

Diffusing Diffusivity: A Model for Anomalous, yet Brownian, Diffusion

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Wang *et al.* [Proc. Natl. Acad. Sci. U.S.A. 106, 15160 (2009)] have found that in several systems the linear time dependence of the mean-square displacement (MSD) of diffusing colloidal particles, typical of normal diffusion, is accompanied by a non-Gaussian displacement distribution $G(x, t)$, with roughly exponential tails at short times, a situation they termed “anomalous yet Brownian” diffusion. The diversity of systems in which this is observed calls for a generic model. We present such a model where there is diffusivity memory but no direction memory in the particle trajectory, and we show that it leads to both a linear MSD and a non-Gaussian $G(x, t)$ at short times. In our model, the diffusivity is undergoing a (perhaps biased) random walk, hence the expression “diffusing diffusivity”. $G(x, t)$ is predicted to be exactly exponential at short times if the distribution of diffusivities is itself exponential, but an exponential remains a good fit for a variety of diffusivity distributions. Moreover, our generic model can be modified to produce subdiffusion.

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In a homogeneous Newtonian fluid like water, the diffusion of microscopic particles obeys laws of Brownian motion known since Einstein [1]: The mean-square displacement (MSD) $\langle x^2 \rangle$ along a particular direction, x , is linear in time t ,

$$\langle x^2 \rangle = 2Dt, \quad (1)$$

where D is the diffusion coefficient, or the diffusivity, while the distribution of displacements is Gaussian [2]. In crowded fluids containing colloidal particles, macromolecules, filaments, etc., Eq. (1) is generally not valid at all times. In many such cases (see, e.g., Refs. [3–13]), experimental data are consistent with $\langle x^2 \rangle \propto t^\nu$, where $\nu < 1$, over a significant time range. This is called anomalous diffusion or, more specifically, subdiffusion.

While much work has concentrated on the MSD, the full displacement distribution (DispD) can be measured using single-particle tracking (SPT) [3–6,11,12,14,15]. In the continuous-time random walk (CTRW) model of anomalous diffusion [16,17], the DispD is significantly non-Gaussian with a characteristic cusp at $x = 0$ [18]. However, the fractional Brownian motion (fBm) model [19–21] demonstrates that the combination of anomalous MSD with a Gaussian DispD is possible.

On the other hand, it is often tacitly assumed that if the DispD is non-Gaussian, then the factors that cause it to deviate from Gaussian should also make the MSD non-linear. Recent SPT experiments by Granick’s group [22,23] show that this is not always the case. Several systems were considered: polystyrene beads on the surface of a lipid bilayer tube [22], beads in an entangled solution of actin filaments [22], and liposomes in a nematic solution of actin filaments [23]. In all three systems, the MSD is linear over

the whole experimental time range (from ~ 0.1 s to \sim seconds). Yet, coexisting with this linear MSD is a strongly non-Gaussian DispD, with approximately exponential tails. When a crossover to Gaussian DispD is observed, the linear MSD dependence continues without any peculiarities. This was termed “anomalous yet Brownian” diffusion [22]. Similar behavior was also observed for diffusion of tracer molecules on polymer thin films [24] and in simulations of a two-dimensional system of discs [25]. Since this is observed in several different cases, it is likely a generic feature of a certain class of systems. The goal of this paper is to show that this may indeed be the case, by proposing a simple and generic *diffusing diffusivity model* that indeed exhibits this behavior.

Consider an unbiased one-dimensional random walk (RW) with particle displacement Δx_i at step i ($i = 1, \dots, N$) and a constant step duration Δt . The total displacement after N steps is $x_N = \sum_{i=1}^N \Delta x_i$. The MSD is

$$\langle x_N^2 \rangle = \sum_{i=1}^N \langle (\Delta x_i)^2 \rangle + 2 \sum_{i=1}^{N-1} \sum_{j=i+1}^N \langle \Delta x_i \Delta x_j \rangle. \quad (2)$$

The second sum in Eq. (2) is zero if the steps are uncorrelated ($\langle \Delta x_i \Delta x_j \rangle = 0$); if the ensemble-averaged magnitude of a step is then time-independent (a condition not satisfied for the CTRW models of anomalous diffusion), the first sum and thus the whole MSD is proportional to N . Note that complete independence of steps is *not necessary* for this to be the case: It is sufficient for the step *directions* to be uncorrelated (given Δx_i , displacements Δx_j and $-\Delta x_j$ should be equiprobable, a condition violated by the fBm). However, correlations of the step *lengths* are

still allowed, in which case a non-Gaussian DispD may result despite a linear MSD.

