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# A probabilistic mechanism hidden behind the universal power law for dielectric relaxation: general relaxation equation

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Abstract. A new model for the explanation of the universal dielectric response law is proposed. It differs significantly from all previous work on the subject. The presented model is based on the assumption that individual dipoles and their environment do not remain independent during the process of relaxation. Therefore the time  $\theta$ , needed for the regression to equilibrium of polarization fluctuations is a random variable that depends for each relaxing dipole on two other random variables: the waiting time  $\eta_i$  and the dissipation rate  $\beta_i$ . As a consequence we derive a general relaxation equation that gives the universal dielectric response in the power-law form (as a special case, the stretched exponential form is obtained).

#### 1. Introduction

The dielectric properties of materials have been the subject of experimental and theoretical investigations for many years. 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 materials.

Not long after Debye had formulated, in 1912, his theory of dipole relaxation in liquids [1], it was realized that his prediction of an exponential decay for polarization fluctuations regressing to equilibrium was not obeyed by most of the systems investigated [2]. The traditional explanation for this behaviour has been to assign a local value to the relaxation time for each dipole, and hence recover the observed regression by suitable distribution [3]. Typically, experimental data are described by empirical functions [4] whose parameters are related to the distribution appropriate to the material investigated. This attempt has the advantage of retaining the stochastic features of Debye's original concept of independently relaxing dipoles in a viscous medium that acts as a random noise source.

Conventional Debye relaxation

$$\varphi(t) = \varphi_0 \exp(-t/\tau_0) \tag{1}$$

is characterized by a single relevant relaxation time  $\tau_0$ . The simplest way to obtain a different result for  $\varphi(t)$  is to postulate a statistical distribution of relaxation times  $\tau$  across

different atoms, clusters or degrees of freedom. Then, with the assumption of additive contributions to the relaxing quantity, it is natural to write

$$\varphi(t) = \int_0^\infty w(\tau) \exp(-t/\tau) \,\mathrm{d}\tau. \tag{2}$$

This approach is microscopically arbitrary and it is also associated with a picture of parallel relaxation, in which each degree of freedom relaxes independently with characteristic time  $\tau_{i}$  [5–15]. Contrary to models that were based on a parallel addition of relaxation contributions, the model presented in [16] proposes a serial summation of a hierarchy of relaxations extending over the same spatial range. The authors pointed out that a group of dipoles must adopt a specific configuration before a subset can relax, which then releases the constraints preventing a further subset from relaxing, and so on. However, although it has been realized in many approaches that the individual dipoles and their environment do not remain independent during the regression of fluctuation, as yet no microscopic model has been based directly on this conclusion. The exception is the cluster model [17-22], which derived entirely new expressions from a consideration of the way in which the energy contained in fluctuation is distributed over a system of interacting clusters. This is also the only theory in which the results obtained are in agreement with empirical functions input to fit the experimental data for  $\varphi(t)$  in the short-time limit  $t \leq \omega_p^{-1}$  and the long-time limit  $t \geq \omega_p^{-1}$ , where  $\omega_p$  is the loss-peak frequency.

However, most proposed probabilistic models derive the experimental results for the short-time limit and agree in inputting the behaviour in this range to the progressive involvement of a hierarchy of self-similar dynamic processes; at times greater than  $\omega_p^{-1}$  agreement either between the models or with the experiment is no longer maintained.

The purpose of this paper is to present a new probabilistic model for dielectric relaxation. The main features of this approach are as follows:

(i) The universal dielectric response law can be derived in a simple analytical form from a general relaxation equation, postulated earlier in a special case in [23].

(ii) The response function agrees with empirical functions input to fit the experimental data in both short- and long-time limits.

(iii) In contrast to the cluster model of Dissado and Hill [17-22] our approach suggests a relation between the experimental parameters m and n defining, respectively, the lowand high-frequency branches of the complex dielectric susceptibility.

