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Hydrodynamic Fluctuations and Damped Oscillating Friction Lead to Active Motion

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Abstract

The fractional Langevin equation is used to analyze the erratic motion of a particle in a complex environment. Hydrodynamic backflows and restoring harmonic forces can work concurrently on the probe particle in addition to the constraints imposed by the complex environment, whose characteristics are represented in the friction force the particle encounters. The friction force, which is used for the modelling, corresponds to a damped oscillating function and it is expressed by a one-parameter Mittag-Leffler function. Analytical solutions for the relaxation functions are extracted, and through them observables like the position autocorrelation function, velocity autocorrelation function and mean square displacement are explicitly given. The competition between hydrodynamic fluctuations and frictional forces establishes a memory whose time scale is larger than the diffusional time scale, and during this time window the motion of the particle is superdiffusive.

Keywords: fractional Langevin equation; anomalous diffusion; colored noises; hydrodynamic fluctuations; one-parameter Mittag-Leffler friction

1. Introduction

The motion of a particle subject to thermal fluctuations in an unrestricted environment, within a stable flow, is described by the archetypal Brownian or normal motion [1]. The same particle within a complex environment undergoes an erratic motion that deviates from the normal one. A time-dependent friction, the presence of hydrodynamic fluctuations, conservative forces acting on the particle, or any combination of these factors could be the source of these deviations [2]. The scaling of the mean square displacement is a widely used indicator that classifies the type of motion. Many studies have shown that $\langle x(t)^2 \rangle \sim t^\alpha$, where α is the scaling exponent; $\alpha = 1$ for Brownian or normal, $0 < \alpha < 1$ for subdiffusion, and $1 < \alpha < 2$ for superdiffusion or active motion [3–6].

The generalized Langevin equation, introduced by Mori and Kubo [7–9], outlines both deterministic and stochastic forces acting on a probe particle, provides a stochastic description of a system close to equilibrium, and has been used extensively to study the properties of anomalous diffusion over the last three decades [10–19]. A generalized Langevin equation that permits non-integer time derivatives to characterize changes in the particle’s position and/or velocity is known as a fractional Langevin Equation [20]. This framework has proven valuable in analyzing and understanding particle dynamics in complex environments [4,5,21–24], as well as in describing the dynamics of fluctuating physical systems such as polymers, membranes, and interfaces [25–27]. Mainardi and Pironi established a fractional Langevin equation, which accounts for retardation effects of



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hydrodynamic fluctuations [28], and presented position and velocity correlation functions in terms of the Mittag-Leffler functions [29,30].

A generic fractional Langevin equation, including hydrodynamic backflows, is given by Equation (1)

$$m_c D_t^\mu x(t) + \int_0^t \gamma(t-t') {}_c D_t^\nu x(t') dt' + F_{con}(t) + F_{per}(t) + F_{BB}(t) = \zeta(t), \quad (1)$$

where ${}_c D_t^\mu$ is the fractional derivative, which, according to Caputo's definition [31,32], reads

$${}_c D_t^\mu x(t) = \frac{1}{\Gamma(n-\mu)} \int_c^t \frac{x^{(n)}(u)}{(t-u)^{\mu-n+1}} du, n-1 < \mu < n, n \in \mathbb{N}^+, t > c. \quad (2)$$

