CONTINUOUS-TIME RANDOM WALK APPROACH TO MODELING OF RELAXATION: THE ROLE OF COMPOUND COUNTING PROCESSES*

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(Received January 4, 2007)

We propose a diffusion, continuous-time random walk (CTRW) scenario which is based on generalization of processes counting the number of jumps performed by a walker. We substitute the renewal counting process, used in the classical CTRW framework, by a compound counting process. The construction of such a compound process involves renormalized clustering of random number of walker's spatio-temporal subsequent steps. The family of the renormalized steps defines a new class of coupled CTRWs. The diffusion front of the studied process exhibits properties which help us to enlarge the class of relaxation models discussed yet in the classical CTRW framework.

PACS numbers: 05.40.Fb, 77.22.Gm

1. Introduction

Investigations of structural and dynamical properties of complex systems, displaying anomalous dynamical behavior [1], play a dominant role in exact and life sciences. For description of relaxation and transport phenomena in such systems as glasses, liquid crystals, polymers, biopolymers,

^{*} Presented at the XX Marian Smoluchowski Symposium on Statistical Physics, Zakopane, Poland, September 22–27, 2007.

proteins or even ecosystems continuous-time random walk (CTRW) processes [2,3] appeared to be one of the useful mathematical tools. By now, following the Montroll–Weiss idea [2], several possible types of the CTRW have been studied and their connection to fractional diffusion or Fokker-Planck equations has been well established [1, 4-7]. However, despite their long history in physics, the CTRWs have not been fully explored yet [8–18]. A possibility of a more detailed analysis of different kinds of the random walks gives the "random-variable" approach to the CTRW [19–21]. Such an approach follows directly from the definition of the CTRW as a cumulative stochastic process [22].

The subject of this paper is to show that the cumulative stochastic processes can be generalized to handle more complicated diffusive scenarios than that leading to the celebrated Mittag-Leffler relaxation [1, 13-19]. In Section 2, we introduce a compound counting process in the random-variable approach to the CTRW. The construction of such a process involves renormalized clustering of random number of walker's spatio-temporal subsequent steps. It defines a new class of coupled CTRWs with diffusion-front properties discussed in Section 3. The diffusion scenario depends on the compound counting process, *i.e.* on the way in which the spatio-temporal steps are grouped into clusters. The results, applied in Section 4 to modeling of relaxation, lead to a generalized Mittag–Leffler relaxation pattern containing the Mittag–Leffler function as a special case.

2. Coupled CTRW given by a compound counting process

The CTRW, *i.e.* the cumulative stochastic process, is characterized by a sequence of independent and identically distributed (i.i.d.) spatio-temporal steps $(R_i, T_i), i \geq 1$. When we assume stochastic independence between the jumps R_i and the waiting times T_i , we obtain a decoupled random walk; otherwise we deal with a coupled CTRW. The total distance reached by a walker at time t is equal to the sum of his jumps performed till time t

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

The number of summands (in general, random) is given by $\nu(t) = \max\{n : t \in \mathbb{N}\}$ $\sum_{i=1}^{n} T_i \leq t$, the renewal counting process determined by the waiting times T_i . In this paper we consider an analog of the process (1)

$$R^M(t) = \sum_{i=1}^{\mu^M(t)} R_i \,,$$

$$R^{M}(t) = \sum_{i=1}^{\mu} R_{i}, \qquad (2)$$

in which the number of summands, instead by the renewal counting process $\nu(t)$, is given by a compound counting process $\{\mu^M(t), t \ge 0\}$ defined as

$$\mu^{M}(t) = \sum_{j=1}^{\nu_{M}(\nu(t))} M_{j}$$
(3)

for $\nu_M(m) = \max\{n : \sum_{j=1}^n M_j \leq m\}$. We assume that $\{M_j\}_{j\geq 1}$ is a sequence of i.i.d. positive integer-valued random variables and that this sequence is independent of the family of the spatio-temporal vectors $\{(R_i, T_i)\}_{i\geq 1}$. The idea of compound counting processes allows us to enlarge the class of the total-distance asymptotics given in the CTRW framework [10]. In particular, it provides a diffusion scenario which leads to the general power-law properties of the empirical "universal relaxation response" [23].

