The Stochastic Nature of Complexity
Evolution in the Fractional Systems

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Abstract

The stochastic scenario of relaxation in the complex systems is presented. It is based on a general probabilistic formalism of limit theorems. The nonexponential relaxation is shown to result from the asymptotic self-similar properties in the temporal behavior of such systems. This model provides a rigorous justification of the energy criterion introduced by Jonscher. The meaning of the parameters into the empirical response functions is clarified. This treatment sheds a fresh light on the nature of not only the dielectric relaxation but also mechanical, luminescent and radiochemical ones. In the case of the Cole-Cole response there exists a direct link between the notation of the fractional derivative (appearing in the fractional macroscopic equation often proposed) and the model. But the macroscopic response equations, relating to the Cole-Davidson and Havriliak-Negami relaxations, have a more general integro-differential form in comparison with the ordinary fractional one.

Key words:
Lévy-stable distributions, Self-Similarity, Subordination, Fractional differential equation, Mittag-Leffler function

1 Introduction

Experimental investigations surely have established the relaxation response of various complex systems (amorphous semiconductors and insulators, polymers, molecular solid solutions, glasses, etc.) to be non-exponential in nature [1,2]. In particular, all types of the empirical functions used to fit the dielectric data exhibit the fractional-power dependence of the dielectric responses on frequency and time. It is worth noticing that this unique property is independent on any special details of examined systems. In the past decades, a

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considerable attention has been paid to find a theoretical explanation of the experimental results [3,4,5,6]. The main feature of all the dynamical processes in the complex systems is their stochastic background. In this framework one can expect that the macroscopic behavior of the complex systems is governed by “averaging principles” like the law of large numbers to be in force. However, to develop the assumption is enough difficult. The point is that their macroscopic evolution is not attributed to any particular object taken from those forming the system. The problem of constructing an ”averaged” object representing the entire relaxing system is not trivial. The description of the relationship between the local random characteristics of complex systems and the universal deterministic empirical laws is of great importance.

Many-body effects play a vital part in such systems. No wonder that there exists a direct relationship, suggested in literature [4,7,8,9], between anomalous relaxation and anomalous diffusion. One of more convenient languages for the description of anomalous diffusion is the continuous random walk (CTRW) theory. It occupies an important place for studying many physical phenomena. The notation of CTRW was first proposed by Montroll and Weiss in 1965 [10]. With their happy touch the CTRW generalized a simple random walk. Although the term “random walk” was introduced by Pierson in 1905, the formalism of simple random walks was known else in the XVII-th century. The random walk approach is based on the assumption that step changes are made through equal time intervals. This was a first approximation in various physical, chemical and economical models. For its turn the CTRW theory allows a random waiting time among subsequent random jumps. To sum up the long-term studies of this problem, the recent, mathematically excellent works of Meerschaert, Scheffler and Becker-Kern [11] have made its details ultimate clear. On the one hand, this has allowed one to recognize the stochastic processes responsible for the anomalous behavior. On the other hand, the approach proposes a description of the anomalous properties. In fact, it has established a close connection between the stable distributions (from the theory of probability) and the fractional calculus. This means that the nondifferentiable nature of microscopic dynamics of components in the complex systems can be transmitted to the macroscopic description of such systems in the form of fractional operators [12,13]. Consequently, the CTRW method is very popular for physical applications connected with anomalous diffusion, transport in disorder media, superslow relaxation, etc. (see the perfect reviews [14,15] and references therein).

In this paper we suggest the probabilistic approach to the analysis of evolution processes. Our approach is based on the probabilistic formalism of limit theorems which provides tools to relate the local random characteristics of complex systems to the deterministic and universal relaxation laws regardless of the specific nature of the systems considered. We attempt to answer the following key questions related to the temporal evolution of complex systems:
what does mean a self-similarity in the evolution of the complex stochastic systems (Section 2);
what to interpret theoretically the empirical deterministic relaxation laws (Section 3);
what characteristics of the internal structure of the complex systems stand behind the empirical responses (Section 4);
what connection is between the micro/meso/macroscopic dynamics of the relaxing systems and the macroscopic energy criterion (Section 5);
what role of the macroscopic response equations is for the description of the relaxation phenomena (Section 6).

Finally, we discuss some alternative models.

## 2 Self-Similarity of Complex Systems

The simplest traditional interpretation of relaxation phenomena is based on the concept of a system of independent exponentially relaxing objects (for example, dipoles) with different (independent) relaxation rates \[16\]. Since any macroscopic system consists of a finite number of objects, the approach gives a discrete set of relaxation rates. Without a doubt, the assumption may be valid for some relaxing systems. However, neither finite nor infinite exponential series with different real rates and contributions can result in the Havriliak-Negami (HN) and Kohlrausch-Williams-Watts (KWW) response laws exactly. The only opportunity to overcome this problem is a mathematical extension of this model. Really, the transition from the discrete “topology” of the relaxation rate set to the continuous one changes the situation in quality. By the integral transform (resembling the Laplace transform and replacing the exponential series) one can obtain the well-known empirical response functions. In this case the temporary relaxation distribution density is a continuous function and has no any narrow peaks that could be interpreted as a manifestation of separate subsystems (objects).

