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Stochastic pathway to anomalous diffusion

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We present an appraisal of differential-equation models for anomalous diffusion, in which the time evolution of the mean-square displacement is $\langle r^2(t)\rangle \sim t^\gamma$ with $\gamma \neq 1$. By comparison, continuous-time random walks lead via generalized master equations to an integro-differential picture. Using Lévy walks and a kernel which couples time and space, we obtain a generalized picture for anomalous transport, which provides a unified framework both for dispersive ($\gamma < 1$) and for enhanced diffusion ($\gamma > 1$).

I. INTRODUCTION

Large classes of dynamical processes in disordered media display diffusionlike behavior. Such processes differ, however, from the simple Brownian motion, a fact manifested through the dependence of the mean-squared displacement $\langle r^2(t)\rangle$ on time. Whereas for simple diffusion $\langle r^2(t)\rangle \sim t$, anomalous diffusion is characterized by

$$\langle r^2(t)\rangle \sim t^\gamma \tag{1}$$

with $\gamma \neq 1$. Examples for Eq. (1) are to be found in chaotic dynamics, which generally leads to enhanced diffusion (e.g., for turbulent motion $\gamma \simeq 3$) on the one hand,[1–9] but also in systems with geometric constraints (doped crystals, glasses, fractals), for which the diffusion is dispersive, i.e., $\gamma < 1$, on the other hand.[10–14] The purpose of this paper is to stress that both patterns of anomalous diffusion follow from an integro-differential approach whose basis are continuous-time random-walk (CTRW) models with coupled memories.[10,15–17] Our formalism connects the region $\gamma > 1$, typical for chaotic dynamics and turbulence, with the region $\gamma < 1$, which obtains from previously discussed temporal and geometric constraints. Interestingly, the *need* for an integral equation approach in the description of turbulence was noted long ago. Batchelor and Townsend remark in Ref. 4, p. 360: "Turbulent diffusion is not a local effect . . . , and a description of the diffusion by some kind of integral equation is more to be expected." However, they did not pursue this formalism.

Here we start by analyzing the differential equations in use, which lead to Eq. (1), and we subsequently introduce the CTRW integrodifferential picture.

II. DIFFERENTIAL EQUATIONS

The diffusion equation for the probability distribution $\rho(\mathbf{r},t)$

$$\frac{\partial\rho(\mathbf{r},t)}{\partial t} = \frac{\partial^2\rho(\mathbf{r},t)}{\partial\mathbf{r}^2} \tag{2}$$

(where $\partial^2/\partial\mathbf{r}^2$ is the d-dimensional Laplace operator and the diffusion constant D is set to unity) leads from an initial $\delta(\mathbf{r})$ form to Gaussian distributions:

$$\rho(\mathbf{r},t) = C_1 t^{-d/2}\exp(-r^2/2t) \ . \tag{3}$$

For such expressions one has $\langle r^2(t)\rangle = Ct$ at all times. Extensions which preserve the structure of Eq. (3) while allowing for $\langle r^2(t)\rangle \sim t^\gamma$ with $\gamma \neq 1$ center mostly on the form[1]

$$\frac{\partial\rho(\mathbf{r},t)}{\partial t} = \frac{\partial}{\partial\mathbf{r}}k(r,t)\frac{\partial}{\partial\mathbf{r}}\rho(\mathbf{r},t) \ . \tag{4}$$

Thus, in order to obtain $\gamma = 3$ Richardson[2] introduced $k(r,t) \equiv k(r) \sim r^{4/3}$, whereas Batchelor[3,4] used $k(r,t) \equiv k(t) \sim t^2$. Later Okubo[5] and Hentschel and Procaccia[6] suggested mixed algebraic forms such as $k(r,t) \sim tr^{2/3}$. The $d=3$ solution for $k(r,t) \sim t^a r^b$ and a $\delta(\mathbf{r})$ source at $t=0$ is given by[1,5,6,18]

