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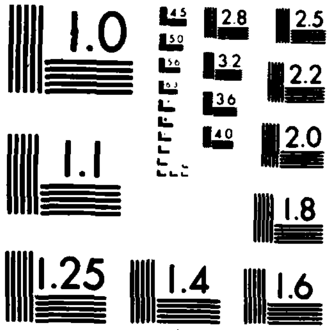
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Department of Statistics
University of North Carolina
Chapel Hill, North Carolina

A Statistical Approach to Relaxation in Glassy Materials

by

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and

Aleksander Weron

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A Statistical Approach to Relaxation in Glassy Materials[1]

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Abstract: Statistical aspects of the Williams-Watts relaxation model are derived explicitly from the limit theorem in probability theory. It is shown that the relaxation rate distribution is governed by completely asymmetric Lévy α-stable distributions, $0 < \alpha < 1$. This gives a rigorous approach to the stretched exponential form of relaxation function and also relates the effective relaxation time to the primitive relaxation time. It is demonstrated how useful is the technique of Lévy α-stable distributions in the study of relaxation phenomena which complements the recent works of Montroll-Bendler and Montroll-Shlesinger.

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1. Introduction.

It has been observed over years (1-5) that experimental relaxation behaviour in many glassy materials is generally well fitted with Williams-Watts function

$$\phi(t) = \exp(-t/\tau_e)^{\alpha}, \qquad 0 < \alpha < 1 \tag{1.1}$$

with α and τ_e constants for a given material, where α is an index of slowness and τ_e is an effective relaxation time. This is in contrast to the conventional Debye exponential form

$$\phi(t) = \exp(-t/\tau_0)$$

where $\alpha = 1$ and τ_0 is the primitive (Debye) relaxation time. Such stretched exponential behaviour has been noted in a wide variety of materials including organic liquids and solutions, polymers, and in particular in many glassy materials. The recent interest in relaxation problem(5-12) provides a number of models explaining the universality of formula (1.1). However, a connection with, the well known in probability theory, class of Lévy α-stable distributions(13) is not realized, except the very recent work of Montroll and Bendler[11].

Many scaling relations for complex systems in the physical sciences involve non-integer exponents, similarly as the Fourier transforms of Lévy α-stable distributions. Consequently, there are many results in statistical physics where Lévy α-stable distributions are invoked, but they are not recognized or kept hidden. For example, Khalfin(14) derived an asymptotic formula for quantum description of decaying physical system which is the Fourier transform of Lévy α-stable distribution of the system energy with $0<\alpha<1$. His paper influenced many physical studies in the decay theory of quantum systems but this relation was not recognized. In the theory of dielectric relaxation of polymeric systems and glasses Williams et al.(3) have derived a series representation of the normalized dielectric parameter. Its imaginary part up to multiplication factor is nothing else as known series representation of Lévy α-stable density. It is also well known(13) that this series representation has poor convergence properties for large arguments. Neverless this fact is rediscovered again by Lindsey and Patterson(6) who were forced to resort to special multiprecision computer programs. Even at that they obtain only numerical results although the functional form of the distribution function for large relaxation times is of equal interest. Helfand(7) continued their work and derived an asymptotic formula for the distribution function for large relaxation times which gives some correction terms to the, known for probabilists, series expansion of Lévy α-stable densities(15). He also made use of Zolotarev(16) explicit expressions of these densities for $\alpha = 1/3$ and $\alpha = 2/3$.

In our opinion a connection between the statistical aspects of Williams-Watts relaxation model and the class of Lévy α-stable distributions is not only a coincident analogy, but it seems to capture the essential statistical nature of relaxation in complex materials. Recall, that Lévy α-stable distributions form a class of universal limit laws in the sense that they describe completely the collective i.e., macroscopic behaviour of a complex system expressed by any normalized sum of independent identically distributed random quantities. As a consequence of this important theorem in probability(13.19) we shall derive in this paper the Williams-Watts form (1.1) of the relaxation function, as well as the existence of second universality relation, first obtained by Ngai(5), connecting τ_e with τ_0.