The construction of processes (2) and (3) is strictly connected with assembling the jumps and waiting times into clusters of random sizes M_1 , M_2,\ldots . Namely, if we transform the sequence of spatio-temporal steps $\{(R_i,T_i)\}_{i\geq 1}$ into a new family $\{(\overline{R_j},\overline{T_j})\}_{j\geq 1}$ by means of the following procedure

$$(\overline{R_1}, \overline{T_1}) = \sum_{i=1}^{M_1} (R_i, T_i), \text{ and } (\overline{R_j}, \overline{T_j}) = \sum_{i=M_1 + \dots + M_{j-1} + 1}^{M_1 + \dots + M_j} (R_i, T_i), \quad j \ge 2,$$
(4)

then the process $R^M(t)$, given by (2), appears to be a coupled CTRW defined by $\{(\overline{R_j}, \overline{T_j})\}_{j \ge 1}$:

$$R^{M}(t) = \sum_{j=1}^{\overline{\nu}(t)} \overline{R_{j}}, \qquad (5)$$

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 [10].

Observe that the compound counting process $\{\mu^M(t)\}_{t\geq 0}$ in (2) is different from the renewal counting process $\{\nu(t)\}_{t\geq 0}$ indicating the number of jumps performed till time t by a walker in the process (1). For example, when $\Pr(M_j = m_0) = 1$ for some constant m_0 , we have $\mu^M(t) = m_0 \left\lfloor \frac{\nu(t)}{m_0} \right\rfloor$ instead of $\nu(t)$, where $\lfloor \cdot \rfloor$ denotes the integer part. However, for $m_0 = 1$ we obtain $\mu^M(t) = \nu(t)$ and, as a consequence, $R^M(t) = R(t)$. Therefore, the proposed construction can be considered as a generalization of (1), *i.e.* of the classical CTRW concept.

3. Diffusion front depending on the clustering procedure

In physical applications of the CTRW ideology, the studies concern the properties of a diffusion front, *i.e.* the limiting behavior of the rescaled total distance

$$R_{\tau_0}(t) = \frac{R(t/\tau_0)}{f(\tau_0)},$$
(6)

when dimensionless time-scale coefficient τ_0 tends to 0 and the space-rescaling function $f(\tau_0)$ is chosen appropriately. Notice that $R_{\tau_0}(t)$ is nothing else but the total distance of the CTRW (1) corresponding to the rescaled spatiotemporal steps $(R_i/f(\tau_0), T_i/\tau_0^{-1})$. Considering, for example, the simple Brownian random walk (*i.e.* $T_i = 1$, $\Pr(R_i = 1) = \Pr(R_i = -1) = \frac{1}{2}$) we obtain that the overall distribution of $R_{\tau_0}(t)$ approaches a Gaussian (normal) law with dispersion t if we take $f(\tau_0) = \tau_0^{-1/2}$. This rescaling means that while the time between steps goes to zero, the length of steps also goes to zero as $a = \tau_0^{1/2}$ while the value of the diffusion coefficient $D = \frac{1}{2}a^2/\tau_0 = \frac{1}{2}$ is retained.

In case of the decoupled CTRW (1) with power-law jump or waitingtime distributions different asymptotic behaviors of $R_{\tau_0}(t)$ have been indicated [24], all strictly related to the Lévy-stable laws [25]. The result is obviously connected with the theory of limit theorems for sequences of i.i.d. random variables [22, 25–27]. Unlike most physical applications of the original Montroll–Weiss idea [1–3], the random-variable approach to the CTRW does not use the Fourier–Laplace transform attempt providing explicit formulas for diffusion fronts only under some restrictive assumptions on the spatio-temporal random walk characteristics. Instead, as we shall show below, it allows us not only to handle complicated diffusive situations but also to find the explicit asymptotic behavior for different kinds of the diffusion fronts from a very restricted knowledge on the jump- and waitingtime distributions.

