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Frequency-Independent Rules for the Dielectric Susceptibility Derived from Two Forms of Self-Similar Dynamical Behavior of Dipolar Systems

A. Jurlewicz[1]

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This paper provides the frequency domain analysis of the probabilistic representation of the cluster model for dielectric relaxation in dipolar systems. It is proved that the restriction (0, 1) experimentally found for both the power-law coefficients n and m is the necessary and sufficient condition to obtain the low- and high-frequency power-law behavior. Consequently, in both frequency regions the Kramers–Krönig-compatible frequency-independent rules are fulfilled. Moreover, in contrast to the empirical functions proposed to fit the experimental data, the dielectric susceptibility derived from the stochastic considerations does cover the full range of the observed dielectric responses.

KEY WORDS: Dipolar materials; dielectric susceptibility; asymmetric Lévy-stable distributions; max-stable distributions; power-law dielectric response.

1. INTRODUCTION

Dielectric relaxation phenomena in complex condensed systems have been the subject of experimental and theoretical investigations for many years.[1–17]. This is not only due to the need for an understanding of the electrical properties of various technological materials, but it has also been realized that the basic physics of the dielectric response leads to interesting questions about the theoretical description of physical phenomena in disordered systems. From the empirical studies of dielectric properties of complex condensed materials it became clear that the functions which describe their dynamical behavior deviate considerably from the predictions of the Debye exponential relaxation laws. Several empirical functions

[1] Hugo Steinhaus Center for Stochastic Methods, Technical University of Wrocław, 50-370 Wrocław, Poland.

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have been proposed[1,2] to describe the observed behavior in the frequency domain, for example:

- The Cole–Cole function

$$\chi(\omega) \propto \frac{1}{1+(i\omega/\omega_p)^\nu} \tag{1}$$

- The Cole–Davidson function

$$\chi(\omega) \propto \frac{1}{(1+i\omega/\omega_p)^\mu} \tag{2}$$

- The Havriliak–Negami function

$$\chi(\omega) \propto \frac{1}{[1+(i\omega/\omega_p)^\nu]^\mu} \tag{3}$$

- The Williams–Watts (stretched exponential) function, being the Fourier transform of $\phi(t)$,

$$\phi(t) = \exp[-(\omega_p t)^\nu] \tag{4}$$

The parameter ω_p is the loss peak frequency and the coefficients ν and μ in (1)–(4), assumed to fall in the range (0, 1), have no physical sense.

On the basis of experimental observations it has been found[1] that the dielectric response of most dipolar systems exhibits the following "universal" power-law response:

$$\chi'(\omega) \propto \chi''(\omega) \propto \omega^{n-1} \qquad \text{for} \quad \omega \gg \omega_p \tag{5}$$

and

$$\Delta\chi'(\omega) \propto \chi''(\omega) \propto \omega^m \qquad \text{for} \quad \omega \ll \omega_p \tag{6}$$

where the polarisation decrement $\Delta\chi'(\omega) = \chi'(0) - \chi'(\omega)$ and the power-law coefficients in the range are $0 < n, m < 1$. Note[1,2] that the empirical functions (1) and (3) both have the power-law properties (5) and (6) with the coefficients $n = 1 - \nu$ and $m = \nu$ for the Cole–Cole function (1), and $n = 1 - \nu\mu$ and $m = \nu$ for the Havriliak–Negami function (3). In contrast, the functions (2) and (4) fulfil only the high-frequency power-law relation (5) with the parameter $n = 1 - \mu$ for the Cole–Davidson function (2) and $n = 1 - \nu$ for the Williams–Watts function (4), while in the low-frequency region they satisfy

$$\Delta\chi'(\omega) \propto \omega^2 \qquad \text{and} \qquad \chi''(\omega) \propto \omega$$

It should be stressed that although the empirical functions (1) and (3) satisfy both the power-law relations (5) and (6), they do not cover the full range $0 < n, m < 1$ of the dielectric responses. Namely, it follows from the restriction $0 < \nu, \mu < 1$ that for these functions only the range $0 < n < 1$, $1 - n \leqslant m < 1$ is attainable.

The interpretation of the physical basis of the observed universality (5) and (6) has occupied many workers(.[1-17] Historically the earliest attempts to reconcile the observed nonexponential relaxation with the classical Debye process was the assumption of the distribution of relaxation times[2] leading to a summation of the contributions of individual entities. The relaxation function of a system was expressed as a weighted average of exponential relaxation functions. There is no mathematical objection to use of this formalism, but the most serious objection lies in the observed universality of the dielectric response, since this requires a proof of why the same form of distribution of relaxation times should apply in all the different systems.

