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Rapid Communications

Brownian diffusion in a dilute field of traps is Fickean but non-Gaussian

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Motivated by experimental observations, we look for the diffusion of Brownian particles in a medium where they can be either trapped in randomly disposed deep traps or where they diffuse by the regular Fick's law outside of the traps. This process can be represented by two coupled equations—one valid inside the traps and another one outside —yielding the probability distribution of the distance run as a function of time. This probability depends on a unique dimensionless parameter λ which is proportional to the product of the (small) density of the traps times the long time of staying in the traps. The mean-square displacement is proportional to the lag time, and for finite or large λ the probability density is no longer Gaussian but exponential on intermediate time scales.

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This work is inspired in part by recent observations of Brownian motion in complex media (colloidal beads on the surface of phospholipid bilayer tubes [1,2] or in entangled actin gels [1], or fluorescent nanoparticles in hard-sphere colloidal suspensions [3]) where the mean-square distance grows linearly with time, as in regular Brownian motion, but where the probability distribution of this distance is markedly non-Gaussian, which differs from the standard theoretical results. Related behaviors have been observed in numerical simulations of particle diffusion in a two-dimensional colloidal system [4] or in diffusion among swimming cells [5]. Any theory of this Fickean but anomalous diffusion will have to introduce a wide range of timescales for the interaction between the diffusing particle and the surrounding medium. This implies that there is a timescale that is long when compared to the standard time of interaction of the particle with most fluid particles. A prime candidate for a long timescale is the trapping of particles in deep potential wells [6–9]. In this Rapid Communication, we put it in the framework of a statistical theory giving the long-term effects of such trapping events on Brownian motion. We study the diffusion of a Brownian particle in a medium where this particle has regular Brownian dynamics, except when it falls in traps disposed randomly but with a low density (Fig. 1). Traps may nevertheless play an important role in the diffusion process because their low density is somehow balanced by a long trapping time. The role of traps is embedded mathematically in a dimensionless parameter called λ, which may be either small or large. The results of our analysis depend in a nontrivial way on the magnitude of λ, and in some range of its values the statistics of the distances run during a fixed amount of time is exponential-like, as observed in experiments [1–3].

We consider dilute and steady traps and we develop a theoretical approach relying on the Langevin equation [10], in which we introduce (i) the physical parameters describing the interaction of a single trap with a diffusing particle and (ii) the nonuniformity in the space of the structure where the particle diffuses. Owing to this we can relate the observed properties with the physical characteristics of the medium where the particles diffuse. Within these assumptions the dynamics of a trap itself is not relevant even if the existence of traps fluctuating in time is consistent with our approach, because fluctuations in the trap properties should most likely lead to Poissonian statistics of the trapping times, although other possibilities may exist, depending on the structure of the potential.

A major constraint on theories of Brownian motion at equilibrium is time-reversal symmetry [11]. It is satisfied by the classical Langevin equation because of its linearity which, as is well known, leads to Gaussian statistics for the fluctuations. Therefore one must introduce nonlinearity somewhere in the equations to find non-Gaussian statistics. Even though the Langevin equation in its original form [12] is linear, it is possible to make it nonlinear without violating this constraint of reversibility [13,14]. For simplicity, we assume that the surrounding fluid is viscous enough so that the inertia term in the Langevin equation is negligible. Let us add to the right-hand side of the overdamped Langevin equation a force equal to minus the gradient $\nabla\Phi$ of a given potential $\Phi(\mathbf{r})$, where $\mathbf{r}$ is the position of the Brownian particle, with vectors in bold,

$$\zeta\frac{d\mathbf{r}}{dt} = -\nabla\Phi + \mathbf{X}(t), \tag{1}$$

where ζ is the Stokes friction coefficient and $\mathbf{X}(t)$ is the Langevin Gaussian white noise. This is a delta correlated function of time with zero average whose pair correlation is $\langle X_i(t)X_j(t')\rangle = 2\zeta\delta_{ij}\delta(t-t')k_BT$, where i and j stand for the Cartesian indices, δ_{ij} is the Kronecker delta, $\delta(t-t')$ is the Dirac delta, k_B is the Boltzmann constant, and T the temperature. Our theory is based on the Langevin equation (1) with a potential $\Phi(\mathbf{r})$, consistent with the constraint of reversibility. The complex surrounding the Brownian particle

