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Abstract: In this paper, we show that spectral functions relevant for commonly used models of the non-Debye relaxation are related to the Stieltjes functions supported on the positive semi-axis. Using only this property, it can be shown that the response and relaxation functions are non-negative. They are connected to each other and obey the time evolution provided by integral equations involving the memory function M(t), which is the Stieltjes function as well. This fact is also due to the Stieltjes character of the spectral function. Stochastic processes-based approach to the relaxation phenomena gives the possibility to identify the memory function M(t) with the Laplace (Lévy) exponent of some infinitely divisible stochastic processes and to introduce its partner memory k(t). Both memories are related by the Sonine equation and lead to equivalent evolution equations which may be freely interchanged in dependence of our knowledge on memories governing the process.

Keywords: non-Debye relaxations; positive definite functions; Sonine equation; Laplace (Lévy) exponent



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1. Introduction

The main information on the nature of dielectric relaxation phenomena comes from broadband dielectric spectroscopy [1], which provides us with data concerning dispersive and absorptive properties of dielectric materials. These properties are encoded in the complex dielectric permittivity $\hat{\varepsilon}(i\omega)$ and its dependence on the frequency of external fields. In particular, data of the spectroscopy experiments enable one to determine the behavior of permittivity $\hat{\varepsilon}(i\omega)$ for asymptotic values of the frequency ω scaled with respect to some relaxation time τ , which characterizes the material under investigation. The pioneer of this research was A. K. Jonscher, who in the 60's and 70's of the previous century, studied with his collaborators a vast majority of available data, and observed that they shared universal asymptotic properties:

$$\hat{\varepsilon}(i\omega) \propto (i\omega\tau)^{a-1}, \quad \omega\tau \ll 1$$

$$\Delta\hat{\varepsilon}(i\omega) = \varepsilon_0 - \hat{\varepsilon}(i\omega) \propto (i\omega\tau)^b, \quad \omega\tau \gg 1,$$
(1)

nowadays known as the Universal Relaxation Law (URL) [2]. In the above, the static permittivity ε_0 denotes the limit of $\hat{\varepsilon}(i\omega)$ for $\omega \to 0$ and the parameters 1 - a and b belong to the range (0, 1). The Jonscher URL agrees with the most commonly used phenomenological models of the relaxation phenomena, namely with the Havrilak–Negami (HN) and the Jurlewicz–Weron–Stanislavski (JWS) models, both depending on a single characteristic time. For the HN model, $b \ge a - 1$, which means that the exponent governing asymptotics at infinity is larger than its counterpart governing asymptotics at zero. The opposite situation occurs for the JWS model, for which b < a - 1.

Recall that the normalized ratio of permittivities $[\hat{\varepsilon}(i\omega) - \varepsilon_{\infty}]/[\varepsilon_0 - \varepsilon_{\infty}]$ (here, ε_{∞} means the infinite frequency limit of $\hat{\varepsilon}(i\omega)$) is named the spectral function $\hat{\phi}(i\omega)$. Through

the Laplace transform, it is related to the time domain response and relaxation functions, denoted as $\phi(t)$ and n(t), respectively,

$$\hat{\phi}(i\omega) = \mathcal{L}[\phi(t);i\omega]$$
 and $[1 - \hat{\phi}(i\omega)]/(i\omega) = \mathcal{L}[n(t);i\omega],$ (2)

which simply correspond to each other

$$\phi(t) = -\dot{n}(t). \tag{3}$$

Actually, the transform in Equation (2) is the Fourier transform restricted to the semiaxis, but following physicists' customs, we call it the Laplace transform [3]. Nevertheless, except Section 3.1 of the manuscript, we do not need to use the complex variables, as we will show that it is enough to make considerations for functions supported on the positive semi-axis. This is the reason why we will restrict ourselves to the Laplace integral $\hat{f}(s) = \int_0^{\infty} e^{-st} f(t) dt$ for s > 0. Its difference with respect to the Laplace transform $\hat{f}(z) = \int_0^{\infty} e^{-zt} f(t) dt$ is that *s* is real, while *z* is complex. For s > 0 and $z \in \mathbb{C} \setminus \mathbb{R}^-$, they can be linked to each other through [4] (Theorem 2.6), repeatedly quoted in [5] (Theorem 1). Because of the latter, we feel free to use the same notation \hat{f} for real and complex functions, as well as to call on the *s* or Laplace domain interchangeably.

The time evolution of the response function $\phi(t)$, which for physical reasons is required to be continuous and vanishing for t < 0, is governed by the equation

$$\phi(t) = BM(t) - B \int_0^t M(t - \xi)\phi(\xi) \,\mathrm{d}\xi,\tag{4}$$

where *B* is a nonnegative transition rate constant, and M(t) is an integral kernel which plays the role of memory. For consistency of its further physical and mathematical interpretation, we assume that $M \in L^1_{loc}(\mathbb{R}^+)$. Due to the conditions given by [6,7], there exists another function $k(t) \in L^1_{loc}(\mathbb{R}^+)$, which, together with M(t), satisfies the Sonine equation [8,9]

$$\int_{0}^{t} k(u)M(t-u)du = \int_{0}^{t} k(t-u)M(u)du = 1.$$
(5)

In addition, assuming the conditions (*) in [6,7] are fulfilled, we can find that

$$\hat{k}(s)\hat{M}(s) = s^{-1}.$$
 (6)

The authors of Refs. [10,11] proposed to employ Equation (6) to justify using an integro-differential equation

$$\frac{\mathrm{d}}{\mathrm{d}t} \int_0^t k(t-\xi)\phi(\xi) \,\mathrm{d}\xi = -B\phi(t) \tag{7}$$

as governing the time evolution of the response function. We emphasize that the mathematical structure of the Equation (7) was the subject of the seminal paper [7] whose results give the conditions under which the Cauchy problem for the Equation (7) is uniquely solved.

