

Communication: A scaling approach to anomalous diffusion

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Communication: A scaling approach to anomalous diffusion

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The paper presents a rigorous derivation of the velocity autocorrelation function for an anomalously diffusing slow solute particle in a bath of fast solvent molecules. The result is obtained within the framework of the generalized Langevin equation and uses only scaling arguments and identities which are based on asymptotic analysis. It agrees with the velocity autocorrelation function of an anomalously diffusing Rayleigh particle whose dynamics is described by a fractional Ornstein-Uhlenbeck process in velocity space. A simple semi-analytical example illustrates under which conditions the latter model is appropriate. © 2014 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4891357>]

The term “anomalous diffusion” has been coined long time ago^{1,2} to label geometrically unconstrained diffusion processes which are not described by Fick's model.^{3,4} Correspondingly, the mean square displacement (MSD) of the diffusing particles does not grow linearly with time, as predicted by Einstein's theory,⁵ but instead $\propto t^\alpha$, where $0 < \alpha < 2$ and $\alpha \neq 1$. The MSD of a tagged particle is an average over all possible realizations of the particle trajectories, $W(t) = \langle (x(t) - x(0))^2 \rangle$, and the diffusive regime refers, strictly speaking, to its asymptotic form for long times,

$$W(t) \stackrel{t \rightarrow \infty}{\sim} 2D_\alpha t^\alpha. \quad (1)$$

The regimes $0 < \alpha < 1$ and $1 < \alpha < 2$ are usually referred to as sub- and superdiffusion, respectively. Here and in the following the tagged particle is supposed to move in an isotropic system and the coordinate x refers to an arbitrary direction in a space-fixed Euclidean reference frame.

A possible route to modeling anomalous diffusion processes is to describe the time evolution of the associated transition probabilities by fractional Fokker-Planck equations (FFPEs).⁶⁻¹⁰ These are generalizations of normal Fokker-Planck equations (FPEs),¹¹ where an additional fractional time derivative enforces the MSD to have the experimentally observed form (1). An interesting model from a theoretical point of view is the anomalously diffusing Rayleigh particle, which describes anomalous Brownian motion on the velocity level. This model has been studied some years ago by Barkai and Silbey⁸ and the corresponding FFPE reads

$$\partial_t p(v, t|v_0, 0) = \eta_\rho {}_0\partial_t^{1-\rho} \left\{ \frac{\partial}{\partial v} v + \frac{k_B T}{m} \frac{\partial^2}{\partial v^2} \right\} p(v, t|v_0, 0). \quad (2)$$

Here $p(v, t|v_0, 0)$ is the conditional probability density for a velocity change $v_0 \rightarrow v$ within time t and the symbol ${}_0\partial_t^{1-\rho}$ denotes a fractional time derivative¹² of order $1 - \rho$. For an

arbitrary function, $f(t)$, the latter is defined as

$${}_0\partial_t^{1-\rho} f(t) = \frac{d}{dt} \int_0^t dt' \frac{(t-t')^{\rho-1}}{\Gamma(\rho)} f(t'), \quad (3)$$

with $\Gamma(\cdot)$ being the generalized factorial¹³ and $\rho > 0$. The parameter η_ρ is a fractional relaxation constant with physical dimension time ^{$-\rho$} and $k_B T/m$ is the mean square velocity of the diffusing particle, where m is its mass, k_B the Boltzmann constant, and T the temperature. The FFPE (2) describes a fractional Ornstein-Uhlenbeck process in velocity space and is to be solved with the initial condition $p(v, 0|v_0, 0) = \delta(v - v_0)$. For long times, the transition probability density tends to the equilibrium (Maxwell) distribution, $\lim_{t \rightarrow \infty} p(v, t|v_0, 0) \equiv p_{eq}(v)$. The link between diffusion in velocity and position space is established through the velocity autocorrelation function (VACF), $c(t) = \langle v(t)v(0) \rangle$, which enables the calculation of the MSD via¹⁴ $W(t) = 2 \int_0^t dt' (t-t')c(t')$. The VACF corresponding to the dynamical model (2) is obtained through $c(t) = \int \int dv dv_0 v v_0 p(v, t|v_0, 0) p_{eq}(v)$, and the resulting form is

$$c(t) = \langle v^2 \rangle E_\rho(-\eta_\rho t^\rho). \quad (4)$$

Here $E_\rho(z) = \sum_{k=0}^{\infty} z^k / \Gamma(1 + \rho k)$ denotes the Mittag-Leffler function of order ρ ,^{15,16} and $\langle v^2 \rangle = \int dv v^2 p_{eq}(v) = k_B T/m$. Setting $\rho = 2 - \alpha$, the resulting MSD has the asymptotic form (1), where

$$D_\alpha = \frac{\langle v^2 \rangle \eta_{2-\alpha}^{-1}}{\Gamma(1 + \alpha)}. \quad (5)$$

