Anomalous diffusion with under- and overshooting subordination: A competition between the very large jumps in physical and operational times

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In this paper we present an approach to anomalous diffusion based on subordination of stochastic processes. Application of such a methodology to analysis of the diffusion processes helps better understanding of physical mechanisms underlying the nonexponential relaxation phenomena. In the subordination framework we analyze a coupling between the very large jumps in physical and two different operational times, modeled by underand overshooting subordinators, respectively. We show that the resulting diffusion processes display features by means of which all experimentally observed two-power-law dielectric relaxation patterns can be explained. We also derive the corresponding fractional equations governing the spatiotemporal evolution of the diffusion front of an excitation mode undergoing diffusion in the system under consideration. The commonly known type of subdiffusion, corresponding to the Mittag-Leffler (or Cole-Cole) relaxation, appears as a special case of the studied anomalous diffusion processes.

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I. INTRODUCTION

Anomalous diffusion is conventionally described by two popular stochastic models in the scientific literature. The first of them is the fractional Brownian motion (FBM) introduced by Kolmogorov in 1940 [1] and further studied by Yaglom [2]. The FBM is a generalization of the ordinary Brownian motion and it is characterized by the Hurst exponent 0 < H < 1 in the mean-squared displacement $\langle x^2 \rangle \sim t^{2H}$ of diffusing particles. For H > 1/2 the anomalous diffusion demonstrates the superdiffusion dynamics, whereas for H < 1/2it is subdiffusive. This study was motivated by Hurst's analysis of annual river discharges [3] and the observations of economic time series [4]. In 1968 Mandelbrot and van Ness developed a stochastic integral representation of the FBM as a fractional integral of the mean-zero Gaussian process [5]. Now the FBM is widely used in many fields (see, for example, [6,7]). However it should be noted that the FBM does not satisfy an important property: it is not a semimartingale; i.e., the FBM cannot be uniquely decomposed into a local martingale (sum of fair-game increments with the zero expectation value) and a finite variation process (drift part) [8]. Nevertheless, many significant processes, including the Brownian motion, Poisson, and Lévy processes, are semimartingales. The important role of semimartingales in description of the anomalous diffusion processes has been shown recently [8].

The second commonly used model of anomalous diffusion is the continuous time random walk (CTRW) and the corresponding fractional generalization (for subdiffusion) of the Fokker-Planck equation. The CTRW formalism has been introduced by Montroll and Weiss in 1965 [9] to describe further the transport of an electric charge in a disordered medium (anomalous diffusion in an amorphous semiconductor) [10]. The approach considers random walks in space and time by means of independent and identically distributed (iid) couples of space and time random steps (R_i, T_i) . The simplest decoupled CTRW considers independent time and space steps. The decoupled CTRWs can be described in terms of a waiting time probability density function. If it is a suitable Mittag-Leffler function, as shown by Hilfer and Anton [11], the time-fractional diffusion equation is obtained. The CTRW model involves stable distributions, and it shows various anomalous behaviors such as subdiffusion (diffusion slower than the normal one), Mittag-Leffler relaxation, and fractional diffusive equations [12-14]. The stochastic behavior of the CTRWs is just a semimartingale. The information that a given physical process belongs to the family of semimartingales permits one to distinguish between different stochastic schemes of subdiffusion (i.e., between the CTRW, FBM, and a fractional Lévy-stable motion) from experimental data (trajectories) [15,16]. As it has been recently shown, this holds also for diffusion in confined media [17]. That is why the notion of the semimartigale to anomalous diffusion is important. By a passage from the discrete (temporal and spatial) steps to a continuous limit, the CTRW description leads to a subordination of one random process by another. It should be pointed out that the idea of the subordination of one process or system by another is not new. Many years ago it was known (and now rather forgotten) under the name of "slaving principle" in synergetics [18]. In the context of stochastic processes the subordinated process $Y(t) = X(S_t)$ consists of the parent random process $X(\tau)$ and the directing one S_t . Here $X(\tau)$ and S_t are assumed to be independent, and the subordinator S_t should be a nondecreasing process in time (recall the deterministic array of time increasing as t). If one takes a strictly increasing α -stable Lévy process $U_{\alpha}(\tau)$ (with Laplace transform $\langle e^{-kU_{\alpha}(\tau)} \rangle = e^{-\tau k^{\alpha}}$ and $0 < \alpha < 1$), the pro-

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cess S_t is defined as the inverse-time α -stable subordinator $S_t = \inf\{\tau | U_{\alpha}(\tau) > t\}$. In this case the resulting process $X(S_t)$ becomes subdiffusion characterized by a power mean-square displacement in time and leading to the Mittag-Leffler (Cole-Cole) relaxation response. The important role of Mittag-Leffler type functions for anomalous relaxation has been recognized in [19,20]. The physical properties, numerical simulation methods, and interpretation of the inverse-time α -stable subordinator are studied in physical literature for almost one decade [21–23]. However, this type of the operational time (subordinator) does not exhaust all possible sources of anomalous diffusion.