$\Gamma()$ is Euler's gamma function and the index c denotes the lower limit of the integral, which in this work is considered zero, and for sake of simplicity we write D_t^μ instead of ${}_0 D_t^\mu$. In Equation (1), the indices μ and ν satisfy the conditions $1 < \mu \leq 2$, $0 < \nu \leq 1$; note that for $\mu = 2$ and $\nu = 1$, Equation (1) returns the generalized Langevin equation. In addition, $F_{con}(t)$ is any conservative force usually taken as a harmonic restoring force, $F_{con}(t) = kx(t)$, with k being the stiffness, and $F_{per}(t)$ is any periodic force acting on the target particle. $F_{BB}(t) = \frac{m_f}{2} \ddot{u}(t) + \gamma_0 \sqrt{\frac{\tau_v}{\pi}} {}_0 D_t^{\frac{1}{2}} \dot{u}(t)$ is the Boussinesq-Basset (BB) force that describes hydrodynamic fluctuations (HFs) [33,34]. The BB force incorporates two terms. The first one stands for the acceleration of the liquid molecules displaced by the tracer particle; m_f is the mass of the fluid element with a volume that equals that of the particle. The second one, written as a fractional semi-derivative, expresses the convolution of the bare friction coefficient, γ_0 , with the acceleration, and is weighted by the square root of the vorticity time, τ_v . Note that $\gamma_0 = 6\pi\eta r$, with η being the dynamic viscosity of the medium and r being the radius of the target particle. The lower limit of the convolution integral in BB force t_0 can be set to $-\infty$, and thus the contribution $\gamma_0 \sqrt{\frac{\tau_v}{\pi}} {}_0 D_t^{\frac{1}{2}} \dot{u}(t) = \gamma_0 \sqrt{\frac{\tau_v}{\pi}} \int_{-\infty}^0 (t-t')^{-\frac{1}{2}} \frac{du(t')}{dt'} dt'$ corresponds to a colored noise caused by BB force, whose correlation can be written as $\langle \zeta_{BB}(t) \zeta_{BB}(0) \rangle \sim t^{-\frac{3}{2}}$ [35,36].

The last two remaining terms in the description of Equation (1) are the friction and noise forces. Both are of common origin when the noise is internal, giving rise to the fluctuation dissipation relation, which holds true in the long-time limit and is unaffected by conservative forces [8].

$$\langle \zeta(t) \zeta(t') \rangle = k_B T \gamma(|t-t'|), \quad (3)$$

where $k_B T$ is Boltzmann's constant times the temperature of the bath. The r.h.s. of Equation (3) can include any term that dissipates energy [37,38]. Equation (3) relates the friction term and the noise. Consequently, if one of the two is known, then the other will be known as well. Let $\zeta(t)$ be the overall random force that can result from both additive and/or multiplicative random forces of different origins. For such a noise, we typically assume that it has zero mean and a variance that is estimated. A lot of research has been done on how internal noises with power-law autocorrelation affect the diffusive motion of a tagged particle [10,11,14–17,19,21,23,39,40]. On the other hand, an intriguing class of internal noises corresponds to Mittag-Leffler (ML) functions with one, two or three parameters [41–45]. The one-parameter ML function is the solution to fractional relaxation processes [46], and it is to fractional kinetics what the exponential function is to classical kinetics. It behaves as a stretched exponential for early times and in the long-time limit behaves as a power law [47,48]. It is defined as a power series, as in Equation (4) [29,30], and converges in the whole complex plane

$$E_\alpha(z) = \sum_{n=0}^{\infty} \frac{z^n}{\Gamma(\alpha n + 1)}, \alpha > 0, z \in \mathbb{C}. \quad (4)$$

For $0 < \alpha < 1$, Equation (4) has been used to describe the friction term of a diffusing particle in the presence of hydrodynamic fluctuations [38]. In addition, for $\alpha = 1$, Equation (4) returns the exponential function, and such a friction term has been used to describe the dynamics of a particle that is also subject to hydrodynamic backflows [49]. A friction term expressed as a one-parameter ML function has the form $\gamma(t) = \frac{\gamma_0}{\tau_m} E_\alpha(-(t/\tau_m)^\alpha)$, where $1 < \alpha < 2$, and τ_m is the characteristic memory of the friction; note that γ_0 can also be a time-dependent variable through its dependence on the dynamic viscosity, η [50]. The Laplace pair $L\{f(t)\}(s) = \int_0^\infty e^{-st} f(t) dt$ of $\gamma(t)$ reads $\gamma(s) = \frac{\gamma_0}{\tau_m} \frac{s^{\alpha-1}}{s^\alpha + \tau_m^{-\alpha}}$. The friction force vanishes in the long-time limit; the use of the Tauberian theorem [51], $\lim_{t \rightarrow \infty} \gamma(t) = \lim_{s \rightarrow 0} s\gamma(s)$, ensures that it goes to zero as s^α . Notably, for $0 < \alpha < 1$, the friction is a completely monotone function converging to zero from positive, while for $1 < \alpha < 2$, the friction force corresponds to a damped oscillating function that has an odd number of roots [52]; see Figure 1.