Ngai's cooperative-relaxation model based on Wigner's random matrices theory of energy-level structures leads to the same formula (1.1) and the effective relaxation time (in his notation) τ_p is related to the primitive relaxation time τ_0 as follows:

$$\tau_p^{1-n} = (1-n)(e^{\gamma^\circ}\omega_c)^n \tau_0, \qquad 0 < n < 1, \qquad (1.3)$$

where $\gamma^\circ = 0.5772$ is Euler's constant and ω_c is the high-frequency cutoff of the linear density of excitations. Recently, Chamberlin et al.(17) have proposed a different empirical relation in the context of the remanent magnetization in spin-glasses

$$\tau_p^{1-n} = (1-n)/C\,\omega^{1-n}, \qquad 0 < n < 1, \tag{1.4}$$

where C and ω are two temperature-independent constants throughout the spin-glass region. The controversy which arises here(17,18) motivates us to seek this relation from purely statistical point of view by using the scaling property of the characteristic function of completely asymmetric Lévy α-stable distributions relevant to the relaxation rates.

Since the statistical aspects of Williams-Watts relaxation model are directly connected with the Lévy α-stable distributions we employ them to study the distribution function of relaxation times which complements the discussion given by Lindsey and Patterson(6), and Helfand(7). Also the statistical properties of dielectric relaxation are discussed from this same point of view.

2. Lévy α-Stable Distributions

In this section we collect some basic facts on Lévy α-stable (LαS in short notation) distributions needed for the purpose of this article(13,19,20). One should not be surprised that probabilistic ideas, in particular limit theorems which provide a macroscopic distribution for a collection of microscopic quantities, play a role in statistical physics. We can refer, for example to Khinchin's(21) treatment of statistical mechanics which relies heavily on so called central limit theorem leading to Gaussian limit distribution. In recent years, inverse power long tails have become more evident in the analysis of physical phenomena of condensed matter(22) and therefore LαS distributions provide useful models. However, the principal feature of these distribution, that we want to stress for our purpose, is that they are completely described as limits of the normalized sums of independent and identically distributed summands. Consequently, LαS distributions express some kind of universality law.

The distribution function F(x) is called stable if for every $a_1 > 0$, b_1, $a_2 > 0$, b_2 there correspond constants $a > 0$, b such that the equation

$$F(a_1 x + b_1) * F(a_2 x + b_2) = F(ax + b) \tag{2.1}$$

holds. The symbol $F_1 * F_2$ indicates the convolution of two distributions in the sense

$$F_1 * F_2 = \int F_1(x-y)\, dF_2(y). \tag{2.2}$$

It turns out that always

$$a = (a_1^{\alpha} + a_2^{\alpha})^{1/\alpha} \text{ with } 0 < \alpha \leq 2 \tag{2.3}$$

and the constant α is called the characteristic exponent of LαS-distribution.

Equation (2.1) can be solved in terms of characteristic functions, i.e., via Fourier transform

$$f(s) = \int_{-\infty}^{\infty} \exp[isx]dF(x).$$

Thus for the distribution function F(x) to be LαS it is necessary and sufficient(19) that its characteristic function f(s) be represented by the formula

$$\log f(s) = \begin{cases} i\gamma s - \sigma |s|^{\alpha} \{1 - i\beta \operatorname{sign}(s)\tan(\pi\alpha/2)\} & \text{if } \alpha \neq 1 \\ i\gamma s - \sigma|s| - i\beta(2/\pi)\sigma s \log |s| & \text{if } \alpha = 1 \end{cases} \tag{2.4}$$

where α, β, γ and σ are real constants with $\sigma \geq 0$, $0 < \alpha \leq 2$ and $|\beta| \leq 1$. Here α is the characteristic exponent, γ and σ determine location and scale. The coefficient β indicates whether the LαS-distribution is symmetric (β=0) or completely asymmetric ($|\beta| = 1$).

