How to obtain the universal response law in the Jonscher screened hopping model for dielectric relaxation

1991 J. Phys.: Condens. Matter 3 221

(http://iopscience.iop.org/0953-8984/3/2/009)

J. Phys.: Condens. Matter 3 (1991) 221–223. Printed in the UK

LETTER TO THE EDITOR

How to obtain the universal response law in the Jonscher screened hopping model for dielectric relaxation

Karina Weron
Institute of Physics, Technical University of Wrocław, 50-370 Wrocław, Poland

Received 13 November 1990

Abstract. The revised screened hopping model is presented for relaxation in condensed matter. It is based on the concept of fractal time. The present model gives the universal dielectric response in the stretched exponential form as well as in the power law form.

A review of the experimental evidence relating to a wide range of dielectric materials (see, e.g., [1]) shows clearly that the relaxation behaviour departs strongly from the 'conventional' Debye exponential form. It is a striking fact that despite the variety of materials used and the experimental techniques employed, the relaxation behaviour is very similar. It was found that almost all the data can be represented in terms of two types of experimental fitting function: the stretched exponential function (the so-called Williams–Watts function) [2]:

$$\varphi(t) \cong \exp[-(t/\tau)^{\alpha}] \qquad 0 < \alpha < 1 \tag{1}$$

or the power-type function (the Jonscher function) [3]:

$$f(t) \cong [(\omega_p t)^n + (\omega_p t)^{m+1}]^{-1} \qquad 0 < n, m < 1 \tag{2}$$

where τ and ω_p are constants for a given material.

The characteristic feature of this universal law of the dielectric response is the fact that the real and imaginary components of the complex susceptibility

$$\chi(\omega) = \int f(t) \exp(-i\omega t)\, dt = \int \left(-\frac{d\varphi}{dt}\right) \exp(-i\omega t)\, dt \tag{3}$$

are the same functions of frequency, so their ratio is independent of frequency. The physical sense is given by the so-called 'energy criterion' [1]. It seems that if a physical mechanism that satisfies the energy criterion can be found, this mechanism would lead to the universal law of the dielectric response. Such a physical mechanism has been proposed by Jonscher [1] and was based on the concept of 'screened hopping', i.e., abrupt transitions of dipoles or charges between localized orientations or positions followed by a gradual adjustment of the surrounding dipoles or charges. This model has the required property of satisfying the energy criterion but does not, however, predict the observed universal law.

There are two features of the polarization process that are common to all materials exhibiting the universal law. The first common property is the presence of interactions

arising from the close proximity of atoms and molecules. As a consequence of many-body interactions the assemblies of charged particles exhibit the phenomenon of self-consistent screening. This leads to the energy criterion, as has been shown by Jonscher [4]: that is, due to self-consistent screening the ratio of energy lost per cycle to that stored at the field peak depends on the screening coefficient only.

The second feature common to all solid dielectrics is the discontinuous nature of the dipolar or charge-carrier transitions between their preferred 'stationary' orientations or positions. The interactive nature of the system implies that any sudden individual transition of a charge or a dipole brings about a series of 'chain' responses stretching both in time and in space beyond the time and position of the initiating transition. This property is equivalent to the existence of two distinctly separate time scales—the practically instantaneous transition time for the individual hopping movements and the relatively much longer screening adjustment time. It turns out that the second of these features is required in order to obtain the universal law of the dielectric response.

Let us consider a system consisting of a discrete series of relaxing modes (dipoles, atoms, ions) $N = 1, 2, 3, \ldots$. The relaxation can be imagined as a quasi-stationary process: the ith mode after the waiting time takes the step from the kth to the $(k + 1)$th stationary step and then stays there until smoothing of the fluctuation induced by the perturbation of the kth relaxation step has occurred. The time dependence of the polarization resulting from this sequence of events can be considered in two steps: at first there is a constant polarization in a waiting time $t_{w,i}$ and, next, after a very 'rapid' change at the moment of the instantaneous transition of the ith mode between the localized sites, a 'slow' change in a screening adjustment time $t_{a,i}$. The relaxation of the ith mode takes an experimental time equal to $t_i = t_{a,i} + t_{w,i}$. In a system of N relaxing modes we assume the ordering of times: $t_1 > t_2 > \ldots > t_i > \ldots > t_N$, choosing the first mode with the longest experimental time, and also that $t_{i+1} = t_{a,i}$.

