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SUBORDINATED RANDOM WALK APPROACH TO ANOMALOUS RELAXATION IN DISORDERED SYSTEMS*

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We study anomalous relaxation properties of the continuous-time random walk model in which the space-jump and waiting-time evolution is given by two random Markov processes. This model describes the subordination of one random process by another. The directing process is inverse to the totally skewed, strictly Lévy process. Owing to the properties of the directing process, the relaxation function in the uncoupled random walk model takes the empirical Cole–Cole form. By means of this theoretical analysis we find that the coupled and uncoupled walks lead to different forms of the relaxation function.

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1. Introduction

The nature of anomalous relaxation in the various complex systems (amorphous semiconductors and insulators, polymers, molecular solid solutions, glasses, *etc.*) is the subject of intensive studies for many years [1,2] (and the references therein). The experimental investigations surely have established the non-exponential evolution of such systems towards equilibrium, *i.e.* the empirical functions used to fit the experimental data exhibit the fractional-power dependence of the relaxation responses on frequency and time. In fact, this important feature is independent on the details of examined systems. Undoubtedly, many-body effects play an appreciable role in such systems. So these effects induce time fluctuations in the potential seen by each particle and essentially act as a noise source. At the same time,

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they form a complex potential fixed landscape with many local minima separated by barriers of all scales, trapping and untrapping the particle orbits in a self-similar hierarchy of cantori. As a result, the trajectory of particles can be very similar to a random walk. It is not surprising the parallels, suggested in literature [3–6], to be drawn between anomalous relaxation and anomalous diffusion.

Now the continuous time random walk (CTRW) concept is widely accepted in physics for the description of anomalous diffusion (see review [7]). Although the CTRW models has the clear advantage in force of their high physical motivation, the master equation for anomalous diffusion is often introduced phenomenologically, and the grounds remain off screen. It is exceptionally important to recognize the stochastic process itself responsible for the anomalous behavior. Without questions, this is a corner stone of the theory. The recent works of Meerschaeft, Scheffler and Becker-Kern [8, 9] have summed up the long-term studies of the problem and made things ultimate clear. The motivation of our work is to explain anomalous relaxation of disordered systems from the latest achievements in the CTRW approach. We show that the applications of CTRW models to relaxing processes can be essentially extended.

The present paper is organized as follows. In Sec. 2 we describe in detail the minimal CTRW model resulting in anomalous diffusion. The main feature of our consideration is that at first we define the corresponding stochastic process, whereupon the density function and the master equation are a direct consequence of the process properties. We analyze the essential aspects of anomalous diffusion and anomalous relaxation in the framework of a one-body picture. As for many-body effects, they are taken into account in terms of randomizing both spatial and temporal degrees of freedom in the one-body picture. Our results towards anomalous diffusion is represented in Sec. 3, and Sec. 4 is devoted to their applications for anomalous relaxation. Our picture embraces the coupled and uncoupled walks. They lead to different forms of diffusion and relaxation. A summary and a discussion are given in Sec. 5.

2. Continuous time random walks

A continuous time random walk describes the stochastic time and space evolution of a walking particle by means of two Markov processes, random waiting-times and random jumps. Successive couples of random time and space steps are usually considered as independent. However, in a couple the time and space steps may be dependent.

Let T_1, T_2, ... be nonnegative independent and identically distributed (i.i.d.) random variables that correspond to the waiting times between jumps of a walking particle. The particle jumps are given by i.i.d. random vectors

$\vec{R}_1, \vec{R}_2, \ldots$ on the d-dimensional space $\boldsymbol{R}^d$ which are assumed independent of (T_i). The position of the particle after the nth jump becomes $\vec{r}_n = \sum_{i=1}^{n} \vec{R}_i$, being $\vec{r}_0 = 0$. For $t \geq 0$ the number of jumps up to time t is $N_t = \max\{n \in \boldsymbol{N} : \sum_{j=1}^{n} T_i \leq t\}$, and the vector $\vec{X}(t) = \vec{r}_{N_t} = \sum_{i=1}^{N_t} \vec{R}_i$ defines the position of the particle at time t. The stochastic process $\{\vec{X}(t)\}_{t\geq 0}$ is called a continuous time random walk (CTRW). We consider CTRW in the d-dimensional space and occurring at the non-negative one-dimensional time. Though such walks consist only of discrete time and space steps, the jump model can be generalized to "continuous steps".