Since f(s) is absolutely integrable the corresponding LαS-distribution has a density. It will be denoted by $p(x;\alpha,\beta)$ and can be calculated from the Fourier inversion formula. It seems impossible to express them in a closed form, but series expansions are available. Only in the case $0 < \alpha < 1$ the LαS-densities with $|\beta| = 1$ are one-sided i.e., their support is $[0,+\infty)$ for $\beta = 1$ and $(-\infty,0]$ for $\beta = -1$.

By means of a linear transformation of the variable in the distribution function F(x), it is possible to make $\sigma = 1$ and $\gamma = 0$. Observe that only the case $\sigma > 0$ is of interest, because $\sigma = 0$ corresponds to a degenerate distribution. We will use the following asymptotic formulas(15) for LαS-densities when $0 < \alpha < 1$, $\gamma = 0$ and $\sigma = 1$.

$$p(x;\alpha,\beta) = (1/\pi) \sum_{n=1}^{\infty} A_n \, x^{-n\alpha-1} \qquad \text{for } x \to \infty, \tag{2.5}$$

where

$$A_n = \frac{(-1)^{n+1}\Gamma(n\alpha+1)}{n!}\left(1+\beta^2\tan^2\frac{\pi\alpha}{2}\right)^{n/2}\sin n\left[\frac{\pi\alpha}{2} + \arctan\left(\beta\tan\frac{\pi\alpha}{2}\right)\right]$$

and

$$p(x;\alpha,1) = \left[\frac{B(\alpha)\ \lambda(\alpha)}{2\pi(1-\alpha)}\right]^{\frac{1}{2}} x^{\frac{-\lambda(\alpha)}{2}-1} \exp(-B(\alpha)x^{-\lambda(\alpha)})$$

$$\times\left[1 + 0\left(x^{\frac{\lambda(\alpha)}{2}-\varepsilon}\right)\right] \quad \text{for } x \to 0, \tag{2.6}$$

where

$$B(\alpha) = (1-\alpha)\alpha^{\frac{\alpha}{1-\alpha}}\left(\cos\frac{\pi\alpha}{2}\right)^{-\frac{1}{1-\alpha}} \tag{2.7}$$

and

$$\lambda(\alpha) = \frac{\alpha}{1-\alpha}\ . \tag{2.8}$$

The most convenient formulation of the limit theorem which gives the description of the distribution law F governing the sum of a large number of mutually independent identically distributed random quantities X_k can be given in the following form$^{(13)}$: *Only stable distributions have a domain of attraction,* i.e., there exist norming constant $a_n > 0$, b_n such that the distribution of $a_n^{-1}(X_1 + X_2 +\ldots+ X_n)-b_n$ tends to F. The normalization constants can be chosen in such a way that $a_n = n^{1/\alpha}$.

If G denotes a common distribution of the random variables X_k put for $x > 0$,

$$u(x) = \int_{-x}^{x} y^2 G(dy). \tag{2.9}$$

Then in order that G belongs to some domain of attraction it is necessary that u be of the form $u(x) \sim x^{2-\alpha}S(x)$, where $0 < \alpha \leq 2$ and S is slowly varying at ∞, that is, for fixed $x > 0$ $S(tx)/S(t) \to 1$ when $t \to \infty$. There are only two cases:

(i) if $\alpha = 2$ then G belongs to the domain of attraction of Gaussian distribution if and only if u is slowly varying,

(ii) if $0 < \alpha < 2$ then G belongs to the domain of attraction of LαS-distribution if and only if $u(x) \sim x^{2-\alpha} S(x)$ and the following two limits exist

$$\lim_{x\to\infty} \frac{1 - G(x)}{1 - G(x) + G(x)} \quad \text{and} \quad \lim_{x\to\infty} \frac{G(-x)}{1 - G(x) + G(-x)} \ . \qquad (2.10)$$

Observe that case (i) holds whenever the second moment of G exists, i.e., $\mathrm{Var}\, G < \infty$. For distributions without variance we have case (ii).