Recently there has been introduced a probabilistic representation[18, 20, 21] of the cluster model[3, 10, 12] for dipolar dielectric relaxation which yields the experimentally observed fractional power-law response (5) and (6) in the time domain

$$f(t) \propto \begin{cases} (\omega_p t)^{-n} & \text{for} \quad t \ll 1/\omega_p \\ (\omega_p t)^{-m-1} & \text{for} \quad t \gg 1/\omega_p \end{cases} \tag{7}$$

where $f(t)$ is the response function. The rigorous mathematical approach to the dielectric relaxation, based on a revised definition of relaxation function,[18] explains why the fractional power-law should be so universally applicable. As the necessary and sufficient conditions for it the probabilistic analysis supplies two forms of self-similarity governing the intra- and inter-cluster dynamics in dipolar systems.[21] Moreover, the time domain analysis[21] leads to the empirically observed restriction $(0, 1)$ for the power-law coefficient n, while for the parameter m only the result $m > 0$ can be derived. It is worth noting that although the Williams–Watts response (4) does not fulfil the power-law relation (7), it can be obtained in this approach by neglecting the intercluster influences.[21, 22]

The purpose of the present paper is to discuss the frequency domain consequences of the self-similar laws obtained in the time domain stochastic analysis[21] of the probabilistic representation of the cluster model for relaxing dipolar systems, namely, the frequency-domain power-law relations (5) and (6) observed experimentally and, consequently, the Kramers–Krönig-compatible frequency-independent rules[1]

$$\frac{\chi''(\omega)}{\chi'(\omega)} = \cot\left(n\,\frac{\pi}{2}\right) \qquad \text{for} \quad \omega \gg \omega_p \tag{8}$$

and

$$\frac{\chi''(\omega)}{\Delta\chi'(\omega)} = \tan\left(m\,\frac{\pi}{2}\right) \qquad \text{for} \quad \omega \ll \omega_p \tag{9}$$

are shown to follow from the probabilistic attempt to relaxation phenomena. The restriction (0, 1) for both the power-law coefficients n and m is proved to be the necessary and sufficient condition for it, and hence the theoretical functions derived from the stochastic analysis cover the full range $0 < n$, $m < 1$ of dielectric responses.

The presented frequency domain analysis not only completes the time domain results,[21] but it also suggests how to extend the Havriliak–Negami function (3) to the full range of the observed dielectric responses. It makes it possible to compare directly the theoretical and empirical functions (1)–(3) by means of numerical methods.[23]

2. PROBABILISTIC REPRESENTATION OF THE CLUSTER MODEL FOR DIELECTRIC RELAXATION

The concept contained in the cluster model for dielectric relaxation represents a radical departure from the traditional picture of relaxation.[3, 10, 12] It is based on a realistic picture of the physical nature of the structure of an imperfectly ordered state and its consequences for the dynamics of its constituent species. The cluster structure of a dipolar system may be considered as a natural consequence of the fact that when the electric field is on, only some of the dipoles have enough energy and time to reach a configuration with the dipole momenta aligned along the field lines. Hence, the dielectric response originates with specific, spatially limited regions containing dipoles with positions altered by the external field and their local (random!) environment. During the relaxation process the strongly coupled local (intracluster) motions are expected to be generated first and then followed by the weakly coupled (intercluster) motions which produce the partial long-range structure. Each of these motions, those leading to the local structure order and those leading to the gross cluster array order, has its own characteristic contribution to the observed features.[3, 10, 12]

Unfortunately, the microscopic physical mechanisms governing relaxation in disordered systems are not known yet. It only can be concluded that such a widespread and specific deviation from exponential ideality implies that the fundamental physical principles governing relaxation must have a general form regardless of the detailed physical and chemical nature of the materials in which it is observed. This suggests also the general, based on statistical methods, mathematical description of relaxation processes in dipolar systems.

This point of view was taken into account in searching for the probabilistic representation of the cluster model for dielectric relaxation.[18–23] The proposed probabilistic concept follows from the simple fact that the time up to which an aligned dipole survives in its initial orientation after removing the external electric field is random and depends on random intra- and inter-cluster influences.