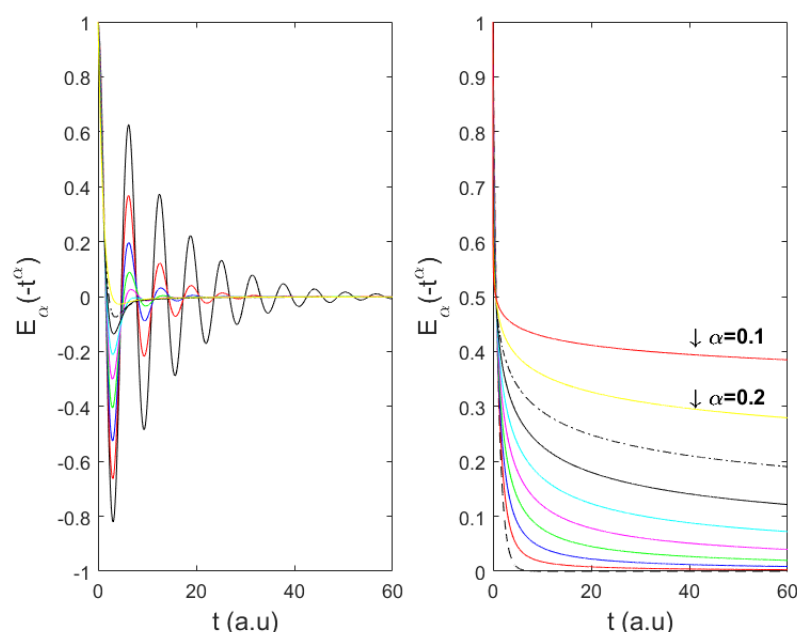


Figure 1. The one-parameter ML as a function of time for various values of the parameter α in the range $0 < \alpha < 2$ is shown; the left panel is for $1 < \alpha < 2$, and the right panel is for $0 < \alpha < 1$.

Even though experimental data and numerical simulations have shown that hydrodynamic fluctuations affect the diffusional particle and provide interesting dynamics [13,53–57], analytical studies of how these fluctuations affect the behaviour of a diffusing particle have received less attention [28,36–38,50,57–62]. Only a few of these works, when hydrodynamic fluctuations are in operation, employ a Mittag-Leffler function to describe either the friction or the noise term [38,61,62]. For $0 < \alpha < 1$, the dynamics of a particle subject to hydrodynamic fluctuations, to a friction force provided by a one-parameter ML function and to a restoring force that can be on or off have been thoroughly studied [38,62]. For $\alpha > 0$, the generalized stochastic resonance, among other properties, has been investigated where the tagged particle is subject to the aforementioned forces and moreover subject to a trichotomous external noise and to a periodic force [61].

In this article, we focus on the unexplored range $1 < \alpha < 2$ of a one-parameter ML function that serves as a friction force; such a friction presents oscillations with attenuating

amplitude. We study the dynamics of a particle that, apart from the friction force, is also subject to (i) hydrodynamic backflows and (ii) a restoring force that can be on or off. We provide analytical solutions for the position and velocity autocorrelation function (PAF/VAF) as well as the mean square displacement (MSD), valid not only at the long-time limit but also over an extended window of time. In each of the cases examined in this work, the motion of the particle is superdiffusive (active motion), with the scaling exponent given by the value of the parameter α of the ML function. A discussion is provided on whether each of the contexts under consideration can depict a physical system in terms of energy balance.