Considering the rescaled spatio-temporal steps $(R_i/f(\tau_0), T_i/\tau_0^{-1})$ we obtain the rescaled total distance of the generalized CTRW (2)

$$R_{\tau_0}^M(t) = \frac{R^M(t/\tau_0)}{f(\tau_0)}$$

analogous to (6). Also in this case, the distribution of the limiting position $\widetilde{R}(t)$ of the walker at time t reached as $\tau_0 \searrow 0$ (*i.e.* $\widetilde{R}(t) \stackrel{d}{=} \lim_{\tau_0 \to 0} R^M_{\tau_0}(t)$, limit in distribution¹) can be evaluated by means of limit theorems of probability theory under very general assumptions set on the distributions of the

¹ The symbol " $\stackrel{d}{=}$ " reads "equal distributions".

variables R_i , T_i and M_i , see [10]. Below, we study in details the classes of the generalized CTRWs corresponding to the heavy-tailed waiting times and positive jumps. We compare the case of identical deterministic jumps with the heavy-tailed jumps which distribution belongs to the normal domain of attraction of some completely asymmetric Lévy-stable law.

Let us assume that the distribution of the waiting times T_i has a heavy tail with the tail exponent $0 < \lambda < 1$, *i.e.*²

$$\Pr(T_i \ge t) \underset{t \to \infty}{\sim} \left(\frac{t}{c_T}\right)^{-\lambda} \tag{7}$$

for some scaling constant $c_T > 0$. Furthermore, let us take into account two different clustering procedures (4): clustering with finite mean value of the spatio-temporal cluster sizes and clustering with heavy-tailed cluster-size distribution with the tail exponent $0 < \gamma < 1$, *i.e.* such that

$$\Pr(M_j \ge m) \underset{m \to \infty}{\sim} \left(\frac{m}{c}\right)^{-\gamma} \tag{8}$$

for some scaling constant c > 0.

In case of identical constant v > 0. $R_i = c_R > 0$ we have $R^M(t) = c_R \mu^M(t)$ so that the total distance is determined directly by the counting process $\mu^M(t)$, defined by (3). If the cluster sizes satisfy (8), then the limiting diffusion front for $f(\tau_0) = \tau_0^{-\lambda}$ approaches the following distribution:

$$\widetilde{R}(t) \stackrel{\mathrm{d}}{=} \left(\frac{t}{A}\right)^{\lambda} \mathcal{T}_{\lambda} \mathcal{B}_{\gamma} \,. \tag{9}$$

The random variable \mathcal{T}_{λ} in (9) has a trans-stable distribution [25], *i.e.* $\mathcal{T}_{\lambda} =$ $1/S_{\lambda}^{\lambda}$, where S_{λ} is distributed according to the completely asymmetric Lévystable law with the index of stability λ . The stable law of S_{λ} is given by the density function $g_{\lambda}(x)$ such that

$$\left\langle e^{-k\mathcal{S}_{\lambda}}\right\rangle = \int_{0}^{\infty} e^{-\mathbf{k}x} g_{\lambda}(x) dx = e^{-k^{\lambda}}.$$

The random variable \mathcal{B}_{γ} in (9) is independent of \mathcal{T}_{λ} and distributed according to the generalized arcsine distribution given by the density function

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

² The symbol " $f(x) \underset{x \to \infty}{\sim} g(x)$ " reads $\lim_{x \to \infty} f(x)/g(x) = 1$.

Constant A equals

$$A = c_T (c_R^{-1} \Gamma(1-\lambda))^{1/\lambda} \,. \tag{10}$$

To derive Eq. (9) we used the following limit theorems: if the waiting-time distribution has the heavy-tail property (7), then [9, 10, 22]

$$\frac{\nu(t/\tau_0)}{(t/\tau_0)^{\lambda}} \xrightarrow[\tau_0 \to 0]{d} \frac{1}{c_T^{\lambda} \Gamma(1-\lambda)} \frac{1}{\mathcal{S}_{\lambda}^{\lambda}}, \qquad (11)$$

and if the cluster sizes satisfy (8), then [22]

$$\frac{1}{s} \sum_{j=1}^{\nu_M(s)} M_j \xrightarrow[s \to \infty]{d} \mathcal{B}_{\gamma}.$$
(12)

From assumed independence of sequences (M_j) and (T_i) , by means of lemma on the limit of a composite random function [28] one obtains (9), see [10] for details.

