Packets of Diffusing Particles Exhibit Universal Exponential Tails

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Brownian motion is a Gaussian process described by the central limit theorem. However, exponential decays of the positional probability density function $P(X,t)$ of packets of spreading random walkers, were observed in numerous situations that include glasses, live cells, and bacteria suspensions. We show that such exponential behavior is generally valid in a large class of problems of transport in random media. By extending the large deviations approach for a continuous time random walk, we uncover a general universal behavior for the decay of the density. It is found that fluctuations in the number of steps of the random walker, performed at finite time, lead to exponential decay (with logarithmic corrections) of $P(X,t)$. This universal behavior also holds for short times, a fact that makes experimental observations readily achievable.

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The emergence of normal Gaussian statistics for various random observables in nature is widespread. Examples range from chest sizes of Scottish soldiers [1] to Brownian motion [2]. This "popularity" is attributed to the central limit theorem (CLT). The statement of the CLT is that, at its center, the distribution of a sum of independent, identically distributed, random variables is Gaussian. However, recently, striking deviations from Gaussian behavior were recorded in a large number of experiments tracking spatial diffusion of tracer particles in various media. Interestingly, in many measurements, the observed probability density function (PDF), $P(X,t)$, attains an exponential (or close to exponential) decay. Examples include colloidal suspensions [3–5], nanoparticles in polymer solutions [6], molecular motion on a solid-liquid interface [7,8], living cells [9], phospholipid fluid tubules and biofilament networks [10,11], active gels [12], financial markets [13], colloidal glasses [14,15], worms [16], and suspensions of swimming microorganisms [17,18] (more examples are provided in [10,19]).

Appearance of a few jumps or excursions that dominate the process is a common feature in a portion of these experiments. For a bead in a bacteria suspension, such "jumps" were attributed to temporal adhesion of the particle to a bacteria flow that establishes short-term motion alongside the bacteria [18]. In the F-actin random network, the displacements are myosin driven excursions [12], while for Lennard-Jones suspensions, there are "cage-breaking" events [3]. The later system is one of four different (experimental and numerical) systems analyzed by Chaudhuri *et al.* [14]. In this work, the authors noticed that particle displacement measured in: dense suspension of colloidal hard spheres, slowly driven dense granular assembly, silica melt, and a binary Lennard-Jones mixture, all showing the same universal feature of exponential decay for $P(X,t)$ of the traced particles. Moreover, the authors used a special variant of the continuous time random walk with two exponential PDFs for waiting times and two Gaussian distributions for the sizes of the jumps in order to reproduce the observed behavior. These findings lead to questions regrading the universality of exponential decay. How can a basic random walk approach give rise to a theory that produces universal exponential decay for systems with various distributions for jump sizes and waiting times? Is there a large class of processes that attains such universality, and if so, what is the precise mathematical description of the mentioned exponential decay? We wish to develop this theory and identify such a broad class of processes by focusing on the role of the randomness of the number of jumps of a particle in an experiment.

In this Letter, we propose to reconcile observed non-Gaussianity by explicitly invoking the continuous time random walk (CTRW) formalism [20], but without restricting ourselves to specific examples. Specifically, we extend the approach of large deviations to both space and time. The random number of measured jumps naturally occurs in CTRW due to random waiting times between the jumps. While, for a constant number of jumps, the large deviations approach fails to produce universal behavior (as we show below), the situation with a random number of jumps that we treat here is quite different. We develop a subordination approach for large deviations and show that, for any process that involves a random number of jumps and can be modeled by CTRW, a very general statement holds: the exponential tails for the positional PDF are rather a rule and not an exception. The exponential decay of the tails is a general feature exactly like the Gaussian behavior (that is dictated by CLT) at the center.