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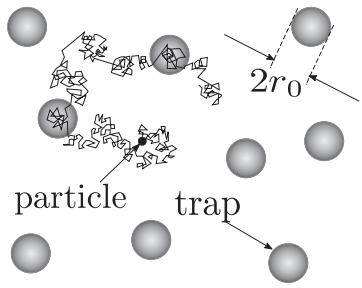

FIG. 1. Sketch of a Brownian particle (represented by the small black disk) in a medium containing spherical traps (represented in gray) of radius r_0.

is represented by a "landscape" of the potential $\Phi(\mathbf{r})$ with randomly distributed wells (or traps) of depth $(-w_0)$, w_0 positive and larger than $k_B T$. We shall assume identical traps all with the same long trapping time, $\Phi(\mathbf{r}) = -\sum_i w(\mathbf{r} - \mathbf{r_i})$, where $-w(\mathbf{r})$ is the potential of a unique trap and $\mathbf{r_i}$ are the random positions of the traps. For simplicity, w is assumed to be isotropic, hence $w(\mathbf{r}) = w(r)$ with r the radial distance.

In previous works [2,15–18], the probability density function of the displacement was assumed to result from a finite distribution of the diffusivity. This distribution was established by considering time-dependent but *spatially homogeneous* diffusivity fluctuations, whereas experiments correspond to diffusion in complex media where the fluctuations (e.g., in density) are also in the space dependence of the physical parameters. Indeed, the dynamics of these fluctuations is driven by short-range interactions, and the involved timescales cannot be neglected with respect to the characteristic times of the diffusion of a particle. Therefore, fluctuations generate spatial heterogeneities in the surrounding material of Brownian particles. In the absence of laws governing the time evolution of a spatially homogeneous diffusivity that are entirely based on physical principles, the authors have found an exponential decay of the probability function as observed in experiments [1–3] by choosing *ad hoc* rules of the random process for the (spatially homogeneous) time-dependent fluctuations of the diffusion coefficient (for instance, in Ref. [17] the diffusivity is chosen to fit with the square of the solution of an Ornstein-Uhlenbeck process so that the decay of the probability function is exponential).

In contrast with these previous models and following Ref. [19], we address here the diffusion of Brownian particles in a *spatially heterogeneous* medium, as expected in the experimental systems. Contrary to other approaches [20,21], our model is free from any makeshift distribution function, and relies only on basic physical laws.

Here, the disorder originates from the random distribution in the traps' location. We consider a unique depth of the traps, and we show that it is enough to predict an exponential decay of the probability density function with the advantage of starting from a unique and clear microscopic mechanism. Of course, our approach can be extended afterwards to more complex situations, e.g., a given distribution of potential depths in order to account for a more complex environment.

Let r_0 be the radius of the traps and ν be their number density. The product νr_0^3 is small by assumption. A diffusing particle can either be trapped, or can freely diffuse over finite distances. We shall here introduce two densities for the diffusing particles (although we have in mind only one particle, it is easier to formulate the problem for a large number of diffusing and noninteracting identical Brownian particles). Let $n_{\rm tr}(\mathbf{r}, t)$ be the density inside the traps: This is the total number of trapped particles divided by the volume of traps in a small region of space but large enough to include many traps. The density $n_u(\mathbf{r}, t)$ is the density outside of the traps: This is the number of untrapped particles divided by the total element of volume they are in, which can be taken as the total volume because the volume of the traps is relatively small. Attention should be paid to the fact that $n_{\rm tr}(\mathbf{r}, t)$ is measured inside traps only: Even if it is large, the density of trapped particles averaged over the whole space, inside or outside the traps, can be very small if there are only very few traps in the system. The number density in the whole space of trapped particles is $\frac{4\pi}{3} n_{\rm tr}(\mathbf{r}, t)\nu r_0^3$. The density inside the traps changes under two effects: First, it decays by a rare escape due to thermal fluctuations, a process taking a long time called τ later. The other process is the refilling of traps by particles coming from the outside, with density $n_u(\mathbf{r}, t)$. Since the traps are fed from the outside by a diffusion process and the traps have a size r_0, the typical timescale for this rate of feeding is $\tau_0 = r_0^2/D$, *a priori* much shorter than τ. The dynamical equation for $n_{\rm tr}(t)$ reads