$$\rho(\mathbf{r},t) = C_1 t^{-3(1+a)/(2-b)}\exp(-C_2 r^{2-b}/t^{1+a}) \ , \tag{5}$$

where C_1 and C_2 are independent of r and t. All forms $\rho(\mathbf{r},t)$ in Eq. (5) with $2a+3b=4$ lead to $\langle r^2(t)\rangle \sim t^3$. Note, however, that distributions $\rho(\mathbf{r},t)$ corresponding to different (a,b) pairs are quite different, a fact which allowed Sullivan[19] to decide experimentally in favor of the Batchelor form ($a=2$, $b=0$) over the Richardson solution ($a=0$, $b=\frac{4}{3}$) for turbulence in Lake Huron.

Furthermore, as remarked by Monin and Yaglom,[1] there is no compelling reason to use as differential equations solely forms related to Eq. (4). Another suggestion is to try [Eq. (24.91), p. 576 of Ref. 1, Vol. II]

$$\frac{\partial^m\rho(\mathbf{r},t)}{\partial t^m} = \frac{\partial^2\rho(\mathbf{r},t)}{\partial\mathbf{r}^2} \tag{6}$$

with $m=3$.

To show the underlying idea, we revert to the Fourier-Laplace space, $(\mathbf{r},t)\to(\mathbf{k},u)$. In this space the requirement $\langle r^2(t)\rangle \sim t^\gamma$ boils down to a simple condition on $\rho(\mathbf{k},u)$ in the limit $\mathbf{k}\to 0$, $u\to 0$.

Whereas the solution of Eq. (2) is

$$\rho(\mathbf{k},u)=(u+k^2)^{-1}\ , \tag{7}$$

one finds by Fourier-Laplace transforming Eq. (6) the formal expression

$$\rho(\mathbf{k},u)=u^{m-1}/(u^m+k^2) \tag{8}$$

under the requirements of conservation of probability $\rho(\mathbf{k}=0,t)=1$ so that $(\partial^p/\partial t^p)\rho(\mathbf{k}=0,t)=0$ for $p\geq 1$. Note that expressions of type (8) are purely formal, since there is in general no guarantee that they correspond to proper functions and that they satisfy all requirements for probability distributions, such as nonnegativity. On the other hand, for well-behaved functions, the mean-squared displacement follows from

$$\langle r^2(t)\rangle = \int \mathbf{r}^2\rho(\mathbf{r},t)d\mathbf{r} = -\frac{\partial^2}{\partial \mathbf{k}^2}\rho(\mathbf{k},t)\bigg|_{\mathbf{k}=0}\ , \tag{9}$$

where the right-hand side (rhs) may be found by an expansion in k^2. Using now the expression (8) we obtain

$$\langle r^2(u)\rangle \sim 1/u^{m+1}\ , \tag{10}$$

which is equivalent to $\langle r^2(t)\rangle \sim t^m$, as may be shown by Tauberian theorems.[20] For $m=3$, the desired relation $\langle r^2(t)\rangle \sim t^3$ follows.

The same argument can be used for the solutions of Eq. (4). The Batchelor solution is

$$\rho(k,t)=e^{-k^2t^3/3} \tag{11}$$

so that with Eq. (9) $\langle r^2(t)\rangle \sim t^3$. In the small $(\mathbf{k},u)$ limit one has from Eq. (11)

$$\rho(k,u)\sim \frac{1}{u} - C\frac{k^2}{u^4} \tag{12}$$

where again, as in Eq. (8), a term k^2/u^4 appears. The same holds for the Green's function of Eq. (4) as may be readily verified by computing the Fourier-Laplace transform of Eq. (5) in the small $(\mathbf{k},u)$ limit. Summarizing this section, we have deduced that in the $(\mathbf{k},u)$ space the requirement $\langle r^2(t)\rangle \sim t^3$ translates into Eq. (12), and that there are many differential equations whose Green's functions obey this form. For general $\langle r^2(t)\rangle \sim t^\gamma$, the requirement is, of course,

$$\rho(k,u)\sim \frac{1}{u} - C\frac{k^2}{u^{\gamma+1}}\ . \tag{13}$$

We now relate this expression to integro-differential equations, which appear naturally in the theory of random systems.