Assume that T_j belongs to the strict domain of attraction of some stable law with index $0 < \beta < 1$. The choice of the index β in the range $0 < \beta < 1$ is conditioned by the support of the time steps T_j on the non-negative semi-axis $\boldsymbol{R}_+$. Then there exist $b_n > 0$ such that the sum $b_n(T_1 + \cdots + T_n)$ has asymptotically (by virtue of a convergence in distribution) the stable distribution with index β, if n tends to infinity. In the discrete model the time of the nth jump is $\Theta(n) = \sum_{j=1}^{n} T_j$, $T_0 = 0$. For $\tau \geq 0$ one can write $\Theta(\tau) = \sum_{j=1}^{\lfloor \tau \rfloor} T_j$ and $b(\tau) = b_{\lfloor \tau \rfloor}$, where $\lfloor \tau \rfloor$ denotes the integer part of τ. As has been stated in [8, 9], $\{b(c)\Theta(c\tau)\}_{\tau\geq 0}$ converges in distribution of all finite dimensional marginal distributions to the process $\{T(\tau)\}_{\tau\geq 0}$ as $c \to \infty$. The process $\{T(\tau)\}$ has stationary independent increments. Furthermore, it is a strictly stable and totally skewed Lévy process satisfying to $\{T(c\tau)\}_{\tau\geq 0} \overset{f.d.}{=} \{c^{1/\beta}T(\tau)\}_{\tau\geq 0}$ for all $c > 0$, where $\overset{f.d.}{=}$ denotes equality of all finite dimensional distributions. The process $\{T(\tau)\}$ depends on the continuous internal time $\tau \geq 0$, but the index is different from the real time t. The label *continuous* indicates just the fact that the index belongs to a continuous set, but does not imply the continuity of the paths. According to [8], the process $\{T(\tau)\}$ is self-similar with exponent $H = 1/\beta > 1$. The sample paths of $\{T(\tau)\}$ are increasing almost surely (a.s). Since $T(\tau) \overset{d}{=} \tau^{1/\beta}T(1)$, where $\overset{d}{=}$ means equal in distribution, it follows that $T(\tau) \to \infty$ in probability as $\tau \to \infty$.

Assume that $(\vec{R}_i)$ are i.i.d. $\boldsymbol{R}^d$-valued random variables independent of (T_j) and let $\vec{R}_i$ belong to the strict generalized domain of attraction of some full operator stable law ν. Then there exists a function $B(c)$ invertible for all $c > 0$ and $B(\lambda c)\, B(c)^{-1} \to \lambda^{-\Upsilon}$ as $c \to \infty$ for any $\lambda > 0$, Υ being a $d \times d$ matrix (real parts of eigenvalues of Υ are greater than or equal to $1/2$), such that $B(n) \sum_{i=1}^{n} \vec{R}_i$ converges in distribution to Y as $n \to \infty$, where Y has distribution ν. Next, using the limit passage $\{B(c) \sum_{i=1}^{\lfloor c\tau \rfloor} \vec{R}_i\}_{\tau\geq 0}$ as $c \to \infty$, we define the stochastic process $\{Y(\tau)\}$ depending on the internal time τ. The process has stationary independent increments with $Y(0) = 0$ a.s. Then $\{Y(\tau)\}$ is continuous in law. Moreover, $\{Y(\tau)\}$ is operator self-similar with exponent $\varUpsilon$: $\{Y(c\tau)\}_{\tau\geq 0} \stackrel{f.d.}{=} \{c^{\varUpsilon}Y(\tau)\}_{\tau\geq 0}$. The process is called *an operator Lévy motion* [8]. If the exponent $\varUpsilon = \tilde{a}I$ is a constant multiplied by the identity, then ν is a stable law with index $\alpha = 1/\tilde{a}$, and $\{Y(\tau)\}$ is a classical d-dimensional Lévy motion. In the particular case, when $\tilde{a} = 1/2$, the process $\{Y(\tau)\}$ is a d-dimensional Brownian motion.

