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A Relationship between Asymmetric Lévy-Stable Distributions and the Dielectric Susceptibility

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This paper, as a complement to the work of Montroll and Bendler, is concerned with the Lévy-stable distributions and their connection to the dielectric response of dipolar materials in the frequency domain. The necessary and sufficient condition for this connection is found. The presented probabilistic analysis is based on the mathematically correct representation of the meaning of the relaxation function of a system of dipoles and shows why the same form of a distribution of relaxation rates, namely, the completely asymmetric Lévy-stable distribution, should apply in all different relaxing systems. This is in contrast to the traditional definition of the relaxation function, expressed as a weighted average of exponential relaxation functions, which does not explain the universality of the dielectric relaxation law. It also follows from the present considerations that not only is the imaginary part $\chi''(\omega)$ of the dielectric susceptibility directly related to the Lévy-stable distribution (as was found by Montroll and Bendler), but so is the real part $\chi'(\omega)$. As a consequence the relation $\chi''(\omega)/\chi'(\omega) = \cot(n\pi/2)$ for $\omega > \omega_p$ and $0 < n < 1$, implied by experimental results, is obtained.

KEY WORDS: Dipolar materials; dielectric susceptibility; asymmetric Lévy-stable distributions; Williams-Watts dielectric response.

1. INTRODUCTION

From studies carried out in recent years on the dielectric relaxation phenomena in complex dipolar systems, it has become clear that the functions which describe their dynamical behavior considerably deviate from the predictions of the Debye exponential relaxation law.[2,3] It was found that the regression of polarization fluctuations to equilibrium proceeds

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faster than exponentially at times shorter than the relaxation time τ_p and slower than exponentially at times greater than τ_p. Over the last decade the physical basis for this specific deviation from the exponential ideality has been the subject of a great deal of interest and a large number of theoretical models involving different physical mechanisms have been proposed.[4-17] Despite differences in physical details, all the proposed models derive the experimental results for the short-time limit and most of them agree in ascribing the behavior in this range to the progressive involvement of a hierarchy of self-similar dynamic processes.[12,15] At times greater than τ_p agreement either between the models or with the experiment is no longer maintained.

The earliest attempts to reconcile the observed non-Debye relaxation were based on the concept of a system of independent exponentially relaxing species with a statistical distribution of relaxation times (or rates). The most serious objection to the acceptance of this way to obtain a different result than that of the conventional Debye relaxation (characterized by a single relevant relaxation time τ_D) lies in the observed universality of the dielectric response, since this would require a proof of why the same form of a distribution of relaxation times should apply in all different systems. This problems has been solved recently.[18-20] It has been shown that the traditional approach based on the concept of a system of independent exponentially relaxing dipoles with different (independent) relaxation rates uniquely leads to the relaxation function of the Williams–Watts form only. It follows from the probabilistic analysis that the Williams–Watts function has to be interpreted as the Laplace transform of the completely asymmetric Lévy-stable distribution of relaxation rates. The only necessary and sufficient condition for this relationship is the self-similar behavior of the nonnegative relaxation rates.

The purpose of the present paper is to discuss the consequences of the traditional approach in the frequency domain. As we shall see below, both the real and imaginary parts of the complex susceptibility $\chi(\omega)$ are directly related to the Lévy-stable distribution of relaxation rates. Due to this relationship the experimentally observed frequency-independent rule, the so-called "energy criterion,"[21] is straightforwardly fulfilled.

2. DIELECTRIC SUSCEPTIBILITY. EXPERIMENTAL EVIDENCES

The frequency dependence of the dielectric susceptibility from orientational polarization of permanent dipoles has been the subject of experimental and theoretical investigations for many years and still there is no generally accepted theory capable of explaining satisfactorily the totality of the observed results. Experimentally it was found that the complex dielectric susceptibility $\chi(\omega)=\chi'(\omega)-i\chi''(\omega)$ of most dipolar materials exhibited a peak in the loss component $\chi''(\omega)$ at a characteristic frequency ω_p, but showed markedly different high- $(\omega>\omega_p)$ and low-frequency $(\omega<\omega_p)$ dependences than those predicted by the Debye model. Thus it was noted[21] that

$$\chi'(\omega) \propto \chi''(\omega) \propto \omega^{n-1} \qquad \text{for} \quad \omega>\omega_p, \quad 0<n<1 \tag{1}$$

in contrast to the Debye (exponential relaxation) result

$$\chi'(\omega) \propto \omega^{-2} \qquad \text{and} \qquad \chi''(\omega) \propto \omega^{-1} \qquad \text{for} \quad \omega>\omega_p$$

