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On a Mathematical Framework for the Constitutive Equations of Anisotropic Dielectric Relaxation

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Abstract Three classes of time-domain non-relativistic anisotropic dielectric constitutive equations of increasing generality are discussed. In each class dissipativity is ensured by the choice of a class of convolution kernels in the **D**-to-**E** constitutive equation expressing the electric field **E** in terms of the electric displacement field **D**. Defining properties of the inverse (**E**-to-**D**) kernels and their Fourier-Laplace transforms (complex dielectric functions) are determined by inversion of the **D**-to-**E** constitutive equation. By this procedure it is shown that dielectric functions of the dipolar dielectrics are tensor-valued Bernstein functions while the dielectric functions of the Drude-Lorentz type are tensor-valued negative definite functions. The properties of the complex dielectric permittivities are also determined for either class. The theory is applied to an exhaustive review of empirical response functions of real dielectric materials encountered in the literature. Each class of convolution kernels is consistent with existence of a conserved energy, but in one case a strictly dissipative energy can be constructed.

Keywords Dielectric · Relaxation · Anisotropy · Energy conservation · Completely monotone function · Bernstein function · Negative definite function · Positive definite function

Notations

$\mathbb{Z}_+$—the set of positive integers
$[a, b[$—the set of real x satisfying $a \le x < b$
$\mathbb{R}$—the set of real numbers
$\mathbb{C}$—the set of complex numbers

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$\mathbb{R}_+ := \{x \in \mathbb{R} \mid x > 0\}$
$\mathbb{R}_- := \{x \in \mathbb{C} \mid \operatorname{Re} x \leq 0, \operatorname{Im} x = 0\}$
$\mathbb{C}_- := \mathbb{C} \setminus \mathbb{R}_-$
δ—Dirac measure
$\theta(t)$—the Heaviside unit step function
$t_+^\alpha := \begin{cases} t^\alpha, & t \geq 0 \\ 0, & t < 0 \end{cases}, \quad \alpha \in \mathbb{C}$
$\overline{z}$—complex conjugate of $z \in \mathbb{C}$
$\mathsf{A} \geq 0$ ($\mathsf{A} \in \mathcal{M}$) if $\mathbf{u}^\dagger \mathsf{A}\mathbf{u} \in \overline{\mathbb{R}_+}$ for all $\mathbf{u} \in \mathbb{C}^d$
$\mathsf{A} > 0$ ($\mathsf{A} \in \mathcal{M}$) if $\mathbf{u}^\dagger \mathsf{A}\mathbf{u} > 0$ for all $\mathbf{u} \in \mathbb{C}^d$, $\mathbf{u} \neq 0$
$\mathrm{D}f(t)$—distributional derivative
$\dot{\phi} := \partial\phi/\partial t$
Fourier transform: $\hat{f}(\omega) := \int_{-\infty}^{\infty} \mathrm{e}^{\mathrm{i}\omega t} f(t)\,\mathrm{d}t$
$\mathfrak{M}$—the set of all the CM functions $\mathsf{F} :]0, \infty[\to \mathcal{M}$
$\mathfrak{D}$—the set of all the CPD functions $\mathsf{F} : [0, \infty[\to \mathcal{M}$
$|\mathsf{H}|$—total variation of a tensor-valued Radon measure H
$\mathcal{D}'(X; V)$—the space of V-valued distributions on X

$\overline{\mathbb{R}_+} = \{x \in \mathbb{R} \mid x \geq 0\}$
$\mathbb{C}^+ := \{z \in \mathbb{C} \mid \operatorname{Im} z > 0\}$
$\mathcal{M}$—the set of all the $m \times m$ complex matrices
$\mathcal{M}_\mathrm{R}$—the set of all the $m \times m$ real matrices
I—the unit matrix
$\mathbf{v} \cdot \mathbf{w} := v_k w_k$
$\mathsf{A}^\dagger$—Hermitian conjugate of matrix A
$\operatorname{Re} \mathsf{A} := (\mathsf{A} + \mathsf{A}^\dagger)/2$
$\langle f, \phi \rangle$—duality form on $\mathcal{D}'$, $\mathcal{D}$
f'—ordinary derivative
$f * g(t) := \int_{-\infty}^{\infty} f(t-s) g(s)\,\mathrm{d}s$
Laplace transform: $\tilde{f}(q) := \int_0^\infty \mathrm{e}^{-qt} f(t)\,\mathrm{d}t$
$\mathfrak{B}$—the set of all the Bernstein functions $\mathsf{F} : [0, \infty[\to \mathcal{M}$
$\mathfrak{P}$—the set of all the functions $\mathsf{F} : [0, \infty[\to \mathcal{M}$ such that the convolution operator $\mathsf{F}*$ is passive
$\mathcal{S}'(X; V)$—Schwartz space of tempered distributions on X with values in V
$\mathcal{E}'(X; V)$—the space of V-valued compactly supported distributions on X

1 Introduction

In view of the appearance of polarizable metamaterials whose properties can be designed many intuitive assumptions about electromagnetic properties of matter have been called in question. Since a large portion of dielectric relaxation theory is phenomenological, a formal analysis of the principles underlying phenomenology and their consequences is needed.

Dielectric relaxation is commonly expressed in terms of complex permittivity [34, 39, 40, 54]. Experimental studies of dielectric relaxation are usually based on measurements of polarization of dielectric samples subject to periodic electric fields. This explains the prevalence of frequency-domain formulation in dielectric relaxation theory. Time-domain approach provides however a more direct approach for the dielectric response to a suddenly applied electric field.

Frequency-domain formulation of material response is inappropriate in problems involving nonlinear expressions which are local in time. In particular, thermodynamic considerations involve bilinear expressions and therefore time domain formulation of dielectric response is more convenient in this context.

Time-domain formulation of the response functions is better adapted for applying thermodynamic constraints. Time domain formulation allows the use of powerful concepts and tools of harmonic analysis (positive definite functions, completely monotone functions) and linear systems theory (passive operators).

There are some analogies between dielectric relaxation (more precisely, gradual increase in polarization following a sudden increase in electric field intensity) and viscoelastic creep. In view of this analogy some methods and ideas developed for viscoelastic constitutive equations can be used in the formulation of dielectric relaxation. In both cases the same molecular mechanisms are involved and the two relaxations are the object of the same experimental studies.

There is an important difference between the two theories.

Empirical models of viscoelastic creep and relaxation in real materials are consistent with the assumption of a non-negative relaxation spectrum. Under this assumption the relaxation modulus is a locally integrable completely monotone (LICM) function and the creep compliance is a Bernstein function. This framework is also appropriate for many phenomenological models of dielectric relaxation in the frequency ranges where dipolar polarization prevails [9, 18, 34, 39, 40, 54]. Dielectric relaxation in such materials can be attributed to a continuum of Debye elements. In this context the convolution kernel in the constitutive **E**-to-**D** equation (10) is a Bernstein function and the kernel in the **D**-to-**E** constitutive equation (16) is LICM.

Ionic and electronic polarization relaxation involves resonance phenomena (Lorentz dispersion) and is therefore inconsistent with a non-negative relaxation spectrum. It therefore requires a wider class of constitutive equations. It will be shown that in this case the convolution kernels in the constitutive **E**-to-**D** equations are causal negative definite function, while the kernel in the **D**-to-**E** constitutive equation is a causal positive definite (CPD) function. In particular, the derivative of the time-domain electric susceptibility function is assumed to be a causal positive definite (CPD) function. The last assumption is consistent with the theory of Tip [60] and some other work on dielectric materials [20]. Our approach provides an explanation of Axiom (A_2) in Tip's theory [60] as well as its generalization to anisotropic dielectrics. Another related theory, due to Glasgow et al. [25], is based on an assumption called "passivity" by the authors. The "passivity" assumption in their paper is equivalent to $\operatorname{Re}\tilde{\mathsf{R}}(\mathrm{i}\omega) < 0$ for $\omega > 0$ (restricted to the isotropic case) and therefore it is a strengthened version of the inequality $\operatorname{Re}\tilde{\mathsf{R}}(\mathrm{i}\omega) \leq 0$ implied by the CPD assumption.

A third option is assuming that the constitutive operator mapping **D** to **E** is passive. This assumption is a strengthened version of the previous one. It leads to the Herglotz-Nevanlinna representation for the complex modulus in viscoelasticity and for the inverse complex dielectric function [21, 41].

It is well-known in the mechanical community that many different constitutive energy functionals can be associated with a given linear viscoelastic stress-strain constitutive relation [7]. The same situation occurs in dielectric relaxation [17]. A strictly dissipative energy U_D [29], derived from the Bernstein theorem [26, 66], a conserved energy U_C [31, 58], derived from the Bochner theorem [26] and the minimum stored energy [16, 17, 33] have attracted most attention. The strictly dissipative energy U_D decays monotonely in a closed system. It can be defined for viscoelastic media with completely monotone relaxation moduli. The conserved energy U_C [58] can be constructed for a much larger class of viscoelastic media with positive definite relaxation moduli. The conserved energy has been used to obtain a Hamiltonian and Lagrangian formulation of viscoelastic theory [31]. The stored energy in either case can be expressed in terms of a quadratic functional of a one-parameter family of auxiliary fields [60], also known as internal variables [29, 30]. The current values of the auxiliary fields represent the past histories of the strain rate. The auxiliary fields have been successfully applied to eliminate histories in numerical wavefield computations (e.g. in [45, 46]) as well as to formulate and prove the existence and uniqueness for the Cauchy problem [27].

Existence of a conserved energy suggests that intrinsic dissipation associated with dielectric relaxation is compatible with a Hamiltonian formulation. A Hamiltonian theory of dielectric relaxation is constructed in [60, 61] on the basis of the Bochner theorem and in [21] on the basis of the Herglotz-Nevanlinna representation of Herglotz functions [24]. Some recent applications of dielectric relaxation theory (e.g. interactions between molecules in the presence of a bulk dielectric material) necessitate quantum theory of dielectric media and therefore also a Hamiltonian formulation of the classical theory of dielectric polarization. Following Tip's method, the energy-momentum tensor of a polarizable and magnetizable medium was constructed by Stallinga in [59]. In our setting energy conservation is rigorously linked to the strengthened dissipativity assumptions underlying the proposed formalism.

In the usual formulation of dielectric relaxation theory the electric displacement field **D** is expressed in terms of the electric field **E**. In the time domain the mapping of **E** to **D** is a convolution operator. The inverse **D**-to-**E** constitutive relation is more appropriate for thermodynamic analysis. The **E**-to-**D** constitutive equation is then obtained by inversion of the **D**-to-**E** constitutive relation. Inversion of the constitutive equations has been the object of many studies in viscoelasticity [28, 32] because experiments involve both creep and stress relaxation data. We now extend to dielectric relaxation our earlier work on inversion of anisotropic viscoelastic creep and relaxation. By this method we are able to show that in dipolar dielectric media and, more generally, in dielectric media with non-negative relaxation spectra, the dielectric response function is a Bernstein function. In order to account for Lorentz-dispersive media a broader class of convolution kernels is needed. By inversion of the constitutive **D**-to-**E** equation we show that dielectric response function is a negative definite function in this more general case.

The CPD and LICM response functions can be associated with two spectral representations, in terms of driven oscillators or in terms of Debye elements. Either of the representations provides a basis for the definition of a formal energy density U. It should be kept in mind that neither the continuum theory nor statistical physics provides an expression for physical energy. The two energies are however useful in mathematical and numerical applications.

In Sects. 3.2–3.3 three alternative frameworks for dielectric relaxation ensuring dissipativity are considered. In Sect. 3.2 passive and positive definite dielectric **D**-to-**E** response functions are considered, while Sect. 3.3 is dedicated to completely monotone **D**-to-**E** response functions. Criteria for the membership of each class are expressed in terms of complex dielectric permittivity. In Sect. 4 inversion of the **D**-to-**E** constitutive equations leads to a precise determination of the class of the corresponding **E**-to-**D** kernels. It is shown in particular that the **E**-to-**D** kernels corresponding to the LICM **D**-to-**E** kernels are tensor-valued Bernstein functions and that every tensor-valued Bernstein function corresponds to a tensor-valued LICM **D**-to-**E** kernel. The **E**-to-**D** kernels corresponding to positive definite **D**-to-**E** kernels are negative definite.

In Sect. 5 the theory is applied to the phenomenological models of dielectric relaxation in real materials. Explicit expressions are obtained for the dielectric response function.

In Sect. 6 two alternative electromagnetic energy density functionals in a dielectric based on the spectral representations are constructed.

The focus of the paper is dielectric relaxation. Due to the variety of magnetizable media magnetization is outside the scope of the paper. It is however considered here inasmuch it is a necessary complement for the Maxwell equations.

The constitutive equations are expressed in terms of convolution operators with respect to the time variable in a fixed Lorentz reference frame, which can be construed as the rest frame of the undeformable medium. The theory is thus non-relativistic.

2 Energy Balance and Constitutive Equations

We shall use the Heaviside-Lorentz units with the time units chosen in such a way that the speed of light $c = 1$. ρ and $\mathbf{J}$ denote the free charge density and the free current density.

The first two Maxwell equations [38, 44, 62]

$$\nabla \times \mathbf{E} = -\dot{\mathbf{B}} \tag{1}$$

$$\nabla \times \mathbf{H} = \dot{\mathbf{D}} + \mathbf{J} \tag{2}$$

imply the Poynting identity

$$\mathbf{E}^\top \dot{\mathbf{D}} + \mathbf{H}^\top \dot{\mathbf{B}} = -\mathrm{div}(\mathbf{E} \times \mathbf{H}) - \mathbf{J}^\top \mathbf{E} \tag{3}$$

where $\mathbf{J}$ denotes the free charge current. The left-hand side of (3) will be considered as electromagnetic power expended in the interaction of the field with the matter. The two additional Maxwell equations are

$$\mathrm{div}\,\mathbf{D} = \rho \tag{4}$$

$$\mathrm{div}\,\mathbf{B} = 0 \tag{5}$$

where ρ denotes the free charge density. The charge conservation equation

$$\dot{\rho} + \mathrm{div}\,\mathbf{J} = 0 \tag{6}$$

follows from (2, 4).

Comparison with viscoelasticity [32] suggests the following constitutive relations of dispersive dielectric and magnetizable rigid bodies

$$E_k = \lambda_{kl} * \dot{D}_l \tag{7}$$

$$H_k = \nu_{kl} * \dot{B}_l \tag{8}$$

$$J_k = \sigma_{kl} * E_l \tag{9}$$

$(k, l = 1, 2, 3)$. In tensor notation

$$\mathbf{E} = \Lambda * \dot{\mathbf{D}} \tag{10}$$

$$\mathbf{H} = \mathsf{N} * \dot{\mathbf{B}} \tag{11}$$

$$\mathbf{J} = \mathsf{S} * \mathbf{E} \tag{12}$$

where N, Λ, S are tensor-valued functions on $\mathbb{R}_+ \times \mathbb{R}^d$ satisfying the causality requirement $\mathsf{N}(t,x) = \Lambda(t,x) = \mathsf{S}(t,x) = 0$ for $t < 0$. The constitutive equations are local and spatial dependence of the response functions does not play any role in our considerations. We shall therefore ignore the dependence on x in the subsequent considerations. The material response kernels N, Λ can be singular at 0.

We shall focus on dissipation associated dielectric polarization and assume that the medium is an insulator ($\mathsf{S} = 0$).