A more complex CTRW model accounts for coupling between time and space steps. The coupled CTRWs were considered in the context of anomalous diffusion and nonexponential relaxation [24-26]. In this case the anomalous diffusion evolution is much richer. Sub- and superdiffusion (faster than normal) may be modeled. However, the analysis is rather exotic for the research, and it is in progress. In particular, the anomalous subdiffusive behavior attracts a great attention in modeling of subdiffusion in space-timedependent force fields beyond the fractional Fokker-Planck equation [27,28]. This approach uses the subordination techniques to represent the force depending on a compound subordinator. It is coupled because of the α -stable Lévy process directed by its inverse. The fractional two-power-law relaxation can also be described in the framework of coupled CTRWs based on subordination of a stochastic process with the heavy-tailed distribution of the waiting times by its inverse [29,30]. Although the papers have a different physical background, they intersect into the application of the coupling between the α -stable Lévy process and its inverse. The main feature of the coupled CTRWs is that it permits one to study physical processes with different power dependencies for short and long times. Undoubtedly, this new random process (subordinator) is of an essential interest for understanding of the anomalous relaxation phenomena and was investigated insufficiently yet. In this paper we are going to make up for the deficiency.

We summarize here our main results, which are fivefold: (i) we introduce in Sec. II notion of the compound subordinator and we show that from the α -stable Lévy process $U_{\alpha}(\tau)$ and its inverse S_t two different compound subordinators can be realized to under- and overestimate the real time t; (ii) following the concept [30] in Sec. III we develop comprehensively a model of anomalous diffusion based on the compound subordinators; (iii) the undershooting subordination leads to subdiffusion, and the overshooting subordination demonstrates a superdiffusive behavior; (iv) we derive diffusion equations corresponding to each case separately; and (v) in Sec. IV we establish that the studied anomalous diffusion is a semimartingale. Section V sums up our investigations.

II. COUPLING BETWEEN THE VERY LARGE JUMPS IN PHYSICAL AND OPERATIONAL TIMES

The probability density of the position vector $\mathbf{r}_t = \mathbf{B}_{S_t}$ (where \mathbf{B}_{τ} is the standard Brownian motion) can be found from a weighted integration of the joint probability density of the couple ($\mathbf{R}_{\tau}, T_{\tau}$) over the internal time parameter τ by subordination. The stochastic time evolution T_{τ} and its (left) inverse process S_t permit one to underestimate or overestimate the physical time *t*.

The sum of iid heavy-tailed random variables T_i

$$\operatorname{Prob}(T_i \ge t) \sim \left(\frac{t}{t_0}\right)^{-\alpha} \quad \text{as} \ t \to \infty, \tag{1}$$

with $0 < \alpha < 1$ and $t_0 > 0$ converges to a stable random variable in distribution as the number of summands tends to infinity. Let $U_n = \sum_{i=0}^n T_i$ with $T_0 = 0$. The counting process $N_t = \max\{n \in \mathbf{N} | U_n \le t\}$ is inverse to U_n which can be defined equivalently as the process satisfying

$$U_{N_t} \le t < U_{N_t+1} \quad \text{for } t \ge 0,$$
 (2)

which follows directly from its definition. In fact, the two processes, U_{N_t} and U_{N_t+1} , correspond to underestimating and overestimating the real time *t* from the random time steps T_i of the CTRWs.

In terminology of Feller's book [31] the variable $Z_t = U_{N_t+1} - t$ is the residual waiting time (lifetime) at the epoch *t* and $Y_t = t - U_{N_t}$ is the spent waiting time (age of the object that is alive at time *t*). The importance of these variables can be explained by one remarkable property. For $t \rightarrow \infty$ the variables Y_t and Z_t have a common proper limit distribution only if their probability distributions F(y) and F(z) have finite expectations. However, if the distribution F(x) satisfies $1 - F(x) = x^{-\alpha}L(x)$, where $0 < \alpha < 1$ and $L(xt)/L(t) \rightarrow 1$ as $x \rightarrow \infty$, then according to [32], the probability density function (pdf) of the normalized variable Y_t/t is given by the generalized arc sine law,

$$p_{\alpha}(x) = \frac{\sin(\pi\alpha)}{\pi} x^{-\alpha} (1-x)^{\alpha-1}, \qquad (3)$$

while Z_t/t obeys

$$q_{\alpha}(x) = \frac{\sin(\pi\alpha)}{\pi} x^{-\alpha} (1+x)^{-1}.$$
 (4)

Since $\sum_{N_t} = t - Y_t$ and $\sum_{N_t+1} = Z_t + t$, the distributions of $\sum_{N_t/t}$ and \sum_{N_t+1}/t can be obtained from Eqs. (3) and (4) by a simple change of variables 1 - x = y and 1 + x = z, respectively.

