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STOCHASTIC FOUNDATIONS OF THE UNIVERSAL DIELECTRIC RESPONSE

Abstract. We present a probabilistic model of the microscopic scenario of dielectric relaxation. We prove a limit theorem for random sums of a special type that appear in the model. By means of the theorem, we show that the presented approach to relaxation phenomena leads to the well known Havriliak–Negami empirical dielectric response provided the physical quantities in the relaxation scheme have heavy-tailed distributions. The mathematical model, presented here in the context of dielectric relaxation, can be applied in the analysis of dynamical properties of other disordered systems.

1. Introduction. Dielectric relaxation in solids, defined as approaching equilibrium of a dipolar system driven out of equilibrium by a step or alternating external electric field, is one of the most intensively researched topics in experimental and theoretical physics (see e.g. [1, 3–5, 10–15, 17–19, 21, 22, 24, 25, 27–30]. The empirical investigations of dielectric properties of different relaxing systems have shown that despite the variety of materials used and of experimental techniques employed, the time or frequency dependencies of dynamic dielectric characteristics are very similar. It has been observed that for most dielectrics the prevailing form of $i(t)$, the time decay of the depolarization current, exhibits the fractional power laws in time; i.e. for some $0 < p_1, p_2 < 1$,

$$i(t)/t^{-p_1} \to \text{const} > 0 \quad \text{as } t \to 0, \tag{1}$$

$$i(t)/t^{-p_2-1} \to \text{const} > 0 \quad \text{as } t \to \infty. \tag{2}$$

This common property of dielectric responses of relaxing systems is known as the "universal relaxation law" [4, 13, 14].

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Furthermore, the statistical analysis of the data for hundreds of materials have shown [4, 11, 13] that the inverse Fourier transform of $i(t)$,

$$\int_0^\infty e^{-\mathrm{i}\omega t} i(t)\,dt, \tag{3}$$

is the complex function

$$\varphi(\omega) = \frac{B}{(1+(\mathrm{i}A\omega)^\alpha)^\gamma} \tag{4}$$

where $A, B > 0$, $0 < \alpha, \gamma \le 1$. In case $\alpha < 1$ and $\gamma \le 1$ the function (4) corresponds to the so-called Havriliak–Negami (if $\gamma < 1$) or Cole–Cole (if $\gamma = 1$) response, and it yields (1) and (2) with $p_1 = 1 - \alpha\gamma$ and $p_2 = \alpha$. If $\gamma < 1$ and $\alpha = 1$ formula (4) yields the so-called Cole–Davidson response that satisfies the short-time power law (1) only (with $p_1 = 1 - \gamma$). For $\alpha = 1$ and $\gamma = 1$ the formula corresponds to the Debye response characterized by the exponential decay of the depolarization current, $i(t) \propto e^{-t/A}$. Notice that although the Cole–Davidson and Debye responses do not satisfy the universal relaxation law, they are observed for some dielectric materials, and often in relaxation processes other than dielectric relaxation.

The empirically established formula (4) characterizes the observed behaviour of the relaxing system without in any way indicating the physical mechanisms involved. Since the experiment probes the ensemble average in the sense that only the net effect of a large number of contributions from different relaxing entities within a sample is measured, in theoretical attempts to explain the observed relaxation laws it is unanimously assumed that they correspond to a kind of general behaviour which is independent of the details of the systems examined. The relaxation process results from an appropriate configuration of the system, imposed by nonequilibrium constraints at time $t = 0$, and is conditioned by specific interactions of different parts of the system [21, 23, 26, 28]. Both the initial nonequilibrium state of a complex system and the internal interactions have, in general, random characteristics. Hence, the question of the origins of the universal relaxation law has to be addressed in terms of probabilistic models which can provide a clue to a better understanding of the physical mechanism of the relaxation processes. The recent advances in the stochastic theory of relaxation [15, 17–19, 28–30] have provided the technique to formulate both the microscopic scenario of relaxation and the resulting effective representation of the system. The approach based on the general probabilistic formalism of limit theorems enables us to treat relaxation of complex systems regardless of the precise nature of local interactions.

