Vol. 37 (2006) ACTA PHYSICA POLONICA B No 5

ANOMALOUS DIFFUSION SCHEMES UNDERLYING THE STRETCHED EXPONENTIAL RELAXATION. THE ROLE OF SUBORDINATORS *

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(Received February 24, 2006)

The random-variable formalism of anomalous diffusion processes is presented. We elucidate the role of the subordinate stochastic processes as the main mathematical tool that allows us to modify the dynamics of the classical, exponential relaxation process. In particular, we discuss the anomalous diffusion schemes underlying the stretched exponential decay of modes.

PACS numbers: 02.50.–r, 05.40.Fb, 61.20.Lc

1. Introduction

Beginning with stochastic formulation [1] of transport phenomena in terms of a continuous-time random-walk (CTRW), the physical community showed a steady interest in the anomalous diffusion, *i.e.*, a diffusion that appears in absence of the second/first moments of the spatio-temporal random-walk jump parameters and with scaling different than that of the classical Gaussian diffusion. Such an attempt to transport phenomena was

* Presented at the XVIII Marian Smoluchowski Symposium on Statistical Physics, Zakopane, Poland, September 3–6, 2005.

fundamental for understanding of the diffusive behaviour of many complex systems, see *e.g.* [2–14]. The successful applications of the anomalous diffusion ideology yielded a development in mathematical techniques of analysis of the properties of the CTRW propagators (*i.e.*, of the diffusion fronts). Usually, following the original concept of Montroll and Weiss [1], the analysis is based on a formal expression for the Fourier–Laplace transform of the asymptotic distribution of the random position of a walker. In this case, the usual explicit formulas are provided only under some restrictive assumptions on the spatio-temporal coupling properties of the CTRW. Otherwise, as a legitimate tool, use of the fractional calculus is required [15–17].

In this paper we present an approach to the random-walk analysis which is based on the definition of the cumulative stochastic process [18]. We demonstrate the power of the random-variable formalism, related directly to limit theorems of probability theory [19], by showing how it can be generalized to handle different diffusive situations in complex systems. Our effort is directed toward bringing into light all stochastic conditions underlying the well-known time-domain stretched exponential relaxation process. We provide a clear random-walk scheme and rigorous analysis of the anomalous diffusion. We also emphasize the possibilities of application of that scenario in stochastic modeling of the nonexponential relaxation phenomena. The proposed approach may serve as a basis for a model which in the framework of the CTRW can lead to the frequency-domain Havriliak–Negami relaxation response.

2. Subordinate processes and continuous-time random-walk

The basic element in construction of subordinate processes is the nonnegative, strictly increasing stochastic process V_t called *subordinator*. If $X(\tau)$ is a Markov process independent of V_t, then $X(V_t)$ is referred to as the subordinate process with the directing process $X(\tau)$ and the subordinator V_t. The stochastic process V_t plays the role of a new, random time τ. It is termed the operational time of a system, see [20]. The prominent example of subordination with wide range of applications in physics [1–14] is the CTRW defined in the following way.

Given a sequence T_i , $i = 1, 2, ...$of nonnegative, independent, identically distributed (i.i.d.) random variables which represent the time intervals between successive jumps of a particle, we define the random time interval of n jumps as

$$T(n) = \sum_{i=1}^{n} T_i\,, \quad T(0) = 0. \tag{1}$$

Then the so-called renewal process, describing the number of the particle jumps performed up to time $t > 0$, takes the form

$$N_t = \max\{n : T(n) \le t\} . \tag{2}$$

N_t is also referred to as the counting process and, by the relation

$$\{T(n) \le t\} = \{N_t \ge n\} ,$$

can be mathematically regarded as a process inverse to $T(n)$. Next, for a sequence R_i , $i = 1, 2, ...$ of i.i.d. random variables, indicating both the length and the direction of the i-th jump, we specify the position of the particle after n jumps by summing up the jumps R_i

$$R(n) = \sum_{i=1}^{n} R_i \quad , \quad R(0) = 0 . \tag{3}$$

Here, the variables R_i , $i = 1, 2, ...$ are assumed to be independent of the sequence T_i , $i = 1, 2, \ldots$. The last assertation assures that the processes N_t and $R(n)$ are independent. In conclusion, the total distance reached by the particle by time $t \ge 0$ is defined as the cumulative stochastic process

$$R(N_t) = \sum_{i=1}^{N_t} R_i \tag{4}$$

known as the CTRW. Thus the CTRW process is clearly an example of subordination with the directing process $R(n)$ and the subordinator N_t. In this case N_t is the operational time of a system describing the number of steps performed by a walker up to time $t > 0$.

