Anomalous diffusion from Brownian motion with random confinement

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We present a model of anomalous diffusion consisting of an ensemble of particles undergoing homogeneous Brownian motion except for confinement by randomly placed reflecting boundaries. For power-law distributed compartment sizes, we calculate exact and asymptotic values of the ensemble averaged mean squared displacement and find that it increases subdiffusively, as either a power or the logarithm of time. Numerical simulations show that the probability density function of the displacement is non-Gaussian. We discuss the relevance of the model for the analysis of single-particle tracking experiments and its relation to other sources of subdiffusion. In particular we discuss an intimate connection with diffusion on percolation processes.

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In the study of diffusion in physical systems it is well known that a variety of processes lead to diffusion that deviates from pure Brownian motion\textsuperscript{[1–6]}. The most commonly studied signature of anomalous diffusion is the mean squared displacement (MSD), which often takes the form of a power law,

\[ \langle x^2(t) \rangle \sim t^\beta, \quad (1) \]

where \( x(t) \) is the displacement of a particle at time \( t \). For Brownian motion \( \beta = 1 \), and for subdiffusive motion \( \beta < 1 \). Subdiffusive systems may be successfully modeled by various random processes such as continuous time random walk (CTRW)\textsuperscript{[7–9]}, fractional Brownian motion\textsuperscript{[10, 11]} (FBM), diffusion on percolation\textsuperscript{[12]} (DOP), Lorentz models\textsuperscript{[5]}, or by a combination of these\textsuperscript{[13, 14]}. The problem of distinguishing which processes contribute to or modify anomalous diffusion is an active area of research\textsuperscript{[15–18]}. In particular, one may need to model complicating factors such as confinement. Confinement plays an important role in modifying or attenuating the subdiffusion manifested by the power-law behavior in (1)\textsuperscript{[19–26]}. But, in this paper we take a different view of the relation of confinement to subdiffusion: We show that random confinement may in fact be the sole cause of observed subdiffusion. While the effect is quite general, the model and analysis are motivated specifically by single particle tracking (SPT) experiments, in particular studies of the biophysics of cells\textsuperscript{[27–30]}. We present a scenario in which disordered confinement produces quantitative signatures of subdiffusion in a typical analysis of SPT data. Given that heterogeneous confining boundaries are often observed in biophysics, it follows that in experiments searching for contributions to the subdiffusion exponent \( \beta \), confinement may not be discarded \textit{a priori} as a candidate.

\textit{Experimental scenario}— Consider a number of Brownian particles uniformly distributed in a space that is partitioned into compartments by a random arrangement of reflecting barriers. Apart from the presence of the boundaries, the motion is diffusive, with parameters homogeneous in space and time. We collect an SPT trajectory (time series) for each of several particles sampled uniformly from the space. The trajectory consists of the displacement of each particle from its starting point recorded at a series of times. Typically, the trajectories are first analyzed via the ensemble averaged mean squared displacement (EMSD), or the time-ensemble averaged mean squared displacement (TEMSD), where the TEMSD consists, operationally, of first computing a time (sliding) averaged MSD (TMSD) for each trajectory and then averaging the result over the trajectories. For the minimal model introduced below, we find

- The EMSD is unbounded and subdiffusive in the sense of (1) if the distribution of the linear size of the compartments has a sufficiently heavy-tail.
- There is no weak ergodicity breaking (if the system is in equilibrium): the EMSD is equal to the TEMSD.
- The TMSD (ie for a single trajectory) tends to a constant. That is, the observables of a trajectory are not self-averaging over disorder.

\textit{The model}— We choose a minimal model that captures the essential features and displays asymptotically subdiffusive motion. We refer to the particular model presented below as the random scale-free confinement model (RSFC). We state the results for the MSD before giving the detailed calculations. In this paper, we treat in detail only one spatial dimension, stating some results for higher dimension at the end, but leaving details to a subsequent paper. The probability density for the displacement of a particle diffusing on a line segment of length \( r \) with reflecting boundary conditions obeys

\[ \partial_t u(x; t) = \partial^2_x u(x; t), \quad (2) \]

with \( \partial_x u(r/2; t) = \partial_x u(-r/2; t) = 0 \). We assume that the probability for a particle to be found in a segment of
In (4) and (5) angle brackets denote averaging over particle trajectories, the tilde denotes averaging over the disorder (ie over $r$), and $K_1, K_2, K_3, K_4$ are constants that depend on dimension, and the initial distribution of particles. Examples of curves for three values of $\alpha$ showing both bounded and unbounded EMSD are shown in Fig. 1.