III. CONTINUOUS-TIME RANDOM WALKS (CTRW)

Another way to treat the dynamics of stochastic processes consists in following the trajectories of discrete particles: For a discrete underlying space this leads to random walks.[21]

Let $\psi(\mathbf{r},t)$ be the probability distribution of making a step of length $\mathbf{r}$ in the time interval t to $t+dt$. The total transition probability in this time interval is

$$\psi(t)=\sum_{\mathbf{r}}\psi(\mathbf{r},t)=\psi(\mathbf{k}=\mathbf{0},t)\ , \tag{14}$$

and the survival probability at the initial site is

$$\Phi(t)=1-\int_0^t \psi(\tau)d\tau\ , \tag{15}$$

so that

$$\Phi(u)=[1-\psi(u)]/u\ . \tag{16}$$

In standard fashion one has for the probability density $\eta(\mathbf{r},t)$ of just arriving at $\mathbf{r}$ in the time interval t to $t+dt$

$$\eta(\mathbf{r},t)=\sum_{\mathbf{r}'}\int_0^t \eta(\mathbf{r}',\tau)\psi(\mathbf{r}-\mathbf{r}',t-\tau)d\tau+\delta(t)\delta_{\mathbf{r},0} \tag{17}$$

in which the initial condition of starting at $t=0$ from $\mathbf{r}=\mathbf{0}$ is incorporated. Equation (17) leads to an integral equation for the probability $\rho(\mathbf{r},t)$ that the particle is at $\mathbf{r}$ at time t, by observing that

$$\rho(\mathbf{r},t)=\int_0^t \eta(\mathbf{r},t-\tau')\Phi(\tau')d\tau'\ . \tag{18}$$

With Eq. (18) and a change in the order of the integrations, Eq. (17) is recast into

$$\rho(\mathbf{r},t)=\sum_{\mathbf{r}'}\int_0^t \rho(\mathbf{r}',\tau)\psi(\mathbf{r}-\mathbf{r}',t-\tau)d\tau+\Phi(t)\delta_{\mathbf{r},0}\ . \tag{19}$$

Reverting to the Fourier-Laplace space Eq. (19) is

$$\rho(\mathbf{k},u)=\rho(\mathbf{k},u)\psi(\mathbf{k},u)+\Phi(u) \tag{20}$$

with the solution

$$\rho(\mathbf{k},u)=\frac{1-\psi(u)}{u}\,\frac{1}{1-\psi(\mathbf{k},u)}\ . \tag{21}$$

Before embarking on the discussion of Eq. (21), let us note that Eq. (19) is formally equivalent to the generalized master equation (GME):[16]

$$\frac{\partial}{\partial t}\rho(\mathbf{r},t)=\sum_{\mathbf{r}'}\int_0^t K(\mathbf{r}-\mathbf{r}',t-\tau)\rho(\mathbf{r}',\tau)d\tau \tag{22}$$

when one takes in Fourier-Laplace space

$$K(\mathbf{k},u)=\frac{\psi(\mathbf{k},u)-\psi(u)}{1-\psi(u)}u\ . \tag{23}$$

The equivalence is immediate when transforming Eq. (22) to $(\mathbf{k},u)$ space and comparing to Eq. (23). Note that Eq. (22) is an integro-differential equation.

Moreover, in Ref. 16 it has been shown that the ensemble averaged transport through substitutionally disordered media obeys *exactly* the CTRW equation (19), with a probability distribution $\psi(\mathbf{r},t)$ in which $\mathbf{r}$ and t are coupled. Decoupling, i.e.,

$$\psi(\mathbf{r},t)=\lambda(\mathbf{r})\psi(t) \tag{24}$$

obtains in special cases, such as ordered arrays.[15] Furthermore, many functional $\psi(\mathbf{r},t)$ forms result from different types of disorder.[16]

We thus view Eqs. (19) and (21) in their general form as the natural extension of diffusive processes to stochastic media. Note that Eq. (19) guarantees that the $\rho(\mathbf{r},t)$ stays nonnegative, a requirement difficult to implement otherwise.