As it was shown in [12–14], $\hat{M}(s)$ can be expressed by the algebraic inverse of the Laplace (Lévy) exponent $\Psi(s)$, that is, $\hat{M}(s) = [\Psi(s)]^{-1}$, of some infinitely divisible stochastic process U underlying the relaxation. Mathematically, the Laplace (Lévy) exponent corresponds to the characteristic function of the process U [12,13]. For instance, the characteristic function of the Lévy stable process X is given by the Lévy–Khintchine formula, which naturally introduces $\Psi(s)$ as a complete Bernstein function (CBF).

Definition 1. A real function $(0, \infty) \rightarrow h(s)$ is CBF if it is a nonnegative infinitely differentiable function h(s) which satisfies

$$(-1)^{n-1}h^{(n)}(s) \ge 0$$
 for $n = 1, 2, 3...$

Thus, Equation (6) illustrates the relation between the CBF and Sonine pair noticed only a few years ago [7].

The completely Bernstein character of $\Psi(s)$ leads to the crucial observation concerning $\hat{M}(s)$ —because the algebraic inverse of a CBF is a Stieltjes function (SF), then $\hat{M}(s)$ does share this property. Following [7,15], we introduce:

Definition 2. A (non-negative) SF is a function $f : (0, \infty) \mapsto [0, \infty)$, which can be written in the form

$$f(s) = a/s + b + \int_0^\infty \sigma(\mathrm{d}u)/(s+u),\tag{8}$$

where $a, b \ge 0$, and σ is a measure $(0, \infty)$ such that $\int_0^\infty \sigma(du)/(1+u) < \infty$.

We remark that if a = 0, then the Definition 2 is the same as given in [16,17], but considered for complex numbers and complex Stieltjes functions. Thus, SF coming from the Definition 2 is a restriction of a complex SF to be supported to the positive semi-axis. As an alternative definition of SF, justified for its convenience for further consideration, we will also use Theorem 7.3 of [15] which says that $f : (0, \infty) \mapsto [0, \infty)$ is SF if, and only if, 1/f is CBF. Note that SFs form a subclass of completely monotonic functions (CMF), defined as:

Definition 3. A real function $\hat{c}(s) : (0, \infty) \mapsto \mathbb{R}$ is CMF if all its derivatives for n = 0, 1, 2, ... exist and $(-1)^n \hat{c}^{(n)}(s) \ge 0$.

The above implies that $\hat{M}(s)$, as a reciprocal of a CBF, is CMF as well and, according to Refs. [18,19], we can call such completely monotonic integral kernels "fading memories".

A physical interpretation of Equations (4) and (7) yields that they should lead to the same physical results. To endow this property with a mathematical meaning, note that the first of them is an integral equation, whereas the next one is an integral equation can be transformed to its differential analogue. In what follows, we are going to demonstrate that the analogical procedure may be performed for Equations (4) and (7). It means that from Equation (4), we should derive Equation (7). Doing that, we will see the importance of the condition (5) or its analogue in the Laplace domain Equation (6). Moreover, to name the integral kernels M(t) and k(t) as "fading memories", it will be essential to prove that they are given by SFs being the subclass of CMFs. In this paper, we will show all these properties by using the fact that the spectral function in the complex domain can be rewritten as SF.

Our presentation goes as follows. Section 2 shows that for commonly used models of non-Debye relaxations, their spectral functions are given by SFs. Thus, it appears that the response and relaxation functions are non-negative. In Section 3 we assume that the Stieltjes character of the spectral functions is enough to obtain two types of integral kernels which govern the evolution of the response functions, and which also are SFs. Hence, we can call them the "memory functions". We will provide the exact and explicit forms of these memories. Requiring that both equations give the same result, we conclude that the memories have to satisfy the Sonine equation. The paper is summarized in Section 5.

2. Basic Models of Non-Debye Relaxations

The spectral functions of HN and JWS models, which in physical literature are also called relaxation patterns, from construction depend on a single characteristic time τ and are given by

$$\hat{\phi}_{HN;\,\alpha,\beta}(\mathrm{i}\omega) = [1 + (\mathrm{i}\omega\tau)^{\alpha}]^{-\beta} \quad \text{and} \quad \hat{\phi}_{JWS;\,\alpha,\beta}(\mathrm{i}\omega) = 1 - (\mathrm{i}\omega\tau)^{\alpha\beta}\,\hat{\phi}_{HN;\,\alpha,\beta}(\mathrm{i}\omega), \quad (9)$$

where $\alpha, \beta \in (0, 1]$. Values of α and β are obtained from the experiment so they correspond to the URL. For the HN relaxation, we have $a = 1 - \alpha\beta$ and $b = \alpha$, whereas for the JWS model, we get $a = 1 - \alpha$ and $b = \alpha \beta$. For special choices of the parameters, the HN and JWS patterns boil down to other widely used models of non-Debye relaxations, namely the Cole-Davidson (CD), the Cole-Cole (CC), and the Debye relaxation (D). The CD model is obtained from the HN model for $\alpha = 1$ and $\beta \in (0, 1]$, whereas the Cole-Cole pattern (CC) is obtained either from the HN or from the JWS model for $\alpha \in (0, 1]$ and $\beta = 1$. The HN and JWS models for $\alpha = \beta = 1$ reduce to the Debye relaxation (D). If the frequency of an applied electric field is of the order 10^5 – 10^{10} Hz, then fitting the experimental data by a single standard relaxation pattern is no longer satisfactory, and it is much more effective to fit them by a (linear) combination of the above-mentioned non-Debye relaxation models or to use models which belong to the excess wing model class (EW). In particular, the latter involves an extended number of parameters, which introduce more than one characteristic time [20]. Thus, in the high-frequency domain, we deal with more than one characteristic time-scale and the Jonscher URL is not satisfied any longer. For instance, in the simplest version of the EW model, we have two characteristic times, τ_1 and τ_2 , built into the spectral function as follows:

$$\hat{\phi}_{EW;\alpha}(i\omega) = \frac{1 + (i\omega\tau_2)^{\alpha}}{1 + i\omega\tau_1 + (i\omega\tau_2)^{\alpha}}, \qquad 0 \le \alpha \le 1.$$
(10)

The HN, JWS, and EW spectral functions, commonly denoted as $\hat{\phi}_{(\cdot)}(i\omega)$, are the (complex) Stieltjes functions which form the subclass of the Nevanlinna-Pick functions P(z), analytic in the upper-half plane and satisfying $\text{Im}P(z) \ge 0$ for Im z > 0 [15–17]. Analyticity in the upper-half plane guarantees that the Kramers–Kronig relations are satisfied [3]. The link between the Nevanlinna-Pick functions and the CMFs is presented by [4] (Theorem 2.6), and repeatedly quoted in [5] (Theorem 1). As has been signalized in Section 1, we will provide our studies in the real domain.