It has been pointed out[21] that the experimental result of Eq. (1) implies that

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

The experimental result[5]

$$\chi'(0)-\chi'(\omega) \propto \chi''(\omega) \propto \omega^{m} \qquad \text{for} \quad \omega<\omega_p, \quad 0<m<1 \tag{3}$$

was also significantly different from that predicted by Debye, namely

$$\chi'(0)-\chi'(\omega) \propto \omega^{2} \qquad \text{and} \qquad \chi''(\omega) \propto \omega \qquad \text{for} \quad \omega<\omega_p$$

The experimental behavior of (3) leads to a similar frequency-independent rule[22] as that of (2):

$$\frac{\chi''(\omega)}{\chi'(0)-\chi'(\omega)}=\tan\left(m\frac{\pi}{2}\right) \qquad \text{for} \quad \omega<\omega_p \tag{4}$$

The relations (2) and (4) underline the differences in the nature of the high- and low-frequency polarization processes. Equation (2) shows that when a system is driven by an AC field the energy recoverable per cycle remains a constant fraction of the work done by the field independent of frequency in the frequency range $\omega<\omega_p$. When a system is driven by an AC field in the frequency range $\omega<\omega_p$, then it follows from Eq. (4) that the energy lost per cycle has a constant relationship to the extra energy that can be stored by a static field. However, it does not seem to have been realized that this must be the rule that defines the nature of the relaxation processes.

As yet no microscopic model has been based directly on the experimental rules, Eqs. (2) and (4), instead most of them have concentrated on the derivation of empirical functions which have been inputted to fit the experimental data for $\chi(\omega)$ [or for the dielectric response function $f(t)$ in the time domain]. The exception is the cluster model,[5,15] which derived entirely new expressions from a consideration of the way in which the energy contained in a fluctuation is distributed over a system of interacting clusters. Although the result is in agreement with the empirical laws, (1) and (3), this model does not convince us of its general applicability.

It is always possible to reproduce the experimental results by means of a choice of a suitable relaxation function $\phi(t)$, since

$$\chi(\omega)=\int_0^\infty e^{-i\omega t}f(t)\,dt \tag{5}$$

where $f(t)=-d\phi(t)/dt$, but such an approach does not explain the universality of the empirical laws. It has been suggested[6-13] that the Williams–Watts relaxation function

$$\phi(t)=\exp[-(\omega_p t)^\alpha], \qquad 0<\alpha<1, \quad \alpha=1-n \tag{6}$$

can mimic a wide variety of behavior because of the slow change in the frequency dependence of its Fourier transform in the region of $\omega<\omega_p$. On the basis of experimental observations it has been argued[15] that this agreement is more apparent than real, and when the frequency range is large enough the value of n determined for $\omega>\omega_p$ is insufficient to define the whole relaxation process. Similarly, measurements of a response function $f(t)$ made in the time domain are often not extended far enough beyond ω_p^{-1} to distinguish between the Williams–Watts function and other alternative expressions. It has also been concluded that, in general, the observed behavior is that of (1) and (3) with exponents $m\neq 1-n$. When the complete frequency dependence can be measured at a constant temperature, the deduced values of n and m are normally found not to change for the portions of the same response obtained at different temperatures.[5] Even when this wide range of measurement is not possible the deduced value of m is usually observed to remain constant over a given temperature range, except for cryogenic temperatures. Therefore, suggestions that deviations at low frequency ($\omega<\omega_p$) are due to a temperature-dependent change in n cannot be accepted and the empirical result can be taken as a true description of the situation.

3. DIELECTRIC SUSCEPTIBILITY. PROBABILISTIC REPRESENTATION

The traditional explanation for non-Debye relaxation has been to assign a local value to the relaxation time τ for each dipole, and hence recover the observed regression of polarization fluctuations by means of the relaxation function, expressed as a weighted average of exponential relaxation functions[1,23]

$$\phi(t) = \int_0^\infty w(\tau) \exp\left(-\frac{t}{\tau}\right) d\tau \tag{7}$$

This attempt has the advantage of retaining the stochastic features of Debye's original concept of independently exponentially relaxing dipoles in a viscous medium that acts as a random noise source. However, the choice of distribution $\omega(\tau)$ is not arbitrary, but defined by the empirical relaxation functions, and such an approach does not explain the universality of dielectric responses. Namely, it can only be concluded that a given empirically observed relaxation law is compatible with a particular distribution of relaxation times (or rates), but it does not explain why the power law should be so universally applicable.