## 2. The experimental evidence

A growing number of dielectric relaxation data show that the classical Debye behaviour is hardly ever observed experimentally [24–31]. It has been found [26] that the asymptotic frequency dependence of the dielectric susceptibility

$$\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$$

follows a common universal pattern for virtually all kinds of materials. Namely, the behaviour

$$\chi''(\omega) \simeq \chi'(\omega) \simeq (\omega/\omega_p)^{n-1}$$
 for  $\omega \ge \omega_p$  (3)

and

$$\chi''(\omega) \simeq \chi'(0) - \chi'(\omega) \simeq (\omega/\omega_p)^m \quad \text{for } \omega \ll \omega_p \quad (4)$$

is observed over many decades of frequency. The parameters 0 < n < 1 and 0 < m < 1

(illustrating the shape of the dielectric response) together with the static polarization  $\chi'(0)$  and  $\omega_p$ , the loss-peak frequency, can give a complete description of the relaxing system. The frequency dependence given by equation (3) implies that the real and imaginary components of the complex susceptibility obey at high frequencies the following relation [32]:

$$\chi''(\omega)/\chi'(\omega) = \cot(n\pi/2) \tag{5}$$

which suggests that  $\chi(\omega)$  has a constant phase independent of frequency. The experimental behaviour of equation (4) leads to a similar frequency-independent rule [33]. Corresponding to the high-frequency relation (5) the following ratio for the low-frequency polarization decrement can be written

$$\chi''(\omega) / [\chi'(0) - \chi'(\omega)] = \tan(m\pi/2).$$
(6)

The relations (5) and (6) underline the differences in the nature of the low- and highfrequency polarization processes. Equation (5) shows that when the 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 the system is driven by an AC field in the frequency range  $\omega < \omega_p$ , then it follows from (6) that the energy lost per cycle has a constant relationship to the extra energy that can be stored by a static field.

The corresponding time dependence of the depolarization current can be given by means of the response function f(t). The response function [20] describes the regression of a natural fluctuation in the property  $\mathcal{P}$ , following its delta function generation  $E(t') = E_0 \delta(t')$  at zero time

$$\Delta \mathcal{P}(t) = \int_{-\infty}^{t} f(t - t') E(t') \, \mathrm{d}t'.$$
<sup>(7)</sup>

The most common method of determining the response function for a chosen property is by measurement of the frequency-dependent susceptibility  $\chi(\omega)$  given by

$$\chi(\omega) = \int_0^\infty e^{-i\omega t} f(t) dt = -\int_0^\infty e^{-i\omega t} \frac{d\varphi(t)}{dt} dt.$$
 (8)

Hence, the early phase of the regression obeys the following time power-law decay

$$f(t) \simeq (\omega_{\rm p} t)^{-n} \qquad t \leqslant \omega_{\rm p}^{-1} \tag{9}$$

and by (5) is governed by a constant relationship, dependent upon n, which defines the partitioning of the original information of the fluctuation into recoverable and irrecoverable portions. The final stage of the regression to equilibrium is governed by a constant relationship between the energy (information) dissipated and that remaining to be dissipated, equation (6), and the fluctuation regresses asymptotically to equilibrium having the form

$$f(t) \simeq (\omega_{\rm p} t)^{-m-1} \qquad t \ge \omega_{\rm p}^{-1}. \tag{10}$$

It has been suggested [8-16] that the Williams-Watts function

$$f(t) \simeq t^{-n} \exp[-(\omega_{\rm p} t)^{1-n}]$$
(11)

can mimic a wide variety of behaviour because of the slow change in the frequency dependence of its Fourier transform in the region of  $\omega < \omega_p$ . However, this agreement

is more apparent than real, and when the frequency range is large enough the value of *n* determined for  $\omega \ge \omega_p$  is insufficient to define the whole process [21, 22]. Similarly measurements of f(t) made in the time domain are often not extended far enough beyond  $\omega_p^{-1}$  to distinguish between the Williams-Watts function and other alternative expressions.

In general the observed behaviour is that of equation (10) with exponent  $m \neq 1 - n$ . 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 [18, 20]. 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 ( $\omega \ll \omega_p$ ) 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. The probabilistic model

It is evident that the universal relation has the property (5), which implies a constant phase angle mentioned above, but which may also be stated in terms of energy stored  $E_s$  and energy lost  $E_t$  per radian

$$E_1/E_s = \cot(n\pi/2). \tag{12}$$

This property is a direct consequence of the power-law relation and it does not in any way depend on any particular physical model; if the power law (3) applies, then equation (12) follows inevitably whatever the mechanism. Equation (12) refers to macroscopic polarization. It has been proposed [34] that the argument could be reversed by stating that if any system displayed the property that the energy lost per reversal of every microscopic polarization was independent of the rate of reversal, then the macroscopic property (12) would likewise be satisfied and the relaxation of the polarization would have to follow a fractional power law.