$$\frac{\partial n_{\rm tr}}{\partial t} = \frac{n_u}{\tau_0} - \frac{n_{\rm tr}}{\tau}. \tag{2}$$

This equation shows already that, at equilibrium [namely, for a steady solution of Eq. (2)] the density inside the traps is much larger, by a factor τ/τ_0 than outside, because of the long time spent there (later on, because of rescaling, the transformed densities will be made "equal" at equilibrium). Moreover, the validity of Eq. (2) requires that the density of the traps is small. Otherwise, correlations between the positions of the traps would create a memory of their location at different times. This would forbid us to consider, as done when writing Eq. (2), that the interaction of the diffusing particle with the traps is always with a trap not met before and located randomly, independent of the path followed by the diffusing particle.

Equation (2) is to be completed by an equation for the untrapped density $n_u(\mathbf{r}, t)$. This equation is of the same general form as Eq. (2). It is constrained by the principle that particles after escaping traps go outside, a way of telling that the total number of particles is conserved. The corresponding number density (trapped and untrapped particles) is $n(\mathbf{r}, t) = \frac{4\pi}{3} n_{\rm tr}(\mathbf{r}, t)\nu r_0^3 + n_u(\mathbf{r}, t)$. The only possibility for an equation of this type with this property is the standard Fick equation with added gain and loss terms balancing the right-hand side of Eq. (2) to guarantee conservation of the number of particles,

$$\frac{\partial n_u}{\partial t} = D\nabla^2 n_u + \frac{4\pi}{3}\nu r_0^3\left(\frac{n_{\rm tr}}{\tau} - \frac{n_u}{\tau_0}\right). \tag{3}$$

Equations (2) and (3) can be written in a dimensionless form by taking τ as the time unit, $(D\tau)^{1/2}$ as the length unit, and by writing $n_{\rm tr}(\mathbf{r}, t)$ as $\frac{\tau}{\tau_0} n_{\rm tr}(\mathbf{r}, t)$. This dimensionless form of

Eqs. (2) and (3) depends on the parameter $\lambda \equiv \frac{4\pi}{3}\frac{\nu r_0^3 \tau}{\tau_0}$,

$$\frac{\partial n_{\rm tr}}{\partial t} = n_u - n_{\rm tr}, \tag{4}$$

$$\frac{\partial n_u}{\partial t} = \nabla^2 n_u + \lambda(n_{\rm tr} - n_u). \tag{5}$$

The parameter λ takes into account two opposite effects: $\frac{4\pi}{3}\nu r_0^3$ is the small amount of space filled by the traps, whereas τ/τ_0 is the large ratio of the trapping time versus the typical time of free diffusion outside of the traps. This turns out to be also the ratio of the total number of particles inside the traps versus the ones outside of the traps at equilibrium. Therefore λ may be either large, finite, or small. Note that the time of escape out of a trap is well defined in three dimensions, but not in one or two dimensions because the return to the place visited on the journey of a Brownian particle then is certain. We shall not consider this kind of situation.