We now return to the processes U_{N_t} and U_{N_t+1} introduced above. Recall that T_i are iid positive random variables with long-tailed distribution (1). In this case U_{N_t}/t tends in distri-

bution (\rightarrow) in the long-time limit to random variable *Y* with density

$$p^{Y}(x) = \frac{\sin(\pi\alpha)}{\pi} x^{\alpha-1} (1-x)^{-\alpha}, \quad 0 < x < 1$$
 (5)

and $U_{N,+1}/t \rightarrow Z$ with the pdf equal to

$$p^{Z}(x) = \frac{\sin(\pi\alpha)}{\pi} x^{-1} (x-1)^{-\alpha}, \quad x > 1.$$
 (6)

The functions $p^{Y}(x)$ and $p^{Z}(x)$ correspond to special cases of the well-known beta density. It should be noticed that the density $p^{Y}(x)$ concentrates near 0 and 1, whereas $p^{Z}(x)$ does near 1. Near 1 both tend to infinity. This means that in the long-time limit the most probable values for U_{N_t} occur near 0 and 1, while for $U_{N,+1}$ they tend to be situated near 1.

As a consequence, the random variable Y has finite moments of any order. They can be calculated directly from density (5) and take the form

$$\langle Y \rangle = \alpha, \quad \langle Y^2 \rangle = \frac{\alpha(1+\alpha)}{2}, \dots, \quad \langle Y^n \rangle$$
$$= \frac{\alpha(1+\alpha)\cdots(\alpha+n-1)}{n!},$$

where $n \in \mathbf{N}$ while even the first moment of *Z* diverges. The divergence of U_{N_i+1} results from the long-tail property [Eq. (1)] of the time steps T_i ($\langle T_i \rangle = \infty$), yielding too long overshot above *t*.

Nonequality (2) can also be represented in a schematic picture of time steps, namely, $T_{\tau}(\Delta \tau) = U_{[\tau/\Delta \tau]}$ and $T_{\tau}^{+}(\Delta \tau) = U_{[\tau/\Delta \tau]+1}$, where [x] indicates the integer part of the real number x so that $[x] \le x < [x]+1$. The inverse process of $T_{\tau}^{+}(\Delta \tau)$ is $S_{t}^{+}(\Delta \tau) = \inf\{\tau \ge 0 \mid T_{\tau}^{+}(\Delta \tau) > t\}$ or equivalently $S_{t}^{+}(\Delta \tau) = \Delta \tau N_{t}$. Therefore, in the limit $\Delta \tau \to 0$ the processes $U_{N_{t}}$ and $U_{N_{t}+1}$ can be expressed through the stochastic process $T(\tau)$ subordinated by its inverse,

$${}^{d}U_{N_{t}} \rightarrow T_{S_{t}^{-}}$$
 and $U_{N_{t}+1} \rightarrow T_{S_{t}^{+}}$,

where S_t^- and S_t^+ are the limiting processes to $S_t^-(\Delta \tau)/\Delta \tau = N_t - 1$ and $S_t^+(\Delta \tau)/\Delta \tau = N_t$, respectively. A passage from the discrete process T_i to the continuous one T_{τ} allows one to reformulate inequality (2) as

$$T_{S_t^-} \le t < T_{S_t^+} \quad \text{for } t \ge 0,$$
 (7)

underestimating or overestimating the real time *t*. From Theorem 1.13 in [33] the joint probability density p(y,z) of $T_{S_t^-}$ and $T_{S_t^+}$ with $0 \le T_{S_t^-} \le t < T_{S_t}^+$ takes the form

$$p(y,z) = \frac{\alpha \sin(\pi \alpha)}{\pi} y^{\alpha - 1} (z - y)^{-1 - \alpha}$$
(8)

for $0 \le y \le t < z$. After integrating Eq. (8) with respect to z in the limits $[t, \infty[$ (or with respect to y in the limits [0, t]) we obtain the densities of $T_{S_t^-}$ and $T_{S_t^+}$, respectively,

$$p^{-}(t,y) = \frac{\sin \pi \alpha}{\pi} y^{\alpha - 1} (t - y)^{-\alpha}, \quad 0 < y < t, \tag{9}$$

$$p^{+}(t,z) = \frac{\sin \pi \alpha}{\pi} z^{-1} t^{\alpha} (z-t)^{-\alpha}, \quad z > t,$$
(10)

valid at all times $t \ge 0$ (see Fig. 1). The moments of $T_{S_t^-}$ and $T_{S_t^+}$ can be calculated directly from the moments of Y and Z by using relations



FIG. 1. (Color online) The probability density $p^{-}(y)$ with support on 0 < y < t and the density $p^{+}(y)$ with support on y > t for different values of the index α .

$$T_{S_t^-} = tY$$
 and $T_{S_t^+} = tZ$,

where = means the equality in distribution. Thus, the process $T_{S_t^-}$ has finite moments of any order, while $T_{S_t^+}$ gives us even no finite the first moment. The overshot of $T_{S_t^+} > t$ is too long also in the limit formulation. Notice that $p^+(t,y) = y^{-2}p^-(t^{-1},y^{-1})$. At this point we should mention that compound subordinators and, in particular, the subordination by an inverse Lévy-stable process via a Lévy-stable process were considered already in [34]. However, the construction of compound subordinators has been based on the statistically independent stochastic processes. This leads to quite different results in comparison with ours. In our construction of the compound subordinators $T_{S_t^-}$ and $T_{S_t^+}$ the processes U_t and S(t) are clearly coupled.