2. Fractional Langevin Equation and Response Functions: Analytical Expressions

Equation (1) for $\mu = 2$ and $\nu = 1$ serves as the foundation for the investigation of the dynamics of a probe particle subject to hydrodynamic backflows and a damped oscillating friction force under the effects of a harmonic restoring force and in the absence of a periodic force.

$$m_{tot}\ddot{x}(t) + \frac{\gamma_0}{\tau_m} \int_0^t E_\alpha\left(-\left(\frac{t-t'}{\tau_m}\right)^\alpha\right) \dot{x}(t') dt' + \gamma_0 \sqrt{\frac{\tau_v}{\pi}} \int_0^t (t-t')^{-\frac{1}{2}} \dot{x}(t') dt' + kx(t) = \xi(t), \quad (5)$$

where $m_{tot} = (m + \frac{m_f}{2})$. By taking the Laplace pair of Equation (5) and by solving with respect to the position, we arrive at the following relation:

$$x(s) = \frac{x_0}{s} - x_0 \omega^2 I(s) + \left(1 + \gamma_0 \sqrt{\tau_u} s^{-1/2}\right) u_0 G(s) + \frac{\xi(s) G(s)}{m_{tot}}, \quad (6)$$

where x_0 and u_0 represent the particle's position and velocity at $t = 0$, respectively. Note that $\gamma_0 = \frac{\gamma_0}{m_{tot}}$, while when a harmonic restoring force is in place, $\omega = \sqrt{\frac{k}{m_{tot}}}$ expresses the frequency of oscillation. Using the definition of the velocity, $u(t) = \dot{x}(t)$, and the Laplace space representation of it, $u(s) = sx(s) - x_0$, together with Equation (6), we obtain

$$u(s) = x_0 \omega^2 G(s) + u_0 \left(1 + \gamma_0 \sqrt{\tau_u} s^{-1/2}\right) g(s) + \frac{\xi(s) g(s)}{m_{tot}}. \quad (7)$$

The response functions $g(s)$, $G(s)$, and $I(s)$ appear in Equations (6) and (7). These functions are interrelated to each other with $g(s) = sG(s)$ and $I(s) = G(s)/s$, with $G(s)$ being given by Equation (8).

$$G(s) = \frac{1}{s^2 + \frac{\gamma_0}{\tau_m} \frac{s^\alpha}{s^\alpha + \tau_m^{-\alpha}} + \gamma_0 \sqrt{\tau_u} s^{3/2} + \omega^2}. \quad (8)$$

For the sake of economy and to avoid having three response functions, we define the generalized response function as

$$R(s) = \frac{s^\delta}{s^2 + \frac{\gamma_0}{\tau_m} \frac{s^\alpha}{s^\alpha + \tau_m^{-\alpha}} + \gamma_0 \sqrt{\tau_u} s^{3/2} + \omega^2}. \quad (9)$$

Equation (9) returns $I(s)$ for $\delta = -1$ and $G(s)$ and $g(s)$ for $\delta = 0, 1$, respectively. Additionally, convolution integrals are computed using the generalized response function (see functions $\lambda(t)$ and $\phi(t)$ in the Appendix A). Equation (9) can be simplified and be written in the form

$$R(s) = \frac{s^{\delta-2} + \nu_2 s^{\delta-2-\alpha}}{1 + \nu_1 s^{-1/2} + \nu_2 s^{-\alpha} + \nu_1 \nu_2 s^{-\alpha-1/2} + \nu_3 s^{-2} + \nu_4 s^{-\alpha-2}}, \quad (10)$$

where ν_1 , ν_2 , ν_3 , and ν_4 are listed in Table 1. We introduce the time scales: (1) the vorticity time scale, $\tau_v = \frac{r^2 \rho_f}{\eta}$, that provides the loss of the hydrodynamic memory, with ρ_f being the density of the fluid molecules; (2) the time period of oscillation, $\tau_h = 1/\omega$; (3) the diffusion time, $\tau_d = 1/\gamma_0$; and (4) the trapping time, $\tau_{tr} = \frac{\gamma_0}{\omega^2}$, that yields the antagonism between friction and stiffness of the harmonic potential. The relationship between the parameters ν_i ($i = 1, 2, 3, 4$) and the different time scales is listed in Table 1.

