s)R(s, u)I_2^t(u)dsdu, \end{aligned} \tag{7.12}$$

$$R(s, u) = L_1(s, u) + L_2(s, u) = R(u, s).$$

The kernels $L(s, u)$ and $R(s, u)$ must be such as to
 render the integral terms in (7.1) and (7.12)₂ non-
 negative.

The work function cannot be expressed in terms of
 $I_2^t(s)$, $s \geq 0$, but can be given in terms of this quantity
 for $s \in \mathbb{R}$. This follows from the frequency represen-
 tation (5.54). We write

$$W(t) = \phi(t) + \frac{1}{2} \int_{-\infty}^\infty I_2^t(s)J(|s-u|)I_2^t(u)dsdu, \tag{7.13}$$

where the kernel $J(|u|)$ is related to the inverse
 transform of the kernel in (5.54)₃. Convergence issues
 in this context must be handled carefully.

Author Proof

886 It follows from (2.10) that the total dissipation must
 887 also depend on $I_2^t(s)$, $s \in \mathbb{R}$. We write

$$\mathfrak{D}(t) = \frac{1}{2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} I_2^t(s) V(s, u) I_2^t(u) ds du,$$

$$V(s, u) = V(u, s), \tag{7.14}$$

889 where, to satisfy (2.10), we must have

$$V(s, u) = \begin{cases} J(|s - u|), & s < 0 \text{ or } u < 0, \\ -L(s, u) + J(|s - u|), & s > 0 \text{ and } u > 0. \end{cases} \tag{7.15}$$

891 Note that $V(s, u)$ is continuous at $s = 0$ and $u = 0$.
 892 Also,

$$V_1(s, u) + V_2(s, u) = -L_1(s, u) - L_2(s, u) = -R(s, u). \tag{7.16}$$

894 Differentiating (7.14) with respect to time and using
 895 (5.34)₂, we obtain

$$\dot{\mathfrak{D}}(t) = D(t), \tag{7.17}$$

897 where $D(t)$ is given by (7.12), provided that

$$\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{\partial^2}{\partial s^2} G(|s|) V(s, u) I_2^t(u) ds du = 0. \tag{7.18}$$

899 This condition must hold for arbitrary histories, which
 900 yields

$$\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{\partial^2}{\partial s^2} G(|s|) V(s, u) \frac{\partial^2}{\partial u^2} G(|u + v|) ds du = 0,$$

$$v \in \mathbb{R}^+. \tag{7.19}$$

902 We see that $Q(s, u)$ in (2.21) is given by

$$Q(s, u) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{\partial^2}{\partial s^2} G(|s + s_1|) V(s_1, u_1)$$

$$\frac{\partial^2}{\partial u^2} G(|u_1 + u|) ds_1 du_1, \tag{7.20}$$

904 so that (7.19) is equivalent to (2.26).

905 Relationships (7.13)–(7.20) are incomplete without
 906 specifying the forms of the kernels more precisely.
 907 This is difficult in the time domain. The natural
 908 framework for a deeper treatment of such issues is the
 909 frequency domain, as is clear from (5.54), and will be
 910 further demonstrated in Sect. 8.

7.1 Free energy kernel in terms of the dissipation kernel 911
 912

Results were obtained in [10] which allowed the 913
 kernel of the quadratic form (2.14) to be determined in 914
 terms of the kernel of (2.18). A corresponding theory 915
 was also given in terms of frequency domain quanti- 916
 ties, which proved more useful for applications. We 917
 now adapt this method to apply to functionals that are 918
 quadratic in I^t . It will emerge that the new technique 919
 does not lead to new free energies. However, it is 920
 useful in the context of dealing with the minimum free 921
 energy. 922

Let us treat (7.12)₃ as a first order partial differential 923
 equation for $L(s, u)$, $s, u \in \mathbb{R}^+$, where $R(s, u)$, $s, u \in 924$
 \mathbb{R}^+ is presumed to be known. We introduce new 925
 variables, 926

$$x = s + u \geq 0, \quad y = s - u, \tag{7.21}$$

in terms of which (7.12)₃ becomes 928

$$\frac{\partial}{\partial x} L_n(x, y) = \frac{1}{2} R_n(x, y), \quad L_n(x, y) = L(s, u),$$

$$R_n(x, y) = R(s, u), \tag{7.22}$$

with general solution 930

$$L_n(x, y) = L_n(x_0, y) + \frac{1}{2} \int_{x_0}^x R_n(x', y) dx' \tag{7.23}$$

where x_0 is an arbitrary non-negative real quantity. It 932
 follows from (7.2) and (7.12)₄ that 933

$$L_n(x, y) = L_n(x, -y) = L_n(x, |y|),$$

$$R_n(x, y) = R_n(x, -y) = R_n(x, |y|). \tag{7.24}$$

Observe that, by virtue of (7.5), 935

$$L_n(u, u) = L_n(u, -u) = L_n(u, |u|) = 0, \quad u \in \mathbb{R}^+. \tag{7.25}$$

Putting 937

$$x' = s' + u' \geq 0, \quad y = s' - u' = s - u, \tag{7.26}$$

we have 939

$$s' = \frac{1}{2}(x' + y), \quad u' = \frac{1}{2}(x' - y),$$

$$R_n(x', y) = R\left(\frac{1}{2}(x' + y), \frac{1}{2}(x' - y)\right), \tag{7.27}$$

so that (7.23) and (7.25) give 941

Author Proof

$$L(s, u) = L_n(x, y) = \frac{1}{2} \int_{|y|}^x R_n(x', y) dx'$$

$$= \int_0^{\min(s, u)} R(s - v, u - v) dv, \tag{7.28}$$

943 which, as expected, obeys (7.5). Relation (7.1) gives

$$\psi(t) = \phi(t) + \frac{1}{2} \int_0^\infty \int_0^\infty I_2^t(s)$$

$$\int_0^{\min(s, u)} R(s - v, u - v) dv I_2^t(u) ds du$$

$$= \phi(t) + \frac{1}{2} \int_0^\infty \int_0^\infty \int_0^\infty I_2^t(s) R(s - v, u - v) I_2^t(u) dv ds du, \tag{7.29}$$

945 since $R(s - v, u - v) = 0$ for $v > \min(s, u)$. Let us
 946 assume that we have chosen $R(\cdot, \cdot)$ so that $D(t)$, given
 947 by (7.12)₂, is non-negative for any choice of I_2^t . For
 948 $v \geq 0$ and arbitrary choices of I_2^t , we have

$$\int_0^\infty \int_0^\infty I_2^t(s) R(s - v, u - v) I_2^t(u) ds du$$

$$= \int_0^\infty \int_0^\infty I_2^t(s_1 + v) R(s_1, u_1) I_2^t(u_1 + v) ds_1 du_1$$

$$= \int_0^\infty \int_0^\infty f(s_1) R(s_1, u_1) f(u_1) ds_1 du_1 \geq 0, \tag{7.30}$$

950 where $f(s_1) = I_2^t(s_1 + v)$ and is therefore arbitrary. It
 951 follows that the integral in (7.29)₂ is also non-
 952 negative. Therefore, $L(\cdot, \cdot)$, given by (7.28), has the
 953 property that the integral term in (7.1) is non-negative.
 954 Thus, the basic strategy developed in [10] is valid here
 955 also. The idea is to assign $R(\cdot, \cdot)$ so that the rate of
 956 dissipation is non-negative. Then, the associated free
 957 energy, *i.e.* that with kernel given by (7.28), also has
 958 the required positivity property. It will emerge how-
 959 ever that the strategy developed in [10] is not useful in
 960 the present case, except in the context of the minimum
 961 free energy.