In fact, such correlations of step magnitudes without correlations of step directions are to be expected in heterogeneous systems where the environment changes slowly in space and time. Over length and time scales smaller than those of these heterogeneities, we can describe a local environment approximately by its effective diffusivity. The idea then is to think of a process where on a short time scale particles undergo regular normal diffusion (but with diffusivities different for different particles depending on their local environment), but on a longer time scale, as the environment changes slowly (either on its own, or because the particle moves to a different environment, or both), the diffusivity of each particle changes gradually. This leads to long-term correlations between step magnitudes: since a long step Δx is more likely to be associated with a region of high diffusivity, subsequent steps of the same particle are also likely to be longer than average, until the environment changes. However, the step directions remain uncorrelated.

In the spirit of the preceding discussion, consider a model in which noninteracting particles diffuse in one dimension, each with its own instantaneous diffusion coefficient that varies with time. Over a fixed $\Delta t = 1$, a specific particle with diffusivity D_i at time step i is displaced by amount Δx_i drawn from the Gaussian distribution

$$P(\Delta x_i) = \frac{1}{\sqrt{4\pi D_i}} \exp\left(-\frac{\Delta x_i^2}{4D_i}\right). \quad (3)$$

In the stationary state, the diffusivity distribution is time-independent, and the ensemble-averaged MSD is linear in the time (or the number of steps $N = t/\Delta t$):

$$\langle x_N^2 \rangle = \sum_{i=1}^N \langle \Delta x_i^2 \rangle = 2 \sum_{i=1}^N \langle D_i \rangle = 2\langle D \rangle N. \quad (4)$$

On the other hand, the fourth moment of the DispD deviates from its Gaussian value, with the deviation being:

$$\begin{aligned} \langle x_N^4 \rangle - 3\langle x_N^2 \rangle^2 &= 12(\langle D^2 \rangle - \langle D \rangle^2)N \\ &+ 24 \sum_{i=1}^{N-1} \sum_{j=i+1}^N (\langle D_i D_j \rangle - \langle D_i \rangle \langle D_j \rangle). \end{aligned} \quad (5)$$

Suppose the largest relaxation time of the correlator $\langle D_i D_j \rangle - \langle D_i \rangle \langle D_j \rangle$ as a function of $|i - j|$ is τ_D . To model a slowly changing environment, D_i should vary little from step to step, thus, $\tau_D \gg 1$. Then the double sum in Eq. (5) dominates. Assuming $\langle D^2 \rangle - \langle D \rangle^2 \sim \langle D \rangle^2$, the non-Gaussianity parameter $\alpha_2 = \langle x^4 \rangle / 3\langle x^2 \rangle^2 - 1 \sim 1$ for $N \lesssim \tau_D$ and $\alpha_2 \sim \tau_D / N$ for $N \gg \tau_D$, which only decays as $1/N$. Thus, significant deviations from Gaussianity are expected well above τ_D , especially when looking at the tails of the

DispD. This is an important point when interpreting experimental results.

Let us now make specific assumptions about the evolution of D for individual particles. Since this evolution is expected to be quasirandom in a complex system, we assume that D undergoes a (perhaps biased) random walk (diffusing diffusivity). Our assumption that D changes little from step to step allows us to switch to a continuous time t . Then the diffusivity distribution (DifD) $\pi(D; t)$ satisfies the usual advection-diffusion equation,

$$\frac{\partial \pi(D; t)}{\partial t} = -\frac{\partial J}{\partial D}, \quad (6)$$

$$-J = \frac{\partial}{\partial D} [d(D)\pi(D; t)] + s(D)\pi(D; t), \quad (7)$$

where $d(D)$ can be referred to as the diffusivity of the diffusivity and $-s(D)$ is the bias of the diffusion of the diffusivity. Since D cannot be negative or higher than the free-solution diffusivity D_{\max} , we add reflecting boundary conditions $J = 0$ at $D = 0$ and $D = D_{\max}$. In what follows, unless stated otherwise, we assume that the system is in the stationary state with the distribution $\pi(D)$ defined by $J(D) = 0$ for all D .