Now we consider the limiting behavior of the counting process $\{N_t\}_{t\geq 0}$ mentioned above. It turns out that the scaling limit of this process is the hitting time process for the Lévy motion $\{T(x)\}_{x\geq 0}$. This hitting time process represents the true time evolution of the position vector of the walking particle and is also self-similar with exponent β. However, it is cardinally different from $\{T(x)\}_{x\geq 0}$. The hitting time process $S(t) = \inf\{x : T(x) > t\}$ is well-defined and dependent on the true time t. Note that $\{S(t)\}_{t\geq 0}$ is actually the inverse of the process $\{T(x)\}_{x\geq 0}$. If $T(x) < t$ then $T(y) < t$ for all $y > x$ sufficiently close to x, so that $S(t) > x$. On the other hand, if $T(x) \geq t$ then $T(y) > t$ for all $y > x$ so that $S(t) \leq x$. It is easily verified that $\{S(t_i) \leq x_i \text{ for } i = 1,\ldots,m\} = \{T(x_i) \geq t_i \text{ for } i = 1,\ldots,m\}$ holds true for any $0 \leq t_1 < \ldots < t_m$ and $x_1,\ldots,x_m \geq 0$. Since $\{T(x)\}_{x\geq 0}$ is strictly increasing, the process $\{S(t)\}_{t\geq 0}$ is continuous and non-decreasing. From the self-similarity of $\{T(x)\}$ it follows the same property for $\{S(t)\}$, *i.e.* $\{S(ct)\}_{t\geq 0} \stackrel{f.d.}{=} \{c^{\beta}S(t)\}_{t\geq 0}$ for any $c > 0$. While $\{T(x)\}_{x\geq 0}$ is a Lévy process, the inverse process $\{S(t)\}_{t\geq 0}$ is no longer a Lévy process, neither a Markov process, but it is a continuous submartingal, as shown in [10].

For a real valued random variable X let $E[X]$ denote its expectation. Collect important properties of the process $\{S(t)\}_{t\geq 0}$

- ◇ the process $\{S(t)\}_{t\geq 0}$ has neither stationary nor independent increments [8];
- ◇ $S(t) \stackrel{d}{=} t^{\beta}S(1)$;
- ◇ for any $\gamma > 0$ the process $\{S(t)\}_{t\geq 0}$ has the finite γ-moment $E[S(t)^{\gamma}] = C(\beta,\gamma)\, t^{\beta\gamma}$, where $C(\beta,\gamma)$ is a positive finite constant [8];
- ◇ $S(T(\tau)) = \tau$ a.s. and $T(S(t)) \geq t$ a.s.;
- ◇ the random variable $S(t)$ has the density

$$p_t(x) = \frac{t}{\beta}\, x^{-1-1/\beta}\, g_\beta\left(t\, x^{-1/\beta}\right)\,,$$

where g_β is the density of the process $T(\tau)$.

The fourth and fifth properties reflect the fact that $\{S(t)\}$ is the left-inverse of the process $\{T(x)\}$. If the probability density of $\{T(x)\}$ has the index $\beta = 1/2$, then the fifth property gives the normal law as a probability density of $\{S(t)\}$. The inverse process to the stochastic time evolution describes the true time evolution of a walking particle. The hitting time $S(t) =$

$\inf\{x : T(x) > t\}$ is called also *a first passage time*. Really, for a fixed time it represents the first passage of the stochastic time evolution above this time level. The sample paths of $\{N_t\}_{t\geq 0}$ and $\{S(t)\}_{t\geq 0}$ are increasing. As shown in [10, 11], the random value $S(t)$ is connected with a Mittag–Leffler distribution via $E[\mathrm{e}^{-vS(t)}] = \sum_{n=0}^{\infty}(-vt^{\beta})^n/\Gamma(1+n\beta) = E_{\beta}(-vt^{\beta})$. This confirms the third property in the special case where γ is a positive integer.