The relaxation, following the Cole-Cole (CC) law, may be developed in a two-state system. Let \( N \) be the common number of dipoles in a dielectric system. If \( N_\uparrow \) is the number of dipoles in the state \( \uparrow \), \( N_\downarrow \) is the number of dipoles in the state \( \downarrow \) so that \( N = N_\uparrow + N_\downarrow \). Assume that for \( t = 0 \) the system is stated in order so that the states \( \uparrow \) dominate, namely

\[
\frac{N_\uparrow(t=0)}{N} = n_\uparrow(0) = 1, \quad \frac{N_\downarrow(t=0)}{N} = n_\downarrow(0) = 0,
\]

where \( n_\uparrow \) is the part of dipoles in the state \( \uparrow \), \( n_\downarrow \) the part in the state \( \downarrow \). Denote the transition rates by \( w \) defined from microscopic properties of the
system (for instance, according to the given Hamiltonian of interaction and the Fermi’s golden rule). In the case the kinetic equation describing the ordinary relaxation (Debye law) takes the form

\[
\begin{align*}
\dot{n}_\uparrow(t) - w \{n_\uparrow(t) - n_\downarrow(t)\} &= 0, \\
\dot{n}_\downarrow(t) - w \{n_\uparrow(t) - n_\downarrow(t)\} &= 0,
\end{align*}
\]

(1)

where, as usual, the dotted symbol means the first-order derivative. The steady state of the system corresponds to equilibrium with \(n_\uparrow(\infty) = n_\downarrow(\infty) = 1/2\).

Clearly its response has an exponential character. However, this happens to be the case for such dipoles that relax irrespective of each other and of their environment. If the dipoles interact with their environment, and the interaction is complex (or random), their behavior already will not be exponential.

Assume that the interaction of dipoles with environment is taken into account with the aid of the temporal subordination. Recall that in the theory of anomalous diffusion the notation of subordination occupies one of the most important places. So, a subordinated process \(Y(U(t))\) is obtained by randomizing the time clock of a random process \(Y(t)\) by means of a random process \(U(t)\) called the directing process. The latter process is also often referred to as the randomized time or operational time [17]. Generally speaking, the process \(Y\) may be both random and deterministic in nature. The anomalous diffusion theory studies, as a rule, the subordination of random processes. We intend to extend this approach to relaxation processes.

Let the time variable be a sum of random temporal intervals \(T_i\) being non-negative independent and identically distributed so that the waiting times \(T_i\) belong to an \(\alpha\)-stable distribution (\(0 < \alpha < 1\)). Then their sum \(n^{-1/\alpha}(T_1 + T_2 + \cdots + T_n)\), \(n \in \mathbb{N}\) converges in distribution to a stable law with the same index \(\alpha\) [11]. To determine a walker position at the true time \(t\), one needs to find the number of jumps up to time \(t\). This discrete counting process is \(\{N_t\}_{t \geq 0} = \max\{n \in \mathbb{N} \mid \sum_{i=1}^{n} T_i \leq t\}\). Denote the continuous limit of \(\{N_t\}_{t \geq 0}\) by \(S(t)\). For a fixed time it represents the first passage of the stochastic time evolution above that time level. The random process is nondecreasing, and it can be chosen as a new time clock (stochastic time arrow) [18]. The probability density of the process \(S(t)\) has the following Laplace image

\[
p^S(t, \tau) = \frac{1}{2\pi j} \int_{Br} e^{u\tau} u^{\alpha-1} du = t^{-\alpha} F_\alpha(\tau/t^\alpha),
\]

(2)

where \(Br\) denotes the Bromwich path. This probability density has a simple physical interpretation. It determines the probability to be at the internal time (or so-called operational time) \(\tau\) on the real (physical) time \(t\) [18]. The function \(F_\alpha(z)\) can be expanded as a Taylor series. Besides, it has the Fox’
H-function representation

\[ F_\alpha(z) = H_{11}^{10} \left( z \left| \begin{array}{c} (1 - \alpha, \alpha) \\ (0, 1) \end{array} \right. \right) = \sum_{k=0}^{\infty} \frac{(-z)^k}{k! \Gamma(1 - \alpha(1 + k))}, \]

where \( \Gamma(x) \) is the ordinary gamma function. In the theory of anomalous diffusion the random process \( S(t) \) is applied for the subordination of Lévy (or Gaussian) random processes [11,19]. The inverse Lévy process \( S(t) \) accounts for the amount of time that a walker does not participate in the motion process [20]. If the walker participates all time in the motion process, the internal time and the physical (external) time would coincide.