This approach was first adapted by Jonscher [26, 35, 36] to hopping electronic systems under the name of 'screened hopping' and was subsequently extended to dipolar situations both below and above  $\omega_p$  [37]. The model relies on the evident notion that in a typical solid the microscopic dipolar, electronic or ionic transitions are very rapid (on the scale of picoseconds or less) and they take place in a system in which electrostatic or strain interactions are invariably present but the adjustment of local equilibrium takes a much longer time to be effected. The consequence of this is a constant energy loss per reversal of microscopic polarization, which leads to the universal relation.

Interactions may be of several types, i.e. electrostatic, mechanical, quantum-mechanical or chemical, and their general consequence is to make the movements of the individual relaxing entities more difficult as the strength of the interactions increases. Moreover, the interactions determine the way in which the relaxation process takes place. Generally, in systems under study, this influence will affect an individual relaxing entity in a random way. The moment of the practically instantaneous transition for the *i*th hopping movement will be random as also will be the screening adjustment time connected with the *i*th abrupt jump. The moment of a transition of the *i*th dipole is fully described by the waiting time,  $\eta_i$ , whereas the screening adjustment time is described by the dissipation rate  $\beta_i = 1/\tau_i$ . Thus the experimental time  $\theta_i$  (i.e. the time needed for





the regression to equilibrium of polarization fluctuations) of the *i*th dipole, as the sum of the waiting time and the screening adjustment time, is stochastically dependent on  $\eta_i$  and  $\beta_i$  (figure 1). For a system consisting of a large number of relaxing dipoles we have to determine the probability that the whole system stays in initially given states up to the experimentally observed time t, with constraints in the form of random waiting time  $\eta_i$  and random dissipation rate  $\beta_i$  for each dipole.

Following the above discussion we introduce formally all the needed quantities:

(i) Let  $\{\eta_i\}$  be a sequence of independent, identically distributed, non-negative random variables with common distribution function

$$F_n(s) = P(\eta_i < s). \tag{13}$$

 $P(\eta_i < s)$  denotes the probability that the waiting time  $\eta_i$  for the *i*th dipole is less than *s*. For simplicity we denote

$$R(s) = 1 - F_n(s) = P(\eta_i \ge s) \tag{14}$$

where R(s) is a continuous decreasing function with properties: R(0) = 1 and  $\lim_{s\to\infty} R(s) = 0$ . Observe that R(s) denotes the probability that the waiting time for the *i*th dipole is not less than *s*. The function R(s), in probability theory, is just known as the reliability function [38].

(ii) Let  $\{\beta_i\}$  be a sequence of independent, identically distributed, non-negative random variables with common distribution function

$$F_{\beta}(b) = P(\beta_i < b). \tag{15}$$

 $P(\beta_i < b)$  denotes the probability that the dissipation rate  $\beta_i$  for the *i*th dipole is less than b. Without loss of generality one can assume that there exists a sequence of positive constants  $\{a_n\}$  such that the *n*th power of the Laplace transform of the distribution function  $F_\beta$  converges to the limit transform G, as  $n \to \infty$ :

$$[\mathscr{L}(F_{\beta};t/a_{n})]^{n} \xrightarrow[n \to \infty]{} G(t).$$
(16)

For convenience, we subsequently use in this paper the following notation:

$$\mathscr{L}(F_{\beta}; t/a_n) = \int_0^\infty \exp(-bt/a_n) \,\mathrm{d}F_{\beta}(b)$$

where the above integral is the Stieltjes integral of  $\exp(-bt/a_n)$  with respect to the

distribution function  $F_{\beta}$ . This notation should not be taken for the traditional meaning of the Laplace transform

$$L(f,t) = \int_0^\infty \exp(-xt)f(x) \,\mathrm{d}x$$

where the last integral is the Lebesque integral of exp(-xt)f(x) with respect to dx.

From the physical point of view such an assumption means the existence of a macroscopic limit dissipation rate for the whole system consisting of *n* relaxing dipoles. The macroscopic dissipation rate is a normalized sum of independent rates  $\beta_i$ , i.e.

$$\sum_{i=1}^n (\beta_i/a_n).$$

(iii) Let  $\{\theta_i\}$  be a sequence of independent, identically distributed, non-negative random variables described by the conditional probability

$$P(\theta_i \ge t | \beta_i = b, \eta_i = s) = \exp[-(b/a_n)\min(t, s)].$$
(17)

Equation (17) means the probability of the event that the *i*th dipole stays in its initial state up to the moment *t*, given the events  $\beta_i = b$  and  $\eta_i = s$ . According to the stochastic features of Debye's original concept of independently relaxing dipoles in a viscous medium, we use the conventional exponential formula. Let us also stress that formula (17) expresses the fact that the experimental time  $\theta_i$  is stochastically dependent on the waiting time  $\eta_i$  and the dissipation rate  $\beta_i$ .

