

## RELAXATION FUNCTIONS IN DIPOLAR MATERIALS

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**Summary:** We compare two simple “cartoons” of relaxation processes in dipolar materials: the “first passage” relaxation function introduced by K. Weron (1991) and the “average” relaxation function expressing the proportion of dipoles which did not change their imposed aligned orientation up to a certain time, the latter providing a description closer to what is experimentally measured. In some cases, the two relaxation functions are proved to coincide.

**KEY WORDS:** Dielectric relaxation function; Kohlrausch–Williams–Watts response; one–sided Lévy stable distribution; ordered statistics.

## 1. Introduction.

The surprising empirical observation that the relaxation decay law for many diverse dipolar systems follows the same stretched exponential pattern

$$\phi(t) = \exp[(-t/\tau)^\alpha],$$

where the parameters  $0 < \alpha < 1$  and  $\tau > 0$  depend on the material and can be a function of external variables such as temperature, already has focussed much theoretical attention.<sup>(1)</sup> Several derivations of this ubiquitous decay law, involving various concepts such as percolation, fractals, hierarchical relaxation of constraints, multipolar interaction transitions or defect-diffusion processes, are known. Klafter and Shlesinger<sup>(2)</sup> analysed the underlying common factors in the case of three different physical models: the Förster direct transfer model, the hierarchically constrained dynamics model, and the defect diffusion model, and found that a common feature responsible for generating the stretched exponential law is the existence of a scale-invariant distribution of relaxation times. The same explanation (a power law distribution) was found for the spin-flip dynamics<sup>(3)</sup> on fractals and percolative structures.

The widely investigated defect-diffusion model<sup>(4)</sup> in the context of dielectric relaxation is based on the concept of migrating defects (e.g. vacancies, local conformational fluctuations, grain boundaries) triggering relaxation of dipoles. Relaxation from an aligned to a random configuration occurs instantaneously when the nearest defect reaches the dipole for the first time. A natural in this case question: *What is the probability that the dipole will first be reached at time  $t$  by one of the diffusing defects ?* can be substituted by a related

question: *What is the probability that the system of dipoles has not changed up to time  $t$  its initial aligned orientation imposed by an external electric field ?*

The latter point of view was adopted in recent probabilistic studies of non-exponential relaxation,<sup>(5-8)</sup> where a related notion of the relaxation function in dipolar materials was introduced. The papers demonstrated that, once one accepts such a definition, the class of possible relaxation functions is essentially restricted to those represented by stretched exponentials or, in another terminology, to Kohlrausch-Williams-Watts (KWW) relaxation responses. The reasoning was based on an observation that switching time  $\theta$  of a single dipole is a random variate, finite with probability 1, and the whole scheme depends on the assumption that the probability  $P(\theta > t)$  of a switching time  $\theta$  of single dipole exceeding  $t$ , is of a certain shape (exponential in the above mentioned papers) common for all dipoles. The concept of such a common distribution, if it is to have a physical meaning, has to be associated with a sort of ergodic type behavior, so that

$$P(\theta > t) = \langle I(\theta > t) \rangle = \lim_{N \rightarrow \infty} \frac{I(\theta_1 > t) + \dots + I(\theta_N > t)}{N}$$

where  $I(\theta_i > t)$  indicates the event that the individual  $i$ -th dipole switched after time  $t$ .

To show the physical significance of the exponent  $\alpha$ , several workers have modeled the system as an interacting particle system on a  $d$ -dimensional lattice, with interaction reflecting a cooperative nature of the phenomenon, and the relaxation function representing a transient behavior of the system in its approach to equilibrium. There is a large physical literature<sup>(3,9-11)</sup> where complex models depending on the underlying spatial (lattice or continuous) structure are provided to explain non-exponential decay laws. In mathematical literature, the results on the transient behavior of interacting particle systems are sparse,

and certainly we could find nothing that would explain an appearance of KWW responses. The work<sup>(12)</sup> that perhaps comes closest, contains deep results of Donsker and Varadhan concerning  $d$ -dimensional volume functional of the so called Wiener (or  $p$ -stable) sausage. Its result might be reinterpreted to give the KWW response with exponent  $d/(d + p)$ . Despite their depth, all these results do not account for the full range of exponents in the experimentally encountered responses<sup>(13,14)</sup>.