Below we present a probabilistic analysis of the idea of the distribution of relaxation times. This idea, while very natural on physical grounds, based on the definition (7) used for 80 years, does not explain the universality of dielectric responses. The present analysis is based on the mathematically correct representation of the meaning of the relaxation function of a system of dipoles.[18] In contrast, the traditional definition (7) does not, in general, represent the relaxation function of a system. Our ultimate aim is (1) to prove that the same form of distribution of relaxation rates will apply in all different systems, and also (2) to show the uniqueness of the form of dielectric susceptibility expressed by Eq. (5).

In the traditional approach the exponential relaxation of an individual dipole is conditioned only by the value taken by its relaxation rate β[18-20] ($\beta = 1/\tau$). 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

$$\Pr(\theta_i \geq t \mid \beta_i = b) = \exp(-bt) \qquad \text{for} \quad t \geq 0, \quad b > 0 \tag{8}$$

The random variable β_i denotes the relaxation rate of the ith dipole and the variable θ_i, the time needed for changing its initial orientation; $\beta_1, \beta_2, \ldots$ and $\theta_1, \theta_2, \ldots$ form sequences of nonnegative, independent, identically distributed random variables with distribution functions F_β and F_θ, respectively.

It follows from Eq. (8) that the total probability that the ith dipole has not changed its initial aligned position up to the moment t equals[19]

$$\Pr(\theta_i \geq t) = \int_0^\infty \exp(-bt)\, dF_\beta(b) \tag{9}$$

The right-hand side of Eq. (9) is a weighted average of exponential relaxations with respect to the distribution F_β of relaxation rates [similarly to the traditional Eq. (7)] and is just the Laplace transform $\mathscr{L}(F_\beta; t)$ of the distribution function F_β. The left-hand side of Eq. (9) has the meaning of the relaxation function $\phi_i(t)$ of an individual dipole taken from the set of independent exponentially relaxing dipoles with different relaxation rates β_i. Therefore

$$\phi_i(t) = \mathscr{L}(F_\beta; t) \tag{10}$$

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]

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

where A_N is a suitable normalizing constant.

According to Eq. (5), the complex susceptibility can be expressed by means of the relaxation function $\phi(t)$ as follows:

$$\chi(\omega) = \int_0^\infty e^{-i\omega t} \left(-\frac{d\phi}{dt}(t) \right) dt$$

Therefore, using Eq. (11), we have

$$\chi(\omega) = \lim_{N \to \infty} \int_0^\infty \cdots \int_0^\infty e^{-i\omega A_N \min(t_1, ..., t_N)} \, dF_\theta(t_1) \cdots dF_\theta(t_N) \tag{12}$$

The above integral, by means of the Fubini theorem,[24] the property of the conditional probability,[25] and Eq. (8), can be expressed as

$$\begin{aligned} &\int_0^\infty \cdots \int_0^\infty e^{-i\omega A_N \min(t_1, ..., t_N)} \, dF_\theta(t_1) \cdots dF_\theta(t_N) \\ &= \int_0^\infty dF_\beta(b_1) \cdots \int_0^\infty dF_\beta(b_N) \int_0^\infty \cdots \int_0^\infty e^{-i\omega A_N \min(t_1, ..., t_N)} \\ &\quad \times \prod_{k=1}^N b_k e^{-b_k t_k} \, dt_1 \cdots dt_N \\ &= \int_0^\infty dF_\beta(b_1) \cdots \int_0^\infty dF_\beta(b_N) \sum_{j=1}^N I_{jN}(b_1, ..., b_N) \end{aligned} \tag{13}$$

where

$$I_{jN}(b_1,..., b_N) = \int_0^\infty dt_j \int_{t_j}^\infty \cdots \int_{t_j}^\infty e^{-i\omega A_N t_j}$$
$$\times \prod_{k=1}^{N} b_k e^{-b_k t_k} dt_1 \cdots dt_{j-1} dt_{j+1} \cdots dt_N$$

After integrating with respect to the variables t_k, $k \neq j$, we obtain that the integral I_{jN} equals

$$I_{jN}(b_1,..., b_N) = -\int_0^\infty e^{-i\omega A_N t_j} \frac{d}{dt_j}(e^{-b_j t_j}) \prod_{k=1, k\neq j}^{N} e^{-b_k t_j} dt_j.$$

and hence

$$\sum_{j=1}^{N} I_{jN}(b_1,..., b_N) = -\int_0^\infty e^{-i\omega A_N s} \frac{d}{ds}\left(\prod_{k=1}^{N} e^{-b_k s}\right) ds \tag{14}$$