Table 1. The parameters ν_1 , ν_2 , ν_3 , and ν_4 are listed together with the time scales τ_m , τ_v , τ_d , τ_h , and τ_{tr} as well as their interrelation. Note that $\gamma_0 = \frac{\gamma_0}{m_{tot}}$ and τ_m is the internal memory with $\tau_m \leq 10^{-7}$ s.

$\nu_1 = \gamma_0 \sqrt{\tau_u}$	$\nu_2 = \tau_m^{-\alpha}$	$\nu_3 = (\gamma_0 \tau_m^{-1} + \omega^2)$	$\nu_4 = \omega^2 \tau_m^{-\alpha}$
$\tau_v = \frac{r^2 \rho_f}{\eta}$	$\tau_d = 1/\gamma_0$	$\tau_h = 1/\omega$	$\tau_{tr} = \frac{\gamma_0}{\omega^2}$
$\nu_1 = \frac{\sqrt{\tau_v}}{\tau_d}$	$\nu_2 = \tau_m^{-\alpha}$	$\nu_3 = (\frac{1}{\tau_m \tau_d} + \frac{1}{\tau_h^2})$	$\nu_4 = \frac{\tau_m^{-\alpha}}{\tau_h^2}$

Figure 1 displays the time dependence of the one-parameter ML function for various values of α in the range $0 < \alpha < 2$. The one-parameter ML, when used to describe friction, avoids divergencies as $t \rightarrow 0$, in contrast to power law types, while it exhibits two distinct behaviours for $1 < \alpha < 2$ and for $0 < \alpha < 1$; see Figure 1. For $1 < \alpha < 2$, it presents an oscillatory attenuating behaviour versus time that converges to zero; see the left panel of Figure 1. Note that for $\alpha = 2$, it is a harmonic function. In contrast, for $0 < \alpha < 1$, the right panel of Figure 1, the function is completely monotone, converging to zero from positive values, and it converges the faster the higher the value of α is.

2.1. Time-Domain Response Functions

The multinomial ML function, which has been introduced as the fundamental solution of the ordinary differential equation of fractional discrete distributed order [63], expresses the inverse Laplace of a function with the form $K(s) = s^{-b} (1 - \sum_{i=1}^k (-\nu_i) s^{\alpha_i})^{-1}$, with the exponents α_i , to satisfy the condition $\alpha_1 > \alpha_2 > \dots > \alpha_k$.

$$K(t) = t^{b-1} E_{(a_1, a_2, a_3, \dots, a_n), b}(z_1, z_2, \dots, z_k), \quad (11)$$

where $z_i = -\nu_i t^{\alpha_i}$ and

$$E_{(a_1, a_2, a_3, \dots, a_n), b}(z_1, z_2, \dots, z_n) = \sum_{k=0}^{\infty} \sum_{m_1+m_2+\dots+m_n=k} \frac{k!}{m_1! m_2! \dots m_n!} \frac{\prod_{i=1}^n z_i^{m_i}}{\Gamma(b + \sum_{i=1}^n a_i m_i)}. \quad (12)$$

The detailed form of the generalized response function, $R_\delta(t)$, is given by Equations (A2)–(A4); see Appendix A. Taking the limit $t \rightarrow 0$, and by keeping only the first order, the expansion of $R_\delta(t)$ yields $R_\delta(t \rightarrow 0) = \frac{t^{1-\delta}}{\Gamma(2-\delta)}$, meaning that $g(t \rightarrow 0) = 1$, $G(t \rightarrow 0) = 0$, and $I(t \rightarrow 0) = 0$.

The analytical forms of $R_\delta(t)$ valid in the entire time domain are challenging to manage since they are expressed as multiple sums. By reducing the time domain and using appropriate expansions (see Appendix C of ref. [38]), we end up with simpler forms.