From equilibrium statistics the parameter λ is given by the integral of the Boltzmann statistical weight computed inside a trap times the density of traps. This yields

$$\lambda = \nu \int d\mathbf{r} e^{w(r)/k_B T} = \nu e^{w_0/k_B T} \int d\mathbf{r} e^{[w(r)-w_0]/k_B T}, \tag{6}$$

where w_0 is the maximum of $w(r)$, a smooth function of the radius r. This is readily estimated in the limit in which $w_0/k_B T$ is large by expanding $w(r)$ near its maximum at $r = 0$, assuming this is a quadratic maximum, such that for small r, $w(r) = w_0(1 - \frac{r^2}{r_0^2} + \cdots)$. This introduces explicitly the radius r_0 of the trap. We shall drop below the next order term. This amounts to taking as the potential $-w(r)$ the exact parabolic form for $0 < r < r_0$ and zero for $r_0 < r$, namely, outside of the trap. In the limit in which $w_0/k_B T$ is large one may estimate this integral by keeping the quadratic term only, to obtain

$$\lambda \approx \nu e^{w_0/k_B T} \int d\mathbf{r} e^{-\frac{w_0 r^2}{k_B T r_0^2}} = \nu r_0^3 e^{w_0/k_B T} \left(\frac{2\pi k_B T}{w_0}\right)^{3/2}. \tag{7}$$

This estimate of λ, even computed at equilibrium, is valid for the out-of-equilibrium situations we shall consider. This is because the relaxation to equilibrium involved there is very quick and depends on fast molecular relaxation inside the traps, whereas the out-of-equilibrium processes we shall consider have much longer timescales, of the order of the Kramers escape time, so that the ratio of population out of and in the traps has enough time to reach its equilibrium value.

Expressed in the Fourier-Laplace domain $(\mathbf{r}, t) \to (\mathbf{k}, z)$, the linear equations (4) and (5) yield

$$n_u(z, k) = \frac{n_u(t = 0, k)(z + 1) + \lambda n_{\rm tr}(t = 0, k)}{P_2(z)}, \tag{8}$$

$$n_{\rm tr}(z, k) = \frac{n_u(t = 0, k) + (z + \lambda + k^2) n_{\rm tr}(t = 0, k)}{P_2(z)}, \tag{9}$$

where k denotes the magnitude of $\mathbf{k}$ and $P_2(z) = (z + k^2 + \lambda)(z + 1) - \lambda$. In order to explicitly derive the probability density function $Q(\mathbf{r}, t)$ of the position of a unique diffusing particle, we consider an initial distribution of trapped and untrapped particles accordingly with the statistical distribution at equilibrium which writes, as already seen, $n_u(\mathbf{r}, t) = n_{tr}(\mathbf{r}, t)$ [see Eq. (4)]. The total density of particles, $n(\mathbf{r}, t) = n_u(\mathbf{r}, t) + \lambda n_{\rm tr}(\mathbf{r}, t)$, is normalized by imposing $\int d\mathbf{r} n(\mathbf{r}, t) = 1$ so that the probability density function is equal to the total density of particles, $Q(\mathbf{r}, t) = n(\mathbf{r}, t)$. With the equilibrium distribution written just above, the normalization yields

$$n_u(\mathbf{r}, t = 0) = n_{\rm tr}(\mathbf{r}, t = 0) = \frac{\delta(\mathbf{r})}{\lambda + 1}. \tag{10}$$

The Fourier-Laplace transform of the probability density function (or, equivalently, of the total density of particles) is obtained from Eqs. (8) and (9) with the initial condition Eq. (10),

$$Q(\mathbf{k}, z) = \frac{(1 + z + \lambda)(\lambda + 1) + \lambda k^2}{(\lambda + 1)[z\lambda + (z + 1)(k^2 + z)]}. \tag{11}$$

One gets the expression of the mean-square displacement in terms of the lag time of the Brownian particle by computing the inverse Laplace transform of the second derivative of $Q(\mathbf{k}, z)$ computed for $k = 0$,

$$\langle \Delta \mathbf{r}^2(t) \rangle = \frac{6Dt}{\lambda + 1}. \tag{12}$$

Since it is a linear function of the lag time, the diffusion process is Fickean with an apparent diffusivity equal to $D_{\rm eff} = D/(\lambda + 1)$.