Let us start with a straight line, namely the time axis, then cut out shorter and shorter constant-polarization gaps $t_{w,i}$. The set yielded by infinite interpolation and extrapolation is self-similar. This procedure is analogous to a construction known in mathematics as a Cantor set [5]. Since a point on the time axis marks an 'event', such a Cantor set is a fractal sequence of events. From the physical point of view, it is just a set of randomly distributed transition times for a given system of N relaxing modes. In order to find the dimension of an inhomogeneous Cantor set (a random weighted construction) we use the following construction [6, 7]. Let the longest relaxation time be

$$t_1 = t_{a,1} + t_{w,1} = (p_1 + q_1)r \tag{4}$$

where $t_{a,1}/t_{t,1} = p_1/q_1$ is a positive rational number, $(p_1 + q_1) > 2$, and r is a time scaling factor. Then

$$1 - t_{a,1}/t_1 = t_{w,1}/t_1 \tag{5}$$

where $t_{a,1}/t_1 = p_1/(p_1 + q_1)$ is the length of the part that remains from the normalized time interval $[0, 1]$ after the first construction stage, and $t_{w,1}/t_1 = q_1/(p_1 + q_1)$ is the length of the part cut out. Hence we get

$$1 - p_1/(p_1 + q_1) = q_1/(p_1 + q_1). \tag{6}$$

After the second construction stage, using the assumption $t_2 = t_{a,1}$ and dividing this time interval into $(p_2 + q_2) > 2$ pieces, we have the following relation:

$$1 - p_1p_2/(p_1 + q_1)(p_2 + q_2) = q_1/(p_1 + q_1) + p_1q_2/(p_1 + q_1)(p_2 + q_2) \tag{7}$$

where the right hand side denotes the part cut out of $[0, 1]$ after the two construction

steps. Denoting the dimension of the remainder set obtained after N steps by D_N, we have

$$D_N = \sum_{i=1}^{N} \log p_i \Big/ \sum_{i=1}^{N} \log(p_i + q_i) \qquad D = \lim_{N} D_N \tag{8}$$

which is a fraction between 0 and 1.

The only observed time is the experimental time t and from (8) it follows that

$$t_a/r = (t/r)^D. \tag{9}$$

By taking account of an intermittent process instead of a continuous one, one can write the relaxation equation in terms of the intrinsic time t_a. Generally, the decay of the polarization can follow the kth-order rate equation

$$\mathrm{d}P/\mathrm{d}t_a = -(C/r)P^k(t_a). \tag{10}$$

The time dependence of the polarization for $k = 1$ has the form:

$$P(t) = P_0 \exp[-C(t/r)^D] \qquad 0 < D < 1 \tag{11}$$

so the relaxation function $\varphi(t) = \exp[-C(t/r)^D]$ has, for $k \neq 1$, the stretched exponential form of (1):

$$P(t) = P_0[1 + (k-1)CP_0^{k-1}(t/r)^D]^{1/(1-k)} \tag{12}$$

so

$$f(t) = -\mathrm{d}\varphi/\mathrm{d}t = (D/r)CP_0^{k-1}(t/r)^{-1}[(t/r)^{D/k-D} + (k-1)CP_0^{k-1}(t/r)^{D/k}]^{k/(1-k)}. \tag{13}$$

Expression (13) brings out the physically significant feature that the time domain response can be made up of two sequential processes (2). The first obeys the relation

$$f(t) \cong (t/r)^{-n} \qquad n = 1 - D \qquad 0 < n < 1 \tag{14}$$

for short times and corresponds to the high-frequency part of the frequency response, while the second obeys the relation

$$f(t) \cong (t/r)^{-m-1} \qquad m = D/(k-1) \qquad 0 < m < 1 \tag{15}$$

for longer times.

In conclusion, the revised screened hopping model, generally applicable to a wide range of physical and chemical conditions, has not only the required property of satisfying the energy criterion but also leads to the universal dielectric response law in both empirically observed forms (1) and (2).

The author gratefully acknowledges Professor A K Jonscher for many stimulating discussions.

References

[1] Jonscher A K 1983 *Dielectric Relaxation in Solids* (London: Chelsea Dielectric Press)
[2] Williams G and Watts D C 1970 *Trans. Faraday Soc.* **66** 80
[3] Jonscher A K 1975 *Colloid. Polym. Sci.* **253** 1231
[4] Jonscher A K 1975 *Nature* **253** 717
[5] Mandelbrot B B 1982 *The Fractal Geometry of Nature* (San Francisco: Freeman)
[6] Chen H, Wu X and Fang J 1986 *J. Phys. C: Solid State Phys.* **19** L499
[7] Weron K and Janicki A 1990 *Proc. COSMEX '89* ed W Kasprzak and A Weron (Singapore: World Scientific) p 440