So we take a different tack in this paper, and try to provide some insight into the situation, by comparing two simple “cartoons” of relaxation processes in dipolar materials: the “first passage” relaxation function<sup>(5)</sup>, and the “average” relaxation function expressing the proportion of dipoles which did not change their imposed aligned orientation up to a certain time, the latter perhaps providing a description closer to what is experimentally measured. In some cases, the two relaxation functions are proved to coincide. The models are mathematically simple and do not take into account the spatial structure. Hence, admittedly, there are obvious limits to how much they can explain.

In Section 2 we present a simple probabilistic explanation of the appearance of the KWW response (with exponent  $0 < \alpha < 1!$ ) in the language of the “first passage” relaxation function  $\phi(t)$ , and provide its graphical interpretation. Let us remark that a broader class of dielectric responses can be obtained<sup>(7)</sup> if the random variate  $\theta$  is permitted to be infinite with a probability  $P_0 > 0$ . Such a modification expresses the well known fact that individual dipoles, and their environments, do not remain independent during the process of relaxation<sup>(13)</sup>, and leads directly to a class of double power laws, which includes the KWW and the Debye responses as special cases<sup>(7)</sup>. In Section 3 we introduce, via the

theory of order statistics, an “average” relaxation function  $\Phi(t)$ , better reflecting the quantities measured in experiments <sup>(14)</sup>. It gives the proportion of dipoles which did not change their imposed aligned orientation up to time  $t$ . We provide the graphical illustration of the “average” relaxation function for a fixed size  $N$  of a dipolar system, and evaluate its limit as  $N \rightarrow \infty$ , which recovers the original distribution of switching time for individual dipoles. Finally, in Section 4 we show that in the KWW case, the “first passage” relaxation function is, essentially, identical with the “average” relaxation function.

## 2. ‘First passage’ relaxation function.

The traditional explanation for a non-exponential (non-Debye) dipolar relaxation has been to assign a local value to the relaxation time  $\tau$  for each dipole, and then to recover the observed regression of polarization fluctuations by means of the relaxation function  $\phi(t)$  expressed as a weighted average of exponential relaxation function <sup>(15)</sup>

$$\phi(t) = \int w(\tau) \exp(-t/\tau) d\tau \quad (2.1).$$

This very natural statistical attempt has the advantage of retaining the stochastic features of Debye’s original concept <sup>(16)</sup> of independently exponentially relaxing dipoles in a viscous medium that acts as a random noise source. However, the choice of the weight function  $w(\tau)$  can not be arbitrary, but has to be determined *ad hoc* by the empirical relaxation functions; such an approach hardly explains universality of dielectric responses. <sup>(17–19)</sup> The best one can conclude is that a given, empirically observed, relaxation law is compatible with a particular distribution of relaxation times  $\tau$ . That approach does not explain why the stretched exponential, or the double power law should be so universally applicable.

The recent probabilistic approach <sup>(5–8)</sup> to non-exponential relaxation responses, based on the concept contained in the cluster model, <sup>(13,18)</sup> and a new definition of relaxation function as the probability that the system as a whole has not changed its initial state up to time  $t$ , uniquely leads to the experimentally observed stretched exponential and double power forms of dielectric responses. In this paper we will refer to that relaxation function as to the “*first passage*” relaxation function. Let us describe the basic results in some detail.