As was shown in [18], the stochastic time arrow can be applied to the general kinetic equation. Then the equation describing a two-state system takes the following form

\[
\begin{align*}
\{ \hat{D}^\alpha n_\uparrow(t) - w \{ n_\downarrow(t) - n_\uparrow(t) \} \} &= 0, \\
\hat{D}^\alpha n_\downarrow(t) - w \{ n_\uparrow(t) - n_\downarrow(t) \} &= 0,
\end{align*}
\]

where \( \hat{D}^\alpha \) is the \( \alpha \)-order fractional derivative with respect to time. Here we use the Caputo derivative [21,22], namely

\[
\hat{D}^\alpha x(t) = \frac{1}{\Gamma(n - \alpha)} \int_0^t \frac{x^{(n)}(\tau)}{(t - \tau)^{\alpha+1-n}} d\tau, \quad n - 1 < \alpha < n,
\]

where \( x^{(n)}(t) = D^n x(t) \) means the \( n \)-derivative of \( x(t) \). The relaxation function for the two-state system is written as

\[
\phi_{CC}(t) = 1 - 2n_\uparrow(t) = 2n_\downarrow(t) - 1 = E_\alpha(-2wt^{\alpha}),
\]

where \( E_\alpha(z) = \sum_{n=0}^{\infty} z^n / \Gamma(1+n\alpha) \) is the one-parameter Mittag-Leffler function [23]. Feller conjectured and Pollard proved in 1948 that the Mittag-Leffler function \( E_\alpha(-t) \) is completely monotonic for \( t \geq 0 \), if \( 0 < \alpha \leq 1 \). Moreover, \( E_\alpha(-t) \) is an entire function of order \( 1/\alpha \) for \( \alpha > 0 \) [17]. It should be pointed out that the relaxation function under interest corresponds to the CC law.

The analysis of this problem will not be complete, unless one consider it on the other hand. It turns out that the same result mentioned above can be also obtained by another way [6,24,25]. If the relaxation rate of the \( i \)-th dipole is equal to the value \( b \), then the probability, that this dipole did not change its initial orientation prior to an instant \( t \), is

\[
Pr \left( \theta_i \geq t \mid \beta_i = b \right) = \exp(-bt) \quad \text{for } t \geq 0, \ b > 0.
\]
The random variable $\beta_i$ denotes the relaxation rate of the $i$-th dipole and the variable $\theta_i$, the time needed for changing its initial orientation. Let $\{\beta_i\}$ and $\{\theta_i\}$ form sequences of nonnegative independent identically distributed random variables. Following [26] and the law of total probability, one define the relaxation function $\phi_i(t)$ for $i$-th dipole as a probability:

$$\phi_i(t) = \Pr (\theta_i \geq t) = \int_0^\infty \exp(-bt) dH_{\beta_i}(b), \quad (5)$$

where $H_{\beta_i}$ is the distribution function of each relaxation rate $\beta_i$. The form of a suitable function $H_{\beta_i}$ should be found. In the system consisting of a large number $N$ of relaxing dipoles, the relaxation function $\phi(t)$ has to express in terms of the probability that the entire system will be without changing its initial state until $t$:

$$\phi(t) = \lim_{N \to \infty} \Pr (A_N \min(\theta_1, \ldots, \theta_N) \geq t), \quad (6)$$

where $A_N$ is a normalizing constant. Let us observe that the expression (5) is the Laplace transform of the distribution function $H_{\beta_i}(b)$:

$$\Pr (\theta_i \geq t) = \mathcal{L}(H_{\beta_i}; t).$$

Because of $\theta_i$ being independent, we get

$$\Pr \left( \min(\theta_1, \ldots, \theta_N) \geq \frac{t}{A_N} \right) = \left( \Pr (\theta_i \geq \frac{t}{A_N}) \right)^N = \left( \mathcal{L}(H_{\beta_i}; \frac{t}{A_N}) \right)^N.$$

When $N$ tends to infinity, the $N$-th power of the Laplace transform of the non-degenerate distribution function $H_{\beta_i}$ converges to a non-degenerate limiting transform, if and only if $H_{\beta_i}$ belongs to the domain of attraction of the Lévy-stable law [17,26,27].

In fact, the above limiting form is only determined by the behavior of the tail of $H_{\beta_i}(b)$ for large $b$, i. e. by asymptotic properties of $H_{\beta_i}(b)$. The detailed knowledge of its other properties is not necessary. It is enough that the distribution function $H_{\beta_i}$ belongs to a domain of attraction of the Lévy-stable law with the index of stability $\alpha$. On the other words [17], the necessary and sufficient condition for any $x > 0$ is

$$\lim_{b \to \infty} \frac{1 - H_{\beta_i}(xb)}{1 - H_{\beta_i}(b)} = x^{-\alpha}. \quad (7)$$
This condition can be interpreted as a type of self-similarity. Really, for any \( x > 0 \) and for large \( b \)
\[
\Pr (\beta_i > xb) \approx x^{-\alpha} \Pr (\beta_i > b).
\] (8)

It is that the self-similarity is suggested as a fundamental feature of relaxation phenomena [7,28,29]. It should be stressed here that in the approach this conclusion arises from the pure probabilistic analysis, independently of the physical details of dipolar systems. Thus, it can be carried over to other similar cases of complex systems.