III. ANOMALOUS DIFFUSION WITH UNDER- AND OVERSHOOTING SUBORDINATION

According to [30], the widely observed fractional twopower relaxation dependencies

$$\chi(\omega) \sim (i\omega/\omega_{\rm p})^{n-1}$$
 for $\omega \gg \omega_{\rm p}$ (11)

and

d

$$\Delta \chi(\omega) \sim (i\omega/\omega_{\rm p})^m \quad \text{for } \omega \ll \omega_{\rm p}$$
 (12)

of the complex susceptibility $\chi(\omega) = \chi'(\omega) - i\chi''(\omega)$, where $\Delta\chi(\omega) = \chi(0) - \chi(\omega)$, the exponent *n* and *m* fall in the range (0,1), and ω_p denotes the loss peak frequency, are closely connected with the under- and overshooting subordination,

$$Z^{U}_{\alpha,\gamma}(t) \le S_{\alpha}(t) < Z^{O}_{\alpha,\gamma}(t) \quad \text{for } t \ge 0,$$

where $Z_{\alpha,\gamma}^{U}(t) = X_{\gamma}^{U}(S_{\alpha}(t))$ and $Z_{\alpha,\gamma}^{O}(t) = X_{\gamma}^{O}(S_{\alpha}(t))$. Here the processes $X_{\gamma}^{U}(t)$ and $X_{\gamma}^{O}(t)$ are nothing else as $T_{S_{t}^{-}}$ and $T_{S_{t}^{+}}$ with the index γ . They are subordinated by an independent inverse α -stable process $S_{\alpha}(t)$ forming the compound subordinators $Z_{\alpha,\gamma}^{U}(t)$ and $Z_{\alpha,\gamma}^{O}(t)$, respectively. The approach enlarges the class of diffusive scenarios in the framework of the

CTRWs. This new type of coupled CTRWs follows from the clustering-jump random walks idea [35]. As it has been rigorously proved [36], the clustering with finite-mean-value cluster sizes leads to the classical decoupled CTRW models, but assuming a heavy-tailed cluster-size distribution with the tail exponent $0 < \gamma < 1$, the coupling between jumps and interjump times tends to the compound operational times $Z_{\alpha,\gamma}^U(t)$ as under- and overshooting subordinators, respectively.

It should be noticed that the physical mechanism underlying the anomalous two-power-law relaxation can be described as a diffusive limit of CTRWs. The resulting relaxation patterns are connected not only with stochastic features of the jumps and the interjump times themselves but also with a stochastic dependence between them. In the framework of linear response theory the temporal decay of a given mode k, representing excitation undergoing diffusion in the system under consideration, is given by the inverse Fourier transform of the diffusion front [37].

On the other hand, the relaxation evolution of complex materials is realized on structural different levels (relaxing entities, e.g., dipoles, clusters, and cooperative regions). The levels can interact between each other, as well as the time evolution toward equilibrium of each of them can be governed by different random processes. To obtain any fractional two-power relaxation law, the diffusion scenario should utilize such a mathematical tool (i.e., the subordination approach) by means of which we are able to incorporate the above. The simplest subordination of a Brownian motion by an inverse α -stable process accounts for the amount of time when a relaxator (dipole) does not participate in a motion, i.e., the motion of relaxators to equilibrium is not uniform. This scenario leads to the Mittag-Leffler (Cole-Cole) relaxation only. The more general Havriliak-Negami (HN) relaxation law, fitting most of the empirical relaxation data, follows, however, from compound subordination. In this case, the anomalous diffusion process combines coupling between the stable process as the real time and its inverse as an operational time. Such a compound subordination helps us to present relaxation in complex systems as a random process in which the interacting dipoles, clusters, and cooperative regions participate at the same time.