An alternative formulation is obtained by integration by parts

$$\mathbf{E}(t) = \Lambda(0+)\mathbf{D}(t) + \int_0^\infty \dot{\Lambda}(s)\mathbf{D}(t-s)\,\mathrm{d}s \tag{13}$$

$$\mathbf{H}(t) = \mathsf{N}(0+)\mathbf{B}(t) + \int_0^\infty \dot{\mathsf{N}}(s)\mathbf{B}(t-s)\,\mathrm{d}s \tag{14}$$

provided $\mathsf{N}(s)$, $\Lambda(s)$ have finite limits at $s \to 0$ and are differentiable on $\mathbb{R}_+$. Note that the kernel functions Λ, N appearing in (10–11) also appear as kernels in the quadratic functional representing the electromagnetic power.

If $\mathsf{R} : \mathbb{R}_+ \to \mathcal{M}_\mathrm{R}$ satisfies the equations

$$\mathsf{R} * \Lambda = \Lambda * \mathsf{R} = t\mathsf{I}, \quad t > 0 \tag{15}$$

then (10) is equivalent to

$$\mathbf{D} = \mathsf{R} * \dot{\mathbf{E}} \tag{16}$$

Similarly, if $\mathsf{Q} : \mathbb{R}_+ \to \mathcal{M}_\mathrm{R}$ satisfies the equations

$$\mathsf{Q} * \mathsf{N} = \mathsf{N} * \mathsf{Q} = t\mathsf{I}, \quad t > 0 \tag{17}$$

then (11) is equivalent to

$$\mathbf{B} = \mathsf{Q} * \dot{\mathbf{H}} \tag{18}$$

Complex dielectric permittivity and complex magnetic permeability are given by the Fourier transforms $(-\mathrm{i}\omega)\hat{\mathsf{R}}(\omega)$ and $(-\mathrm{i}\omega)\hat{\mathsf{Q}}(\omega)$ of DR and DQ.

The Laplace transforms of the material response functions R and Q satisfy the equations

$$\tilde{\mathsf{R}}(p)\tilde{\Lambda}(p) = \tilde{\Lambda}(p)\tilde{\mathsf{R}}(p) = p^{-2}\mathsf{I} \tag{19}$$

$$\tilde{\mathsf{Q}}(p)\tilde{\mathsf{N}}(p) = \tilde{\mathsf{N}}(p)\tilde{\mathsf{Q}}(p) = p^{-2}\mathsf{I} \tag{20}$$

Equations (8–7) are valid in the rest system of the body. Time-domain formulation of dispersive constitutive equations allows covariant formulations if desirable. In addition, (8–7) are also tailored to introduce two concepts of harmonic analysis which have proved very useful in viscoelasticity [28, 29, 32]: positive definiteness and complete monotonicity, as well as a related concept from the system theory: passivity [41, 63, 69].

3 Admissible D-to-E Kernels

3.1 A Necessary Condition for Dissipativity

Consider a non-deformable electromagnetically polarizable medium. The power expended by the external agents on the medium is given by the expression

$$W := W_0 + W_1 \tag{21}$$

where

$$W_0 := \mathbf{E}^\top\dot{\mathbf{D}} + \mathbf{H}^\top\dot{\mathbf{B}} \tag{22}$$

$$W_1 := \mathbf{E}^\top\mathbf{J} \tag{23}$$

The dissipativity condition

$$\forall T \in \mathbb{R} \quad \int_{-\infty}^{T} W(t)\,\mathrm{d}t \geq 0 \tag{24}$$

[17] is not sufficiently strong to rule out some non-physical processes. In a homogeneous polarizable medium it does not rule out the time-periodic homogeneous process $\mathbf{E}(t), \mathbf{D}(t), \mathbf{J}(t) = -\dot{\mathbf{D}}(t)$, $\mathbf{H} = \mathbf{B} = 0$ with zero energy flux, zero power W and without external sources. We shall therefore assume that

$$\forall T \in \mathbb{R} \quad \int_{-\infty}^{T} W_0(t)\,\mathrm{d}t \geq 0 \tag{25}$$

$$\forall T \in \mathbb{R} \quad \int_{-\infty}^{T} W_1(t)\,\mathrm{d}t \geq 0 \tag{26}$$

If $\mathbf{D} = \cos(\omega t)\mathbf{D}_0$, $\mathbf{D}_0 \in \mathbb{R}^3$, then

$$\mathbf{E} = 2\omega \left[\operatorname{Im} \tilde{\Lambda}(\mathrm{i}\omega)\cos(\omega t) - \operatorname{Re} \tilde{\Lambda}(\mathrm{i}\omega)\sin(\omega t)\right]\mathbf{D}_0$$

and the average power over a period $T = 2\pi/\omega$ is given by the expression

$$P := \frac{1}{T}\int_0^T \mathbf{E}(t)^\top \dot{\mathbf{D}}(t)\,\mathrm{d}t = \omega^2 \mathbf{D}_0^\top \operatorname{Re} \hat{\Lambda}(\omega)\mathbf{D}_0$$

Equation (25) is satisfied for arbitrary $\mathbf{D}_0 \in \mathbb{R}^3$ and $\omega \in \mathbb{R}$ if

$$\operatorname{Re} \tilde{\Lambda}(\mathrm{i}\omega) \geq 0 \quad \forall \omega \in \mathbb{R} \tag{27}$$

In a smooth periodic electric field $P = -\int_0^T \mathbf{D}(t)^\top \dot{\mathbf{E}}(t)\,\mathrm{d}t/T$. Consequently, if the constitutive equation (10) can be expressed in the form $\mathbf{D} = \mathsf{R} * \dot{\mathbf{E}}$ then

$$\operatorname{Re} \tilde{\mathsf{R}}(\mathrm{i}\omega) \leq 0 \quad \forall \omega \in \mathbb{R} \tag{28}$$

Equation (28) also follows from (27) and (19). Inequality (28) is a generalization of an inequality derived in [44]. Inequalities (28) and (27) are necessary but not sufficient to ensure dissipativity.

3.2 Passive and Positive Definite Time-Domain Response Functions

Inequalities (25) and (26) are justified by the fact that they immediately translate into easily verifiable spectral conditions. We shall focus on dielectric dissipation and set the resistivity $\mathsf{S} \equiv 0$.

We shall recast (25) in a more explicit form

$$\frac{1}{2}\int_{-\infty}^{T}\int_{-\infty}^{t} \left[\dot{\mathbf{D}}(t)^\top \Lambda(t-s)\dot{\mathbf{D}}(s) + \dot{\mathbf{B}}(t)^\top \mathsf{N}(t-s)\dot{\mathbf{B}}(t-s)\right]\mathrm{d}s\,\mathrm{d}t \geq 0 \tag{29}$$

In order to obtain more restrictions on material response (29) will be extended to arbitrary smooth compactly supported vector-valued test functions $\mathbf{w}$:

$$\int_{-\infty}^{T}\int_{-\infty}^{t} \left[\dot{\mathbf{w}}(t)^\top \Lambda(t-s)\dot{\mathbf{w}}(s)\right]\mathrm{d}s\,\mathrm{d}t \geq 0 \tag{30}$$

$$\int_{-\infty}^{T}\int_{-\infty}^{t}\left[\dot{\mathbf{w}}(t)^{\top}\mathsf{N}(t-s)\dot{\mathbf{w}}(s)\right]\mathrm{d}s\,\mathrm{d}t \geq 0 \tag{31}$$

for all $\mathbf{w} \in \mathcal{D}(\mathbb{R};\mathbb{R}^m)$.

By $\mathcal{D}'_+(\mathbb{R};V)$ we denote the set of all the V-valued distributions on $\mathbb{R}$ with support contained in $[0,\infty[$. We shall often use the vector space $V=\mathbb{R}^d$. In the following the term matrix refers to an arbitrary $d\times d$ complex matrix.

Definition 3.1 ([41, 63, 69]) A convolution operator $L\mathbf{u} := \mathsf{F} * \mathbf{u}$, $\mathsf{F} \in \mathcal{D}'_+(\mathbb{R};\mathcal{M}_{\mathrm{R}})$, satisfying

$$\int_{-\infty}^{T}\mathbf{u}(t)^{\top}(\mathsf{F}*\mathbf{u})(t)\,\mathrm{d}t \geq 0 \quad \forall T\in\mathbb{R}\ \forall \mathbf{u}\in\mathcal{D}\big(\mathbb{R};\mathbb{R}^d\big) \tag{32}$$

is called *passive*.[1]

The set of distributions which are kernels of passive operators will be denoted by $\mathfrak{P}$.

The following theorem follows from the theory developed in [63, 69]:

Theorem 3.1 *If the operator* $L=\mathsf{F}*$ *is passive*, *then* $\mathsf{F}\in\mathcal{S}'_+(\mathbb{R};\mathcal{M}_{\mathrm{R}})$.

Theorem 3.1 implies that the tensor-valued distribution F has a Fourier-Laplace transform $\tilde{\mathsf{F}}$, defined as a holomorphic function $\tilde{\mathsf{F}}(p+\mathrm{i}q)$ satisfying

$$\int \tilde{\mathsf{F}}(q+\mathrm{i}p)\hat{\phi}(p)\,\mathrm{d}p = \langle \mathsf{F}, \mathrm{e}_q\hat{\phi}\rangle$$

for every function $\phi\in\mathcal{S}(\mathbb{R};\mathbb{R}^d)$ and $\mathrm{e}_q(t) := \mathrm{e}^{-qt}$.

If the convolution operator $\Lambda *$, defined by the constitutive equation (10), is passive, then the electromagnetic work performed on the system in a cyclical process is non-negative.

Definition 3.2 A tensor-valued function $\mathsf{F}:\mathbb{R}_+\to\mathcal{M}_{\mathrm{R}}$ is said to be *causal positive definite* (CPD) if

$$\int_{-\infty}^{\infty}\mathbf{v}(s)^{\top}\int_{-\infty}^{s}\mathsf{F}(s-\xi)\mathbf{v}(\xi)\,\mathrm{d}\xi\,\mathrm{d}s \geq 0 \tag{33}$$

for every $\mathbf{v}\in\mathcal{D}(\mathbb{R}_+;\mathbb{R}^d)$.

Definition 3.3 A distribution $\mathsf{F}\in\mathcal{D}'_+(\mathbb{R};\mathcal{M}_{\mathrm{R}})$ is said to be causal positive definite (CPD) if it satisfies

$$\int_{-\infty}^{\infty}\mathbf{u}(t)^{\top}\mathsf{F}*\mathbf{u}(t)\,\mathrm{d}t \geq 0 \quad \forall\mathbf{u}\in\mathcal{D}\big(\mathbb{R};\mathbb{R}^d\big) \tag{34}$$

The set of causal positive definite distributions will be denoted by $\mathfrak{D}$.

In [69] such distributions are called semi-passive. In [63] they are called dissipative. In [26] measures which are CPD distributions are called of positive type. Every CPD distribution is a tempered distribution [63].

[1] Note that the integrand is a continuous integrable function because $\mathsf{F}*\mathbf{u}\in\mathcal{E}(\mathbb{R};\mathbb{R}^d)$.

The kernel of a passive operator is a CPD distribution. The limit $T \to \infty$ can be taken in (32) for every $\mathbf{u} \in \mathcal{D}(\mathbb{R}; \mathbb{R}^d)$.

Theorem 3.2 ([69], Theorem 8.12-1; [63], Theorem I in Sect. 3.17.3) *F is the kernel of a passive operator if and only if* (i) $\mathsf{F} \in \mathcal{S}'_+(\mathbb{R}; \mathcal{M}_\mathrm{R})$, (ii) *its Fourier-Laplace transform* $\tilde{\mathsf{F}}(z)$ *is holomorphic in the right half of the complex plane* ($\operatorname{Re} z > 0$) *and* (iii) $\operatorname{Re} \tilde{\mathsf{F}}(z) \geq 0$ *for* $\operatorname{Re} z > 0$.

The Herglotz-Nevanlinna representation theorem [24] implies the following formula for a general tensor-valued function $\tilde{\mathsf{F}}$ satisfying (iii):

$$\tilde{\mathsf{F}}(z) = \mathrm{i}\mathsf{A} + z\mathsf{B} + \int_{-\infty}^{\infty} \frac{1 + \mathrm{i}yz}{z + \mathrm{i}y} \mathsf{H}(\mathrm{d}y)$$

where $\mathsf{A} = \operatorname{Im} \tilde{\mathsf{F}}(1)$, $\mathsf{B} \geq 0$,

$$\mathsf{B} = \lim_{\substack{y \to \infty \\ \operatorname{Im} z = 0}} z^{-1} \tilde{\mathsf{F}}(z)$$

and H is a finite tensor-valued measure on $\mathbb{R}$.

Theorem 3.3 ([69], Sect. 8.12) *If $\mathsf{F}*$ is a passive operator, then*

$$\langle \mathsf{F}, \mathbf{v} \rangle = \mathsf{W}\mathbf{v}(0) - \mathsf{A}\mathbf{v}'(0) + \int_0^{\infty} \mathsf{J}(t)\mathbf{v}(t)\,\mathrm{d}t + \int_0^{\infty} [\mathsf{J}(0) - \mathsf{J}(t)]\,\mathbf{v}''(t)\,\mathrm{d}t \quad \forall \mathbf{v} \in \mathcal{D} \tag{35}$$

where W is a skew-symmetric matrix, A is a positive semi-definite matrix,

$$\mathsf{J}(t) = \int_{]-\infty,\infty[} \mathrm{e}^{\mathrm{i}\omega t} \mathsf{M}(\mathrm{d}\omega) \tag{36}$$

and M is a finite $\mathcal{M}_\mathrm{R}$-valued Radon measure on $\mathbb{R}$ such that $\mathsf{M}(\mathcal{U}) \geq 0$ for every subset $\mathcal{U}$ of $\mathbb{R}$.

A tensorial version of the Bochner theorem [66] states that every CPD tensor-valued function $\mathsf{F}(t)$ is the Fourier-Stieltjes transform of a positive Radon measure $\mathsf{G}(\zeta)$ (or, equivalently, a non-negative distribution):

Theorem 3.4 ([26, 56]) *If $\mathsf{F}(t)$ is a CPD tensor-valued distribution, then it is the distributional Fourier transform of a positive semi-definite tensor-valued tempered distribution G:*

$$\langle \mathsf{F}, \mathbf{v} \rangle = \left\langle \mathsf{G}(\zeta), \int \mathrm{e}^{\mathrm{i}\zeta t} \mathbf{v}(t)\,\mathrm{d}t \right\rangle \quad \forall \mathbf{v} \in \mathcal{S}(\mathbb{R}; V) \tag{37}$$

where $\mathsf{G}(\zeta) = \operatorname{Re} \hat{\mathsf{F}}(\zeta)$ and $\hat{\mathsf{F}}$ denotes the distributional transform of F.

If the tensor-valued function $\mathsf{F}(t)$ is locally bounded on $\overline{\mathbb{R}_+}$, then it is the Fourier transform of a positive tensor-valued Radon measure M

$$\mathsf{F}(t) = \int_{-\infty}^{\infty} \mathrm{e}^{\mathrm{i}\zeta t} \mathsf{M}(\mathrm{d}\zeta). \tag{38}$$

The Radon measure M *is the real part of the distributional Fourier transform of* F: $\mathsf{M}(\zeta) = \operatorname{Re}\hat{\mathsf{F}}(\zeta)$.

The class of passive operators is fully characterized by the following theorem:

Theorem 3.5 ([69], Theorem 8.14-1; [63], Theorem II in Sect. 3.17.4) $\mathsf{F} \in \mathcal{D}'$ *is the kernel of a passive operator if and only if it satisfies the following conditions*:

(i) F *is CPD*;
(ii) $\mathsf{F} = \mathrm{D}^2\mathsf{F}_1 + \mathsf{A}\mathrm{D}\delta$, $\mathsf{F}_1 \in \mathcal{C}(\mathbb{R}; \mathcal{M}_\mathrm{R})$ *has tempered growth and* $\mathsf{A} \in \mathcal{M}_\mathrm{R}$, $\mathsf{A} \geq 0$.