In case of normal diffusion, where $\rho = \alpha = 1$, Eq. (2) reduces to the FPE for a normal Rayleigh particle⁴ and (4) to an exponentially decaying function. Alternatively, anomalous diffusion may be described by generalizations of the Langevin equation,¹⁷ where the friction constant is replaced by a memory kernel and the stochastic force is modified accordingly. These generalizations may be motivated by essentially mathematical arguments¹⁸ or by particular physical models for the bath in which the diffusing particle is immersed.^{19,20} It

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is worthwhile mentioning that the underlying assumptions of different models for subdiffusion maybe quite different. The stochastic processes described by the generalized Langevin equations presented in Refs. 18–20 are, for example, Gaussian, whereas the FFPE (2) describes a non-gaussian process. An overview of stochastic models for anomalous diffusion can be found in Refs. 10 and 21.

Any stochastic model for a dynamical system encodes a separation of time scales corresponding to “slow” and “fast” dynamical variables, where the latter are modeled as noise. This is true for normal and anomalous diffusion. Computer simulation experiments on a simple liquid²² have shown that the exponentially decaying VACF of a normally diffusing Rayleigh particle emerges if the mass of a tagged “solute” is systematically increased, slowing its motions down with respect to those of the solvent molecules. A formal proof for this observation has been given *a posteriori*²³ in the framework of the generalized Langevin equation (GLE) introduced by Zwanzig,^{14,24–26} turning the mass scaling into an amplitude scaling of the memory function. The aim of the present paper is to extend the scope of the latter work and to derive the VACF of an *anomalously* diffusing heavy particle in a bath of light solvent molecules from “first principles.” The starting point is again the GLE

$$\dot{v}(t) + \int_0^t d\tau \kappa(t - \tau)v(\tau) = f^+(t), \quad (6)$$

which is an *exact*, deterministic equation of motion for a tagged particle in an interacting many-body system. It is not to be confused with the generalizations of the Langevin equation mentioned earlier (see, e.g., Refs. 18–20), which are *stochastic* equations of motion implying a model for the solvent. In the GLE (6), the memory kernel, κ , and the projected acceleration, f^+ , fully represent the effect of the environment on the dynamics of the tagged particle. Assuming that the time evolution of the total system is described by Hamiltonian dynamics, the projected acceleration has the form

$$f^+(t) = \exp((1 - \mathcal{P})\mathcal{L}t)(1 - \mathcal{P})\mathcal{L}v, \quad (7)$$

where \mathcal{L} is the Liouville operator and \mathcal{P} is a projector whose action on an arbitrary phase function f is defined through $\mathcal{P}f = (\langle vf \rangle / \langle v^2 \rangle)v$. In this context, the brackets $\langle \dots \rangle$ represent an ensemble average over the phase space variables. With the above definitions, the memory kernel can be expressed as autocorrelation function of f^+ ,

$$\kappa(t) = \frac{\langle f^+(0)f^+(t) \rangle}{\langle v^2 \rangle}. \quad (8)$$

Since $\langle v(0)f^+(t) \rangle = 0$ by construction, it follows from (6) that the time evolution of the VACF is described by the integro equation

$$\dot{c}(t) + \int_0^t d\tau \kappa(t - \tau)c(\tau) = 0. \quad (9)$$

The memory kernel is to be considered as a purely formal quantity, since its calculation is in practice as impossible as the explicit solution of the equations of motions for the solute and the solvent molecules. For the following considerations its exact form is, however, not needed and it only matters that

the VACF verifies an equation of motion of the form (9). The normalized solution,

$$\psi(t) = c(t)/c(0), \quad (10)$$

of this integro-differential equation can be expressed by the contour integral

$$\psi(t) = \frac{1}{2\pi i} \oint ds \frac{\exp(st)}{s + \hat{\kappa}(s)}, \quad (11)$$

where $\hat{\kappa}(s) = \int_0^\infty dt \exp(-st)\kappa(t)$ ($\Re\{s\} > 0$) denotes the Laplace-transformed memory function. Expression (11) is simply the inverse Laplace transform of the solution of (9) in Laplace space, setting $c(0) = 1$.

