PHYSICAL REVIEW E **78**, 060102(R) (2008)

Continuous-time random walks with internal dynamics and subdiffusive reaction-diffusion equations

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(Received 26 June 2008; published 5 December 2008)

We formulate the generalized master equation for a class of continuous-time random walks in the presence of a prescribed deterministic evolution between successive transitions. This formulation is exemplified by means of an advection-diffusion and a jump-diffusion scheme. Based on this master equation, we also derive reaction-diffusion equations for subdiffusive chemical species, using a mean-field approximation.

DOI: 10.1103/PhysRevE.78.060102 PACS number(s): 05.40.Fb, 05.10.Gg, 52.65.Ff

While transport in equilibrium systems occurs due to Brownian motion of the microscopic particles following Fick's law, systems far from equilibrium often show anomalous—i.e., non-Fickian and non-Gaussian—diffusion [1–3]. Here, the mean–square displacement of a particle is given by a power law $\langle x^2(t)\rangle \propto t^\alpha$ with $\alpha \neq 1$. As a model of such anomalous behavior, the continuous-time random walk (CTRW) introduced by Montroll and Weiss in 1965 [4] and extended by Scher and Montroll to explain anomalous diffusion [5] has attracted much attention during the last few decades. The Scher-Montroll CTRW process corresponds to the situation in which a particle (walker) is trapped on a site for a time τ distributed with a power-law probability density $W(\tau)$ and then makes a jump to another site. In between the jumps it neither moves nor changes its identity—i.e., does not exhibit any dynamics of its own. The probability distribution of the walker's positions is determined by a generalized master equation [6], which, under certain assumptions, takes the form of a diffusion equation with a time memory [1]. Even this simple model exhibits extremely interesting properties connected with its intrinsic nonstationarity [7] and is still under extensive investigation both theoretically and experimentally.

In many situations, however, the system undergoes additional, internal dynamics also during waiting periods, due, e.g., to a deterministic drift generated by an external force or due to chemical reactions among the diffusing particles. The purpose of the present article is to perform a treatment of such processes based on the derivation of a generalized master equation. We shall argue that, generally, these types of stochastic processes cannot be captured by a subordination procedure along the lines of Fogedby [8]. Our approach will allow us to formulate subdiffusive reaction-diffusion equations, which are of utmost importance for pattern formation in biological systems, due to the ubiquity of subdiffusive transport in biological systems (see, e.g., Ref. [9] and references therein).

Our treatment extends earlier work of Shlesinger *et al.* [10] on Lévy walks, which have been developed for the description of particle transport in chaotic and turbulent flows, the work of Metzler *et al.* [11] on passively transported particles in flows undergoing jumps with respect to the moving fluid, and the work of Eliazar and Klafter [12] on the behavior of overdamped particles exposed to random impacts, a special case of a shot noise situation.

Generalized master (or Fokker-Planck) equations can be obtained in a straightforward manner by the method of subordination proposed by Fogedby [8]. Here, one considers a Markov process $x(s)$ for a variable x depending on an internal (operational) time variable s. The corresponding probability distribution $f(x,s)$ obeys the Fokker-Planck equation

$$\frac{\partial}{\partial s} f_0(x,s) = L_{\mathrm{FP}} f_0(x,s), \tag{1}$$

where the Fokker-Planck operator L_{FP} is defined, e.g., in Ref. [13]. A related stochastic process $y(t)=x(s(t))$ is created by a time transformation $s(t)$, which is assumed to be an independent random process with non-negative increments. It can be shown that the probability distribution of the process $y(t)$, $f(y,t)$, obeys a generalized Fokker-Planck equation

$$\frac{\partial}{\partial t} f(y,t) = \int_0^t dt' Q(t-t') L_{\mathrm{FP}} f(y,t'), \tag{2}$$

whose solution reads $f(y,t)=\int_0^t p(s,t) f_0(y,s) ds$ [8]. In particular, it was shown that the quantity $p(s,t)$ is the probability distribution of the process $s(t)$, which is related to the kernel $Q(t)$ by the relation

$$\frac{\partial}{\partial t} p(s,t) = -\int_0^t dt' Q(t-t') \frac{\partial}{\partial s} p(s,t'). \tag{3}$$

The choice $Q(t-t') \propto (t-t')^{\delta-1}$ leads to fractional equations [1]. Independently of the work by Fogedby, one of us (I.S.) arrived at a similar conclusion for a much broader class of stochastic processes and clarified the meaning of the kernel $Q(t-t')$ [14].