**Comparison with other sources of subdiffusion**—Our random confinement model does not manifest weak ergodicity breaking [31], a characteristic whose presence is known to imply dramatic differences between EMSD and TEMSD observed in SPT experiments [32–37]. On the contrary, the model is in equilibrium at $t = 0$, and the increments of $x(t)$ are stationary so that the TEMSD and EMSD are equal at all times. Thus, the subdiffusion is due to correlated increments, as is the case for both DOP and FBM. Our simulations show that the PDF of the displacement for large times is non-Gaussian as shown in Fig. 2, a feature shared by DOP, and CTRW, but not FBM. RSFC and DOP (but not FBM and CTRW) are examples of diffusion on disordered, or more broadly, heterogeneous media. A model of this type that is closely related to RSFC considers domains of random scale-free size and diffusivity [38]. Despite this similarity, the latter model has deeper similarities to other models of heterogeneous transport coefficients [6, 39–45]. When these models do show subdiffusion, it arises from non-stationary increments, and they are thus more closely related to CTRW.

**RSFC and percolation**—It is interesting to compare RSFC more closely to DOP. We distinguish two cases: The first is diffusion of a particle starting at a randomly chosen site on a percolation process at the critical threshold [12] (DOP I). In the second case the initial site is a random site on the critical infinite cluster (DOP II). Both DOP I and DOP II show subdiffusive EMSD, but with differing values of the anomalous diffusion exponent $\beta$ in (1) [12, 46, 47]. For DOP II, subdiffusion is due to the fractal properties of the infinite cluster. However, because the volume fraction of the critical infinite cluster is zero, it gives no weight in DOP I where the initial position may be any point on the lattice. As is the case with the pure random confinement of RSFC, in DOP I every particle is confined to a region of finite size. Thus, for both RSFC and DOP I, the time-averaged mean squared displacement (TMSD)—the time average of a single trajectory—tends to a constant at long times. In DOP I, the size of the confinement regions (the finite clusters) has a heavy-tailed, power-law distribution. Thus, as for RSFC, averaging over an ensemble of uniformly distributed particles gives an unbounded, subdiffusive MSD, with an additional contribution from the fractal structure of large clusters. In other words, The fact that $\beta$ differs between (I) the walk on all clusters and (II) the walk on the infinite cluster, is due to random, scale-free confinement and RSFC represents an abstraction of this phenomenon.

We address a potential issue in simulations and experiments. In CTRW, a finite number of trajectories show a subdiffusive EMSD no matter how long their duration, though this implies an ever increasing spatial domain. In DOP I and RSFC, the average over a finite number of trajectories will tend to a constant EMSD at long times, even for $\alpha < 2$. But, simulations and SPT trajectories are typically limited to times shorter than the time $T$ required to explore the entire experimental domain. Since a heavy-tail ($\alpha < 2$) implies that there are compartments whose area is of the order of the experimental domain, the time required to cross these domains will be of the same order as $T$, and thus power-law subdiffusion will be observed as shown in Fig. 1. Note that: In CTRW, no particle is trapped for an infinite time, yet the step rate decreases toward zero. In RSFC, the motion of every...
The EMSD averaged over the disorder is given by

\[ \langle u^2 \rangle = \int_0^\infty \langle u^2(t; r) \rangle P(r) \, dr. \]