Now, integrating by parts the integral in Eq. (14) and substituting $t = A_N s$, we get

$$\sum_{j=1}^{N} I_{jN}(b_1,..., b_N) = 1 - i\omega \int_0^\infty e^{-i\omega t}\left(\prod_{k=1}^{N} e^{-b_k t/A_N}\right) dt \tag{15}$$

From Eqs. (13) and (15), the susceptibility function $\chi(\omega)$ defined by Eq. (12) equals

$$\chi(\omega) = \lim_{N\to\infty} \left\{1 - i\omega \int_0^\infty e^{-i\omega t}\left[\mathscr{L}\left(F_\beta; \frac{t}{A_N}\right)\right]^N dt\right\} \tag{16}$$

and after integrating by parts, Eq. (16) obtains the form

$$\chi(\omega) = \lim_{N\to\infty} \left\{-\int_0^\infty e^{-i\omega t} \frac{d}{dt}\left[\mathscr{L}\left(F_\beta; \frac{t}{A_N}\right)\right]^N dt\right\} \tag{17}$$

The limit in Eq. (17) exists if and only if $[\mathscr{L}(F_\beta; t/A_N)]^N$ tends to some limit. Moreover, it follows from the theory of the Lévy-stable laws[26] that the only possible nondegenerate form of the limit of $[\mathscr{L}(F_\beta; t/A_N)]^N$ is the Laplace transform of the completely asymmetric Lévy-stable distribution function F supported on the nonnegative half-line, with an index of stability α, $0 < \alpha < 1$, and $A_N = N^{1/\alpha}$, i.e.,

$$\lim_{N\to\infty} \left[\mathscr{L}\left(F_\beta; \frac{t}{N^{1/\alpha}}\right)\right]^N = \mathscr{L}(F; t) \tag{18}$$

Consequently, from Eq. (17),

$$\chi(\omega) = -\int_0^\infty e^{-i\omega t} \frac{d}{dt} \mathscr{L}(F; t)\, dt \tag{19}$$

or, from Eq. (16),

$$\chi(\omega) = 1 - i\omega \int_0^\infty e^{-i\omega t} \mathscr{L}(F; t)\, dt \tag{20}$$

Equation (20) shows the explicit relationship between the Lévy-stable distributions and the dielectric response in the frequency domain.

It has been shown[20] that the limiting Laplace transform in Eq. (18) is just the relaxation function $\phi(t)$ given by Eq. (11) and therefore

$$\phi(t) = \int_0^\infty w_{st}(b) \exp(-bt)\, db \tag{21}$$

where $w_{st}(b)$ denotes the completely asymmetric Lévy-stable probability density function. Comparing the result (21) with Eqs. (9) and (10), we get the representation of a system consisting of a large number of relaxing dipoles by one "averaged dipole" with the completely asymmetric Lévy-stable distribution of its relaxation rate. Concluding, a system of $N \to \infty$ independent exponentially relaxing dipoles with different relaxation rates β_i can be represented by the averaged dipole with relaxation rate[18] $\tilde{\beta} = \lim_{N \to \infty} \sum_{i=1}^{N} \beta_i / A_N$ the distribution of which is a completely asymmetric Lévy-stable one. Hence, only in this case does the relaxation function of a whole system satisfy Eq. (7), but with $w(\tau) = b^2 w_{st}(b)$, $b = 1/\tau$.

It is worth to note that it is not necessary to know the detailed nature of F_β to obtain the limiting form (18). In fact, this is determined only by the behavior of the tail of $F_\beta(b)$ for large b, and so a good deal may be said about the asymptotic properties based on rather limited knowledge of the properties of F_β. It can be shown[26] that the necessary and sufficient condition for the distribution function $F_\beta(b) = 1 - \Pr(\beta_i > b)$ to have the limit in Eq. (18) can be expressed by the following scaling law:

$$\text{for any} \quad x > 0 \qquad \Pr(\beta_i > xb) = x^{-\alpha} \Pr(\beta_i > b) \quad \text{for large } b \tag{22}$$

In other words, the self-similar property (22) in taking by a relaxation rate the value greater than b and the value greater than xb is the necessary and sufficient condition for the distribution of β_i to have the limit in Eq. (18). It has been suggested[15,19,20] that self-similarity (fractal behavior) is a fundamental feature of relaxation in real materials. This result, obtained here by means of purely probabilistic techniques independent of physical details, is in agreement with models[12] identifying this region of fractal behavior.