With the help of the just-mentioned theorem, Equations (9) and (10) can be rewritten as

$$\hat{\phi}_{HN;\alpha,\beta}(s) = [1 + (s\tau)^{\alpha}]^{-\beta} \quad \text{and} \quad \hat{\phi}_{JWS;\alpha,\beta}(s) = 1 - (s\tau)^{\alpha\beta} \,\hat{\phi}_{HN;\alpha,\beta}(s), \tag{11}$$

while

$$\hat{\phi}_{EW;\alpha}(s) = \frac{1 + (s\tau_2)^{\alpha}}{1 + s\tau_1 + (s\tau_2)^{\alpha}} = \frac{1}{1 + \tau_1 s / [1 + (s\tau_2)^{\alpha}]},$$
(12)

where s > 0 and $0 < \alpha, \beta \le 1$.

All these functions are SFs, and their Stieltjes character can be shown using the power function s^{μ} which, due to the value of exponent μ , is either CMF, or SF, or CBF. Namely, for $\mu \leq 0$ it is CMF, for $\mu \in [-1, 0]$ it is SF, and for $\mu \in [0, 1]$ it is CBF. Let us now check if the considered spectral functions really belong to SFs.

Fact 1. From (cb1) it appears that the convex sum $1 + \tau^{\alpha}s^{\alpha}$, $\alpha \in (0, 1]$ is CBF. Moreover, (cb2) gives that the composition of SF (here, σ^{-1} with $\sigma > 0$) and CBF (here, $1 + \tau^{\alpha}s^{\alpha}$) is SF. Thus, $\hat{\phi}_{HN;\alpha,\beta}(s)$ is SF.

The HN spectral function is bounded. Its maximal value equals 1 and the function $\hat{\phi}_{HN;\alpha,\beta}$ decreases to 0 at infinity.

Fact 2. $f(s) = [1 + (s\tau)^{\alpha}]^{\beta}$ is CBF for $0 < \alpha, \beta \le 1$, and the property (cb3) implies that

$$\frac{1}{f(1/s)} = \frac{1}{[1+(s\tau)^{-\alpha}]^{\beta}} = \frac{(s\tau)^{\alpha\beta}}{[1+(s\tau)^{\alpha}]^{\beta}} = (s\tau)^{\alpha\beta}\,\hat{\phi}_{HN;\alpha,\beta}(s)$$

is CBF because it is a linear combination of CBFs with all weight functions equal to 1. The pointwise limit of this combination gives the series for $(s\tau)^{\alpha\beta}\hat{\phi}_{HN;\alpha\beta}(s)$. Then,

$$\sum_{r=0}^{\infty} [(s\tau)^{\alpha\beta} \hat{\phi}_{HN;\,\alpha\beta}(s)]^r = [1 - (s\tau)^{\alpha\beta} \hat{\phi}_{HN;\,\alpha\beta}(s)]^{-1}.$$
(13)

The nonnegative function $(s\tau)^{\alpha\beta} \hat{\phi}_{HN;\alpha,\beta}(s)$ varies from 0 to 1 with increasing s > 0. Thus, we do not need any further restrictions put on the elements of the series in Equation (13)—the equality in Equation (13) is universally true. With a little help of the property (cb7), we obtain that Equation (13) is CBF. Thus, use of the definition of SF given by [15] (Theorem 7.3) or property (cb2) gives that $\hat{\phi}_{IWS;\alpha,\beta}(s)$ is SF.

The JWS spectral function for s > 0 decreases from $\hat{\phi}_{JWS;\alpha,\beta}(0) = 1$ (which is its maximal value) to $0 = \lim_{s \to \infty} \hat{\phi}_{JWS;\alpha,\beta}(s)$ being the minimal value of $\hat{\phi}_{JWS;\alpha,\beta}(s)$.

Fact 3. $1 + (s\tau_2)^{\alpha}$ is CFB for $0 \le \alpha \le 1$, and from the property (cb5) it appears that $\tau_1 s / [1 + (s\tau_2)^{\alpha}]$ for $\tau_1 > 0$ is also CBF. From the property (cb8) we conclude that the EW spectral function $\{1 + \tau_1 s / [1 + (s\tau_2)^{\alpha}]\}^{-1}$ is SF, which decreases from 1 to 0 with increasing *s*.

Assumption 1. The spectral function $\hat{\phi}_{(\cdot)}(s)$ for s > 0 is given by a bounded SF.

At this point of our considerations, we make a comment concerning the physical meaning of our work. Studying measured experimental data, we are able to determine only the relaxation function, its first time-derivative, and eventually the second one. Therefore, the requirement that all derivatives exist and alternate for s > 0, as it is needed by the definition of CMF, is impossible to be verified in practice. Nevertheless, from just listed facts 1–3, we learn that the spectral functions used to fit the data are modelled by SFs, and thus, CMFs. Moreover, Assumption 1 simplifies many calculations because we know that any CMF is uniquely represented by a Laplace integral of a nonnegative function. Indeed, the Bernstein theorem (also called the Bernstein–Widder theorem), according to the classical book by D. V. Widder [21] (Theorem 12a), reads

Theorem 4. A necessary and sufficient condition that f(x) should be completely monotonic in $0 \le x < \infty$ is that

$$f(x) = \int_0^\infty e^{-x\xi} \, \mathrm{d}\alpha(\xi),\tag{14}$$

where $\alpha(\xi)$ is bounded and non-decreasing and the integral converges for $0 \le x < \infty$.