We consider now the situation that the memory function is scaled according to

$$\kappa(t) \rightarrow \lambda\kappa(t), \quad (12)$$

where $\lambda \rightarrow 0$ and $\lambda > 0$. Under the assumptions to be discussed in the following, the above scaling corresponds to changing the mass of the tagged particle as

$$m \rightarrow m/\lambda. \quad (13)$$

In case that $t = 0$, the equivalence of (12) and (13) is strictly valid if one assumes that the Hamiltonian of the full system has the standard form $H = \sum_{i=1}^n p_i^2/(2m_i) + V(x_1, \dots, x_n)$, where n is the total number of degrees of freedom of the system, x_i are the particle coordinates, and p_i the associated momenta. This is seen by using that the Liouville operator has the general form, $\mathcal{L} = \sum_{i=1}^n \{(\partial H/\partial p_i)\partial/\partial x_i - (\partial H/\partial x_i)\partial/\partial p_i\}$, such that $\mathcal{L}v = -(1/m)\partial V/\partial x$. Since $\langle v^2 \rangle = k_B T/m$, it follows then from relations (7) and (8) that $\kappa(0) \propto 1/m$. Defining k to be the index of the tagged particle, the Hamiltonian takes the form $H = \lambda p_k^2/(2m_k) + \sum_{i \neq k} p_i^2/(2m_i) + V(x_1, \dots, x_n)$, which shows that the dynamics of the tagged particle is frozen out in the limit $\lambda \rightarrow 0$. If one can assume that the dynamics of the remaining particles is not affected by this process, as far as the calculation of ensemble averages is concerned, relation (13) is also true for $t > 0$.

Since the Laplace transform is a linear integral transform, the VACF corresponding to the scaled memory kernel (12) is given by

$$\begin{aligned} \psi_\lambda(t) &= \frac{1}{2\pi i} \oint ds \frac{\exp(st)}{s + \lambda\hat{\kappa}(s)} \\ &= \frac{1}{2\pi i} \oint ds \frac{\exp(s\lambda t)}{s + \hat{\kappa}(\lambda s)}, \end{aligned} \quad (14)$$

where the variable change $s \rightarrow s/\lambda$ has been performed to go from the first to the second line. In the limit $\lambda \rightarrow 0$ one thus needs only the *asymptotic* form of $\hat{\kappa}(s)$ for small arguments s . Using a theorem from asymptotic analysis,²⁷ it has been shown in Ref. 28 that this form is entirely determined by the MSD for large times. If the latter has the form (1) the Laplace transformed memory function behaves as

$$\hat{\kappa}(s) \stackrel{s \rightarrow 0}{\sim} \frac{\langle v^2 \rangle}{D_\alpha \Gamma(\alpha + 1)} s^{\alpha-1}. \quad (15)$$

Introducing here the characteristic time scale

$$\tau = \left(\frac{D_\alpha \Gamma(\alpha + 1)}{\langle v^2 \rangle} \right)^{1/(2-\alpha)}, \quad (16)$$

the dimensionless Laplace variable $u = s\tau$, and the dimensionless asymptotic memory function

$$K(u) = u^{\alpha-1}, \quad (17)$$

the normalized VACF takes a form which shows that τ sets the time scale for the velocity of the tagged particle,

$$\psi_\lambda(t) \stackrel{\lambda \rightarrow 0}{\sim} \frac{1}{2\pi i} \oint du \frac{\exp(\lambda u [t/\tau])}{u + K(\lambda u)}. \quad (18)$$

Due to the power-law form of $K(u)$, a scaling factor for its argument may be turned into a scaling factor for its amplitude, $K(\lambda u) = \lambda^{\alpha-1} K(u)$. A new variable change $u \rightarrow u/\lambda^{\alpha-1}$ shifts the latter back into the argument of $K(u)$ and repeating this procedure n times leads to

$$\psi_\lambda(t) \stackrel{\lambda \rightarrow 0}{\sim} \frac{1}{2\pi i} \oint du \frac{\exp(\lambda \lambda^{\alpha-1} \dots \lambda^{(\alpha-1)^{n-1}} u [t/\tau])}{u + K(\lambda^{(\alpha-1)^{n-1}} u)}.$$