In the recent paper of Piryatinska *et al.* [21], authors consider more general class of subordinations, where the random process $R(n)$ is replaced by a Lévy diffusion $X(\tau)$ and $T(n)$ is substituted by the continuous parameter process $T(\tau)$ with the corresponding inverse random process $V_t = \inf \{\tau : T(\tau) > t\}$. The relationship between $T(\tau)$ and V_t is in principle the same as the one between $T(n)$ and the counting process N_t. The subordinate process $X(V_t)$, which is the generalization of the CTRW process $R(N_t)$, is called the *anomalous diffusion*.

The anomalous diffusion $X(V_t)$ exhibits some important mathematical properties, which will frequently be used in our further considerations. As $X(\tau)$ and V_t are independent processes, the probability density function (p.d.f.) $p(x,t)$ of $X(V_t)$, obtained via the total probability formula, equals

$$p(x,t) = \int_0^{\infty} f(x,\tau) g(\tau,t) d\tau , \tag{5}$$

where $f(x,\tau)$ and $g(\tau,t)$ are the p.d.f.s of $X(\tau)$ and V_t, respectively. Similarly, the Fourier transform $\widetilde{p}(k,t) = \langle \exp(ikX(V_t))\rangle$ and the Laplace transform $\widehat{p}(k,t) = \langle \exp(-kX(V_t))\rangle$ are given by

$$\begin{aligned} \widetilde{p}(k,t) &= \int_0^\infty \widetilde{f}(k,\tau) g(\tau,t) d\tau \,, \\ \widehat{p}(k,t) &= \int_0^\infty \widehat{f}(k,\tau) g(\tau,t) d\tau \,. \end{aligned} \tag{6}$$

Here $k > 0$ has the physical sense of a wave number.

Recently, it has been explored [22] a special case of the CTRW, where the interjump time intervals T_i belong to the domain of attraction of a completely asymmetric stable distribution $S_{\alpha,1}(t)$ [1] (*i.e.* $P(T_i > t) \propto t^{-\alpha}$ as $t \to \infty$ for some $0 < \alpha < 1$, see [23, 24]), and the jumps R_i belong to the domain of attraction of a γ-stable distribution $S_{\gamma,\beta}(x)$ with $0 < \gamma \leq 2$, $|\beta| \leq 1$. If the above assumptions are satisfied, then the appropriately rescaled CTRW process $R(N_t)$ tends in distribution to the subordinate process $X(V_t)$, where $X(\tau)$ is the standard γ-stable Lévy motion and V_t is the so-called inverse-time α-stable subordinator (or inverse-time α-stable process) defined via its Laplace transform

$$\langle \exp(-uV_t)\rangle = E_\alpha(-c_\alpha u t^\alpha) \ , \ c_\alpha > 0 \ . \tag{7}$$

Here

$$E_\alpha(z) = \sum_{n=0}^{\infty} \frac{z^n}{\Gamma(n\alpha + 1)} \tag{8}$$

is the Mittag–Leffler function [25].

In the aforementioned paper, the authors elucidate the role of the inverse-time α-stable subordinator V_t and show that this random process is responsible for the nonexponential Cole–Cole relaxation. In the next section we introduce a new type of a subordinator and, in a similar manner, point out its relationship with another significant type of relaxation, namely with the stretched exponential relaxation response.

[1] Here, for a stable distribution we use the notation $S_{\alpha,\beta}(t)$, where $0 < \alpha \leq 2$ denotes the index of stability and $|\beta| \leq 1$ denotes the skewness parameter.