First, let us stress out why, mathematically speaking, the numerously observed exponential decay is unexpected from the stand point of a regular random walk and standard large deviation approach [21–26]. The random walk definition is as follows, at each step, a particle can perform a step of size x, while the PDF of x is given by $f(x)$. After N steps, the position X is simply the sum of all random and independent steps $X = \sum_{i=1}^{N} x_i$. We will concentrate on the case when $f(x)$ is symmetric and decays as $f(x) \sim \exp[-(|x|/\delta)^{\beta}]$ when $|x| \to \infty$, $(\beta > 1)$. Namely, here, we exclude the power law decay of $f(x)$, in particular, we are in the domain of attraction of the Gaussian CLT. Indeed, the first moment $\langle x \rangle$ is 0 due to symmetry and the second moment, $\langle x^2 \rangle = \int_{-\infty}^{\infty} x^2 f(x) dx$, is finite. Thus, according to CLT, when $|X|/N$ is not large, the PDF to find the particle at position X after N steps, i.e., $\mathcal{P}_N(X) \sim \exp\{-N[(1/\sqrt{2\langle x^2 \rangle})(|X|/N)]^2\}$. All the different properties of $f(x)$ enter solely via $\langle x^2 \rangle$, the functional Gaussian form is universal. The situation for the tails is quite different. According to the theory of large deviations and, more specifically, Cramér's Theorem [21], for large $N \to \infty$, $\mathcal{P}_N(X) \sim \exp[-NI(|X|/N)]$ where the rate function $I(a) = \sup_{\Omega}\{\Omega a - \log[\langle \exp(\Omega x) \rangle]\}$. For small $|X|/N$, this leads to the quadratic form of $I(|X|/N)$ and the already mentioned Gaussian behavior of $\mathcal{P}_N(X)$ at the center. But for the tails, a straight-forward result of this theorem is that, when $|X|/N \to \infty$, the rate function takes the form $I(a) \sim a^{\beta}$ and

$$\mathcal{P}_N(X) \underset{N\to\infty}{\sim} \exp\left(-N\left[\frac{1}{\delta}\frac{|X|}{N}\right]^{\beta}\right) \quad \frac{|X|}{N} \to \infty. \tag{1}$$

Indeed, if $\beta = 2$, $\mathcal{P}_N(X)$ is Gaussian, and hence, exponential tails are not present. From Eq. (1), it becomes quite obvious that the functional form of the decay is β dependent and nonuniversal. It is natural to expect that the decay of the tails is very specific. This is why, based on the regular random walk perspective, the large number of different experiments that show very similar functional decay (i.e., exponential) is definitely unexpected. Unless one assumes intrinsic exponential distribution for the jumps of the particle ($\beta = 1$) in all the different experiments, which is unlikely.

We already mentioned our intention to resolve this issue by using the fact that the number of steps in most experiments is random for any finite measurement time t. Probably the simplest assumption is that the particle will wait for some random time τ between successive steps. This assumption is exactly the framework of CTRW, and it leads to the randomization of N [27]. The CTRW is a widely applicable model for transport in disordered media [27,28] that describes a particle that performs random independent steps x, determined by the PDF $f(x)$, and between two successive steps, the particle waits a random time τ that is distributed according to $\psi(\tau)$. All the waiting times are independent. The probability of observing N steps at time t, $Q_t(N)$, is fully determined by $\psi(\tau)$ (see below). For CTRW, the position $X = \sum_{i=1}^{N} x_i$ depends both on the random $\{x_i\}$s and the random N. By conditioning on the specific outcomes of N steps, the PDF to find the particle at X at time t is

$$P(X,t) = \sum_{N=0}^{\infty} \mathcal{P}_N(X) Q_t(N). \tag{2}$$

Equation (2) is also known as the subordination of the spatial process for X by the temporal process for N [27, 29–32]. The regular approach for CTRW without anomalously large jumps [28] is to replace $\mathcal{P}_N(X)$ in Eq. (2) by the Gaussian approximation. From Eq. (1), it is clear that, for the tails, the Gaussian approximation is simply incorrect, unless $\beta = 2$. In order to accomplish the calculation of $P(X,t)$ for large $|X|$, the first thing to do is to insert the form of $\mathcal{P}_N(X)$ in Eq. (1) into Eq. (2), instead of the Gaussian approximation, i.e., subordination of large deviations. In Eq. (2), one notices that, for large $|X|$, the form of large deviations for $\mathcal{P}_N(X)$ [i.e., Eq. (1)] states that all the small N contributions of $\mathcal{P}_N(X)$ are negligible, as compared to large N. So the sum in Eq. (2), with Eq. (1) for $\mathcal{P}_N(X)$, is affected only by large N values of $Q_t(N)$ when $|X|$ is large [see Supplemental Material [33] (SM)]. Indeed, for any fixed t, the position $|X|$ can be chosen arbitrarily large in order to suppress all the contributions of $\mathcal{P}_N(X)Q_t(N)$, for any finite N. Thus, it is crucial to obtain the large N behavior of $Q_t(N)$.

