

Relaxation Processes and Fractional Differential Equations

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Relaxation properties of different media (dielectrics, semiconductors, ferromagnetics, and so on) are normally expressed in terms of response function $f(t)$ or of real and imaginary components of its Fourier transform dependent on the frequency ω . It had been recently recognized that most of real materials show deviation from classical Debye process. There exist a few empirical approximations of non-Debye response functions. One of them is the two-power approximation containing ω^α and ω^β , where α and β belong to the interval (0, 1). This formula gives the basis for introducing of fractional differential equation considered in this paper. A stochastic interpretation of this equation is offered; its solution is found and investigated. The results are in agreement with experimental data.

KEY WORDS: relaxation processes; fractional differential equations.

1. INTRODUCTION

Relaxation properties of different media (dielectrics, semiconductors, ferromagnetics, and so on) are normally expressed in terms of time-domain response function $f(t)$ which represents the current flowing under the action of a step-function electric field, or of the frequency-dependent real and imaginary components of its Fourier transform

$$\tilde{f}(i\omega) \equiv \int_0^\infty e^{-i\omega t} f(t) dt = \varphi'(\omega) - i\varphi''(\omega). \quad (1)$$

The classical Debye expression for a system of noninteracting randomly oriented dipoles freely floating in a neutral viscous liquid (see, for example, Kubo, 1965) is

$$\tilde{f}(i\omega) = \frac{1}{1/\tau + i\omega}, \quad (2)$$

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where τ is the temperature-dependent relaxation time characterizing the Debye process:

$$f(t) = f(0) e^{-t/\tau}, \quad t > 0. \quad (3)$$

The latter function obeys the simple differential equation

$$df/dt + \tau^{-1} f(t) = 0. \quad (4)$$

Numerous experimental data gathered, for instance, in books (Jonscher, 1983, 1996; Ramakrishnan and Lakshmi, 1987) convincingly show that this theory is not able to describe relaxation processes in solids. There exist a few other empirical response functions for solids: the Cole–Cole (CC) function (Cole and Cole, 1941)

$$\tilde{f}_\alpha(i\omega) = \frac{1}{a_0 + a(i\omega)^\alpha}, \quad 0 < \alpha < 1, \quad (5)$$

the Cole–Davidson (CD) function (Jonscher, 1983)

$$\tilde{f}_\beta(i\omega) = \frac{1}{[a_0 + a(i\omega)]^\beta}, \quad 0 < \beta < 1, \quad (6)$$

and the Havriliak–Negami (HN) function (Havriliak and Negami, 1966)

$$\tilde{f}_\alpha^\beta(i\omega) = \frac{1}{[a_0 + a(i\omega)^\alpha]^{\beta/\alpha}}, \quad 0 < \alpha < 1, \quad 0 < \beta < 1, \quad (7)$$

and some others.

The HN function (7) covers the previous cases (2), (5), and (6), and is considered as a general expression for the *universal relaxation law* (Jonscher, 1983). A schematic representation of various types of dielectric response in the α, β plane is shown in Fig. 1. The full vertex of the square represents the Debye relaxation, the full diagonal and the full side display the CC relaxation and the CD relaxation respectively.

The “universality” means that any other relaxation process can be depicted by a point on the α, β plane. This universality is observed in dielectric relaxation in dipolar and nonpolar materials, conduction in hopping electronic semiconductors, conduction in ionic conductors, trapping in semiconductors, decay of delayed luminescence, surface conduction on insulators, chemical reaction kinetics, mechanical relaxation, magnetic relaxation. Despite of quite different intrinsic mechanisms, the processes manifest astonishing similarity (Jonscher, 1996).