In the first approximation, the intercluster interactions are neglected and the exponential relaxation of an individual dipole is conditioned only by the value taken by its relaxation rate β which reflects the random intracluster influence. So, if the relaxation rate of the ith dipole has taken the value b, then the probability that this dipole has not changed its initial aligned position up to the moment t, is

$$\Pr(\theta_i \geqslant t \mid \beta_i = b) = \exp(-bt) \qquad \text{for} \quad t \geqslant 0, \quad b > 0 \tag{10}$$

The random variable β_i denotes the relaxation rate of the ith dipole and the variable θ_i, is the time needed for changing its initial orientation (waiting time); $\beta_1, \beta_2,...$ and $\theta_1, \theta_2,...$ form sequences of nonnegative, independent, identically distributed random variables.

In a system consisting of a large number N of relaxing dipoles, the relaxation function $\phi(t)$ has to express the probability that the whole system has not changed its initial state until the time t. So[18]

$$\phi(t) = \lim_{N \to \infty} \Pr(A_N \min(\theta_1,..., \theta_N) \geqslant t) \tag{11}$$

where A_N is a suitable normalizing constant.

It has been shown recently[21, 22] that the assumption (10) leads to the unique form

$$\phi(t) = \exp[-(At)^\alpha] \tag{12}$$

of the relaxation function (11). Moreover, it follows from the probabilistic analysis[22] that the function (12) has to be interpreted as the Laplace transform of the completely asymmetric Lévy-stable distribution[24] and thus the parameter α has to be in the range (0, 1). The parameter A is a positive constant. Consequently, in the first approximation, i.e., when the intercluster influences are neglected, the relaxation function (11) cannot have any other than the stretched exponential form (4). The main mathematical reason for this is that, independent of a statistical distribution of relaxation rates β_i, the random variable θ_i is finite with probability 1:

$$\Pr(\theta_i \geqslant t \mid \beta_i = b) = \begin{cases} 1 & \text{for} \quad t = 0 \\ 0 & \text{for} \quad t \to \infty \end{cases} \tag{13}$$

A broader class of dielectric responses is then available when the random time θ_i depends on both the intra- and inter-cluster influences in such a way that it is infinite with some nonzero probability. If instead of (10) we postulate[21]

$$\Pr(\theta_{iN} \geqslant t \mid \beta_i = b, a_N^{-1} \max(\eta_1, ..., \eta_{i-1}, \eta_{i+1}, ..., \eta_N) = s) \\ = \exp[-b \min(t, s)] \tag{14}$$

for $b > 0$, $s > 0$, $t \geqslant 0$, then in contrast to (13) we have

$$\Pr(\theta_{iN} \geqslant t \mid \beta_i = b, a_N^{-1} \max(\eta_1, ..., \eta_{i-1}, \eta_{i+1}, ..., \eta_N) = s) \\ = \begin{cases} 1 & \text{for} \quad t = 0 \\ \exp(-bt) & \text{for} \quad t < s \\ \exp(-bs) = \text{const} > 0 & \text{for} \quad t \to \infty \end{cases}$$

i.e., the random variable θ_{iN} can be infinite with some nonzero probability. This is in accordance with the cluster model,[3, 10, 12] in which aggregates require an infinite time for relaxation. The random variable β_i denotes the relaxation rate of the ith dipole and η_i is the time needed for the structural reorganization of the ith cluster (cluster relaxation time); $\beta_1, \beta_2, ...$ and $\eta_1, \eta_2, ...$ form independent sequences of nonnegative, independent, identically distributed random variables. The random variable

$$\eta_{i,N} \equiv a_N^{-1} \max(\eta_1, ..., \eta_{i-1}, \eta_{i+1}, ..., \eta_N)$$

constructed from the sequence $\{\eta_i\}$ has the meaning of a stopping time for exponentially decaying conditional probability (10). The variable θ_{iN} denotes the time needed for changing orientation by the ith dipole (waiting time) in the system consisting of N relaxing dipoles; $\theta_{1N}, ..., \theta_{NN}$ are nonnegative, independent, identically distributed for each N; however, each θ_{iN} depends on the relaxation rate β_i and on the stopping time $\eta_{i,N}$.

It has been shown by time domain analysis[21] that for the whole system, satisfying (14), the relaxation function (11),

$$\phi(t) = \lim_{N \to \infty} \Pr(A_N \min(\theta_{1N}, ..., \theta_{NN}) \geqslant t)$$

takes on a nondegenerate form if:

- The distribution of relaxation rates β_i belongs to the domain of attraction of the completely asymmetric Lévy-stable law,[24] i.e., for some $0 < \alpha < 1$ and any $x > 0$

$$\Pr(\beta_i > xb) = x^{-\alpha} \Pr(\beta_i > b) \qquad \text{for large } b \tag{15}$$