Separating time and space variables, the solution to (2) may be written as an eigenfunction expansion

\[
u(x; t) = \sum_{n=0}^\infty [a_n e^{-4\pi^2 n^2 t} \cos(2\pi nx) + b_n e^{-4\pi^2 (n+1/2)^2 t} \sin(2\pi [n + 1/2])],
\]

with coefficients determined by the initial condition and orthogonality of the eigenfunctions. Using the initial condition \( u(x, 0) = \delta(x - x_0) \), we find

\[
u(x; x_0, t) = 1 + 2 \sum_{n=0}^\infty e^{-4\pi^2 n^2 t} \cos(2\pi nx) \cos(2\pi nx_0)
+ 2 \sum_{n=0}^\infty e^{-4\pi^2 (n+1/2)^2 t} \sin(2\pi [n + 1/2]x) \sin(2\pi [n + 1/2]x_0).
\]

(8)

Evaluating the integrals obtained by substituting (8) into (6) is straightforward and results in

\[
\pi(y, t) = \left(1 - |y|\right) + \sum_{n=1}^\infty e^{-\pi^2 n^2 t} \left[ (1 - |y|) \cos(\pi n|y|) - \frac{\sin(\pi n|y|)}{\pi n} \right].
\]

(9)

The asymptotic shape of \( \pi(y, t) \) is reflected in the central portion of the disorder-averaged density as seen in Fig. 2.

Inserting (9) into (7) we find

\[
\langle x^2(t; 1) \rangle = \frac{1}{6} - \frac{1}{\pi^4} \sum_{n=0}^\infty \frac{1}{(n+1/2)^4} \frac{1}{(n + 1/2)^4}.
\]

(10)

We now consider a segment of length \( r \), rather than 1, and use (10) to write

\[
\langle x^2(t; r) \rangle = \frac{r^2}{\pi^4} \sum_{n=0}^\infty \left(1 - e^{-4\pi^2 (n+1/2)^2 t/r^2}\right) \frac{1}{(n + 1/2)^4},
\]

where we have absorbed the initial term into the sum in order to cancel two diverging quantities below. Averaging over the disorder we have

\[
\langle x^2(t) \rangle = \frac{4}{\pi} \sum_{n=0}^\infty \frac{1}{(n + 1/2)^4} \times 
\int_0^\infty \left(1 - e^{-4\pi^2 (n+1/2)^2 t/r^2}\right) r^2 P(r) \, dr 
\]

\[
= \frac{4}{\pi^{3/2}} \sum_{n=0}^\infty \frac{1}{(n + 1/2)^4} \times 
\int_0^\infty \left(1 - e^{-y}\right) y^{-5/2} P \left(2\pi(n + 1/2)\sqrt{t/y}\right) \, dy.
\]

(11)
Inserting the Pareto distribution (3) into (11), we have
\[
\langle x^2(t) \rangle = \frac{\alpha r_0^2}{2\pi^2} \sum_{n=0}^{\infty} \frac{1}{(n+1/2)^2} z^{2-n} \int_0^z (1 - e^{-y}) y^{\alpha-2-1} dy \\
= \frac{\alpha r_0^2}{2\pi^2} \sum_{n=0}^{\infty} \frac{1}{(n+1/2)^2} \text{Ein} \left( \frac{\alpha - 2 - 4\pi^2(n+1/2)^2 t}{\alpha r_0^2} \right),
\]
where \( z = 4\pi^2(n+1/2)^2 t/r_0^2 \) and we have defined a generalized entire exponential integral by
\[
\text{Ein}(s, z) = z^{-s} \int_0^z (1 - e^{-y}) y^{s-1} dy.
\]
The series obtained by expanding the exponential[48] is easily seen to be analytic for \( s, z \in \mathbb{C} \) except for simple poles at \( s = -1, -2, \ldots \). In particular, (13) is analytic at \( s = 0 \), where \( \text{Ein}(0, z) = \text{Ein}(z) \), the usual entire exponential integral. This corresponds to the critical value of the power-law exponent \( \alpha = 2 \). It can be shown that for \( \alpha \neq 2 \), \( \text{Ein}(s, z) = 1/s - z^{-s} \gamma(s, z) \), where \( \gamma(s, z) \) is the lower incomplete gamma function. Large \( t \) corresponds to large \( z \), so, that using \( \lim_{z \to \infty} \gamma(s, z) = \Gamma(s) \), we find the asymptotic form
\[
\langle x^2(t) \rangle = \frac{\alpha r_0^2}{\alpha - 2} \frac{\alpha - 2}{4} - 2 \alpha \pi^{\alpha-2} - 2 \alpha r_0^2 \Gamma \left( \frac{\alpha - 2}{2} \right) \zeta(2 + \alpha, 1/2) t^{\frac{2-\alpha}{2}},
\]
where \( \zeta(s, z) \) is the Hurwitz Zeta function. Eq. (14) shows that for \( \alpha > 2 \), \( \langle x^2(t) \rangle \) converges to a constant at long times, while for \( 1 < \alpha < 2 \), \( \langle x^2(t) \rangle \) shows sub-diffusion. For \( \alpha = 2 \), we use \( \text{Ein}(0, z) = \text{Ein}(z) = \ln(z) + \gamma + \Gamma(0, z) \) and \( \lim_{z \to \infty} \Gamma(s, z) = 0 \) to find the asymptotic solution
\[
\langle x^2(t) \rangle = \frac{r_0^2}{6} \left[ \gamma + 2 \ln \left( \frac{2\pi}{r_0} \right) - \frac{12\partial_\eta \zeta(4, 1/2)}{\pi^4} + \ln(t) \right],
\]
where \( \partial_\eta \zeta(s, 1/2) \big|_{s=4} \approx 10.9697 \). Equations (14) and (15) are the goals of the calculations and are shown in Fig. 1.