3. The Williams-Watts Function.

One of the most characteristic features of glassy materials is the non-exponential character of relaxation as a function of time. This has been shown to be true in a wide variety of materials, including organic liquids(23) and solutions(24), polymers(4,5,25), and in particular, many glass forming materials(26). It is a striking fact that, despite the variety of materials used and of the experimental techniques employed, the relaxation behaviour is universal(5). Good fits to the observations have been obtained with a functional form termed the Williams-Watts function given by formula (1.1).

It is common(6,7,11) to attempt to interpret general relaxation in terms of exponential relaxations, writing

$$\phi(t) = \int_0^\infty e^{-t/\tau} \rho(\tau)\, d\tau,$$

where $\rho(\tau)$ is the density of a statistical distribution, i.e., $\int_0^\infty \rho(\tau)\, d\tau = 1$, of relaxation times τ across different atoms, clusters, or degrees of freedom. If $\mu = \tau_0/\tau$ where τ_0 is a single relevant relaxation time associated with Debye relaxation, then μ is called a relaxation rate and is interpretable as dimensionless time. Substituting $s = t/\tau_0$ in formula (3.1) we have

$$\phi(\tau_0 s) = \int_0^\infty e^{-s/\mu}\, \tau_0 \mu^{-2} \rho(\tau_0/\mu) d\mu = \int_0^\infty e^{-s\mu} w(\mu) d\mu, \tag{3.2}$$

where

$$w(\mu) = \tau_0 \mu^{-2} \rho(\tau_0/\mu) \tag{3.3}$$

is the density of a distribution of dimensionless relaxation rates. Since this approach is microscopically arbitrary we may consider the random variables

$\mu_i = \tau_0^i/\tau$ as the possible relaxation rates of elements in a given complex material. The index i indicates the number of the elements in the system and μ_i are mutually independent and identically distributed. The problem is then to determine a random variable $\tilde{\mu}$ for describing the collective, i.e., macroscopic, behaviour of the relaxing system. The idea is to introduce a suitable normalized block-variable.

Suppose we divide our system in blocks of length n. The total relaxation rate of each block will be equal to $\sum_{i=1}^{n} \mu_i$. Clearly this is not a good collective variable since for large n it goes to infinity. Therefore we need to consider normalization

$$\tilde{\mu} = a_n^{-1} \sum_{i=1}^{n} \mu_i , \qquad a_n > 0. \tag{3.4}$$

Thus it follows from the limit theorem discussed in Section 2 that the distribution $w(\mu)d\mu$ of the collective, i.e., macroscopic relaxation rate, belongs to the class of LαS-distributions $0<\alpha\leq 2$. Since relaxation rates are non-negative, $w(\mu)$ has to have only non-negative support. Consequently, $w(\mu)$ is the LαS density with $0<\alpha<1$ and $\beta=1$, (cf. Section 2),

$$w(\mu) = p(\mu;\alpha,1), \qquad 0<\alpha<1. \tag{3.5}$$

We have then shown that the macroscopic behaviour of relaxation rates of our system is described by a distribution belonging to a rather restricted class of LαS-distributions with parameters $0<\alpha<1$ and $\beta=1$. The possible initial distributions of the relaxation rates split naturally into universality classes, parametrized by the characteristic exponent α, according to which domain of attraction they belong. Observe that all relaxation rates have an infinite variance and mean value, since for LαS distributions only p-th moments for $p<\alpha$ exist.

Now using (3.2) we get

$$\phi(\tau_0 s) = \int_0^\infty e^{-s\mu}\, p(\mu;\alpha,1)d\mu, \tag{3.6}$$

i.e., $\phi(\tau_0 s)$ is the Laplace transform of LαS density $p(\mu;\alpha,1)$. Recall, that from (2.4) the Fourier transform of $p(\mu;\alpha,1)$ can be written as

$$\begin{aligned} f(s) &= \exp\{i\gamma s - \sigma s^\alpha (1 - i\tan(\pi\alpha/2))\} \\ &= \exp\{i\gamma s - (\sigma/\cos(\pi\alpha/2))s^\alpha(\cos(\pi\alpha/2) - i\sin(\pi\alpha/2))\} \\ &= \exp\{i\gamma s - (\sigma/\cos(\pi\alpha/2))(-is)^\alpha\} \quad . \end{aligned} \tag{3.7}$$