The following spectral criterion is more useful for verifying whether a frequency domain material response is the Fourier transform of a CPD function:

Theorem 3.6 ([26], Theorem 16.2.4) *If* $\mathsf{F} : [0, \infty[\to \mathcal{M}_\mathrm{R}$ *satisfies the condition*

$$\int_0^\infty \mathrm{e}^{-pt} |\mathsf{F}(t)| \, \mathrm{d}t < \infty \quad \textit{for every } p > 0$$

then the following statements are equivalent:

(i) F *is CPD*;
(ii) $\operatorname{Re}\tilde{\mathsf{F}}(p) \geq 0$ *in a strip* $0 < \operatorname{Re} p < \varepsilon$ *for some* $\varepsilon > 0$;
(iii) $\liminf_{\substack{p \to \mathrm{i}\omega \\ \operatorname{Re} p > 0}} \operatorname{Re}\tilde{\mathsf{F}}(p) \geq 0$ *for* $\omega \in \mathbb{R}$ *and* $\liminf_{\substack{|p| \to \infty \\ \operatorname{Re} p > 0}} \tilde{\mathsf{F}}(p) \geq 0$.

If the dielectric response function $\Lambda \in \mathfrak{D}$, then $\operatorname{Re}\tilde{\Lambda}(p) \geq 0$ on the imaginary axis $p = -\mathrm{i}\omega$, $\omega \in \mathbb{R}$ and, in particular, (27) is satisfied. For comparison with the more familiar isotropic case, assume that

$$\Lambda(t) = \sum_{k=1}^{3} \lambda_k(t)\mathsf{P}_k \tag{39}$$

$$\mathsf{R}(t) = \sum_{k=1}^{3} r_k(t)\mathsf{P}_k \tag{40}$$

where P_k, $k = 1, 2, 3$, are projections, $\sum_{k=1}^{3} \mathsf{P}_k = \mathsf{I}$. In this case $\operatorname{Re}\tilde{\lambda}_k(\mathrm{i}\omega) \geq 0$, provided the limit $p \to \mathrm{i}\omega$, for real ω, exists, and therefore

$$\operatorname{Re}\tilde{r}_k(\mathrm{i}\omega) = -\frac{1}{\omega^2}\frac{\operatorname{Re}\tilde{\lambda}_k(\mathrm{i}\omega)}{|\tilde{\lambda}_k(\mathrm{i}\omega)|^2} \leq 0, \quad k = 1, 2, 3 \tag{41}$$

The complex permittivity tensor $[p\tilde{\mathsf{R}}(p)]_{p=\mathrm{i}\omega} \equiv p^{-1}\tilde{\Lambda}(p)^{-1}$ has a non-positive imaginary part. In an isotropic dielectric with a complex permittivity ε

$$\tilde{r}_k(-\mathrm{i}\omega) = \frac{1}{-\mathrm{i}\omega}\varepsilon(\omega) = \varepsilon'(\omega) + \mathrm{i}\varepsilon''(\omega) \tag{42}$$

and

$$-\operatorname{Re}\tilde{r}_k(-\mathrm{i}\omega) = \frac{\varepsilon''(\omega)}{\omega} \geq 0 \quad \text{for all non-zero } \omega \in \mathbb{R} \tag{43}$$

The functions $r_k(t)$ are real-valued, hence $\tilde{r}_k(\mathrm{i}\omega) = \overline{\tilde{r}_k(\mathrm{i}\omega)}$ and therefore $\varepsilon''(\omega)$ is an odd function. Consequently (43) can be replaced by

$$\frac{\varepsilon''(\omega)}{\omega} \geq 0 \quad \forall \omega > 0 \tag{44}$$

In [44] the last inequality is derived from the condition of non-negative average attenuation in time-periodic wavefields.

3.3 Completely Monotone Time-Domain Response Functions

A tensor-valued function B is positive semi-definite (non-decreasing) if the function $\mathbf{v}^\dagger \mathsf{B}(\cdot)\mathbf{v}$ is non-negative (non-decreasing, respectively) for every $\mathbf{v} \in \mathbb{C}^d$.

Definition 3.4 A tensor-valued function $\mathsf{F} : \mathbb{R}_+ \to \mathcal{M}_\mathrm{R}$ is said to be *completely monotone* (CM) if it is infinitely differentiable and for every vector $\mathbf{v} \in \mathbb{C}^d$

$$\forall n \in \mathbb{Z}_+ \quad (-1)^n \mathbf{v}^\top \mathrm{D}^n \mathsf{F}(t)\mathbf{v} \geq 0 \tag{45}$$

The set of CM tensor-valued functions on $\mathbb{R}_+$ is denoted by $\mathfrak{M}$.

This definition has an obvious extension to functions defined on $\mathbb{R}$ with support in $\overline{\mathbb{R}_+}$.

Proposition 3.7 *If $\mathsf{A} \in \mathcal{M}_\mathrm{R}$ and $\mathsf{A} \geq 0$, then A is a symmetric matrix.*

Proof If $\mathsf{A} \in \mathcal{M}_\mathrm{R}$ and $\mathsf{A} \geq 0$ then, substituting $\mathbf{v} = \mathbf{v}_\mathrm{R} + \mathrm{i}\mathbf{v}_\mathrm{I}$ with arbitrary $\mathbf{v}_\mathrm{R}, \mathbf{v}_\mathrm{I} \in \mathbb{R}^d$, we have $\mathbf{v}_\mathrm{R}^\top \mathsf{A}\mathbf{v}_\mathrm{I} - \mathbf{v}_\mathrm{I}^\top \mathsf{A}\mathbf{v}_\mathrm{R} \geq 0$ and $\mathsf{A}^\top = \mathsf{A}$. □

Corollary 3.8 *If $\mathsf{A} \in \mathfrak{M}$, then $\mathsf{A}(t)^\top = \mathsf{A}(t)$ for all $t > 0$.*

Theorem 3.9 (Bernstein Theorem, [26]) *A tensor-valued function $\mathsf{A} : \mathbb{R}_+ \to \mathcal{M}_\mathrm{R}$ is CM if and only if it is the Laplace transform of a positive semi-definite tensor-valued Radon measure C,*

$$\mathsf{A}(t) = \int_{[0,\infty[} \mathrm{e}^{-rt} \mathsf{C}(\mathrm{d}r) \tag{46}$$

which satisfies the inequality

$$\int_{[0,\infty[} \mathrm{e}^{-rt} |\mathsf{C}|(\mathrm{d}r) < \infty \quad \textit{for } t > 0$$

In the above theorem $|\mathsf{C}|$ denotes the total variation of the Radon measure C (cf. e.g. [26]). An $\mathcal{M}$-valued Radon measure B is positive if $\mathbf{v}^\dagger \mathsf{B}(\mathcal{U})\mathbf{v} \geq 0$ for every bounded measurable set $\mathcal{U} \subset [0, \infty[$ and for every vector $\mathbf{v} \in \mathbb{C}^d$.

Theorem 3.10 *Every positive semi-definite real tensor-valued Radon measure C on a measurable space Q can be expressed as the product of a real Radon measure m and a positive semi-definite tensor-valued density B defined m-almost everywhere and satisfying the inequalities $|\mathsf{B}(r)| \leq 1$ and $\mathbf{v}^\top \mathsf{B}(r)\mathbf{v} \geq 0$ for m-almost all r in Q:*

$$\mathsf{C}(\mathcal{U}) = \int_{\mathcal{U}} \mathsf{B}(r) m(\mathrm{d}r)$$

Proof Let $m(\mathcal{U})$ denote the trace $\mathrm{tr}(\mathsf{C}(\mathcal{U}))$ of $\mathsf{C}(\mathcal{U})$ for an arbitrary bounded measurable subset $\mathcal{U}$ of Q. m is a positive real-valued Radon measure. For an arbitrary bounded measurable subset $\mathcal{U}$ of Q and $\mathbf{v}, \mathbf{w} \in \mathbb{R}^d$ the quadratic form on $\mathbb{R}^2$

$$\mathbf{v}^\top \mathsf{C}(\mathcal{U})\mathbf{v}\xi^2 + 2\mathbf{v}^\top \mathsf{C}(\mathcal{U})\mathbf{w}\xi\zeta + \mathbf{w}^\top \mathsf{C}(\mathcal{U})\mathbf{w}\zeta^2 \equiv (\xi\mathbf{v} + \zeta\mathbf{w})^\top \mathsf{C}(\mathcal{U})(\xi\mathbf{v} + \zeta\mathbf{w})$$

is positive semi-definite. Consequently $|\mathbf{w}^\top \mathsf{C}(\mathcal{U})\mathbf{v}| \le \sqrt{\mathbf{v}^\top \mathsf{C}(\mathcal{U})\mathbf{v}\mathbf{w}^\top \mathsf{C}(\mathcal{U})\mathbf{w}}$. Since $\mathbf{v}^\top \mathsf{C}(\mathcal{U})\mathbf{v} \le m(\mathcal{U})\mathbf{v}^\top \mathbf{v}$ for arbitrary $\mathbf{v}, \mathbf{w} \in \mathbb{R}^d$,

$$|\mathbf{w}^\dagger \mathsf{C}(\mathcal{U})\mathbf{v}| \le m(\mathcal{U})|\mathbf{v}||\mathbf{w}|$$

and therefore the Radon measure $\mathbf{w}^\top \mathsf{C}(\mathcal{U})\mathbf{v}$ has a Radon-Nikodym derivative $B(\mathbf{v}, \mathbf{w})(r)$ with respect to m, which is uniquely defined with a possible exception of a subset of $[0, \infty[$ of measure 0. For m-almost-every value of r the expression $B(\mathbf{v}, \mathbf{w})(r)$ is a bilinear function of its vectorial arguments. Choosing a basis $\mathbf{e}_1, \mathbf{e}_2, \ldots, \mathbf{e}_d$ and expressing $\mathbf{v}, \mathbf{w}$ in terms of this basis a tensor-valued function $\mathsf{B}(r)$ can be constructed in such a way that $B(\mathbf{v}, \mathbf{w})(r) = \mathbf{v}^\top \mathsf{B}(r)\mathbf{w}$. It is clear that $|\mathsf{B}(r)| \le 1$ and $\mathbf{v}^\top \mathsf{B}(r)\mathbf{v} \ge 0$ for m-almost all $r \ge 0$ and all $\mathbf{v} \in \mathbb{R}^d$. □

Theorem 3.10 allows factoring the tensor-valued measure $\mathsf{C}(\mathrm{d}r)$ into a scalar spectral density $m(\mathrm{d}r)$ and a positive semi-definite tensorial density B, which characterizes the anisotropic properties of the medium.

Applying Theorem 3.10 the tensor-valued Radon measure can be replaced by a positive (scalar) Radon measure and a positive semi-definite tensor-valued density:

Theorem 3.11 *A tensor-valued function* $\mathsf{A}(t)$ *is CM if and only if it is the Laplace transform of a positive semi-definite tensor-valued function* $\mathsf{B} : \overline{\mathbb{R}_+} \to \mathcal{M}_\mathrm{R}$

$$\mathsf{A}(t) = \int_{[0,\infty[} \mathrm{e}^{-rt} \mathsf{B}(r) m(\mathrm{d}r) \tag{47}$$

where m is a positive Radon measure satisfying the inequality

$$\int_{[0,\infty[} \mathrm{e}^{-rt} |\mathsf{B}(r)| m(\mathrm{d}r) < \infty \quad \textit{for some } t > 0$$

while the function B *is locally integrable with respect to* m, *positive semi-definite* m*-almost everywhere and satisfies the condition*

$$|\mathsf{B}(r)| \le 1 \quad m\textit{-almost everywhere}$$

The integral in (47) can be replaced by the Lebesgue-Stieltjes integral with respect to the non-decreasing right-continuous function μ defined by the formula $\mu(t) = m([0, t])$. The jump discontinuities of μ constitute a discrete relaxation spectrum, possibly embedded in a continuous spectrum.

Phenomenological models of viscoelastic response in real materials assume that $m(\mathrm{d}r) = h(r)\,\mathrm{d}\ln(r)$, where the function h is the relaxation spectrum density. In this case Theorem 3.11 implies that $h(r) \ge 0$. Relaxation in dipolar dielectrics has similar properties. The idea of a distribution of relaxation times has been used in dielectric relaxation theory since the pioneering work of Schweidler (1907) and Wagner (1913) [23, 54]. A more general approach is adopted in this paper because it results in a manageable time-domain characterization of the material response.

A function $\mathsf{F} \in \mathfrak{M}$ is continuous on $]0, \infty[$. It is therefore locally integrable (LI) everywhere except, perhaps, in a neighborhood of 0.

Definition 3.5 A tensor-valued function $\mathsf{F} : \mathbb{R}_+ \to \mathcal{M}_\mathrm{R}$ is said to be *locally integrable completely monotone* (LICM) ($\mathsf{F} \in \mathfrak{M}_\mathrm{LI}$) if it is completely monotone and

$$\int_0^1 |\mathsf{F}(t)|\,\mathrm{d}t < \infty$$

The LICM functions satisfy a sharper version of the Bernstein theorem:

Theorem 3.12 ([26]) *If* $\mathsf{F} : \mathbb{R}_+ \to \mathcal{M}_\mathrm{R}$ *is LICM, then* (46) *and* (47) *hold with*

$$\int_{[0,\infty[} \frac{|\mathsf{C}|(\mathrm{d}r)}{1+r} < \infty$$

and

$$\int_{[0,\infty[} \frac{m(\mathrm{d}r)}{1+r} < \infty$$

respectively.

If F is LICM, then, by Theorem 3.12 and a theorem in [66], the double Laplace transform

$$\int_0^\infty \mathrm{e}^{-pt} \left[\int_0^\infty \mathrm{e}^{-rt} \mathsf{F}(r) |C|(\mathrm{d}r) \right] \mathrm{d}t = \int_{[0,\infty[} \frac{1}{p+r} |C|(\mathrm{d}r) \tag{48}$$

exists for $t > 0$. Consequently every LICM function possesses a Laplace transform.

Given a complex dielectric permittivity, the LICM property can be verified by means of the following theorem:

Theorem 3.13 ([26], Theorem 5.2.6) *The Laplace transform* $\tilde{\mathsf{A}}$ *of an LICM tensor-valued function* A *has the following properties*:

(i) $\tilde{\mathsf{A}}$ *has an analytic extension to* $\mathbb{C}^- := \overline{\mathbb{C} \setminus \mathbb{R}_-}$;
(ii) $\tilde{\mathsf{A}}$ *is real on* $\mathbb{R}_+$;
(iii) $\tilde{\mathsf{A}}(p) \to 0$ *for* $p \to \infty$ *on* $\mathbb{R}_+$;
(iv) $\operatorname{Im} \tilde{\mathsf{A}}(p) \le 0$ *for* $\operatorname{Im} p > 0$;
(v) $\operatorname{Im}[p\tilde{\mathsf{A}}(p)] \ge 0$ *for* $\operatorname{Im} p > 0$ *and* $\tilde{\mathbf{A}}(p) \ge 0$ *for* $p \in \overline{\mathbb{R}_+}$;
(vi) $\operatorname{Re} \tilde{\mathsf{A}}(p) \ge 0$ *for* $\operatorname{Re} p > 0$.

