Generalized Mittag-Leffler relaxation: Clustering-jump continuous-time random walk approach

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A stochastic generalization of renormalization-group transformation for continuous-time random walk processes is proposed. The renormalization consists in replacing the jump events from a randomly sized cluster by a single renormalized (i.e., overall) jump. The clustering of the jumps, followed by the corresponding transformation of the interjump time intervals, yields a new class of coupled continuous-time random walks which, applied to modeling of relaxation, lead to the general power-law properties usually fitted with the empirical Havriliak-Negami function.

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I. INTRODUCTION

Continuous-time random walk (CTRW), introduced by Montroll and Weiss more than 40 years ago [1], has proven to be a powerful mathematical tool for description and analysis of relaxation and transport phenomena in complex systems in which the temporal evolution strongly deviates from the corresponding standard laws: Relaxation from the classical (exponential) Debye law, and diffusion from the normal one given by the Gaussian statistics. Today, the list of phenomena displaying anomalous dynamical behavior is quite extensive. It contains examples such as charge carrier transport in amorphous semiconductors, rebinding kinetics in proteins, NMR diffusometry in percolative and porous media, transport on fractal geometries, diffusion of contaminants in complex geological formations, diffusion of pollutants in large ecosystems, or transport in micelle systems (for a detailed list see [2] and references therein). Usually, in physical applications of the CTRW methodology, analysis of the diffusion front properties is presented within the classical Montroll-Weiss approach [1–3] that is based on a formal expression for the Fourier-Laplace transform of the total distance reached at time $t \geq 0$ by a randomly moving particle. Alternatively, the fractional calculus is proposed as a legitimate tool (see, e.g., [2,4–7]). In such approaches, explicit formulas can be provided only under some restrictive assumptions on the spatiotemporal random walk characteristics. Here, we present the random-variable approach [8–10] which is based directly on the definition of the CTRW as a cumulative stochastic process. Our aim is to show that despite the extensive studies on CTRWs, and their long history in physics, they have not been fully explored yet [11–19]. We show how the CTRW tool can be generalized to handle complicated diffusive situations. In particular, we are interested in a diffusion scenario which can lead to a class of power-law functions able to describe all cases of the empirical "universal relaxation response" [20].

The CTRW process is characterized by a sequence $\{(R_i, T_i)\}_{i \geq 1}$ of independent and identically distributed (i.i.d.) random vectors indicating the length and the direction of the subsequent jumps (by means of R_i), as well as the waiting time, T_i, between them [8–10]. The case when we assume stochastic independence between the space steps, R_i, and the time steps, T_i, is referred to as a decoupled CTRW; otherwise we deal with a coupled CTRW. The total distance, $R(t)$, reached at time t is equal to the random sum of space steps, R_i, with the random number of summands given by the counting process $\nu(t)$,

$$R(t) = \sum_{i=1}^{\nu(t)} R_i. \tag{1}$$

The counting process $\nu(t)$ is determined by the waiting times T_i in the following way $\nu(t) = \max\{n : \Sigma_{i=1}^{n} T_i \leq t\}$. The value of $\nu(t)$ is equal to n if we need exactly $n+1$ time steps for exceeding t, and the probability distribution of $\nu(t)$ can be easily expressed in terms of the waiting-time distribution $F_T(t) = \Pr(T_i \leq t)$, since we have $\Pr[\nu(t) = n] = F_T^{*n}(t) - F_T^{*(n+1)}(t)$, where F_T^{*n} is the convolution of n identical distribution functions F_T. Moreover, in the decoupled-CTRW case the process $\{\nu(t)\}_{t \geq 0}$ is independent of the space-step sequence $\{R_i\}_{i \geq 1}$, and as a consequence the Fourier transform of (1) reads as

$$\langle e^{ikR(t)} \rangle = \sum_{n=0}^{\infty} (\langle e^{ikR_1} \rangle)^n \Pr[\nu(t) = n] = g_{\nu(t)}[\varphi_R(k)],$$

where $g_{\nu(t)}(z) = \Sigma_{n=1}^{\infty} z^n P[\nu(t) = n]$ is the generating function of the random index $\nu(t)$, and $\varphi_R(k) = \langle e^{ikR_1} \rangle$.

Important and well-known examples of the decoupled CTRW's are the compound Poisson process [21] and the Lévy flight [2]. The first corresponds to the exponentially distributed waiting times T_i and is known to have independent and stationary increments. The second refers to the case when the mean value $\langle T_i \rangle$ of the waiting times is finite, and the space steps R_i are symmetric Lévy-stable random variables [22–24] with the index of stability α falling in the range (1,2) so that the mean $\langle R_i \rangle = 0$ and the variance $D^2 R_i = \infty$. Among the coupled CTRW's the most popular is the Lévy walk [2,25,26], obtained for $R_i = Y_i T_i^a + m$, where $a \geq 0$ and m are constants; $\{Y_i\}_{i \geq 1}$ is a sequence of i.i.d. ran-

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dom variables, independent of $\{T_i\}_{i\geq 1}$, such that $\Pr(Y_i=1)=1-\Pr(Y_i=-1)=p$ for some $0<p<1$; and the waiting times are such that for some $0<\alpha<2$ the tails satisfy $\Pr(T_i>x) \underset{x\to\infty}{\propto} x^{-\alpha}$ [where symbol "$f(x) \underset{x\to\infty}{\propto} g(x)$" means that $\lim_{x\to\infty}\frac{f(x)}{g(x)}=\text{const}>0$].