#### 4. General relaxation equation

From the law of total probability [39], which is one of the most fundamental formulae of probability analysis, we have

$$P(\theta_i \ge t | \beta_i = b) = \int_0^\infty P(\theta_i \ge t | \beta_i = \eta_i = s) \, \mathrm{d}F_\eta(s) \tag{18}$$

where  $F_{\eta}(s)$  is the waiting time distribution function. Now substituting expression (17) and  $dF_{\eta}(s) = -R'(s) ds$ , cf (14), we get

$$\frac{\mathrm{d}}{\mathrm{d}t}P(\theta_i \ge t | \beta_i = b) = -\frac{\mathrm{d}}{\mathrm{d}t} \int_0^\infty \exp[-(b/a_n)\min(t,s)]R'(s) \,\mathrm{d}s. \tag{19}$$

Now splitting the interval of the integration  $[0, +\infty)$  into two sub-intervals [0, t] and  $[t, +\infty)$ , and using the fact that the function min(t, s) is equal to s on the first sub-interval and t on the second, we find that the above integral is

$$-\frac{d}{dt}\int_{0}^{t}\exp\left(-\frac{b}{a_{n}}s\right)R'(s)\,ds - \frac{d}{dt}\int_{t}^{\infty}\exp\left(-\frac{b}{a_{n}}t\right)R'(s)\,ds$$
$$= -\exp\left(-\frac{b}{a_{n}}t\right)R'(t) - \frac{d}{dt}\exp\left(-\frac{b}{a_{n}}t\right)\int_{t}^{\infty}R'(s)\,ds$$
$$= -\exp\left(-\frac{b}{a_{n}}t\right)R'(t) - \frac{d}{dt}\exp\left(-\frac{b}{a_{n}}t\right)[R(\infty) - R(t)]$$

Dielectric relaxation equation

$$= -\exp\left(-\frac{b}{a_n}t\right)R'(t) + \frac{d}{dt}\exp\left(-\frac{b}{a_n}t\right)R(t).$$

Finally we have

$$\frac{\mathrm{d}}{\mathrm{d}t}P(\theta_i \ge t|\beta_i = b) = R(t)\frac{\mathrm{d}}{\mathrm{d}t}\exp\left(-\frac{b}{a_n}t\right). \tag{20}$$

Applying once again the law of total probability to equation (20), we get

$$\frac{\mathrm{d}}{\mathrm{d}t}P(\theta_i \ge t) = R(t) \int_0^\infty \frac{\mathrm{d}}{\mathrm{d}t} \exp\left(-\frac{b}{a_n}t\right) \mathrm{d}F_\beta(b).$$

As the above integral is uniformly convergent with respect to t, by the Leibnitz rule we can thus interchange the order of differentiation and integration in the above formula. Hence

$$\frac{\mathrm{d}}{\mathrm{d}t}P(\theta_i \ge t) = R(t)\frac{\mathrm{d}}{\mathrm{d}t}\int_0^\infty \exp\left(-\frac{b}{a_n}t\right)\mathrm{d}F_\beta(b) = R(t)\frac{\mathrm{d}}{\mathrm{d}t}\mathcal{L}(F_\beta;t/a_n).$$
(21)

 $\mathscr{L}(F_{\beta}; t/a_n)$  denotes here the Laplace transform of the distribution function  $F_{\beta}$  in the sense of the Stieltjes integral. Since

$$\frac{\mathrm{d}}{\mathrm{d}t}[P(\theta_i \ge t)]^n = n[P(\theta_i \ge t)]^{n-1} \frac{\mathrm{d}}{\mathrm{d}t}[P(\theta_i \ge t)]$$
(22)

then introducing expression (21) with the derivative

$$\frac{\mathrm{d}}{\mathrm{d}t}\mathscr{L}(F_{\beta};t/a_n)$$

evaluated from a formula similar to (22), we have

$$\frac{\mathrm{d}}{\mathrm{d}t}[P(\theta_i \ge t)^n = [P(\theta_i \ge t)]^{n-1}R(t)[\mathscr{L}(F_\beta; t/a_n)]^{-n+1} \frac{\mathrm{d}}{\mathrm{d}t}[\mathscr{L}(F_\beta; t/a_n)]^n.$$
(23)