$Q_t(N)$ in the $N \to \infty$ limit is the probability of occurrence of a rare event, i.e., a large number of steps, in finite time. The distribution of the dwell time τ between two steps, $\psi(\tau)$, is independent of previous or following waiting times. The probability $Q_t(N)$ is the probability that $\sum_{i=1}^{N} \tau_i < t$ while $\sum_{i=1}^{N+1} \tau_i > t$, where $\{\tau_i\}$ are the waiting times. Because of the convolution property of Laplace transform, $\hat{Q}_s(N) = \int_0^{\infty} \exp(-st) Q_t(N) dt$ is [37,38]

$$\hat{Q}_s(N) = \hat{\psi}(s)^N [1 - \hat{\psi}(s)]/s, \tag{3}$$

where $\hat{\psi}(s) = \int_0^{\infty} \psi(t) \exp(-st) dt$. It is assumed that the short time $(\tau \to 0)$ Taylor expansion of $\psi(\tau)$ is

$$\psi(\tau) \underset{\tau\to 0}{\sim} \sum_{j=0}^{\infty} C_{A+j} \tau^{A+j}, \tag{4}$$

where $A \geq 0$ is an integer. This is a very natural assumption, as it merely demands that $\psi(\tau)$ will be analytic at the vicinity of $\tau = 0$. In the SM, we use a power series expansion of $\hat{Q}_s(N)$ to show that the leading term of $Q_t(N)$ in the large N limit is

$$Q_t(N) \underset{N\to\infty}{\sim} \frac{\{[C_A \Gamma(A+1)]^{1/(A+1)} t\}^{N(A+1)}}{\Gamma[N(A+1)+1]} e^{(C_{A+1}/C_A)t}, \tag{5}$$

see Fig. 1. $Q_t(N)$ attains the form of the large deviation principle, $Q_t(N) \sim \exp[-NI_T(t/N)]$ (see SM). This general result holds for any $\psi(\tau)$, while t is kept constant and $N \to \infty$. It is not affected by the large τ behavior of $\psi(\tau)$ and includes situations when $\langle\tau\rangle \to \infty$, i.e., anomalous diffusion [27,28]. For the case when $\psi(\tau)$ is exponential, $Q_t(N)$ is a Poisson distribution and Eq. (5) agrees perfectly with this fact. If $A = 0$, namely, $\lim_{\tau\to 0}\psi(\tau) = C_0$, then C_0 is acting like an effective rate, similar to $1/\langle\tau\rangle$ for the Poisson distribution.

Supplemented with the general result for $Q_t(N)$ and using Eq. (1) for $\mathcal{P}_N(X)$, we finally obtain the tail behavior of $P(X,t)$. We plug Eq. (1) and Eq. (5) into Eq. (2), approximate the sum by an integral over N, and obtain

$$P(X,t) \sim \int_0^\infty \exp\Bigg(\overbrace{-N\Bigg\{\left[\frac{1}{\delta}\frac{|X|}{N}\right]^\beta - \frac{C_{A+1}}{C_A}\frac{t}{N} - (A+1)\left[\log\left\{\frac{[C_A\Gamma(A+1)]^{1/(A+1)}}{A+1}\frac{t}{N}\right\}+1\right]\Bigg\}}^{K(N)}\Bigg)dN. \quad (6)$$

Clearly, this represents a subordination of the large deviations result, i.e., Cramér's theorem with the just obtained universal $Q_t(N)$. Now, we use the saddle point approximation in order to calculate the integral for $|X| \to \infty$. We find that the maximum of $K(N)$ is achieved for

$$N^* = |X| g_0 W_0\left[\left(g_1\frac{|X|}{t}\right)^\beta\right]^{-1/\beta}, \quad (7)$$

where $g_0 = [\beta(\beta-1)/(A+1)]^{1/\beta}/\delta$, $g_1 = \{g_0(A+1)/[C_A\Gamma(A+1)]^{1/(A+1)}\}$ and $W_0(y)$ is the principal branch of a Lambert W function [39–42], i.e., a solution of the equation $W(y)\exp[W(y)] = y$. Therefore, the asymptotic behavior of $P(X,t)$ in the $|X| \to \infty$ limit is provided by