The general properties of different possible types of process (1), as well as, related to them the Debye or Mittag-Leffler relaxation patterns are by now well explored, and what has been also well established is their connection to fractional diffusion or Fokker-Planck equations [2,4–8,16–19]. The related studies [8–10] on the cumulative processes concern, in fact, the limiting behavior of the total distance $R(t)$. In case of the decoupled CTRW (1), with power-law jump or waiting-time distributions, the limiting distributions of $R(t)$ have been shown [26] all strictly related to the Lévy-stable laws (i.e., to the Lévy-stable laws themselves or to the *trans*-stable and fractional stable distributions [22,27]). The intimate relation between the CTRW with power-law waiting-time distributions and Lévy-stable laws was clear practically from the very beginning and was stressed in the approach based on the concept of fractal time [28,29]. The relation with the Lévy-stable laws is connected with the theory of limit theorems for sums of i.i.d. random variables [21–24,30], which allows us to answer the question of asymptotic behavior for different kinds of CTRW's. The limit theorems help us to handle not only with diffusive situations based on the most celebrated Lévy flights or Lévy walks but also with more complicated situations [12,13].

In this paper, in Sec. II, we present a stochastic analog of renormalization-group transformation for the CTRW's. This concept has been introduced [28] by analogy with the theory of critical phenomena [31] for fractal stochastic processes, used in physics for description of the anomalous dynamical behavior of the complex systems. The renormalizationlike transformations have been shown to possess common features with the dynamical behavior of hierarchical systems [32]. As a consequence they lead to scale invariance and hence to the absence of a fundamentally space or time scale of the studied processes. The hierarchical clustering-jump transformation [33] appears to be very useful for the analysis of branched chain processes, describing a broad class of growth phenomena from physics, chemistry, and biology (such as nuclear or chemical chain reactions, high-energy hadron collisions, stimulated emission of photons, polymer or crack growth, population growth, growth of cellular aggregates, etc., see references in [33]); applied to the CTRW [32] has led to the class of diffusion fronts related strictly with the Lévy-stable laws. The renormalization transformation $\mathbf{T}_N$ of a sequence $\{X_i\}_{i\geq 1}$ of i.i.d. random variables defined as $\mathbf{T}_N(\{X_i\})=\{\overline{X_{j,N}}\}_{j\geq 1}$, where the block size N is a positive integer number and

$$\overline{X_{j,N}}=\frac{1}{N^{\delta}}\sum_{i=(j-1)N+1}^{jN} X_i, \tag{2}$$

can be also treated as a way to characterize stable distributions [34]. Namely, the only fixed point for transformations $\mathbf{T}_N$, $N\geq 1$, with parameter δ is a sequence of independent strictly stable distributed random variables with the index of stability $\alpha=1/\delta$. Moreover, such stable distributions only may appear as the limiting distribution resulting from applications of transformation $\mathbf{T}_N$ with increasing N (or successive applications of the transformation with fixed N). However, the larger class of geometric-stable (or more general ν-stable) laws corresponds to the limiting behavior for the summation scheme with random number of summands [instead of the deterministic block size N in Eq. (2)], and it cannot be obtained as a fixed point [35,36].

In what follows we are interested in the latter, i.e., in properties of a process which is constructed by means of random clustering of the random walker jumps. Our work is motivated by the problem of evaluating possible steplike processes from noisy time series data sets. The importance of this problem has been recently discussed [37] in the context of assembly dynamics of microtubules (i.e., highly dynamic protein polymers forming a crucial part of the cytoskeleton in all eukaryotic cells). The analysis of the growth and shrinkage of microtubules is based on clustering of the noisy displacement time series data, and the results depend on the level of experimental resolutions. In this paper we show that the random clustering of the walker's jumps involves also the clustering of the corresponding interjump time intervals. The sequence of the renormalized spatiotemporal steps defines a new class of the coupled CTRW's, different than the well-known Lévy walks. Using then the powerful tool of limit theorems, see the Appendix, we study the diffusion front of the renormalized process for waiting-time and symmetric-jump distributions, both from the domains of attraction of the appropriate Lévy-stable laws. We show that the properties of the resulting process depend on the way in which the jumps are clustered. Our goal is to enlarge the class of the relaxation responses derived yet in the continuous-time random walk framework. In Sec. III, we present a diffusion scenario which can lead not only to the well-known Mittag-Leffler (or to the corresponding Cole-Cole) relaxation pattern but also to a more general two-power-law behavior [20] consistent with the properties of the empirical Havriliak-Negami function.

II. COUPLED CTRW GIVEN BY RANDOM RENORMALIZATION-GROUP TRANSFORMATION

To enlarge the class of the total-distance asymptotics given in the classical CTRW framework, we propose generalization of the compound-counting-process idea [13]. The construction of such processes involves agglutination of a random number of walker's spatiotemporal subsequent steps or, in other words, introduction of a stochastic analog of the renormalization-group transformation [34].