Except in the limiting cases of large or short lag times (see Sec. C in the Supplemental Material (SM) [22]) or small λ (SM Sec. D [22]), the expression of $Q(\mathbf{k}, z)$ [Eq. (11)] does not match with the Fourier transform of a Gaussian distribution (SM Sec. E [22]): Hence, the diffusion of a Brownian particle in the dilute field of traps is Fickean but non-Gaussian.

We first look at the case of large λ, the one where the effects of the traps are the biggest, and we show that the Fickean diffusion is anomalous with an exponential tail of the probability density function. In this limit the Fourier-Laplace transform of the density probability function [Eq. (11)] simplifies in

$$Q(\mathbf{k}, z) \simeq \frac{1}{z + k^2/(k^2 + \lambda)}, \tag{13}$$

which is the Laplace transform of

$$Q(\mathbf{k}, t) = \exp\left(-\frac{k^2 t}{k^2 + \lambda}\right). \tag{14}$$

Developing the exponential in a Taylor expansion, one gets

$$Q(\mathbf{r}, t) = e^{-t}\delta(\mathbf{r}) + \frac{\lambda e^{-t} e^{-\sqrt{\lambda} r}}{2\pi^2 r} \sum_{n=1}^{\infty} \frac{t^n}{n!} P_n(\sqrt{\lambda} r), \tag{15}$$

where

$$P_n(r) = e^r \int_0^\infty dk \frac{k}{(k^2 + 1)^n} \sin(kr) \tag{16}$$

is a polynomial of degree $n - 1$. The first term in Eq. (15) corresponds to the particles trapped initially and remaining so. It is a delta function in space because we neglected until now the width of the traps compared to any other length scale. This is only approximate and the thermal fluctuations widen the probability distribution of the particles inside the traps, an effect considered in SM Sec. A [22]. $Q(\mathbf{r}, t)$ is plotted in Fig. 2 for $t = 0.1$, 1, and 2. One observes a significant

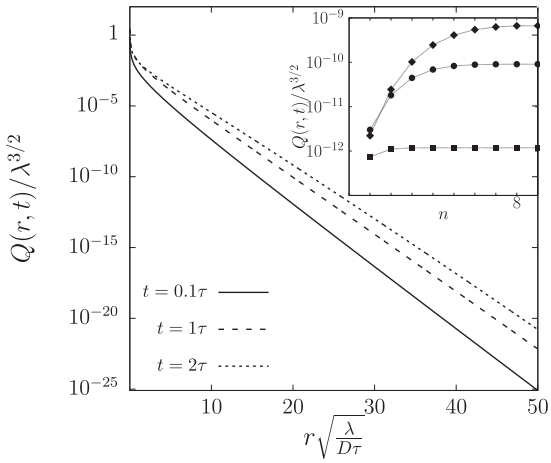

FIG. 2. Large λ limit. Probability density function normalized by $\lambda^{3/2}$ as a function of r for $t = 0.1\tau$, $t = \tau$, and $t = 2\tau$, calculated from Eq. (15) with the sum truncated beyond $n = 8$. No significant differences were observable if one had taken $n = 7$ of $n = 9$ instead. Inset: $Q(\mathbf{r}, t)/\lambda^{3/2}$ calculated from Eq. (15) with the sum truncated at different n, for $r = 20\sqrt{D\tau/\lambda}$, $t = 0.1\tau$ (squares), $t = \tau$ (circles), and $t = 2\tau$ (diamonds).

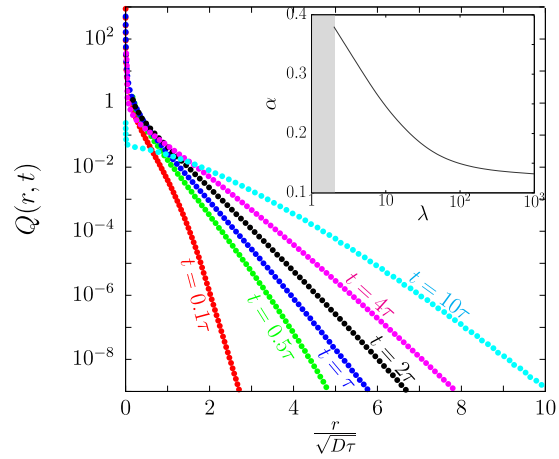

FIG. 3. Colored disks: Probability density function $Q(\mathbf{r}, t)$ as a function of $r/\sqrt{Dt}$, computed from the complete expression (11) for $\lambda = 10$ at various diffusion times t. Inset: Exponent α in the time variation of the characteristic length ℓ appearing in the exponential-like decay of the probability density function $Q(\mathbf{r}, t)$, as a function of λ: $\ell \simeq t^{\alpha}$.