3. Anomalous diffusion

Let us investigate the relationship between the probability density of the position vector $\vec{\boldsymbol{r}}_t$ of a walking particle at real time t and the couple $(Y(\tau), T(\tau))$. The safe mathematical construction of the process $\vec{\boldsymbol{r}}_t$ is shown in [11]. The limit process $\vec{\boldsymbol{r}}_t = Y(S(t))$ is an operator Lévy motion $Y(\tau)$ subordinated to the hitting process $S(t)$ of a classical stable subordinator $T(\tau)$. We briefly recall that a subordinated process is obtained by randomizing the time clock of a stationary process $\boldsymbol{X}(t)$ using a new clock $U(t)$, where $U(t)$ is a random process with nonnegative independent increments. The resulting process $\boldsymbol{X}(U(t))$ is said to be subordinated to $\boldsymbol{X}(t)$, called *the parent process*, and is directed by $U(t)$, called *the directing process*. The directing process is often referred to as the randomized time or operational time. In general, the subordinated process $\boldsymbol{X}(U(t))$ can become non-Markovian, though its parent process is Markovian. The process $\vec{\boldsymbol{r}}_t$ is a scaling limit of CTRW.

The new process $\{\vec{\boldsymbol{r}}_t\}_{t\geq 0}$ subordinated to the Markov process $\{Y(\tau)\}_{\tau\geq 0}$ and directed by the randomized time process $\{S(t)\}_{t\geq 0}$ is well defined. If the processes $\{Y(\tau)\}_{\tau\geq 0}$ and $\{T(\tau)\}_{t\geq 0}$ is uncoupled (*i.e.* independent on each other), the probability density of $\vec{\boldsymbol{r}}_t$ with $t \geq 0$ can be written as a weighted integration over the internal time τ so that

$$p^{\vec{\boldsymbol{r}}_t}(t, \vec{\boldsymbol{x}}) = \int_0^{\infty} p^Y(\tau, \vec{\boldsymbol{x}})\, p^S(t, \tau)\, d\tau, \tag{1}$$

where $p^Y(\tau, \vec{\boldsymbol{x}})$ represents the probability to find the parent process $Y(\tau)$ at $\vec{\boldsymbol{x}}$ on operational time τ and $p^S(t, \tau)$ is the probability to be at the operational time τ on real time t. The process $Y(S(t))$ is self-similar with index $\beta\Upsilon$ such that $\{Y(S(ct))\}_{t\geq 0} \overset{f.d.}{=} \{c^{\beta\Upsilon}Y(S(t))\}_{t\geq 0}$ is for all $c > 0$. According to [8, 9], the limiting process does not have stationary increments and is not operator stable for any $t > 0$. In Laplace space the probability density $p^{\vec{\boldsymbol{r}}_t}(t, \vec{\boldsymbol{x}})$ takes the most simple form $u^{\beta-1}\bar{p}^Y(u^{\beta}, \vec{\boldsymbol{x}})$, since $\bar{p}^S(u, \tau) = u^{\beta-1}\exp(-u^{\beta}\tau)$ and $\bar{p}^Y(u^{\beta}, \vec{\boldsymbol{x}}) = \int_0^{\infty} p^Y(\tau, \vec{\boldsymbol{x}})\,\exp(-u^{\beta}\tau)\, d\tau$.

If the operator Lévy motion $Y(\tau)$ on $\boldsymbol{R}^d$ has the probability distribution $p\,(x,\tau)$, the linear operator $\hat{T}_\tau f(x) = \int f(x-y)\,p\,(y,\tau)\,dy$ forms a convolution semigroup with generator $\hat{L} = \lim_{\tau\downarrow 0}\tau^{-1}(\hat{T}_\tau - \hat{T}_0)$ [12]. Then the mapping $q(x,\tau) = \hat{T}_\tau f(x)$ solves the abstract Cauchy problem $\partial q(x,\tau)/\partial\tau = \hat{L}\,q(x,\tau)$ with the initial condition $q(x,0) = f(x)$. The distinguishing feature of the process $\{Y(\tau)\}$ is that the generator $\hat{L}$ is time-independent. In all other respects its exact form can be quite arbitrary. If $\{Y(\tau)\}$ is an α-stable Lévy motion, the operator $\hat{L}$ is a multidimensional fractional derivative of order α [13]. In general, for an operator Lévy motion it even represents a generalized fractional derivative on $\boldsymbol{R}^d$ whose order of differentiation can vary with coordinate [14]. When the operator $\hat{L}$ acts on the Laplace image $\bar{p}^{\vec{\boldsymbol{r}}_t}(u,\vec{\boldsymbol{x}}) = u^{\beta-1}\bar{p}^Y(u^\beta,\vec{\boldsymbol{x}})$, we obtain $[\hat{L}\,\bar{p}^{\vec{\boldsymbol{r}}_t}](u,\vec{\boldsymbol{x}}) = u^\beta\,\bar{p}^{\vec{\boldsymbol{r}}_t}(u,\vec{\boldsymbol{x}}) - f(\vec{\boldsymbol{x}})\,u^{\beta-1}$, where $f(\vec{\boldsymbol{x}})$ is the initial condition. The inverse Laplace transform of the latter expression gives the abstract partial differential equation with the fractional derivative of time:

$$p^{\vec{\boldsymbol{r}}_t}(t,\vec{\boldsymbol{x}}) = f(\vec{\boldsymbol{x}}) + \frac{1}{\Gamma(\beta)}\int_0^t d\tau (t-\tau)^{\beta-1}\left[\hat{L}\,p^{\vec{\boldsymbol{r}}_t}\right](\tau,\vec{\boldsymbol{x}})\,. \tag{2}$$

The solution of (2) is directly connected with the solution of $\partial p^Y(\tau,\vec{\boldsymbol{x}})/\partial\tau = \hat{L}\,p^Y(\tau,\vec{\boldsymbol{x}})$. The probability density $p^S(t,\tau)$ is written as $t^{-\beta}F_\beta(z)$ with $z = \tau/t^\beta$, and the function $F_\beta(z)$ has the H-function representation

$$F_\beta(z) = H^{10}_{11}\left(z\left|\begin{matrix}(1-\beta,\beta)\\(0,1)\end{matrix}\right.\right) = \sum_{k=0}^{\infty}\frac{(-z)^k}{k!\Gamma(1-\beta-k\beta)}\,,$$

where $\Gamma(x)$ is the ordinary Gamma function. Then the probability density $p^{\vec{\boldsymbol{r}}_t}(t,\vec{\boldsymbol{x}})$ is expressed in terms of the following integral relation

$$p^{\vec{\boldsymbol{r}}_t}(t,\vec{\boldsymbol{x}}) = \int_0^\infty F_\beta(z)\,f^Y(t^\beta z,\vec{\boldsymbol{x}})\,dz\,. \tag{3}$$

The formula is especially useful for the probability density $p^Y(\tau,\vec{\boldsymbol{x}})$, known either in a closed form (for example, an harmonic potential in the standard Fokker–Planck equation) or in the context of separation of variables. Moreover, each functional of such a solution can be obtained from the solution by immediate integration, only if the functional exists. It should be noted here that the probability density $p^S(t,\tau)$ has finite moments of any order, although the subordinator $T(\tau)$ has not the property. The function $F_\beta(z)$ vanishes exponentially for large positive z and strictly positive on $(0,\infty)$.

Now assume that the waiting time between jumps and the jumps sizes are no more independent. The CTRWs are called *coupled*. Then the subordination relation (1) takes a more general form

$$p^{\vec{\boldsymbol{r}}_t}(t,\vec{\boldsymbol{x}}) = \int_0^\infty P\{Y(\tau) = \vec{\boldsymbol{x}} \mid S(t) = \tau\}\, p^S(t,\tau)\, d\tau, \tag{4}$$

where $P\{Y(\tau) = \vec{\boldsymbol{x}} \mid S(t) = \tau\}$ describes the conditional distribution between $T(\tau)$ and $Y(\tau)$. Following the arguments of [9], the probability density of $\{\vec{\boldsymbol{r}}_t\}$ is written as

$$p^{\vec{\boldsymbol{r}}_t}(t,\vec{\boldsymbol{x}}) = \int_0^\infty \int_0^t f_u(\vec{\boldsymbol{x}}, t-\tau)\, d\tau\,, \tag{5}$$