We note the similarity between the expression 962
 (7.28) and the kernel of the expression for the total 963
 dissipation in [10]. 964

8 Double integral quadratic forms in terms of I^t derivatives: frequency domain representations 965
 966

The initial results presented here are analogous to 967
 those in [10]. We define 968

$$L_{+-}(\omega_1, \omega_2) = \int_0^\infty \int_0^\infty L(s, u) e^{-i\omega_1 s + i\omega_2 u} ds du$$

$$= \overline{L_{+-}}(\omega_2, \omega_1),$$

$$R_{+-}(\omega_1, \omega_2) = \int_0^\infty \int_0^\infty R(s, u) e^{-i\omega_1 s + i\omega_2 u} ds du$$

$$= \overline{R_{+-}}(\omega_2, \omega_1),$$

$$V_F(\omega_1, \omega_2) = \int_{-\infty}^\infty \int_{-\infty}^\infty V(s, u) e^{-i\omega_1 s + i\omega_2 u} ds du$$

$$= \overline{V_F}(\omega_2, \omega_1), \tag{8.1}$$

where L is introduced in (7.1), R is defined by (7.12)₃ 970
 and V by (7.15). The functions $L_{+-}(\omega_1, \omega_2)$ and 971
 $R_{+-}(\omega_1, \omega_2)$ are analytic in the lower half of the ω_1 972
 complex plane and in the upper half of the ω_2 plane. 973
 The quantity $V_F(\omega_1, \omega_2)$ may have singularities 974
 anywhere in the ω_1 and ω_2 complex planes. Inverting 975
 Fourier transforms in (8.1) yields that 976

$$L(s, u) = \frac{1}{4\pi^2} \int_{-\infty}^\infty \int_{-\infty}^\infty L_{+-}(\omega_1, \omega_2) e^{i\omega_1 s - i\omega_2 u} d\omega_1 d\omega_2,$$

$$R(s, u) = \frac{1}{4\pi^2} \int_{-\infty}^\infty \int_{-\infty}^\infty R_{+-}(\omega_1, \omega_2) e^{i\omega_1 s - i\omega_2 u} d\omega_1 d\omega_2,$$

$$V(s, u) = \frac{1}{4\pi^2} \int_{-\infty}^\infty \int_{-\infty}^\infty V_F(\omega_1, \omega_2) e^{i\omega_1 s - i\omega_2 u} d\omega_1 d\omega_2. \tag{8.2}$$

Note that, for complex values of the frequencies, 978

$$\overline{L_{+-}(\omega_1, \omega_2)} = L_{+-}(-\overline{\omega_1}, -\overline{\omega_2}) = L_{+-}(\overline{\omega_2}, \overline{\omega_1}), \tag{8.3}$$

Author Proof

980 with analogous relations for $R_{+-}(\omega_1, \omega_2)$ and
 981 $V_F(\omega_1, \omega_2)$. We define

$$L_0(s) = L_1(0, s) = L_2(s, 0),$$

$$R(s, 0) = R(0, s) = R(s) = L_0(s),$$

$$L_{0+}(\omega) = \int_0^\infty L_0(s)e^{-i\omega s} ds, \tag{8.4}$$

$$R_+(\omega) = \int_0^\infty R(s)e^{-i\omega s} ds = L_{0+}(\omega).$$

983 Relations (7.5) and (7.12)₃ have been used in deriving
 984 these connections. We have

$$\lim_{\omega \rightarrow \infty} i\omega L_{0+}(\omega) = L_0(0) = R(0, 0). \tag{8.5}$$

986 Equations (7.5), (7.12)₃ and (8.1) give

$$i(\omega_1 - \omega_2)L_{+-}(\omega_1, \omega_2) = R_{+-}(\omega_1, \omega_2), \tag{8.6}$$

988 which yields

$$L_{+-}(\omega_1, \omega_2) = \frac{R_{+-}(\omega_1, \omega_2)}{i(\omega_1^- - \omega_2^+)}, \tag{8.7}$$

990 on using the notation of (4.8). This choice, rather than
 991 that in (4.7), is dictated by the analytic properties of
 992 $L_{+-}(\omega_1, \omega_2)$. We refer to the analogous formula for
 993 the kernel of the total dissipation in [10].

994 Also

$$i(\omega_1 - \omega_2)V_F(\omega_1, \omega_2) = -R_{+-}(\omega_1, \omega_2), \tag{8.8}$$

996 by virtue of (7.16). This gives an equation for
 997 $V_F(\omega_1, \omega_2)$ similar to (8.7) for $L_{+-}(\omega_1, \omega_2)$. The
 998 question which arises is whether the quantity in the
 999 denominator is $\omega_1^- - \omega_2^+$, as in (8.7), or $\omega_1^+ - \omega_2^-$.
 1000 These are the only two possibilities. What they mean
 1001 respectively is specified after (4.7). Now, the first
 1002 choice would yield a quadratic form for the total
 1003 dissipation equal to the negative of the integral term in
 1004 the expression for the free energy (see (8.19) below).
 1005 This would yield a meaningless result, so we take

$$V_F(\omega_1, \omega_2) = -\frac{R_{+-}(\omega_1, \omega_2)}{i(\omega_1^+ - \omega_2^-)}. \tag{8.9}$$

1007 Another derivation of this result is given below; see
 1008 (8.21).

1009 Relation (8.1)₂ and the asymptotic behaviour of
 1010 Fourier transforms [1, 10] yield that

$$R_{+-}(\omega_1, \omega_2) \sim \begin{cases} \frac{L_{0+}(\omega_1)}{-i\omega_2} & \text{as } \omega_2 \rightarrow \infty, \\ \frac{L_{0+}(\omega_2)}{i\omega_1} & \text{as } \omega_1 \rightarrow \infty, \end{cases} \tag{8.10}$$

where $L_{0+}(\omega)$ is defined in (8.4). It follows from (8.7) that

$$L_{+-}(\omega_1, \omega_2) \sim \begin{cases} -\frac{L_{0+}(\omega_1)}{\omega_2^2} & \text{as } \omega_2 \rightarrow \infty, \\ -\frac{L_{0+}(\omega_2)}{\omega_1^2} & \text{as } \omega_1 \rightarrow \infty. \end{cases} \tag{8.11}$$

The asymptotic behaviour of $V_F(\omega_1, \omega)$ is similar to (8.11), by virtue of (8.9). The condition corresponding to (7.5) is

$$\int_{-\infty}^\infty L_{+-}(\omega_1, \omega) d\omega_1 = 0 \tag{8.12}$$

$$= \int_{-\infty}^\infty L_{+-}(\omega, \omega_2) d\omega_2 = 0 \quad \forall \omega \in \mathbb{R},$$

which follows from Cauchy's theorem and (8.11).

It is shown in [10] that the free energy, the rate of dissipation and total dissipation, in terms of histories, are given by

$$\begin{aligned} \psi(t) &= \phi(t) + \frac{1}{8\pi^2} \int_{-\infty}^\infty \int_{-\infty}^\infty \overline{\dot{E}_+^t(\omega_1)} \tilde{G}_{+-}(\omega_1, \omega_2) \\ &\quad \dot{E}_+^t(\omega_2) d\omega_1 d\omega_2, \\ D(t) &= -\frac{1}{8\pi^2} \int_{-\infty}^\infty \int_{-\infty}^\infty \overline{\dot{E}_+^t(\omega_1)} K_{+-}(\omega_1, \omega_2) \dot{E}_+^t(\omega_2) d\omega_1 d\omega_2, \\ \mathfrak{D}(t) &= \frac{1}{8\pi^2} \int_{-\infty}^\infty \int_{-\infty}^\infty \overline{\dot{E}_+^t(\omega_1)} Q_{+-}(\omega_1, \omega_2) \dot{E}_+^t(\omega_2) d\omega_1 d\omega_2, \\ &= \frac{i}{8\pi^2} \int_{-\infty}^\infty \int_{-\infty}^\infty \frac{\overline{\dot{E}_+^t(\omega_1)} K_{+-}(\omega_1, \omega_2) \dot{E}_+^t(\omega_2)}{\omega_1^- - \omega_2^+} d\omega_1 d\omega_2, \end{aligned} \tag{8.13}$$

where $\tilde{G}_{+-}(\omega_1, \omega_2)$, $K_{+-}(\omega_1, \omega_2)$ and $Q_{+-}(\omega_1, \omega_2)$ are the Fourier transforms of $\tilde{G}(s, u)$ in (2.14), $K(s, u)$ in (2.18), (2.19) and $Q(s, u)$ in (2.21). These are Fourier transforms as defined in (8.1).