The situation seems to be similar to diffusion processes. Random movements of small pollen grain visible under a microscope, neutrons in nuclear reactors, electrons in semiconductors are quite different processes from physical point of view but they are the same process of Brownian motion from stochastic point of view. This analogy stimulates search of an appropriate stochastic model for the universal relaxation law. A few attempts of such kinds have been undertaken

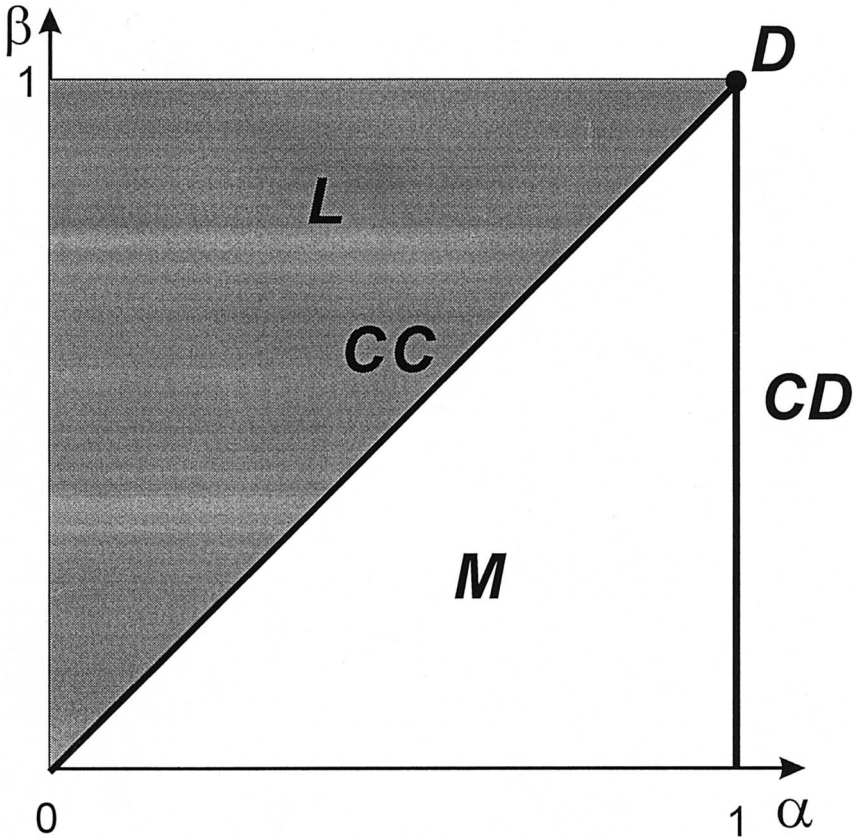


Fig. 1. Various types of dielectric response. Abbreviation: D – Debye law, CC – Cole–Cole law, CD – Cole–Davidson law, M – region of more typical behavior, L – region of less typical behavior.

in the works (Glöckle and Nonnenmacher, 1993; Jurlewicz and Weron, 2000; Nigmatullin and Ryabov, 1997; Weron and Kotulski, 1996, 1997) and for description of CC and CD relaxations (see full lines in Fig. 1).

The purpose of my report is to extend the stochastically interpreted area by including the region L (shaded region in Fig. 1).

2. FRACTAL PROCESS AND FRACTIONAL DERIVATIVE

Let us consider a simple point process (Prochorov, 1999, p. 727)

$$\dots < X_{-2} < X_{-1} < X_0 = 0 < X_1 < X_2 < \dots, \quad X_j \in \mathbf{R}^1.$$

We will call it the conditionally homogeneous point process if the conditional counting process

$$N(t|X_j) = \sum_{j=1}^{\infty} 1_{[X_j, X_{j+t}]}(X_j), \quad t \geq 0,$$

where

$$1_{[a, b]}(x) = \begin{cases} 1, & a \leq x \leq b, \\ 0, & x \geq b, \end{cases}$$

does not depend on X_j . The point process will be called conditionally stationary if

$$EN(t|X_i) \equiv \bar{N}(t) = N(1)t.$$

The conditional density of the process

$$n(t) = d\bar{N}(t)/dt$$

is constant:

$$\frac{dn(t)}{dt} = 0, \quad t > 0.$$

Call this property the first-order conditional stationarity and introduce the concept of ν -order conditional stationarity $0 < \nu \leq 1$ via relation

$$\frac{d^\nu n_\nu(t)}{dt^\nu} = 0, \quad t > 0. \quad (8)$$

Here

$$\frac{d^\nu n_\nu(t)}{dt^\nu} = \frac{1}{\Gamma(1-\nu)} \frac{d}{dt} \int_0^t \frac{n_\nu(\tau) d\tau}{(t-\tau)^\nu}, \quad 0 < \nu \leq 1, \quad (9)$$

is the Riemann–Liouville fractional derivative (Miller and Ross, 1993; Saichev and Woyczynski, 1996; Samko *et al.*, 1993).

It is known that

$$\int_0^\infty e^{-\lambda t} \frac{d^\nu n(t)}{dt^\nu} dt = \lambda^\nu \int_0^\infty e^{-\lambda t} n(t) dt, \quad (10)$$

and

$$\frac{d^\nu t^\mu}{dt^\nu} = \frac{\Gamma(\mu+1)}{\Gamma(\mu-\nu+1)} t^{\mu-\nu}, \quad \mu > -1.$$

In case $\mu = \nu - 1$, the latter equation yields

$$\frac{d^\nu t^{\nu-1}}{dt^\nu} = 0.$$

This means that the solution of Eq. (8) has the form

$$n_\nu(t) = ct^{\nu-1},$$

and

$$\bar{N}_\nu(t) = \int_0^t n_\nu(t) dt = \bar{N}_\nu(1)t^\nu, \quad 0 < \nu \leq 1. \tag{11}$$

They say that process $N(t|X_j)$ obeying the condition (11) is a fractal process with fractal dimension ν (Feder, 1988). This dimension coincides with the order ν of fractional derivative (9), and this coincidence stresses the link between fractals and fractional calculus.

There exist two interpretations of fractional derivatives: geometrical and dynamical (Kubarev, 1993; Mehaute *et al.*, 1998; Nigmatullin, 1992; Rutman, 1994, 1995). The above example shows that a third interpretation is possible: this is the stochastic interpretation, leading to stochastic fractals.

3. INTEGRAL EQUATION FOR THE RESPONSE FUNCTION

Consider a simple killed renewal process. Let T_1, T_2, T_3, \dots be nonnegative independent identically distributed random variables with common distribution function $F(t)$ and let $\Theta_1, \Theta_2, \Theta_3, \dots$ be random variables independent of T_j and among themselves with common distribution

$$P\{\Theta_j = 1\} = c, \quad P\{\Theta_j = 0\} = 1 - c, \quad 0 < c \leq 1.$$

Define stochastic process $N(t)$ via the relation

$$N(t) = 1_{[0,t]}(T_1) + \sum_{j=2}^{\infty} \Theta_1 \dots \Theta_{j-1} 1_{[0,t]}(T_1 + T_2 + T_3 + \dots + T_j). \tag{12}$$

Its probabilistic sense is very clear. If, for example, we keep in mind some physical system relaxing toward an equilibrium state by means of jumps, then T_1 is a moment of the first jump of the system, $T_1 + T_2$ is a moment of its second jump, and so on. If $c < 1$, the full number of jumps J is a.s. finite and

$$\text{Prob}\{J = k\} = c^{k-1}(1 - c).$$

In other words the sequence $T_1, T_1 + T_2, \dots, T_1 + T_2 + \dots + T_J$ is nothing but a killed Markov chain on positive semiaxis t .