$$P(X,t) \underset{|X|\to\infty}{\sim} \frac{\exp\{-t[\frac{|X|}{t}Z(\frac{|X|}{t}) + C]\}}{\sqrt{K''(N^*)}}, \quad (8)$$

where

$$Z(y) = \frac{(\frac{g_0(A+1)}{\beta} + \frac{1}{g_0^{\beta-1}\delta^\beta})W_0[g_1 y^\beta] - g_0(A+1)}{W_0[g_1 y^\beta]^{\frac{1}{\beta}}}, \quad (9)$$

and $C = -C_{A+1}/C_A$. The function $W_0(y)$ $(y \geq -1/e)$ is a monotonically increasing function with sublogarithmic slow growth, $\log(y) - \log[\log(y)] \leq W_0(y) \leq \log(y) - \frac{1}{2}\log[\log(y)]$ for $e \leq y$ [43]. The asymptotic expansion for $y \to \infty$ is $W_0(y) \sim \log(y) - \log[\log(y)]$ and in the limit $|X|/t \to \infty$, Eq. (8) obtains the form

$$P(X,t) \underset{|X|/t\to\infty}{\sim} \exp\left\{-t\left[\kappa\log\left(\frac{|X|}{t}\right)^{1-1/\beta}\frac{|X|}{t} + C\right]\right\}, \quad (10)$$

and $\kappa = [g_0(A+1)/\beta + 1/g_0^{\beta-1}\delta^\beta]\beta^{1-1/\beta}$. This result states that the tails of $P(X,t)$ will exhibit almost exponential decay. The logarithmic corrections, due to slow sublogarithmic growth of $Z(\ldots)$ in Eq. (8), will cause small deviations from pure exponential behavior, and overall, it would seem that $yZ(y) + C$ converges to linear form. In Fig. 2, this (approximately) exponential behavior of $P(X,t)$ is displayed for two different pairs of $f(x)$ and $\psi(\tau)$. An argument that explains the appearance of exponential decay is related to the observation that N^* is proportional to $|X|$ (neglecting the logarithmic corrections). According to the saddle point approximation, N^* is the dominating N for $P(X,t)$ (an exactly solvable example is provided below). Hence, replacing N with N^* in Eq. (1), we obtain a universal exponential decay. Namely, the β dependence disappears since $(|X|/N^*)^\beta$ is constant. As we already mentioned, in the case of $P(X,t)$, t is not limited to the domain of large values. If t can take small enough values, while keeping the values of $Z(|X|/t)|X|/t + C$ not too large, the exponential behavior can be readily observed in an experimental situation (see below).

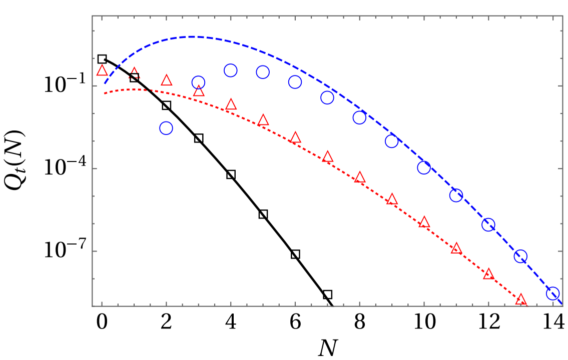

FIG. 1. Numerical simulations (symbols) of $Q_t(N)$ are compared to Eq. (5) (lines) for three different $\psi(\tau)$s. Square is the Half Gaussian Distribution $\psi(\tau) = (2/5\pi)\exp(-\tau^2/25\pi)$, measurement time is $t = 1.5$. Circle is a special form of Beta distribution $\psi(\tau) = 6\tau(1-\tau)$, $0 \leq \tau \leq 1$ $(t = 1.5)$, and Triangle is the Dagum distribution $\psi(\tau) = 1/(1+\tau)^2$, while measurement time $t = 2.5$.