We can write the frequency domain version of (7.12)₂ in the form

$$\begin{aligned}
 D(t) &= \frac{1}{8\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \overline{I_{2+}^t}(\omega_1) R_{+-}(\omega_1, \omega_2) \\
 &\quad I_{2+}^t(\omega_2) d\omega_1 d\omega_2 \\
 &= \frac{1}{8\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \overline{I_{2F}^t}(\omega_1) R_{+-}(\omega_1, \omega_2) \\
 &\quad I_{2F}^t(\omega_2) d\omega_1 d\omega_2 \\
 &= \frac{1}{8\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \overline{I_F^t}(\omega_1) \omega_1^2 \omega_2^2 R_{+-}(\omega_1, \omega_2) \\
 &\quad I_F^t(\omega_2) d\omega_1 d\omega_2.
 \end{aligned}
 \tag{8.14}$$

1031 where I_{2+}^t, I_F^t and I_{2F}^t are defined in (5.50)_{2,4} and (5.44)
 1032 respectively. The second form of (8.14) relies on
 1033 (5.51) and the fact that

$$\begin{aligned}
 &\int_{-\infty}^{\infty} R_{+-}(\omega_1, \omega_2) I_{2-}^t(\omega_2) d\omega_2 \\
 &= \int_{-\infty}^{\infty} \overline{I_{2-}^t}(\omega_1) R_{+-}(\omega_1, \omega_2) d\omega_1 = 0,
 \end{aligned}
 \tag{8.15}$$

1035 which are consequences of (8.10) and Cauchy's
 1036 theorem. Using (5.44)₃, we can write (8.14)₃ as

$$\begin{aligned}
 D(t) &= \frac{1}{2\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \dot{E}_+^t(\omega_1) H(\omega_1) H(\omega_2) \\
 &\quad R_{+-}(\omega_1, \omega_2) \overline{\dot{E}_+^t}(\omega_2) d\omega_1 d\omega_2 \\
 &= \frac{1}{2\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \overline{\dot{E}_+^t}(\omega_1) H(\omega_1) H(\omega_2) \\
 &\quad R_{+-}(\omega_2, \omega_1) \dot{E}_+^t(\omega_2) d\omega_1 d\omega_2,
 \end{aligned}
 \tag{8.16}$$

1038 on interchanging integration variables. Comparing
 1039 with (8.13)₂, we deduce that

$$\begin{aligned}
 -4H(\omega_1)H(\omega_2)R_{+-}(\omega_2, \omega_1) &= K_{+-}(\omega_1, \omega_2) \\
 +k_{2+}(\omega_1, \omega_2) + k_{1-}(\omega_1, \omega_2),
 \end{aligned}
 \tag{8.17}$$

1041 where $k_{2+}(\omega_1, \omega_2)$ has singularities on the ω_2 com-
 1042 plex plane only in $\Omega^{(+)}$ and $k_{1-}(\omega_1, \omega_2)$ has singular-
 1043 ities on the ω_1 plane only in $\Omega^{(-)}$. They must also

decay to zero at large ω_1, ω_2 but are otherwise
 arbitrary. This is an expression of the non-uniqueness
 of the kernels in the frequency domain, which is
 explored in [10], and which indeed apply to
 $R_{+-}(\omega_1, \omega_2)$ and $L_{+-}(\omega_1, \omega_2)$ in the present context.
 Using such non-uniqueness leads however to kernels
 that do not have the analytic properties possessed by
 R_{+-} and L_{+-} .

By analogy with (8.14) and (8.15), the frequency
 domain version of (7.1) takes the forms

$$\begin{aligned}
 \psi(t) &= \phi(t) + \frac{1}{8\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \overline{I_{2+}^t}(\omega_1) L_{+-}(\omega_1, \omega_2) \\
 &\quad I_{2+}^t(\omega_2) d\omega_1 d\omega_2 \\
 &= \phi(t) + \frac{1}{8\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \overline{I_{2F}^t}(\omega_1) L_{+-}(\omega_1, \omega_2) \\
 &\quad I_{2F}^t(\omega_2) d\omega_1 d\omega_2 \\
 &= \phi(t) + \frac{1}{8\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \overline{I_F^t}(\omega_1) \omega_1^2 \omega_2^2 L_{+-}(\omega_1, \omega_2) \\
 &\quad I_F^t(\omega_2) d\omega_1 d\omega_2.
 \end{aligned}
 \tag{8.18}$$

Note the all free energies and dissipations of the form
 (8.13) are expressible as quadratic forms in $I_F^t(\omega)$, by
 virtue of (5.44). However, in general, the analytic
 properties of the resulting kernels will not be given as
 in (8.14) and (8.18), so that the special forms (8.14)₁
 and (8.18)₁ do not hold. It follows from (8.7) and
 (8.18) that

$$\begin{aligned}
 \psi(t) &= \phi(t) - \frac{i}{8\pi^2} \\
 &\quad \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{\overline{I_{2+}^t}(\omega_1) R_{+-}(\omega_1, \omega_2) I_{2+}^t(\omega_2)}{\omega_1^- - \omega_2^+} d\omega_1 d\omega_2 \\
 &= \phi(t) - \frac{i}{8\pi^2} \\
 &\quad \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{\overline{I_{2F}^t}(\omega_1) R_{+-}(\omega_1, \omega_2) I_{2F}^t(\omega_2)}{\omega_1^- - \omega_2^+} d\omega_1 d\omega_2.
 \end{aligned}
 \tag{8.19}$$

By virtue of the result proved in subsection 7.1, if R_{+-}
 is such that $D(t)$, given by (8.14), is non-negative, then

Author Proof

1065 $\psi(t) - \phi(t)$, given by (8.19), is also non-negative. Let
 1066 us use (3.19) with respect to the integral in (8.19)₂ over
 1067 ω_1 to obtain

$$\begin{aligned} \psi(t) &= \phi(t) - \frac{i}{8\pi^2} P \\ &\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{\overline{I_{2F}^t(\omega_1)} R_{+-}(\omega_1, \omega_2) I_{2F}^t(\omega_2)}{\omega_1 - \omega_2} d\omega_1 d\omega_2 \\ &+ \frac{1}{8\pi} \int_{-\infty}^{\infty} \overline{I_{2F}^t(\omega)} R_{+-}(\omega, \omega) I_{2F}^t(\omega) d\omega. \end{aligned} \tag{8.20}$$

1069 The frequency domain version of (7.14), combined
 1070 with (8.9), yields

$$\begin{aligned} \mathfrak{D}(t) &= \frac{i}{8\pi^2} \\ &\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{\overline{I_{2F}^t(\omega_1)} R_{+-}(\omega_1, \omega_2) I_{2F}^t(\omega_2)}{\omega_1^+ - \omega_2^-} d\omega_1 d\omega_2 \\ &= \frac{i}{8\pi^2} P \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{\overline{I_{2F}^t(\omega_1)} R_{+-}(\omega_1, \omega_2) I_{2F}^t(\omega_2)}{\omega_1 - \omega_2} d\omega_1 d\omega_2 \\ &+ \frac{1}{8\pi} \int_{-\infty}^{\infty} \overline{I_{2F}^t(\omega)} R_{+-}(\omega, \omega) I_{2F}^t(\omega) d\omega. \end{aligned} \tag{8.21}$$

1072 Alternatively, we can obtain this result by substituting
 1073 for $K_{+-}(\omega_1, \omega_2)$ in (8.13)₄ from (8.17), noting that
 1074 $k_{2+}(\omega_1, \omega_2)$ and $k_{1-}(\omega_1, \omega_2)$ do not contribute. This
 1075 expression cannot be reduced to a quadratic form in
 1076 $I_{2+}^t(\omega)$.