Let us consider an analog of process (1),

$$R_M(t)=\sum_{i=1}^{\mu_M(t)} R_i, \tag{3}$$

which differs from Eq. (1) in the number of summands that now [instead by the renewal counting process $\nu(t)$] is given by a compound counting process $\{\mu^M(t),t\geq 0\}$ defined as

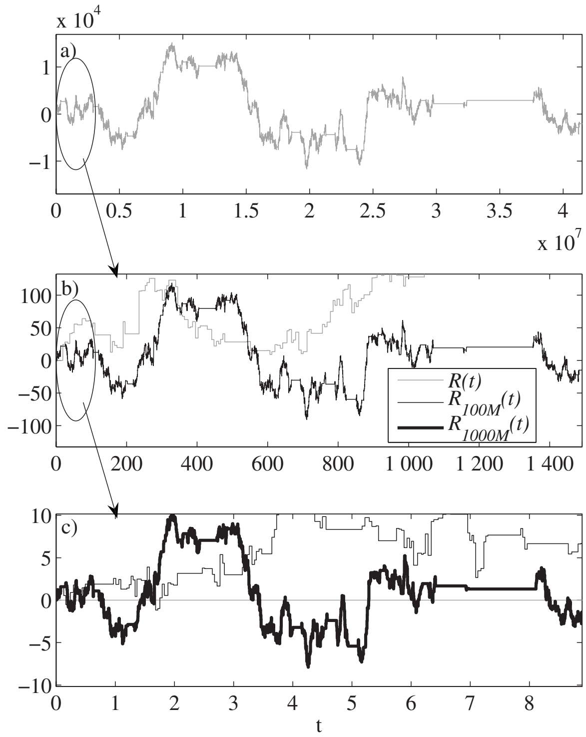

FIG. 1. Exemplary trajectory of the transformed CTRW: The classical renormalization case ($M_j=1$).

$$\mu_M(t)=\sum_{j=1}^{\nu_M[\nu(t)]} M_j \tag{4}$$

for $\nu_M(m)=\max\{n:\sum_{j=1}^{n} M_j \le m\}$. We assume that $\{M_j\}_{j\ge 1}$ is a sequence of i.i.d. positive integer-valued random variables and that this sequence is independent of the family of the spatiotemporal vectors $\{(R_i,T_i)\}_{i\ge 1}$. The construction of processes (3) and (4) is strictly connected with assembling the jumps and waiting times $\{(R_i,T_i)\}_{i\ge 1}$ into clusters $\{(\overline{R_j},\overline{T_j})\}_{j\ge 1}$ of random sizes $M_1, M_2, \ldots$ by means of the following procedure:

$$\overline{R_1}=\sum_{i=1}^{M_1} R_i,$$

$$\overline{T_1}=\sum_{i=1}^{M_1} T_i,$$

$$\overline{R_j}=\sum_{i=1}^{M_j} R_{i+M_1+\cdots+M_{j-1}},$$

$$\overline{T_j}=\sum_{i=1}^{M_j} T_{i+M_1+\cdots+M_{j-1}}$$

$$\text{for } j\ge 2.$$

The introduced process $R_M(t)$, given by Eq. (3), appears to be a coupled CTRW defined by $\{(\overline{R_j},\overline{T_j})\}_{j\ge 1}$. Hence, it can be expressed by the following formula, equivalent to Eq. (3):

$$R_M(t)=\sum_{j=1}^{\overline{\nu}(t)} \overline{R_j}, \tag{5}$$

where $\overline{\nu}(t)=\max\{n:\sum_{j=1}^{n}\overline{T_j}\le t\}$. The dependence between the jumps $\overline{R_j}$ and the waiting times $\overline{T_j}$ of the coupled process $R^M(t)$ is determined by the distribution of cluster sizes M_j [13].

In order to study properties of the coupled process $R_M(t)$, Eq. (5), we introduce the following transformation of spatiotemporal steps:

$$\overline{R_{1,N}}=\frac{1}{f_R(N)}\sum_{i=1}^{NM_1} R_i,$$

$$\overline{T_{1,N}}=\frac{1}{f_T(N)}\sum_{i=1}^{NM_1} T_i,$$

$$\overline{R_{j,N}}=\frac{1}{f_R(N)}\sum_{i=1}^{NM_j} R_{i+N(M_1+\cdots+M_{j-1})},$$

$$\overline{T_{j,N}}=\frac{1}{f_T(N)}\sum_{i=1}^{NM_j} T_{i+N(M_1+\cdots+M_{j-1})}$$

$$\text{for } j\ge 2, \tag{6}$$

where N is a positive integer cluster-size rescaling constant, and $f_R(N)$ and $f_T(N)$ are appropriately chosen dimensionless space- and time-rescaling functions. Formula (6) describes assembling of the renormalized spatiotemporal steps into clusters of random sizes NM_j. The family $\{(\overline{R_{j,N}},\overline{T_{j,N}})\}_{j\ge 1}$ of the clustered steps defines a renormalized CTRW, say $\{R_{NM}(t)\}_{t\ge 0}$. In analogy to Eq. (5), the new process reads as

$$R_{NM}(t)=\sum_{j=1}^{\overline{\nu_N}(t)} \overline{R_{j,N}}, \tag{7}$$

where $\overline{\nu_N}(t)=\max\{n:\sum_{j=1}^{n}\overline{T_{j,N}}\le t\}$. As far as M_j is essentially random (i.e., it is not a fixed constant), the renormalized CTRW $\{R_{NM}(t)\}_{t\ge 0}$ is coupled. In such a case, the dependence between the renormalized jumps $\overline{R_{j,N}}$ and waiting times $\overline{T_{j,N}}$ is determined by the distribution of cluster sizes NM_j [13]. For $M_j=1$ we have $R_M(t)=R(t)$, and the proposed procedure coincides with the classical renormalization-group transformation (2) discussed in [34] and valid for the CTRW process (1). However, for random M_j the proposed transformation (6) is essentially different from that followed from Eq. (2).