The overshooting subordinator yields the anomalous diffusion scenario leading to the well-known HN relaxation pattern [38], and the undershooting subordinator leads to a new relaxation law given by the generalized Mittag-Leffler relaxation function [29,30]. These results are in agreement with the idea of a superposition of the classical (exponential) Debye relaxations. Thus, the stochastic mechanism underlying the anomalous relaxation is quite clear, but the corresponding diffusion analysis requires some additional clarity. Let B(t)be the parent process that is subordinated either by $Z^U_{\alpha,\gamma}(t)$ or $Z^O_{\alpha,\gamma}(t)$. Then the subordination relation, expressed by means of a mixture of pdfs, takes the form

$$p^{r}(x,t) = \int_{0}^{\infty} \int_{0}^{\infty} p^{B}(x,y) p^{\pm}(y,\tau) p^{S}(\tau,t) dy d\tau, \quad (13)$$

where $p^{r}(x,t)$ is the probability density of the subordinated process $B(Z_{\alpha,\gamma}^{U}(t))$ [or $B(Z_{\alpha,\gamma}^{O}(t))$] with respect to the coordi-

nate x and time t, $p^{B}(x, \tau)$ is the probability density of the parent process, $p^{\pm}(y, \tau)$ is the probability density of $T_{S_{t}^{-}}$ and $T_{S_{t}^{+}}$, respectively, and $p^{S}(\tau, t)$ is the probability density of S(t). Recall that for the subdiffusion B(S(t)), by taking the Laplace transform from the corresponding subordination relation, we can derive the celebrated fractional Fokker-Planck equation [37]. It is therefore reasonable to ask is it possible to find a diffusion equation corresponding to relation (13). In the Laplace space

$$\overline{f}(u) = \int_0^\infty e^{-ut} f(t) dt$$

we obtain

$$\overline{p}^{r}(x,u) = u^{\alpha-1} \int_{1}^{\infty} \overline{p}^{B}(x,u^{\alpha}/z) p_{0}^{+}(z) \frac{dz}{z},$$
 (14)

with $p_0^+(z) = \sin(\pi \gamma) z^{-1} (z-1)^{-\gamma} / \pi$ for z > 1, as well as

$$\bar{p}^{r}(x,u) = u^{\alpha-1} \int_{0}^{1} \bar{p}^{B}(x,u^{\alpha/z}) p_{0}^{-}(z) \frac{dz}{z},$$
(15)

with $p_0^{-}(z) = \sin(\pi \gamma) z^{\gamma-1} (1-z)^{-\gamma} / \pi$ for 0 < z < 1. The Laplace image of the pdf of the subordinated process B(S(t)) can be simply expressed in terms of an algebraic form with the Laplace image of the parent process pdf. This allows one to get the fractional Fokker-Planck equation driving the spatiotemporal evolution of the propagator of the anomalous diffusion underlying the Mittag-Leffler relaxation [14,26,37]. However, expressions (14) and (15) are not similar to the latter. They have an integral form. Nevertheless, derivation of the corresponding Fokker-Planck equation is also possible.

If we take the Laplace transform with respect to t and the Fourier transform with respect to x for $p^r(x,t)$ in Eq. (13), the Fourier-Laplace (FL) image reads

$$\mathcal{I}_{\mathbf{FL}}(p^r)(k,s) = s^{\alpha-1} \int_0^\infty \int_0^\infty e^{-\psi(k)y} p^{\pm}(y,\tau) e^{-\tau s^{\alpha}} dy d\tau,$$
(16)

where $\psi(k)$ is the logarithmic-Fourier transform of the parent process pdf $p^B(x,y)$. Consider the case of $p^-(y,\tau)$. After changing variables $y=z\tau$ we take the integral

$$\int_0^\infty e^{-\tau(s^\alpha + \psi(k)z)} d\tau = \frac{1}{s^\alpha + \psi(k)z}.$$

Next, the change of variables t=z/(1-z) maps [0,1] onto $[0,\infty)$. This helps us to derive

$$\mathcal{I}_{\mathbf{FL}}(p^r)(k,s) = \frac{s^{\alpha-1}}{\Gamma(\gamma)\Gamma(1-\gamma)} \int_0^\infty \frac{t^{\gamma-1}dt}{(s^\alpha + \psi(k))t + s^\alpha}.$$

The last expression can be easily calculated from the integral [39]

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$$\int_0^\infty \frac{t^{\gamma-1}}{t+1} dt = \Gamma(\gamma) \Gamma(1-\gamma).$$

The FL image of $p^r(x,t)$ with the undershooting directing process $Z^U_{\alpha,\gamma}(t) = X^U_{\gamma}(S_{\alpha}(t))$ is of the form

$$\mathcal{I}_{\mathbf{FL}}(p^r)(k,s) = \frac{s^{\alpha\gamma - 1}}{(s^\alpha + \psi(k))^\gamma}.$$
(17)

Finally, we invert the Fourier and Laplace transforms to get the pseudodifferential diffusion equation

$$\left(\frac{\partial^{\alpha}}{\partial t^{\alpha}} + L_{\rm FP}(x)\right)^{\gamma} p^{r}(x,t) = \delta(x) \frac{t^{-\alpha\gamma}}{\Gamma(1-\alpha\gamma)},\tag{18}$$

where $L_{\rm FP}(x)$ is the Fokker-Planck operator, $\delta(x)$ is the Dirac function, and $\partial^{\alpha}/\partial t^{\alpha}$ denotes the Riemann-Louiville derivative. The corresponding Fokker-Planck equation can also be obtained in the case when the overshooting directing process $Z^O_{\alpha,\gamma}(t) = X^O_{\gamma}(S_{\alpha}(t))$ is taken into account. Unfortunately, the derivation is more complicated as we present below.