Since $|\alpha - 1| < 1$, the limit $n \rightarrow \infty$ can be performed, yielding $\lim_{n \rightarrow \infty} (\alpha - 1)^{n-1} = 0$ for the exponent of λ in the argument of $K(\cdot)$ and $\sum_{k=0}^{\infty} (\alpha - 1)^k = 1/(2 - \alpha)$ for the series of the exponents of λ in the argument of the exponential function. The resulting VACF has then the form

$$\psi_\lambda(t) \stackrel{\lambda \rightarrow 0}{\sim} \frac{1}{2\pi i} \oint du \frac{\exp(\lambda^{1/(2-\alpha)} u [t/\tau])}{u + K(u)}. \quad (19)$$

At this point one can use that

$$E_\rho(-t^\rho) = \frac{1}{2\pi i} \oint du \frac{\exp(ut)}{u(1 + u^{-\rho})} \quad (20)$$

is the contour integral representation of the “stretched” Mittag-Leffler (ML) function,^{15,16} and combining relations (19) and (20) thus leads to

$$\psi_\lambda(t) \stackrel{\lambda \rightarrow 0}{\sim} E_{2-\alpha}(-\lambda [t/\tau]^{2-\alpha}). \quad (21)$$

Obviously, the limit $\lambda \rightarrow 0$ in (21) cannot be strictly performed for finite time arguments and should be interpreted as $\lambda \ll 1$, where $t \gg \tau$ must be fulfilled to obtain appreciable variations of the VACF. In a simple liquid, where all molecules are identical, τ is same for all molecules. Applying now the scaling procedure described above for one of them turns the selected “solute” molecule into a heavy, anomalously diffusing Rayleigh particle, which moves on much longer time scales than the remaining “solvent” molecules. According to relation (21), this time scale is given by

$$\tau_\lambda = \frac{\tau}{\lambda^{1/(2-\alpha)}} \gg \tau, \quad (22)$$

such that

$$\psi_\lambda(t) \approx E_{2-\alpha}(-[t/\tau_\lambda]^{2-\alpha}). \quad (23)$$

Setting here $\eta_\rho = \tau_\lambda^{-\rho}$ and $\rho = 2 - \alpha$, the normalized version of the VACF given in (4) is retrieved.

The scaling procedure described above and the resulting time scale separation can be illustrated with a simple model

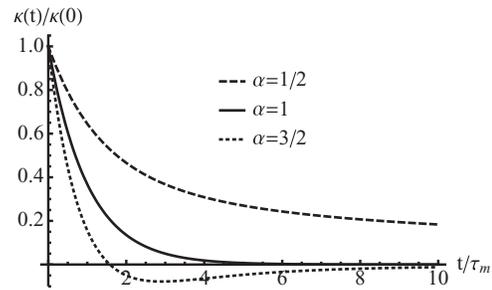


FIG. 1. Model memory function (24) for different values of α . Reprinted with permission from J. Chem. Phys. **134**, 224106 (2011). Copyright 2011 AIP Publishing LLC.

system, where the memory function has the form²⁸

$$\kappa(t) = \Omega^2 M(\alpha, 1, -t/\tau_m). \quad (24)$$

Here, $M(a, b, z)$ is Kummer’s hypergeometric function,¹³ Ω has the dimension of a frequency, and τ_m is the characteristic time scale of the memory function. Fig. 1 shows the function for $\alpha = 1/2, 1, 3/2$. The Laplace transformed memory function,

$$\hat{\kappa}(s) = \Omega^2 \left\{ \frac{\tau_m^\alpha}{s^{1-\alpha}} \frac{1}{(s\tau_m + 1)^\alpha} \right\}, \quad (25)$$

has the requested form (15) for small s , where

$$D_\alpha = \frac{\tau_m^{-\alpha} \langle v^2 \rangle}{\Omega^2 \Gamma(\alpha + 1)}, \quad (26)$$

and the characteristic velocity time scale is

$$\tau = \left(\frac{\tau_m^{-\alpha}}{\Omega^2} \right)^{\frac{1}{2-\alpha}}. \quad (27)$$