To illustrate the introduced clustering transformation (6) of the CTRW processes, in Figs. 1–3 we present exemplary trajectories of the obtained CTRW (7) for $N=100$ and $N=1000$ and for different cluster size distributions. The trajectory $R(t)$ of the initial walk (1) is the same for each considered case, and it results from a completely asymmetric Lévy-stable waiting time T_i and from a symmetric Lévy-stable jump R_i. In Fig. 1 the effect of the classical renormalization-group transformation (i.e., $M_j=1$) is shown. The transformation applied to the initial CTRW trajectory in Fig. 2 corre-

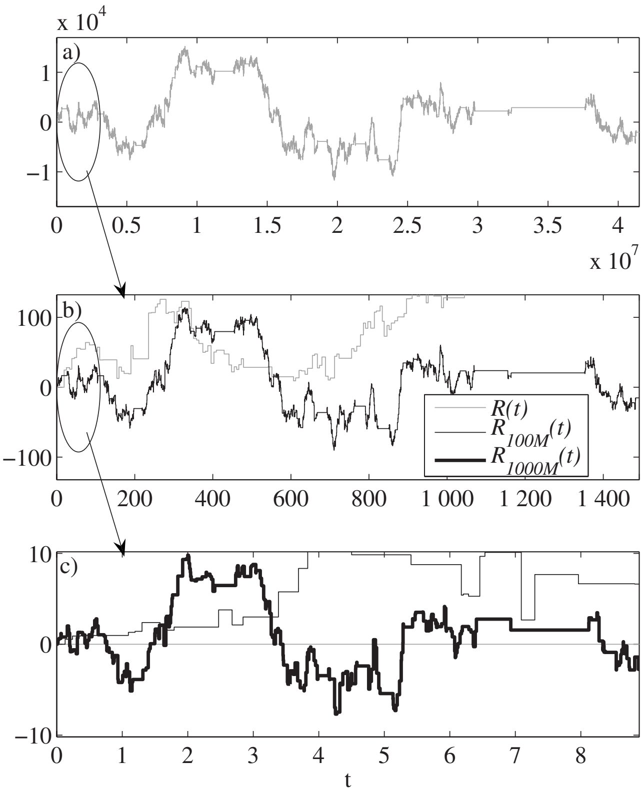

FIG. 2. Exemplary trajectory of the transformed CTRW: The renormalization transformation with finite-mean-value cluster sizes.

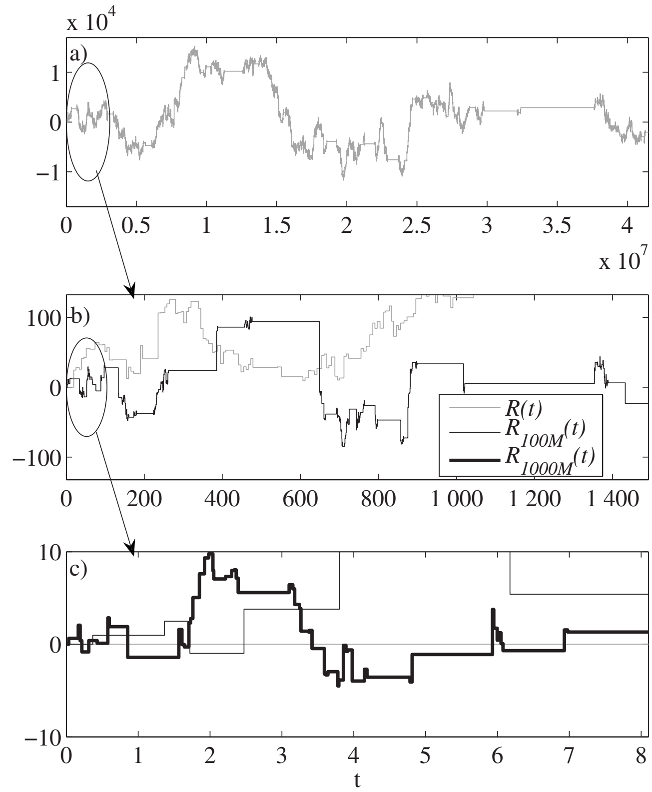

FIG. 3. Exemplary trajectory of the transformed CTRW: The renormalization transformation with heavy-tailed cluster sizes.

sponds to cluster sizes' random components M_j's drawn according to the discretized exponential distribution, having finite mean value. Finally, in Fig. 3 we present the transformed CTRW trajectories resulting from the heavy-tailed discretized Lévy-stable distribution of cluster sizes. As we see, the transformed CTRW trajectory keeps less and less information on the initial-walk trajectory as we pass from the classical renormalization case through random clustering with finite-mean-value to heavy-tailed distribution of cluster sizes. On the other hand, the transformation "smoothes" the trajectory more and more with enlarging the rescaling parameter N.