The proof of the Bernstein theorem can be found in [21,22]. We notice that the formulation of the Bernstein theorem can confuse the reader because some of the authors use the Laplace transform (e.g., [7]), but in Widder's and Pollard's approach, the theorem is formulated just in terms of the real valued integral. For the majority of physicists, the name "integral transform" means that we can invert Equation (14), which usually demands using methods of the complex analysis. Here, such considerations are not needed. Hence, we will use Widder's and his student, Pollard's, definition [21,22]. Thanks to the Bernstein theorem, we can claim that:

Proposition 5. The response function $\phi(t)$ and the relaxation function n(t) are nonnegative.

Proof. The proof of nonnegativity of $\phi(t)$ flows immediately from the Bernstein theorem applied to the first formula of Equations (2) which, written in the *s* domain, reads

$$\hat{\phi}(s) = \int_0^\infty e^{-st} \,\phi(t) \,\mathrm{d}t, \qquad \text{where} \quad \phi(t) \ge 0. \tag{15}$$

To show that the relaxation function n(t) is given by a nonnegative function, we use the second formula of Equation (2) in the *s* domain:

$$[1 - \hat{\phi}(s)]/s = \int_0^\infty e^{-st} n(t) \,\mathrm{d}t.$$
 (16)

Note that the function $1 - \hat{\phi}(s)$ can be rewritten as $\hat{\phi}(0) - \hat{\phi}(s)$, where $\hat{\phi}(0)$ is bounded and $\hat{\phi}(0) = 1$ is the maximum of $\hat{\phi}(s)$. Then, the property (cb4) implies that it is CBF, and then, from the definition of CBF, it appears that $[1 - \hat{\phi}(s)]/s$ is SF (see the property (cb6)). Furthermore, Equation (16) means that the Laplace integral of n(t) is equal to $[1 - \hat{\phi}(s)]/s$, which is SF and thus CMF. Then, the Bernstein theorem implies that n(t) is non-negative. \Box

The exact forms of the response function $\phi(t)$ and the relaxation function n(t) for the HN and JWS models can be found in, for example, [23,24]. Relevant formulae are

$$n_{HN;\alpha,\beta}(t) = 1 - (t/\tau)^{\alpha\beta} E^{\beta}_{\alpha,1+\alpha\beta}[-(t/\tau)^{\alpha}], \qquad \phi_{HN;\alpha,\beta}(t) = \tau^{-1} (t/\tau)^{\alpha\beta-1} E^{\beta}_{\alpha,\alpha\beta}[-(t/\tau)^{\alpha}], \tag{17}$$

and

$$n_{JWS;\,\alpha,\beta}(t) = E^{\beta}_{\alpha,1}[-(t/\tau)^{\alpha}], \qquad \phi_{JWS;\,\alpha,\beta}(t) = \delta(t) - \tau^{-1}(t/\tau)^{-1}E^{\beta}_{\alpha,0}[-(t/\tau)^{\alpha}].$$
(18)

Using the response and relaxation functions for the HN model, we can check the correctness of Equation (3), which immediately flows out from Equation (A4). From Equations (3) and (18), one finds the formula which describes the first derivative of $E_{\nu,1}^{\lambda}(ax^{\nu})$. Observe that, in this case, Equation (A4) cannot be used for the JWS relaxation because it works only if the order of derivative is larger than the value of the second lower parameter in the Mittag–Leffler function $E_{\nu,\mu}^{\lambda}(ax^{\nu})$. Equations (3) and (18) give

$$\frac{\mathrm{d}}{\mathrm{d}x}E_{\nu,1}^{\lambda}(ax^{\nu}) = x^{-1}E_{\nu,0}^{\lambda}(ax^{\nu}) - \delta(x), \qquad x \in \mathbb{R}.$$
(19)

For the EW model, only the relaxation function $n_{EW;\alpha}(t)$ is known. It can be expressed through the binomial Mittag–Leffler function [20]

$$n_{EW;\alpha}(t) = E_{(1,1-\alpha),1}(-t/\tau_1, -\tau_2^{\alpha} t^{1-\alpha}/\tau_1)$$
(20)

or as the series of three parameter Mittag–Leffler functions [24] (Equations (3.71) or (3.73)). These series lead to Equation (20) with the help of Equation (A6). To calculate the response function $\phi_{EW;\alpha}(t)$ we make the inverse Laplace transform of the spectral function Equation (12), which can be easily obtained with the help of Equation (A9). That enables us to write

$$\phi_{EW;\alpha}(t) = \delta(t) - t^{-1} E_{(1,1-\alpha),0}(-t/\tau_1, -\tau_2^{\alpha} t^{1-\alpha}/\tau_1).$$
(21)

Analogically, as in the derivation of Equation (19), we can obtain the first derivative of the binomial Mittag–Leffler function $E_{(\nu_1,\nu_2),1}(ax^{\nu_1}, bx^{\nu_2})$. It reads

$$\frac{\mathrm{d}}{\mathrm{d}x}E_{(\nu_1,\nu_2),1}(ax^{\nu_1},bx^{\nu_2}) = x^{-1}E_{(\nu_1,\nu_2),0}(ax^{\nu_1},bx^{\nu_2}) - \delta(x), \qquad x \in \mathbb{R}.$$
(22)

Equation (22) can also be derived by employing the representation of the binomial Mittag–Leffler function in terms of the series of three parameter Mittag–Leffler functions given by Equation (A6) and, next, separate from these series the zero term, that is, $E_{\nu_1,1}^1(bx^{\nu_2})$ or $E_{\nu_2,1}^1(ax^{\nu_1})$. The calculation goes as follows:

$$\begin{aligned} \frac{\mathrm{d}}{\mathrm{d}x} E_{(\nu_1,\nu_2),1}(ax^{\nu_1},bx^{\nu_2}) &= \frac{\mathrm{d}}{\mathrm{d}x} \sum_{r \ge 0} (ax^{\nu_1})^r E_{\nu_2,\nu_1 r+1}^{1+r}(bx^{\nu_2}) \\ &= \frac{\mathrm{d}}{\mathrm{d}x} E_{\nu_2,1}^1(bx^{\nu_2}) + \sum_{r \ge 1} a^r \frac{\mathrm{d}}{\mathrm{d}x} [x^{\nu_1 r} E_{\nu_2,\nu_1 r+1}^{1+r}(bx^{\nu_2})]. \end{aligned}$$