1077 Relations (8.20), (8.21) and (5.54)₃ give (2.10) or

$$\begin{aligned} \psi(t) + \mathfrak{D}(t) &= \phi(t) + \frac{1}{4\pi} \\ &\int_{-\infty}^{\infty} \overline{I_{2F}^t(\omega)} R_{+-}(\omega, \omega) I_{2F}^t(\omega) d\omega = W(t), \end{aligned} \tag{8.22}$$

1079 provided we put

$$R_{+-}(\omega, \omega) = \frac{1}{2\omega^2 H(\omega)}, \tag{8.23}$$

1081 which is similar to a relation for $K_{+-}(\omega, \omega)$, derived in
 1082 [10]. Indeed, it can be seen from (8.17) that the two

conditions are consistent if and only if $k_{2+}(\omega, \omega)$ 1083
 $+ k_{1-}(\omega, \omega) = 0$. Furthermore, if $R_{+-}(\omega_1, \omega_2)$ 1084
 is replaced by an equivalent kernel, using the non- 1085
 uniqueness arguments referred to after (8.17), then 1086
 (8.23) is typically no longer valid. 1087

From (5.45), (8.14)_{2,3} and (5.50)₄, we obtain 1088

$$\begin{aligned} \mathfrak{D}(t) &= D(t) = \frac{1}{8\pi^2} \\ &\int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \overline{I_{2F}^t(\omega_1)} R_{+-}(\omega_1, \omega_2) I_{2F}^t(\omega_2) d\omega_1 d\omega_2, \end{aligned} \tag{8.24}$$

if 1090

$$\begin{aligned} \frac{i}{8\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{H(\omega_1) R_{+-}(\omega_1, \omega_2) I_{2F}^t(\omega_2)}{\omega_1^+ - \omega_2^-} d\omega_1 d\omega_2 \\ + \frac{i}{8\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{\overline{I_{2F}^t(\omega_1)} R_{+-}(\omega_1, \omega_2) H(\omega_2)}{\omega_1^+ - \omega_2^-} d\omega_1 d\omega_2 = 0. \end{aligned} \tag{8.25}$$

The two terms on the left are complex conjugates of 1092
 each other, and can be shown to be individually real, so 1093
 that we can express this condition as 1094

$$\frac{i}{8\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{H(\omega_1) R_{+-}(\omega_1, \omega_2) I_{2F}^t(\omega_2)}{\omega_1^+ - \omega_2^-} d\omega_1 d\omega_2 = 0. \tag{8.26}$$

Let us apply (3.20) to the integral over ω_1 in (8.26). 1096
 This gives, with the aid of (8.23) and (5.50)₄, 1097

$$\begin{aligned} \frac{i}{8\pi^2} P \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{H(\omega_1) R_{+-}(\omega_1, \omega_2) I_{2F}^t(\omega_2)}{\omega_1 - \omega_2} d\omega_1 d\omega_2 \\ = -\frac{1}{8\pi} \int_{-\infty}^{\infty} H(\omega) R_{+-}(\omega, \omega) I_{2F}^t(\omega) d\omega \\ = \frac{1}{16\pi} \int_{-\infty}^{\infty} I_F^t(\omega) d\omega \end{aligned} \tag{8.27}$$

It follows from (8.19)₂, (5.45) and (2.13) that 1099

Author Proof

$$\begin{aligned} \dot{\psi}(t) &= -\frac{1}{8\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \overline{I'_{2F}}(\omega_1) R_{+-}(\omega_1, \omega_2) \\ &I'_{2F}(\omega_2) d\omega_1 d\omega_2 + \dot{E}(t) \left[T_e(t) + \frac{i}{2\pi^2} \right. \\ &\left. \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{H(\omega_1) R_{+-}(\omega_1, \omega_2) I'_{2F}(\omega_2)}{\omega_1^- - \omega_2^+} d\omega_1 d\omega_2 \right], \end{aligned} \tag{8.28}$$

where the reality of the last integral has been invoked. Since (2.9) or (7.12)₁ must be satisfied, we require that

$$\begin{aligned} \frac{i}{2\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{H(\omega_1) R_{+-}(\omega_1, \omega_2) I'_{2F}(\omega_2)}{\omega_1^- - \omega_2^+} d\omega_1 d\omega_2 \\ = \frac{1}{2\pi} \int_{-\infty}^{\infty} I'_F(\omega) d\omega = [T(t) - T_e(t)] \dot{E}(t), \end{aligned} \tag{8.29}$$

by virtue of (5.47). Now, using (3.19), we find that

$$\begin{aligned} \frac{i}{2\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{H(\omega_1) R_{+-}(\omega_1, \omega_2) I'_{2F}(\omega_2)}{\omega_1^- - \omega_2^+} d\omega_1 d\omega_2 \\ = \frac{i}{2\pi^2} P \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{H(\omega_1) R_{+-}(\omega_1, \omega_2) I'_{2F}(\omega_2)}{\omega_1 - \omega_2} d\omega_1 d\omega_2 \\ + \frac{1}{2\pi} \int_{-\infty}^{\infty} H(\omega) R_{+-}(\omega, \omega) I'_{2F}(\omega) d\omega \\ = \frac{i}{2\pi^2} P \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{H(\omega_1) R_{+-}(\omega_1, \omega_2) I'_{2F}(\omega_2)}{\omega_1 - \omega_2} d\omega_1 d\omega_2 \\ + \frac{1}{4\pi} \int_{-\infty}^{\infty} I'_F(\omega) d\omega. \end{aligned} \tag{8.30}$$

Using (8.27), we see that (8.29) is satisfied.

Of the relations (8.23), (8.25) and (8.29), any two implies the third.

We can show directly that (8.29) is the frequency domain equivalent of (7.7). Using (8.2)₁ and (5.47), we can write (7.7) as

$$\begin{aligned} \frac{1}{4\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \overline{G''_+}(\omega_1) L_{+-}(\omega_1, \omega_2) \\ I'_{2+}(\omega_2) d\omega_1 d\omega_2 = \frac{1}{2\pi} \int_{-\infty}^{\infty} I'_F(\omega) d\omega. \end{aligned} \tag{8.31}$$

With the help of (8.11), (8.12) and the property 1113

$$\int_{-\infty}^{\infty} G''_+(\omega_1) L_{+-}(\omega_1, \omega_2) d\omega_1 = 0, \tag{8.32}$$

which follows by closing the integral on $\Omega^{(-)}$, we conclude from (3.5) that $\overline{G''_+}(\omega_1)$ can be replaced by $-2H(\omega_1)$. Also, we can replace I'_{2+} by I'_{2F} , as concluded in relation to (8.18). Thus, the left-hand side of (8.31) becomes 1115
1116
1117
1118
1119

$$\begin{aligned} -\frac{1}{2\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} H(\omega_1) L_{+-}(\omega_1, \omega_2) I'_{2F}(\omega_2) d\omega_1 d\omega_2 \\ = \frac{i}{2\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{H(\omega_1) R_{+-}(\omega_1, \omega_2) I'_{2F}(\omega_2)}{\omega_1^- - \omega_2^+} d\omega_1 d\omega_2, \end{aligned} \tag{8.33}$$

where (8.7) has been invoked. Therefore, (8.31) is equivalent to (8.29). 1121
1122

Similarly, we can show, using (8.9), that (8.26) is the frequency domain equivalent of (7.18). 1123
1124

We can write (8.29) in the form 1125

$$\begin{aligned} \frac{1}{2\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} H(\omega_1) L_{+-}(\omega_1, \omega_2) \omega_2^2 \\ I'_F(\omega_2) d\omega_1 d\omega_2 = \frac{1}{2\pi} \int_{-\infty}^{\infty} I'_F(\omega) d\omega, \end{aligned} \tag{8.34}$$

with the aid of (5.50)₄. 1127

Let us now explore possible solutions of (8.34), leading to new free energies. This equation must be true for an arbitrary history, so that, on using (5.44), we obtain the relations 1128
1129
1130
1131

$$\frac{1}{\pi} \int_{-\infty}^{\infty} H(\omega_1) L_{+-}(\omega_1, \omega) H(\omega) d\omega_1 = \frac{H(\omega)}{\omega^2} + S_-(\omega), \tag{8.35}$$

1133 where $S_-(\omega)$ is an arbitrary function that is analytic in
 1134 Ω^+ and goes to zero at infinity, since, by Cauchy's theorem,

$$\int_{-\infty}^{\infty} S_-(\omega) \overline{\dot{E}_+^t(\omega)} d\omega = 0. \tag{8.36}$$

1136 Recall that (7.8) has the same relationship with (7.7)
 1137 that (8.35) has with (8.34).

1138 The frequency version of (7.11) has the same form
 1139 as (8.35) and indeed (6.7). Comparing these latter two
 1140 equations, we see that

$$\begin{aligned} \overline{f_+}(\omega) &= \frac{\omega}{\pi i} \int_{-\infty}^{\infty} H(\omega_1) L_{+-}(\omega_1, \omega) d\omega_1 - \frac{1}{i\omega^+} \\ &= -\frac{\omega}{\pi} \int_{-\infty}^{\infty} \frac{H(\omega_1) R_{+-}(\omega_1, \omega)}{\omega_1 - \omega^+} d\omega_1 - \frac{1}{i\omega^+}, \\ S_-(\omega) &= -\frac{1}{2} \overline{J_+}(\omega). \end{aligned} \tag{8.37}$$

1142 Relations (8.37)_{1,2} and (8.23) are constraints on
 1143 $L_{+-}(\omega_1, \omega)$ and $R_{+-}(\omega_1, \omega)$, which derive from
 1144 (7.11) or ultimately (2.16).