In analogy to Eq. (3), the renormalized process $\{R_{NM}(t)\}_{t\geq 0}$ can be expressed directly in terms of (R_i,T_i)'s, the spatiotemporal steps of the initial CTRW (1). In this case

$$R_{NM}(t) = \frac{1}{f_R(N)} \sum_{i=1}^{\mu_{NM}(t)} R_i, \tag{8}$$

where the number of summands is equal to

$$\mu_{NM}(t) = \sum_{j=1}^{\nu_M\{\nu[f_T(N)t]/N\}} NM_j .$$

Comparing Eqs. (8), (3), and (1), we see that the clustering transformation essentially changes the number of steps performed until time t and simply rescales the space steps. Indeed, the compound counting process $\{\mu_{NM}(t)\}_{t\geq 0}$ being the number of jumps in Eq. (8) is different from the compound counting process $\{\mu_M(t)\}_{t\geq 0}$ in Eq. (3) and from the renewal counting process $\{\nu(t)\}_{t\geq 0}$ in (1). For example, in the simplest case when $\Pr(M_j=m_0)=1$ for some integer constant m_0, we have $\mu_{NM}(t)=Nm_0\lfloor\frac{\nu(f_T(N)t)}{Nm_0}\rfloor$ instead of $\mu_M(t)=m_0\lfloor\frac{\nu(t)}{m_0}\rfloor$ and of $\nu(t)$, where $\lfloor\cdots\rfloor$ denotes the integer part.

The limiting distribution of the particle position at time t, i.e., the properties of the diffusion front $\widetilde{R}(t)$ can be now evaluated by means of the limit in distribution

$$\widetilde{R}(t) = \lim_{N\to\infty} R_{NM}(t). \tag{9}$$

Its explicit form depends obviously on assumptions set on the distributions of the variables (R_i,T_i) and M_j, see [8,13]. Here we focus our attention on the class of the generalized CTRW's which result from the decoupled initial walks with waiting-time and symmetric jump distributions, both taken from the normal domains of attraction of appropriate Lévy-stable laws (completely asymmetric for the waiting times and symmetric, including Gaussian law as a special case, for the space steps). For this class we study the spatiotemporal clustering transformation (6) for cluster sizes that have either finite-mean-value or heavy-tailed distribution. The resulting diffusion fronts are presented in Table I (for detailed derivations, see the Appendix). Observe that the random clustering of the subsequent steps of the initial decoupled CTRW does not influence at all the limiting law if the spatiotemporal clusters have sizes with a finite mean value. In such a case the distribution of the diffusion front $\widetilde{R}(t)$ is given by a symmetric fractional stable law $\mathcal{F}^{(0)}_{\alpha,\lambda}$ and is exactly the same as

TABLE I. Diffusion front $\tilde{R}(t)$ resulting from different diffusion scenarios. Here $\mathcal{F}^{(0)}_{\alpha,\lambda}$ denotes a symmetric fractional stable random variable, independent of generalized arcsine random variable $\mathcal{B}_\gamma$, and A is a positive constant, see the Appendix for details.

Assumptions	R_i	Symmetric distribution from normal domain of attraction of symmetric stable law $(0<\alpha\leq 2)$	
	T_i	Heavy-tailed distribution $(0<\lambda<1)$	
	M_j	Finite mean value	Heavy-tailed distribution $(0<\gamma<1)$
Results	$\tilde{R}(t)$	Symmetric fractional stable law $(t/A)^{\lambda/\alpha}\mathcal{F}^{(0)}_{\alpha,\lambda}$	"Shrinked" symmetric fractional stable law $(t/A)^{\lambda/\alpha}\mathcal{F}^{(0)}_{\alpha,\lambda}\mathcal{B}^{1/\alpha}_\gamma$

for $M_j=1$, i.e., when formulas (6) refer to the classical renormalization-group approach [34]. As a consequence, asymptotic properties of $R_M(t)$, Eq. (3), are the same as for the initial CTRW $R(t)$, Eq. (1). In contrary, the heavy-tail property of the cluster-size distribution results in appearance of new properties of the diffusion front $\tilde{R}(t)$, related to process $R_M(t)$ only. In this case the limiting distribution is given by a "shrinked" symmetric fractional stable law which is expressed by a mixture of the symmetric fractional stable $\mathcal{F}^{(0)}_{\alpha,\lambda}$ and generalized arcsine $\mathcal{B}_\gamma$ random variables. The generalized arcsine law, because of its support falling in the range (0,1), plays a role of a constricting term.

III. EFFECTIVE RELAXATION RATE: POWER-LAW PROPERTIES

Wide-ranging experimental information has led to the conclusion that the classical phenomenology of relaxation breaks down in complex systems. It has been found that the pure Debye (exponential) response is hardly ever found in nature, and the deviations from it may be relatively large. It appears to be a general rule that the response function $f(t) = -\frac{d\Phi(t)}{dt}$ [i.e., a negative time derivative of the relaxation function $\Phi(t)$] exhibits [20] the following fractional power-law asymptotics:

$$f(t) \propto \begin{cases} (t/\tau_p)^{-n} & \text{for } t \ll \tau_p, \\ (t/\tau_p)^{-m-1} & \text{for } t \gg \tau_p, \end{cases} \tag{10}$$

for some power-law exponents $0<n,m<1$ and the characteristic relaxation time τ_p.