The limiting law of $\widetilde{R}(t)$ in (9) is a mixture of the trans-stable and the generalized arcsine distributions, and hence its density function can be given by the following integral representation:

$$f(x) = \begin{cases} \frac{A^{1-\lambda}(t/A)^{1-\lambda}}{\Gamma(\gamma)\Gamma(1-\gamma)} x^{\gamma-1} \int_{0}^{(t/A)x^{-1/\lambda}} u^{\lambda\gamma}((t/A)^{\lambda} - xu^{\lambda})^{-\gamma}g_{\lambda}(u)du & \text{for } x > 0, \\ 0 & \text{otherwise.} \end{cases}$$

Since \mathcal{B}_{γ} takes values from the range (0, 1), the limiting law in (9) can be interpreted as a "shrinked" trans-stable distribution.

If instead of (8) we take into account the cluster sizes M_j having finite mean value $\langle M_j \rangle$, then³ [29]

$$\frac{1}{s} \sum_{j=1}^{\nu_M(s)} M_j \xrightarrow[s \to \infty]{\text{w.p.1}} 1$$
(13)

and as a consequence, formula (9) simplifies to the trans-stable law

$$\widetilde{R}(t) \stackrel{\mathrm{d}}{=} \left(\frac{t}{A}\right)^{\lambda} \mathcal{T}_{\lambda} \,, \tag{14}$$

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³ The symbol $\xrightarrow{\text{w.p.1}}$ reads "tends with probability 1".

and the limiting total-distance distribution in (14) is simply given by the density function of the trans-stable law [24, 25]

$$f(x) = \frac{1}{\lambda} \left(\frac{t}{A}\right) x^{-1/\lambda - 1} g_{\lambda} \left(\left(\frac{t}{A}\right) x^{-1/\lambda} \right) \,.$$

Let us now consider positive space steps R_i with a distribution that satisfies the heavy-tail condition with the tail exponent $0 < \alpha < 1$

$$P(R_i > x) \underset{x \to \infty}{\sim} \left(\frac{x}{c_R}\right)^{-\alpha} \tag{15}$$

for some scaling constant $c_R > 0$. The above condition means that the distribution of R_i belongs to the normal domain of attraction of the completely asymmetric Lévy-stable distribution with the index of stability α [25, 27]. Namely, if we represent the stable law by a random variable S_{α} , then (15) leads to

$$\frac{1}{n^{1/\alpha}} \sum_{i=1}^{n} R_i \xrightarrow[n \to \infty]{d} c_R \left(\Gamma(1-\alpha) \right)^{1/\alpha} \mathcal{S}_{\alpha} \,. \tag{16}$$

Assume moreover that R_i and T_i are independent, and take into account cluster sizes M_j with property (8). Then, using again the lemma on the limit of a composite random function [28] and limit theorems (11), (12) and (16), one obtains (for details, see [10])

$$\widetilde{R}(t) \stackrel{\mathrm{d}}{=} \left(\frac{t}{A_1}\right)^{\lambda/\alpha} \mathcal{F}_{\alpha,\lambda} \left(\mathcal{B}_{\gamma}\right)^{1/\alpha}$$
(17)

with the constant

$$A_1 = \frac{c_T}{c_R^{\alpha/\lambda}} \left(\frac{\Gamma(1-\lambda)}{\Gamma(1-\alpha)}\right)^{1/\lambda} .$$
(18)