Calculate the mean value $\bar{N}(t)$ by averaging relation (8) over all possible realizations of the process:

$$\bar{N}(t) = F(t) + \sum_{j=2}^{\infty} c^{j-1} F_j(t), \tag{13}$$

where

$$F_2(t) = \int_0^t F(t - \tau) dF(\tau) \equiv F(t) * F(t)$$

and

$$F_j(t) = F(t) * F_{j-1}(t), \quad j = 3, 4, \dots$$

are multifold convolutions of the distribution function $F(t)$. Rewriting Eq. (13) in the form

$$\bar{N}(t) = F(t) + cF(t) * \left[F(t) + \sum_{j=2}^{\infty} c^{j-1} F_j(t) \right] = F(t) + cF(t) * \bar{N}(t),$$

we arrive at the integral equation,

$$\bar{N}(t) = F(t) + c \int_0^t F(\tau - t) d\bar{N}(\tau) d\tau,$$

with the substochastic kernel $cF(t)$ (Feller, 1971). Identifying the response function $f(t)$ with the rate of increasing $\bar{N}(t)$,

$$f(t) = d\bar{N}/dt,$$

we get

$$f(t) = p(t) + c \int_0^t p(t - \tau) f(\tau) d\tau, \quad (14)$$

where $p(t) = dF/dt$. Choosing different kernels for the equation, we obtain different kinds of relaxation processes. To make the calculations more simple, we pass to the Laplace transform of Eq. (10):

$$\tilde{f}(\lambda) = \tilde{p}(\lambda) + c\tilde{p}(\lambda)\tilde{f}(\lambda), \quad (15)$$

where

$$\tilde{p}(\lambda) = \int_0^{\infty} e^{-\lambda t} p(t) dt,$$

and so is $\tilde{f}(\lambda)$. Notice that for $\lambda = i\omega$ $\tilde{f}(i\omega)$ coincides with Eq. (1).

The Debye process emerges now as a result of choosing the kernel in the exponential form

$$p(t) = (1/a)e^{-t/a}, \quad a > 0.$$

Its Laplace transform reads

$$\tilde{p}(\lambda) = 1/(1 + \lambda a). \quad (16)$$

On substituting Eq. (16) in Eq. (15), we get

$$a\lambda\tilde{f}(\lambda) + a_0\tilde{f}(\lambda) = 1,$$

where $a_0 = 1 - c$. This is the Laplace transform of the above differential Eq. (4).

The most important property of the Debye time-process is absence of memory: the ratio,

$$f(t' + t)/f(t') = e^{-t/a},$$

does not depend on t' . From this point of view the Debye process and only it is the simplest stochastic process of the kind under consideration.

4. THE COLE-COLE PROCESS

Let us consider a process with memory whose waiting time probability density has a power-type asymptotics

$$\int_t^\infty p(t') dt' \equiv \int_t^\infty p - (t') dt' \sim At^{-\alpha}, \quad t \rightarrow \infty, \quad 0 < \alpha < 1. \quad (17)$$

According to Tauberian theorem, the condition (17) leads to the asymptotic relation

$$1 - \tilde{p}_\alpha(\lambda) \sim A\Gamma(1 - \alpha)\lambda^\alpha, \quad \alpha \rightarrow 0. \quad (18)$$

Substitution (18) into (15) yields the following equation for the asymptotical term

$$a\lambda^\alpha \tilde{f}_\alpha^{as}(\lambda) + a_0 \tilde{f}_\alpha^{as}(\lambda) = 1, \quad (19)$$

where $a_0 = 1 - c$ and

$$a = cA\Gamma(1 - \alpha). \quad (20)$$

According to (10), Eq. (15) is the Laplace image of the fractional differential equation

$$aD^\alpha f_\alpha^{as}(t) + a_0 f_\alpha^{as}(t) = \delta(t), \quad (21)$$

describing the CC relaxation model.