The randomness of the relaxation rates \( \beta_i \) \((1 \leq i \leq N)\) is motivated by the fact that in the complex systems an object has not the only equilibrium state, but their states form a whole set of metastable substates. Their configuration changes in a very complicated way during their evolution. Each of the objects is locked into a substate, and the distribution of relaxation rates rejects any deterministic behavior of an individual object in the complex system. The total survival probability of the whole system has formally the same form
\[
\Pr (\theta_i \geq t) = \langle \exp(-\beta_i t) \rangle = \int_0^\infty \exp(-bt) dH_\alpha(b) \] (9)

like (5). However, now the form of d.f. \( H_\alpha \) is strictly fixed (i.e. it adheres to the Lévy-stable law), and the information about the distribution functions \( H_{\beta_i} \) is concentrated in the index \( \alpha \) of the stable law.

3 Probabilistic Interpretation of Empirical Laws

Since the relaxation rate \( b \) cannot be negative, the Lévy-stable laws are completely asymmetric (supported on the nonnegative half-line) with \( 0 < \alpha < 1 \). In this case the relaxation function (6) with \( A_N = N^{1/\alpha} \) is well defined and takes the KWW form
\[
\phi_{\text{KWW}}(t) = \lim_{N \to \infty} \left( \mathcal{L}(H_{\beta_i}; \frac{t}{N^{1/\alpha}}) \right)^N = \exp\left(\frac{-A}{\alpha} t\right),
\] (10)

where \( A \) is a positive constant [30,31]. If \( \alpha = 1 \), the relaxation function (10) becomes \( \phi_D(t) = \exp(-t/\tau_D) \) (Debye form), where \( \tau_D = A^{-1} \) is the D relaxation time. Mathematically, this case corresponds to the degenerate case in (9). For any fixed (deterministic) constant \( A \) we obtain the only expression
In general, the feature of $A$ is not necessarily true. It is therefore reasonable to ask what will be, if the constant becomes random. We will get the other relaxation laws.

To find the “scenario” leading to the observable relaxation laws different from the KWW form, let us note of the fact that the relaxation function for the CC response can write in the form

$$\phi_{\text{CC}}(t) = \int_0^\infty \exp(- (t/\lambda)^a) \, dT_a(\lambda/\tau_{\text{CC}}) = E_a(-(t/\tau_{\text{CC}})^a), \quad (11)$$

where $E_a(z)$ is the one-parameter Mittag-Leffler function, $T_a(\lambda/\tau_{\text{CC}})$ the one-sided Lévy-stable probability distribution with the index $0 < a \leq 1$, and $\tau_{\text{CC}}$ is constant. It is useful to recall that from the subordination approach the CC relaxation response is expressed as

$$\phi_{\text{CC}}(t) = \int_0^\infty \phi_D(\tau) \, p^S(t, \tau) \, d\tau = \int_0^\infty \exp(-\mu \tau) \, p^S(t, \tau) \, d\tau = E_a(-\mu t^a). \quad (12)$$

Here the parameter $\mu$ is constant, and the contribution of irregular changing dipole orientations in the macroscopic evolution of the system is derived from the probability density of the directing process $S(t)$. The approach of Weron and Jurlewich [24,25] is based on the other conception. It is that each individual dipole in a complex system relaxes exponentially, but their relaxation rates are different and obey a probability distribution (continuous function). However, the subordination approach brings advantages in deriving a rather simple macroscopic equation for the description of the CC relaxation response.

The result (11) may be interpreted as a weighted average (or as randomizing the parameter $\lambda$) of the stretched exponential relaxation (10) respect to the distribution function $T_a(\lambda)$ of the scale parameter $\lambda$. This idea works not only for the CC relaxation. Really, let $Q_a$ be such a random value that its Laplace transform is the stretched exponential function

$$\langle e^{-s Q_a} \rangle = \int_0^\infty \exp(-st) \, h_a(t) \, dt, \quad 0 < a \leq 1. \quad (13)$$

Then the random value $Q_a$ is distributed according to the one-side Lévy-stable law with the probability distribution function $h_a(t)$ with $0 < a < 1$ (see details, for example, [31]). Now let the random value $G_b$ be independent of $Q_a$ and distributed according to the gamma law [32] defined by the probability
distribution function

\[ g_b(t) = \frac{1}{\Gamma(b)} t^{b-1} e^{-t}, \quad b > 0, \ t > 0. \]

In this connection it should be pointed out that the Laplace transform of \( G_b \) takes the form

\[ \langle e^{-s G_a} \rangle = \int_0^\infty \exp(-st) g_b(t) \, dt = \frac{1}{(1 + s)^b}. \]  

(14)

For the random value \( B Q_a (G_b)^{1/b} \) one obtains

\[ \langle e^{-s B Q_a G_a} \rangle = \left( \int_0^\infty \exp\left(-Bst^{1/a}\right) h_a(s) \, ds \right) g_b(t) \, dt \]

\[ = \int_0^\infty \exp\left(-s(B)^a t\right) g_b(t) \, dt = \frac{1}{(1 + (Bs)^a)^b}, \]