In the case of $p^+(y, \tau)$, after the substitution $y=z\tau$, we map $[1,\infty)$ onto [0,1] by the change of variables z=1/x. Then we obtain the corresponding FL image,

$$\mathcal{I}_{\mathbf{FL}}(p^{r})(k,s) = \frac{s^{\alpha-1}}{\Gamma(\gamma)\Gamma(1-\gamma)} \int_{0}^{1} \frac{x^{\gamma-1}(1-x)^{-\gamma}dt}{s^{\alpha} + \psi(k)/x}.$$

The mapping t=x/(1-x) transforms the latter expression to the form

$$\mathcal{I}_{\mathbf{FL}}(p^r)(k,s) = \frac{s^{\alpha-1}}{\Gamma(\gamma)\Gamma(1-\gamma)} \int_0^\infty \frac{t^{\gamma}dt}{(1+t)[(s^{\alpha}+\psi(k))t+\psi(k)]}.$$

This integral can be calculated exactly,

$$\int_0^\infty \frac{t^{\gamma}}{(t+1)(at+b)} dt = \frac{\Gamma(\gamma)\Gamma(1-\gamma)}{(a-b)} [1-(b/a)^{\gamma}].$$

As a result, the FL image of $p^r(x,t)$ with the directing process $Z^O_{\alpha,\gamma}(t) = X^O_{\gamma}(S_{\alpha}(t))$ can be written as

$$\mathcal{I}_{\mathbf{FL}}(p^r)(k,s) = \frac{1}{s} \left\{ 1 - \left(\frac{\psi(k)}{s^{\alpha} + \psi(k)}\right)^{\gamma} \right\}.$$
 (19)

Now we invert the Fourier and Laplace transforms to get the pseudodifferential diffusion equation,

$$\left(\frac{\partial^{\alpha}}{\partial t^{\alpha}} + L_{\rm FP}(x)\right)^{\gamma} p^{r}(x,t) = f_{\alpha,\gamma}(x,t), \qquad (20)$$

where

$$f_{\alpha,\gamma}(x,t) = \left\{ \left(\frac{\partial^{\alpha}}{\partial t^{\alpha}} + L_{\rm FP}(x) \right)^{\gamma} - (L_{\rm FP}(x))^{\gamma} \right\} \delta(x)$$

is a function depending on the probability density $p^B(x,y)$. The exact form of $f_{\alpha,\gamma}(x,t)$ is quite different from the rightside term of Eq. (18). In this connection it should be pointed out the work [40] where the derivation of a fractional Fokker-Planck underlying the HN type of relaxation is based on the entirely phenomenological approach in [41]. However, the stochastic background leading to the anomalous dif-



FIG. 2. (Color online) Propagator $p^r(x,t)$ of the under- and overshooting anomalous diffusion with a constant potential, $\alpha = 2/3$ and $\gamma = 2/3$, drawn for consecutive dimensionless instances of time t=1,3,10. The cusp shape of the pdfs appears.

fusion yielding the HN pattern has remained behind these works.

To calculate the moments of the processes $B(Z^U_{\alpha,\gamma}(t))$ and $B(Z^O_{\alpha,\gamma}(t))$, assume for simplicity that the parent process *B* is a one-dimensional Brownian motion. Its moments are written as

$$I_{2n}(t) = \frac{1}{\sqrt{4\pi Dt}} \int_{-\infty}^{\infty} x^{2n} \exp\left(-\frac{x^2}{4Dt}\right) dx = \frac{(2n)!}{n!} (Dt)^n,$$

where *D* is the diffusion coefficient. If the subordinator $Z^{U}_{\alpha,\gamma}(t)$ governs the Brownian motion, then the moment integral reads

$$\langle x^{2n} \rangle = \int_{-\infty}^{\infty} x^{2n} p^r(x,t) dx$$

$$= B_n \int_0^1 z^n p_0^{-}(z) dz \int_0^{\infty} \tau^n p^S(\tau,y) d\tau$$

$$= \frac{(2n)!}{n!} D^n \frac{(\gamma,n)}{n!} \frac{t^{n\alpha}}{\Gamma(1+n\alpha)},$$

$$(21)$$

where $(\gamma, n) = \gamma(\gamma+1)(\gamma+2)\cdots(\gamma+n-1)$ is Appell's symbol with $(\gamma, 0) = 1$. When another subordinator $Z^{O}_{\alpha,\gamma}(t)$ is used, even the first moment of the subordinated process $B(Z^{O}_{\alpha,\gamma}(t))$