where $f_u(\vec{\boldsymbol{x}},t)$ has the Fourier–Laplace transform

$$\mathcal{FL}[f_u](\vec{\boldsymbol{x}},t) = \int_{\boldsymbol{R}^d} \int_0^\infty \mathrm{e}^{i\langle\vec{\boldsymbol{x}}\cdot\vec{\boldsymbol{k}}\rangle}\, \mathrm{e}^{-st}\, f_u(\vec{\boldsymbol{x}},t)\, dt\, d\vec{\boldsymbol{x}} = \exp\{-\psi(\vec{\boldsymbol{k}},s)\}$$

well defined for all $(\vec{\boldsymbol{k}},s) \in \boldsymbol{R}^d \times \boldsymbol{R}_+$, and $\psi(\vec{\boldsymbol{k}},s)$ is the log-characteristic function of (Y,T). Recall that infinitely divisible distributions are just determined by its log-characteristic function (Lèvy-Khinchin formula). If Q is a positive quadratic form on $\boldsymbol{R}^d$, $(\vec{\boldsymbol{a}},b) \in \boldsymbol{R}^d \times \boldsymbol{R}_+$ and ϕ is a Lévy measure of (Y,T) on $\boldsymbol{R}^d \times \boldsymbol{R}_+/\{(0,0)\}$, the log-characteristic function is

$$\psi(\vec{\boldsymbol{k}},s) = i\langle\vec{\boldsymbol{a}}\cdot\vec{\boldsymbol{k}}\rangle + Q(\vec{\boldsymbol{k}}) + \int_{\boldsymbol{R}^d\times\boldsymbol{R}_+/\{(0,0)\}} \left(1 - \mathrm{e}^{i\langle\vec{\boldsymbol{x}}\cdot\vec{\boldsymbol{k}}\rangle - st} + \frac{i\langle\vec{\boldsymbol{x}}\cdot\vec{\boldsymbol{k}}\rangle}{1+\|\vec{\boldsymbol{x}}\|^2}\right) \phi(d\vec{\boldsymbol{x}},dt).$$

It should be pointed out that the log-characteristic function of an infinitely divisible distribution is the symbol of pseudo-differential operator defined by the generator of the corresponding convolution semigroup [12]. Thus, in our notations we have

$$\mathcal{FL}[\psi(iD_x,\partial_t)\, p^{\vec{\boldsymbol{r}}_t}](\vec{\boldsymbol{x}},t) = \psi(\vec{\boldsymbol{k}},s)\, \mathcal{FL}[p^{\vec{\boldsymbol{r}}_t}](\vec{\boldsymbol{x}},t)\,. \tag{6}$$

If ψ does not grow too fast at infinite, the function $\psi(iD_x,\partial_t)$ is pointwise defined and can be extended to larger spaces of functions (or even distributions) [9]. Then we can write (5) in the form $\mathcal{FL}[p^{\vec{\boldsymbol{r}}_t}](\vec{\boldsymbol{x}},t) = s^{\beta-1}/\psi(\vec{\boldsymbol{k}},s)$.

The inverse Fourier–Laplace transform $s^{\beta-1}$ gives the distribution $\delta(\vec{\boldsymbol{x}})\, t^{-\beta}/\Gamma(1-\beta)$, where $\delta(x)$ is the Dirac δ-function. At last formally the expression of the corresponding pseudo-differential equation can be written as

$$\psi(iD_x, \partial_t)\, p^{\vec{\boldsymbol{r}}_t}(\vec{\boldsymbol{x}}, t) = \delta(\vec{\boldsymbol{x}}) \frac{t^{-\beta}}{\Gamma(1-\beta)} \,. \tag{7}$$

In fact, Eq. (2) is a particular case of (7), where $\psi(\vec{\boldsymbol{k}}, s) = \psi(\vec{\boldsymbol{k}}) + s^\beta$. However, the coupled model is more flexible for anomalous diffusion than uncoupled one. As has been shown in [9], the effect of subordinating $Y(\tau)$ is to lighten the tail of $p^{\vec{\boldsymbol{r}}_t}(\vec{\boldsymbol{x}}, t)$ and slow the spreading rate, whereas in the uncoupled case the subordinated process $\vec{\boldsymbol{r}}_t$ spreads slowly, but has the same tail behavior as $Y(\tau)$. Therefore, the uncoupled model behaves as subdiffusion, and the coupled model can set also in anomalous superdiffusion.