The aim of this paper is to present a limit theorem for random sums of a special type that explains applicability of formula (4) as a fitting function for dielectric data and brings to light the underlying reason for the wide occurrence of the universal relaxation law. The article is structured as follows: In Section 2 we introduce a general idea of probabilistic representations for dielectric responses of relaxing systems, and we derive the representation corresponding to formula (4). In Section 3 we present a model of the microscopic scenario of dielectric relaxation. In Section 4 we prove the limit theorem, and we show by means of it that the model leads, under appropriate general assumptions, to the dielectric responses related to function (4). We thus indicate stochastic origins of the responses observed empirically.

2. Probabilistic representation of dielectric characteristics. A function $\phi(t)$ such that $i(t) \propto -d\phi/dt$ and $\phi(0) = 1$, called a *relaxation function*, is usually considered instead of $i(t)$ in theoretical approaches to model relaxation. In probabilistic terms $\phi(t)$ is interpreted as the survival probability of the nonequilibrium initial state of the relaxing system until time t (see [20]). Introducing $\widetilde{\boldsymbol{\theta}}$, the random waiting time of the entire system for the transition from its initial state, we obtain

$$\phi(t) = \Pr(\widetilde{\boldsymbol{\theta}} \geq t) \tag{5}$$

and, equivalently,

$$\int_0^\infty e^{-\mathrm{i}\omega t} i(t)\, dt \propto \mathrm{E} e^{-\mathrm{i}\omega \widetilde{\boldsymbol{\theta}}}. \tag{6}$$

On the other hand, following the historically oldest approach to relaxation, the relaxation function $\phi(t)$ is commonly assumed to take the form of a weighted average of an exponential decay e^{-bt} with respect to the distribution of relaxation rate b, a physical quantity characterizing the speed of the transition [4]. This means that

$$\phi(t) = \mathcal{L}(\widetilde{\boldsymbol{\beta}}; t) \tag{7}$$

where the random variable $\widetilde{\boldsymbol{\beta}}$ is distributed as the relaxation rate considered. (Here and throughout this paper $\mathcal{L}(X; t) := \mathrm{E} e^{-tX}$ denotes the Laplace transform of a random variable X.)

Summing up, the dielectric response of the relaxing system can be associated with $\widetilde{\boldsymbol{\theta}}$, the waiting time of the system for the transition from the initial state, and $\widetilde{\boldsymbol{\beta}}$, the relaxation rate of the system. The random variables $\widetilde{\boldsymbol{\theta}}$ and $\widetilde{\boldsymbol{\beta}}$ are strictly connected since from (5) and (7) we have $\Pr(\widetilde{\boldsymbol{\theta}} \geq t) = \mathcal{L}(\widetilde{\boldsymbol{\beta}}; t)$. Each of them can unequivocally represent the shape of $i(t)$, as well as of its inverse Fourier transform (3).

We now derive the explicit forms of the waiting time $\widetilde{\boldsymbol{\theta}}$ and the relaxation rate $\widetilde{\boldsymbol{\beta}}$ for the empirically observed dielectric responses corresponding to (4).

THEOREM 1. *For the relaxation function* $\phi(t)$ *related to* (4) *with parameters* $A, B > 0$, $0 < \alpha, \gamma \le 1$ *we have*

$$\phi(t) = \Pr(A\mathcal{G}_\gamma^{1/\alpha}\mathcal{S}_\alpha \ge t) = \mathcal{L}\left(\frac{1}{A}\,\frac{\mathcal{S}'_\alpha}{\mathcal{S}_\alpha}\left(\frac{1}{\mathcal{B}_\gamma}\right)^{1/\alpha};t\right), \tag{8}$$

where

- *for* $\alpha \ne 1$ *the random variables* $\mathcal{S}_\alpha$ *and* $\mathcal{S}'_\alpha$ *are identically distributed according to the completely asymmetric* α*-stable law such that* $\mathcal{L}(\mathcal{S}_\alpha;t) = e^{-t^\alpha}$;
- *for any* $0 < \gamma \le 1$ *the random variable* $\mathcal{G}_\gamma$ *is distributed according to the gamma distribution with scale parameter* 1 *and shape parameter* γ;
- *for* $\gamma \ne 1$ *the random variable* $\mathcal{B}_\gamma$ *is distributed according to the generalized arcsine distribution with parameter* γ, *i.e., the beta distribution with parameters* $p = \gamma$ *and* $q = 1-\gamma$;
- $\mathcal{B}_1 = 1$, $\mathcal{S}_1 = 1$, *and* $\mathcal{S}'_1 = 1$ *with probability* 1;
- *for any* $0 < \alpha, \gamma \le 1$ *the random variable* $\mathcal{G}_\gamma$ *is independent of* $\mathcal{S}_\alpha$;
- *for any* $0 < \alpha, \gamma \le 1$ *the random variables* $\mathcal{B}_\gamma$, $\mathcal{S}_\alpha$, *and* $\mathcal{S}'_\alpha$ *are independent.*