The solution of this equation is found in (Podlubny, 1994):

$$f_\alpha(t) = \frac{1}{\alpha} t^{\alpha-1} E_{\alpha\alpha}(-\alpha_0 t^\alpha/a),$$

where

$$E_{\alpha\beta}(z) = \sum_{k=0}^\infty \frac{z^k}{\Gamma(\alpha k + \beta)}, \quad \alpha > 0, \quad \beta > 0,$$

is the two-parameter function of the Mittag–Leffler type. To analyze the solution at large times, we come back to Eq. (15) and replace $\tilde{p}_\alpha(\lambda)$ by the function

$$\tilde{p}_\alpha(\lambda) = e^{-b\lambda^\alpha}$$

with

$$b = A\Gamma(1 - \alpha), \quad (22)$$

as required by Eq. (18). In this case, $p(t)$ is connected with one-sided stable density

$$g^{(\alpha)}(t) = \frac{1}{2\pi i} \int_C e^{\lambda t - \lambda^\alpha} d\lambda$$

via relation

$$p_\alpha(t) = b^{-1/\alpha} g^{(\alpha)}(tb^{-1/\alpha}).$$

Writing the solution of Eq. (14) in the form of Neumann's series and taking into account the basic property of a stable density, we get

$$f_\alpha(t) = \sum_{j=1}^{\infty} c^{j-1} (jb)^{-1/\alpha} g^{(\alpha)}(t(jb)^{-1/\alpha}). \quad (23)$$

Using the convergent series for stable density,

$$g^{(\alpha)}(t) = \alpha \sum_{n=1}^{\infty} \frac{(-1)^{n-1} t^{-\alpha n - 1}}{(n-1)! \Gamma(1 - \alpha n)},$$

and changing the order of summation in the case $c < 1$, we obtain

$$f_\alpha(t) = \alpha \sum_{n=1}^{\infty} \frac{(-1)^{n-1} b^n}{(n-1)! \Gamma(1 - \alpha n)} \Phi(c, -n, 1) t^{-\alpha n - 1}, \quad (24)$$

where

$$\Phi(c, -n, 1) = \sum_{j=1}^{\infty} c^{j-1} j^n.$$

In particular,

$$\Phi(c, -1, 1) = (1 - c)^{-2},$$

$$\Phi(c, -2, 1) = (c + 1)(1 - c)^{-3},$$

$$\Phi(c, -3, 1) = (c^2 + 4c + 1)(1 - c)^{-4},$$

$$\Phi(c, -4, 1) = (c^3 + 11c^2 + 11c + 1)(1 - c)^{-5},$$

$$\Phi(c, -5, 1) = (c^4 + 26c^3 + 66c^2 + 26c + 1)(1 - c)^{-6}.$$

The asymptotic behavior of (24) far away from the origin is governed by the first term of the series

$$f_\alpha^{as}(t) = \alpha A(1 - c)^{-2} t^{-\alpha-1}, \quad c < 1. \tag{25}$$

Evidently, this result is not applicable to the case $c = 1$, which should be treated separately. Setting $c = 1$ in (23) and applying the Euler–Maclaurin summation formula, we obtain

$$f_\alpha(t) = \sum_{j=1}^{\infty} (jb)^{-1/\alpha} g^{(\alpha)}(t(jb)^{-1/\alpha}) = \int_1^{\infty} \psi(x) dx + \frac{1}{2} \psi(1) - \sum_{m=0}^{\infty} \frac{B_{2m}}{(2m)!} \psi^{(2m-1)}(1),$$

where

$$\psi(x) = (xb)^{-1/\alpha} g^{(\alpha)}(t(xb)^{-1/\alpha}) = \alpha \sum_{n=1}^{\infty} \left[\frac{(-1)^{n-1} t^{-\alpha n-1}}{(n-1)! \Gamma(1 - \alpha n)} \right] (bx)^n$$

and B_{2m} are the Bernoulli numbers. The leading asymptotic term is due to the integral

$$f_\alpha(t) \sim \int_1^{\infty} \psi(x) dx = (\alpha/b)t^{\alpha-1} \int_0^{tb^{-1/\alpha}} g^{(\alpha)}(\tau)\tau^{-\alpha} d\tau \tag{26}$$

$$\sim (\alpha/b)t^{\alpha-1} \int_0^{\infty} g^{(\alpha)}(\tau)\tau^{-\alpha} d\tau = \alpha [b\Gamma(1 + \alpha)]^{-1} t^{\alpha-1} = f_\alpha^{as}(t), \quad c = 1.$$