Over times $t \ll \tau_D$ the diffusivity of a particle can be assumed constant. The DispD for an ensemble of particles over such times does not depend on the diffusivity diffusion, but only on $\pi(D)$, and is given by [23,26]

$$G(x; t) = \int_0^{D_{\max}} \frac{\pi(D)}{2\sqrt{\pi D t}} \exp\left(-\frac{x^2}{4Dt}\right) dD, \quad t \ll \tau_D. \quad (8)$$

In the simplest reasonable case $d(D) = \text{const}$, $s(D) = \text{const}$ ($s > 0$ is necessary to bias D towards lower values, as it should be in crowded systems). For $D_{\max} \rightarrow \infty$, this gives

$$\pi(D) = \frac{1}{D_0} \exp(-D/D_0), \quad D_0 = d/s, \quad (9)$$

and [27]

$$G(x; t) = \frac{1}{2\sqrt{D_0 t}} \exp\left(-\frac{|x|}{\sqrt{D_0 t}}\right), \quad t \ll \tau_D, \quad (10)$$

an exactly exponential distribution. Even for a finite cutoff $D_{\max} \gg D_0$, $G(x; t)$ is going to be close to exponential if $|x|$ is not too large.

We simulate the model with $d(D) = \text{const}$, $s(D) = \text{const}$ by a Monte Carlo procedure with $\Delta t = 1$. At each time step, the particle displacement is drawn from the Gaussian distribution (3) and the diffusivity change is drawn from the Gaussian distribution with variance $2d$

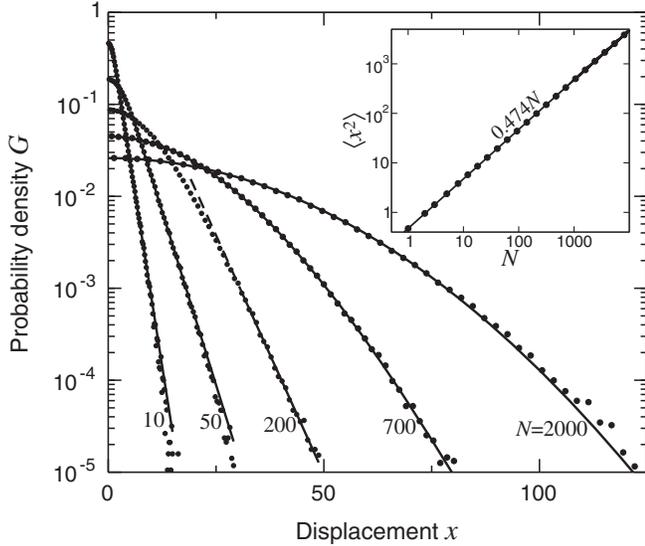


FIG. 1. The displacement distributions after different numbers of steps N for the diffusing diffusivity model [Eqs. (3), (6), and (7)] with $d = 0.0025$ and $s = 0.01$ simulated as described in the text. The solid line fits are exponential for the three smallest values of N , Gaussian for $N = 2000$, and the interpolating function $G(x) = A \exp(-B\sqrt{1 + (x/X_0)^2})$ for $N = 700$. The inset shows the MSD and a linear fit.

shifted by $-s$. Whenever D exits the interval from 0 to $D_{\max} = 1$ it is reflected back into this interval. We chose small values $d = 0.0025$ and $s = 0.01$ so the change of D during one step is likewise small. Drawing the initial diffusivities from the uniform distribution on $[0;1]$, we let them evolve for 1000 steps before taking displacement data, which is sufficient for convergence of the DifD towards that approximately given by Eq. (9). Figure 1 shows that exponential fits to the DispD tails at short t are very successful, despite the fact that $D_0 = 0.25$ only weakly satisfies the condition $D_0 \ll D_{\max} = 1$. Wang *et al.* [22] found the observation of anomalous yet Brownian diffusion on lipid tubules particularly surprising, since the ratio of the effective diffusivity in the system and the free-resolution diffusivity (about 1/5) was atypically large for systems with anomalous diffusion. Remarkably, the ratio of the mean diffusivity $\langle D \rangle \approx D_0$ to D_{\max} is even higher in our example.

While the above provides a simple model for anomalous yet Brownian diffusion, one may argue that $\pi(D)$ is unlikely to be exponential in all cases studied by Wang *et al.* [22,23]. We note, however, that the exponential fits to Wang *et al.* are not perfect. Therefore, showing that other types of $\pi(D)$ produce $G(x;t)$ with tails that appear exponential would go some way towards addressing this concern. For a particular $\pi(D)$, the integral (8) can be done numerically and the asymptotic behavior can also be estimated using the steepest-descent method [28]. For instance, for $\pi(D) \sim D^\alpha \exp(-D/D_0)$, there is a power-law prefactor in the form of the tail,