Now, applying Equations (19) and (A4), we get

$$\frac{\mathrm{d}}{\mathrm{d}x}E_{(\nu_1,\nu_2),1}(ax^{\nu_1},bx^{\nu_2}) = x^{-1}E^1_{\nu_2,0}(bx^{\nu_2}) + x^{-1}\sum_{r\geq 1}(ax^{\nu_2})^r E^{1+r}_{\nu_1,\nu_2r}(bx^{\nu_2}) - \delta(x),$$

The series form of the three-parameter Mittag–Leffler function $E_{\nu,\mu}^{\lambda}(x)$ for $x \in \mathbb{R}$ and the binomial Mittag–Leffler function $E_{(\nu_1,\nu_2),\mu}(x,y)$ for $x, y \in \mathbb{R}$, where their properties are recalled in Appendix B.

3. $\hat{M}(s)$ and $\hat{k}(s)$ and the Laplace (Lévy) Exponents $\Psi(s)$ Related to Them

In this section, we shall show that to determine the Stieltjes character of $\hat{M}(s)$ and $\hat{k}(s)$, as well as to explain the CBF nature of the Laplace (Lévy) exponent $\Psi(s)$, it is enough to demand that Assumption 1 is fulfilled.

Equations (4) and (16) allow one to express the memory M(t) in terms of the spectral function $\hat{\phi}(s)$. From them, we have

$$\hat{M}(s) = B^{-1}\hat{\phi}(s)[1-\hat{\phi}(s)]^{-1},$$
(23)

which is SF.

Proof. The proof of the Stieltjes character of $\hat{M}(s)$ goes as follows. We begin with $\sum_{r=0}^{n} [\hat{\phi}(s)]^{r+1}$, which is SF as a linear combination of SFs. Then, taking the pointwise limit $n \to \infty$, we obtain the series $\sum_{r=0}^{\infty} [\hat{\phi}(s)]^{r+1}$, which tends to $B\hat{M}(s)$ for all $\hat{\phi}(s)$ (we remind that $\hat{\phi}(s)$ is a bounded SF which maximum is $\hat{\phi}(0) = 1$ and which vanishes for $s \to \infty$). Due to property (s1), we end up with the proof. \Box

The algebraic inverse of $\hat{M}(s)$ gives the Laplace (Lévy) exponent $\Psi(s)$:

$$\Psi(s) = B[1 - \hat{\phi}(s)]/\hat{\phi}(s), \tag{24}$$

which is CBF. The complete Bernstein character of $\Psi(s)$ is confirmed by the standard non-Debye relaxation patterns, that is, by the HN, JWS, and EW models. That is illustrated in the Examples 1–3 below.

Example 1. For the HN relaxation, $\Psi_{HN;\alpha,\beta}(s)$ reads $B\{[1 + (\tau s)^{\alpha}]^{\beta} - 1\}$. Thus, $[1 + \Psi_{HN;\alpha,\beta}(s)/B]^{-1}$ gives $[1 + (\tau s)^{\alpha}]^{-\beta}$, which is SF for $\alpha, \beta \in (0, 1]$. The property (s2) and nonnegative B implies that $\Psi_{HN;\alpha,\beta}(s)$ is CBF.

Example 2. In the case of the JWS model, we express the Laplace (Lévy) exponent as $\Psi_{JWS;\alpha,\beta}(s) = [\Psi_{HN;\alpha,\beta}(1/s)]^{-1}$. From the property (cb3), it emerges that it is CBF.

Example 3. The EW Laplace exponent reads $\Psi_{EW;\alpha}(s) = Bs\tau_1/[1 + (\tau_2 s)^{\alpha}]$, and it is CBF. $1 + (\tau_2 s)^{\alpha}$ is CBF, and grace to the property (cb5) $s/[1 + (\tau_2 s)^{\alpha}]$ is also CBF.

The Laplace (Lévy) exponent $\Psi(s)$ can also be employed for calculating $\hat{k}(s)$, which, through Equation (6) is coupled to $\hat{M}(s)$. It reads

$$\hat{k}(s) = \Psi(s)/s = B[1 - \hat{\phi}(s)]/[s\hat{\phi}(s)], \text{ and it is SF.}$$
 (25)

The Stieltjes character of $\hat{k}(s)$ flows out from the fact that $\Psi(s)$ is CBF and the property (cb6). Moreover, from the property (cb5), it occurs that $s/\Psi(s) = \Phi(s)$ is CBF, such that it can be treated as another Laplace (Lévy) exponent.

3.1. Examples of Memories M(t) and k(t)

The examples of $\hat{M}(s)$ and $\hat{k}(s)$ in the *s* domain for the HN, JWS, and EW models are listed in Table 1.

	$\hat{M}(s)$	$\hat{k}(s)$
HN	$B^{-1}\{[1+(\tau s)^{\alpha}]^{\beta}-1\}^{-1}$	$Bs^{-1}[1+(\tau s)^{\alpha}]^{\beta}-s^{-1}$
JWS	$B^{-1}[1+(\tau s)^{-lpha}]^{eta}-1$	$Bs^{-1}\{[1+(\tau s)^{-\alpha}]^{\beta}-1\}^{-1}$
EW	$B^{-1}(\tau_2^{-\alpha}+s^{\alpha})/s$	$B(\tau_2^{-\alpha}+s^{\alpha})^{-1}$

Table 1. $\hat{M}(s)$ and $\hat{k}(s)$ for the HN, JWS, and EW models.

In the time *t* domain, we have Table 2.

Table 2. M(t) and k(t) for the HN, JWS, and EW models.