- The distribution of times of structural reorganization of the clusters η_i belongs to the domain of attraction of the max-stable law,[25] i.e., for some $\gamma > 0$ and any $x > 0$

$$\Pr(\eta_i > xs) = x^{-\gamma} \Pr(\eta_i > s) \qquad \text{for large } s \tag{16}$$

The conditions (15) and (16) can be recognized as two forms of self-similar (fractal) dynamical behavior assumed to be a fundamental feature of the power-law dielectric response in the cluster model.[3, 10, 12] The first form is identified with the internal dynamics of the clusters[12] and the second form refs to the way in which the response of the macroscopic system is built up from its cluster components.[12] Other models[7–8, 11, 13] identify only one region of fractal behavior, i.e., adequate to form (15). However, on the basis of experimental observations, it has been argued[3, 10] that the relaxation of dipolar systems involves two different self-similar regimes which are a natural consequence of interwoven cluster groups rather than site dipoles.

The relaxation function $\phi(t)$, Eq. (11), has been shown[21] to fulfil the following relaxation equation (a generalized master equation):

$$\frac{d\phi}{dt}(t) = -\alpha A(At)^{\alpha-1} \left\{1 - \exp\left[-\frac{(At)^{-\gamma}}{k}\right]\right\} \phi(t) \tag{17}$$

where the parameters $0 < \alpha < 1$, $\gamma > 0$, $A > 0$, and $k > 0$ are defined by the Lévy-stable and max-stable laws. When $k \to 0$, Eq. (17) takes the well-known form[8, 10, 13]

$$\frac{d\phi}{dt}(t) = -\alpha A(At)^{\alpha-1} \phi(t)$$

with the solution (12) recognized as the Williams–Watts response (4). In the general case we get the solution of Eq. (17) in the form

$$\phi(t) = \exp[-cS(t)]$$

where $c = k^{-\alpha/\gamma}$ and

$$S(t) = \int_0^{(k^{1/\gamma}At)^\alpha} [1 - \exp(-s^{-\gamma/\alpha})]\, ds$$

A similar form has been obtained from different approaches[8] (the Förster direct-transfer model, the hierarchically constrained dynamics model, and the defect-diffusion model) analyzing nonexponential relaxations, with emphasis on the stretched exponential Williams–Watts form (4). The probabilistic representation of the cluster model for dipolar systems yields also the Williams–Watts relaxation function (4), but as a special case when $k \to 0$. Although each model describes a different mechanism, they have the same underlying reason for the stretched exponential pattern: the existence of scale-invariant relaxation rates.

The response function $f(t)$, $f(t) = -d\phi(t)/dt$, obtained from Eq. (17), has the fractional power-law form (7) only when $\gamma \geqslant \alpha$. The power-law coefficients n and m take the following forms:

$$
\begin{aligned}
n &= 1 - \alpha \\
m &= \begin{cases} \alpha/k & \text{if} \quad \gamma = \alpha \\ \gamma - \alpha & \text{if} \quad \gamma > \alpha \end{cases}
\end{aligned}
\tag{18}
$$

Observe that since $0 < \alpha < 1$ the parameter n is in the range $(0, 1)$. In contrast, for the parameter m only the restriction $m > 0$ can be concluded.

3. POWER-LAW DIELECTRIC RESPONSE IN THE FREQUENCY DOMAIN

The mathematical basis for the treatement of the frequency domain response rests on the Fourier transformation of the response function $f(t)$, which defines the complex frequency-dependent susceptibility $\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$. Hence, in terms of the result (17), the dielectric susceptibility $\chi(\omega)$ has the form

$$
\chi(\omega) = \int_0^\infty \alpha A (At)^{\alpha - 1} \left\{ 1 - \exp\left[-\frac{(At)^{-\gamma}}{k} \right] \right\} \phi(t) \exp(-i\omega t)\, dt \tag{19}
$$

Consequently, in the high-frequency region we have

$$
\begin{aligned}
\omega^\alpha \chi(\omega) &= \int_0^\infty \alpha A (At)^{\alpha - 1} \left[\exp(-it)\right] \left\{ 1 - \exp\left[-\frac{(At)^{-\gamma}}{k} \omega^\gamma \right] \right\} \phi\left(\frac{t}{\omega}\right) dt \\
&\xrightarrow[\omega \to \infty]{} C_1 \int_0^\infty t^{\alpha - 1} \exp(-it)\, dt
\end{aligned}
$$

where C_1 is a positive real constant. Therefore we get

$$
\frac{\chi(\omega)}{\omega^{n-1}} \xrightarrow[\omega \to \infty]{} C_1 \int_0^\infty t^{-n} e^{-it}\, dt
$$