(i) For $1 < \alpha < 3/2$ and for $\frac{\nu_3}{\nu_1 \nu_2} t^{\frac{3}{2}-\alpha} > 1$, and by using the recurrence relation of a two-parameter ML function, $E_{a,b}(z) = z E_{a,a+b}(z) + \frac{1}{\Gamma(b)}$, Equation (A2) reads

$$R_\delta(t) = \frac{t^{-1-\delta}}{\nu_3} \left\{ \left(\nu_2 - \frac{\nu_4}{\nu_3} \right) t^\alpha E_{\alpha, \alpha-\delta} \left(-\frac{\nu_4}{\nu_3} t^\alpha \right) + \frac{1}{\Gamma(-\delta)} \right\}, \quad (13)$$

while for $t > (\frac{\nu_3}{\nu_4})^{1/\alpha}$, Equation (13) takes the form

$$R_\delta(t) = \frac{\nu_2}{\nu_4} \left\{ \frac{t^{-1-\delta}}{\Gamma(-\delta)} - \left(\frac{\nu_3}{\nu_4} - \frac{1}{\nu_2} \right) \frac{t^{1-\delta-\alpha}}{\Gamma(-\alpha-\delta)} \right\}. \quad (14)$$

Equation (14) describes the long-time behaviour of the response function and it is in line with previous findings [38].

(ii) For $\alpha = 3/2$ and for $\frac{\nu_2}{\nu_1} t > 1$, using the recurrence relation once more, Equation (A3) is simplified to

$$R_\delta(t) = \frac{t^{-1-\delta}}{(\nu_1\nu_2 + \nu_3)} \left\{ \frac{1}{\Gamma(-\delta)} + \left(\nu_2 - \frac{\nu_4}{\nu_1\nu_2 + \nu_3} \right) t^{\frac{3}{2}} E_{\frac{3}{2}, \frac{3}{2}-\delta} \left(-\frac{\nu_4 t^{\frac{3}{2}}}{\nu_1\nu_2 + \nu_3} \right) \right\}, \quad (15)$$

while for $t > (\frac{\nu_1\nu_2 + \nu_3}{\nu_4})^{1/\alpha}$, Equation (15) takes the form

$$R_\delta(t) = \frac{\nu_2}{\nu_4} \left\{ \frac{t^{-1-\delta}}{\Gamma(-\delta)} - \left(\frac{\nu_1\nu_2 + \nu_3}{\nu_4} - \frac{1}{\nu_2} \right) \frac{t^{-5/2-\delta}}{\Gamma(-\frac{3}{2}-\delta)} \right\}. \quad (16)$$

Equation (16) describes the long-time behaviour of the generalized response function when $\alpha = 3/2$.

(iii) For $3/2 < \alpha < 2$ and for $\frac{\nu_1\nu_2}{\nu_3} t^{\alpha-\frac{3}{2}} > 1$, Equation (A4) reads

$$R_\delta(t) = \frac{t^{\frac{1}{2}-\delta-\alpha}}{\nu_1\nu_2} \left\{ E_{\frac{3}{2}, \frac{3}{2}-\alpha-\delta} \left(-\frac{\nu_4}{\nu_1\nu_2} t^{\frac{3}{2}} \right) + \nu_2 t^\alpha E_{\frac{3}{2}, \frac{3}{2}-\delta} \left(-\frac{\nu_4}{\nu_1\nu_2} t^{\frac{3}{2}} \right) \right\}, \quad (17)$$

while for $t > (\frac{\nu_1\nu_2}{\nu_4})^{\frac{2}{3}}$, Equation (17) takes the form

$$R_\delta(t) = \frac{\nu_2}{\nu_4} \frac{t^{-1-\delta}}{\Gamma(\frac{3}{2}-\delta-\alpha)} + \frac{\nu_2}{\nu_4} \frac{t^{-\alpha}}{\Gamma(\frac{3}{2}-\delta-2\alpha)} \left(\frac{1}{\nu_2} - \frac{\nu_1\nu_2}{\nu_4} t^{-\frac{3}{2}+\alpha} \right). \quad (18)$$