To derive (17) the following scaling function $f(\tau_0) = \tau_0^{-\lambda/\alpha}$ has to be assumed. Random variables \mathcal{B}_{γ} and $\mathcal{F}_{\alpha,\lambda}$ are independent, and $\mathcal{F}_{\alpha,\lambda}$ has a fractional stable distribution [30–32], *i.e.* $\mathcal{F}_{\alpha,\lambda} = \mathcal{S}_{\alpha}/\mathcal{S}_{\lambda}^{\lambda/\alpha}$, where \mathcal{S}_{α} is independent of \mathcal{S}_{λ} . The limiting total-distance distribution in (17) is therefore a mixture of the fractional stable and the generalized arcsine distributions, or in other words, "shrinked" fractional stable law. It has the following integral representation of the density function:

$$f(x) = \frac{\alpha}{\Gamma(\gamma)\Gamma(1-\gamma)} \int_{0}^{\infty} dz \int_{0}^{(t/A_1)z^{-\alpha/\lambda}} u^{\lambda\gamma} ((t/A_1)^{\lambda} - z^{\alpha}u^{\lambda})^{-\gamma} z^{\alpha\gamma-2} g_{\lambda}(u) g_{\alpha}(x/z) du \,,$$

where $g_{\alpha}(x)$ denotes the density function of S_{α} . If the cluster sizes M_j have a finite mean value, then (13) holds instead of (12) and for the same scaling function $f(\tau_0) = \tau_0^{-\lambda/\alpha}$ we get the fractional stable limiting law

$$\widetilde{R}(t) \stackrel{\mathrm{d}}{=} \left(\frac{t}{A_1}\right)^{\lambda/\alpha} \mathcal{F}_{\alpha,\lambda}$$
 (19)

with the density function

$$f(x) = \left(\frac{t}{A_1}\right)^{-\lambda/\alpha} \int_0^\infty g_\alpha \left(\left(\frac{t}{A_1}\right)^{-\lambda/\alpha} x u^{\lambda/\alpha}\right) u^{\lambda/\alpha} g_\lambda(u) \, du \, .$$

As we see, when in procedure (4) we take into account the spatiotemporal clusters that have sizes with finite mean value, then the limiting distribution of $\tilde{R}(t)$ is exactly the same as for $M_j = 1$, *i.e.* as if no agglutination of jumps takes place. In such a case the clustering transformation does not at all influence the limiting law, providing hence the same results as the classical CTRW (1). In contrary, the heavy-tail property (8) of the clustersize distribution results in appearance of the generalized arcsine term \mathcal{B}_{γ} , constricting the trans-stable and the fractional stable law in (9) and (17), respectively.

In case of identical deterministic jumps the resulting diffusion fronts, Eqs. (9) and (14), reflect in fact the limiting behavior of the rescaled compound counting process $\mu^{M}(t)$, Eq. (3). If instead, the heavy-tail distributed jumps are considered, property (15) of the jump distribution yields the Lévystable term S_{α} of the fractional stable distribution in (17) and (19). It also influences the scaling constant A_{1} , Eq. (18).

4. General Mittag–Leffler relaxation pattern

Theoretical modeling of relaxation in the CTRW framework is based on the idea of an excitation undergoing diffusion in the system under consideration [1,11,13,14,19]. Consequently, the relaxation function is connected with the temporal decay of a given macroscopic mode k and, in case of positive space steps, defined as the Laplace transform of the diffusion front $\tilde{R}(t)$

$$\Phi(t) = \langle e^{-kR(t)} \rangle \,. \tag{20}$$

Below, we study the properties of the above function corresponding to the diffusion fronts (9), (14), (17), (19), obtained in Section 3. Examining influence of the spatio-temporal clustering procedure (4) on the relaxation

process, we observe distinct relaxation behavior for finite mean value and for heavy-tailed cluster sizes M_i .