If an analytic tensor-valued function $\mathsf{F}(p)$ *enjoys the properties* (i)–(ii),

$$\limsup_{p\to\infty} |\mathsf{F}(p)| < \infty$$

and either (iv) *or* (v), *then*

(I) *the limit*

$$\lim_{p\to\infty} \mathsf{F}(p) = \mathsf{F}_0$$

exists and is finite;

(II) $\mathsf{F}(p) - \mathsf{F}_0$ *is the Laplace transform of an LICM function.*

Remark If condition (ii) is relaxed to $\limsup_{p\to\infty} |\tilde{\mathsf{A}}(p)| < \infty$ along $\mathbb{R}_+$, then $\lim_{p\to\infty} \tilde{\mathsf{A}}(p) = \mathsf{A}_0$ exists, $\mathsf{A} = \mathsf{A}_0\delta + \mathsf{G}$ and G is LICM.

Corollary 3.14 *If the tensor-valued function* A *is LICM, then* A *is CPD.*

Proof By Theorem 3.12 the Laplace transform $\tilde{\mathsf{A}}(p)$ is absolutely convergent for $p > 0$. Consequently (vi) in Theorem 3.13 combined with Theorem 3.6 (ii) implies that the function A is CPD. □

4 E-to-D Constitutive Equations

4.1 Inversion of LICM Material Response Functions

Given an LICM tensor-valued function $\Lambda : \mathbb{R}_+ \to \mathcal{M}$, we shall consider the solutions $\mathsf{R} : \mathbb{R}_+ \to \mathcal{M}$ of the equation

$$\Lambda * \mathsf{R}(t) = t\mathsf{I}, \quad t > 0 \tag{49}$$

The problem can also be rephrased in terms of functions defined over the entire real axis. Given a function $\Lambda : \mathbb{R} \to \mathcal{M}$ with support on $\overline{\mathbb{R}_+}$, find a function $\mathsf{R} : \mathbb{R} \to \mathcal{M}$ with support on $\overline{\mathbb{R}_+}$ satisfying the equation $\Lambda * \mathsf{R}(t) = \mathsf{I}$ for $t > 0$.

Equation (19) and Corollary 3.8 imply that the tensor-valued function $\tilde{\mathsf{R}}(p)$ is symmetric, hence the complex dielectric function $\tilde{\Lambda}(p)$ is a symmetric tensor.

Let R' denote the ordinary derivative of the function $\mathsf{R} : \mathbb{R}_+ \to \mathcal{M}$.

Definition 4.1 A tensor-valued function $\mathsf{F} : [0, \infty[\to \mathcal{M}_{\mathrm{R}}$ is said to be a *Bernstein function* if it is infinitely differentiable and $\mathsf{F}(t) \geq 0$ for all $t \geq 0$ and $(-1)^n \mathrm{D}^n \mathsf{F}(t) \leq 0$ for $t > 0$ and all the positive integers n.

The set of all the $\mathcal{M}_{\mathrm{R}}$-valued Bernstein functions on $[0, \infty[$ is denoted by $\mathfrak{B}$.

The derivative of a Bernstein function is thus a completely monotone function.

We shall prove in this section that the dielectric relaxation function Λ is a Bernstein function. We shall begin with a representation theorem for tensor-valued Bernstein functions.

Proposition 4.1 *If* $\mathsf{R} \in \mathfrak{B}$, *then the limits* $\mathsf{R}_0 := \lim_{t\to 0+} \mathsf{R}(t)$, $\mathsf{R}_1 := \lim_{t\to 0+} \mathsf{R}'(t)$, $\mathsf{R}_\infty := \lim_{t\to\infty} \mathsf{R}'(t)$ *exist and*

$$\mathsf{R}(t) = \mathsf{R}_0 + t\mathsf{R}_\infty + \mathsf{R}_2(t), \quad t > 0 \tag{50}$$

$$\mathsf{R}_2(t) := \int_{]0,\infty[} \left[1 - \mathrm{e}^{-rt}\right] \mathsf{T}(\mathrm{d}r) \tag{51}$$

where T *is a positive tensor-valued Radon measure satisfying the inequality*

$$\int_{]0,\infty[} \frac{r}{1+r} |\mathsf{T}|(\mathrm{d}r) < \infty \tag{52}$$

Conversely, if the function $\mathsf{R} : \mathbb{R}_+ \to \mathcal{M}$ *is defined by* (50–51), *with an* $\mathcal{M}$*-valued Radon measure* T *on* $]0, \infty[$ *satisfying* (52), *then* $\mathsf{R} \in \mathfrak{B}$.

Proof Assume that $\mathsf{R} \in \mathfrak{B}([0, \infty[; \mathcal{M})$.

For every $\mathbf{v} \in \mathbb{C}^d$ the function $t \to \mathbf{v}^\dagger \mathsf{R}(t)\mathbf{v}$ is non-negative, non-decreasing on $\mathbb{R}_+$, hence it has a finite limit $\mathsf{R}_0(\mathbf{v})$. Therefore for every $\mathbf{v}$ and $\mathbf{w}$ in $\mathbb{C}^d$

$$\mathbf{v}^\dagger \mathsf{R}(t)\mathbf{w} \equiv \frac{1}{2}\left[(\mathbf{v}+\mathbf{w})^\dagger \mathsf{R}(t)(\mathbf{v}+\mathbf{w}) - \mathbf{v}^\dagger \mathsf{R}(t)\mathbf{v} - \mathbf{w}^\dagger \mathsf{R}(t)\mathbf{w}\right]$$

has a finite limit $\frac{1}{2}[\mathsf{R}_0(\mathbf{v}+\mathbf{w}) - \mathsf{R}_0(\mathbf{v}) - \mathsf{R}_0(\mathbf{w})]$ where $\mathsf{R}_0(\mathbf{v}) = \mathbf{v}^\dagger \mathsf{R}_0 \mathbf{v}$ and $\mathsf{R}_0 \in \mathcal{M}$, $\mathsf{R}_0 \geq 0$.

Similarly, for every $\mathbf{v} \in \mathbb{C}^d$ the function $t \to \mathbf{v}^\dagger \mathsf{R}'(t)\mathbf{v}$, $t \in \mathbb{R}_+$, is non-negative and non-decreasing. Repeating the argument of the previous paragraph, we conclude that $\mathsf{R}'(t)$ has a limit R_1 at $t \to 0+$.

By the Bernstein theorem

$$\mathsf{R}'(t) = \int_{[0,\infty[} \mathrm{e}^{-rt} \mathsf{M}(\mathrm{d}r) = \mathsf{R}_\infty + \int_{]0,\infty[} \mathrm{e}^{-rt} \mathsf{M}(\mathrm{d}r) \tag{53}$$

where $\mathsf{R}_\infty = \mathsf{M}(\{0\}) \geq 0$ and

$$\int_{]0,\infty[} \mathrm{e}^{-r} |\mathsf{M}|(\mathrm{d}r) < \infty$$

$\mathsf{R}(t)$ will now be obtained by integrating the right-hand side of (53) over $[0, t]$ and applying the Fubini theorem. The formula

$$\mathsf{T}(]0, R]) := \int_{]0,R]} \frac{\mathsf{M}(\mathrm{d}r)}{r}$$

defines a Radon measure T satisfying

$$\int_{]0,\infty[} r\mathrm{e}^{-r} |\mathsf{T}|(\mathrm{d}r) < \infty$$

such that (50) holds.

Since T is a positive tensor-valued measure, (52) is equivalent to the pair of inequalities

$$\int_{]0,1[} \frac{r}{1+r} \langle \mathbf{v}, \mathsf{T}(\mathrm{d}r)\mathbf{v}\rangle < \infty \tag{54}$$

$$\int_{[1,\infty[} \frac{r}{1+r} \langle \mathbf{v}, \mathsf{T}(\mathrm{d}r)\mathbf{v}\rangle < \infty \tag{55}$$

Since $1/2 < 1/(1+r) < 1$ on $]0, 1[$, the first inequality is equivalent to

$$\left\langle \mathbf{v}, \int_{]0,1[} r\mathsf{T}(\mathrm{d}r)\mathbf{v} \right\rangle < \infty$$

and follows from the inequality $\langle \mathbf{v}, \mathsf{M}(]0, 1[)\mathbf{v}\rangle < \infty$. Since $1/2 \leq r/(1+r) < 1$ on $[1, \infty[$, the second inequality is equivalent to

$$\left\langle \mathbf{v}, \int_{[1,\infty[} \mathsf{T}(\mathrm{d}r)\mathbf{v} \right\rangle < \infty \tag{56}$$

Inequality (56) follows from the following chain of inequalities:

$$\left(1 - \frac{1}{\mathrm{e}}\right)\left\langle \mathbf{v}, \int_1^\infty r^{-1}\mathsf{M}(\mathrm{d}r)\mathbf{v}\right\rangle \leq \left\langle \mathbf{v}, \int_1^\infty \frac{1-\mathrm{e}^{-rt}}{r}\mathsf{M}(\mathrm{d}r)\mathbf{v}\right\rangle \leq \langle \mathbf{v}, \mathsf{R}(1)\mathbf{v}\rangle < \infty$$

Equation (50), the inequalities $\mathsf{R}_0, \mathsf{R}_\infty \geq 0$ and the fact that M is a positive $\mathcal{M}$-valued measure have been used here.

In view of (52) and the inequality $\mathrm{e}^{-rt} \leq 1/(1+rt)$, the derivative $\mathsf{R}_2'(t)$ of the third term $\mathsf{R}_2(t)$ in (50) tends to 0 for $t \to \infty$, whence $\mathsf{R}'(t)$ has a limit R_∞ for $t \to \infty$.

Conversely, assume that the tensor-valued function $\mathsf{R}(t)$ is given by (50) with R_2 given by (51) with a Radon measure T on $]0,\infty[$ satisfying the inequality (52). Let $\mathsf{R}_2(t)$ denote the third term on the right-hand side of (50). We shall now prove that R_2 has a derivative which is a tensor-valued Bernstein function. Let $h > 0$,

$$\frac{\mathsf{R}_2(t+h) - \mathsf{R}_2(t)}{h} = \int_{]0,\infty[} \frac{[1-\mathrm{e}^{-rh}]\mathrm{e}^{-rt}}{h} \mathsf{T}(\mathrm{d}r) \tag{57}$$

The function $r\mathrm{e}^{-rt}$ is integrable with respect to the measure $\langle \mathbf{v}, \mathsf{T}(\mathrm{d}r)\mathbf{v}\rangle$ on account of the inequalities (52) and $\mathrm{e}^{-x} \leq 1/(1+x)$, $x \geq 0$. The inequality

$$1 - \mathrm{e}^{-x} \leq x, \quad \forall x \geq 0$$

implies that the integrand of (57) is also integrable with respect to the measure $\langle \mathbf{v}, \mathsf{T}(\mathrm{d}r)\mathbf{v}\rangle$. By the Lebesgue Dominated Convergence Theorem the left-hand side of (57) tends to

$$\frac{\mathrm{d}\langle \mathbf{v}, \mathsf{R}_2(t)\mathbf{v}\rangle}{\mathrm{d}t} = \int_{]0,\infty[} r\mathrm{e}^{-rt}\langle \mathbf{v}, \mathsf{M}(\mathrm{d}r)\mathbf{v}\rangle$$

which is a CM function by the Bernstein theorem. Consequently R_2 is a Bernstein function. □

R_1 and R_∞ denote the instantaneous and relaxed dielectric permittivity, respectively. Note that R has a finite limit at infinity if and only if $\int_{]0,\infty[} |\mathsf{T}(\mathrm{d}r)| < \infty$ and $\mathsf{R}_\infty = 0$.

Theorem 4.2 *If* $\Lambda : \mathbb{R}_+ \to \mathcal{M}_\mathrm{R}$ *is LICM and*

$$\Lambda(t_1) > 0 \tag{58}$$

for some $t_1 > 0$, *then there is a unique tensor-valued function* R *satisfying* (15).

$\mathsf{R} : \overline{\mathbb{R}_+} \to \mathcal{M}_\mathrm{R}$ *is a tensor-valued Bernstein function satisfying the equations*

$$\mathsf{R}(t) = \mathsf{R}_0 + \mathsf{R}_1(t), \quad t \geq 0 \tag{59}$$

$$\lim_{t\to 0} \mathsf{R}(t) = \mathsf{R}_0 \geq 0, \quad \lim_{t\to\infty} \mathsf{R}_1(t) = 0, \quad t \geq 0 \tag{60}$$

$$\mathsf{R}_1(t) = \int_{]0,\infty[} \left[1 - \mathrm{e}^{-rt}\right] \mathsf{T}(\mathrm{d}t) \tag{61}$$

where T *is a positive tensor-valued Radon measure satisfying* (52).

If $\Lambda_0 := \lim_{t\to 0} \Lambda(t) > 0$, *then* $\lim_{t\to 0} \mathsf{R}(t) = \Lambda_0^{-1}$.

Proof Since $\Lambda \in \mathfrak{M}$ the Laplace transform $\tilde{\Lambda}(p)$ exists for $p \in \mathbb{C}_-$.

By the Bernstein theorem Λ is the Laplace transform of a positive tensor-valued Radon measure M. Consequently $\tilde{\Lambda}$ is the Stieltjes transform of M [66] and

$$p\tilde{\Lambda}(p) = \int_{[0,\infty[} \frac{p}{p+r} \mathsf{M}(\mathrm{d}r) \tag{62}$$

In particular $\mathbf{v}^\dagger p\tilde{\Lambda}(p)\mathbf{v} \geq 0$. The integrand of (62) is an increasing function of $p \in \mathbb{R}_+$ and the measure M is positive, hence $\mathbf{v}^\dagger p\tilde{\Lambda}(p)\mathbf{v}$ is a non-decreasing function of $p \in \mathbb{R}_+$.

We now prove that the matrix $\tilde{\Lambda}(p)$ is invertible for $p \in \mathbb{C}_-$.

Suppose that $\mathbf{v}^\dagger p_2\tilde{\Lambda}(p_2)\mathbf{v} = 0$ for some $p_2 > 0$ and $\mathbf{v} \in \mathbb{C}^d$. The Radon measure $m = \mathbf{v}^\dagger\mathsf{M}\mathbf{v}$ is positive, hence (62) implies that the function $g(r) := p_2/(r + p_2)$ vanishes almost everywhere on $[0,\infty[$ with respect to the measure m. The function g is everywhere positive, hence $m([a,b]) = 0$ for every segment $[a,b]$ and $\mathbf{v}^\dagger\Lambda(t_1)\mathbf{v} = 0$, contrary to the assumption (58).

Assume now that $\mathbf{v}^\dagger\tilde{\Lambda}(p_2)\mathbf{v} = 0$ for some $p_2 \in \mathbb{C}_-$, $\operatorname{Im} p_2 \neq 0$. Since $\operatorname{Im}[p_2/(p_2+r)] \equiv r\operatorname{Im} p_2/|p_2+r|^2 > 0$ for $\operatorname{Im} p_2 > 0$ and $\operatorname{Im}[p_2/(p_2+r)] < 0$ for $\operatorname{Im} p_2 < 0$, it follows again from (58) that $m([a,b]) = 0$ for every $b > a > 0$, which in turn implies that $\mathbf{v}^\dagger\Lambda(t_1)\mathbf{v} = 0$, in contradiction with (62). This concludes the proof of invertibility of the operator $p\tilde{\Lambda}(p)$ for $p \in \mathbb{C}_-$.