3. Stretched exponential response and anomalous diffusion

The classical Debye pattern of dielectric ralaxation

$$\phi(t) = e^{-\omega_p t}\,,$$

where ω_p is the loss-peak frequency (a characteristic material constant), has found for many years a widespread acceptance in representing the relaxation data of various physical systems. However, as extensive time-and frequency-domain experimental investigations prove, the exponential model of relaxation hardly ever fits the relaxation data [26, 27]. As an alternative, the deviation from the classical pattern observed in the time-dependent response of dielectric systems to a steady electric field is often described by the stretched exponential relaxation function

$$\phi(t) = e^{-(\omega_p t)^\alpha}\,,\ \alpha \in (0,1). \tag{9}$$

It is commonly assumed [16–18] that the theoretical attempts to model the nonexponential relaxation can be based on the idea of relaxation of an excitation representing the diffusive behaviour of the system as a whole. In the framework of the one-dimensional nonbiased CTRWs, the time-domain relaxation function for a given mode k is introduced in a standard way as the following Fourier transform

$$\phi(t) = \left\langle e^{ikR(t)} \right\rangle \quad ,$$

where $R(t)$ denotes the diffusion front — the scaling limit of the CTRW. If $R(t)$ takes values only on the positive half-line, the above definition must be modified [18] by replacing the Fourier transform with the Laplace transform. Hence we obtain

$$\phi(t) = \left\langle e^{-kR(t)} \right\rangle$$

for the biased walk. The above two formulas give the temporal relaxation of a macroscopic excitation.

Let us now consider the subordinate process with the directing process $X(\tau)$ belonging to the family of Lévy stable processes and the subordinator $V_t^{(\alpha)}$ being the fully asymmetric Lévy α-stable process with the following Laplace transform

$$\left\langle e^{-kV_t^{(\alpha)}} \right\rangle = e^{-c_\alpha k^\alpha t}\ \ ,\ 0 < \alpha < 1\ .$$

As it has been already shown in [20], such an operational time $V_t^{(\alpha)}$ does not change the type of relaxation — $\phi(t)$ remains a simple exponentially decaying function. To win a model undergoing a stretched exponential relaxation pattern, we have to modify the subordinator $V_t^{(\alpha)}$. Since $V_t^{(\alpha)}$ is $1/\alpha$-selfsimilar, for $b > 0$ and $t > 0$ we get

$$V_{bt}^{(\alpha)} \stackrel{\mathrm{d}}{=} (bt)^{1/\alpha} V_1^{(\alpha)}\,,$$

where "$\stackrel{\mathrm{d}}{=}$" stands for "equal in law". Note that for $\alpha \to 1$, $V_t^{(\alpha)}$ converges to the degenerate deterministic process. Now we transform the above formula by "stretching" the spatial axis and introduce a new subordinator $\overline{V}_t^{(\alpha)}$ by defining its finite-dimensional distributions

$$\overline{V}_t^{(\alpha)} :\stackrel{\mathrm{d}}{=} ct V_1^{(\alpha)}\,, \quad c > 0\,. \tag{10}$$

$\overline{V}_t^{(\alpha)}$ is clearly a positive, strictly increasing process. Additionally, it is 1-selfsimilar and its Laplace transform is given by the stretched exponential function

$$\left\langle e^{-k\overline{V}_t^{(\alpha)}} \right\rangle = e^{-c_\alpha t^\alpha k^\alpha}\,, 0 < \alpha < 1\,,\ c_\alpha > 0\,. \tag{11}$$

Clearly, when $\alpha \to 1$, then $\overline{V}_t^{(\alpha)}$ becomes deterministic. In what follows we illustrate the relationship between the stretched exponential relaxation pattern and the anomalous diffusion $X\left(\overline{V}_t^{(\alpha)}\right)$, where $X(\tau)$ belongs to the general class of Lévy stable diffusions and $\overline{V}_t^{(\alpha)}$ is defined by (10).