By using a general relation between Laplace and Fourier transform for functions with the non-negative support $L(p(\mu);s) = F(p(\mu);is)$, (3.6) and (3.7) give

$$\phi(\tau_0 s) = \exp\{-\gamma s - (\sigma/\cos(\pi\alpha/2))\, s^\alpha\} \; . \tag{3.8}$$

Since in our notation $\tau_0 s = t$ thus (3.8) can be rewritten as

$$\phi(t) = \exp\{-\gamma t/\tau_0 - (\sigma/\cos(\pi\alpha/2))\, (t/\tau_0)^\alpha\} \tag{3.9}$$

As we observed in Section 2 one can choose $\gamma = 0$ and hence

$$\phi(t) = \exp\{-(t/\tau_e)^\alpha\}, \tag{3.10}$$

where

$$\tau_e^\alpha = [(1/\sigma)\cos(\pi\alpha/2)]\, \tau_0^\alpha \quad . \tag{3.11}$$

Thus (3.10) gives the Williams-Watts form of the relaxation function and (3.11) gives the relation between the effective relaxation time τ_e and the primitive relaxation time τ_0. Let's remark that formulas (3.10) and (3.11) have universal character as a consequence of the use of universal limit law in macroscopic behaviour of the relaxing system. This provides an answer to the question which has been posed recently by Palmer et al$^{(12)}$.

Since $\cos(\pi\alpha/2) \sim 1 - \alpha$ for $0<\alpha<1$ then (3.11) can be approximated by

$$\tau_e^\alpha \sim \frac{1 - \alpha}{\sigma} \tau_0^\alpha \qquad (3.12)$$

which differs from (1.4) only by changing the numerator $1 - \alpha$ in (3.12) into α, if one has made an unnecessary assumption$^{(17)}$ that $1-n = \alpha$ and $\omega \sim 1/\tau_0$. It seems to be that our relation (3.11) or (3.12) is not comparable directly with (1.3).

4. The Distribution Function of Relaxation Times

If the statistical approach to explaining nonexponential relaxation behaviour is taken, it becomes important to determine the distribution of exponential relaxation times which produce the Williams-Watts function(6,7,11). In this section we will employ the known asymptotic formulas for one-sided LαS-densities to derive the asymptotic behaviour of the distribution function (in probabilistic terminology density) $\rho(\tau)$, defined in (3.1).

Since $\rho(\tau) = \tau_0^{-1}\mu^2 w(\mu)$ according to (3.3), thus by (2.5), if we put $\beta = 1$, we have for $\tau \to 0$

$$\rho(\tau) = \frac{1}{\pi\tau_e} \sum_{n=1}^{\infty} \frac{(-1)^{n+1}\Gamma(n\alpha + 1)}{n!} \left(\frac{\tau}{\tau_e}\right)^{\alpha n-1} \sin n\pi\alpha, \tag{4.1}$$

where

$$\tau_e = [\cos(\pi\alpha/2)]^{1/\alpha}\tau_0 .$$

For $\tau \to \infty$ we get from (2.6)

$$\rho(\tau) = \frac{\lambda(\alpha)}{\tau_e\sqrt{2\pi\alpha}} \left(\frac{\tau}{\tau_e}\right)^{\frac{1}{2}\lambda(\alpha)-1} \exp\left\{-\left(\frac{\tau}{\tau_e}\right)^{\lambda(\alpha)}\right\} , \tag{4.2}$$

where $\tau_e = B(\alpha)^{-\lambda^{-1}(\alpha)}\tau_0$, $B(\alpha)$ and $\lambda(\alpha)$ are given by (2.7) and (2.8), respectively.