FIG. 3. (Color online) Left panel: the propagator p'(x,t) of under- (a) and overshooting (b) anomalous diffusion with $\alpha = 2/3$ and $\gamma = 2/3$ for t = 1. The line (c) displays the propagator of ordinary subdiffusion with $\alpha = 2/3$ and $\gamma = 1$ for t = 1. Right panel: diagram shows the interrelation between $B(Z_{\alpha,\gamma}^U(t))$, $B(S_{\alpha}(t))$, and $B(Z_{\alpha,\gamma}^O(t))$. Here *m* and 1-n denote the power-law exponents defined in formulas (11) and (12).

diverges because the probability density $p_0^+(z)$ gives no finite moments. Thus, the process $B(Z_{\alpha,\gamma}^U(t))$ is a subdiffusion and $B(Z_{\alpha,\gamma}^O(t))$ is a superdiffusion. In Fig. 2, as an example, the propagator $p^r(x,t)$ for the under- and overshooting anomalous diffusion with $\alpha = 2/3$ and $\gamma = 2/3$ is drawn.

It should be noticed that the ordinary subdiffusion $B(S_{\alpha}(t))$ takes an intermediate place between the under- and overshooting anomalous diffusion $B(Z_{\alpha,\gamma}^U(t))$ and $B(Z_{\alpha,\gamma}^O(t))$. The feature is illustrated in Fig. 3. This allows one to compare an asymptotic behavior of the temporal evolution of diffusion fronts. From that one can see that the diffusion front of $B(Z_{\alpha,\gamma}^U(t))$ is more stretched than the front of $B(S_{\alpha}(t))$, whereas the diffusion front of $B(Z_{\alpha,\gamma}^O(t))$ is more contracted in comparison with the front of $B(S_{\alpha}(t))$.

One of interesting questions is what interpretation can be assigned to the subordinators $Z^U_{\alpha,\gamma}(t) = X^U_{\gamma}(S_{\alpha}(t))$ and $Z^O_{\alpha,\gamma}(t) = X^O_{\gamma}(S_{\alpha}(t))$. As the processes $X^U_{\gamma}(\tau)$ and $X^O_{\gamma}(\tau)$ are independent on $S_{\alpha}(t)$, they can be considered separately. The inverse Lévy-stable process $S_{\alpha}(t)$ accounts for the amount of time when a walker does not participate in motion. The pdf of the subordinated process $B(X^U_{\gamma}(\tau))$ is a special case of the Dirichlet average, namely,

$$F(\gamma, x, \tau) = \frac{\sin \pi \alpha}{\pi} \int_0^1 p^B(x, \tau z) z^{\gamma - 1} (1 - z)^{-\gamma} dz.$$

Recall that many of important special and elementary functions can be represented as Dirichlet averages of continuous functions (see more details in [42]). The Dirichlet average includes the well-known means (arithmetic, geometric, and others) as special cases. The process $X_{\gamma}^{U}(t)$ evolves to infinity like time *t*. Its contribution in the subordinated process $B(X_{\gamma}^{U}(t))$ is taken into account by the Dirichlet average of the probability density of the parent process *B*. The similar reasoning can be developed for the process $X_{\gamma}^{O}(t)$.

IV. LANGEVIN EQUATION WITH COMPOUND SUBORDINATORS

Description of the anomalous diffusion with under- and overshooting subordination in the forms of Eqs. (18) and (20) is not unique. These diffusion processes can be repre-

sented equivalently in the language of Langevin equation. As it is well known, the ordinary Brownian motion satisfies the stochastic differential equation (called the Langevin equation),

$$dR(\tau) = F(R(\tau), \tau)d\tau + G(R(\tau), \tau)d\mathcal{B}_{\tau},$$

where $R(\tau)$ describes the space position of a diffusing particle and *F* and *G* are some functions. The Brownian diffusion with a drift is a semimartingale, where the first term describes a local martingale, and the second one a finite variation process [8]. It is interesting that the subordination operation can save the property. Really, the process subordinated to the Brownian motion $R(S_t)=\mathcal{B}_{S_t}$ is a semimartingale and obeys the following equation:

$$d\mathbf{r}_t = F(\mathbf{r}_t)dS_t + G(\mathbf{r}_t)d\mathcal{B}_{S_t},$$

where \mathbf{r}_t is the position vector with $t \ge 0$. From the fact that the processes $Z_{\alpha,\gamma}^U(t)$ and $Z_{\alpha,\gamma}^O(t)$ bound the subordinator S_t , i.e., $Z_{\alpha,\gamma}^U(t) \le S_{\alpha}(t) < Z_{\alpha,\gamma}^O(t)$, the Langevin equation framework holds also for the compound subordinators studied in the present work. This means that the corresponding Langevin equation reads