This result can be obtained also immediately from Eq. (21) under condition that $a_0 \equiv 1 - c = 0$:

$$aD^\alpha f_\alpha^{as}(t) = \delta(t), \quad t \geq 0.$$

Acting on the both sides of the equation by the integral fractional operator $I^\alpha \equiv D^{-\alpha}$, we have

$$f_\alpha^{as}(t) = \frac{1}{a_\alpha \gamma(\alpha)} \int_0^{\infty} \frac{\delta(t) dt}{(t - \tau)^{1-\alpha}} = \frac{1}{a\Gamma(\alpha)} t^{\alpha-1}. \tag{27}$$

Substitution (22) into (26) and (20) into (27) shows that results (26) and (27) are identical and we have

$$f_\alpha^{as}(t) \propto \begin{cases} t^{-\alpha-1} & \text{as } c < 1, \\ t^{\alpha-1} & \text{as } c = 1. \end{cases}$$

Numerical calculations show, that, in the case $c < 1$ but c being very closely to 1,

say $c = 0.9999$ or so, the following picture is observed:

$$f_{\alpha}^{as}(t) \propto \begin{cases} t^{\alpha-1} & \text{as } t < t^*, \\ t^{-\alpha-1} & \text{as } t > t^*. \end{cases}$$

It is important to note that the exponents in different regions are not independent:

$$(\alpha - 1) - (-\alpha - 1) = 2\alpha.$$

This effect is observed in experiments (Pfister, 1974, Sharfe, 1973).

5. THREE-POWER APPROXIMATION OF HN PROCESS IN REGION L

One can see in (Nigmatullin, 1997) what kind of difficulties are to be found on the way of direct transformation of HN function (7) into a fractional differential equation for the response function $f_{\alpha}^{\beta}(t)$. The problem arises because of the nonlinear dependence of the left side of equation

$$[a_0 + a(i\omega)^{\alpha}]^{\beta/\alpha} \tilde{f}_{\alpha}^{\beta}(i\omega) = 1$$

on term $(i\omega)^{\alpha}$ being Fourier transform of fractional differential operator D^{α} .

However, there exists a simple way to avoid the trouble, at least for region L . Indeed, the HN function is an empirical one and as a result it is an approximate expression for real dependence. Thus, it is not necessary to work with namely this expression: one can find another approximate formula fitting the same experimental data, for example,

$$[c_0 + c(i\omega)^{\gamma} + d(i\omega)^{\delta} + e(i\omega)^{\varepsilon}] \tilde{f}_{\gamma\delta\varepsilon}(i\omega) = 1. \quad (28)$$

Numerical calculations show that the HN function

$$\tilde{f}_{\alpha}^{\beta}(z) = \frac{1}{[1 + z^{\alpha}]^{\beta/\alpha}} \quad (29)$$

can be replaced by the function

$$\tilde{f}_{\gamma\delta\varepsilon}(z) = \frac{1}{c_0 + cz^{\gamma} + dz^{\delta} + ez^{\varepsilon}}, \quad \gamma < \delta < \varepsilon \quad (30)$$

in the case $\alpha < \beta$. The constants $c_0, c, d, e, \gamma, \delta$, and ε can be determined from the conditions

$$\lim_{z \rightarrow \infty} [\tilde{f}_{\gamma\delta\varepsilon}(z) / \tilde{f}_{\alpha}^{\beta}(z)] = 1$$

and

$$\begin{aligned} \lim_{z \rightarrow 0} [1 - \tilde{f}_{\gamma\delta\varepsilon}(z)] / [1 - \tilde{f}_{\alpha}^{\beta}(z)] &= 1, \\ \tilde{f}_{\gamma\delta\varepsilon}(1) &= \tilde{f}_{\alpha}^{\beta}, \end{aligned}$$