In the CTRW framework, the theoretical attempt to relaxation is based on the idea of an excitation undergoing diffusion in the system under consideration [2,8,14–17]. Consequently, the relaxation function is connected with the temporal decay of a given mode k and defined as the inverse Fourier transform of the diffusion front $\tilde{R}(t)$,

$$\Phi(t) = \langle e^{-ik\tilde{R}(t)}\rangle.$$

On the other hand, following the historically oldest attempt to nonexponential relaxation [20], the relaxation function can be also expressed [12,14,16,17] as a weighted average of the random effective relaxation rate $\tilde{\beta}$,

$$\Phi(t) = \langle e^{-t\tilde{\beta}}\rangle.$$

Taking now into account the equivalence between the above two forms of the relaxation function, we examine the properties of the effective relaxation rate distribution influenced by the clustering procedure (6) in the diffusion scenarios considered in Table I. We study the relaxation behavior depending on the way in which the spatiotemporal steps are grouped into clusters. In what follows, we show two distinct cases resulting from finite-mean-value and heavy-tailed distributions of the cluster sizes NM_j.

If the spatiotemporal clusters have sizes with a finite mean value, then the diffusion front reads as

$$\tilde{R}(t) \overset{d}{=} \left(\frac{t}{A}\right)^{\lambda/\alpha} \mathcal{F}^{(0)}_{\alpha,\lambda}, \tag{11}$$

where the symbol "$\overset{d}{=}$" denotes equal distributions, $\mathcal{F}^{(0)}_{\alpha,\lambda}$ is a symmetric fractional stable random variable, and A is an appropriately chosen positive constant (for details and derivations, see the Appendix). The corresponding relaxation function takes the Mittag-Leffler form

$$\Phi_{\rm ML}(t) = 1 - \Lambda_\lambda\left(\frac{|k|^{\alpha/\lambda}}{A}t\right), \tag{12}$$

where

$$\Lambda_\lambda(x) = \begin{cases} 1 - \sum_{n=0}^{\infty} \frac{(-1)^n x^{\lambda n}}{\Gamma(1+\lambda n)} & \text{for } x>0, \\ 0 & \text{for } x\leq 0; \end{cases}$$

is the distribution function of the Mittag-Leffler law [8,22]. This result yields

$$\tilde{\beta}_{\rm ML} \overset{d}{=} \frac{1}{\tau_p}\frac{\mathcal{S}'_\lambda}{\mathcal{S}_\lambda},$$

where $\mathcal{S}'_\lambda$ and $\mathcal{S}_\lambda$ are independent variables distributed with the same Lévy-stable law, and

$$\tau_p = \frac{A}{|k|^{\alpha/\lambda}}.$$

The density function of the above effective relaxation rate $\tilde{\beta}_{\rm ML}$ equals [12,38]

$$g_{\rm ML}(b) = \begin{cases} \dfrac{\sin(\pi\lambda)(\pi b)^{-1}}{(\tau_p b)^{\lambda} + (\tau_p b)^{-\lambda} + 2\cos(\pi\lambda)}, & b>0, \\ 0, & b\leq 0, \end{cases}$$

and has the following power-law asymptotics:

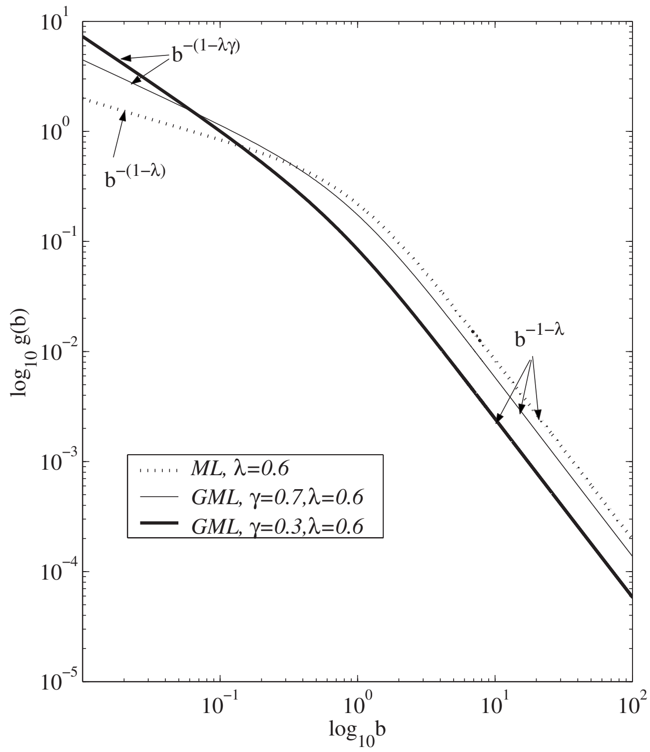

FIG. 4. Exemplary log-log plots of the effective relaxation rate density functions: $g_{\mathrm{GML}}(b)$ (solid lines) and $g_{\mathrm{ML}}(b)$ (dotted line).

$$g_{\mathrm{ML}}(b) \underset{b\to 0+}{\propto} b^{-(1-\lambda)}$$

$$g_{\mathrm{ML}}(b) \underset{b\to\infty}{\propto} b^{-\lambda-1},$$

see Fig. 4. By means of Tauberian theorems [21], the respective response function $f_{\mathrm{ML}}(t)$ exhibits the power-law property (10) with $n=1-\lambda$ and $m=\lambda$, corresponding to the frequency-domain Cole-Cole function [8].