Due to various applications briefly mentioned above, the case of anomalous diffusion in the presence of an additional deterministic process is of significant interest. However, Fogedby's approach can only be extended to this situation in a straightforward manner if the deterministic dynamics depends on the *internal* time s, not on the *physical* time t. In the latter case one expects a quite different behavior, and the derivation of a respective generalized Fokker-Planck equation—which is the main goal of the present article—

turns out to be somewhat involved. Before we address this issue in a general way, it is useful to first look at two concrete examples.

In the first one, we consider a particle with coordinate q which is advected with a constant velocity v and subjected to transitions occurring at randomly distributed time intervals. The resulting generalized Fokker-Planck equation reads

$$\left[\frac{\partial}{\partial t}+v\frac{\partial}{\partial q}\right]f(q,t)=\int dt' Q(t-t')\frac{\partial^2}{\partial q^2}f(q-v(t-t'),t'). \tag{4}$$

In this context, we would like to point out the occurrence of a retardation effect in the diffusion term (cf. Refs. [15,16]) which is due to the advective force acting in *real* time on the anomalously diffusing particle. The above equation can be solved by the ansatz $f(q,t)=F(q-vt,t)$. The probability distribution is then governed by

$$\frac{\partial}{\partial t}F(\xi,t)=\int dt' Q(t-t')\frac{\partial^2}{\partial \xi^2}F(\xi,t'), \tag{5}$$

with $\xi=q-vt$. Thus, in the present case, a transition to a comoving reference frame leads to a conventional CTRW for the variable ξ, in contrast to the behavior for a subordinated process described by Eq. (2). Similar arguments have been given in Ref. [11]. However, the generalized Fokker-Planck equation proposed therein is only valid to order v^2.

Let us now proceed to our second example. Here, we consider the motion of an overdamped particle ($\dot{q}=-\gamma q$), subjected to random transitions with a suitably defined waiting time distribution $W(t)$. The evolution equation for the probability distribution $f(q,t)$ describing such a process takes the form

$$\frac{\partial}{\partial t}f(q,t)=\gamma\frac{\partial}{\partial q}qf(q,t)+\int_0^t dt' Q(t-t')\frac{\partial^2}{\partial q^2}\frac{1}{e^{-\gamma(t-t')}}f\left(\frac{q}{e^{-\gamma(t-t')}},t'\right). \tag{6}$$

Using this equation, we can derive an expression for the moments of this process to arbitrary order, in the limit of long times. Considering symmetric initial conditions one obtains for the moments of even order the rather strange behavior $\langle q^{2k}(t)\rangle\approx Q(t)$. This behavior differs from the one obtained for the subordinated case, in which the moments tend to constants.

Having considered two specific examples, we now describe the derivation of a generalized master equation for the class of processes under consideration. We assume that the variable q undergoes a purely deterministic (or a Markovian random) process which is characterized by the transition probability (or propagator) $p(q,q';t-t')$. For the two examples considered above, we have, respectively, $p(q,q',t-t')=\delta(q-q'-v(t-t'))$ and $p(q,q',t-t')=\delta(q-q'e^{-\gamma(t-t')})$. However, the considered class also includes diffusion processes described by a Fokker-Planck operator. We further assume that the variable q undergoes sudden transitions from state q to state q' after time intervals τ which are characterized by a waiting-time distribution $W(\tau)$. Let us denote the transition probability from state q' to state q by $F(q,q')$. Then the probability that such a jump between q and q' occurs in the time interval τ is just $F(q,q')W(\tau)$. Factorization of this quantity demonstrates that waiting times and transitions are statistically independent, which is characteristic for decoupled CTRWs.