and

$$\tilde{f}'_{\gamma\delta\varepsilon}(1) = \tilde{f}^\beta_\alpha(1).$$

The two first conditions yield $c_0 = e = 1$, $c = \beta/\alpha$, and $\gamma = \alpha$. From the third we have

$$d = 2^{\beta/\alpha} - 2 - \beta/\alpha,$$

and from the fourth

$$\delta = \beta[2^{(\beta-\alpha)} - 2/d].$$

The case $\beta = 2\alpha$ is obtained by the limit transition leads to the exact result

$$\tilde{f}_{\gamma\delta\varepsilon}(z) = \frac{1}{(1 + z^\alpha)^2}.$$

Results of comparative calculations for real and imaginary components of function $\tilde{f}(i\omega)$ plotted in Fig. 2 show that Eq. (30) fits HN function with an acceptable accuracy.

6. RESULTS

Equation (28) represents the differential equation with fractional derivatives

$$[c_0 + cD^\gamma + dD^\delta + eD^\varepsilon]f_{\gamma\delta\varepsilon}(t) = \delta(t),$$

in Fourier space. Taking into account $\gamma < \delta < \varepsilon$, we can write $\tilde{f}_{\gamma\delta\varepsilon}(z)$ in the form (Podlubny, 1994)

$$\begin{aligned} f_{\gamma\delta\varepsilon}(z) &= \frac{1}{ez^\varepsilon + dz^\delta} \frac{1}{1 + \frac{cz^\gamma + c_0}{ez^\varepsilon + dz^\delta}} = \frac{e^{-1}z^{-\delta}}{z^{\varepsilon-\delta} + e^{-1}d} \frac{1}{1 + \frac{e^{-1}cz^\gamma + e^{-1}c_0z^{-\delta}}{z^{\varepsilon-\delta} + e^{-1}d}} = \\ &= \sum_{m=0}^{\infty} (-1)^m \frac{e^{-1}z^{-\delta}}{(z^{\varepsilon-\delta} + e^{-1}d)^{m+1}} \left(\frac{c}{e}z^{\gamma-\delta} + \frac{c_0}{e}z^{-\delta} \right)^m = \sum_{m=0}^{\infty} (-1)^m \frac{e^{-1}z^{-\delta}}{(z^{\varepsilon-\delta} + e^{-1}d)^{m+1}} \\ &= \sum_{k=0}^m \binom{m}{k} \frac{c^k c_0^{m-k}}{e^m} z^{\gamma k - \delta m} = \frac{1}{e} \sum_{m=0}^{\infty} (-1)^m \left(\frac{c_0}{e} \right)^m \sum_{k=0}^m \binom{m}{k} \left(\frac{c}{c_0} \right)^k \frac{z^{\gamma k - \delta m - \delta}}{(z^{\varepsilon-\delta} + e^{-1}d)^{m+1}} \end{aligned}$$

The term-by-term inversion based on the general expansion theorem for the Laplace transform yields the final expression for the version of HN function under consideration

$$\begin{aligned} f_{\gamma\delta\varepsilon}(t) &= \frac{1}{e} \sum_{m=0}^{\infty} \frac{1}{m!} \left(\frac{-c_0}{e} \right)^m \sum_{k=0}^m \binom{m}{k} \left(\frac{c}{c_0} \right)^k t^{\varepsilon(m+1) - \gamma k - 1} E_{\varepsilon-\delta, \varepsilon + \delta m - \gamma k}^{(m)} \\ &\quad \times \left(-\frac{d}{e} t^{\varepsilon-\delta} \right). \end{aligned}$$