Let us turn to Eq. (21) and look at the influence of the moments

$$\tau_1=\int dt\, t \int \psi(\mathbf{r},t)d\mathbf{r} \tag{25}$$

and

$$\sigma^2=\int dt \int r^2\psi(\mathbf{r},t)d\mathbf{r} \ . \tag{26}$$

For finite τ_1 and σ^2 Eq. (19) corresponds for small $(\mathbf{k},u)$ to a simple diffusion equation. One has

$$\psi(\mathbf{k},u)\sim 1-\tau_1 u - k^2\sigma^2/2 \ , \tag{27}$$

which inserted into Eq. (21) gives $\rho(\mathbf{k},u)\sim(u+Ck^2)^{-1}$ with $C=\sigma^2/2\tau_1$, i.e., Eqs. (7) and (2), from which $\langle r^2(t)\rangle \sim Ct$ follows. Furthermore, when σ^2 is finite, but τ_1 is infinite, say

$$\psi(\mathbf{k},u)\sim 1-C_1u^{\gamma}-k^2\sigma^2/2 \tag{28}$$

with $0<\gamma<1$, and from Eq. (21) it follows that

$$\rho(k,u)\simeq\frac{u^{\gamma-1}}{u^{\gamma}+Ck^2} \tag{29}$$

with $C=\sigma^2/2C_1$. Eq. (29) may be identified with Eq. (8) by setting $\gamma=m$, and hence, following Eqs. (9)ff one finds $\langle r^2(t)\rangle \sim t^{\gamma}$, with $\gamma<1$, i.e., dispersive transport.

On the other hand, for finite τ_1 but infinite σ^2, say

$$\psi(\mathbf{k},u)\sim 1-\tau_1 u - C_2 k^{\beta} \tag{30}$$

with $0<\beta<2$, one has from Eq. (21)

$$\rho(\mathbf{k},u)\sim\frac{1}{u+Ck^{\beta}} \tag{31}$$

with $C=C_2/\tau_1$. Now the second moment of $\rho(\mathbf{r},t)$ diverges, as is immediate by applying Eq. (9) to $\rho(\mathbf{k},t)\sim \exp(-Ck^{\beta}t)$. If the equality obtained in Eq. (31), for $\beta=\frac{2}{3}$, $d=3$ the function $\rho(\mathbf{r},t)$ can be expressed in closed form [Monin and Yaglom, Ref. 1, Eq. (24.89)]. Then, indeed, $\langle r^2(t)\rangle$ is divergent for all $t>0$.

In the same vein, for infinite τ_1 and σ^2, say

$$\psi(\mathbf{k},u)\sim 1-C_1u^{\gamma}-C_2k^{\beta} \tag{32}$$

one verifies easily that $\langle r^2(t)\rangle$ is again divergent for all $t>0$. Note that Eq. (32) always follows from a decoupled picture $\psi(\mathbf{k},u)=\lambda(\mathbf{k})\psi(u)$ with infinite τ_1 and σ^2.

In order to obtain finite $\langle r^2(t)\rangle$ which follow t^{γ} with $\gamma>1$ it is thus imperative to use *coupled* $\psi(\mathbf{r},t)$ forms.[9,16,17] A suitable function is[22,23]

$$\psi(\mathbf{r},t)=Cr^{-\mu}\delta(r-t^{\nu}) \tag{33}$$

where, through the δ function, r and t are coupled. Equation (33) allows steps of arbitrary length, but long steps are penalized by requiring more time to be performed. Or, stated differently, in a given time window only a finite shell of points may be reached: hierarchically, nearer points are no more and farther points not yet accessible. Now,