If the spatiotemporal clusters have heavy-tailed sizes, then in the framework of the considered diffusion scenario the diffusion front reads as

$$\tilde{R}(t) \overset{d}{=} \left(\frac{t}{A}\right)^{\lambda/\alpha} \mathcal{F}^{(0)}_{\alpha,\lambda} \mathcal{B}^{1/\alpha}_{\gamma}, \tag{13}$$

where the generalized arcsine random variable $\mathcal{B}_\gamma$ is independent of the symmetric fractional stable random variable $\mathcal{F}^{(0)}_{\alpha,\lambda}$, and A is as in Eq. (11) (for details and derivations, see the Appendix). In this case we obtain a generalization of the Mittag-Leffler relaxation function (12), expressed as

$$\Phi_{\mathrm{GML}}(t) = \int_0^1 \left[1 - \Lambda_\lambda\left(\frac{|k|^{\alpha/\lambda}}{A} t x^{1/\lambda}\right)\right] h_\gamma(x) dx.$$

The corresponding effective relaxation rate is of the form

$$\tilde{\beta}_{\mathrm{GML}} \overset{d}{=} \frac{1}{\tau_p} \frac{\mathcal{S}'_\lambda}{\mathcal{S}_\lambda} \mathcal{B}^{1/\lambda}_\gamma,$$

where $\mathcal{S}'_\lambda$, $\mathcal{S}_\lambda$, and $\mathcal{B}_\gamma$ are independent, and its density is given by [12,38]

$$g_{\mathrm{GML}}(b) = \begin{cases} \dfrac{\sin[\gamma\psi(b)](\pi b)^{-1}}{[(\tau_p b)^{-2\lambda} + 2(\tau_p b)^{-\lambda}\cos(\pi\lambda) + 1]^{\gamma/2}}, & b > 0, \\ 0, & b \le 0, \end{cases}$$

where $\psi(b) = \frac{\pi}{2} - \arctan\left(\frac{(\tau_p b)^\lambda + \cos(\pi\lambda)}{\sin(\pi\lambda)}\right)$. This density has the following power-law asymptotics

$$g_{\mathrm{GML}}(b) \underset{b\to 0+}{\propto} b^{-(1-\lambda\gamma)},$$

$$g_{\mathrm{GML}}(b) \underset{b\to\infty}{\propto} b^{-\lambda-1},$$

see Fig. 4. By Tauberian theorems, the respective response function $f_{\mathrm{GML}}(t)$ exhibits the power-law property (10) with $n=1-\lambda$ and $m=\lambda\gamma$, corresponding to the most general case of the "universal relaxation response" [20].

IV. CONCLUSIONS

The paper introduces a diffusion scenario which leads to the generalized Mittag-Leffler relaxation with the well-known Mittag-Leffler pattern as a special case. The approach is based on the idea of stochastic renormalization-group transformation of a decoupled CTRW. We start with combining the subsequent jumps of a walker into hierarchical clusters renormalized by a suitable rescaling function. The renormalized clustering of jumps is followed by the corresponding transformation of the interjump time intervals. Unlike most renormalization methods, our approach does not use a constant decimation measure. Instead, the size of a "block" in the renormalization transformation is assumed to be a random variable. The stochastic generalization of the classical approach contains the "deterministic" group transformation as a special case when the cluster sizes with probability 1 take a constant value.

The sequence of the renormalized spatiotemporal steps defines a new class of the coupled CTRW's. The dependence between the jumps and waiting times of the renormalized process is introduced by the clustering-jump procedure (6). As a consequence, the asymptotic distribution of the diffusion front depends on the way in which the jumps are grouped into clusters. If the cluster sizes have finite mean value (or with probability 1 take a constant value), then the asymptotic properties of the decoupled walk with power-law jump and waiting-time distributions are not changed by the clustering procedure. In this case the limiting distribution of the diffusion front is related to the Lévy-stable laws only. Such a diffusion scenario does not lead beyond the well-known Mittag-Lefler (or Cole-Cole) relaxation. If, however, the heavy-tailed distribution of the cluster sizes is assumed, the limiting distribution of the diffusion front is related to the mixture of Lévy-stable and generalized arcsine laws. This scenario leads to the generalized Mittag-Leffler relaxation pattern consistent with the more general "universal relaxation response" (usually fitted with the Havriliak-Negami function). It is worth noticing that in both cases the characteristic time constants do not contain information on the clustering-jump procedure, and the power laws do not depend on the properties of the jump distribution.

The further investigations should be directed toward clarification of the relationship between the renormalized CTRWs and the fractional-equation attempts to anomalous diffusion.

APPENDIX: LIMITING DISTRIBUTIONS OF THE PARTICLE POSITION

In this appendix, using limit theorems of probability theory, we study limiting distributions of the particle position resulting from the renormalized CTRW. We derive explicit formulas for diffusion fronts defined by Eq. (9) in two distinct cases depending on the way in which the spatiotemporal steps are grouped in the random clusters.