Let $\mu_n(p)$, $n = 1,\ldots,d$, be the eigenvalues of the operator $p\tilde{\Lambda}(p)$. Since $p\tilde{\Lambda}(p)$ is real, positive and non-decreasing on $]0,\infty[$, the eigenvalues $\mu_n(p)$ are real and positive. We shall assume that $\mu_n(p)$, $p \in \mathbb{R}_+$, are arranged in the increasing order, with account of their multiplicities. It is shown in Appendix A that μ_n are non-decreasing functions of $p \in]0,\infty[$. Consequently, the eigenvalues $1/\mu_n(p)$, $1 \leq n \leq d$, of the inverse matrix

$$\mathsf{F}(p) := \left[p\tilde{\Lambda}(p)\right]^{-1} \tag{63}$$

are positive and non-increasing on $]0,\infty[$. Hence $\limsup_{p\to\infty} 1/\mu_n(p) < \infty$, $1 \leq n \leq d$, and $\limsup_{p\to\infty} |\mathsf{F}(p)| < \infty$. Furthermore, the tensor-valued function $\mathsf{F}(p)$ is real on $]0,\infty[$ and analytic on $\mathbb{C}_-$. By Theorem 3.13 (I) the limit $\lim_{p\to\infty}\mathsf{F}(p) =: \mathsf{A}$ exists.

Let $\mathbf{e}_n$, $1 \leq n \leq d$, denote a base of unit eigenvectors of $\mathsf{F}(p)$ at a fixed p, where $\mathbf{e}_n$ is the eigenvector corresponding to the eigenvalue $\mu_n(p)$. Expanding the vector $\mathbf{v}$ in the eigenvector base

$$\mathbf{v} = \sum_{n=1}^d c_n\mathbf{e}_n$$

we obtain

$$\operatorname{Im}\left[p\mathbf{v}^\dagger\mathsf{F}(p)\mathbf{v}\right] = \sum_{n=1}^d |c_n|^2\frac{\operatorname{Im}\overline{\lambda_n(p)}}{|\lambda_n(p)|^2} \tag{64}$$

where $\lambda_n(p) = \mu_n(p)/p$ is an eigenvalue of the matrix $\tilde{\Lambda}(p)$. By Theorem 3.13 (iv) $\operatorname{Im}[\mathbf{v}^\dagger\tilde{\Lambda}(p)\mathbf{v}] \leq 0$ for every $\mathbf{v} \in \mathbb{C}^d$. Hence $\operatorname{Im}\lambda_n(p) \leq 0$ and $\operatorname{Im}[p\mathbf{v}^\dagger\mathsf{F}(p)\mathbf{v}] \geq 0$. By the second part of Theorem 3.13

$$\mathsf{F}(p) = \mathsf{A} + \int_0^\infty \mathrm{e}^{-pt}\mathsf{K}(t)\,\mathrm{d}t$$

where K is LICM, and $\lim_{p\to\infty}\tilde{K}(p) = 0$. By (19)

$$\tilde{\mathsf{R}}(p) = \frac{1}{p}\left[\mathsf{A} + \tilde{\mathsf{K}}(p)\right]$$

and

$$\mathsf{R}(t) = \mathsf{A} + \mathsf{R}_1(t), \quad t \geq 0$$

with $\mathsf{R}_1(t) := \int_0^t \mathsf{K}(s)\,\mathrm{d}s$. The functions R_1 and R are Bernstein functions. Equation (61) follows from Lemma 4.1. Equation $(60)_1$ follows from $(60)_2$.

The last statement follows from the identity

$$\Lambda_0 = \lim_{p\to\infty} p\tilde{\Lambda}(p) = \lim_{p\to\infty} [p\tilde{\mathsf{R}}(p)]^{-1} = \mathsf{R}_0^{-1}$$ □

In an isotropic polarizable medium $\Lambda(t) = \lambda(t)\mathsf{I}$, $\mathsf{N}(t) = n(t)\mathsf{I}$, $\mathsf{Q}(t) = q(t)\mathsf{I}$, $\mathsf{R}(t) = \mathcal{E}(t)\mathsf{I}$ and

$$\lambda * \mathcal{E} = t \tag{65}$$

$$n * q = t \tag{66}$$

for $t > 0$. If Λ, N are CM, then the functions λ, n are CM. By a theorem in [29] the functions $q, \mathcal{E} : \mathbb{R}_+ \to \mathbb{R}$ are non-negative and their derivatives are CM.

Theorem 4.3 *If* $\mathsf{R} \in \mathfrak{B}$, $\mathsf{R}'(t_2) > 0$ *for some* $t_2 > 0$ *and* $\mathsf{R}_0 := \lim_{t\to 0} \mathsf{R}(t)$ *has an inverse, then* (15) *has a unique solution* Λ, Λ *is LICM and* $\lim_{t\to 0} \Lambda(t) = \mathsf{R}_0^{-1}$.

Proof Since $\mathsf{R}' \in \mathfrak{M}$, Theorem 3.13 (iv) implies that $\mathrm{Im}[\mathbf{v}^\dagger p\tilde{\mathsf{R}}(p)\mathbf{v}] \le 0$ for every $\mathbf{v} \in \mathbb{C}^d$ and $p \in \mathbb{C}^+$. By Theorem 3.13 $p^2\tilde{\mathsf{R}}(p)$ is analytic on $\mathbb{C}_-$. Repeating an argument used in the proof of Theorem 4.2, $p^2\tilde{\mathsf{R}}(p)$ is invertible for all $p \in \mathbb{C}_-$. In view of (19) $\tilde{\Lambda}(p)$ is analytic on $\mathbb{C}_-$. All the eigenvalues of the matrix $\tilde{\mathsf{R}}(p)$, $p > 0$, are non-negative by Theorem 3.13 (ii) and (vi). For all $p > 0$ the matrix $\tilde{\Lambda}(p)$ is symmetric and therefore also positive semidefinite.

By Theorem 3.13 (v)

$$\mathrm{Im}\left[\mathbf{v}^\dagger p\tilde{\Lambda}(p)\mathbf{v}\right] \equiv -\frac{\mathrm{Im}[\mathbf{v}^\dagger p\tilde{\mathsf{R}}(p)\mathbf{v}]}{|\mathbf{v}^\dagger p\tilde{\mathsf{R}}(p)\mathbf{v}|^2} \ge 0$$

Furthermore, $\mathsf{R} \in \mathfrak{B}$ implies that $\mathsf{R}(t) \ge \mathsf{R}_0$ and therefore

$$p^2\tilde{\mathsf{R}}(p) \equiv p^2 \int_0^\infty \mathrm{e}^{-pt}\mathsf{R}(t)\,\mathrm{d}t \ge p\mathsf{R}_0$$

Let $\lambda_n(p)$, $\mathbf{e}_n(p)$ be an eigenvalue and the corresponding unit eigenvector of $p^2\tilde{\mathsf{R}}(p)$. The smallest eigenvalue ρ of R_0 is positive, hence

$$\lambda_n(p) \ge p\mathbf{e}_n(p)^\dagger \mathsf{R}_0 \mathbf{e}_n(p) \ge \rho p$$

and $\lambda_n(p)^{-1} \le \rho^{-1}p^{-1} \underset{p\to\infty}{\longrightarrow} 0$. The eigenvalues of $\tilde{\Lambda}(p) = [p^2\tilde{\mathsf{R}}(p)]^{-1}$ are $\lambda_n(p)^{-1}$, $n = 1, \ldots, d$, hence $\lim_{p\to\infty} \tilde{\Lambda}(p) = 0$. Hence, using Theorem 3.13, Λ is LICM.

Finally

$$\Lambda_0 = \lim_{t\to 0} \Lambda(t) = \lim_{p\to\infty} \left[p\tilde{\Lambda}(p)\right] = \left[\lim_{p\to\infty} p\tilde{\mathsf{R}}(p)\right]^{-1} = \mathsf{R}_0^{-1}$$ □

Material response of a real dielectric medium is usually represented in terms of the complex permittivity $\mathsf{E}(\omega)$.

Corollary 4.4 *If the function* $\Lambda :]0, \infty[\to \mathcal{M}_\mathrm{R}$ *is LICM, then*

(i) *the complex dielectric permittivity* $\mathsf{E}(\omega) \equiv \widetilde{\mathsf{R}'}(-\mathrm{i}\omega)$ *is analytic in the complex plane cut along the negative imaginary axis*;

(ii) *on the positive imaginary axis* $\mathsf{E}(\omega)$ *is real positive semi-definite and tends to zero as* $|\omega| \to \infty$;

(iii) $\operatorname{Im}\mathsf{E}(\omega) \geq 0$ *for* $\operatorname{Re}\omega > 0$.

Proof Under the hypotheses of the corollary $\mathsf{R}' \in \mathfrak{M}$. The statements (i) and (ii) of the corollary thus follow from Theorem 3.13 (i) and (ii). (iii) follows from Theorem 3.13 (v) combined with the identity $\mathsf{E}(\omega) = \overline{\mathsf{E}(-\omega)}$. □

Since $\mathbf{v}^\top \mathsf{R}(t)\mathbf{v}$ is non-decreasing and non-negative for every $\mathbf{v} \in \mathbb{R}^d$, the existence of the limit R_0 is granted. R_0 however may not be invertible. If R_0 is invertible, then $\Lambda_0 := \lim_{t\to 0} \Lambda(t) = \mathsf{R}_0^{-1}$. If $\Lambda :]0, \infty[\to \mathcal{M}_\mathrm{R}$ is an arbitrary tensor-valued LICM function, then the existence of the limit $\Lambda_0 = \lim_{t\to 0+} \Lambda(t)$ is not granted. It is easy to show that (i) $R_0 = 0$ if and only if the largest eigenvalue of $\Lambda(t)$ is unbounded for $t \to 0+$; (ii) R_0 is invertible if and only if all the eigenvalues of $\Lambda(t)$ are bounded in a neighborhood of 0.

The function R has the properties of the viscoelastic creep compliance. In particular it is a non-decreasing function, either bounded or unbounded. In physical terms the dielectric polarization tends to increase as an increasing proportion of dipoles tends to adjust to those already polarized by the external field.

If R is bounded, then for every vector $\mathbf{w} \in \mathrm{R}^3$ the function $\mathbf{w}^\top \mathsf{R}(\cdot)\mathbf{w}$ is non-decreasing and bounded hence it has a finite limit $f(\mathbf{w})$. Consequently

$$2\mathbf{w}_1^\top \mathsf{R}(t)\mathbf{w}_2 = (\mathbf{w}_1 + \mathbf{w}_2)^\top \mathsf{R}(t)(\mathbf{w}_1 + \mathbf{w}_2) - \mathbf{w}_1^\top \mathsf{R}(t)\mathbf{w}_1 - \mathbf{w}_2^\top \mathsf{R}(t)\mathbf{w}_2$$

has a finite limit at ∞ and therefore R has a finite limit R_∞ at infinity. Since $\mathsf{R}_\infty \geq \mathsf{R}_0$, $\mathsf{R}_0 > 0$ implies that $\mathsf{R}_\infty > 0$. From these inequalities the inequalities for the complex permittivity at high and low frequencies follow. Assume for definiteness that R_∞ exists.

$$\mathsf{E}_\mathrm{s} := \lim_{\omega\to 0} \mathsf{E}(\omega) = \lim_{p\to 0} \left[p\tilde{\mathsf{R}}(p)\right] = \mathsf{R}_\infty$$

$$\mathsf{E}_\infty := \lim_{\omega\to \infty} \mathsf{E}(\omega) = \lim_{p\to \infty} \left[p\tilde{\mathsf{R}}(p)\right] = \mathsf{R}_0$$

by an Abelian theorem [66], and therefore

$$\mathsf{E}_\infty \leq \mathsf{E}_\mathrm{s} \tag{67}$$

where the subscript "s" stands for "static".

The dielectric after-response of a finite isotropic homogeneous sample can be expressed in terms of the function $\varphi(t)$. If the electric field $\mathbf{E}$ is kept at a fixed value for a sufficiently long period and then instantaneously switched off, then the polarization vector of the sample decays according to the law:

$$\mathbf{P}(t) = \varphi(t)\mathbf{P}(0) \tag{68}$$

The function φ satisfies the constraint $\varphi(0) = 1$ and therefore it satisfies the inequalities $0 \leq \varphi(t) \leq 1$. Let $\Phi(t) = 1 - \varphi(t)$. The dielectric response can be expressed in the form

$$\mathbf{D}(t) = \varepsilon_0\mathbf{E}(t) + \mathbf{P}(t) = \varepsilon_\infty\mathbf{E}(t) + \Delta_\varepsilon \int_0^\infty \Phi(s)\dot{\mathbf{E}}(t-s)\,\mathrm{d}s \tag{69}$$

where $\Delta_\varepsilon = \varepsilon_\mathrm{s} - \varepsilon_\infty$ [54] and $\mathsf{R}(t) = \Delta_\varepsilon \Phi(t)\mathsf{I}$. Since $\Phi(0) = 0$, (69) can also be expressed in the following form

$$\mathbf{D}(t) = \varepsilon_\infty \mathbf{E}(t) + [\varepsilon_\mathrm{s} - \varepsilon_\infty] \int_0^\infty \dot{\Phi}(s)\mathbf{E}(t-s)\,\mathrm{d}s \tag{70}$$

The following theorem follows easily from the definition of Bernstein functions:

Theorem 4.5 *If $f \in \mathfrak{M}$ and $0 \le f(t) \le 1$ for $t > 0$, then the function g defined on $[0,\infty[$ by the equations $g(t) := 1 - f(t), t > 0$ and $g(0) = 1 - f(0+)$ is a Bernstein function.*

Consequently Φ (and therefore also R) is a Bernstein function if $\varphi \in \mathfrak{M}$ and $0 \le \varphi(t) \le 1$. As an example, φ can be the stretched exponential function $\varphi(t) = \mathrm{e}^{-(t/\tau)^\alpha}$, $\tau > 0, 0 < \alpha \le 1$ [18, 57].

4.2 Inversion of Passive Material Response Functions

The fundamental solution R_2 of (49) is given by the equation in the distributions sense on $\mathbb{R}$

$$\Lambda(t) * \mathsf{R}_2(t) = \delta(t)\mathsf{I} \tag{71}$$

If a solution R of (49) exists, then $\mathsf{R}_2 = \mathrm{D}^2\mathsf{R}$ is the second-order derivative of R in the distributions sense.

If the function Λ is CPD (in particular, if the convolution operator $\Lambda*$ is passive), then the Laplace transform $\tilde{\Lambda}$ exists and (71) for a tempered distribution R_2 is equivalent to the algebraic equation

$$\tilde{\Lambda}(p)\widetilde{\mathsf{R}_2}(p) = \mathsf{I} \tag{72}$$

Assume that Λ is passive and $\det\tilde{\Lambda}(p) \ne 0$ for $\operatorname{Re} p > 0$. It follows that

$$\operatorname{Re}\widetilde{\mathsf{R}_2}(p) = \widetilde{\mathsf{R}_2}(p)^\dagger \left[\operatorname{Re}\tilde{\Lambda}(p)\right] \widetilde{\mathsf{R}_2}(p) \ge 0$$

hence the convolution operator R_2* is passive and

$$\mathsf{R}_2(t) = \delta(t)\mathsf{W} + \mathrm{D}\delta(t)\mathsf{A} + \mathsf{K}(t) + \delta''(t)\,[\mathsf{K}(0) - \mathsf{K}(t)]$$

where

$$\mathsf{K}(t) = \int \mathrm{e}^{\mathrm{i}\omega t}\mathsf{M}(\mathrm{d}\omega)$$

is CPD, M is a finite Borel measure, $\mathsf{W}^\dagger = -\mathsf{W}$, $\mathsf{A} = \mathsf{A}^\dagger \ge 0$. Since the function K is continuous, the function $\mathsf{L}(t) = \int_0^t \mathrm{d}s \int_0^s \mathsf{K}(u)\,\mathrm{d}u$ is well defined.