$$\begin{aligned}\psi(u)&=\int_0^{\infty}dt\int_{\delta}d\mathbf{r}\,\psi(\mathbf{r},t)e^{-ut}\\ &\sim\int_0^{\infty}dr\,r^{d-\mu-1}\int_{\delta_1}^{\infty}\delta(r-t^{\nu})e^{-ut}dt\\ &=\int_{\delta_2}^{\infty}t^{\nu(d-\mu-1)}e^{-ut}dt=\int_{\delta_2}^{\infty}t^{-\nu\mu^*}e^{-ut}dt \ ,\end{aligned} \tag{34}$$

where we set $\mu^*=\mu-d+1$, and indicated the lower bound cutoffs by δ_i.

From the requirement $\psi(u=0)=1$ one must have $\nu(\mu-d+1)=\nu\mu^*>1$. Furthermore, for $\nu(\mu-d+1)=\nu\mu^*>2$ one obtains a finite τ_1. Calculating $\psi(\mathbf{k},u)$ we find

$$\begin{aligned}\psi(\mathbf{k},u)-\psi(u)&=\int_0^{\infty}dt\int_{\delta}^{\infty}d\mathbf{r}[e^{i\mathbf{k}\cdot\mathbf{r}}-1]\psi(\mathbf{r},t)e^{-ut}\\ &\sim k^2\int_0^{\infty}\int_{\delta}^{\infty}dr\,r^{-\mu^*+2}\delta(r-t^{\nu})e^{-ut}dt\\ &=-k^2\int_{\delta_2}^{\infty}t^{-\nu(\mu^*-2)}e^{-ut}dt\equiv -k^2I(u) \ .\end{aligned} \tag{35}$$

For $\nu(\mu^*-2)>1$ the integral $I(u)$ exists even for $u=0$. In this case, with finite τ_1 one has

$$\psi(\mathbf{k},u)\sim 1-\tau_1 u - C_1k^2 \ , \tag{36}$$

and we recover Brownian behavior, for which $\langle r^2(t)\rangle \sim t$. For $\nu(\mu^*-2)<1$ we find that $I(u)$ diverges for $u=0$. Only for $u>0$ the integral converges, and then

$$\psi(\mathbf{k},u)-\psi(u)\sim -k^2u^{\nu(\mu^*-2)-1}$$

such that

$$\psi(\mathbf{k},u)\sim 1-\tau_1 u - C_1k^2u^{\nu(\mu^*-2)-1} \ . \tag{37}$$

One may note the coupled form of Eq. (37), namely that the first k^2 term involves u. From Eqs. (21) and (37) it follows that

$$\rho(\mathbf{k},u)\sim\frac{u^{\nu(2-\mu^*)+1}}{\tau_1 u^{\nu(2-\mu^*)+2}+C_1k^2} \tag{38}$$

and hence

$$\langle r^2(t)\rangle \sim t^{\nu(2-\mu^*)+2} \ . \tag{39}$$

Turning now to the case of infinite τ_1 we obtain from Eq. (34) for $1<\nu\mu^*<2$:

$$\psi(u)\sim 1-Cu^{\nu\mu^*-1} \ . \tag{40}$$

For $\nu(\mu^*-2)>1$ the integral in Eq. (35) is finite for $u=0$.

Thus,

$$\psi(\mathbf{k},u)\sim 1-Cu^{\nu\mu^*-1}-C_1k^2 \ , \tag{41}$$

and we have $\langle r^2(t)\rangle \sim t^{\nu\mu^*-1}$, i.e., dispersive transport.[10,13] For $\nu(\mu^*-2)<1$ the integral $I(u)$ converges only for $u>0$, and we recover Eq. (38). Now,

$$\psi(\mathbf{k},u)\sim 1-Cu^{\nu\mu^*-1}-C_1k^2u^{\nu(\mu^*-2)-1} \tag{42}$$

and thus,

$$\rho(\mathbf{k},u)\sim\frac{u^{\nu\mu^*-2}}{u^{\nu\mu^*-1}+C_1k^2u^{\nu(\mu^*-2)-1}}=\frac{u^{2\nu-1}}{u^{2\nu}+C_1k^2} \ . \tag{43}$$

TABLE I. Behavior of $\langle r^2(t)\rangle$ as a function of ν and of $\mu^*=\mu-d+1$ when the $\psi(\mathbf{r},t)$ follows Eq. (33).