Let the space steps R_i have a symmetric distribution from the normal domain of attraction of a symmetric Lévy-stable law with the index of stability $0<\alpha\leq 2$ [22,23]. For $\alpha=2$ (Gaussian law) it is equivalent to the existence of finite dispersion of the space steps, i.e.,

$$D^2 R_i = c_R^2 < \infty .$$

For $0<\alpha<2$ the distribution of R_i exhibits the following tail condition:

$$P(|R_i| > x) \underset{x\to\infty}{\sim} (x/c_R)^{-\alpha}$$

for some scale parameter $c_R>0$ [where symbol "$f(x) \underset{x\to\infty}{\sim} g(x)$" reads as $\lim_{x\to\infty}\frac{f(x)}{g(x)}=1$]. For such a jump distribution we have

$$\frac{1}{n^{1/\alpha}}\sum_{i=1}^{n} R_i \overset{d}{\underset{n\to\infty}{\to}} c_R q_\alpha^{1/\alpha}\mathcal{S}_\alpha^{(0)}, \tag{A1}$$

where $\mathcal{S}_\alpha^{(0)}$ is distributed according to the symmetric α-stable law such that

$$\langle e^{ix\mathcal{S}_\alpha^{(0)}}\rangle = e^{-|x|^\alpha},$$

and

$$q_\alpha = \begin{cases} \Gamma(1-\alpha)\cos(\pi\alpha/2) & \text{for } 0<\alpha<1, \\ \pi/2 & \text{for } \alpha=1, \\ \Gamma(2-\alpha)\cos(\pi\alpha/2)/(1-\alpha) & \text{for } 1<\alpha<2, \\ 1/2 & \text{for } \alpha=2. \end{cases}$$

(Here "$\overset{d}{\to}$" reads as "tends in distribution.")

Assume now that the waiting times T_i, independent of the jumps R_i, have a distribution from the normal domain of attraction of the completely asymmetric Lévy-stable law with the index of stability $0<\lambda<1$ [22,23]. It means that the waiting-time distribution satisfies the following tail condition:

$$\Pr(T_i \geq s) \underset{s\to\infty}{\sim} (s/c_T)^{-\lambda}$$

for some scaling constant $c_T>0$. In such a case the rescaled renewal counting process $\nu(s)/s^\lambda$ approaches the *trans*-stable distribution as $s\to\infty$ [12,21],

$$\frac{\nu(s)}{s^\lambda} \overset{d}{\underset{s\to\infty}{\to}} \frac{1}{c_T^\lambda\Gamma(1-\lambda)}\frac{1}{\mathcal{S}_\lambda^\lambda}, \tag{A2}$$

where the random variable $\mathcal{S}_\lambda$ is distributed according to the completely asymmetric Lévy-stable law such that

$$\langle e^{-x\mathcal{S}_\lambda}\rangle = e^{-x^\lambda}.$$

Since for the cluster sizes M_j having a finite mean value $\langle M_j\rangle$ we have [30]

$$\frac{1}{s}\sum_{j=1}^{\nu_M(s)} M_j \overset{w.p.1}{\underset{s\to\infty}{\to}} 1 \tag{A3}$$

(where "$\overset{w.p.1}{\to}$" reads as "tends with probability 1"), then from the independence of sequences (M_j), (R_i), and (T_i), the limits (A1)–(A3) yield [39] the symmetric fractional stable diffusion front (11), where the symmetric fractional stable random variable $\mathcal{F}_{\alpha,\lambda}^{(0)}=\frac{1}{\mathcal{S}_\lambda^{\lambda/\alpha}}\mathcal{S}_\alpha^{(0)}$ and the positive constant reads as

$$A = c_T\left(\frac{\Gamma(1-\lambda)}{q_\alpha c_R^\alpha}\right)^{1/\lambda}. \tag{A4}$$

The fractional stable law is expressed by a mixture of independent stable variables $\mathcal{S}_\alpha^{(0)}$ and $\mathcal{S}_\lambda$ and has been derived by using in Eq. (6) the scaling functions $f_R(N)=N^{(1+\delta)/\alpha}$ and $f_T(N)=N^{(1+\delta)/\lambda}$ for some $\delta>0$. Let us note that the diffusion front (11) is the same as the one derived for the classical decoupled CTRW (1), see [8,26].

If, instead of the finite mean value, the distribution of the cluster sizes has a heavy tail with exponent

$$0<\gamma<1,$$

i.e.,

$$\Pr(M_j \geq m) \underset{m\to\infty}{\sim} (m/c)^{-\gamma} \tag{A5}$$

for some scaling constant $c>0$, then [21]

$$\frac{1}{s}\sum_{j=1}^{\nu_M(s)} M_j \overset{d}{\underset{s\to\infty}{\to}} \mathcal{B}_\gamma, \tag{A6}$$

where $\mathcal{B}_\gamma$ is distributed according to the generalized arcsine distribution given by the density function

$$h_\gamma(x) = \begin{cases} \dfrac{x^{\gamma-1}(1-x)^{-\gamma}}{\Gamma(\gamma)\Gamma(1-\gamma)}, & 0<x<1, \\ 0 & \text{otherwise}, \end{cases}$$

with parameter γ. From Eqs. (A1), (A2), and (A6) we obtain [39] that for $f_R(N)=N^{(1+\delta)/\alpha}$ and $f_T(N)=N^{(1+\delta)/\lambda}$ (for some $\delta>0$) the diffusion front approaches the limiting form (13) given by a mixture of the symmetric fractional stable and generalized arcsine laws, where the generalized arcsine random variable $\mathcal{B}_\gamma$ is independent of the symmetric fractional stable random variable $\mathcal{F}_{\alpha,\lambda}^{(0)}$, and constant A is given by Eq. (A4). Notice that condition (A5) means that cluster-size distribution is taken from the normal domain of attraction of the completely asymmetric Lévy-stable law with the index of stability γ.

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