Observe that the existence of average relaxation time $<\tau>$, and higher moments, $<\tau^n>$, $n \geq 1$ follow without any trouble since by (3.3) and (2.5)

$$\begin{aligned} <\tau^n> &= \int_0^\infty \tau^n \rho(\tau)d\tau = \tau_0^n \int_0^\infty \mu^{-n} w(\mu)d\mu \\ &\approx \tau_0^n \int_0^\infty \mu^{-\alpha-1-n}\, d\mu < \infty . \end{aligned} \tag{4.3}$$

This is in sharp contrast to the momentless distribution of the relaxation rates $\mu = \tau_0/\tau$.

The relationship between the relaxation function $\phi(t)$ and the LαS-density $w(\mu)$, see (3.2), permits to find the relaxation time $<\tau>$ and higher moments without explicit knowledge of $\rho(\tau)$. Namely, since

$$w(\mu) = \frac{1}{2\pi i} \int_{-i\infty}^{i\infty} e^{\mu s} \phi(\tau_0 s) \, ds,$$

where $s = t/\tau_0$ and $\mu = \tau_0/\tau$, one has from (4.3)

$$<\tau^n> = \tau_0^n \int_0^\infty \mu^{-n} \left[\frac{1}{2\pi i} \int_{-i\infty}^{i\infty} e^{\mu s} \phi(\tau_0 s) ds\right] d\mu.$$

Moving the μ^{-n} term inside the integral over ds, interchanging the order of integration, and moving the $\phi(\tau_0 s)$ term outside of the integration over $d\mu$ yields

$$<\tau^n> = \tau_0^n \int_0^\infty \phi(\tau_0 s) \left[\frac{1}{2\pi i} \int_{-i\infty}^{i\infty} \mu^{-n} e^{\mu s} d\mu\right] ds$$

$$= \{\tau_0^n/\Gamma(n)\} \int_0^\infty \phi(\tau_0 s) \, s^{n-1} ds = \{1/\Gamma(n)\} \int_0^\infty \phi(t) t^{n-1} \, dt$$

$$= \{1/\Gamma(n)\} \int_0^\infty t^{n-1} \exp \{-(\frac{t}{\tau_e})^\alpha\} \, dt$$

$$= \tau_e^n \, \Gamma(\tfrac{n}{\alpha})/(\alpha\Gamma(n)) \qquad (4.4)$$

$$= \tau_0^n \, [(1/\sigma) \cos (\pi\alpha/2)]^{n/\alpha} \, \Gamma(\tfrac{n}{\alpha})/(\alpha\Gamma(n)), \qquad (4.5)$$

where we have used formulas (3.10) and (3.11). Formula (4.4) corresponds to the result in Lindsey and Patterson$^{(6)}$.

5. Dielectric Relaxation

The complex dielectric permittivity $\varepsilon^*(\omega)$ is related to the relaxation function $\phi(t)$ by a one-sided Fourier transform(27)

$$\frac{\varepsilon^*(\omega) - \varepsilon_\infty}{\varepsilon_0 - \varepsilon_\infty} = \int_0^\infty e^{-i\omega t} \left(-\frac{d}{dt} \phi(t)\right) dt, \tag{5.1}$$

where $[-(d/dt)\phi(t)]$ is the normalized transient decay current which follows when a steady electric field is removed from a sample. Here ε_0 is the static dielectrical constant and ε_∞ the high-frequency limit of the dielectric constant. One generally writes

$$\frac{\varepsilon^*(\omega) - \varepsilon_\infty}{\varepsilon_0 - \varepsilon_\infty} = \varepsilon'(\omega) - i\varepsilon''(\omega) , \tag{5.2}$$

where $\varepsilon'(\omega)$ and $\varepsilon''(\omega)$ are, respectively, the real and imaginary parts of the normalized dielectric parameter. Williams et al.(3) derived an expression for $\varepsilon^*(\omega)$ when $\phi(t) = \exp\{-(t/\tau_e)^\alpha\}$ in the form

$$\varepsilon'(\omega) - i\varepsilon''(\omega) = \sum_{n=1}^{\infty} (-1)^{n-1} \frac{1}{(\omega\tau_e)^{n\alpha}} \frac{\Gamma(n\alpha + 1)}{n!} \times \\ \times\left(\cos n \frac{\pi 2}{2} - i \sin n \frac{\pi 2}{2}\right). \tag{5.3}$$