The diffusion scheme, based on clustering procedure (4) with finite mean value cluster sizes, has led to the diffusion fronts (14) and (19). In both cases, independently of the distribution of jumps, the time behavior of the relaxation function (20) follows the same Mittag–Leffler pattern. Indeed, for $\tilde{R}(t)$ given by (14), referred to identical space steps and hence reflecting in fact asymptotics of the compound counting process $\mu^{M}(t)$, we have

$$\Phi_{\rm ML}(t) = \left\langle e^{-k\widetilde{R}(t)} \right\rangle = 1 - \Lambda_{\lambda} \left(\left(\frac{t}{A} \right) k^{1/\lambda} \right) \,, \tag{21}$$

where

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

is the Mittag–Leffler distribution function [19], and the constant A is given by (10). Relaxation function $\Phi_{ML}(t)$ exhibits the short- and long-time powerlaw properties

$$\Phi_{\rm ML}(t) \propto \begin{cases} (t/\tau_p)^{\lambda} & \text{for } t \ll \tau_p ,\\ (t/\tau_p)^{-\lambda} & \text{for } t \gg \tau_p , \end{cases}$$
(22)

for the characteristic relaxation time

$$\tau_p = \frac{A}{k^{1/\lambda}}.$$

We obtain almost the same formula for the relaxation function in case of the diffusion front (19) connected with positive heavy-tailed space steps. We need only to substitute the time constant A in (21) by A_1 and τ_p in (22) by

$$\tau_{p,1} = \frac{A_1}{k^{\alpha/\lambda}} \,.$$

When the heavy-tailed clustering has been applied in (4), the diffusion fronts of the generalized CTRW have been shown to take forms (9) and (17). In the case of (9), obtained for deterministic space steps, the relaxation function reads

$$\Phi_{\rm GML}(t) = \left\langle e^{-k\widetilde{R}(t)} \right\rangle = \int_{0}^{1} \left(1 - \Lambda_{\lambda} \left(\left(\frac{t}{A} \right) (kx)^{1/\lambda} \right) \right) h_{\gamma}(x) dx \,. \tag{23}$$

The above, generalized Mittag–Leffler relaxation function exhibits the shortand long-time power-law properties

$$\Phi_{\rm GML}(t) \propto \begin{cases} (t/\tau_p)^{\lambda} & \text{for } t \ll \tau_p ,\\ (t/\tau_p)^{-\lambda\gamma} & \text{for } t \gg \tau_p . \end{cases}$$
(24)

more suitable for description of the empirical relaxation evidence [23] than those of the Mittag-Leffler function (22). Similarly, the diffusion front (17), referred to positive heavy-tailed space steps, yields the relaxation function (23) with A replaced by A_1 , satisfying power laws (24) with $\tau_{p,1}$ instead of τ_p .

Notice that the Mittag-Leffler and the generalized Mittag-Leffler relaxation patterns derived for considered different diffusion scenarios, see Table I, are essentially determined by asymptotics of the compound counting process $\mu^{M}(t)$. The properties of the jump distribution influence only the value of the characteristic material constant (relaxation time).

TABLE I

	Identical constant jumps	Heavy-tailed positive jumps	Relaxation function
Clusters of finite mean value	Trans-stable diffusion front	Fractional stable diffusion front	Mittag–Leffler
Heavy-tailed clusters	Shrinked trans-stable diffusion front	Shrinked fractional stable diffusion front	Generalized Mittag–Leffler

Different diffusion scenarios and the resulting relaxation patterns.

5. Concluding remarks

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 a generalization of the classical CTRW concept.

We start with substituting the renewal counting process, simply indicating the number of steps performed by a walker, by a compound counting process. Construction of such a process involves renormalized clustering of random number of walker's spatio-temporal subsequent steps. The clustering procedure, applied to a decoupled CTRW, provides a new class of the coupled CTRW's in which the dependence between the jumps and waiting times is determined by the cluster-size distribution. As a consequence, the asymptotic distribution of the diffusion front depends on the way in which the spatio-temporal steps 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 initial 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-Leffler 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 consistent with the general empirical relaxation law. In both cases the characteristic time constants do not contain information on the clustering procedure, and the asymptotic power laws do not depend on the properties of the jump distribution. Indeed, despite assumed randomness in the space jumps, the resulting relaxation patterns appear to be the same as in case of deterministic identical jumps. The specific forms of the relaxation function are hence determined mainly by the asymptotics of the compound counting process $\mu^{M}(t)$. The jump distribution influences only the characteristic scale constants.

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