$$d\mathbf{r}_{t} = F(\mathbf{r}_{t})dZ_{\alpha,\gamma}^{U}(t) + G(\mathbf{r}_{t})d\mathcal{B}_{Z_{\alpha,\gamma}^{U}(t)},$$
(22)

and the similar equation can also be written for the case of $Z_{\alpha,\gamma}^{0}(t)$. Since the mathematical theory of semimartingales is well developed, it can help us to understand better various properties of the anomalous diffusion. In particular, the trajectories of semimartingales are always of finite quadratic variation. If, e.g., Y(t) is a stochastic process observed on time interval $t \in [0,T]$, then the quadratic variation $V^{(2)}(t)$, corresponding to Y(t), is defined as $V^{(2)}(t) = \lim_{n\to\infty} V_n^{(2)}(t)$, where $V_n^{(2)}(t)$ is the partial sum of the squares of increments of the process Y(t) given by

$$V_n^{(2)}(t) = \sum_{j=0}^{2^n - 1} |Y(T(j+1)/2^n \wedge t) - Y(Tj/2^n \wedge t)|^2$$

with $a \wedge b = \min\{a, b\}$.

According to [30], using the self-similar properties of the subordinators and their independency from the parent process $B(\tau)$, we have

$$B(Z^{U}_{\alpha,\gamma}(t)) \stackrel{d}{=} (Z^{U}_{\alpha,\gamma}(t))^{1/2} B(1) \stackrel{d}{=} t^{\alpha/2} (L_{\alpha}(1))^{-\alpha/2} (X^{U}_{\gamma}(1))^{1/2} B(1),$$

$$B(Z^{O}_{\alpha,\gamma}(t)) \stackrel{d}{=} (Z^{O}_{\alpha,\gamma}(t))^{1/2} B(1) \stackrel{d}{=} t^{\alpha/2} (L_{\alpha}(1))^{-\alpha/2} (X^{O}_{\gamma}(1))^{1/2} B(1),$$

where the Brownian motion $B(\tau)$ is 1/2-self-similar [and hence $B(\tau) = \tau^{1/2}B(1)$], the α -stable Lévy motion $L_{\alpha}(\tau)$ yields $L_{\alpha}(\tau) = \tau^{1/\alpha}L_{\alpha}(1)$, and $X_{\gamma}^{O}(1) = 1/X_{\gamma}^{U}(1)$. Thus, both processes $B(Z_{\alpha,\gamma}^{U}(t))$ and $B(Z_{\alpha,\gamma}^{O}(t))$ are semimartingales. The result is

 $B(Z_{\alpha,\gamma}^{o}(t))$ and $B(Z_{\alpha,\gamma}^{o}(t))$ are semimartingales. The result is very important because the different behavior of quadratic variations can be used to construct efficient statistical tests which distinguish different types of anomalous dynamics in experimental data [15–17,28].

V. CONCLUSIONS

The paper introduces an approach to study the coupling between the very large jumps in physical and operational times. It is based on the compound subordination of a Lévy-stable process $T(\tau)$ by its inverse S(t). The inverse Lévy-stable process is actually the left-inverse process of the Lévy-stable one. In fact, we have $S(T(\tau)) = \tau$, while T(S(t)) > t holds. In the framework of CTRWs the compound subordinator provides a direct coupling of physical and operational times. The subordination scenario leads to two types of the operational time: the spent lifetime and the residual age. In the first random process all the moments are finite, whereas the second process has no finite moments. We have shown that the approach is useful for analysis of anomalous diffusion underlying all empirical fractional twopower-law relaxation responses. Due to the two types of the operational time the diffusion can display as well the subdiffusive and superdiffusive characters. The anomalous diffusion with under- and overshooting subordination discovers a novel law of relaxation accompanied by the well-known HN function [43]. The point is that the original HN relaxation [38] with exponents $0 < \alpha$, $\gamma \le 1$ satisfies $m \ge 1-n$. Its

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modified version [44], proposed to fit relaxation data with power-law exponents satisfying m < 1-n, assumes $0 < \alpha$, $\alpha \gamma \le 1$. Unfortunately, the HN function with $\gamma > 1$ cannot be derived within the framework of diffusive relaxation mechanisms. The problem succeeds in overcoming by the undershooting subordination [30]. The difference in physical mechanisms underlying the typical (m > 1-n) and the less typical (m < 1-n) sets of the two-power-law relaxation responses is clearly seen in behavior of the corresponding operational times. In the language of subordinators the process $X_{\gamma}^{U}(t)$ makes a rescaling for small times resulting in "compressing" the operational time characteristic for the Mittag-Leffler relaxation [i.e., the inverse Lévy-stable process $S_{\alpha}(t)$] while the process $X_{\gamma}^{O}(t)$ turns on a similar rescaling for long time "stretching" hence $S_{\alpha}(t)$.

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