1145 The quantity $f_+(\omega)$ is given by (6.9) for discrete
 1146 spectrum materials, and is zero if the material has
 1147 branch points.

1148 Alternatively, we can argue that (8.26) must be true
 1149 for arbitrary history $\overline{\dot{E}_+^t(\omega)}$, so that, instead of (8.35),
 1150 we have

$$\frac{1}{i\pi} \int_{-\infty}^{\infty} \frac{H(\omega_1) R_{+-}(\omega_1, \omega) H(\omega)}{\omega_1 - \omega^-} d\omega_1 = S_-(\omega), \tag{8.38}$$

1152 and (8.37)₂ is replaced by

$$\overline{f_+}(\omega) = -\frac{\omega}{\pi} \int_{-\infty}^{\infty} \frac{H(\omega_1) R_{+-}(\omega_1, \omega)}{\omega_1 - \omega^-} d\omega_1. \tag{8.39}$$

1154 Using (8.23), (3.19) and (3.20), we see that (8.39) is
 1155 equivalent to (8.37)₂.

1156 **9 Quadratic forms for $\psi_f(t)$ in terms of I^f**

1157 Consider the quadratic forms (4.7) and (4.9). These
 1158 can be replaced by quadratic forms in terms of $I_{2F}^f(\omega)$,

using (5.51)₁. The question discussed in this section is: 1159
 can they be expressed as quadratic forms in $I_{2+}^f(\omega)$, 1160
 which would provide examples of (8.14)₁ and (8.19)₁ 1161
 or, in the time domain, (7.1) and (7.12)₂. It emerges in 1162
 Sect. 9.1 that only the minimum free energy $\psi_m(t)$ 1163
 corresponding to $f = 1$ can be expressed in such a 1164
 manner. This property of $\psi_m(t)$ is discussed in detail in 1165
 Sect. 9.2. 1166

This is consistent with the fact that $\psi_m(t)$ is a FMS. 1167
 However, it is also true that all the $\psi_f(t)$ are FMSs. It 1168
 will be shown how this property holds even though the 1169
 $\psi_f(t)$ for $f > 1$ are not expressible as quadratic func- 1170
 tionals of $I_{2+}^f(\omega)$ or in the time domain, $I_2^f(s)$, $s > 0$. 1171

9.1 Quadratic forms for $\psi_f(t)$ 1172

We will base our discussion on (4.2) and (4.3). 1173
 Referring to (4.3) and (5.51), we put 1174

$$p^{ft}(\omega) = \frac{iH_-^f(\omega)}{\omega} \dot{E}_+^t(\omega) = \left[\frac{1}{2i\omega^- H_+^f(\omega)} \right] \overline{[I_{2F}^f(\omega)]}. \tag{9.1}$$

There is no singularity at $\omega = 0$ because of the factor 1176
 ω^2 in $I_{2F}^f(\omega)$, given by (5.50)₄. The superscript on ω^- 1177
 is chosen for convenience. The last form of p^{ft} is the 1178
 product of two functions both in $L^2(\mathbb{R})$. For $f = 1$, the 1179
 first factor has all its singularities in $\Omega^{(+)}$, by virtue of 1180
 the property that the zeros of H_+^f are in $\Omega^{(+)}$. However, 1181
 for other values of f , the zeros of H_+^f can be in $\Omega^{(+)}$ or 1182
 $\Omega^{(-)}$. Using (5.51)₂, we obtain 1183

$$p^{ft}(\omega) = \frac{1}{2i\omega^- H_+^f(\omega)} \overline{[I_{2+}^f(\omega) + I_{2-}^f(\omega)]} \tag{9.2}$$

The quantity $p^{ft}(\omega)$ in (4.2) and (4.3) will now be 1185
 considered in more detail. Let us write 1186

$$\frac{1}{2i\omega^- H_+^f(\omega)} = A_+(\omega) + A_-(\omega), \tag{9.3}$$

where, as indicated by the notation, $A_{\pm}(\omega)$ has all its 1188
 singularities in $\Omega^{(\pm)}$ respectively. For discrete spec- 1189
 trum materials, $H_+^f(\omega)$ is given by (4.20) and 1190

$$\begin{aligned} \frac{1}{H_+^f(\omega)} &= \frac{1}{h_\infty} + \sum_{i=1}^n \frac{V_i^f}{\omega - i\rho_i^f}, \\ V_i^f &= \lim_{\omega \rightarrow i\rho_i^f} \frac{\omega - i\rho_i^f}{H_+^f(\omega)}, \quad i = 1, 2, \dots, n. \end{aligned} \tag{9.4}$$

Author Proof

1192 Thus, $2i\omega A_+(\omega)$ is equal to the sum of terms with
 1193 $\rho_i^f = +\gamma_i$ and $2i\omega A_-(\omega)$ consists of terms where
 1194 $\rho_i^f = -\gamma_i$.

1195 If $f = 1$, then $A_-(\omega)$ will vanish, while for $f = N$
 1196 (yielding the maximum free energy referred to after
 1197 (4.9); see also remark 7.1 of [10] and [1], p 343) $A_+(\omega)$
 1198 is zero. For all values of f , $p_{\pm}^{(f)}(\omega)$ will be given by (4.3)
 1199 with

$$p^{(f)}(\omega') = A_+(\omega')\overline{I_{2+}^f(\omega')} + A_-(\omega')\overline{I_{2+}^f(\omega')} \\ + A_+(\omega')\overline{I_{2-}^f(\omega')} + A_-(\omega')\overline{I_{2-}^f(\omega')}.$$

(9.5)

1201 The relation for $p_-^{(f)}(\omega)$ can be simplified to give

$$p_-^{(f)}(\omega) = \frac{1}{2\pi i} \int_{-\infty}^{\infty} \frac{A_+(\omega')\overline{I_{2+}^f(\omega')} + A_-(\omega')\overline{I_{2+}^f(\omega')} + A_-(\omega')\overline{I_{2-}^f(\omega')}}{\omega' - \omega^+} d\omega' \\ = \frac{1}{2\pi i} \int_{-\infty}^{\infty} \frac{A_+(\omega')\overline{I_{2+}^f(\omega')} + A_-(\omega')\overline{I_{2F}^f(\omega')}}{\omega' - \omega^+} d\omega'.$$

(9.6)

1203 The first form follows by observing that if we evaluate
 1204 the term with $A_+(\omega')\overline{I_{2-}^f(\omega')}$ by closing the contour on
 1205 $\Omega^{(-)}$ then, by Cauchy's theorem, the result is zero.

1206 Consider the second form. For the case of the
 1207 minimum free energy, only the first term of the
 1208 integrand is non-zero and it follows immediately that
 1209 $\psi_m(t)$ can be expressed as a quadratic form in $I_{2+}^f(\omega)$,
 1210 as noted above.

1211 We now seek to show that $p_-^{(f)}(\omega)$ (and therefore
 1212 $\psi_f(t)$) is a FMS even if $f > 1$, for which the second
 1213 term in the denominator of (9.6)₂ is non-zero. The
 1214 argument will be presented for discrete spectrum
 1215 materials (Remark 5.2) but is in fact more general.