4. Anomalous relaxation

In a many-body system, the relaxation function is the self part of the density autocorrelation function [3]. Within the framework of the one-body picture, this function corresponds to the characteristic function $G_k(t)$ of the position vector $\boldsymbol{r}_t$ of a walking particle at time t

$$G_k(t) = E[\exp(i\,\vec{\boldsymbol{k}} \cdot \vec{\boldsymbol{r}}_t)] = E\left[\exp\left(-\vec{\boldsymbol{k}}^2 \frac{S(t)}{2}\right)\right] , \tag{8}$$

where $\vec{\boldsymbol{k}}$ is the wave number. Therefore, we consider $G_k(t)$ as the relaxation function. The frequency-domain response $\phi^\star(\omega)$ is connected with the relaxation function $G_k(t)$ via the one-sided Fourier transform

$$\phi^\star(\omega) = 1 - i\omega \int_0^\infty \mathrm{e}^{-i\omega t} G_k(t)\, dt. \tag{9}$$

As is well known [15], the (dielectric) susceptibility $\chi(\omega)$ is defined by the formula:

$$\phi^\star(\omega) = \frac{\chi(\omega) - \chi_\infty}{\chi_0 - \chi_\infty},$$

where the constant χ_∞ represents the asymptotic value of $\chi(\omega)$, and χ_0 is the value of the opposite limit. In the case of a normal random walk these functions indicate the relaxation of Debye type $\exp(-t/\tau)$ with the constant τ. The formula (3) under consideration of CTRWs leads to the Cole–Cole relaxation with $\phi^\star_{\mathrm{CC}}(\omega) = 1/(1 + (iA\omega)^a)$, where A and a are some constants. It should be noted that though the result first was obtained for one-dimensional CTRW model in [5, 6], it remains valid for the multidimensional case and for non-normal distributions of space steps. Here it is important for the waiting time between successive jumps and space jumps to be independent of each other. When the condition is invalid, the relaxation functions can be other types.

Consider a concrete coupled example [9], where the necessary calculations can be carried out completely. Let T be a stable subordinator with Laplace transform $E[\mathrm{e}^{-sT}] = \exp(-s^\beta)$, $0 < \beta < 1$, and the conditional distribution of $Y \mid T = t$ is normal with mean zero and variance $2t$. The log-characteristic function of (Y, T) is given by

$$\psi(\vec{\boldsymbol{k}}, s) = \left(\vec{\boldsymbol{k}}^2 + s\right)^\beta .$$

Next we use the result (6) and write the Fourier–Laplace image of the probability density of the process $\vec{\boldsymbol{r}}_t = Y(S(t))$

$$\mathcal{FL}\left[p^{\vec{\boldsymbol{r}}_t}\right](\vec{\boldsymbol{x}}, t) = \frac{s^{\beta-1}}{\left(\vec{\boldsymbol{k}}^2 + s\right)^\beta} .$$

The inverse Laplace transform yields

$$G_k(t) = \int_0^t \mathrm{e}^{-\vec{\boldsymbol{k}}^2 u} \frac{u^{\beta-1}\,(t-u)^{-\beta}}{\Gamma(\beta)\,\Gamma(1-\beta)}\, du\,, \tag{10}$$

Expanding $\exp(-\vec{\boldsymbol{k}}^2 t)$ into a Taylor series and integrating (10) with respect to u leads to

$$G_k(t) = \sum_{n=0}^{\infty} \frac{\Gamma(\beta+n)}{\Gamma(\beta)\,(n!)^2} \left(-\vec{\boldsymbol{k}}^2 t\right)^n .$$

This model describes a coupled space-time diffusion having the same scaling index as Brownian motion [9]. However, the relaxation response based on the CTRW model is different of the exponential law typical for the ordinary Brownian motion.