**Non-equilibrium initial conditions**— Although we assumed equilibrium above, subdiffusion is also observed for other initial distributions. We performed calculations analogous to those above for the initial distribution \( u(x, 0) = \delta(x) \), that is, each particle begins at the center of the confinement domain, and obtained the asymptotic form for \( \alpha \neq 2 \)
\[
\langle x^2(t) \rangle = \frac{\alpha r_0^2}{\alpha - 2} - 2 \alpha \pi^{\alpha-2} - 2 \alpha r_0^2 \Gamma \left( \frac{\alpha - 2}{2} \right) \eta(\alpha) t^{\frac{2-\alpha}{2}},
\]
where \( \eta(x) \) is the Dirichlet eta function. For \( \alpha = 2 \) we found
\[
\langle x^2(t) \rangle = \frac{r_0^2}{6} \left( \frac{\ln(t)}{2} - \frac{6\zeta(2)}{\pi^2} + \ln(\pi) + \frac{\gamma}{2} - \ln(r_0) \right),
\]
where \( \zeta(x) \) is the Riemann zeta function. In two dimensions, ie diffusion on disks of random radius, we obtained the asymptotic forms, for \( \alpha \neq 2 \)
\[
\langle \rho^2(t) \rangle = \frac{\alpha r_0^2}{\alpha - 2} - 2 \alpha r_0^2 \Gamma \left( \frac{\alpha - 2}{2} \right) \zeta_{11}(\alpha) t^{\frac{2-\alpha}{2}},
\]
where \( j_{\nu,n} \) is the \( n \)th positive zero of the Bessel function of the first kind \( J_\nu \), and \( \zeta_{11}(\beta) \) is
\[
\zeta_{11}(\beta) = \sum_{n=1}^{\infty} \frac{1}{J_{\nu+1}(j_{\nu,n})^2 \nu + \Gamma},
\]
and \( \rho(t) \) is the radial density, ie averaged over azimuthal angle. For \( \alpha = 2 \), we found
\[
\langle \rho^2(t) \rangle = \frac{r_0^2}{6} \left( \gamma - \ln(r_0^2) - 16\partial_\beta \zeta_{11}(2) + \ln(t) \right),
\]
where \( \partial_\beta \zeta_{11}(2) \approx 0.147342 \).

**Conclusion**— We have demonstrated that heavy-tailed random scale-free confinement gives rise to a subdiffusive EMSD. We have discussed its relation to other sources of anomalous diffusion, in particular its presence in certain diffusive processes on percolation. A number of questions remain, including: Are there random potential fields that lead to RSFC? What is nature of the propagator (PDF) of RSFC? ... or of autocorrelation of the displacement? What is the anomalous exponent when RSFC is combined with other sources of anomalous diffusion?

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