	M(t)	k(t)
HN	$(Bt)^{-1}\sum_{r\geq 0}(t/\tau)^{\alpha\beta(r+1)}E^{\beta(r+1)}_{\alpha,\alpha\beta(r+1)}[-(t/\tau)^{\alpha}]$	$B(\tau/t)^{\alpha\beta}E_{\alpha,1-\alpha\beta}^{-\beta}[-(t/\tau)^{\alpha}]-B$
JWS	$(Bt)^{-1}E_{\alpha,0}^{-\beta}[-(t/\tau)^{\alpha}] - B^{-1}\delta(t)$	$B\sum_{r\geq 0} E_{\alpha,1}^{\beta(r+1)} \left[-(t/\tau)^{\alpha}\right]$
EW	$B^{-1}\tau_2^{-\alpha} + B^{-1}t^{-\alpha}/\Gamma(1-\alpha)$	$Bt^{\alpha-1}E_{\alpha,\alpha}[-(t/\tau_2)^{\alpha}]$

Relations between functions listed in Tables 1 and 2 are obtained through the Laplace transform, in which we use the complex z instead of s > 0.

4. Equivalence of Equations (4) and (7)

Lemma 6. Equations (4) and (7) are equivalent.

Proof. To show that Lemma 6 is true, we integrate both sides of Equation (4) by $\int_0^T k(T - t) \cdots dt$, where instead of " \cdots " we substitute all terms of Equation (4). In this way, we obtain

$$\int_{0}^{T} k(T-t)\phi(t) \, \mathrm{d}t = B \int_{0}^{T} k(T-t)M(t) \, \mathrm{d}t - B \int_{0}^{T} k(T-t) \left[\int_{0}^{t} M(t-\tau)\phi(\tau) \, \mathrm{d}\tau \right] \mathrm{d}t.$$
(26)

Afterwards, due to the Dirichlet formula, the double-integral $\int_0^T dt \int_0^t d\tau$ can be changed to $\int_0^T d\tau \int_{\tau}^T dt$. Consequently, Equation (26) is expressed as

$$\int_{0}^{T} k(T-t)\phi(t)dt = B \int_{0}^{T} k(T-t)M(t) dt - B \int_{0}^{T} \left[\int_{\tau}^{T} k(T-t)M(t-\tau)\phi(\tau) dt \right] d\tau.$$
(27)

Setting T - t = u, we transform Equation (27) as

$$\int_{0}^{T} k(T-t)\phi(t) dt = B \int_{0}^{T} k(T-t)M(t) dt - B \int_{0}^{T} \left[\int_{0}^{T-\tau} k(u)M(T-\tau-u) du \right] \phi(\tau) d\tau$$

$$= B - B \int_{0}^{T} \phi(\tau) d\tau,$$
(28)

where the passage from the upper to lower formulas goes by using Equation (5) twice. Next, we differentiate it with respect to *T*. That ends the proof. \Box

Example 4. In the case of the HN model, only the memory $k_{HN;\alpha,\beta}(t)$ is known explicitly. Thus, Equation (7) is much simpler to find for the equation governing behavior of the HN response function. The substitution of $k_{HN;\alpha,\beta}(t)$ into Equation (7) leads to [24] (Equation (3.36)), namely,

$$(_{0}D_{t}^{\alpha} + \tau^{-\alpha})^{\beta}\phi(t) = \frac{\mathrm{d}}{\mathrm{d}t}\int_{0}^{t} \left(\frac{t-\xi}{\tau}\right)^{-\alpha\beta} E_{\alpha,1-\alpha\beta}^{-\beta} \left[-\left(\frac{t-\xi}{\tau}\right)^{\alpha}\right]\phi(\xi)\,\mathrm{d}\xi = 0.$$
(29)

The symbol $_{0}D_{t}^{\alpha}$ denotes the fractional derivative in the Riemann–Liouville sense. The right (also called the upper) fractional derivative in the Riemann–Liouville sense $_{0}D_{t}^{\alpha}$ can be defined through the fractional integral $(_{0}I_{t}^{\alpha}f)(t) = (_{0}D_{t}^{-\alpha}f)(t) = [\Gamma(\alpha)]^{-1}\int_{0}^{t}(t - \xi)^{\alpha-1}f(\xi) d\xi$ as follows: $(_{0}D_{t}^{\alpha}f)(t) = d^{n}(_{0}I_{t}^{n-\alpha}f)(t)/dt^{n}$. Furthermore, the fractional derivative in the Caputo sense $(_{0}^{C}D_{t}^{\alpha}f)(t)$ is equal to $_{0}I_{t}^{n-\alpha} d^{n}f(t)/dt^{n}$. Complete information on how to understand the pseudo-operator $(_{0}D_{t}^{\alpha} + \tau^{-\alpha})^{\beta}$ can be found in [24] (Appendix B) and in [25] (Section 5).

Example 5. For the JWS model, the memory $M_{JWS;\alpha,\beta}(t)$ is fully known. It makes possible to get Equation (4), which is relevant for this model. To achieve this goal, we represent $M_{JWS;\alpha,\beta}(t)$ taken from the Table 2 as the fractional derivative in the Riemann–Liouville sense minus a term being the Dirac δ distribution, that is,

$$M_{JWS;\alpha,\beta}(t) = {}_{0}D_{t}^{1-\alpha\beta} \{ t^{-\alpha\beta} E_{\alpha,1-\alpha\beta}^{-\beta} [-(t/\tau)^{\alpha}] \} - \delta(t),$$
(30)

and expressing $M_{JWS;\alpha,\beta}(t)$ with the help of Equation (19) as the first derivative. Then, substituting it into Equation (4), we get

$$\int_{0}^{t} {}_{0}D_{t}^{1-\alpha\beta}\{(t-\xi)^{-\alpha\beta}E_{\alpha,1-\alpha\beta}^{-\beta}[-(t-\xi)^{\alpha}/\tau^{\alpha}]\phi(\xi)\}\,\mathrm{d}\xi = \frac{\mathrm{d}}{\mathrm{d}t}E_{\alpha,1}^{-\beta}[-(t/\tau)^{\alpha}].\tag{31}$$

Integrating both sides of this equation with ${}_{0}D_{t}^{-(1-\alpha\beta)}$ and using the fact that ${}_{0}D_{t}^{-\mu}[({}_{0}D_{t}^{\mu}f)(t)] = f(t) - t^{\mu-1}/\Gamma(\mu) \times [({}_{0}D_{t}^{\mu-1}f)(t)]_{t=0}$ given in [26] (Equation (2.113) on p. 70), we obtain

$$\int_0^t (t-\xi)^{-\alpha\beta} E_{\alpha,1-\alpha\beta}^{-\beta} [-(t-\xi)^{\alpha}/\tau^{\alpha}] \phi(\xi) \,\mathrm{d}\xi = t^{-\alpha\beta} E_{\alpha,1-\alpha\beta}^{-\beta} [-(t/\tau)^{\alpha}] - \frac{t^{-\alpha\beta}}{\Gamma(1-\alpha\beta)}.$$
 (32)

This formula is the same as [24] (Equation (3.36)).