(15)

where the positive (arbitrary) constant \( B \) is a scale parameter. The frequency-domain response \( \phi^*(\omega) \) is related to the relaxation function \( \phi(t) \) by the one-sided Fourier transform:

\[ \phi^*(\omega) = \int_0^\infty e^{i\omega t} \left( -\frac{d\phi(t)}{dt} \right) \, dt. \]

(16)

As it is well known [7], the (dielectric) susceptibility \( \chi(\omega) \) is directly connected with \( \phi^*(\omega) \) by the formula:

\[ \phi^*(\omega) = \frac{\chi(\omega) - \chi_\infty}{\chi_0 - \chi_\infty}, \]

where the constant \( \chi_\infty \) represents the asymptotic value of \( \chi(\omega) \), and \( \chi_0 \) is the value of the opposite limit. Clearly, the process \( B Q_a (G_b)^{1/b} \) leads to the following time-frequency response

\[ \phi_{HN}^*(\omega) = \frac{1}{(1 + (iB\omega)^a)^b}. \]

(17)

This is just the HN relaxation response. Evidently, for \( b = 1 \) the expression corresponds to the Cole-Davidson (CD) empirical law. When the random value \((G_1)^{1/c}\) follows the Weibull distribution [32] with the shape parameter equal to
c, we arrive at the CC relaxation. Thus, the KWW, CC, CD and HN relaxation functions are very close in connection from the probabilistic point of view to the random processes associated with the relaxation.

In this connection it should be pointed out that the evolution of \( n_\uparrow(t) \) and \( n_\downarrow \) in Eq. (2) can be connected with the Mittag-Leffler distribution. Let \( Z_n \) denote the sum of \( n \) independent random values with the Mittag-Leffler distribution. Then the Laplace transform of \( n^{-1/\alpha}Z_n \) is \( (1+s^\alpha/n)^{-n} \), which tends to \( e^{-s^\alpha} \) as \( n \) tends to infinity. Following Pillai [33], this demonstrates an infinity divisibility of the Mittag-Leffler distribution. By virtue of the power asymptotic form (long tail) the distribution with parameter \( \alpha \) is attracted to the stable distribution with exponent \( \alpha \), \( 0 < \alpha < 1 \). The property of the Mittag-Leffler distribution allows one to develop a corresponding stochastic process. The process (called Mittag-Leffler’s) arises of subordinating a stable process by a directing gamma process [33]. In this case the relaxation function has the Havriliak-Negami form

\[
\phi_{HN}(t) = 1 - \sum_{k=0}^{\infty} \frac{(-1)^k \Gamma(b+k)}{k! \Gamma(b) \Gamma(1+ab+ak)} \left( \frac{t}{\tau_{HN}} \right)^{ab+ak}, \tag{18}
\]

where \( a, b, \tau_{HN} \) are constant. The one-side Fourier transformation of the relaxation function gives

\[
\phi_{HN}^*(\omega) = \int_0^\infty e^{-i\omega t} \left( -\frac{d\phi_{HN}(t)}{dt} \right) dt = \frac{1}{(1+(i\omega\tau_{HN})^b).} \tag{19}
\]

This result also corresponds to the well-know HN empirical law. Thus, the HN relaxation can be explained from the subordination approach, if the hitting time process of dipole orientations transforms into the Mittag-Leffler process. For that the hitting time process has an appropriate distribution attracted to the stable distribution. The subordination of the latter results just in the Mittag-Leffler process. It is interesting to observe that the Lévy process subordinated by another Lévy one leads again to the Lévy process, but with other index [34]. Observe that in this point the subordination approach is almost equivalent to the approach studied in [24].

4 Internal Structure of Complex Systems

In any dielectric (complex) system under an week external electric field (external action) only a part (active dipoles or objects) of the total number \( N \) of dipoles is directly governed by changes of the field. But even those dipoles,
not contributing to the relaxation dynamics, can have an effect on the behavior of active dipoles. This means that the $i$-the active dipole interacts with $N_i - 1$ inactive neighbors forming a cluster of size $N_i$. The number $K_N$ of active dipoles in the system is equal to the number of clusters. The sum of the clusters exceeds $N$, the size of the system. Because of the screening effects the active dipoles can “see” only some of their active neighbors. If so, the cooperative regions built upon the active dipoles will appear. The number of the such mesoscopic regions is determined by their sizes $M_1, M_2, \ldots$. The contribution of each region to the total relaxation rate is a sum of the contributions of all active dipoles over the region. Generally speaking, the sums are random. Hence, the $j$-th region has its relaxation rate, say $\beta_{jN}$, equal to

$$\overline{\beta_{jN}} = \sum_{i=M_1+\ldots+M_{j-1}+1}^{M_1+\ldots+M_j} \beta_{jN}.$$  

For $j = 1$ the latter expression is simply the sum $\overline{\beta_{1N}} = \sum_{i=1}^{M_1} \beta_{iN}$. Next for $j = 2$ it takes the form $\overline{\beta_{2N}} = \sum_{i=M_1+1}^{M_1+M_2} \beta_{iN}$ and so on. The relaxation function of the whole system

$$\phi(t) = \langle e^{-t\overline{\beta_N}} \rangle$$

is provided by the total relaxation rate $\overline{\beta_N}$ as the sum of the contributions over all cooperative regions:

$$\overline{\beta_N} = \sum_{j=1}^{L_N} \overline{\beta_{jN}}.$$  

As a rule, the relaxing systems consist of a large number of dipoles so that the limit transition $\overline{\beta} = \lim_{N \to \infty} \overline{\beta_N}$ is valid (in practice, $N \approx 10^5$ and more is enough). Limit theorems for the random sums have been recently established in [35].