The generalized response function can clearly be written as $\lim_{t \rightarrow \infty} R_\delta(t) \sim \frac{\nu_2}{\nu_4} t^{-1-\delta}$ in the long-time limit for all instances presented here. For a longer time window, the response functions $g(t)$, $G(t)$, and $I(t)$, which are the detailed forms of $R_\delta(t)$ for $\delta = 1, 0, -1$, are listed in Table 2. It is worthwhile to demonstrate the generalized response function for a scenario in which the hydrodynamic backflows and the restoring force are both turned off. This response function demonstrates how a damped oscillating friction force affects the diffusing behaviour of a particle. Starting from Equation (9) and setting $\nu_1 = \nu_4 = 0$, we obtain $R_{\delta, Fr}(s) = \frac{s^{\delta-2}}{1 - (-\frac{1}{\tau_d \tau_m} \frac{s^{\alpha-2}}{s^\alpha + \tau_m^\alpha})}$, and taking the inverse Laplace pair, we obtain $R_{\delta, Fr}(t) = \sum_{n=0}^{\infty} \left(-\frac{1}{\tau_d \tau_m} \right)^n t^{2n+1-\delta} E_{a, 2n+2-\delta}^n \left(-(t/\tau_m)^\alpha \right)$. For $t > \tau_m$, we arrive at the final form, which reads

$$R_{\delta, Fr}(t) = t^{1-\delta} E_{2-\alpha, 2-\delta} \left(-\frac{\tau_m^{\alpha-1}}{\tau_d} t^{2-\alpha} \right). \quad (19)$$

2.2. Analytical Solutions for PAF, VAF and MSD

Based on the analytical expressions of the generalized response function, Equations (13)–(18), the PAF, VAF, and MSD, are explicitly expressed in the time domain for an extended range of time as in Equations (A5)–(A7); see Appendix A, and for more details refer to Appendix B of ref. [38]. The long-time behaviour of the PAF, VAF, and MSD is provided by Equations (14), (16) and (18). Assuming that $t \rightarrow \infty$, the generalized response function, independent of the values the parameter α takes, can be written as

$R_\delta(t \rightarrow \infty) \frac{\nu_2}{\nu_4} \frac{t^{-1-\delta}}{\Gamma(-\delta)}$. Thus only the response function $I(t) = R_{-1}(t)$ converges to the value of $\frac{\nu_2}{\nu_4} = \frac{1}{\omega^2}$, while $G(t)$ and $g(t)$ go to zero.

Table 2. The response functions $g(t)$, $G(t)$, and $I(t)$ are listed for various value ranges of the parameter α ; each function is valid for a specific extended time window.