As usual in the theory of CTRWs, the next step is to introduce the probability density for having arrived at times t' at an infinitesimal interval close to the position q' shortly after a jump. The respective quantity shall be denoted by $\eta(q',t')$. The probability density of arriving at q at time t after another jump is then given by

$$\eta(q,t)=\int_0^t dt'\int dq''\int dq' F(q,q'')W(t-t')\times p(q'',q',t-t')\eta(q',t')+\delta(t)f(q,0). \tag{7}$$

Here, it has been taken into account that the system evolves after the first jump according to the process described by the propagator $p(q'',q',t-t')$, performing another jump from q'' to q at time t. The probability density $f(q,t)$ related to the probability for finding a particle in the interval dq close to q is then given by

$$f(q,t)=\int_0^t dt'\int dq' p(q,q',t-t')w(t-t')\eta(q',t'). \tag{8}$$

After having arrived after the jump at time t' in the interval close to q', the position of the particle changes according to the propagator p from q' to q. The quantity $w(t-t')$ is the probability that no jump occurs in the time interval $t-t'$. It is related to the quantity $W(t-t')$ according to

$$w(t-t')=1-\int_0^{t-t'} d\tau W(\tau). \tag{9}$$

The operator representation $p(q,q',t-t')=\{e^{L(t-t')}\}_{q,q'}$ of the propagator $p(q,q',t-t')$ allows for the determination of the Laplace-transformed equations

$$f(q,\lambda)=w(\lambda-L)\eta(q,\lambda) \tag{10}$$

and

$$\eta(q,\lambda)=f(q,0)+\int dq' F(q,q')W(\lambda-L)\eta(q',\lambda). \tag{11}$$

Here, the Laplace transforms of $f(q,t)$, $w(t)$, and $W(t)$ are denoted, respectively, by $f(q,\lambda)$, $w(\lambda)$, and $W(\lambda)$. Combining both relationships, we arrive at the generalized master equation for the process under consideration:

$$\left[\frac{\partial}{\partial t}-L\right]f(q,t)=\int_0^t dt' Q(t-t')\times\int dq'[F(q,q')-\delta(q-q')]e^{L(t-t')}f(q',t'). \tag{12}$$

Here, we have introduced the kernel $Q(t-t')$, which is defined by its Laplace transform [6]

$$Q(\lambda) = \frac{\lambda W(\lambda)}{1 - W(\lambda)}. \tag{13}$$

For example, an exponential waiting-time distribution $W(t) = \Gamma e^{-\Gamma t}$ leads to the kernel $Q(t) = \delta(t)$. The representation of $Q(t)$ in terms of the waiting time is necessary in order to obtain generalized master equations which define a nonnegative probability density $f(q,t)$ for all times t [14].

At this point, it is useful to recall that, allowing only for nearby jumps, the transition probability $F(q,q')$ in the master equation yields a generalized Fokker-Planck equation [17]. In our case, it is of the form

$$\left[\frac{\partial}{\partial t} - L\right] f(q,t) = \int_0^t dt' L_{\mathrm{FP}} Q(t-t') e^{L(t-t')} f(q,t'). \tag{14}$$

Here, the two operators L and L_{FP} arise. The Fokker-Planck operator L_{FP} is connected with the transition probability $F(q,q')$ of the time-random sudden jumps [17], whereas the operator L is connected with the propagator $p(q,q',t,t')$ and describes the continuously evolving process. This is the desired Fokker-Planck-type equation for the class of stochastic processes under consideration—a key result of the present paper—representing a nontrivial generalization of Eq. (2) with the operator L, describing the time evolution of the system between successive jumps, entering on both sides of the equation.