1216 The first term in (9.6)₂ contributes a sum of simple
 1217 poles at the points $-i\alpha_l$, $l = 1, 2, \dots, n$ by virtue of
 1218 (5.53)₂, in an expression involving $\dot{E}_+^f(\omega)$ evaluated
 1219 only at $\omega = -i\alpha_l$. This can be seen by closing the
 1220 contour on $\Omega^{(-)}$. In the second term, the singularities
 1221 of $A_-(\omega')$ are cancelled by $\overline{I_{2F}^f(\omega')}$ because of the
 1222 factor $H(\omega')$ in this quantity, defined by (5.51). This
 1223 can be shown by using (9.4) to evaluate $A_-(\omega)$, and by
 1224 taking the product of $H_{\pm}^f(\omega)$, given by (4.20). The

cancellation would not be manifest if $\overline{I_{2F}^f}$ were 1225
 expressed in terms of $\overline{I_{2\pm}^f}$. Closing on $\Omega^{(-)}$ again, we 1226
 find that the only contributing singularities are those at 1227
 $-i\alpha_i$ in $H(\omega)$, in spite of the fact that $\overline{I_{2F}^f}$ is not a FMS. 1228
 One again obtains an expression where the only 1229
 dependence on $\dot{E}_+^f(\omega)$ is through $\dot{E}_+^f(-i\alpha_j)$, 1230
 $j = 1, 2, \dots, n$, as required by Remark 5.3. 1231

1232 However, the point we wish to emphasize here is 1232
 that $p_-^{(f)}$ for $f \neq 1$ or $f \neq N$ is linear in both $\overline{I_{2+}^f}$ and $\overline{I_{2F}^f}$, 1233
 so that ψ_f is quadratic in these quantities, as we see 1234
 from (4.2). 1235

1236 One could also have approached the above argu- 1236
 ment from another point of view, by expressing (4.7) 1237
 as a quadratic functional in I_{2F}^f , using (5.51). With the 1238
 aid of arguments similar to those after (9.6), one again 1239
 obtains a quadratic functional of I_{2+}^f and I_{2F}^f . This 1240
 approach is developed explicitly for the minimum free 1241
 energy in Sect. 9.2. 1242

1243 These quadratic functionals can be expressed also 1243
 in terms of time domain quantities, as shown for the 1244
 minimum free energy in Sect. 9.2. 1245

1246 For $f = N$, giving the maximum free energy, the 1246
 quadratic form depends only on I_{2F}^f . 1247

1248 Thus, for all linear combinations of the $\psi_f(t)$ 1248
 involving terms with $f > 1$, we need to include $\overline{I_{2F}^f}$, 1249
 and the property of being a FMS is dependent on a 1250
 special cancellation, which is a specific property of the 1251
 kernel associated with those given by (4.10), where at 1252
 least one λ_f for $f > 1$ is non-zero. This will not 1253
 necessarily hold for a quadratic form in I_{2+}^f and I_{2F}^f 1254
 with a general kernel. 1255

9.2 The minimum free energy as an explicit 1256
 functional of I^f 1257

1258 It has already been shown in subsection 9.1 that the 1258
 minimum free energy can be expressed as a quadratic 1259
 form in $I_{2+}^f(\omega)$ or $I_2^f(\tau)$, $\tau \in \mathbb{R}^+$. Derivations of the 1260
 explicit form of this functional were given in [1, 6]. 1261
 We give a different derivation of this result here. Also, 1262
 we show that the conditions (8.23) and (8.29) are 1263
 obeyed. 1264

1265 Consider firstly the frequency domain representa- 1265
 tion. Recalling (5.51), we can write (4.7)–(4.9) (for 1266
 $f = 1$, corresponding to the minimum free energy) in 1267
 the form (after exchanging ω_1 and ω_2) 1268

Author Proof

$$\begin{aligned} \psi_m(t) &= \phi(t) - \frac{i}{8\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{\overline{I_{2F}^-(\omega_1)} R_{m+-}(\omega_1, \omega_2) I_{2F}^+(\omega_2)}{\omega_1^- - \omega_2^+} d\omega_1 d\omega_2, \\ D_m(t) &= \frac{1}{8\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \overline{I_{2F}^-(\omega_1)} R_{m+-}(\omega_1, \omega_2) I_{2F}^+(\omega_2) d\omega_1 d\omega_2, \\ \mathfrak{D}_m(t) &= \frac{i}{8\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{\overline{I_{2F}^-(\omega_1)} R_{m+-}(\omega_1, \omega_2) I_{2F}^+(\omega_2)}{\omega_1^+ - \omega_2^-} d\omega_1 d\omega_2, \\ R_{m+-}(\omega_1, \omega_2) &= \frac{1}{2\omega_1^- H_+(\omega_1) \omega_2^+ H_-(\omega_2)}. \end{aligned} \tag{9.7}$$

1270 The quantity $R_{m+-}(\omega_1, \omega_2)$ is analytic with respect to
 1271 ω_1 in Ω^+ and with respect to ω_2 in Ω^- . We now
 1272 replace I_{2F}^+ in these two relations by the right-hand side
 1273 of (5.51)₂. It follows from Cauchy's theorem, by
 1274 closing the contour on $\Omega^{(+)}$, that

$$\int_{-\infty}^{\infty} \frac{R_{m+-}(\omega_1, \omega_2) I_{2-}^-(\omega_2)}{\omega_1^- - \omega_2} d\omega_2 = 0. \tag{9.8}$$

1276 Similarly, $\overline{I_{2-}^-(\omega_1)}$ may be dropped from (9.7)₁ on
 1277 integration over ω_1 and we obtain

$$\begin{aligned} \psi_m(t) &= \phi(t) - \frac{i}{8\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{\overline{I_{2+}^-(\omega_1)} R_{m+-}(\omega_1, \omega_2) I_{2+}^+(\omega_2)}{\omega_1^- - \omega_2^+} d\omega_1 d\omega_2 \\ &= \phi(t) + \frac{1}{8\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \overline{I_{2+}^-(\omega_1)} L_{m+-}(\omega_1, \omega_2) I_{2+}^+(\omega_2) d\omega_1 d\omega_2, \\ L_{m+-}(\omega_1, \omega_2) &= \frac{R_{m+-}(\omega_1, \omega_2)}{i(\omega_1^- - \omega_2^+)}, \end{aligned} \tag{9.9}$$

1279 which is the explicit quadratic form implied by (9.6)
 1280 for $f = 1$. A similar argument yields that

$$\begin{aligned} D_m(t) &= \frac{1}{4\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \overline{I_{2+}^-(\omega_1)} R_{m+-}(\omega_1, \omega_2) I_{2+}^+(\omega_2) d\omega_1 d\omega_2 \\ &= \frac{1}{4\pi^2} \left| \int_{-\infty}^{\infty} \frac{I_{2+}^-(\omega)}{2\omega^+ H_-(\omega)} d\omega \right|^2 \\ &= \frac{1}{4\pi^2} \left| \int_{-\infty}^{\infty} \frac{I_{2F}^-(\omega)}{2\omega H_-(\omega)} d\omega \right|^2. \end{aligned} \tag{9.10}$$

Observe that (8.23) is true for (9.7)₄. 1282

Consider now the time domain representations. We 1283
 seek to express $D_m(t)$ and $\psi_m(t)$ as quadratic func- 1284
 tionals of $I^t(s)$, $s \in \mathbb{R}^+$. Let us define the quantity 1285
 $M(s)$ by 1286

$$M(s) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{1}{2i\omega^- H_+(\omega)} e^{i\omega s} d\omega, \quad s \in \mathbb{R}. \tag{9.11}$$

This is a real quantity which vanishes for $s \in \mathbb{R}^{--}$. 1288
 The integrand has a quadratic singularity near the 1289
 origin, due to the explicit pole term and the factor ω in 1290
 $H_+(\omega)$ which is taken, for consistency, to be ω^- . This 1291
 gives a finite contribution. 1292

Let us write the time domain version of (9.9)₂ in the 1293
 form 1294

$$\psi_m(t) = \phi(t) + \frac{1}{2} \int_0^{\infty} \int_0^{\infty} I_2^t(u) L_m(u, v) I_2^t(v) dudv, \tag{9.12}$$

corresponding to (7.1), where $L_m(u, v)$ is given by 1296
 (8.2)₁ in terms of $L_{+-}(\omega_1, \omega_2)$. The rate of dissipation 1297
 given by (9.10) becomes, in the time domain, (c.f. 1298
 (4.6)) 1299

$$D_m(t) = |K(t)|^2, \quad K(t) = \int_0^{\infty} M(u) I_2^t(u) du, \tag{9.13}$$

on using Parseval's formula. Therefore 1301

$$\begin{aligned} D_m(t) &= \left| \int_0^{\infty} M(u) I_2^t(u) du \right|^2 \\ &= \int_0^{\infty} \int_0^{\infty} I_2^t(u) M(u) M(v) I_2^t(v) dudv, \end{aligned} \tag{9.14}$$

1303 so that

$$R(s, u) = 2M(s)M(u). \tag{9.15}$$

1305 It follows from (7.28) that

$$L_m(u, v) = 2 \int_0^{\min(u, v)} M(u - z)M(v - z)dz = L_m(v, u). \tag{9.16}$$

1307 The following two results are of interest.

1308 **Proposition 9.1** We seek to show that (8.29)₁ holds
 1309 for the minimum free energy. This implies that the
 1310 equivalent time domain version (7.7) is also true.