The number of dipoles directly engaged in the relaxation process is random as well as their locations are random too. Obviously, all the quantities $N_i$’s, $M_j$’s, $\beta_{iN}$’s and those defined by them, are random values. Their stochastic characteristics determine the total relaxation rate $\overline{\beta_N}$, but they are not known. Nevertheless, on the basis of the limit theorems of probability theory, the distribution of the limit $\overline{\beta}$ (for the large relaxing systems) can be defined, even with rather information about the distributions of micro/mesoscopic quantities.

In the approach it is quite enough to consider stochastically independent sequences of random values $N_i$’s, $M_j$’s, $\beta_{iN}$’s. Each sequence consists of indepen-
dent and identically distributed nonnegative random values that have either finite expected value finite or long-tailed distribution. Then the total relaxation rate \( \tilde{\beta} \) takes the form corresponding to one of the empirical responses (see Section 3). It should be noted that the distribution of a nonnegative random value, say \( X \), has a long tail, if and only if the tail \( \Pr(X > x) \) fulfills the condition
\[
\lim_{x \to \infty} \frac{\Pr(X > x)}{x^{-\gamma}} = \text{const} > 0
\]
for some \( 0 < \gamma < 1 \) so that for value \( x \) the tail exhibits the fractional power law \( x^{-\gamma} \) \cite{17,20}. Many different continuous and discrete distributions are well known to satisfy the condition \( 20 \). Classical examples are completely asymmetric Lévy-stable laws as well as the Pareto and Burr distributions with an appropriate choice of their parameters \cite{17}. To obtain the discrete distributions with long tails, one should apply a quantization procedure to the above continuous examples \cite{26}. If the distribution of random value \( X \) has long tail, then the expected value \( \langle X \rangle \) is infinite. The finiteness of the expected value and long-tail property \( 20 \) can be presented only on different levels (theirs are three: an active dipole \( \to \) a cluster \( \Rightarrow \) a cooperative region) of the complex system. To sum up, Table II shows the connection between the internal properties of complex system’s dynamics and the empirical relaxation responses, as well as the physical sense of the parameters characterizing the responses. The proposed approach leads to a very general scenario of relaxation, from the stochastic nature of microscopic dynamics through the hierarchical structure of parallel multi-channel processes to the deterministic macroscopic laws of relaxation given by \( 10 \) and \( 17 \).

5 Energy Criterion

The common property of the empirical relaxation laws is that they exhibit the high-frequency power law in the susceptibility:
\[
\chi(\omega) \propto \left( \frac{i\omega}{\omega_p} \right)^n \quad \text{for } \omega \gg \omega_p,
\]
where the exponent \( n \) falls in range \((0,1)\) and the constant \( \omega_p \) is the loss peak frequency. As a consequence, for large \( \omega \) the ratio of the imaginary to real components of the susceptibility \( \chi(\omega) = \chi'(\omega) - i\chi''(\omega) \) becomes a constant of degree \( n \):
\[
\frac{\chi''(\omega)}{\chi'(\omega)} = \cot \left( n \frac{\pi}{2} \right) \quad \text{for } \omega \gg \omega_p.
\]
Table 1
The connection between the internal properties of complex systems and their relaxation response (the notations of the column “Parameters” correspond to (10) and (17); the constant $\gamma$ according to (20)).

| Law | Parameters | $N_i$ | $M_j$ | $\beta_{iN}$ |
|-----|------------|------|------|--------------|
| D   | $a = 1$    | $b = 1$ | $\langle N_i \rangle < \infty$ | $\langle M_j \rangle < \infty$ | $\langle \beta_{iN} \rangle < \infty$ |
| KWW | $0 < \alpha < 1$ | $\langle N_i \rangle < \infty$ | $\langle M_j \rangle < \infty$ | long tail | $\gamma = \alpha$ |
| CD  | $a = 1$    | $0 < b < 1$ | long tail | $\gamma = b$ | $\langle \beta_{iN} \rangle < \infty$ |
| CC  | $0 < a < 1$ | $b = 1$ | long tail | $\gamma = a$ | long tail |
| HN  | $0 < a < 1$ | $0 < b < 1$ | long tail | $\gamma = a$ | long tail |

However, the D response has not the property. It should be noted the physical significance of expression (21). At high frequencies the ration of the macroscopic energy lost per radian to the energy stored at the peak is independent of frequency.