Example 6. In the case of EW relaxation $M_{EW;\alpha}(t)$ and $k_{EW;\alpha}(t)$ are known in compact forms, so Equations (4) and (7) can be found as well. After substituting $M_{EW;\alpha}(t)$ from Table 2, Equation (4) reads

$$\phi_{EW;\alpha}(t) = \tau_2^{-\alpha} \Big[1 - \int_0^t \phi_{EW;\alpha}(\xi) \, \mathrm{d}\xi \Big] + \frac{t^{-\alpha}}{\Gamma(1-\alpha)} + \Big({}_0 D_t^{-(1-\alpha)} \phi_{EW;\alpha} \Big)(t), \tag{33}$$

where ${}_{0}D_{t}^{-(1-\alpha)}$ is a fractional integral given below Equation (29). Using Equation (3), we can find the evolution equation for $n_{EW;\alpha}(t)$. Namely, $1 - \int_{0}^{t} \phi_{EW;\alpha}(\xi) d\xi = n_{EW;\alpha}(t)$ and $({}_{0}D_{t}^{-(1-\alpha)}\phi_{EW;\alpha})(t) = ({}_{0}^{C}D_{t}^{\alpha} n_{EW;\alpha})(t)$. Moreover, $t^{-\alpha}/\Gamma(1-\alpha) + ({}_{0}^{C}D_{t}^{\alpha} n_{EW;\alpha})(t)$ is equal to the fractional derivative of the Riemman–Liouville sense, that is, $({}_{0}D_{t}^{\alpha} n_{EW;\alpha})(t)$. Hence, Equation (33) can be rewritten as

$$-\tau_2^{-\alpha} n_{EW;\alpha}(t) = \left({}_0 D_t^{\alpha} n_{EW;\alpha}\right)(t) + \frac{\mathrm{d}}{\mathrm{d}t} n_{EW;\alpha}(t). \tag{34}$$

Equation (7) for the EW model is obtained by substituting $k_{EW;\alpha}(t)$ into it. That allows one to obtain

$$\frac{\mathrm{d}}{\mathrm{d}t} \int_0^t (t-\xi)^{\alpha-1} E_{\alpha,\alpha} [-(t-\xi)^{\alpha}/\tau^{\alpha}] \phi_{EW,\alpha}(\xi) \,\mathrm{d}\xi = -\phi_{EW,\alpha}(t). \tag{35}$$

5. Conclusions

In this paper, we have studied the most popular models of the non-Debye relaxations, namely the HN, JWS, and EW ones. We have shown that using only one assumption concerning the Stieltjes character of the spectral function guarantees the nonnegativeness

of the response and relaxation functions. Such a conclusion is not surprising because SFs are the subclass of CMFs, and in fact, it can be deduced from the survey paper [24]. Our new results attract readers attentions to the fact that we use SFs instead of CMFs, and connect the Laplace (Lévy) exponent $\Psi(s)$ with the integral kernels M(t) and k(t), basic objects which through the evolution equations, govern the behavior of the response and relaxation functions. This observation is grace to the fact that $\Psi(s)$ is CBF, and hence, $s/\Psi(s)$ is also CBF. That allows us to join these functions with M(t) and k(t). Namely, $M(t) = [\Psi(s)]^{-1}$ and $k(t) = \Psi(s)/s$. Hence, one stochastic process underlying the relaxation can be described in two-fold way—either by M(t) or by k(t). Throughout the paper, we were able to reconstruct the previously known form of M(t) for the HN relaxation model [27,28] and to find M(t) for the JWS and EW models, as well as give k(t) for all investigated models. Relevant kernels are itemized in Table 2, whereas their shapes in the s domain are presented in Table 1. We have also shown that M(t) and k(t) are SFs, so they can be called fading memories. Moreover, M(t) and k(t) satisfy the classical Sonine equation and have integrable singularities at zero, and thus form the Sonine pairs. We provided three examples of them: $(k_{HN}(t), M_{HN}(t)), (k_{JWS}(t), M_{JWS}(t))$, and

 $(k_{EW}(t), M_{EW}(t))$. Each of these pairs lead to two equations: the integral, and the integrodifferential, which are mutually coupled by the Sonine equation for the memories M(t) and k(t).

A byproduct of our considerations is providing explicit expressions for functions belonging to the Mittag–Leffler family, namely for the first derivative of $E_{\nu,1}^{\lambda}(x)$ and $E_{(\nu_1,\nu_2),1}(x)$. We emphasize that these formulae were derived using physical arguments—a relationship between the response and relaxation functions.

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Abbreviations

The following abbreviations are used in this manuscript:

- CMF Completely Monotonic Function
- SF Stieltjes function support by positive semi-axis
- CBF Complete Bernstein Function
- HN Havrillak-Negami
- JWS Jurlewicz-Weron-Stanislavsky
- EW excess wings

Appendix A. Properties of SFs and CBFs

In this Appendix we list the properties of SFs and CBFs which we have extensively used in our construction and proofs.