As a consequence, the dielectric response corresponding to (4) *has the following probabilistic representations*:

$$\widetilde{\boldsymbol{\theta}} \stackrel{d}{=} A\mathcal{G}_\gamma^{1/\alpha}\mathcal{S}_\alpha, \tag{9}$$

$$\widetilde{\boldsymbol{\beta}} \stackrel{d}{=} \frac{1}{A}\,\frac{\mathcal{S}'_\alpha}{\mathcal{S}_\alpha}\left(\frac{1}{\mathcal{B}_\gamma}\right)^{1/\alpha}. \tag{10}$$

Proof. For any $0 < \alpha \le 1$,

$$\mathcal{L}(\mathcal{S}_\alpha;t) = \mathcal{L}(\mathcal{S}'_\alpha;t) = e^{-t^\alpha}. \tag{11}$$

For $\widetilde{\boldsymbol{\theta}}$ of the form (9), applying the technique of conditional expected value we obtain

$$\begin{aligned}\mathrm{E}e^{-\mathrm{i}\omega\widetilde{\boldsymbol{\theta}}} &= \mathrm{E}e^{-\mathrm{i}\omega A\mathcal{G}_\gamma^{1/\alpha}\mathcal{S}_\alpha} = \mathrm{E}(\mathrm{E}(e^{-\mathrm{i}A\omega x^{1/\alpha}\mathcal{S}_\alpha})|_{x=\mathcal{G}_\gamma})\\ &= \mathrm{E}e^{-(\mathrm{i}A\omega)^\alpha\mathcal{G}_\gamma} = \frac{1}{(1+(\mathrm{i}A\omega)^\alpha)^\gamma}.\end{aligned}$$

As a consequence, (6) holds for

$$\int_0^\infty e^{-\mathrm{i}\omega t} i(t)\,dt = \varphi(\omega),$$

where $\varphi(\omega)$ is given by (4). This leads to the first equality in (8). To show the second, observe that for any $0 < \gamma \le 1$,

$$\mathcal{L}(1/\mathcal{B}_\gamma;t) = \Pr(\mathcal{G}_\gamma \ge t). \tag{12}$$

Indeed, it is obvious that (12) holds for $\gamma = 1$. In case $\gamma \neq 1$, the density function of $\mathcal{B}_\gamma$ is

$$f_\gamma(x) = \begin{cases} \dfrac{1}{\Gamma(\gamma)\Gamma(1-\gamma)}\, x^{\gamma-1}(1-x)^{-\gamma} & \text{for } 0 < x < 1, \\ 0 & \text{otherwise;} \end{cases}$$

and hence,

$$\begin{aligned}
\mathcal{L}(1/\mathcal{B}_\gamma; t) &= \int_0^1 e^{-t/x} f_\gamma(x)\, dx = [{\scriptstyle y=-1+1/x}] \\
&= \frac{1}{\Gamma(\gamma)\Gamma(1-\gamma)} \int_0^\infty e^{-t(y+1)}(y+1)^{-1} y^{-\gamma}\, dy \\
&= \frac{1}{\Gamma(\gamma)\Gamma(1-\gamma)} \int_0^\infty \Big(\int_1^\infty t e^{-st(y+1)}\, ds \Big) y^{-\gamma}\, dy \\
&\overset{(*)}{=} \frac{1}{\Gamma(\gamma)\Gamma(1-\gamma)} \int_1^\infty \Big(\int_0^\infty t e^{-st(y+1)} y^{-\gamma}\, dy \Big)\, ds = \frac{1}{\Gamma(\gamma)} \int_1^\infty t e^{-st}(st)^{\gamma-1}\, ds \\
&= [{\scriptstyle x=st}] = \frac{1}{\Gamma(\gamma)} \int_t^\infty e^{-x} x^{\gamma-1}\, dx = \Pr(\mathcal{G}_\gamma \geq t),
\end{aligned}$$

where $(*)$ follows from [8, XIV, §3, 521].