We first consider the problem of finding the relaxation function $\phi(t)$ for the anomalous diffusion $X\left(\overline{V}_t^{(\alpha)}\right)$. Recall that $\overline{V}_t^{(\alpha)}$ has the Laplace transform (11) and $X(\tau)$ belongs to the class of γ-stable Lévy processes. In the case of the nonbiased random-walk, *i.e.* when the process $X(\tau)$ has the following characteristic function

$$\left\langle e^{ikX(\tau)} \right\rangle = e^{-c_\gamma k^\gamma \tau}\,,\ c_\gamma > 0\,,$$

the above formula together with (6) and (11) imply that the relaxation function takes the form

$$\phi(t) = \left\langle e^{ikX(\overline{V}_t^{(\alpha)})} \right\rangle = \int_0^\infty e^{-c_\gamma k^\gamma \tau} g(\tau, t)\, d\tau = \exp(-c_{\alpha,\gamma} t^\alpha k^{\alpha\gamma})\,,$$

where $g(\tau, t)$ is the p.d.f. of $\overline{V}_t^{(\alpha)}$ and $c_{\alpha,\gamma} = c_\alpha \cdot c_\gamma^\alpha$. Hence for $\omega_p = k^\gamma \cdot c_{\alpha,\gamma}^{1/\alpha}$ we obtain the stretched exponential time-domain relaxation function (9).

Even if the directing process $X(\tau)$ is the classical Brownian motion (that is when $\gamma = 2$), the anomalous diffusion $X(\overline{V}_t^{(\alpha)})$ leads to the stretched exponential function

$$\phi(t) = \exp(-c_{\alpha,2} t^{\alpha} k^{2\alpha})$$

as well.

Similarly, in the case of the biased random-walk, when the totally asymmetric process $X(\tau)$ has the Laplace transform

$$\left\langle e^{-kX(\tau)} \right\rangle = e^{-c_\gamma k^\gamma \tau}\,, \quad 0 < \gamma \leq 1\,,$$

it can be shown in an analogous way that

$$\phi(t) = \left\langle e^{-kX(\overline{V}_t^{(\alpha)})} \right\rangle = \int\limits_0^\infty e^{-c_\gamma k^\gamma \tau} g(\tau, t) d\tau = \exp(-c_{\alpha,\gamma} t^{\alpha} k^{\alpha\gamma})\,.$$

Once more, for $\omega_p = k^\gamma \cdot c_{\alpha,\gamma}^{1/\alpha}$, we get the stretched exponential function (9). In even more general case, when $X(\tau)$ is a general Lévy process with characteristic function

$$\left\langle e^{ikX(\tau)} \right\rangle = e^{-C\psi(k)\tau}\,,$$

here $\psi(k)$ is the logarithm of the characteristic function of the random variable $X(1)$, some standard calculations lead to the stretched exponential response function as well.

4. Conclusions

We have introduced a new type of subordinator $\overline{V}_t^{(\alpha)}$ by modifying the well known strictly increasing α-stable process. We have demonstrated how the empirical time-domain stretched exponential function can be obtained from the anomalous diffusion model $X(\overline{V}_t^{(\alpha)})$ and illustrated the role of the operational time $\tau = \overline{V}_t^{(\alpha)}$ in this probabilistic model. Our considerations show that the type of relaxation function derived from the anomalous diffusion process depends entirely on the subordinator. The Lévy-stable directing process $X(\tau)$ influences only the constant ω_p in the stretched exponential function (9) and determines the spatial properties of $X(\overline{V}_t^{(\alpha)})$, see Table I. Our results together with the ones presented in [20] and [22] confirm that the subordination being the transformation from the physical time t to the operational time τ is responsible for the anomalous behaviour of a system.

TABLE I

Stochastic schemes of the stretched exponential time-domain response with subordinator $\overline{V}_t^{(\alpha)}$. Let us note that the Brownian motion in combination with $\overline{V}_t^{(\alpha)}$ also leads to the nonexponential relaxation of a system.

$X(\tau),\ 0<\gamma\le 2,\ \lvert\beta\rvert\le 1$	$\phi(t)$
symmetric γ-stable Lévy processes $(0<\gamma<2,\ \beta=0)$	$\exp(-c_{\alpha,\gamma}t^{\alpha}k^{\alpha\gamma})$
strictly increasing α-stable Lévy processes $(0<\gamma<1,\ \beta=1)$	$\exp(-c_{\alpha,\gamma}t^{\alpha}k^{\alpha\gamma})$
Brownian motion $(\gamma=2,\ \beta=0)$	$\exp(-c_{\alpha,2}t^{\alpha}k^{2\alpha})$
deterministic process linear in operational time $(\gamma=1,\ \beta=1)$	$\exp(-c_{\alpha,1}t^{\alpha}k^{\alpha})$

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