By the comparison with (2.5) one obtains immediately that

$$\varepsilon''(\omega) = x\pi p(x;\alpha,0), \tag{5.4}$$

where $x = \omega\tau_e$. This direct relation between the imaginary part ε'' of the normalized dielectric parameter and the symmetric LαS density $p(x;\alpha,0)$ has been also shown recently by a different method(11). It is interesting to explain why here in contrast to Sections 3 and 4 symmetric ($\beta=0$) LαS distributions arise.

For this observe that if we put $\gamma = 0$ and $\sigma = 1$ in formula (2.4) then for $0 < \alpha < 1$

$$\log f(s) = - |s|^{\alpha} \{1 - i\beta \operatorname{sign} (s) \tan (\pi\alpha/2)\}. \qquad (5.5)$$

Knowing that the density $p(x;\alpha,\beta)$ of such LαS distribution is real, it can be written, after a short calculation, in the form

$$p(x;\alpha,\beta) = \frac{1}{2\pi} \operatorname{Re} \int_{-\infty}^{\infty} e^{-isx} f(s)\, ds =$$

$$= \frac{1}{\pi} \int_{0}^{\infty} e^{-s^{\alpha}} \cos [xs-\beta s^{\alpha} \tan(\pi\alpha/2)]\, ds. \qquad (5.6)$$

On the other hand the integration by parts in (5.1) yields

$$[\varepsilon'(\omega) - 1] - i\varepsilon''(\omega) = -x \int_{0}^{\infty} e^{-s^{\alpha}} \sin(sx) dx - ix \int_{0}^{\infty} e^{-s^{\alpha}} \cos(sx) ds, \qquad (5.7)$$

where $s = t/\tau_c$ and $x = \omega\tau_c$. The comparison of (5.6) and (5.7) shows that the imaginary part $\varepsilon''(\omega)$ of the normalized dielectric parameter is related to LαS distribution $p(x;\alpha,\beta)$ if and only if $\beta = 0$, i.e., when the LαS distribution is a symmetric one. Namely one obtains from (2.5) and (3.11)

$$\begin{aligned} \varepsilon''(\omega) &= \pi\omega\tau_c\, p(\omega\tau_c;\alpha,0) \\ &= \sum_{n=1}^{\infty} (-1)^{n+1} \frac{\Gamma(n\alpha + 1)}{n!} (\omega\tau_0)^{-n\alpha} [\cos(\pi\alpha/2)]^{-n} \sin (n\pi\alpha/2). \end{aligned} \qquad (5.8)$$

6. Concluding Remarks

There are several approaches to explaining nonexponential relaxation behaviour in glassy materials$^{(1-12)}$. The statistical approach interprets the nonexponential relaxation behaviour of the material in terms of a superposition of exponentially relaxing processes which then leads to a distribution of relaxation times. In this paper we derived the Williams-Watts formula directly from the limit theorem in probability theory that explains the universality of this formula, as well as provides a relation between the effective relaxation time τ_e and the primitive relaxation time τ_0. This procedure is different from those in the existing literature and provides the main departure from all the previous work on the statistical approach to the relaxation problem. The most important consequence is that the theory of Lévy α-stable distributions can be directly employed to the study of the problem.

We do not propose that the statistical approach is the physically more correct one; instead we discuss the mathematical foundation and consequences of this approach which must be understood when physical interpretation is attached to the distribution functions underlying the relaxation. We would like to conclude the paper by quotation from E.P. Wigner$^{(28)}$: "The first point is that mathematical concepts turn up in entirely unexpected connections. Moreover, they often permit an unexpectedly close and accurate description of the phenomena in these connections. Secondly, just because of this circumstance, and because we do not understand the reasons of their usefulness, we cannot know whether a theory formulated in terms of mathematical concepts is uniquely appropriate." Let it be an excuse for the authors, if they were not sufficiently modest in some formulation of this paper.

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