Then, applying the technique of conditional expected value, we deduce from (11) and (12) that

$$\begin{aligned}
\mathcal{L}\bigg(\frac{\mathcal{S}'_\alpha}{\mathcal{S}_\alpha} \Big(\frac{1}{\mathcal{B}_\gamma} \Big)^{1/\alpha}; t \bigg) &= \mathrm{E}\bigg(\mathrm{E}\bigg(e^{-t\left(\frac{1}{\mathcal{S}_\alpha}\left(\frac{1}{\mathcal{B}_\gamma}\right)^{1/\alpha}\right)\mathcal{S}'_\alpha} \,\bigg|\, \frac{1}{\mathcal{S}_\alpha} \Big(\frac{1}{\mathcal{B}_\gamma} \Big)^{1/\alpha} \bigg) \bigg) \\
&= \mathrm{E}\Big(\mathrm{E}(e^{-tx\mathcal{S}'_\alpha}) \Big|_{x=\frac{1}{\mathcal{S}_\alpha}\left(\frac{1}{\mathcal{B}_\gamma}\right)^{1/\alpha}} \Big) = \mathrm{E}(e^{-(t/\mathcal{S}_\alpha)^\alpha/\mathcal{B}_\gamma}) \\
&= \mathrm{E}(\mathrm{E}(e^{-(t/x)^\alpha/\mathcal{B}_\gamma})|_{x=\mathcal{S}_\alpha}) = \mathrm{E}(\Pr(\mathcal{G}_\gamma \geq (t/x)^\alpha)|_{x=\mathcal{S}_\alpha}) \\
&= \Pr(\mathcal{G}_\gamma^{1/\alpha} \mathcal{S}_\alpha \geq t),
\end{aligned}$$

which yields the second equality in (8). ■

It is worth noting that it follows from [2, 4] that for the random variable $\widetilde{\boldsymbol{\beta}}$ of the form (10) the density function $g(b)$ is given by the following analytical formula:

- for $0 < \alpha, \gamma < 1$,

$$g(b) = \begin{cases} \dfrac{1}{\pi b} \cdot \dfrac{\sin(\gamma\psi(b))}{((Ab)^{2\alpha} + 2(Ab)^\alpha \cos(\pi\alpha) + 1)^{\gamma/2}} & \text{for } b > 0, \\ 0 & \text{for } b \leq 0, \end{cases}$$

where

$$\psi(b) = \frac{\pi}{2} - \arctan\left(\frac{(Ab)^{-\alpha} + \cos(\pi\alpha)}{\sin(\pi\alpha)}\right);$$

- for $0 < \alpha < 1$, $\gamma = 1$,

$$g(b) = \begin{cases} \dfrac{1}{\pi b} \cdot \dfrac{\sin(\pi\alpha)}{(Ab)^{\alpha} + (Ab)^{-\alpha} + 2\cos(\pi\alpha)} & \text{for } b > 0, \\ 0 & \text{for } b \le 0; \end{cases}$$

- for $\alpha = 1$, $0 < \gamma < 1$,

$$g(b) = \begin{cases} \dfrac{1}{\pi b} \cdot \dfrac{\sin(\pi\gamma)}{(Ab-1)^{\gamma}} & \text{for } b > 1/A, \\ 0 & \text{for } b \le 1/A. \end{cases}$$

The second case ($0 < \alpha < 1$, $\gamma = 1$) is especially interesting. It provides a simple form of the density function for the quotient $\mathcal{S}_\alpha/\mathcal{S}'_\alpha$ of independent and identically distributed positive α-stable random variables while, in general, the density functions of $\mathcal{S}_\alpha$ and $\mathcal{S}'_\alpha$ themselves cannot be given by analytical formulas.

3. Model for the microscopic scenario of dielectric relaxation. To explain the wide applicability of formula (4) in representing dielectric data, the information on the microscopic scenario which relates the local random characteristics of complex systems to the deterministic and universally valid empirical relaxation laws is of great importance. We now present a model for the detailed scheme of relaxation process (proposed recently in [15, 19, 30]) that provides the internal structure of the relaxation rate representing the response of the relaxing system.

In any dielectric complex system capable of responding to an external electric field it is possible that only a part of the total number n of dipoles in the system are able to follow changes of the field [13, 14]. However, even if some dipoles do not directly contribute to the relaxation dynamics, they may affect the stochastic transition of the active dipole. This influence is reflected, for example, in the properties of individual relaxation rates $\beta_{1n}, \beta_{2n}, \ldots$ of the active entities in the system. According to the rate-theory concept [27], the individual relaxation rates are considered here as the contributions of the dipoles to the total relaxation rate representing the response of the system as a whole. They are often assumed to take the form $\beta_{jn} = \beta_j/a_n$ with β_j independent of the system size n and the same normalizing constant a_n for each dipole.