Now taking the limit as  $n \rightarrow \infty$ , and using the assumption (16), we obtain

$$\frac{\mathrm{d}}{\mathrm{d}t}\lim_{n\to\infty} \left[P(\theta_i \ge t)\right]^n = \lim_{n\to\infty} \left[P(\theta_i \ge t)\right]^{n-1} R(t) \frac{\mathrm{d}}{\mathrm{d}t} \log G(t).$$
(24)

Observe that we can write

$$\left\{\min_{1\leq i\leq n}\theta_i\geq t\right\}=\bigcap_{1\leq i\leq n}(\theta_i\geq t).$$

Using the assumption that  $\theta_i$  are independent, identically distributed we have

$$\lim_{n \to \infty} P\left\{\min_{1 \le i \le n} \theta_i \ge t\right\} = \lim_{n \to \infty} P\left\{\bigcap_{1 \le i \le n} (\theta_i \ge t)\right\} = \lim_{n \to \infty} [P(\theta_i \ge t)]^n.$$
(25)

The left-hand side of formula (25), which is the probability that the whole system,

consisting of a large number n of relaxing dipoles, stays in its initial state up to the experimentally observed time t, defines the relaxation function  $\varphi(t)$ :

$$\varphi(t) = \lim_{n \to \infty} P\left\{\min_{1 \le i \le n} \theta_i \ge t\right\}.$$
(26)

Thus, substituting equation (26) into (24), we obtain a general relaxation equation

$$\frac{\mathrm{d}}{\mathrm{d}t}\varphi(t) = \varphi(t)R(t)\frac{\mathrm{d}}{\mathrm{d}t}\log G(t). \tag{27}$$

Recall that G(t) is the Laplace transform of a distribution function F, which is just the distribution function of a limit random variable to which the sequence of normalized sums  $\Sigma \beta_i/a_n$  converges in law. So, in other words,  $F_\beta$  is attracted to F. The set of all distribution functions that are attracted to F is called the domain of attraction of the distribution F. It is well known in probability theory [40] that only Lévy-stable distributions have domains of attraction, so the Laplace transform of the non-negative Lévy-stable distribution can be written, cf [14], as

$$G(t) = \exp(-t^{\alpha}) \qquad 0 < \alpha < 1.$$
(28)

Thus the relaxation equation has the following form

$$\frac{\mathrm{d}}{\mathrm{d}t}\varphi(t) = -\alpha t^{\alpha-1}R(t)\varphi(t) \tag{29}$$

and consequently the relaxation function is given by

$$\varphi(t) = \varphi_0 \exp\left(-\alpha \int_0^{\omega_p t} R(s) s^{\alpha - 1} \, \mathrm{d}s\right) \tag{30}$$

where  $\omega_{\rm p}$  is the loss-peak frequency.

#### 5. The universal dielectric response

In order to obtain the response function

$$f(t) = -\frac{\mathrm{d}}{\mathrm{d}t}\varphi(t) \tag{31}$$

in an explicit form one has to choose the function R(s) defined by formula (14). From the mathematical point of view it is just a decreasing reliability function. From the physical point of view, for disordered dipolar systems it is obvious that the intensity function

$$\lambda(s) = - dR(s)/ds \tag{32}$$

cannot be constant. So for the non-negative random variable we can take the reliability function given, for example, by the Weibull distribution [38], namely,

$$R(s) = \exp(-ks^{\delta}) \tag{33}$$

where k is a positive real number, and  $0 < \delta < 1$ . From the analysis of experimental data [22] it follows that in the short-time limit both the power law as well as the Williams-

Watts function can be used. Hence we have to take  $\delta = \alpha$  and k equal to any small number. The reliability function (33) can be written now in the following form:

$$R(s) \simeq 1/(1+ks^{\alpha})$$
  $k > 0$   $0 < \alpha < 1.$  (34)

Substituting (34) into equation (30) we obtain the following relaxation function:

$$\varphi(t) = \varphi_0 [1 + k(\omega_p t)^{\alpha}]^{-1/k}.$$
(35)

Observe that

$$\lim_{k \to 0} \varphi(t) \to \exp[-(\omega_{\rm p} t)^{\alpha}] \tag{36}$$

i.e. the relaxation function (35) tends to the stretched exponential form, predicted by a number of probabilistic models [5–16]. The stretched exponential form (36) of the relaxation function is always obtained when only one probabilistic mechanism is considered: long-tail distribution of waiting times or relaxation times.