On the other hand, the Cole–Cole relaxation function is related to the processes having the Mittag–Leffler distribution. Let Z_n denote the sum of n independent random values with Mittag–Leffler distribution. Then the Laplace transform of $n^{-1/\beta} Z_n$ is $(1 + s^\beta/n)^{-n}$, which tends to e^{-s^β} as n tends to infinity. This indicates the infinity divisibility of the Mittag–Leffler distribution [16]. By virtue of the power asymptotic form (long tail) the distribution with parameter β is attracted to the stable distribution with exponent β, $0 < \beta < 1$. The property of Mittag–Leffler distribution enables one to develop a corresponding stochastic process. The stochastic process (called *Mittag–Leffler's*) is subordinated to a stable process by the directing gamma process [16]. This directing process can be connected with channel switching in such a system, where each channel develops as an independent random process with the Mittag–Leffler distribution. The most of the theoretical models describing the relaxation response takes into consideration a cooperative nature of the phenomenon (for example, the dipole–dipole interaction and the interaction of different polar regions in conformity with the dielectric relaxation) [1,2]. The Mittag–Leffler process after its substitution in (8) gives the relaxation function of the well-know Havriliak–Negami empirical law. Thus, the Havriliak–Negami relaxation response can be also explained from the CTRW approach, if the hitting time process of a walking particle transforms into the Mittag–Leffler process. For that the hitting time process has an appropriate distribution attracted to a stable distribution. The subordination of the latter results just in the Mittag–Leffler process. In this connection it should be mentioned that the Lévy process subordinated by another Lévy one leads again to the Lévy process, but with other index [12]. The more detailed analysis of the Mittag–Leffler process as applied to the CTRW model will be considered in the future work.

As for the stretched-exponential form of the relaxation function [17], the type of relaxation cannot be explained from the above CTRW models. The appropriate random process must be other, namely the fractional Brownian motion. A mean-zero Gaussian process $\{B_H(t), t \geq 0\}$ is called *fractional Brownian motion*, if

$$E[B_H(t)\, B_H(s)] = \tfrac{1}{2}\left\{t^{2H} + s^{2H} - \mid t - s \mid^{2H}\right\} E\left[B_H(1)^2\right]$$

with $0 < H < 1$. This process is self-similar with stationary increments, but has independent increments only for $H = 1/2$ [18]. Using the Gaussian property of the fractional Brownian motion, in the simplest case the characteristic function of the process is written as $G_k(t) = \exp(-\vec{\boldsymbol{k}}^2 t^{2H})$. Within the framework of the one-body picture we consider $G_k(t)$ as the relaxation function. Thus, the fractional Brownian motion leads to the Kohlrausch law.

It is interesting to observe that the CTRW approach to anomalous relaxation in disordered systems does not assume an obligatory real travel of a real particle. In particular, for the dielectric systems under a week external electric field the active dipoles change their direction during the relaxation dynamics so that the macroscopic effect is defined by the resulting polarization vector. This vector can be imagined as a position vector of a "virtual" traveling particle toward that end to apply an appropriate CTRW model. The arguments support the wide application of CTRW models to relaxing processes.

5. Summary and discussion

In the light of the latest achievements the CTRW approach has extended appreciably its potentialities. It suggests to describe anomalous diffusion and anomalous relaxation in terms of subordinated random processes so that the index of the original Markov parent process is randomized by continuous, increasing and non-Markovian process, which is the inverse to a totally skewed, strictly Lévy process. The new process represents the limit process generalizing "discrete steps" of the CTRW to "continuous steps". The randomized time clock shows both small and large periods of resting in jumps of the stochastic time evolution occur with no finite mean. This reflects the absence of a characteristic time scale typical for the ordinary relaxation and diffusion. The anomalous behavior originates from the slowly decaying and self-similar distribution of the stochastic time evolution.

The coupled and uncoupled models of the CTRW describe different processes of relaxation and diffusion. So, the relaxation function in the uncoupled CTRW model has the Cole–Cole type, whereas the coupled CTRW model gives rise to a more general law. The models and their modifications allow one to obtain many well-know empirical functions fitted for experimental data of relaxation and diffusive phenomena. This emphasizes the universality of the CTRW approach. It should be also noted here the other universal approach to anomalous relaxation, suggested in [19–21]. It proceeds from the randomization of parameters of distributions that describes the relaxation rates in disordered systems. Although the point of view is enough close to ours in this paper, there are differences. Their comparison will be carried out elsewhere.

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