4. FREQUENCY DOMAIN WILLIAMS–WATTS RESPONSE

From the probability theory[26] the limiting Laplace transform in Eq. (18) has the following form:

$$\mathscr{L}(F; t) = \exp[-(At)^\alpha] \tag{23}$$

and so the dielectric response, Eq. (19), in the frequency domain equals

$$\chi(\omega) = \int_0^\infty e^{-i\omega t}\left(-\frac{d}{dt}e^{-(At)^\alpha}\right)dt \tag{24}$$

where $0 < \alpha < 1$ and A is a positive constant. Hence, the complex susceptibility in Eq. (20) corresponds to the Williams–Watts form (6) of the relaxation function. It is also possible to obtain the Debye response taking $\alpha \to 1$. However, from the mathematical point of view this is fulfilled in the case of a degenerate limiting distribution function F, i.e., in the case when the random relaxation rate $\tilde{\beta}$ of the averaged dipole representing a relaxing system can take only one value.

Equations (20) and (23) give us

$$\chi'(\omega) = 1 - \omega \int_0^\infty \mathscr{L}(F; t)\sin(\omega t)\,dt = 1 - \omega \int_0^\infty e^{-(At)^\alpha}\sin(\omega t)\,dt \tag{25}$$

and

$$\chi''(\omega) = \omega \int_0^\infty \mathscr{L}(F; t)\cos(\omega t)\,dt = \omega \int_0^\infty e^{-(At)^\alpha}\cos(\omega t)\,dt \tag{26}$$

So, it has been proved that not only is the imaginary part $\chi''(\omega)$ of the dielectric susceptibility directly related to the Lévy-stable laws (as was found by Montroll and Bendler[11]), but so is the real part $\chi'(\omega)$. The difference in the result obtained here and in the mentioned paper is a consequence of the following fact: The Fourier transform of the symmetric Lévy-stable distribution {for which the parameter α can be taken from the range $(0, 2]$} and the Laplace transform of the completely asymmetric Lévy-stable law [for which α has to be in the range $(0, 1)$] are of the same form, $\exp[-(At)^\alpha]$, for $t \geqslant 0$. Therefore, for $0 < \alpha < 1$ in the theory of the Lévy-stable distributions any function of such a form can be interpreted in two different ways. If the function $\exp[-(At)^\alpha]$ is taken as the Fourier transform (as in Montroll and Bendler[11]), then it is connected with a random variable which takes both positive and negative values in the range $(-\infty, \infty)$. But if this function is the Laplace transform, then the random variable connected with it is nonnegative.

The most natural way to describe the relaxing dipolar systems is to assume the randomness of, obviously, nonnegative relaxation times. Hence, from the physical point of view the proper case is the second one, i.e., when the connection between the Lévy-stable distribution and the dielectric susceptibility is given by the Laplace transform, Eqs. (25) and (26).

Now, let us examine the high- and low-frequency behavior of the dielectric susceptibility given by Eq. (24). As we will see below, $\chi(\omega)$ is proportional to $\omega^{-\alpha}$ for large ω and, consequently, $\chi''(\omega)/\chi'(\omega)$ tends to a positive constant as $\omega \to \infty$. In contrast, for $\omega \to 0$, $\chi''(\omega)/[\chi'(0)-\chi'(\omega)]$ tends to infinity and $[\chi(0)-\chi(\omega)]$ cannot have any "power-law" property for small ω.

Substituting $s=\omega t$ in Eq. (24), we get

$$\frac{\chi(\omega)}{\omega^{-\alpha}}=\alpha A \int_0^\infty (As)^{\alpha-1} e^{-is} e^{-(As/\omega)^\alpha} ds$$

Since $\exp[-(At/\omega)^\alpha]$, as a function of ω, monotonically increases to 1 as $\omega \to \infty$, we obtain

$$\lim_{\omega \to \infty} \frac{\chi(\omega)}{\omega^{-\alpha}}=\alpha A^\alpha \int_0^\infty t^{\alpha-1} e^{-it}\, dt \tag{27}$$

From the theory of complex functions we have[27]

$$\int_0^\infty t^{\alpha-1} e^{-it}\, dt=(i)^{-\alpha}\,\Gamma(\alpha)=[\cos(\alpha\pi/2)-i\sin(\alpha\pi/2)]\,\Gamma(\alpha) \tag{28}$$

where

$$\Gamma(\alpha)=\int_0^\infty t^{\alpha-1} e^{-t}\, dt$$

From Eqs. (27) and (28) it follows that

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

where $n=1-\alpha$, which is in agreement with the experimental results [Eq. (2)].