1311 *Proof* Substitute $R_{m+-}(\omega_1, \omega_2)$, given by (9.7)₄, into
 1312 the left-hand side of (8.29). By integrating around
 1313 $\Omega^{(+)}$, we obtain

$$\frac{i}{2\pi^2} \int_{-\infty}^{\infty} \frac{H_-(\omega_1)}{\omega_1(\omega_1 - \omega_2^+)} d\omega_1 = -\frac{1}{\pi} \frac{H_-(\omega_2)}{\omega_2}, \tag{9.17}$$

1315 and (8.29)₁ follows immediately, on noting the last
 1316 relation of (5.50). \square

1317 **Proposition 9.2** The quantity $\overline{f}_+(\omega)$ in (8.37) or
 1318 (8.39) vanishes in the case of the minimum free energy

1319 *Proof* For (8.39), closing the ω_1 contour over $\Omega^{(+)}$
 1320 gives zero. For (8.37)₂, the two terms cancel. \square

1321 Thus, this property, which is true for all free
 1322 energies in materials with branch cut singularities,
 1323 holds also for materials with only isolated singularities
 1324 in the case of the minimum free energy.

1325 **Proposition 9.3** The minimum free energy is the
 1326 only free energy functional for which the rate of
 1327 dissipation is given by a simple product. This is in
 1328 effect the result that the factorization of $H(\omega)$, given
 1329 by (3.8) and (3.9), where both zeros and singularities
 1330 of $H_{\pm}(\omega)$ are in Ω^{\pm} respectively, is unique up to a sign
 1331 ([1], p 240).

1332 *Proof* Let

$$R_{+-}(\omega_1, \omega_2) = r_+(\omega_1)r_-(\omega_2), \tag{9.18}$$

1334 under the condition

$$|r_+(\omega)|^2 = \frac{1}{2\omega^2 H(\omega)}. \tag{9.19}$$

Equation (8.39) reduces to

1336

$$\int_{-\infty}^{\infty} \frac{H(\omega_1)r_+(\omega_1)}{\omega_1 - \omega^-} d\omega_1 = -\frac{\overline{f}_+(\omega)\pi}{\omega r_-(\omega)} = F_-(\omega), \tag{9.20}$$

since the zeros of $r_-(\omega)$ are in $\Omega^{(-)}$. Using the Plemelj
 1338 formulae (3.19) and (3.20), we can write (cf. (4.3)) 1339

$$H(\omega_1)r_+(\omega_1) = \rho_-(\omega_1) - \rho_+(\omega_1),$$

$$\rho_{\pm}(\omega_1) = \frac{1}{2\pi i} \int_{-\infty}^{\infty} \frac{H(\omega_1)r_+(\omega_1)}{\omega_1 - \omega^{\mp}} d\omega_1, \tag{9.21}$$

and (9.20) is the requirement that $\rho_+(\omega) = F_-(\omega)$. 1341
 Both sides vanish at infinity, so that both must be zero 1342
 everywhere, by Liouville's theorem (for example, [1], 1343
 p 534). Thus, we have that 1344

$$H_+(\omega_1)r_+(\omega_1) = \frac{\rho_-(\omega_1)}{H_-(\omega_1)}. \tag{9.22}$$

Multiplying across by a factor ω_1 , we see that both 1346
 sides must be equal to a constant k , by Liouville's 1347
 theorem, giving 1348

$$r_+(\omega_1) = \frac{k}{\omega H_+(\omega_1)}. \tag{9.23}$$

It follows from (9.19) that $|k|^2 = 1/2$, and (9.23), 1350
 substituted into (9.18), yields (9.7)₄. Thus, the mini- 1351
 mum free energy is the only possibility associated with 1352
 (9.18). The requirement that $F_-(\omega)$ vanishes implies 1353
 that, in agreement with proposition 9.2, we have 1354
 $\overline{f}_+(\omega) = 0$. \square 1355

10 General form of free energies that are FMSs: discrete spectrum materials 1356 1357

We now present quadratic forms in terms of the 1358
 minimal state functionals I^l for discrete spectrum 1359
 materials, just as (5.25) and (5.28) apply to 1360
 quadratic forms in terms of histories. Let us 1361
 consider the form (8.14)₁ for $I_{2+}^l(\omega)$ given by 1362
 (5.53)₂. We obtain 1363

$$\begin{aligned}
 D(t) &= \frac{1}{2} \mathbf{w}^\top(t) \mathbf{R} \mathbf{w}(t) \\
 \mathbf{w}(t) &= (w_1(t), w_2(t), \dots, w_n(t)), \quad w_i(t) = \alpha_i^2 G_i e_i(t), \\
 R_{ij} &= \frac{1}{4\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{R_{+-}(\omega_1, \omega_2)}{(\omega_1 + i\alpha_i)(\omega_2 - i\alpha_j)} d\omega_1 d\omega_2 \\
 &= R_{+-}(-i\alpha_i, i\alpha_j), \quad i, j = 1, 2, \dots, n,
 \end{aligned} \tag{10.1}$$

where $e_i(t)$ is defined by (5.24) and the last relation is deduced by integrating over $\Omega^{(-)}$ on the ω_1 plane and $\Omega^{(+)}$ on the ω_2 plane. Relations (10.1) can also be obtained from (7.12) and (5.52).

The free energy functional (7.1) has the form

$$\begin{aligned}
 \psi(t) &= \phi(t) + \frac{1}{2} \mathbf{w}^\top(t) \mathbf{L} \mathbf{w}(t) \\
 L_{ij} &= \frac{1}{4\pi^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{L_{+-}(\omega_1, \omega_2)}{(\omega_1 + i\alpha_i)(\omega_2 - i\alpha_j)} d\omega_1 d\omega_2 \\
 &= L_{+-}(-i\alpha_i, i\alpha_j) = \frac{R_{ij}}{\alpha_i + \alpha_j}, \quad i, j = 1, 2, \dots, n,
 \end{aligned} \tag{10.2}$$

by virtue of (8.7). The quantities \mathbf{R} and \mathbf{L} are symmetric. Using (5.27), we see that

$$\begin{aligned}
 \dot{w}_i(t) &= -\alpha_i w_i(t) + z_i \dot{E}(t), \\
 z_i &= \alpha_i^2 G_i, \quad i = 1, 2, \dots, n.
 \end{aligned} \tag{10.3}$$

It follows that (2.9) holds, provided that

$$\sum_{i=1}^n \frac{w_i(t)}{\alpha_i^2} \left[1 - \sum_{j=1}^n \alpha_i^2 L_{ij} \alpha_j^2 G_j \right] = 0, \tag{10.4}$$

which is (7.7) for discrete spectrum materials. Let us put

$$l_{ij} = \frac{l_{ij}}{\alpha_i^2 \alpha_j^2}, \quad i, j = 1, 2, \dots, n, \tag{10.5}$$

in terms of the matrix \mathbf{l} . Relation (10.4) holds for all histories, so that we must have

$$\sum_{j=1}^n l_{ij} G_j = 1, \quad i = 1, 2, \dots, n. \tag{10.6}$$

Referring to (5.26), we see that if $\mathbf{l} = \mathbf{C}^{-1}$, then (10.6) holds. The form (10.6) corresponds to the Laplace

transform of (7.11)₃ for discrete spectrum materials, at the points $i\alpha_i$, where, from (6.9), we know that $\overline{f}_+(i\alpha_i) = 0, i = 1, 2, \dots, n.$

We can also see that (8.37)₁ gives

$$\begin{aligned}
 \overline{f}_+(\omega) &= i\omega \sum_{i=1}^n \alpha_i^2 G_i L_{+-}(-i\alpha_j, \omega) - \frac{1}{i\omega^+} \\
 &= -\omega \sum_{i=1}^n \frac{\alpha_i^2 G_i R_{+-}(-i\alpha_j, \omega)}{\omega + i\alpha_i} - \frac{1}{i\omega^+}
 \end{aligned} \tag{10.7}$$

on using (4.14)₂, (8.12) and by closing the contour on $\Omega^{(-)}$. Putting $\omega = i\alpha_j$ yields (10.6).