We are now ready to show that

$$\mathsf{R}(t) = t_+\mathsf{W} + \theta(t)\mathsf{A} + \mathsf{L}(t) + \mathsf{K}(0) - \mathsf{K}(t) \tag{73}$$

Indeed, for an arbitrary test function $\mathbf{v} \in \mathcal{S}(\mathbb{R}; V)$,

$$\langle \mathsf{R}, \mathrm{D}^2\mathbf{v}\rangle = \mathsf{W}\mathbf{v}(0) + \mathsf{A}\mathbf{v}'(0) + \langle \mathsf{L}, \mathbf{v}\rangle + \langle [\mathsf{K}(0) - \mathsf{K}(t)], \mathrm{D}^2\mathbf{v}\rangle = \langle \mathsf{R}_2, \mathbf{v}\rangle$$

Theorem 4.6 *The class of distributional solutions of* (49) *for* $\Lambda \in \mathfrak{P}$ *is given by* (73) *with* $\mathsf{W}^\dagger = -\mathsf{W}$, $\mathsf{A}^\dagger = \mathsf{A} \geq 0$, $\mathsf{K} \in \mathfrak{D}$, $\mathsf{L} = \int_0^t \mathrm{d}s \int_0^s \mathsf{K}(u)\,\mathrm{d}u$.

Note that $\operatorname{Re}\tilde{\mathsf{R}}(\mathrm{i}\omega) = -(1+\omega^2)\operatorname{Re}\tilde{\mathsf{K}}(\mathrm{i}\omega)/\omega^2 \leq 0$.

4.3 Inversion of Positive Definite Material Response Functions

If Λ is CPD, then the second-order distributional derivative $\mathrm{D}^2\mathsf{R}$ is CPD and, by Bochner's theorem, there is a positive Radon tensor-valued measure H with the support contained in $[0,\infty[$, such that

$$\mathrm{D}^2\mathsf{R}(t) = \int_{-\infty}^{\infty} \mathrm{e}^{\mathrm{i}\omega t}\mathsf{H}(\mathrm{d}\omega) = \Delta\mathsf{H} + \mathsf{R}_3(t) \tag{74}$$

where $\Delta\mathsf{H} := \mathsf{H}(\{0\}) \geq 0$ and

$$\mathsf{R}_3(t) :\equiv 2\operatorname{Re}\int_{]0,\infty[} \mathrm{e}^{\mathrm{i}\omega t}\mathsf{H}(\mathrm{d}\omega) \tag{75}$$

Hence, for $t > 0$,

$$\begin{aligned}\mathrm{D}\mathsf{R}(t) &= \mathsf{B} + t\Delta\mathsf{H} + 2\operatorname{Re}\int_{]0,\infty[} \frac{1}{\mathrm{i}\omega}\mathrm{e}^{\mathrm{i}\omega t}\mathsf{H}(\mathrm{d}\omega)\\ &= \mathsf{B} + t\Delta\mathsf{H} + \int_{]0,\infty[} \frac{\sin(\omega t)}{\omega}\mathsf{H}(\mathrm{d}\omega) = \mathsf{B} + \int_{[0,\infty[} \frac{\sin(\omega t)}{\omega}\mathsf{H}(\mathrm{d}\omega)\\ &= \mathsf{B} + \int_{-\infty}^{\infty} \frac{\sin(\omega t)}{\omega}\mathsf{H}_1(\mathrm{d}\omega)\end{aligned} \tag{76}$$

where $\mathsf{H}_1(\mathcal{U}) = \mathsf{H}(\mathcal{U})/2$ for every Borel subset of $\mathbb{R}$ not containing 0, and $\mathsf{H}_1(\{0\}) = \mathsf{H}(\{0\})$.

This observation suggests the following form of the response function R:

$$\mathsf{R}(t) = \mathsf{A} + t\mathsf{B} + \int_{[0,\infty[} \frac{1-\cos(\omega t)}{\omega^2}\mathsf{H}(\mathrm{d}\omega), \quad t \geq 0 \tag{77}$$

where $\mathsf{A} = \mathsf{A}^\top$, $\mathsf{B} = \mathsf{B}^\top \geq 0$. (76) is a generalization of Axiom (A_2) in [60]. Note that (49) implies that $\mathsf{A} \neq 0$. If the total variation $|\mathsf{H}|([0,\infty[)$ of H is finite, then $\mathrm{D}\mathsf{R}(t) = \delta(t)\mathsf{A} + \mathsf{R}_1(t)$ and $\mathsf{B} = \lim_{t\to 0+}\mathsf{R}_1(t)$. R_1 is locally integrable if $\int_{[0,\infty[} |\mathsf{H}|(\mathrm{d}\omega)/(1+\omega^2) < \infty$ [58].

The tensor A is the dielectric constant. The tensor B plays the role of the Newtonian viscosity tensor in viscoelasticity. The inverse of $\Lambda(t) = \delta(t)\mathsf{B}$ is $t_+\mathsf{B}$.

If $\mathsf{B} = 0$, then the right-hand side of (77) can be extended to a negative definite tensor-valued function $\mathsf{R}_\mathrm{e} : \mathbb{R} \setminus \{0\} \to \mathcal{M}$:

$$\mathsf{R}_\mathrm{e}(t) = \mathsf{A} + t^2\mathsf{C} + \int_{]0,\infty[} \frac{1-\cos(\omega t)}{\omega^2}\mathsf{H}(\mathrm{d}\omega) \tag{78}$$

where $\mathsf{C} \geq 0$ and H is a Radon measure on $\mathbb{R}_+$ satisfying the inequality

$$\int_{]0,\infty[} \frac{|\mathsf{H}|(\mathrm{d}\omega)}{\omega^2}[1-\cos(\omega t)] < \infty$$

and $\mathsf{C} = \mathsf{H}(\{0\})$. The function R_e is negative definite function on $\mathbb{R}$ [2] and the right-hand side of (78) is similar to the Lévy-Khinchin representation of a real continuous negative definite function [2, 22]. In the theory of convolution semigroups negative definite functions play the same role with respect to the positive definite functions as the Bernstein functions do with respect to the CM functions.

If the total variation $|\mathsf{H}|([0,\infty[)$ of the Radon measure H is finite, then the dielectric response function is the distributional derivative of R

$$\mathsf{DR}(t) = \delta(t)\mathsf{A} + \theta(t)\mathsf{B} + \int_{[0,\infty[} \frac{\sin(\omega t)}{\omega} \mathsf{H}(\mathrm{d}\omega), \quad t \geq 0 \tag{79}$$

The second-order distributional derivative is given by the formula

$$\mathrm{D}^2\mathsf{R}(t) = \mathrm{D}\delta(t)\mathsf{A} + \delta(t)\mathsf{B} + \int_{[0,\infty[} \cos(\omega t)\mathsf{H}(\mathrm{d}\omega) \tag{80}$$

Note that $\mathrm{D}\delta\mathsf{A}$ is positive definite (or, equivalently, semi-passive):

$$\int_{-\infty}^{\infty} \phi(t)^\top \mathsf{A}\mathrm{D}\phi(t)\,\mathrm{d}t = \frac{1}{2}\left[\phi(t)^\top \mathsf{A}\phi(t)\right]_{t=-\infty}^{\infty} = 0$$

5 Application to Empirical Dielectric Response Functions

5.1 Introduction

Viscoelastic creep compliances of real media are without exception Bernstein functions. Dielectric functions of dipolar dielectric media have the same property. On the other hand Lorentz-dispersive dielectric functions have a non-monotone oscillatory behavior attributable to molecular inertia [9, 10, 54]. Lorentz dispersion is however consistent with passivity and therefore also with positive definiteness of dielectric relaxation functions. The effects of molecular inertia are usually ignored in statistical models of viscoelastic relaxation in polymers, based on the Smoluchowski equation [4, 14].

There is no convincing general argument in support of positivity of the relaxation spectrum, although some ad hoc statistical arguments in favor of specific models have been advanced [49, 64, 65]. On the other hand statistical physics provides a fairly general explanation of the positive definite property of Λ. According to the fluctuation-dissipation theorem [42] the relaxation kernel is proportional to an auto-correlation function. If the auto-correlation function can be expressed in terms of time averaging and if it is additionally a continuous function, then it is a positive definite function ([19], Chap. 19). An example of the fluctuation-dissipation formula in dielectric relaxation theory is the following relation for the dipole moment **m** [10, 54]:

$$\mathbf{m}(t) = \mathsf{A}_0\mathbf{E}(t) + \int_0^t \mathsf{A}(s)\mathbf{E}(t-s)\,\mathrm{d}s \tag{81}$$

with $\mathsf{A}_\infty - \mathsf{A}(s) > 0$, where $\mathsf{A}_\infty := \lim_{s\to\infty}\mathsf{A}(s)$. Statistical arguments yield the after-effect function

$$\mathbf{B}(s) := \mathsf{A}_\infty - \mathsf{A}(s) = \frac{1}{3\mathrm{k}T}\mathsf{C}_\mathbf{m}(s), \quad s \geq 0 \tag{82}$$

where T denotes the absolute temperature, k is the Boltzmann constant and $\mathsf{C}_\mathbf{m}$ is the auto-correlation function

$$\mathsf{C}_\mathbf{m}(s) := \lim_{\tau\to\infty} \frac{1}{2\tau}\int_{-\tau}^{\tau} \mathbf{m}(t+s) \otimes \mathbf{m}(t)\,\mathrm{d}t \tag{83}$$

The integral in the above equation is positive definite and hence the limit is also positive definite.

In the following subsections we give a few examples of phenomenological models of dielectric relaxation functions which are either monotone or oscillatory and fit into one of the formulations given above. A general dielectric loss exhibits several peaks (the α peak, possibly a β peak, and the phonon peak) [47]. Each of these peaks is attributable to a different molecular relaxation mechanism and lies in a different frequency range. The dielectric susceptibility is obtained by summing the component dielectric susceptibilities associated with different molecular relaxation mechanisms. The explicit models discussed below represent the contributions of a single relaxation mechanism to the total relaxation spectrum and the corresponding dielectric loss functions exhibit a single peak. The peaks of the dielectric loss are however broadened with respect to the Debye model, which indicates that more than one relaxation time is involved.

5.2 Dielectric Media with Positive Relaxation Spectra

In the Cole-Cole model the complex dielectric function has the following form

$$\tilde{\mathcal{E}}_{\mathrm{CC}}(p) = \frac{\tilde{\varepsilon}(p)}{p} = \frac{\varepsilon_\infty}{p} + \frac{1}{p}\frac{\varepsilon_{\mathrm{s}} - \varepsilon_\infty}{1 + (p\tau_0)^\alpha} = \frac{\varepsilon_{\mathrm{s}}}{p} - \frac{1}{p}\frac{(\varepsilon_{\mathrm{s}} - \varepsilon_\infty)(p\tau_0)^\alpha}{1 + (p\tau_0)^\alpha} \tag{84}$$

where $0 \le \alpha \le 1$ and $\varepsilon_{\mathrm{s}} \ge \varepsilon_\infty$ in accordance with (67). The time-domain dielectric relaxation function can be expressed in terms of the Mittag-Leffler function

$$E_\alpha(z) := \sum_{n=0}^{\infty} \frac{z^n}{\Gamma(\alpha n + 1)} \tag{85}$$

[15]. Applying the integral representation

$$\int_0^\infty \mathrm{e}^{-pt} E_\alpha\left(-a t^\alpha\right) \mathrm{d}t = \frac{p^{\alpha-1}}{a + p^\alpha} \tag{86}$$

[52] to (84), it is easily shown that

$$\mathcal{E}_{\mathrm{CC}}(t) = \varepsilon_\infty \theta(t) + (\varepsilon_{\mathrm{s}} - \varepsilon_\infty)\left[1 - E_\alpha\left(-(t/\tau_0)^\alpha\right)\right]\theta(t) \tag{87}$$

The Mittag-Leffler function in the second term is completely monotone [55] and bounded by 1, hence $\mathcal{E}_{\mathrm{CC}}$ is a Bernstein function. Since $E_1(z) = \mathrm{e}^z$, the limit $\alpha = 1$ corresponds to the Debye relaxation.

The same result can be obtained by applying Theorem 3.13. Let $p = r\mathrm{e}^{\mathrm{i}\phi}$, $0 < \phi < \pi$. Since

$$\mathrm{Im}[p\tilde{\mathcal{E}}_{\mathrm{CC}}(p)] = -\frac{(\tau_0|p|)^\alpha \sin(\alpha\phi)\varepsilon_\infty}{|1 + (\tau_0 p)^\alpha|^2} \le 0$$

the derivative $\mathcal{E}'_{\mathrm{CC}}$ of $\mathcal{E}_{\mathrm{CC}}$ is completely monotone. Since $\mathcal{E}_{\mathrm{CC}}$ is also non-negative, it is a Bernstein function.

From (19)

$$\tilde{\Lambda}_{\mathrm{CC}}(p) = \frac{1}{\varepsilon_{\mathrm{s}}}\left[\frac{1}{p} + (1 - \Delta)\frac{p^{\alpha-1}}{1 + \Delta(p\tau_0)^\alpha}\right] \tag{88}$$

where

$$\Delta := 1 - \frac{\varepsilon_\infty}{\varepsilon_{\mathrm{s}}} \tag{89}$$

satisfies the inequalities $0 < \Delta < 1$. In the time domain

$$\Lambda_{\mathrm{CC}}(t) = \frac{1}{\varepsilon_{\mathrm{s}}}\left[1 + \frac{1-\Delta}{\Delta} E_\alpha\left(-\left(\frac{t}{\tau_1}\right)^\alpha\right)\right]\theta(t) \tag{90}$$

with $\tau_1 := \Delta^{1/\alpha}\tau_0$, which is a completely monotone function [55]. The function $\mathcal{E}_{\mathrm{CC}}(t)$ is a Bernstein function.

The Havriliak-Negami relaxation is a generalization of the Cole-Cole relaxation and it is used to account for the asymmetry of the α peak in glass-forming materials:

$$\tilde{\mathcal{E}}_{\mathrm{HN}}(p) = \frac{\tilde{\varepsilon}(p)}{p} = \frac{\varepsilon_\infty}{p} + \frac{1}{p}\frac{\varepsilon_{\mathrm{s}} - \varepsilon_\infty}{[1 + (p\tau_0)^\alpha]^\beta} \tag{91}$$

Let $p = r\mathrm{e}^{\mathrm{i}\phi}$, $r > 0$, $0 < \phi < \pi$. For $r \to \infty$ the right-hand side of $\mathrm{Im}[p\tilde{\mathcal{E}}(p)]$ is asymptotically equal to $-\sin(\alpha\beta\phi)/(r\tau_0)^{\alpha\beta}$. Hence for $\alpha\beta > 1$ the function $\mathcal{E}'$ is not completely monotone and $\mathcal{E}$ is not a Bernstein function.

For $\beta = 1$ the Havriliak-Negami relaxation reduces to the Cole-Cole model, while for $\alpha = 1$ it is known as the Cole-Davidson model [11]. The latter is often used to represent the asymmetric α peak in glass-forming materials, in contrast to the Cole-Cole relaxation modeling symmetric non-Debye peaks of the imaginary part of the complex dielectric permittivity.