This is why, in any system where the CTRW description is applicable, the exponential decay is expected. The CTRW framework requires the existence of local trapping (or suppressed motion) and relatively fast spatial transitions, i.e., jumps, between the trapping regions. Specifically relevant are the glassy systems [44], where the CTRW

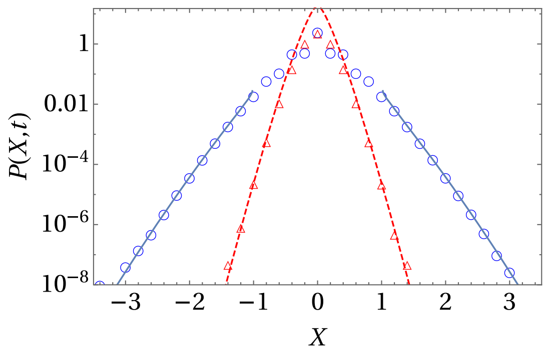

FIG. 2. Universality of exponential tails. Comparison of $P(X,t)$ obtained from CTRW simulations (symbols) and theoretical prediction, Eq. (8), without fitting (see SM). Circle displays CTRW where $f(x)$ is uniform between −0.5 and 0.5 (and zero everywhere else), $\psi(\tau) = 1/(1+\tau)^2$, $t = 1.5$, and the theory is the thick line. Triangle is for $f(x) = \exp(-50x^2)/\sqrt{\pi/50}$, $\psi(\tau)$ is uniform between 0 and 1 (and zero everywhere else), $t = 1.5$, and the theory is the dashed line. Notice the log-linear coordinates, indicating that $P(X,t)$ is exponential as in many experiments mentioned in the introduction.

approach [14,44,45], proved to be useful. The dynamics of single glass formers, e.g., colloids, often presents itself in the form of fast cage-breaking events [3], while between the fast events the colloid is trapped by its neighbors. In the case of molecular motion on a solid-liquid surface [7,8], the molecules are switching between periods of immobilization (trapping) on the surface and fast displacements (jumps) produced by excursions through the liquid bulk. The motion of a bead in a suspension of eukaryotic microswimmers is composed of periods of diffusion in the liquid media (suppressed motion) and short periods of fast and extensive motion (jumps) due to entrainment by nearby swimming microswimmers [17,18]. Another example is the transport of charged carriers in amorphous semiconductors where the charges tend to get trapped by imperfections of the surrounding media and the motion is a combination of periods of trapping and fast transitions (i.e., jumps) between those traps [46]. Experimental and theoretical studies of motion of molecular motors on top of cytoskeletal networks also show that, for this case, the motion is described by trapping at points of high concentration of filaments and relatively fast transitions between such regions [47,48]. The randomness in the number of such N jumps is what makes it an important factor behind the observed universal exponential decay of $P(X,t)$, and the convergence to exponential behavior occurs even on time scales when the average number of jumps is small. For example, in glassy systems [14], the number of cage breaking events was small. As discussed below, when observation time is of the order of the typical waiting time, the exponential tails are readily achieved, while when the measurement time is much longer, the effect is found in far tails of $P(X,t)$. We must note that the exponential tails were also observed for systems with memory, i.e., fractional Brownian motion [49], so there is a possibility that the CTRW universality class can be further extended.

When the exponential decay of the tails of $P(X,t)$ is compared to Gaussian behavior at the center, it is important to stress out the different timescales when these two behaviors will take place. The Gaussian behavior will appear only when the measurement time is sufficiently long, while the exponential decay will take place for any time (as we already mentioned). Both features are based on statistics of large numbers of events. While for the center of $P(X,t)$, large numbers of events are sampled only for long enough time, the tails that describe the rare events are, by themselves, a manifestation of appearance of a large group of events. Then, it is expected that, in an experimental situation, the exponential decay will show itself long before the convergence to Gaussian behavior will appear [11].

As previously mentioned, the exponential behavior is expected to appear when N^* becomes proportional to $|X|$ [Eq. (7)]. Let us further investigate this for the case of exponential $\psi(\tau) = \exp(-\tau)/\langle\tau\rangle$ and Gaussian $f(x) = \exp(-x^2/2\sigma^2)/\sqrt{2\pi\sigma^2}$ PDFs. For this case, the exact solution for $|X| \neq 0$ is

$$P(X,t) = \sum_{N=1}^{\infty} \frac{(t/\langle\tau\rangle)^N e^{-t/\langle\tau\rangle}}{N!} \frac{e^{-X^2/2N\sigma^2}}{\sqrt{2\pi N\sigma^2}}, \tag{11}$$

For fixed X and t, we search for the N that gives the largest term in the sum. In the inset of Fig. 3, we plot this N^* as a function of X and find a nearly linear behavior with X.