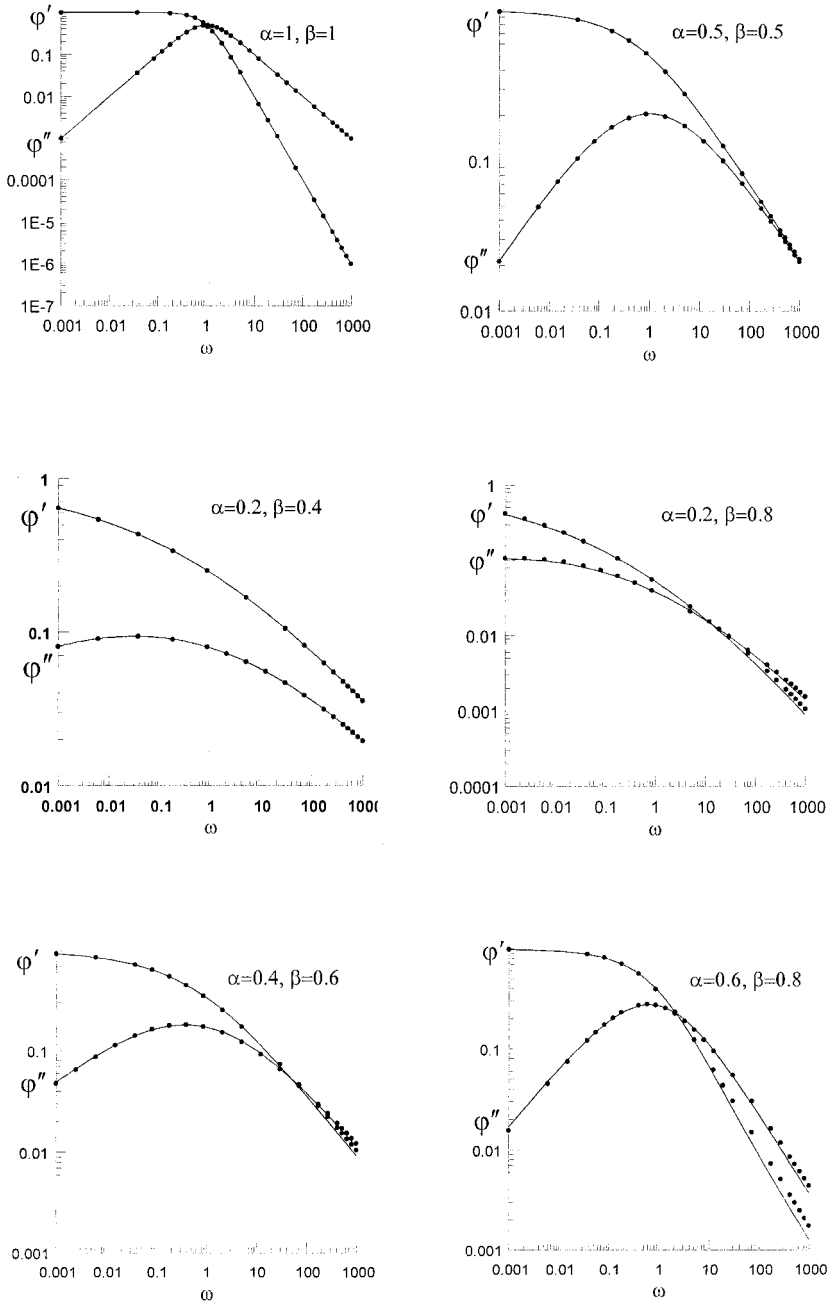


Fig. 2. Comparison approximation (25) of the Havriliak–Negami law (circles) with exact expression (24) (lines).

This is a general solution for all relaxation processes belonging to the region L .

What have we got from the obtained results? The equation with three fractional derivatives of different orders γ , δ , and ε allows us to propose that three different mechanisms are involved in the relaxation process. All of them manifest themselves because of jumps of the observed physical characteristics as quantum mechanics dictates, and the response function is an average function. One can suppose that one of them is the change in orientation of macrodipoles in solids, and the second of this mechanisms is the decay of macrodipole moments in magnitude (this process is absent in Debye relaxation). But what is the third of them? I hope that further investigations on this way can throw light upon this question.

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