Jonscher [2] has advanced a hypothesis that the fact is based on the identical property of individual structural elements of the systems. This explains the universality in the large scale behavior of complex systems, but needs for the
precise derivation. In the framework of the proposed and mentioned-above model the physical intuition can be strictly argumentative. Really, the condition \( \beta \) applied to any relaxation rate \( \beta \) leads to the scaling property of the relaxation-rate distribution at large \( b \) (see also [3]). The asymptotic behavior of the distribution is connected with the short-time asymptotic properties of the associated relaxation function \( \phi(t) \), and the response function as its derivative \( f(t) = -d\phi(t)/dt \) takes the form

\[
 f(t) \propto t^{\gamma-1} U(t)
\]

for \( t \to 0 \), where \( U(t) \) is a slowly varying function so that \( \lim_{t \to 0} U(ct)/U(t) = 1 \) for any constant \( c > 0 \). It may be easily verified that the short-time behavior of \( f(t) \) corresponds to the high-frequency properties of the susceptibility \( \chi(\omega) \):

\[
 \chi(\omega) = \chi'(\omega) - i\chi''(\omega) \propto (i\omega)^{-\gamma} U(1/\omega).
\]

The result yields straightforwardly the energy criterion (21) with \( n = 1 - \gamma \). The long-tail property of micro/meso/macroscopic relaxation rate with the parameter \( \gamma \) leads to micro/meso/macroscopic energy criterion with the characteristic constant \( 1 - \gamma \). The analysis of the model shows [25] that in the HN, CC and KWW responses the energy criterion is the case for all micro/meso/macro levels, and the constant \( n \) for the HN case is defined not only the long-tail property of the distribution of cluster sizes, but one of cooperative-region sizes. In the CD case the microscopic energy criterion is absent. The high-frequency power law of this response results only from the long-tail property of the distribution of cooperative-region sizes.

6 Macroscopic Response Equations

It is well known (see e. g. [36]) that the relaxation function \( \phi(t) \) has to fulfil the two-state master equation

\[
 \frac{d\phi(t)}{dt} = -r(t) \phi(t), \quad \phi(0) = 1,
\]

where the nonnegative, time-dependent value \( r(t) \) is the transition rate of the relaxing system (i. e. the probability of transition per unit time). This is a macroscopic deterministic equation. Although the equation does not contain any (for example, micro/mesoscopic and so on) details about relaxation processes, it is convenient for practical purpose because of its simplicity. So, in the case of the KWW relaxation \( r(t) = a A^{t^{a-1}} \). For the D relaxation the
equation (22) has the simplest form \( r(t) = r_0 = \text{const}. \) Then, both relaxation function \( \phi(t) \) and response function \( f(t) = -d\phi(t)/dt \) satisfy the same equation, and their expressions coincide. However, this equivalence is wrong for nonexponential relaxations. Following the old notation ([16], p. 8), the response function is a pulse-response function of the polarization. Any relaxation function or “decay function of the polarization” (by definition, see [16]) tends to 1 for \( t \to 0 \). But some response functions have a singularity in zero, as appears, for example, from the CD response function. In general, the response function \( f(t) \) can be a solution of the first-order differential equation with variable coefficients:

\[
\left[ r^2(t) - \frac{dr(t)}{dt} \right] f(t) + r(t) \frac{f(t)}{dt} = 0. \tag{23}
\]

When the relaxation obeys the CC, CD, HN laws, the equations (22) and (23) are not just simple because of a sufficiently complicated expression for \( r(t) \). It may be attempted to transform (22) and (23) in an integro-differential form simpler than the input ones. Really, the way gives some progress. The CC relaxation and response functions can be expressed as a solution of the fractional differential equation.

In this case we have

\[
\phi_{\text{CC}}(t) = E_a(- (t/\tau)^a),
\]

and the response function is written in the following series representation

\[
f_{\text{CC}}(t) = \sum_{k=0}^{\infty} \frac{(-1)^k(t/\tau)^{a(1-k)-1}}{\tau^a \Gamma[a(1+k)]}.
\]

According to the book of Miller and Ross [37], the one-parameter Mittag-Leffler function \( E_a(-(t/\tau)^a) \) satisfies the identity

\[
J^{1-a}[D E_a(-(t/\tau)^a)] = -\frac{1}{\tau^a} E_a(-(t/\tau)^a),
\]

where \( D \) denotes the usual differentiation operator \( d/dt \), and the operator \( J^{1-a} \) is the Riemann-Liouville fractional integral having the form

\[
J^{\nu}x(t) = \frac{1}{\Gamma(\nu)} \int_0^t (t-s)^{\nu-1} x(s) \, ds. \tag{24}
\]
From this it follows that for the CC relaxation the relaxation function fulfills
\[ J^{1-a}[D \phi_{CC}(t)] = -\frac{1}{\tau^a} \phi_{CC}(t), \]  
(25)
and the response function does
\[ D[J^{1-a}f_{CC}(t)] = -\frac{1}{\tau^a} f_{CC}(t). \]  
(26)

In this connection it should be pointed out that equation (25) is expressed in terms of the fractional integral of the ordinary derivative, and (26) is in terms of the ordinary derivative of the fractional integral. The equations (25) and (26) are fully equivalent to (22) and (23) with the transition rate
\[ r_{CC}(t) = -[E_a(-(t/\tau)^a)]^{-1}D[E_a(-(t/\tau)^a)]. \]

In fact, this is only another formulation of (22) and (23), in terms of the Green function (see more details, for example, in [38]).