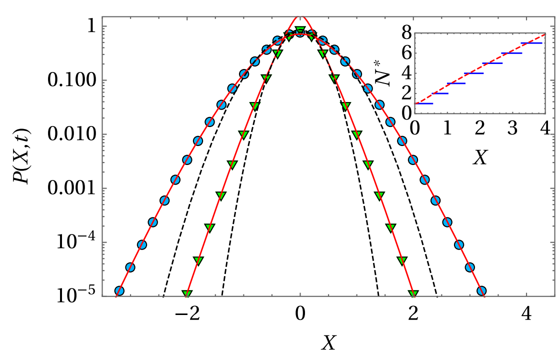

FIG. 3. Appearance of exponential tails for the case of $\psi(\tau) = \exp(-\tau)$, $f(x) = \exp(-x^2/2\sigma^2)/\sqrt{2\pi\sigma^2}$, and $\sigma = 0.25$. Inverted triangle are simulations for $t = 1$ and circle are for $t = 5$. The dashed lines are Gaussian approximations at the center while the thick lines are predictions due to Eq. (8). The deviations from the Gaussian start for $|X| > t$ and the trajectories with $N > t$ contribute to the tail behavior, where the average number of jumps per trajectory is t and $N > \langle N\rangle$. The inset describes the N that gives rise to the maximal term in the sum $\sum_{N=0}^{\infty} \mathcal{P}_N(X) Q_t(N)$ for $t = 1$. Because of the discrete nature of N^* the growth appears in a steplike fashion. The dashed line is the behavior of N^* according to Eq. (7) and is approximately linear.

The result agrees perfectly with Eq. (7). In this example, N^* is a linear function of $|X|$ when $|X|/\sigma > t/\langle\tau\rangle$ and trajectories with $N > t/\langle\tau\rangle$ contribute to the exponential regime (see SM for more details). The average numbers of jumps that are used in Fig. 3 are 1 and 5. In several experiments [14], where exponential decay is evident, $\langle N\rangle$ was also recorded to be small. For long measurement times, Eq. (8) also holds, and the exponential tails are simply pushed towards really small values of $P(X,t)$, i.e., far from the center.

We presented a space-time theory for large deviations of the widely applicable continuous time random walk. This theory provides an explanation for a large class of recent experimental observations of diffusion processes. In this sense, the reported universal behavior is likely to establish a link between experiments and the theory of large deviations. The large deviation principle for space, $\mathcal{P}_N(X) \sim \exp[-N I_S(X/N)]$ Eq. (1), and for time $Q_t(N) \sim \exp[-N I_T(t/N)]$ Eq. (5), were described by rate functions $I_S(\dots)$ and $I_T(\dots)$, respectively. The subordination approach yielded our main result, Eq. (8), where the rate function $(|X|/t)Z(|X|/t) + C$ controls the decay of the PDF. However, it is remarkable that our theory works for any t. This stems from the fact that, once X is large, a large number of jumps is needed to arrive at this position. When the number of jumps is fixed, as in a standard random walk, the widely observed universal decay is completely missed. In the mathematical literature, the CTRW is known as the renewal-reward process. The large deviation behavior of the renewal-reward process was studied in several works [50–52] but in the $t\to\infty$ limit. Deviations from the presented theory are expected when $\psi(\tau)$ is nonanalytic in the vicinity of $\tau = 0$ or when the decay of $f(x)$ is broad, e.g., power law.

It is also worth mentioning that the CTRW formalism will present a Fickian diffusion, i.e., linear growth of the mean squared distance with time, as long as $\langle\tau\rangle$ is finite [53]. This means that the presented broad class of models investigated here will also show the widely investigated Fickian yet non-Gaussian behavior [10,11,19,54–56]. The presented results can be generalized to the case of asymmetric $f(x)$, where we expect to observe asymmetrical exponential decay. Finally, we notice that the presented results are expected to have a high impact on the field of triggered reactions in physics, chemistry, and biology [57–59]. In any situation where a reaction occurs as a result of a first arrival, the universal rare behavior described here will dominate due to the simple fact that exponential decay is significantly slower than the Gaussian case of simple diffusion. This has crucial consequences for transport in such systems as the living cell [59,60].

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