As is the case in the traditional approach, neglecting the inter-cluster motions, we may assume that the probability that the dipole has not changed its initial aligned position, imposed by an external field, during time  $t$  following removal of that field, is exponential

$$\phi_D(t, b) = P(\theta \geq t \mid \beta = b) = \exp(-bt) \quad \text{for} \quad t \geq 0, b > 0, \quad (2.2)$$

conditioned only by the value  $b$  taken by the relaxation rate  $\beta = 1/\tau$ . The latter reflects the random intra-cluster influence. Here  $\theta$  represents the random switching time of the individual dipole. In the system of  $N$  aligned dipoles the random variable  $\beta_i$  will denote the relaxation rate of the  $i$ -th dipole and the variable  $\theta_i$ , the “switching time” – the time needed for changing its initial orientation. The basic assumption is that  $\beta_1, \beta_2, \dots$  and  $\theta_1, \theta_2, \dots$  form sequences of non-negative, independent, identically distributed random variables with distribution functions  $F_\beta(b)$  and  $F(t)$ , respectively. The total probability that the  $i$ -th dipole has not changed its initial aligned position up to time  $t$  equals

$$P(\theta_i \geq t) = \int_0^\infty \exp(-bt) dF_\beta(b) = \langle \phi_D(t, \beta_i) \rangle. \quad (2.3)$$

The right-hand side of equation (2.3) is a weighted average of exponential relaxations with respect to the distribution of relaxation rates  $\beta$ , and corresponds to the weighted average

in (2.1). The crucial observation is that it is also the Laplace transform  $\mathcal{L}(F_\beta; t)$  of the distribution function  $F_\beta$ . It has to be stressed, that so far *there are no restrictions on*  $F_\beta$ ; the only requirement is that it is a distribution function supported on the nonnegative half-line.

For a finite ensemble of  $N$  dipoles one can then define a “first passage” relaxation function as

$$\begin{aligned}\phi_N(t) &= P\left(\min(\theta_1, \theta_2, \dots, \theta_N) \geq t\right) \\ &= \left[P(\theta_i \geq t)\right]^N = \left[\mathcal{L}(F_\beta; t)\right]^N,\end{aligned}\tag{2.4}$$

which expresses the probability that the whole system has not changed its initial state, imposed by an external electric field, up to time  $t$ . The second equality is justified by the assumption that  $\theta_i$ 's are independent, identically distributed random variables. However, the above quantity converges to zero as  $N \rightarrow \infty$  so, to get a meaningful relaxation function, one has to further rescale relaxation times. This leads to the following definition of the “first passage” relaxation function of macroscopic dipolar system:

$$\phi(t) = \lim_{N \rightarrow \infty} P\left(A_N \min(\theta_{1N}, \dots, \theta_{NN}) \geq t\right)\tag{2.5}$$

$$= \lim_{N \rightarrow \infty} \left[P\left(\theta_i \geq \frac{t}{A_N}\right)\right]^N = \lim_{N \rightarrow \infty} \left[\mathcal{L}\left(F_\beta; \frac{t}{A_N}\right)\right]^N.\tag{2.6}$$

where  $A_N$  is a sequence of suitable normalizing constants.

The “first passage” relaxation function  $\phi(t)$  is defined by formula (2.5) in terms of the time of switch of the first i.e., the fastest dipole, that is

$$T_N := \min(\theta_1, \dots, \theta_N)$$

which is pictured on Fig. 1.

*Fig. 1.*

The  $N$ -th power of the Laplace transform of the nondegenerate distribution function  $F_\beta$  converges to the non-degenerate limiting transform if and only if  $F_\beta$  belongs to the domain of attraction of a completely asymmetric Lévy  $\alpha$ -stable law <sup>(20,21)</sup> with  $0 < \alpha < 1$ . Then, we get the “first passage” relaxation function as a weighted average of exponential relaxations with respect to the distribution  $F_{\tilde{\beta}}$  of the relaxation rate  $\tilde{\beta}$

$$\phi(t) = \mathcal{L}(F_{\tilde{\beta}}; t) = \int_0^\infty \exp(-bt) dF_{\tilde{\beta}}(b) = \langle \phi_D(t, \tilde{\beta}) \rangle, \quad (2.7)$$

where  $\tilde{\beta}$  is an  $\alpha$ -stable random variable

$$\tilde{\beta} = \lim_{N \rightarrow \infty} \frac{\sum_{i=1}^N \beta_i}{A_N}.$$

The above result allow us to represent a system of independent, exponentially relaxing dipoles with an unknown distribution  $F_\beta$  of the relaxation rates  $\beta_i$ , i.e., a system “chosen” by the external field, by an “average” dipole with the relaxation rate  $\tilde{\beta}$  being  $\alpha$ -stable random variable.