With macroscopic equations for the CD and HN responses the situation is more intricate. From the above consideration it is seen that the responses are results of the (not simple) integral transformations. This means that probably, the CD and HN responses satisfy enough complicated macroscopic equations. Therefore, it should be given consideration.

An interesting idea was suggested in [5]. It proceeds from the fact that the ordinary equation of exponential relaxation can be written in the form
\[ \exp(-\Omega_0 t) \frac{d}{dt} \exp(\Omega_0 t)f_D(t) = 0, \]
where \( \Omega_0 \) is constant. By the direct substitution [5] it is easily verified that the CD response function is a solution the following equation
\[ \exp(-\Omega_0 t)D^{\nu}[\exp(\Omega_0 t)f_{CD}(t)] = 0. \]  
(27)

Here the fractional derivative
\[ D^{\nu}[x(t)] = 1/\Gamma(1-\nu) \frac{d}{dt} \int_0^t (t-s)^{-\nu} x(\tau) d\tau \]
is defined as well as in [5]. Next, Eq. (27) can be again generalized, namely
\[ \exp(-\Omega_0 t^\mu)D^{\nu}[\exp(\Omega_0 t^\mu)f_{gen}(t)] = 0. \]  
(28)
Now its solution
\[ f_{\text{gen}}(t) \sim t^{\nu - 1}\exp(-\Omega_0 t^\mu), \]
describes both CD ($\mu = 1$ and $0 < \nu < 1$) and KWW ($0 < \mu = \nu < 1$) response functions.

In [5] it is proved that the macroscopic equation like $(D^\varepsilon + \Omega)^{\alpha/\varepsilon}f(x) = 0$ is well for the HN response. Their conclusion is based on the proof of the operator relation (Appendix A)
\[ \exp(-\Omega u D^{1-\varepsilon}) D^\alpha \exp(\Omega u D^{1-\varepsilon}) = (D^\varepsilon + \Omega)^{\alpha/\varepsilon}, \quad 0 < \varepsilon \leq 1, \quad \alpha \leq \varepsilon, \]
where $u$ is a variable, and $\Omega$ a constant (all the notations follow [5]). However, the commutator (A2) in [5] does not hold true for any continuous functions. In particular, by direct calculations one can found
\[ [D^\alpha, \Omega t D^{1-\varepsilon}] t^{\alpha - 1}\Gamma(\alpha) = \Omega (\alpha + \varepsilon - 1) \frac{t^{\varepsilon - 1}}{\Gamma(\varepsilon)} \]
rather than $\alpha \Omega t^{\varepsilon - 1}/\Gamma(\varepsilon)$ that it follows from [5]. The cause lies in vanishing the term $D^\alpha t^{\alpha - 1} = 0$. On the other hand, if in time domain one expands the Havriluk-Negami (Cole-Cole) response function as an infinite power series, its first term is just $t^{\alpha - 1}/\Gamma(\alpha)$ (in notation of [5]). Thus the expression (A5) is invalid for the HN response. The commutator (A2) remains true only for the case, when $\varepsilon = 1$ (CD relaxation). The expression (A5) for the Cole-Davidson case can be derived directly by the Leibniz’s formula for fractional derivatives without the operator identity.

In this connection it should also be recalled about difficulties with the interpretation of fractional operators in terms of the fractal Cantor set. Following [5,39], the memory function is represented by a Cantor fractal function. However, such a memory function possesses only a power-like property asymptotically, and the approach itself requires else some average procedure for the log-periodic term appearing together with the power [40]. The formalism [39] is exactly macroscopic, but not stochastic. The main feature of the complex system evolution lies just in a stochastic background of dynamical processes. In the framework of the approach [39], to find the relationship between the local random characteristics of complex systems and the universal deterministic empirical laws is hardly possible.
7 Concluding Remarks

In this paper we have shown the outlook of the probabilistic approach proposed to the analysis of relaxation phenomena in the complex systems. The approach permits ones to consider the observable relaxation law on the unique theoretical base, the limits theorem of probability theory. The general probabilistic formalism treats the relaxation of the complex systems regardless of the precise nature of local interactions. In a natural way, it gives an efficient method for calculating the dynamical evaluating averages of the relaxation processes. We have obtained all (known up to now) the empirical relaxation laws, characterized their parameters, connected the parameters with local random characteristics of the relaxation processes, reconstructed the internal structure of relaxing systems, justified the energy criterion, demonstrated the transition from the analysis of the microscopic random dynamics in the systems to the macroscopic deterministic description by integro-differential equations. As a rule, the classical methods of statistical physics take into account the Central Limit Theorem in respect to the probability distributions having finite invariance. However, the assumption does not help to clarify the nature of relaxation phenomena. The above approach has the advantage over the traditional models and goes behind the classical statistical physics. The preliminary results presented in this paper are promising and give a confidence for a fundamental understanding of the relaxation processes in the framework.

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