	$\tau_1<\infty$ $\nu\mu^*>2$		$\tau_1=\infty$ $1<\nu\mu^*<2$	
$I(0)<\infty$ in Eq. (35) $\nu\mu^*-2\nu>1$	t		$t^{\nu\mu^*-1}$	
		$\nu=\frac{1}{2}$ $\mu^*=4$		$\mu^*=2+1/\nu$
$I(0)=\infty$ in Eq. (35) $\nu\mu^*-2\nu<1$	$t^{2-\nu\mu^*+2\nu}$		$t^{2\nu}$	
		$\mu^*=2/\nu$		

Hence $\langle r^2(t)\rangle \sim t^{2\nu}$. This is again our expression for anomalous transport; for $\nu<\frac{1}{2}$ the transport is dispersive while for $\nu>\frac{1}{2}$ it is enhanced.

The $\langle r^2(t)\rangle$ behavior in the different (ν,μ^*) regions is summarized in Table I. We note that the transitions between the regions are smooth, when abstraction is made of possible logarithmic corrections.

A few remarks on Table I are in order. Keeping ν fixed while varying μ^* from large ($\mu^* \gg 2/\nu$) to small ($\mu^* \ll 2/\nu$) values permits to cover the region from Brownian to anomalous behavior. The marginal value for ν is $\frac{1}{2}$, for which one passes for $\mu^*=4$ through the central point of the table. For $\nu<\frac{1}{2}$ the small μ^* behavior is dispersive and one verifies readily that in the intermediate μ^* region $t^{\nu\mu^*-1}$ is obeyed. For $\nu>\frac{1}{2}$ the diffusion is enhanced for small μ^* values, and the intermediate behavior is $t^{2-\nu\mu^*+2\nu}$. Two special cases for $\nu>\frac{1}{2}$ were already discussed in Refs. 9 and 22: The value $\nu=\frac{3}{2}$ corresponds to models for turbulence,[22] whereas $\nu=1$ was used to describe the behavior of chaotic maps.[9]

IV. CONCLUSIONS

In this work we have shown that CTRW allows the straightforward extension of Brownian motion to anomalous transport, both in the dispersive and in the enhanced cases. Whereas in the CTRW framework the dispersive transport may also be obtained from decoupled kernels,[10,13] the enhanced diffusion can only be achieved by using kernels which couple time and space, e.g., Eq. (33). In our opinion random walk models offer a series of advantages over approaches which utilize extensions of diffusion-type equations to mimic anomalous transport. Firstly, random walks offer a dynamical picture of the motion, which allows to follow the course of chemical reactions in complex situations, as we have already demonstrated for dispersive motion.[24] Second, the CTRW description ensures that the time development of a delta-pulse remains at all times a probabilistically well-defined object (non-negative, integrable, and normalized), a feature which cannot always be taken for granted when manipulating partial differential equations. One may also note that the introduction of time or distance dependent diffusion coefficients in Eq. (4) renders the basic picture inhomogeneous, whereas the CTRW Eq. (21) is homogeneous in space and time.

The particular kernel used, Eq. (33), shows a very rich pattern. In the enhanced diffusion regime the chaotic dynamics and also aspects of turbulent flow appear as special cases: The transitions show an intermediate zone between the Brownian motion and the fully developed enhanced diffusion. Interestingly, an intermediate zone is also found for dispersive transport, where the transition between Brownian behavior and largest possible dispersion is gradual. This finding is comparable to the transition from Brownian to dispersive motion in ultrametric spaces.[8,14]

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