Assume that the jth active dipole interacts with $N_j - 1$ inactive neighbours forming a cluster of size N_j. The number K_n of active dipoles in the system coincides with the number of clusters determined by the local interactions. The latter is equal to the first index k for which the sum $N_1+\ldots+N_k$ of the cluster sizes exceeds n, the size of the system; i.e.

$$K_n = \min\Big\{k : \sum_{j=1}^{k} N_j > n\Big\}.$$

Depending on the screening mechanisms [13], the active dipoles may "see" some of their active neighbours. If so, the cooperative regions built up from the active dipoles may appear. The number L_n of such mesoscopic regions is determined by their sizes $M_1, M_2, \ldots$:

$$L_n = \min\Big\{l : \sum_{m=1}^{l} M_m > K_n\Big\},$$

where M_m is the number of interacting active dipoles in the mth cooperative region. The contribution of each region to the total relaxation rate is the sum of the contributions of all active dipoles over the region. Hence, for the mth region its relaxation rate, say $\overline{\beta}_{mn}$, is equal to

$$\overline{\beta}_{mn} = \sum_{j=M_1+\ldots+M_{m-1}+1}^{M_1+\ldots+M_m} \beta_j/a_n.$$

The probabilistic representation of the dielectric response for the relaxing system as a whole is provided by the total relaxation rate $\widetilde{\boldsymbol{\beta}}_n$, that is, the sum of the contributions over all cooperative regions:

$$\widetilde{\boldsymbol{\beta}}_n = \sum_{m=1}^{L_n} \overline{\beta}_{mn}. \tag{13}$$

In general, the number of dipoles directly engaged in the relaxation process, as well as their locations, are random. Therefore, all the quantities N_j, M_m, β_{jn}, and those defined by them, have to be considered as random variables. Their stochastic characteristics would obviously determine the properties of the total relaxation rate $\widetilde{\boldsymbol{\beta}}_n$ if they were known. But they are not, in general. Nevertheless, as we shall show in the next section, on the basis of limit theorems of probability theory, it is possible to define the distribution of the weak limit $\widetilde{\boldsymbol{\beta}}_\infty = \lim_{n\to\infty} \widetilde{\boldsymbol{\beta}}_n$ even with rather limited knowledge of the distributions of the introduced micro/mesoscopic quantities.

4. Limit theorem for the total relaxation rate. Assume that $M = \{M_m,\, m = 1,2,\ldots\}$, $N = \{N_j,\, j = 1,2,\ldots\}$, and $\beta = \{\beta_j,\, j = 1,2,\ldots\}$ are independent sequences, each consisting of independent and identically distributed positive random variables. (M_m and N_j are integer-valued.) It is easy to show that $\widetilde{\boldsymbol{\beta}}_n$ given by (13) can be rewritten in the form

$$\widetilde{\boldsymbol{\beta}}_n = S_\beta(I_n)/a_n \tag{14}$$

with

$$I_n = S_M(\nu_M(\nu_N(n))),$$

where for $X = \{X_j,\ j = 1, 2, \ldots\}$ being $X = M, N$, or β,

$$S_X(0) = 0, \qquad S_X(k) = \sum_{j=1}^{k} X_j \quad \text{for } k = 1, 2, \ldots,$$

$$\nu_X(n) = \min\{k : S_X(k) > n\}.$$

Observe that $\widetilde{\boldsymbol{\beta}}_n$ in (14) has the form of a normalized random sum with the random index I_n independent of the components β_j. Below we discuss its asymptotic properties (as $n \to \infty$) for the cases connected with formula (4). (More general considerations will be presented in [16].)

Throughout this paper, we will say that the distribution of a positive random variable X_j has a *heavy tail* if for some $c > 0$ and $0 < r < 1$,

$$\lim_{x\to\infty} \frac{\Pr(X_j > x)}{(x/c)^{-r}} = 1.$$

For such a distribution the expected value $\mathrm{E}X_j$ is infinite.