The normalized VACF corresponding to the scaled memory function is computed by evaluating the contour integral (14). An analytical solution exists, however, only for $\alpha = 1$, where $\kappa(t) = \Omega^2 \exp(-t/\tau_m)$. For $\alpha \neq 1$ one can follow Ref. 28 and resort to a Padé-approximation for the Laplace-transformed memory function,

$$\hat{\kappa}(s) \approx \frac{\sum_{k=0}^{M_a} a_k (s - s_0)^k}{\sum_{k=0}^{M_b} b_k (s - s_0)^k}, \quad (28)$$

where $\{a_k\}$ and $\{b_k\}$ are constants. The VACF is then approximated by a multi-exponential function which can be evaluated with computer algebra programs. The calculations presented here have been performed with MATHEMATICA,²⁹ setting $M_a = M_b = 10$, and $s_0 = 0.05/\tau_m$. These values were chosen empirically, in order to maintain the relative error of the memory function below 5% for $t/\tau_m \in [0, 1000]$. The amplitude for the memory function was set to $\Omega = 1/\tau_m$, such that $\tau = \tau_m$. Figure 2 shows the normalized VACFs (solid lines) for $\alpha = 1/2, 1, 3/2$ (from top to bottom), and in each row the scaling parameter for the memory function is set to $\lambda = 1, 0.2, 0.02$, respectively, from left to right. In each panel the corresponding asymptotic VACF, which is defined by the rhs of Eq. (23), is displayed for comparison (dashed lines). The results show that the model for an anomalously diffusing Rayleigh particle implies a separation of time scales, as its

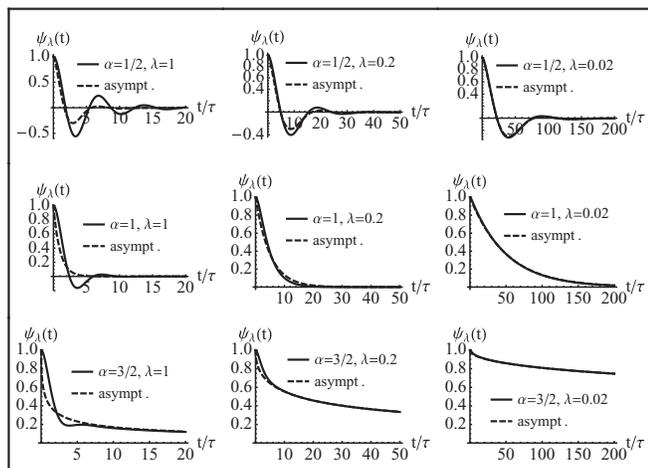


FIG. 2. Velocity autocorrelation functions $\psi_\lambda(t)$ for different scaling factors λ (solid lines) and corresponding asymptotic approximations (23) (dashed lines). From top to bottom $\alpha = 1/2, 1, 3/2$, from left to right $\lambda = 1, 0.2, 0.02$. The amplitude of the memory function (Eq. (24)) is chosen as $\Omega = 1/\tau_m$, such that $\tau = \tau_m$.

counterpart for normal diffusion, and that the model leads to a nearly perfect approximation of the “real” VACF if the time scales τ_λ and τ are separated by about two orders of magnitude. Computer simulations of lipid bilayers, where the lipid molecules exhibit anomalous lateral diffusion, have shown that τ is of the order of picoseconds,³⁰ which is extremely short compared to the millisecond time scale on which these motions are usually studied experimentally.³¹ In this situation the anomalously diffusing Rayleigh article is an excellent model, but it should be kept in mind that it cannot be valid on arbitrarily short time scales, where it becomes even unphysical since the derivatives $c^{(k)}(0)$ all diverge, although they represent physical quantities.^{14,26}

In this Communication, an exact model-free derivation of the VACF for an anomalously diffusing particle has been presented for the case that the motions of the particle are much more slower than those of the molecules in the surrounding solvent. Here the asymptotic form of its MSD is supposed to be known. The effect of the time scale separation on the VACF

has been illustrated for a simple model system. At present, the *prediction* of anomalous diffusion on the basis of general physical properties of a solute–solvent system is still a challenge.

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