The Havriliak-Negami relaxation function $\mathcal{E}_{\mathrm{HN}}$ can be expressed in terms of the Prabhakar generalized Mittag-Leffler function [53]

$$E^\gamma_{\alpha,\beta}(z) := \sum_{n=0}^{\infty} \frac{\Gamma(\gamma + n)}{\Gamma(\gamma)\Gamma(\alpha n + \beta) n!} z^n \tag{92}$$

for $\mathrm{Re}\,\alpha > 0$, $\mathrm{Re}\,\beta > 0$ (Appendix B). Applying the identity (132) to (91) yields an explicit expression for the Havriliak-Negami relaxation:

$$\mathcal{E}_{\mathrm{HN}}(t) = \left\{\varepsilon_\infty + (\varepsilon_{\mathrm{s}} - \varepsilon_\infty)\left[1 - (t/\tau_0)^{\alpha\beta} E^\beta_{\alpha,1+\alpha\beta}\left(-\left(\frac{t}{\tau_0}\right)^\alpha\right)\right]\right\}\theta(t) \tag{93}$$

The function $f(t) := E^\beta_{\alpha,1}(-t^\alpha)$ is completely monotone if $0 \le \alpha \le 1$, $0 \le \beta \le 1$. Indeed, the Laplace transform $\tilde{f}$ of f is

$$\tilde{f}(p) = \frac{1}{p\,(1 + p^{-\alpha})^\beta}$$

Hence $\mathrm{Im}[p\tilde{f}(p)] = \mathrm{Im}[1 + r^{-\alpha}\mathrm{e}^{\mathrm{i}\alpha\phi}]^\beta / |1 + p^{-\alpha}|^{2\beta}$, where $p = r\mathrm{e}^{\mathrm{i}\phi}$. In the upper half $\mathbb{C}_+$ of the principal Riemann sheet of the complex p-plane $0 < \phi < \pi$. Since $0 \le \alpha \le 1$, the complex number $z := r^{-\alpha}\mathrm{e}^{\mathrm{i}\alpha\phi}$ as well as $1 + z$ lies in $\mathbb{C}_+$. Hence $(1 + z)^\beta \in \mathbb{C}_+$, and $\mathrm{Im}[p\tilde{f}(p)] \ge 0$. Consequently f is CM under the restrictions on α, β assumed above. Since $E^\beta_{\alpha,1}(0) = 1$, the function $1 - E^\beta_{\alpha,1}(-(t/\tau_0)^\alpha)$ is nonnegative and its derivative is completely monotone. This completes the proof that $\mathcal{E}_{\mathrm{HN}}$ is a Bernstein function if $0 \le \alpha, \beta \le 1$ and its counterpart Λ_{HN} is completely monotone.

The relaxation models of Cole-Cole, Cole-Davidson and Havriliak-Negami are distinguishable by inspection of their Cole-Cole plots. The Cole-Davidson relaxation differs from the Cole-Cole and Havriliak-Negami by an exponential decay. In fact, for each of these relaxation models the relaxation function can be expressed in terms of a complex contour integral along a Hankel loop $\mathcal{H}$ encircling the cut on the negative real axis and hence to the Laplace transform of the jump of the complex permittivity on the cut:

$$\mathcal{E}(t) = \frac{1}{\pi} \int_0^\infty \frac{\operatorname{Im} \varepsilon(r\mathrm{e}^{\mathrm{i}\pi})}{r} \mathrm{e}^{-rt} \, \mathrm{d}r \tag{94}$$

In contrast to the Cole-Cole and Havriliak-Negami models the cut for the Cole-Davidson model starts at $-1/\tau_0$ which implies that the lower limit of integration in (94) can be shifted to $1/\tau_0$. It follows immediately that the Cole-Davidson relaxation $\mathcal{E}_{\mathrm{CD}}$ decays faster that e^{-t/τ_0}. On the other hand the algebraic decay rates of the Cole-Cole and Havriliak-Negami models can be obtained from the behavior of the complex dielectric permittivity at low frequency by applying the Karamata Abelian theorems [13, 66].

Dielectric response functions can also be expressed in terms of the Fox function in both time and frequency domain [35].

The Kohlrausch-Watts-Williams (KWW) relaxation function, also known as the stretched exponential,

$$\Lambda(t) = \Lambda_{\mathrm{KWW}}(t) := b\theta(t)\mathrm{e}^{-(t/\tau_0)^\alpha} \tag{95}$$

$\tau_0, b > 0$, was originally introduced by Williams and Watts for the phenomenological description of dielectric relaxation in [67, 68], cf. [12] for more recent references. While most phenomenological models of the dielectric response are primarily expressed in the frequency domain, the KWW response is an exception. Its Laplace transform is a rather complicated expression involving the hypergeometric functions, except for $\alpha = 1/2$, in which case

$$\tilde{\Lambda}(p) = \frac{1}{p} \left[1 - \frac{\mathrm{e}^{1/(4p)} \pi^{1/2} \operatorname{erfc}(-1/(2p^{1/2}))}{2p^{1/2}} \right] \tag{96}$$

The KWW response function is completely monotone if $0 \le \alpha \le 1$. Indeed, the function $\mathrm{e}^{-(t/\tau_0)^\alpha}$ is the Laplace transform of a Lévy stable one-sided probability density p_α [48]

$$\Lambda_{\mathrm{KWW}}(t) = \tau_0 \int_0^\infty p_\alpha(r\tau_0)\mathrm{e}^{-rt} \, \mathrm{d}r, \quad t \ge 0 \tag{97}$$

and hence the Bernstein theorem implies that Λ_{KWW} is a CM function. The behavior of Λ_{KWW} for small t is similar to Λ_{CC}. For large t the KWW relaxation function has an almost exponential decay, as opposed to the algebraic decay of its Cole-Cole counterpart. The Fourier transform of the KWW relaxation function is often used as an alternative model of the asymmetric α peak in glass-forming materials [37].

For $\alpha \ge 1$ the stretched exponential is no longer a completely monotone function. For $0 \le \alpha \le 2$ the stretched exponential is a CPD function. For $\alpha = 2$ this follows from the Bochner theorem. For $1 < \alpha < 2$ the function $|x|^\alpha$ is a negative definite function and the stretched exponential is CPD by a theorem of Schoenberg (a proof can be found in Appendix D). For $\alpha > 2$ the stretched exponential is not a CPD function.

It is also worth noting that $f(t) := 1 - \exp(-(t/\tau)^\alpha)$, $t \ge 0$, is a Bernstein function for $0 < \alpha \le 1$. Indeed, $f(t) \ge 0$ and $f'(t) = \alpha(t/\tau)^{\alpha-1} \exp(-(t/\tau)^\alpha)$ is the product of two CM functions, hence it is CM.

A detailed investigation of the properties of the stretched exponential can be found in [1, 36].

A related example is the generalized Bordewijk model [5]

$$\Lambda(t) = \Lambda_{\mathrm{B}}(t) := b\mathrm{e}^{-t/\tau_1 - (t/\tau_0)^\alpha} \tag{98}$$

with $b, \tau_0, \tau_1 > 0$. The function Λ_{B} can be expressed in terms of the α-stable Lévy probability density p_α

$$\Lambda_{\mathrm{KWW}}(t) = \tau_0 \int_{1/\tau_1}^{\infty} p_\alpha((r - 1/\tau_1)\tau_0)\mathrm{e}^{-rt}\,\mathrm{d}r \tag{99}$$

which demonstrates that it is also completely monotone for $0 < \alpha < 1$.

5.3 Anisotropic Dielectric Response Functions with Positive Relaxation Spectrum

The simplest anisotropic relaxation models of dielectric relaxation can be constructed by superposing scalar relaxations:

$$\Lambda(t) = \sum_{k=1}^{3} \lambda_k(t)\mathsf{P}_k \tag{100}$$

$$\mathsf{R}(t) = \sum_{k=1}^{3} \varepsilon_k(t)\mathsf{P}_k \tag{101}$$

where P_k are constant projection operators on $\mathbb{R}^3$, $\mathsf{P}_k\mathsf{P}_l = 0$ if $k \neq l$, $\mathsf{P}_k^2 = \mathsf{P}_k$, $\sum_{k=1}^3 \mathsf{P}_k = \mathsf{I}$ and $\lambda_k * \varepsilon_k(t) = t_+$, $k = 1, 2, 3$. Λ (R) is CM (BF, respectively) if and only if the functions λ_k (ε_k, respectively) are CM. The theory developed in Sects. 3.3 and 4 allows for more general CM (BF) anisotropic functions Λ (R, respectively), as demonstrated by an example in [32].

5.4 Dielectric Response with Non-positive Relaxation Spectra (Lorentz-Dispersive Dielectric Media)

Inertial effects associated with molecule rotation play an important role in metals and ionic crystals. They are accounted for in the following generalization of the Van Vleck-Weisskopf-Fröhlich model [54]

$$\mathcal{E}(t) := \varDelta\big[a - f(t)\big]\theta(t), \qquad \mu, \gamma > 0 \tag{102}$$

$f(t) = \mathrm{e}^{-\gamma t}\cos(\mu t)$, $\varDelta > 0$, $a \geq 1$ (in the Van Vleck-Weisskopf-Fröhlich model $a = 1$). The function $\mathcal{E}$ is negative definite (Theorem C.1). The Laplace transform of $\mathcal{E}$ is

$$\tilde{\mathcal{E}}(p) = \left[\frac{a}{p} - \frac{p+\gamma}{(p+\gamma)^2 + \mu^2}\right]\varDelta$$

Equation (41) is satisfied if $\gamma \geq 0$. Since

$$\tilde{\lambda}(\mathrm{i}\omega) = -1/\big[\omega^2\tilde{\mathcal{E}}(\mathrm{i}\omega)\big]$$

it is easy to see from Theorem 3.6 that $\lambda \in \mathfrak{D}$. $\mathcal{E}$ is a Bernstein function if $a \geq 1$ and $\mu = 0$. If $\mu = 0$, then

$$\tilde{\lambda}(p) = \frac{p+\gamma}{p[(a-1)p + a\gamma]} \tag{103}$$

Hence $\mathrm{Re}\,\tilde{\lambda}(\mathrm{i}\omega) = \gamma/[(a-1)^2\omega^2 + a^2\gamma^2] \geq 0$ for all $\omega \in \mathbb{R}$, in accordance with (41) and with Theorem 3.6 (ii). If $a \neq 1$, then $\tilde{\lambda}(\mathrm{i}\omega) \to 0$.

In particular, if $a = 1$ then

$$\mathcal{E}(t) = 1 - \mathrm{e}^{-\gamma t}, \quad \gamma > 0$$
$$\tilde{\Lambda}(p) = \frac{1}{\gamma} + \frac{1}{p}$$
$$\Lambda(t) = \frac{1}{\gamma}\delta(t) + \theta(t)$$

The Drude-Lorentz complex permittivity is in the simplest case given by the formula

$$\varepsilon(p) = \varepsilon_0 + \frac{a}{b + \tau^2 p^2 + \tau p} \tag{104}$$

with $a, b, \varepsilon_0 > 0$. Since $\mathrm{Re}[p\varepsilon(p)] \geq 0$ for $\mathrm{Re}\, p > 0$, $\mathrm{D}^2\mathsf{R}$ is CPD and R is negative definite.

6 Energy Density Functionals Based on Spectral Properties of the Dielectric and Magnetic Response Functions

6.1 The Energy Functional

We shall attempt to define the energy density functional U of the electromagnetic field interacting with matter in such a way that the constitutive equations imply the inequality

$$W \geq \dot{U} \tag{105}$$

where W is the electromagnetic power, defined in Sect. 2. The difference $D := W - \dot{U}$ is identified as the energy dissipation rate.

We note here that an alternative choice of the electromagnetic energy density functional, such as

$$U_1 = \frac{\varepsilon_0}{2}\mathbf{E}^2 + \frac{1}{2\varepsilon_0}\mathbf{B}^2$$

[51] with the energy flux density

$$\mathbf{S}_1 = \varepsilon_0\,[\mathbf{E} \times \mathbf{H}]$$

is not appropriate for our approach because

$$\dot{U}_1 + \mathrm{div}\,\mathbf{S}_1 = -\mathbf{E}^\top \cdot \mathbf{J}^{\mathrm{tot}} = -\mathbf{E}^\top \cdot \mathbf{J} - \mathbf{E}^\top \cdot \mathbf{J}^{\mathrm{bound}}$$

involves the displacement current $\mathbf{J}^{\mathrm{bound}}$, which is not a controllable variable.

We shall give rigorous derivations of two different energy density functionals. The first one (Sect. 6.2) leads to a total energy which is strictly dissipated, while the second one (Sect. 6.3) is such that the total energy is a constant of motion.

By isolating the energy density from the energy-momentum density we have implicitly committed ourselves to a fixed reference frame. This frame can be thought of as the rest frame of the material medium. The theory is however implicitly non-relativistic for yet another reason: the constitutive equations involve convolution in the time variable only.

6.2 A Dissipative Energy Density and a Dissipation Rate Functional

It is well-known that given a linear constitutive equations of a dispersive medium, (105) does not determine a unique energy density functional [6, 44]. A special energy density functional can however be associated with the LICM class of response functions Λ, N. According to the Bernstein theorem there are two non-decreasing tensor-valued functions Ω, P such that

$$\Lambda(t) = \int_{[0,\infty[} \mathrm{e}^{-\xi t} \Omega(\mathrm{d}\xi) \tag{106}$$

$$\mathsf{N}(t) = \int_{[0,\infty[} \mathrm{e}^{-\xi t} \mathsf{P}(\mathrm{d}\xi) \tag{107}$$

Since the matrices $\Lambda(t), \mathsf{N}(t)$ are symmetric for $t \geq 0$, the theorem about the inversion of the Laplace-Stieltjes transform [66] implies that the matrices $\Omega(\xi)$, $\mathsf{P}(\xi)$ are symmetric for all $\xi \geq 0$. We shall define two auxiliary fields

$$\mathbf{y}(t,\xi) := \int_{-\infty}^{t} \mathrm{e}^{-\xi(t-s)} \dot{\mathbf{D}}(s)\, \mathrm{d}s \tag{108}$$

$$\mathbf{z}(t,\xi) := \int_{-\infty}^{t} \mathrm{e}^{-\xi(t-s)} \dot{\mathbf{B}}(s)\, \mathrm{d}s \tag{109}$$

so that, in view of (106, 107),

$$\mathbf{E}(t) = \int_{[0,\infty[} \Omega(\mathrm{d}\xi) \mathbf{y}(t,\xi) \tag{110}$$

$$\mathbf{H}(t) = \int_{[0,\infty[} \mathsf{P}(\mathrm{d}\xi) \mathbf{z}(t,\xi) \tag{111}$$

The auxiliary fields $\mathbf{y}, \mathbf{z}$ satisfy the differential equations

$$\frac{\mathrm{d}\mathbf{y}}{\mathrm{d}t} + \xi \mathbf{y}(t,\xi) = \dot{\mathbf{D}}(t) \tag{112}$$

$$\frac{\mathrm{d}\mathbf{z}}{\mathrm{d}t} + \xi \mathbf{z}(t,\xi) = \dot{\mathbf{B}}(t) \tag{113}$$

The energy density functional U_{D} is defined by the formula

$$U_{\mathrm{D}}(t) = \frac{1}{2} \int_{[0,\infty[} \left[\mathbf{y}(t,\xi)^{\dagger} \Omega(\mathrm{d}\xi) \mathbf{y}(t,\xi) + \mathbf{z}(t,\xi)^{\dagger} \mathsf{P}(\mathrm{d}\xi) \mathbf{z}(t,\xi) \right] \tag{114}$$

The rate of the energy density

$$\begin{aligned} \dot{U}_{\mathrm{D}}(t) &= \int_{[0,\infty[} \left[\mathbf{y}_{,t}(t,\xi)^{\dagger} \Omega(\mathrm{d}\xi) \mathbf{y}(t,\xi) + \mathbf{z}_{,t}(t,\xi)^{\dagger} \mathsf{P}(\mathrm{d}\xi) \mathbf{z}(t,\xi) \right] \\ &= \mathbf{E}^{\top} \dot{\mathbf{D}} + \mathbf{H}^{\top} \dot{\mathbf{B}} - D(t) \end{aligned}$$

where the intrinsic dissipation rate D defined by the formula

$$D(t) := \int_{[0,\infty[} \xi \left[\mathbf{y}(t,\xi)^{\dagger} \Omega(\mathrm{d}\xi) \mathbf{y}(t,\xi) + \mathbf{z}(t,\xi)^{\dagger} \mathsf{P}(\mathrm{d}\xi) \mathbf{z}(t,\xi) \right] \geq 0 \tag{115}$$

represents the dissipation associated with the dispersive properties of the dielectric medium. On account of the Poynting identity

$$\dot{U} = \operatorname{div}(\mathbf{E} \times \mathbf{H}) - D - \mathbf{J}^{\top}\mathbf{E}, \quad D \geq 0 \tag{116}$$

6.3 A Conserved Energy Density Functional for Positive Definite Response Functions

We shall show that in the absence of electric current and external energy sources the Maxwell equations of motion have an integral of motion which can be interpreted as an energy [58]. The appropriate energy density functional will be derived from the Fourier integral representation of Λ. In contrast to the Laplace-Stieltjes representation, which applies only to LICM **D**-to-**E** kernels, the Fourier representation remains valid in the dielectric media with a bounded response function Λ in the CPD class. In particular, a bounded LICM function is CPD and satisfies the Bochner theorem. This means that an arbitrary dielectric which is a perfect insulator admits a conserved energy. With an appropriate choice of canonical variables this energy becomes a Hamiltonian with respect to a canonical Poisson bracket, but we shall not pursue this subject here.