$G(x;t) \sim t^{-(\alpha+1)/2} |x|^\alpha \exp(-|x|/\sqrt{D_0 t})$. This prefactor is just a logarithmic correction on a semilogarithmic plot, and if $|\alpha|$ is small (say, below 2), fitting with an exponential over several decades in G is still adequate. When $\pi(D) \sim \exp[-(D/D_0)^\beta]$, that is, a stretched or compressed exponential, the tail of $G(x;t)$ is likewise a stretched or compressed exponential (up to a power-law prefactor) but with the exponent $\gamma = 2\beta/(\beta + 1)$, which is always closer to unity than β . For instance, for a Gaussian $\pi(D)$ ($\beta = 2$), $\gamma = 4/3$, and thus the tails are closer to being exponential than Gaussian. It is only for $\beta \rightarrow \infty$, when $\pi(D)$ approaches a step function, that $\gamma \rightarrow 2$ and the tails become Gaussian-like.

Another case of near-exponential tails of $G(x;t)$ is provided by a physically motivated variant of the diffusing diffusivity model with diffusivity changes coupled to particle displacement. Let us make the simplest assumption that the changes in the particle environment alone would lead to a constant diffusivity of the diffusivity *without bias*. As for the motion of the particle itself between different environments, it is logical to assume that the longer the particle step, the larger the typical diffusivity change during that step. With this in mind, consider the model in which for each step Δx_i the associated diffusivity change $\Delta D_i = D_{i+1} - D_i$ is Gaussian with half-variance

$$d = d_0 + f(\Delta x_i)^2, \quad (11)$$

where d_0 and f are constants. The first term reflects the random fluctuations of the local environment and the second term is due to the particle's moving between different environments. As the mean-square step size is itself proportional to the diffusivity ($\langle \Delta x_i^2 \rangle = 2D_i$), this is approximately equivalent to having $d(D) = d_0 + 2fD$ and $s(D) = 0$ in Eq. (6). For these $d(D)$ and $s(D)$ with $D_{\max} = 1$, it follows from the condition $J = 0$ with J given by Eq. (7) that

$$\pi(D) \approx \frac{2f}{\ln(1 + 2f/d_0)} \times \frac{1}{d_0 + 2fD}, \quad (12)$$

quite different from Eq. (9). Yet, the resulting $G(x;t)$ can still be fitted with exponentials at short times over a significant region, as shown in Fig. 2. This is despite the fact that the exponential is not an asymptotic solution at either small or large x . For Fig. 2 we have used $d_0 = 5 \times 10^{-5}$ and $f = 10^{-3}$, for which Eq. (12) gives $\langle D \rangle \approx 0.244$, about the same as in the first version of the model (as before, we let the diffusivities evolve, this time for 2000 steps, before collecting data). On the other hand, it is clear that for $\langle D \rangle$ even closer to D_{\max} , exponential fits will not be as good since the expected crossover to a Gaussian-like distribution at $\sim \sqrt{D_{\max} t}$ will be too close to the root-mean-square displacement $\sim \sqrt{\langle D \rangle t}$. We have checked, in particular, that this is indeed

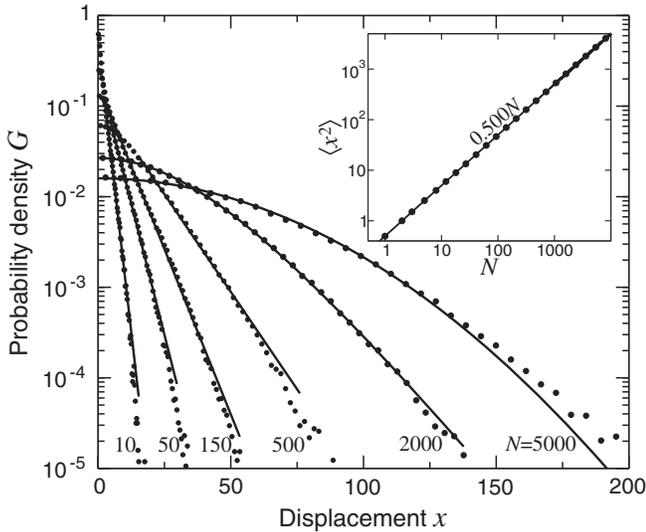


FIG. 2. The displacement distributions after different numbers of steps N for the diffusing diffusivity model with coupling to particle displacement with $d_0 = 5 \times 10^{-5}$ and $f = 10^{-3}$ in Eq. (11), simulated as described in the text. The solid line fits are exponential for the four smallest values of N , Gaussian for $N = 5000$, and the interpolating function $G(x) = A \exp(-B\sqrt{1 + (x/X_0)^2})$ for $N = 2000$. The inset shows the MSD and a linear fit.