At first glance, Eq. (14) appears to be rather complicated, maybe not analytically tractable. However, as the above examples show, analytical solutions are indeed possible. In this context, we would like to briefly mention the following strategy to solve this generalized Fokker-Planck equation. It turns out to be convenient to switch to a kind of interaction picture by employing the ansatz

$$f(q,t) = e^{Lt} g(q,t). \tag{15}$$

Then, the resulting problem to be solved is

$$\frac{\partial}{\partial t} g(q,t) = e^{-Lt} L_{\mathrm{FP}} e^{Lt} \int_0^t dt' Q(t-t') g(q,t'). \tag{16}$$

Provided the two Fokker-Planck operators L and L_{FP} commute, we (only) have to solve the simpler problem

$$\frac{\partial}{\partial t} g(q,t) = L_{\mathrm{FP}} \int_0^t dt' Q(t-t') g(q,t'). \tag{17}$$

Comparing this result with the Fokker-Planck equation (2), it is evident that $g(q,t)$ describes a subordinated process.

The first of the above-mentioned examples falls into this category. As a further example, we would like to consider the two operators $L = Q_1 \frac{\partial^2}{\partial q^2}$ and $L_{\mathrm{FP}} = Q_0 \frac{\partial^2}{\partial q^2}$ describing pure diffusion interrupted by randomly distributed sudden transitions—i.e., a jump-diffusion process. The corresponding probability distribution takes the form

$$f(q,t) = \int_0^\infty ds\, p(s,t) \exp\left[-\frac{q^2}{2(Q_0 s + Q_1 t)}\right]. \tag{18}$$

Calculating the even moments we find that their long-term behavior is dominated by the scaling behavior $\langle q(t)^{2k} \rangle \propto t^k$. The additional jumps result in subdominated scalings.

The master equation just derived can also be used to obtain reaction-subdiffusion equations, which happens to be a particularly interesting application. The subdiffusive transport is modeled by the introduction of random waiting times between the jumps of particles. The reaction among the chemical species evolves, however, continuously in time, changing the concentrations also between the jumps. Our derivation starts from extending the procedure leading to Eq. (12) to multiple dimensions, leading to a master equation for a multidimensional state vector, and the reaction-subdiffusion equations are then derived along the lines of the derivation of the reaction-diffusion equations from the usual Fokker-Planck equations.

We partition real space into small compartments labeled by the index i and consider N different chemical species (labeled by $\alpha = 1, \ldots, N$), which locally react according to the deterministic reaction kinetics

$$\dot{c}_{\alpha,i} = R_\alpha(c_{\alpha,i}). \tag{19}$$

Further, we allow for diffusive transitions between neighboring cells. For the sake of simplicity, we assume that the transitions of all particles occur simultaneously ("global update") at random times, characterized by the waiting-time distribution $W(t)$. The situations with independent particle jumps ("local update") are also treatable, but lead to considerably more complicated calculations. Lumping all concentrations $c_{\alpha,i}$ into the state vector $\mathbf{c}$, we can formulate the generalized master equation

$$\begin{aligned}\frac{\partial}{\partial t} f(\mathbf{c},t) = &- \frac{\partial}{\partial \mathbf{c}} \mathbf{R}(\mathbf{c}) f(\mathbf{c},t) + \int_0^t dt' Q(t-t') \\ &\times \int d\mathbf{c}' \int d\mathbf{c}'' [F(\mathbf{c},\mathbf{c}') - \delta(\mathbf{c}-\mathbf{c}')] \\ &\times \delta(\mathbf{c}' - \mathbf{G}(\mathbf{c}'', t-t')) f(\mathbf{c}'',t').\end{aligned} \tag{20}$$

Here the function $\mathbf{G}(\mathbf{c}', t-t')$ is the solution of the kinetic equation (19) with the initial condition $\mathbf{G}(\mathbf{c}',0) = \mathbf{c}'$.