Hence, the “first passage” relaxation function of a macroscopic dipolar system consisting of independent , conditionally exponentially relaxing dipoles with different relaxation rates  $\beta_i$  is uniquely determined and equals

$$\phi(t) = \exp[-(At)^\alpha], \quad 0 < \alpha < 1, \quad (2.8)$$

where  $A$  is a positive constant. The relaxation function obtained in equation (2.8) is of the form of the well known empirical KWW relaxation function. In the case when  $\alpha \rightarrow 1$  one

obtains the relaxation function in the Debye's exponential form and this corresponds to the case of degenerate limiting distribution function of relaxation rates  $\tilde{\beta}$ .<sup>(7,8)</sup>

It is not necessary to know the detailed nature of  $F_\beta$  to obtain the limiting form (2.8). 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 properties of  $F_\beta$ . Namely, the necessary and sufficient condition for the limit (2.7) to exist, and thus for the "first passage" relaxation function to be well defined, is that:<sup>(12)</sup>

$$\lim_{b \rightarrow \infty} \frac{1 - F_\beta(xb)}{1 - F_\beta(b)} = x^{-\alpha}, \text{ for each } x > 0 \text{ and } 0 < \alpha < 1, \quad (2.9)$$

what can be interpreted as a requirement that the individual relaxation rates have, approximately, a self-similar behavior:

$$P(\beta_i > xb) = x^{-\alpha} P(\beta_i > b) \quad \text{for any } x > 0, 0 < \alpha < 1, \text{ and large } b. \quad (2.10)$$

This result, obtained here by means of purely probabilistic techniques, is in agreement with physical models<sup>(2)</sup> in which the scale-invariant relaxation rates has been identified as an underlying common mathematical feature.

In conclusion, if (2.1) has to represent the macroscopic relaxation function, then the weight function  $w(\tau)$  of relaxation times has to be of the form

$$w(\tau) = \frac{1}{\tau^2} w_\alpha(1/\tau),$$

where  $w_\alpha = p(1/\tau; \alpha, 1)$  is the completely asymmetric Lévy-stable probability density with  $0 < \alpha < 1$ . This corresponds to the stretched exponential relaxation function.

### 3. “Average” relaxation function via order statistics.

Consider a sample of independent, identically distributed random variables

$$\theta_{1N}, \dots, \theta_{NN}$$

representing switching times of  $N$  dipoles, with a common distribution function  $F(t)$ . The dipoles switch in a certain order, and in our model in this section that order will be reflected in the notion of the order statistics <sup>(22)</sup>

$$\theta_{1:N}, \dots, \theta_{N:N},$$

which is simply a nondecreasing rearrangement of the above switching times  $\theta_{1N}, \dots, \theta_{NN}$ .

In other words,

$$\theta_{1:N} \leq \dots \leq \theta_{N:N}.$$

Traditionally,  $\theta_{k:N}$  is called the  $k$ -th order statistics of the above sample. Note that

$$\theta_{1:N} = T_N = \min(\theta_{1N}, \dots, \theta_{NN}),$$

which corresponds to the quantity introduced in the last section where the “first passage” relaxation function was discussed, and that

$$\theta_{N:N} = \max\{\theta_{1N}, \dots, \theta_{NN}\}.$$

In terms of the above ordered switching times, the relaxation function for a fixed size  $N$  of the system, can be defined as

$$\Phi_N(t) := 1 - \frac{k}{N} \quad \text{if } \theta_{k:N} \leq t < \theta_{k+1:N},$$

where, by definition  $\theta_{0:N} = 0$ . A sample path of such a relaxation function is pictured on Fig. 2.