the case when $\pi(D)$ is uniform on $[0; D_{\max}]$, with $\langle D \rangle = D_{\max}/2$. Therefore, the nonexponential DispDs seen in Ref. [29], where the observed diffusivity is $> 70\%$ of D_{\max} , are entirely expected. There may be other situations where exponential fits fail, for instance, bimodal DifDs [30]. An interesting situation was observed by Leptos *et al.* [31] who studied diffusion of passive tracers in suspensions of swimming microorganisms: since in addition to being moved by the flow created by the microorganisms the tracers undergo Brownian motion, the effective diffusivity has a nonzero lower bound, which gives rise to a combination of a Gaussian DispD at small $|x|$ and exponential tails at large $|x|$, even for small t .

Interestingly, the same general approach can be used to produce subdiffusion. Let $s(D) = 0$ and $d(D) \propto D^b$ in Eq. (7), where $b > 3$ is a constant. In this case, there is no stationary solution, except for the trivial $\pi(D) = \delta(D)$. Instead, there is a quasistationary solution of the form $\pi(D; t) = t^c f(Dt^c)$, with $c = 1/(b-2)$, that corresponds to ageing with $\langle D \rangle$ decreasing gradually to zero as t^{-c} and the anomalous diffusion exponent being $\nu = 1 - c = (b-3)/(b-2)$. Our simulations confirm this result (data not shown). We have also found that for $2 < b < 3$, $\langle D \rangle$ decays faster than t^{-1} , and the MSD instead approaches a constant—the particle remains trapped forever. We note that Massignan *et al.* [32] have recently published a similar model of subdiffusion, likewise assuming a power-law $\pi(D)$; the difference is that D is piecewise constant as a function of time in their model.

To summarize, we have proposed a class of one-dimensional models of anomalous yet Brownian diffusion. Recognizing the fact that the linear MSD combined with a non-Gaussian DispD should be observed for random walks without direction memory, but with diffusivity memory, we have assumed that the instantaneous effective diffusion coefficient of a particle changes gradually at random, or diffuses (with or without bias)—hence our expression “diffusing diffusivity.” Physically, this corresponds to the environment of the particle changing slowly—either on its own or because the particle moves to a different environment. Yet another possibility is a particle slowly changing its own properties (e.g., a protein changing its conformation). The short-time DispD is determined solely by the stationary DifD. This DispD is exactly exponential when the DifD is exponential, but an exponential remains a good fit to a significant part of the tail of the DispD for a wide variety of DifDs, which may explain the available experimental results. The same approach can produce subdiffusion, which may provide another possible route to subdiffusion in addition to the CTRW, the fBm, and obstructed diffusion [33], although the peculiar power-law dependence of the diffusivity of the diffusivity required needs to be justified on physical grounds.

Just like the CTRW, our approach is mean-field since random diffusivity changes neglect the possibility of returning to the same environment. This is a better approximation in higher dimensions and for environments changing rapidly on their own. To what extent the conclusions are modified, in particular, in the least favorable case of a static diffusivity distribution in one dimension will be a subject of future studies. Also, while we have assumed a gradual change of the diffusivity, we expect the results to be applicable qualitatively to cases with sharp boundaries between regions of different diffusivities, as in [32]. Again, this will be tested in the future.

While our model is very generic, we do not claim that it can account for all possible cases of anomalous yet Brownian diffusion. For instance, if in the CTRW model of anomalous diffusion the waiting time distribution is modified by introducing a finite cutoff, then the MSD measured after equilibrating for much longer than the cutoff time will be perfectly linear [34,35], and our calculations show that exponential tails in the DispD are possible. While there is some similarity to diffusing diffusivity (waiting periods can be interpreted as periods of low diffusivity that changes slowly), it is a distinct model [for instance, it is not obvious how to apply Eq. (8)] that deserves further study. We believe, however, that our model fits better at least some of the observed cases of anomalous yet Brownian diffusion, for example, the diffusion on a lipid tube where a relatively high global diffusion coefficient makes long trapping characteristic of CTRW unlikely and an additional peak at zero in the DispD corresponding to particles that have not moved yet is absent. Another model producing exponential

tails has been discussed by Chaudhuri *et al.* [36], although in the case in which the MSD would be linear ($\alpha = 1$, $\ell = 0$ in the authors' notation) the exponential tails are not particularly prominent.

Finally, we note the similarity of our model to some models of market price fluctuations (see, e.g., [37]), although the details differ.

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