change in the shape of $Q(\mathbf{r}, t)$ compared with the Gaussian probability density function of standard diffusion processes. Indeed, the main variation of $Q(\mathbf{r}, t)$ is driven by the prefactor $e^{-r\sqrt{\lambda}}$ for large r, hence the exponential-like decay observed in Fig. 2 over numerous decades. This property is reminiscent of the experimental observations of Refs. [1–3].

We now release the assumption $\lambda \gg 1$. The inverse Laplace transform of $Q(\mathbf{k}, z)$ is first computed without any approximation from Eq. (11) (SM Sec. B [22]) and the probability density function is obtained by calculating (numerically) the inverse Fourier transform of $Q(\mathbf{k}, t)$,

$$Q(\mathbf{r}, t) = \frac{1}{2\pi^2} \int_0^{\infty} dk\, Q(k, t) k \sin(kr). \tag{17}$$

$Q(\mathbf{r}, t)$ is plotted in Fig. 3 for $\lambda = 10$, at various diffusing times, showing an exponential-like decay for $t = 0.5, 1, 2$, and 4 over more than seven decades. The characteristic lengths ℓ of the exponential-like parts of $Q(\mathbf{r}, t)$ are found to be well described by the power law $\ell \sim t^{\alpha}$ for any $\lambda \geqslant 2$ and t in ranges including $[0.2; 4]$. Exponent α depends on λ, as shown in the inset of Fig. 3. Because our model considers identical traps, we do not recover the exponent $\alpha = 0.5$ observed over two decades in experiments [1–3]. It is likely that the observed scaling behavior ($\alpha = 0.5$) could be recovered by choosing a proper statistical distribution in the size or depth of the traps. For $\lambda < 2$, any exponential decay would fit $Q(\mathbf{r}, t)$ over two decades with a root-mean-square relative deviation larger than 5%.

The exponential tail predicted in our model originates from a well-identified microscopic mechanism, namely, the interaction of the Brownian particle with a diluted field of deep wells. No *ad hoc* hypothesis on the distribution of relaxation times [19,23,24] or diffusion coefficient distributions [2,15] is necessary to capture this important characteristic of anomalous Fickean diffusion. Indeed, our theory is fundamentally different from the theories relying on an average of standard Gaussian probability resulting from a set of weighted diffusivities [2,15–18]. Our approach mixes two domains, the deep traps where the diffusivity is zero and the rest of the system where it has a finite value. The result of the averaged solutions of the diffusion equation weighted by the probability that the particle is in free space or inside traps, would be the sum of a delta function corresponding to trapped particles plus a regular Gaussian. This average does not correspond at all to our solution of the phenomenon of diffusion in a random field of traps. The explanation for this discrepancy comes from the consideration, in our model, of the exchange between the two domains of small (or even zero) diffusion and of regular diffusion. This highlights the crucial importance of these exchanges.

To conclude, we introduced a model of diffusion of a particle in a dilute field of deep traps and solved the relevant equations. This model has some flexibility because it relies on a unique dimensionless parameter that just depends on the density, the size, and the depth of the traps, and on temperature, but may change widely with the physical conditions. The resulting diffusion is Fickean but with a non-Gaussian probability density function of the displacement in large ranges of time and of dimensionless parameter. This gives a unique possibility of comparing the present theory and experimental data in a range of physical parameters. On a more fundamental side, the theory presented makes the Kramers escape time directly observable owing to its occurrence in the single parameter of the problem.

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