*Fig. 2.*

In other words,

$$\Phi_N(t) = 1 - \frac{k_N(t)}{N}$$

where

$$k_N(t) = \min\{k : \theta_{k+1:N} > t\} = \max\{k : \theta_{k:N} \leq t\}.$$

Then the classical relaxation function  $\Phi(t)$  that measures the average proportion of unswitched dipoles at time  $t$  can be defined as

$$\Phi(t) := \lim_{N \rightarrow \infty} \langle \Phi_N(t) \rangle. \quad (3.1)$$

Consider a fixed  $k$  and  $N$ . The critical observation which makes order statistics useful for our purposes is that, for any  $t > 0$  the event

$$\begin{aligned} & \{\theta_{k:N} \geq t\} \\ &= \{\text{at least } N - (k - 1) \text{ of the switching times } \theta_{iN} \geq t \text{ for } 1 \leq i \leq N\} \\ &= \{e_N(t) \geq N - (k - 1)\}, \end{aligned} \quad (3.2)$$

where, after an increasing rearrangement of the above switching times of  $N$  dipoles,  $e_N(t) = N - k_N(t)$  represents the total number of dipoles the switching times thereof exceeded <sup>(23)</sup> time  $t$ .

In this context, to evaluate (3.1), we need to look first at what can be called the finite ensemble “average” relaxation function expressing the proportion of dipoles which did not

change their orientation up to the moment  $t$ . We have

$$\begin{aligned}
\langle \Phi_N(t) \rangle &= \frac{\langle e_N(t) \rangle}{N} \\
&= \frac{1}{N} \sum_{k=1}^N k P(e_N(t) = k) = \frac{1}{N} \sum_{k=1}^N P(e_N(t) \geq k) \\
&= \frac{1}{N} \sum_{k=1}^N P(e_N(t) N - (k-1)) = \frac{1}{N} \sum_{k=1}^N P(\theta_{k:N} \geq t).
\end{aligned} \tag{3.3}$$

However, the distribution of the  $k$ -th order statistics is well known <sup>(22,23)</sup> and expressible in terms of the common distribution function  $F(t)$  of individual switching times  $\theta_{k:N}$  so that we get

$$\langle \Phi_N(t) \rangle = \frac{1}{N} \sum_{k=1}^N \sum_{l=0}^{k-1} \binom{N}{l} F^l(t) (1-F(t))^{N-l}, \tag{3.4}$$

and after changing the order of summation

$$\begin{aligned}
\langle \Phi_N(t) \rangle &= \frac{1}{N} \sum_{l=0}^{N-1} \sum_{k=l+1}^N \binom{N}{l} F^l(t) (1-F(t))^{N-l} \\
&= \frac{1}{N} \sum_{l=0}^{N-1} (N-l-1) \binom{N}{l} F^l(t) (1-F(t))^{N-l} \\
&= \sum_{l=0}^{N-1} \binom{N}{l} F^l(t) (1-F(t))^{N-l} \\
&\quad - \frac{1}{N} \sum_{l=0}^{N-1} l \binom{N}{l} F^l(t) (1-F(t))^{N-l} - \frac{1}{N} \sum_{l=0}^{N-1} \binom{N}{l} F^l(t) (1-F(t))^{N-l} \\
&= (1 - F^N(t)) - \frac{1}{N} (NF(t) - NF^N(t)) - \frac{1}{N} (1 - F^N(t)) \\
&= 1 - F(t) - \frac{1}{N} (1 - F^N(t)).
\end{aligned}$$

Now, as  $N \rightarrow \infty$  we obtain that for the ‘‘average’’ relaxation function based on the above order statistics model

$$\Phi(t) = \lim_{N \rightarrow \infty} \langle \Phi_N(t) \rangle = 1 - F(t). \tag{3.5}$$

This recovers the original distribution of switching time for individual dipoles.

## 4. Remarks and Conclusions

4.1. *When do the ‘first passage’ and the ‘average’ relaxation functions coincide?* The “first passage” and “average” relaxation functions  $\phi(t)$  and  $\Phi(t)$  coincide if, and only if the distribution  $F_\beta(t)$  of relaxation rates on the microscopic level is equal to the distribution  $F_{\tilde{\beta}}(t)$  on the macroscopic level. Then, in view of (2.7) and (2.8), both of them have to be  $\alpha$ -stable with  $0 < \alpha < 1$ .