Note also that the relaxation function (35) satisfies the following equation

$$\frac{\mathrm{d}}{\mathrm{d}t}\varphi(t) = -\alpha\omega_{\mathrm{p}}(\omega_{\mathrm{p}}t)^{\alpha-1}[\varphi(t)]^{k+1}$$
(37)

which agrees with the equation introduced to describe high-order chemical reactions [41], when k is a natural number, and has the form of a unimolecular fractal equation of motion [11, 13, 42, 43], with a solution in the form (36) when k = 0.

It is easy to show that the response function, obtained from the derived relaxation function (35),

$$f(t) = \varphi_0 \alpha \omega_p (\omega_p t)^{\alpha - 1} [1 + k(\omega_p t)^{\alpha}]^{-(1+k)/k}$$
(38)

has the short-time limit

$$f(t) \simeq (\omega_{\rm p} t)^{\alpha - 1} = (\omega_{\rm p} t)^{-n} \tag{39}$$

where  $n = 1 - \alpha$ , and obviously 0 < n < 1; and the long-time limit

$$f(t) \simeq (\omega_{\rm p} t)^{-(\alpha+k)/k} = (\omega_{\rm p} t)^{-m-1}$$
(40)

where  $m = \alpha/k$ , and 0 < m < 1 if  $\alpha < k$ .

Thus the derived formula (38), for the response function, agrees with experimental results in both limits:  $t \ll \omega_p^{-1}$  and  $t \gg \omega_p^{-1}$ . But, in contrast to common opinion that the parameters *m* and *n* are not related [44, 45], we get the following relationship

$$m = (1 - n)/k \tag{41}$$

where k > 1 - n and 0 < n < 1. We can differentiate the following two cases.

(i) If  $1 - n \le k \le 1$ , then  $1 - n \le m \le 1$  (as is observed in most analysed experimental data [45]): for k = 1, m = 1 - n (this case is recognized as the Cole-Cole response; for k = 1 - n, m = 1 (this case is recognized as the Cole-Davidson response).

(ii) If k > 1, then 0 < m < 1 - n (as is observed only in a small number of analysed data [45]).



Figure 2. The time dependence of the response functions for a dipolar system with an exponential (e) and universal peak characteristics for  $\alpha = (1 - n) = 0.65$  (other curves). The behaviour of universal response functions as  $k \rightarrow 0$  is shown.

Figure 2 shows the difference between the classical Debye (1) and the universal timedomain response (38). In the case when  $k \rightarrow 0$  the Williams–Watts response (11) is obtained.

Since k is connected with the waiting time distribution for the relaxing dipoles, cf equation (33), it may express the strength of interactions. The interactions are considered as primarily constraints for dipolar motions [16] (one dipole cannot move until another one moves out of the way). The parameter k, introduced in equation (34), plays an important role in determining the probability  $R(s) = P(\eta_i \ge s)$  that the waiting time for the *i*th dipole is not less than s. Namely, the greater the parameter k, the smaller is the probability R(s). It can be shown that the function R(s) corresponding to waiting times  $\eta_i$  will always have the approximate form (34). This is a consequence of the extreme value theorem, cf [46]. For a more physical discussion of the significance of the parameter k, we refer to [47].

#### 6. Concluding remarks

We have shown that it is possible to describe the universal dielectric response by means of a probabilistic method. The model presented is based on the observation that the time  $\theta_i$  needed for the regression to equilibrium of polarization fluctuations is a random variable for each relaxing dipole. Since individual dipoles are dependent on their environment during the process of relaxation,  $\theta_i$  depends stochastically on the waiting time  $\eta_i$  and on the dissipation rate  $\beta_i$ .

Using one of the basic formulae of probability analysis, i.e. the law of total probability, we derived the new relaxation equation for the probability  $\varphi(t)$  of finding the whole relaxing system in its initial state up to the experimentally observed time t. Next, we have shown that taking a particular waiting time distribution function, the solution  $\varphi(t)$  can be obtained in a simple analytical form

$$\varphi(t) = \varphi(t, 1-n, k)$$
  $0 < 1-n < 1$   $k > 0$ 

which, in the case when the distribution of waiting times is neglected  $(k \rightarrow 0)$ , tends to the stretched exponential relaxation function. The stretched exponential form, as shown in figure 2, is of no use for describing the relaxation process in real systems.

The response function f(t) obtained in our model agrees in the short- as well as the long-time limit with the empirically postulated power law. Moreover, the relationship between the parameters m and n has been derived: m = m(k, n). This relation underlines the influence of the waiting time distribution on the dissipation rate distribution in the long-time limit only.

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