where n is the high-frequency power-law coefficient; see Eq. (18). From the time domain analysis[21] we have $0<n<1$, and so, from the theory of complex functions,[26] there exists the integral

$$\int_0^{\infty} t^{-n} e^{-it}\, dt = C_2 \left[\sin\left(n\frac{\pi}{2}\right) - i\cos\left(n\frac{\pi}{2}\right)\right]$$

where C_2 is a positive real constant. Hence the dielectric susceptibility $\chi(\omega)$ given by Eq. (19) fulfils the high-frequency relation (5) and

$$\lim_{\omega\to\infty} \frac{\chi''(\omega)}{\chi'(\omega)} = \cot\left(n\frac{\pi}{2}\right)$$

The above result, which holds also for the Williams–Watts relaxation function,[22] is in agreement with the experimental rule (8).

In the low-frequency region we have to investigate two distinct cases: $\gamma>\alpha$ and $\gamma=\alpha$. When $\gamma>\alpha$

$$\frac{\chi(0)-\chi(\omega)}{\omega^{\gamma-\alpha}} = \int_0^{\infty} \alpha A/k(At)^{\alpha-\gamma-1}\,[1-\exp(-it)]\,\frac{1-\exp[-\omega^{\gamma}(At)^{-\gamma}/k]}{\omega^{\gamma}(At)^{-\gamma}/k}\,\phi\left(\frac{t}{\omega}\right) dt$$

and

$$\frac{\chi(0)-\chi(\omega)}{\omega^{\gamma-\alpha}} \xrightarrow[\omega\to 0]{} C_3 \int_0^{\infty} t^{\alpha-\gamma-1}(1-e^{-it})\, dt \tag{20}$$

where C_3 is a positive real constant. When $\gamma=\alpha$

$$\frac{\chi(0)-\chi(\omega)}{\omega^{\alpha/k}} = \int_0^{\infty} \alpha A/k(At)^{-\alpha/k-1}\,[1-\exp(-it)]\,\frac{1-\exp[-\omega^{\gamma}(At)^{-\gamma}/k]}{\omega^{\gamma}(At)^{-\gamma}/k} \times\left(\frac{t}{\omega}\right)^{\alpha/k}\phi\left(\frac{t}{\omega}\right) dt$$

and

$$\frac{\chi(0)-\chi(\omega)}{\omega^{\alpha/k}} \xrightarrow[\omega\to 0]{} C_4 \int_0^{\infty} t^{-\alpha/k-1}(1-e^{-it})\, dt \tag{21}$$

where C_4 is a positive real constant. From the above results, Eqs. (20) and (21), we get

$$\frac{\chi(0)-\chi(\omega)}{\omega^m} \xrightarrow[\omega \to 0]{} C \int_0^{\infty} t^{-m-1}(1-e^{-it})\,dt \tag{22}$$

where m is the low-frequency power-law coefficient [see Eq. (18)] and $C = C_3$ or $C = C_4$. Hence the limit (22) depends on the value taken by the integral

$$\int_0^{\infty} t^{-m-1}(1-e^{-it})\,dt$$

which, from the theory of complex functions,[26] exists if and only if $0 < m < 1$, and then equals $C_5[\cos(m(\pi/2)) + i\sin(m(\pi/2))]$; C_5 is a positive real constant. Consequently, when $0 < m < 1$ the low-frequency relation (6) is fulfilled and

$$\lim_{\omega \to 0} \frac{\chi''(\omega)}{\chi'(0)-\chi'(\omega)} = \tan\left(m\frac{\pi}{2}\right)$$

which is in agreement with the low-frequency empirical result (9).

In this paper we have presented the frequency domain analysis of the probabilistic representation of the cluster model for a relaxing dipolar system.[18, 19, 21, 22] The time domain considerations[21] have led to two forms of self-similar dynamical behavior of the system, Eqs. (15) and (16), yielding the time-domain power-law response (7) with the coefficients $0 < n < 1$ and $m > 0$. We have shown that both the restrictions $0 < n < 1$ (obtained earlier) and $0 < m < 1$ are necessary and sufficient for the dielectric susceptibility derived from the stochastic analysis to fulfil the power-law relations (5) and (6), and consequently the Kramers–Krönig-compatible high- and low-frequency rules (8) and (9). Therefore, the theoretically obtained functions cover the full experimentally found range $0 < n, m < 1$ of dielectric responses, while the empirical functions (1)–(4) cannot fall in the range $0 < n < 1$, $0 < m < 1 - n$.

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