Indeed, if we assume that the “first passage” relaxation function is well defined, then we have from formula (2.3) that

$$1 - F(t) = P(\theta_i \geq t) = \int_0^\infty \exp(-bt) dF_\beta = \mathcal{L}(F_\beta; t).$$

Hence, formula (3.5) for the “average” relaxation function can be rewritten as follows  $\Phi(t) = \mathcal{L}(F_\beta; t)$ . On the other hand, by (2.7),  $\phi(t) = \mathcal{L}(F_{\tilde{\beta}}; t)$ , and we can see that  $\Phi(t) = \phi(t)$  if and only if  $\mathcal{L}(F_\beta; t) = \mathcal{L}(F_{\tilde{\beta}}; t)$ . Since the Laplace transform uniquely determines the distribution function, the necessary and sufficient condition for the equality of the two relaxation functions is that the distribution functions

$$F_\beta(t) = F_{\tilde{\beta}}(t).$$

Once we decide to use the “first passage” relaxation function, its very existence implies that the microscopic distribution function  $F_\beta(t)$  of relaxation rates is in the domain of normal attraction of the  $\alpha$ -stable law and has the  $t^{-\alpha}$  asymptotics at  $+\infty$  described in (2.9), the same as the  $\alpha$ -stable distribution function  $F_{\tilde{\beta}}(t)$  itself. So, in practical terms, we can just say that the equality of microscopic and macroscopic relaxation rates is a necessary

and sufficient condition for the equality of the “first passage” and “average” relaxation functions.

Note that among other commonly used one-sided distribution functions <sup>(24)</sup> such as Burr, Pareto, generalized Pareto, transformed gamma, gamma, loggamma, lognormal, and Weibull distribution functions, only the Burr and Pareto distribution functions satisfy condition (2.9).

*4.2. A hierarchy of relaxation functions.* Notice, that the order statistics discussed in Section 3, permit introduction of a whole hierarchy of relaxation functions corresponding to “first passage”, “second passage” , etc., transition functions, thus extending the ideas of Section 2. They all lead to a KWW-type relaxation functions, but with polynomial corrections. Indeed, if  $k = 1, 2, \dots, N$ , and  $N$  is the number of dipoles in the finite ensemble, then the “ $k$ -th passage” transition function

$$\phi^{(k)}(t) = \lim_{N \rightarrow \infty} P(A_N \theta_{k:N} \geq t) = e^{-(At)^\alpha} \sum_{i=1}^{k-1} \frac{(At)^\alpha}{i!},$$

where  $A_N$  is a sequence of suitable normalizing constants. <sup>(22,23)</sup> Obviously

$$\phi^{(1)}(t) = \phi(t)$$

corresponds to the “first passage” relaxation function introduced in Section 2.

4.3. *Fractal scaling of time.* The appearance of the KWW relaxational response can also be partially justified by presupposing a fractal time scaling. On the physical level this line of reasoning has been pursued by several authors <sup>(25–30)</sup>. Mathematically, it can be summarized as the following characterization of the KWW response function, which is a fractal generalization of the well known characterization of the exponential (Debye) distribution by its memoryless property: *If  $\theta$  is a nonnegative random variable and  $\alpha > 0$  then statements*

$$(i) P(\theta > (t^\alpha + s^\alpha)^{1/\alpha} \mid \theta > s) = P(\theta > t) \text{ for all } s, t \geq 0, \text{ and}$$

$$(ii) \text{ There exists a } \lambda > 0 \text{ such that } P(\theta > t) = \exp(-\lambda t^\alpha) \text{ for all } t \geq 0,$$

*are equivalent.*

As its exponential special case, the above characterization can be immediately obtained from the Sierpiński's theorem on solutions of the Cauchy functional equation  $f(t + s) = f(t)f(s)$ .

However, it is important to notice that the fractal time argument is unable to account for the fact that in empirical data always  $0 < \alpha < 1$ . On the other hand, models discussed in Sections 2 and 3, provide also a satisfactory mathematical explanation of that fact.

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