The desired subdiffusive reaction-diffusion equation is an evolution equation for the mean value of the quantity $\mathbf{c}$, $\mathbf{C} = \int d\mathbf{c}\, \mathbf{c} f(\mathbf{c},t)$. It reads

$$\begin{aligned}\frac{\partial}{\partial t} \mathbf{C} = &\int \mathbf{R}(\mathbf{c}) f(\mathbf{c},t) + \int_0^t dt' Q(t-t') \\ &\times \left(\int d\mathbf{c}\, d\mathbf{c}'\, \mathbf{c} F(\mathbf{c}, \mathbf{G}(\mathbf{c}', t-t')) f(\mathbf{c}',t') \right. \\ &\left. - \int d\mathbf{c}'\, \mathbf{G}(\mathbf{c}', t-t') f(\mathbf{c}',t') \right).\end{aligned} \tag{21}$$

The mean-field approximation $\int d\mathbf{c}\, H(\mathbf{c}) f(\mathbf{c},t) \approx H(\mathbf{C})$ leads then to a closed equation for the mean concentrations. Al-

lowing only for nearest-neighbor transitions, we obtain the reaction-subdiffusion equation

$$\frac{\partial}{\partial t}\mathbf{C}(\mathbf{x},t) = \mathbf{R}(\mathbf{C}(\mathbf{x},t)) + D\Delta_x \int_0^t dt' Q(t-t')\mathbf{G}(\mathbf{C}(\mathbf{x},t'),t-t'). \tag{22}$$

There have been several attempts to establish reaction-diffusion equations for subdiffusive chemical species (see [18] and references therein). *Ad hoc* formulations can be obtained from Eq. (22) by replacing $G(C(\mathbf{x},t'),t-t')$ with $C(\mathbf{x},t')$. However, such equations show serious inconsistencies; in some cases, even mass conservation is violated [18]. The correct way to proceed was discussed in [18] for a special example of a linear bimolecular reaction scheme $A \rightleftharpoons B$. This case is contained in our general subdiffusive reaction-diffusion equation as follows. If we take the reaction rates $\mathbf{R}=M\mathbf{C}$ to be linear in $\mathbf{C}$, we obtain $\mathbf{G}(\mathbf{C}(\mathbf{x},t'),t-t') = e^{M(t-t')}\mathbf{C}(\mathbf{x},t')$. For a two-component vector $\mathbf{C}(\mathbf{x},t)$ we end up directly with the system considered in [18]. Since the particles in this scheme do not interact, the global update assumption leads to the same results as the independent-particle motion considered in Ref. [18]. An important application is the treatment of radiaoctive decay in flows through porous media.

Finally, as a nonlinear example, we consider the subdiffusive Fisher-Kolmogorov-Petrovskii-Piskunov (FKPP) equation, which has been proposed as a model equation for the propagation of favorable genes in a population [19]. The diffusive version of the FKPP equation is one of the basic equations for the investigation of reaction fronts in nonlinear reaction kinetics. Our subdiffusive version for the $A+A \rightleftharpoons A$ reaction reads

$$\frac{\partial}{\partial t}C(x,t) = C(x,t)[1-C(x,t)] + \frac{\partial^2}{\partial x^2}\int_0^t dt' Q(t-t') \times \frac{C(x,t')}{[1-e^{-(t-t')}]C(x,t') + e^{-(t-t')}}. \tag{23}$$

Note the emergence of a nonlinear diffusion term in addition to a temporal memory. We stress that the equation for a reversible reaction under global update differs from the one obtained in Ref. [20] for the locally updated irreversible $A+B \rightarrow 2B$ reaction scheme.

We have considered stochastic processes which are partly generated by Markovian processes and which, *additionally*, are subjected to the impact of fluctuations randomly occurring in time. These impacts are treated in the framework of continuous-time random walk processes by a waiting-time distribution. We have derived the generalized master equation for this class of processes and have been able to formulate subdiffusive reaction-diffusion systems also for nonlinear reaction kinetics, which are of relevance for pattern formation in biological systems.

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