For small ω it is easy to see that

$$\lim_{\omega \to 0} \frac{\chi''(\omega)}{\omega} = \frac{\Gamma(1 + 1/\alpha)}{A}$$

and

$$\lim_{\omega \to 0} \frac{\chi'(0) - \chi'(\omega)}{\omega^2} = \frac{\Gamma(1 + 2/\alpha)}{2A^2}$$

Therefore, we have

$$\lim_{\omega \to 0} \frac{\chi''(\omega)}{\chi'(0) - \chi'(\omega)} = \infty$$

which is contrary to the experimental results [Eq. (4)].

In this paper we have discussed the mathematical foundation of the generalized concept of the distribution of relaxation times (rates) introduced in the time domain analysis of dielectric relaxations[18-20] and its consequences in the frequency domain. Here we have restricted our considerations to a system of independent exponentially relaxing dipoles with different relaxation rates β_i determined by local environments. We have based our analysis on a new definition of the relaxation function[18] $\phi(t)$, Eq. (11), which provides the mathematically correct representation of the meaning of the relaxation function of a system. By contrast, the definition (7), used since Wagner (1913)[23] in terms of the distribution of relaxation times $w(\tau)$, which has served to determine $\omega(\tau)$ from the empirical function $\phi(t)$, does not, in general, represent the relaxation function of a system.

Equation (7) is equivalent to (11) only in the case discussed in this paper and then, if and only if the scaling law (22) is fulfilled does the relaxation function take the only possible form (6). The scaling law (22) determines the behavior of the tail of the unknown distribution function F_β of relaxation rates and guarantees the existence of the nondegenerate limiting distribution F and hence the existence of the macroscopic relaxation function (11).

So, we have shown that the generalized concept of the distribution of relaxation times (rates) in the case of a system of independent exponentially relaxing dipoles with different (independent) relaxation rates β_i uniquely leads to the relaxation function of the Williams–Watts form (6). It follows from the probabilistic analysis that the Williams–Watts function has to be interpreted as the Laplace transform of the complete asymmetric Lévy-stable distribution and thus the parameter α has to be in the range (0, 1). The only necessary and sufficient condition for this relationship is the self-similar behavior of the nonnegative relaxation rates [Eq. (22)].

Several theories based on different physical mechanisms have been also successful in deriving the stretched exponential decay law (6). Three of these theories, the direct-transfer model,[4] the hierarchically constrained dynamics model,[6] and the defect-diffusion model,[7] have been shown[12] to have an underlying common mathematical structure. The existence of scale-invariant relaxation rates generated in each model was found to be the unifying feature of the theories and the sufficient condition to obtain the nonexponential relaxation function.

Here we have demonstrated that the mathematical framework leading in a natural way to a stretched exponential relaxation function is based on the representation of a relaxing dipolar system by independent exponential relaxations, which seems to be hidden in the models[4,7] mentioned above. In this case the system can be represented by one "averaged dipole" (as was the starting point in the defect-diffusion model[7]) and the necessary and sufficient condition for this is the existence of the scale-invariant relaxation rates, Eq. (22). Moreover, we have found that the distribution of the relaxation rate of the averaged dipole is the completely asymmetric Lévy-stable one and hence is scale invariant.

In the case discussed in this paper it has also been proved that both the real and imaginary parts of the complex susceptibility $\chi(\omega)$ are directly related to the Lévy-stable distributions [Eqs. (25) and (26)]. As a consequence, only the experimentally observed high-frequency result [Eq. (2)] is fulfilled.

The main reason the relaxation function (11) cannot have any other than the stretched exponential form (6) is that the random variable θ_i is finite with probability 1. In order to obtain a broader class of dielectric responses one should define a random variable θ_i infinite with some nonzero probability.[18-20] Such a modification expresses the well-known fact that the individual dipoles and their environments do not remain independent during the process of relaxation[5] and leads directly to a class of double power laws, which includes the Williams–Watts and the Debye responses as special cases.[20] It leads also to both the frequency-independent rules, Eqs. (2) and (4), experimentally observed for the dielectric response in the frequency domain.

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