THEOREM 2. *Let $0 < \alpha, \gamma < 1$, $c_1, c_2 > 0$ be fixed.*

(a) *Assume that both N_j and B_j have heavy-tailed distributions with $c = c_1$ and $c = c_2$, respectively, and the same $r = \alpha$.*

- *If the distribution of M_m has a heavy tail with some $c > 0$ and $r = \gamma$, then*

$$\frac{S_\beta(I_n)}{n} \xrightarrow[n\to\infty]{d} \frac{c_2}{c_1} \frac{\mathcal{S}'_\alpha}{\mathcal{S}_\alpha} \left(\frac{1}{\mathcal{B}_\gamma}\right)^{1/\alpha}.$$

- *If $\mathrm{E}M_m < \infty$, then*

$$\frac{S_\beta(I_n)}{n} \xrightarrow[n\to\infty]{d} \frac{c_2}{c_1} \frac{\mathcal{S}'_\alpha}{\mathcal{S}_\alpha} = \frac{c_2}{c_1} \frac{\mathcal{S}'_\alpha}{\mathcal{S}_\alpha} \left(\frac{1}{\mathcal{B}_1}\right)^{1/\alpha}.$$

(b) *Assume that the expected values of both N_j and B_j are finite, and $\mathrm{E}N_j = c_1$, $\mathrm{E}B_j = c_2$.*

- *If the distribution of M_m has a heavy tail with some $c > 0$ and $r = \gamma$, then*

$$\frac{S_\beta(I_n)}{n} \xrightarrow[n\to\infty]{d} \frac{c_2}{c_1} \frac{1}{\mathcal{B}_\gamma} = \frac{c_2}{c_1} \frac{\mathcal{S}'_1}{\mathcal{S}_1} \left(\frac{1}{\mathcal{B}_\gamma}\right)^{1/1}.$$

- *If* $\mathrm{E}M_m < \infty$, *then*

$$\frac{S_\beta(I_n)}{n} \xrightarrow[n\to\infty]{\text{a.s.}} \frac{c_2}{c_1} = \frac{c_2}{c_1}\frac{\mathcal{S}_1'}{\mathcal{S}_1}\left(\frac{1}{\mathcal{B}_1}\right)^{1/1}.$$

The random variables $\mathcal{B}_\gamma$, $\mathcal{S}_\alpha$, *and* $\mathcal{S}_\alpha'$ *are as in Theorem* 1.

Proof. Under the assumptions of part (a), from [7, Theorem XIII.6.2] we obtain

$$\frac{S_N(n)}{n^{1/\alpha}} \xrightarrow[n\to\infty]{d} c_1\,(\Gamma(1-\alpha))^{1/\alpha}\mathcal{S}_\alpha, \tag{15}$$

$$\frac{S_\beta(n)}{n^{1/\alpha}} \xrightarrow[n\to\infty]{d} c_2\,(\Gamma(1-\alpha))^{1/\alpha}\mathcal{S}_\alpha'. \tag{16}$$

Since for any $n, k = 1, 2, \ldots,$

$$\{\nu_N(n) > k\} = \{S_N(k) \le n\}, \tag{17}$$

we have $\nu_N(n) \to \infty$ a.s. as $n \to \infty$. Moreover,

$$\frac{\nu_N(n)}{n^\alpha} \xrightarrow[n\to\infty]{d} \frac{1}{c_1^\alpha\,\Gamma(1-\alpha)}\left(\frac{1}{\mathcal{S}_\alpha}\right)^\alpha. \tag{18}$$

Indeed, from (17), (15) and Cramér's theorem, for any $x > 0$ we have

$$\begin{aligned}\Pr\left(\frac{\nu_N(n)}{n^\alpha} > x\right) &= \Pr(\nu_N(n) > [xn^\alpha]) = \Pr(S_N([xn^\alpha]) \le n)\\ &= \Pr\left(\left(\frac{[xn^\alpha]}{xn^\alpha}\right)^{1/\alpha}\frac{S_N([xn^\alpha])}{[xn^\alpha]^{1/\alpha}} \le \frac{1}{x^{1/\alpha}}\right)\\ &\xrightarrow[n\to\infty]{}\Pr(c_1\,(\Gamma(1-\alpha))^{1/\alpha}\mathcal{S}_\alpha \le 1/x^{1/\alpha}),\end{aligned}$$

which leads to (18).