The CPD kernels $\Lambda(t)$ and $\mathsf{N}(t)$ are real tensor-valued and therefore they can be expressed as the real parts of one-sided Fourier integrals of positive definite tensor-valued Radon measures S, T:

$$\Lambda(t) = \operatorname{Re} \int_{[0,\infty[} \mathrm{e}^{\mathrm{i}\zeta t} \mathsf{S}(\mathrm{d}\zeta)$$

$$\mathsf{N}(t) = \operatorname{Re} \int_{[0,\infty[} \mathrm{e}^{\mathrm{i}\zeta t} \mathsf{T}(\mathrm{d}\zeta)$$

Define the energy density functional U_{C} by the formula

$$U_{\mathrm{C}}(t) := \frac{1}{2} \int_{[0,\infty[} \left[\mathbf{u}(t,\zeta)^{\dagger} \mathsf{S}(\mathrm{d}\zeta) \mathbf{u}(t,\zeta) + \mathbf{v}(t,\zeta)^{\dagger} \mathsf{T}(\mathrm{d}\zeta) \mathbf{v}(t,\zeta)\right] \tag{117}$$

where the auxiliary fields

$$\mathbf{u}(t,\zeta) := \int_{-\infty}^{t} \mathrm{e}^{\mathrm{i}\zeta(t-s)} \dot{\mathbf{D}}(s)\, \mathrm{d}s \tag{118}$$

$$\mathbf{v}(t,\zeta) := \int_{-\infty}^{t} \mathrm{e}^{\mathrm{i}\zeta(t-s)} \dot{\mathbf{B}}(s)\, \mathrm{d}s \tag{119}$$

satisfy the ordinary differential equations

$$\dot{\mathbf{u}} + \mathrm{i}\zeta \mathbf{u} = \dot{\mathbf{D}} \tag{120}$$

$$\dot{\mathbf{v}} + \mathrm{i}\zeta \mathbf{v} = \dot{\mathbf{B}} \tag{121}$$

It is easy to see that

$$\frac{\mathrm{d}U_{\mathrm{C}}}{\mathrm{d}t} = \mathbf{E}^{\top}\dot{\mathbf{D}} + \mathbf{H}^{\top}\dot{\mathbf{B}} = \operatorname{div}(\mathbf{E} \times \mathbf{H}) - \mathbf{J}^{\top}\mathbf{E} \tag{122}$$

Note that there is no intrinsic dissipation associated with this notion of energy density. The real part $\mathbf{u}_{\mathrm{R}}$ and the imaginary part $\mathbf{u}_{\mathrm{I}}$ of $\mathbf{u}$ satisfy the equations

$$\ddot{\mathbf{u}}_{\mathrm{R}} + \zeta^2 \mathbf{u}_{\mathrm{R}} = \ddot{\mathbf{D}} \tag{123}$$

$$\ddot{\mathbf{u}}_{\mathrm{I}} + \zeta^2 \mathbf{u}_{\mathrm{I}} = -\zeta \dot{\mathbf{D}} \tag{124}$$

which shows that the auxiliary fields represent a one-parameter family of field-driven oscillators.

A conserved viscoelastic energy functional is considered in [31]. It is shown that the dissipated energy resides in a continuum of oscillators driven by the strain rate. A similar result was obtained for dielectrics by Tip [60] by a different argument. In [60] the oscillators are interpreted as representing the energy of matter interacting with the pure electromagnetic field. The rate of the energy density can be decomposed into two terms

$$\frac{\mathrm{d}U_{\mathrm{D}}}{\mathrm{d}t} = \frac{\mathrm{d}U_0}{\mathrm{d}t} + \mathbf{E}^\top \dot{\mathbf{P}} + \mathbf{H}^\top \mathsf{Q}_0 \dot{\mathbf{M}}$$

where $U_0 := (1/2)(\mathbf{E}^\top \mathsf{R}_0 \mathbf{E} + \mathbf{H}^\top \mathsf{Q}_0 \mathbf{H})$ by splitting the electric displacement field and magnetic induction field into the free space fields, polarization and magnetization,

$$\mathbf{D} = \mathsf{R}_0 \mathbf{E} + \mathbf{P} \tag{125}$$

$$\mathbf{B} = \mathsf{Q}_0 (\mathbf{H} + \mathbf{M}) \tag{126}$$

where $\mathsf{R}_0 = \mathsf{R}(0)$, $\mathsf{Q}_0 = \mathsf{Q}(0)$ [59]. U_0 can be interpreted as the energy density of the pure electromagnetic field. Existence of a conserved total energy implies that the sum of the second and third term on the right-hand side represents the rate of the energy of the matter subsystem.

A fairly straightforward derivation of the conserved energy can be found in [25]. It is based on a different assumption and is not sufficiently rigorous. An important contribution of [25] is showing that the energy U_0 of the pure field may propagate at superluminal speed.

Stallinga extended Tip's method to energy-momentum conservation in [59]. Notwithstanding the extension to energy-momentum conservation, Stallinga's theory remains non-relativistic because the constitutive equations are formulated in terms of time convolutions.

7 Conclusions

Dissipative properties of dielectric relaxation can be ensured by assuming that the dielectric response functions have some particular spectral properties. These assumptions are necessarily much stronger than positive semi-definiteness of the dielectric loss. It has been shown that in the case of Lorentz dispersion the dielectric response function is negative definite. A narrower class of admissible response functions is obtained from the assumption that the **D**-to-**E** constitutive relation is a passive system. Dipolar dielectrics have a non-negative relaxation spectrum and therefore the corresponding dielectric relaxation functions belong to the class of Bernstein functions, which is a proper subset of the class of negative definite functions. A relation between the **D**-to-**E** constitutive equation and the more familiar **E**-to-**D** constitutive equation is established. It is shown that empirical and theoretical models of dielectric response are consistent with the proposed theoretical framework.

All the three classes of dielectric response are compatible with the existence of a conserved energy. The expression for the conserved energy obtained here is general and rigorous, as opposed to the Brillouin energy [8, 44]. LICM response functions additionally admit an energy that decays monotonely in a closed system.

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Appendix A: Two Lemmas Needed in the Proof of Theorem 4.2

Lemma A.1 *If $f, g : U \to \mathbb{R}$ and $f(x) \leq g(x)$ for all in $U \subset \mathbb{R}$ then*

$$\sup_U f(x) \leq \sup_U g(x) \tag{127}$$

$$\inf_U f(x) \leq \inf_U g(x) \tag{128}$$

Proof If the supremum of f is reached at $x_0 \in U$ then $\sup f(x) = f(x_0) \leq g(x_0) \leq g(x)$, else there is a sequence $x_n \in U$ such that

$$\sup f(x) \leq f(x_n) + 1/n \leq g(x_n) + 1/n \leq \sup g(x) + 1/n$$

for all $n \in \mathbb{Z}_+$, which proves (127).

For the infimum we have $\inf f(x) \leq f(x_0) \leq g(x_0) = \inf g(x)$ if $g(x)$ reaches its minimum at $x_0 \in U$ and

$$\inf f(x) - 1/n \leq f(x_n) - 1/n \leq g(x_n) - 1/n \leq \inf g(x)$$

otherwise. This proves (128). □

Lemma A.2 *The eigenvalues μ_n of $p\tilde{\Lambda}(p)$ are non-decreasing functions of $p \in]0, \infty[$.*

Proof Let $0 < p_1 < p_2$. For every $\mathbf{v} \in \mathbb{C}^d$

$$\mathbf{v}^\dagger p_1 \tilde{\Lambda}(p_1)\mathbf{v} \leq \mathbf{v}^\dagger p_2 \tilde{\Lambda}(p_2)\mathbf{v}$$

hence, by the Courant-Fischer formula,

$$\mu_n(p) = \sup_{\dim V = n} \inf_{\mathbf{v} \in V \cap S_1} \langle \mathbf{v}, p\tilde{\Lambda}(p)\mathbf{v}\rangle \tag{129}$$

[43] and Lemma A.1, $\mu_n(p_1) \leq \mu_n(p_2)$, which proves that μ_n is a non-decreasing function of p. □

Appendix B: The Prabhakar Mittag-Leffler Function and its Laplace Transform

Let $\alpha, \beta > 0$. Substituting the series representation of the Prabhakar generalized Mittag-Leffler function in the Laplace transformation yields the identity

$$\int_0^\infty \mathrm{e}^{-pt} t^{\beta-1} E^\gamma_{\alpha,\beta}(at^\alpha)\, \mathrm{d}t = p^{-\beta} \sum_{n=0}^\infty \frac{\Gamma(\gamma+n)}{\Gamma(\gamma)n!} \left(\frac{a}{p^\alpha}\right)^n \tag{130}$$

On the other hand

$$(1+z)^{-\gamma} = \sum_{n=0}^\infty \frac{\Gamma(1-\gamma)}{\Gamma(1-\gamma-n)n!} z^n = \sum_{n=0}^\infty (-1)^n \frac{\Gamma(\gamma+n)}{\Gamma(\gamma)n!} z^n \tag{131}$$

Comparison of (130) and (131) yields the Laplace transform

$$\int_0^\infty \mathrm{e}^{-pt} t^{\beta-1} E^{\gamma}_{\alpha,\beta}\,(at^{\alpha})\,\mathrm{d}t = p^{-\beta}\frac{1}{(1-ap^{-\alpha})^{\gamma}} \tag{132}$$

Equation (132) holds for $\operatorname{Re}\alpha > 0$, $\operatorname{Re}\beta > 0$.

Appendix C: Positive Definite and Negative Definite Functions

A function $f : \mathbb{R} \to \mathcal{M}$ is *positive definite* if

$$\sum_{k,l=1}^{N} f(x_k - x_l)\overline{y}_k y_l \geq 0$$

for all $N \in \mathbb{Z}_+$, all $x_k \in \mathbb{R}$ and all $y_k \in \mathbb{C}$, $1 \leq k \leq N$.

A function $g : \mathbb{R} \to \mathcal{M}$ is *negative definite* if

$$\sum_{k,l=1}^{N} f(x_k - x_l)\overline{y}_k y_l \leq 0$$

for all $N \in \mathbb{Z}_+$, all $x_k \in \mathbb{R}$ and all $y_k \in \mathbb{C}$, $1 \leq k \leq N$, such that $\sum_{k=1}^{N} y_k = 0$.

Both definitions can be extended to Abelian groups with involution ($x \to -x$ is the involution in $\mathbb{R}$ in the definition given above). Taking the involution to be the identity, the positive definite functions and the negative definite functions become the completely monotone functions and the Bernstein functions respectively under some provisions.

Theorem 4.5 has a counterpart in the class of negative and positive definite functions [3]:

Theorem C.1 *If the function f is PD and $f(t) \leq a$, then $a - f(t)$ is negative definite.*

Theorem C.1 is a trivial generalization of Corollary 7.7 in [3].

A real-valued negative definite function g bounded from below has the Lévy-Khinchin integral representation:

$$g(x) = g(0) + ax^2 + \int_{]0,\infty[} \left[1 - \cos(xy)\right] m(\mathrm{d}y) \tag{133}$$

where $a \geq 0$ and the Lévy measure m is a positive measure on $\mathbb{R}_+$ satisfying the inequality

$$\int_{]0,\infty[} \left[1 - \cos(xy)\right] m(\mathrm{d}y) < \infty$$

[2, 22]. Equation (133) follows from Theorem 4.3.19 in [2]. In terms of the measure $\mu(\mathrm{d}y) = y^2 m(\mathrm{d}y)$, with $\mu(\{0\}) = 2a$, we have

$$g(x) = g(0) + \int_{[0,\infty[} \frac{1-\cos(xy)}{y^2}\mu(\mathrm{d}y)$$

The concept of negative definite functions and the representation (133) can be extended to matrix- and tensor-valued functions by the usual argument.

Let $\mathsf{F} : \overline{\mathbb{R}_+} \to \mathcal{M}$ be a continuous causal positive definite function. Let

$$\mathsf{G}(s) = \begin{cases} \mathsf{F}(s), & s \geq 0 \\ \mathsf{F}(-s)^\dagger, & s < 0 \end{cases} \tag{134}$$

Equation (33) is equivalent to

$$\int_{-\infty}^{\infty} \left[\int_{-\infty}^{\infty} \mathbf{v}(t)^\dagger \mathsf{G}(s) \mathbf{v}(t-s)\, \mathrm{d}s \right] \mathrm{d}t \geq 0 \tag{135}$$

Appendix D: Proof of a Theorem on the Stretched Exponential

It is easy to see that x^2 is a negative definite function on $\mathbb{R}$. Consequently $x \to \exp(-ax^2)$ is positive definite on $\mathbb{R}$ and $x \to 1 - \exp(-ax^2)$ is negative definite on $\mathbb{R}$ for every $a \geq 0$.

The measure $m(\mathrm{d}a) := a^{-1-\alpha/2}\, \mathrm{d}a$ satisfies the inequality

$$\int_{[0,\infty[} \frac{a}{1+a} m(\mathrm{d}a) < \infty$$

hence the integral in

$$x^\alpha = (x^2)^{\alpha/2} = \frac{\alpha}{2\Gamma(1-\alpha/2)} \int_0^\infty \left(1 - \mathrm{e}^{-ax^2}\right) m(\mathrm{d}a)$$

is well-defined and is negative definite on $\mathbb{R}$ for $0 < \alpha \leq 2$. Consequently $x \to \mathrm{e}^{-x^\alpha}$ is positive definite on $\mathbb{R}$ and, therefore, the function $x \to \theta(x)\mathrm{e}^{-x^\alpha}$ is positive definite if $0 < \alpha \leq 2$.

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