Appendix A.1. Properties of SFs

- (s1) The set of SFs is closed under pointwise limits: if $(f_n)_{n \in \mathbb{N}}$ is a SF and if the limit $\lim_{n \to \infty} f_n(s) = f(s)$ exist for all s > 0, then f is also a SF, see [15] (Theorem 2.2 (iii))
- (s2) *f* is a CBF if, and only if, $(u + f)^{-1}$ is a SF for every u > 0, see [15] (Theorem 7.5)

Appendix A.2. Properties of CBFs

(cb1)The set CBF is a convex cone: $sf_1 + tf_2 \in CBF$ for all $s, t \gg 0$ and $f_1, f_2 \in CBF$, see [15] (Corollary 7.6 (i)).

(cb2) According to [15] (Corollary 7.9 (i) and (ii)) $CBF \circ SF \subset SF$ and $SF \circ CBF \subset SF$.

 $(cb3) f(s) \in CBF$ if, and only if, $1/f(1/s) \in CBF$, see [15] (Equation (7.1)).

(cb4)[15] (Proposition 7.7) says that if $g \in SF$ is bounded, then $g(0+) - g \in CBF$. Conversely, if the function $f \in CBF$ is bounded, there exist some constant c > 0 and some bounded function $g \in SF$, $\lim_{s \in \infty} g(s) = 0$, such that f = c - g; then c = f(0+) + g(0+).

(cb5)[15] (Proposition 7.1) says that f(s) is in *CBF*, $f \neq 0$, if, and only if s/f(s) is in *CBF*. (cb6)[15] (Theorem 6.2 (ii)) says that if f(s) is a CBF then f(s)/s is a SF.

(cb7) The set of CBFs is closed under pointwise limits, see [15] (Corollary 7.6 (ii)) (cb8) If *f* is a CBF then $(u + f)^{-1}$ is a SF for every u > 0.

Appendix B. Mittag-Leffler Function and Binomial Mittag-Leffler Function

Appendix B.1. Three Parameter Mittag-Leffler Function

The series form of three parameter Mittag-Leffler function $E_{\nu,\mu}^{\lambda}(z)$ [5,23,29] reads

$$E_{\nu,\mu}^{\lambda}(z) = \frac{1}{\Gamma(\lambda)} \sum_{r \ge 0} \frac{\Gamma(\lambda+r)z^r}{r!\Gamma(\mu+\nu r)}$$
(A1)

 $z \in \mathbb{C}$ and for the real argument it is involved in the Prabhakar function $t^{\mu-1}E_{\nu,\mu}^{\lambda}(-at^{\alpha})$ where $t \ge 0$. The Laplace transform of Prabhakar function reads

$$\mathcal{L}[t^{\mu-1}E^{\lambda}_{\nu,\mu}(-at^{\nu});z] = z^{\nu\lambda-\mu}(a+z^{\nu})^{-\lambda} \quad \text{for} \quad \Re(\mu), \Re(z) > 0, \ |z| > |a|^{1/\Re(\nu)}$$
(A2)

[29] whereas the integral representation of Prabhakar function can be found in [23] (Equation (11)) or [10] (Equation (17))

$$t^{\nu\mu-1}E^{\mu}_{\nu,\nu\mu}(-at^{\nu}) = \frac{1}{\Gamma(\mu)}\int_{0}^{\infty} e^{-au} u^{\mu-1}g_{\nu}(u,t) \,\mathrm{d}u.$$
(A3)

For $\Re(\nu)$, $\Re(\lambda) > 0$, $\Re(\mu) > n$ and $n \in \mathbb{N}$ the Formula (11.5) of [30] holds

$$\frac{d^{n}}{dx^{n}}[x^{\mu-1}E^{\lambda}_{\nu,\mu}(ax^{\nu})] = x^{\mu-1-n}E^{\lambda}_{\nu,\mu-n}(ax^{\nu}).$$
(A4)

Appendix B.2. Binomial Mittag-Leffler Function

The binomial Mittag-Leffler function $E_{(\nu_1,\nu_2),\mu}(x_1,x_2)$ [20,31] yields

$$E_{(\nu_1,\nu_2),\mu}(x_1,x_2) = \sum_{k\geq 0} \sum_{\substack{l_1,l_2\geq 0\\l_1+l_2=k}} \frac{k!}{l_1!l_2!} \frac{x_1^{l_1}x_2^{l_2}}{\Gamma(\mu+\nu_1l_1+\nu_2l_2)}$$
(A5)

with x_1 and x_2 being real. It can be expressed as the series of three parameter Mittag-Leffler functions, namely

$$E_{(\nu_1,\nu_2),\mu}(x_1,x_2) = \sum_{r\geq 0} x_1^r E_{\nu_2,\nu_1r+\mu}^{1+r}(x_2)$$

= $\sum_{r\geq 0} x_2^r E_{\nu_1,\nu_2r+\mu}^{1+r}(x_1).$ (A6)

$$E_{(\nu_1,\nu_2),\mu}(x_1,x_2) = \sum_{k\geq 0} \sum_{l_1=0}^k \binom{k}{l_1} \frac{x_1^{l_1} x_2^{k-l_1}}{\Gamma[\mu + \nu_1 l_1 + \nu_2(k-l_1)]}$$

$$= \sum_{l_1\geq 0} \sum_{k\geq l_1} \binom{k}{l_1} \frac{x_1^{l_1} x_2^{k-l_1}}{\Gamma[\mu + \nu_1 l_1 + \nu_2(k-l_1)]}.$$
(A7)

Changing now the summation index $k - l_1$ onto r we have

$$E_{(\nu_1,\nu_2),\mu}(x_1,x_2) = \sum_{l_1 \ge 0} \sum_{r \ge 0} \binom{r+l_1}{l_1} \frac{x_1^{l_1} x_2^r}{\Gamma(\mu+\nu_1 l_1+\nu_2 r)}$$
(A8)

and using the series expression of the three parameter Mittag-Leffler function we can obtain Equations (A6). \Box

Its Laplace transform can be found in [31] and it reads

$$\mathcal{L}[t^{\beta-1}E_{(\alpha_1,\alpha_2),\beta}(-a_1t^{\alpha_1},-a_2t^{\alpha_2});t] = \frac{s^{-\beta}}{1+a_1s^{-\alpha_1}+a_2s^{-\alpha_2}}.$$
(A9)

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