If the distribution of M_m has a heavy tail, we have

$$\frac{S_M(\nu_M(n))}{n} \xrightarrow[n\to\infty]{d} \frac{1}{\mathcal{B}_\gamma} \tag{19}$$

(see [7, XIV 3]). Since the sequences $\{S_M(\nu_M(n))\}$ and $\{\nu_N(n)\}$ are independent, it follows from [6, Theorem, part (V)] that (18) and (19) yield

$$\frac{I_n}{n^\alpha} \xrightarrow[n\to\infty]{d} \frac{1}{c_1^\alpha\,\Gamma(1-\alpha)}\left(\frac{1}{\mathcal{S}_\alpha}\right)^\alpha\frac{1}{\mathcal{B}_\gamma}. \tag{20}$$

In case $\mathrm{E}M_m < \infty$, from [9, Theorems I.2.3 and II.5.1] we have

$$\frac{S_M(\nu_M(n))}{n} \xrightarrow[n\to\infty]{\text{a.s.}} 1.$$

Hence, from (18), [9, Theorem I.1.1], and Cramér's theorem we get

$$\frac{I_n}{n^\alpha} \xrightarrow[n\to\infty]{d} \frac{1}{c_1^\alpha\,\Gamma(1-\alpha)}\left(\frac{1}{\mathcal{S}_\alpha}\right)^\alpha. \tag{21}$$

From (16), (20), (21), by means of the theorem proved in [6] we obtain the assertions of part (a).

Under the assumptions of part (b) we have

$$\nu_N(n) \xrightarrow[n\to\infty]{\text{a.s.}} \infty, \qquad \frac{\nu_N(n)}{n} \xrightarrow[n\to\infty]{\text{a.s.}} \frac{1}{c_1}$$

(see [9, II 5]), and from Kolmogorov's strong law of large numbers

$$\frac{S_\beta(n)}{n} \xrightarrow[n\to\infty]{\text{a.s.}} c_2;$$

hence, the proof of the statements of this part is parallel to that of (a). ■

Studying relaxation phenomena, one usually deals with systems consisting of a large number of dipoles (of order greater than 10^{23}). Within experimental error one cannot distinguish the observed response of the entire system, represented by unknown $\widetilde{\boldsymbol{\beta}}_n$, from its approximation resulting from the weak limit $\widetilde{\boldsymbol{\beta}}_\infty = \lim_{n\to\infty} \widetilde{\boldsymbol{\beta}}_n$. For the $\widetilde{\boldsymbol{\beta}}_n$ given by (13) with $a_n = n$, under the assumptions of Theorem 2, the limit $\widetilde{\boldsymbol{\beta}}_\infty$ has been shown to have the form (10) with $A = c_1/c_2$, corresponding to the dielectric response related to (4). Hence, in the presented approach to dielectric relaxation, the response related to (4) appears as a result of statistical rules the system follows during its spatio-temporal evolution. This indicates the stochastic reasons for applicability of (4) as a fitting function for dielectric data.

Table 1. Assumptions for which the model leads to particular responses given by (4) with $A, B > 0$, $0 < \alpha, \gamma \le 1$ (c_0 is a positive constant)

Assumptions			Response
β_j	N_j	M_m	
heavy tail with $r = \alpha$ and $c = c_0$	heavy tail with $r = \alpha$ and $c = Ac_0$	heavy tail with $r = \gamma$ and $c > 0$	Havriliak–Negami $\alpha, \gamma < 1$
heavy tail with $r = \alpha$ and $c = c_0$	heavy tail with $r = \alpha$ and $c = Ac_0$	$\mathrm{E}M_i < \infty$	Cole–Cole $\alpha < 1$, $\gamma = 1$
$\mathrm{E}B_i = c_0 < \infty$	$\mathrm{E}N_i = Ac_0 < \infty$	heavy tail with $r = \gamma$ and $c > 0$	Cole–Davidson $\alpha = 1$, $\gamma < 1$
$\mathrm{E}B_i = c_0 < \infty$	$\mathrm{E}N_i = Ac_0 < \infty$	$\mathrm{E}M_i < \infty$	Debye $\alpha = 1$, $\gamma = 1$

Detailed assumptions for which the model leads to the Havriliak–Negami, Cole–Cole, Cole–Davidson, and Debye responses are collected in Table 1.

Let us add that the heavy-tail property has been recognized [15] as directly related to the spatial (if referred to N_j and M_m, the cluster and cooperative-region sizes) or temporal (if referred to the relaxation rate β_j) scaling properties of the system.

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