The expressions (10.1) and (10.2) are not helpful in characterizing quadratic forms in terms of $I_2^f(s), s \in \mathbb{R}^+$ because they are, in effect, quadratic forms in the $e_i(t)$; while the free energies ψ^f , given by (4.7), and discussed in Sect. 9, can also be expressed as such quadratic forms, even though they depend on $\overline{I}_{2F}^f(\omega)$ in the frequency domain, or $I_2^f(s), s \in \mathbb{R}$, in the time domain.

11 Proof that no new free energies can be expressed in terms of I^f

The approach adopted in [10] was based on product formulae in the time domain, and more particularly in the frequency domain, for the kernel of the rate of dissipation, which ensure that this quantity is non-negative. They also ensure that the resulting free energy has the correct non-negativity properties. In principle, the same approach should apply in the present context, as demonstrated in Sect. 7.1. However, as we will now show, there are no free energy functionals expressible as quadratic forms in I^f other than the minimum free energy. This is a generalization of the conclusion of Sect. 9.1 that, of the family $\psi_f(t)$, only $\psi_m(t)$ has this property. It further indicates how restrictive the requirement is that a free energy functional be expressible in the form (7.1) or (8.18)₁.

Proposition 11.1 The only possible choice of $L_{+-}(\omega_1, \omega_2)$ obeying (8.37) is the kernel $L_{m+-}(\omega_1, \omega_2)$, given by (9.9)₃.

Proof We express $L_{+-}(\omega_1, \omega_2)$ in the form

$$L_{+-}(\omega_1, \omega_2) = L_{m+-}(\omega_1, \omega_2) + L_{l+-}(\omega_1, \omega_2). \tag{11.1}$$

1421 The case of materials with only discrete spectrum
 1422 singularities (remark 5.2) will be considered first. The
 1423 quantity $L_{m+-}(\omega_1, \omega_2)$ is a solution of (8.37)_{1,2} for
 1424 $\bar{f}_+(w) = 0$ (proposition 9.2), so that we have

$$\begin{aligned} \bar{f}_+(w) &= U(w), \\ U(w) &= \frac{\omega}{\pi i} \int_{-\infty}^{\infty} H(\omega_1) L_{1+-}(\omega_1, w) d\omega_1 \\ &= \frac{\omega}{\pi i} \int_{-\infty}^{\infty} H_+(\omega_1) H_-(\omega_1) L_{1+-}(\omega_1, w) d\omega_1, \\ &\forall \omega \in \mathbb{R}. \end{aligned} \tag{11.2}$$

1426 The quantity $f_+(w)$ is given by (6.9); it vanishes at
 1427 $-i\alpha_i$, $i = 1, 2, \dots, n$, and has singularities at $i\chi_i$,
 1428 $i = 0, 1, \dots, n$, where the parameters χ_i are arbitrary
 1429 positive quantities. The kernel $L_{1+-}(\omega_1, w)$ must
 1430 depend on the χ_i , since $H(\omega_1)$ is independent of them.
 1431 Let us seek forms of $L_{1+-}(\cdot, \cdot)$ which are solutions of
 1432 (11.2)₁, for any choices of the χ_i .

1433 The simplest way of ensuring that the zeros of $U(w)$
 1434 are consistent with the location of the zeros of $\bar{f}_+(w)$ is
 1435 to assume that $L_{1+-}(\omega_1, w)$ vanishes at each point
 1436 $w = i\alpha_i$. Alternatively, if $L_{1+-}(\omega_1, w)$ is not zero at a
 1437 given point $w = i\alpha_i$, then it is still possible that $U(i\alpha_i)$
 1438 could vanish, for given values of χ_i , thus achieving
 1439 consistency with (11.2)₁. Thus, we take the quantity
 1440 $L_{1+-}(\omega_1, w)$ to be zero at each point $w = i\alpha_i$ for most
 1441 values of the parameters χ_i , $i = 1, 2, \dots, n$.

1442 Let us consider a given set of values χ_j , $j \neq k$ as
 1443 fixed parameters, and regard $U(w)$ as a function of χ_k ,
 1444 denoted by $U(w, \chi_k)$. Now, $U(i\alpha_i, \chi_k)$ may have
 1445 discrete roots, in other words, may vanish at discrete
 1446 values of χ_k . However, this does not allow us to drop
 1447 the assumption that $L_{1+-}(\omega_1, i\alpha_i)$ is zero at these
 1448 values of χ_k , since such an assumption would intro-
 1449 duce anomalous discontinuities in the function
 1450 $L_{1+-}(\omega_1, i\alpha_i)$, regarded as a function of χ_k , because
 1451 it is zero for almost all choices of this parameter and
 1452 non-zero at certain isolated values.

1453 It follows that $L_{1+-}(\omega_1, w)$ must be taken to vanish
 1454 at each point $w = i\alpha_i$, $i = 1, 2, \dots, n$. Relation (8.3)
 1455 then implies that it is zero at each point $\omega_1 = -i\alpha_i$,
 1456 $i = 1, 2, \dots, n$, and the singularities of $H_-(\omega_1)$, as
 1457 given by (4.18)₃, are cancelled by $L_{1+-}(\omega_1, w)$ in
 1458 (11.2)₃. The remaining singularities of the integrand

are all in $\Omega^{(+)}$. Therefore, by closing the contour on
 $\Omega^{(-)}$ and recalling (8.11), we find that the right-hand
 side of (11.2) vanishes.

Thus, there are no kernels that are consistent with a
 non-zero choice of $f_+(w)$. Any acceptable choice of
 $L_{1+-}(\omega_1, w)$ must obey the equation

$$\int_{-\infty}^{\infty} H_+(\omega_1) H_-(\omega_1) L_{1+-}(\omega_1, w) d\omega_1 = 0, \quad \forall \omega \in \mathbb{R}. \tag{11.3}$$

The only way to ensure this condition for all w is to
 assign to $L_{1+-}(\omega_1, w)$ the property that it vanishes at
 each point $\omega_1 = -i\alpha_i$, and thereby cancels the singu-
 larities in $H_-(\omega_1)$. But these points are the singular-
 ities of $\bar{I}_{2+}(\omega_1)$ in (8.18), so that the quadratic form
 with kernel $L_{1+-}(\omega_1, w)$ would give a zero contribu-
 tion to the free energy, as can be seen by integrating ω_1
 over a contour on $\Omega^{(-)}$.

We conclude that $f_+(w)$ must be zero, even for
 materials with only isolated singularities and
 $L_{1+-}(\omega_1, w)$ in (11.1) makes no contribution to the
 free energy functional.

For materials with some branch cuts, the quantity
 $f_+(w)$ vanishes, in any case, and we must have a
 relation of the same form as (11.3). Then, there will be
 some branch cuts in $L_{1+-}(\omega_1, w)$ as a function of ω_1 .
 These must be in $\Omega^{(+)}$. There will also be branch cuts
 in $H_-(\omega_1)$, which must be in $\Omega^{(-)}$. There is no
 mechanism whereby these can neutralize or cancel
 each other. The only remaining possibility is that
 $L_{1+-}(\omega_1, w)$ vanishes. \square

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