$0 < \alpha < \frac{3}{2}$	For $t > (\frac{\nu_1 \nu_2}{\nu_3})^{\frac{1}{2-\alpha}}$
$g(t)$	$\frac{t^{\alpha-2}}{\nu_3} (\nu_2 - \frac{\nu_4}{\nu_3}) E_{\alpha, \alpha-1}(-\frac{\nu_4}{\nu_3} t^\alpha)$
$G(t)$	$\frac{t^{\alpha-1}}{\nu_3} (\nu_2 - \frac{\nu_4}{\nu_3}) E_{\alpha, \alpha}(-\frac{\nu_4}{\nu_3} t^\alpha)$
$I(t)$	$\frac{1}{\nu_3} \{1 + (\nu_2 - \frac{\nu_4}{\nu_3}) t^\alpha E_{\alpha, \alpha+1}(-\frac{\nu_4}{\nu_3} t^\alpha)\}$
$\alpha = \frac{3}{2}$	For $t > \frac{\nu_1}{\nu_2}$
$g(t)$	$\frac{t^{-\frac{1}{2}}}{\nu_1 \nu_2 + \nu_3} (\nu_2 - \frac{\nu_4}{\nu_1 \nu_2 + \nu_3}) E_{\frac{3}{2}, \frac{1}{2}}(-\frac{\nu_4}{\nu_1 \nu_2 + \nu_3} t^\alpha)$
$G(t)$	$\frac{t^{\frac{1}{2}}}{\nu_1 \nu_2 + \nu_3} (\nu_2 - \frac{\nu_4}{\nu_1 \nu_2 + \nu_3}) E_{\frac{3}{2}, \frac{3}{2}}(-\frac{\nu_4}{\nu_1 \nu_2 + \nu_3} t^\alpha)$
$I(t)$	$\frac{1}{\nu_1 \nu_2 + \nu_3} \{1 + (\nu_2 - \frac{\nu_4}{\nu_1 \nu_2 + \nu_3}) t^{\frac{3}{2}} E_{\frac{3}{2}, \frac{5}{2}}(-\frac{\nu_4}{\nu_1 \nu_2 + \nu_3} t^\alpha)\}$
$\frac{3}{2} < \alpha < 2$	For $t > (\frac{\nu_3}{\nu_1 \nu_2})^{\alpha-\frac{3}{2}}$
$g(t)$	$\frac{t^{-\frac{1}{2}}}{\nu_1} E_{\frac{3}{2}, \frac{1}{2}}(-\frac{\nu_4}{\nu_1 \nu_2} t^{\frac{3}{2}}) + \frac{t^{-\frac{1}{2}-\alpha}}{\nu_1 \nu_2 \Gamma(\frac{1}{2}-\alpha)} - \frac{\nu_4 t^{1-\alpha}}{(\nu_1 \nu_2)^2} E_{\frac{3}{2}, 2-\alpha}(-\frac{\nu_4}{\nu_1 \nu_2} t^{\frac{3}{2}})$
$G(t)$	$\frac{t^{\frac{1}{2}}}{\nu_1} E_{\frac{3}{2}, \frac{3}{2}}(-\frac{\nu_4}{\nu_1 \nu_2} t^{\frac{3}{2}}) + \frac{t^{\frac{1}{2}-\alpha}}{\nu_1 \nu_2 \Gamma(\frac{3}{2}-\alpha)} - \frac{\nu_4 t^{2-\alpha}}{(\nu_1 \nu_2)^2} E_{\frac{3}{2}, 3-\alpha}(-\frac{\nu_4}{\nu_1 \nu_2} t^{\frac{3}{2}})$
$I(t)$	$\frac{t^{\frac{3}{2}}}{\nu_1} E_{\frac{3}{2}, \frac{5}{2}}(-\frac{\nu_4}{\nu_1 \nu_2} t^{\frac{3}{2}}) + \frac{t^{\frac{3}{2}-\alpha}}{\nu_1 \nu_2} E_{\frac{3}{2}, \frac{5}{2}-\alpha}(-\frac{\nu_4}{\nu_1 \nu_2} t^{\frac{3}{2}})$

The same is also true when t and t' take large values simultaneously and their difference, which is defined as time lag, remains finite. Considering thermal equilibrium at the beginning of the motion, i.e., $\langle x_0 \xi(t) \rangle = 0$, $\langle x_0 u_0 \rangle = 0$, $u_0^2 = \frac{k_B T}{m_p}$, where m_p is the mass of the diffusing particle, and $m_{tot} x_0^2 \omega^2 = k_B T$, we end up with

$$\lim_{t \rightarrow \infty} \frac{\langle x(t)x(t+\tau) \rangle}{x_0^2} = 1 - \omega^2 I(\tau), \quad (20)$$

$$\lim_{t \rightarrow \infty} \frac{\langle u(t)u(t+\tau) \rangle}{\frac{m_p}{m_{tot}} u_0^2} = g(\tau), \quad (21)$$

and

$$\lim_{t \rightarrow \infty} \langle (x(t) - x(t+\tau))^2 \rangle = \frac{2k_B T}{m_{tot}} I(\tau). \quad (22)$$

Moreover, simplified forms of the PAF, VAF, and MSD in the entire time window can be provided by considering the limit $\lim_{t' \rightarrow 0} R_\delta(t')$ in Equations (A5)–(A7) while $t > 0$. This limit takes the value of 1 for $\delta = 1$, and it equals zero for the other values.

$$\langle x(t)x(0) \rangle = x_0^2 - x_0^2 \omega^2 I(t) + x_0 u_0 G(t) + \frac{x_0 u_0}{m_{tot}} \lambda(t), \quad (23)$$

$$\langle u(t)u(0) \rangle = u_0^2 g(t) - x_0 u_0 \omega^2 G(t) + \frac{u_0^2}{m_{tot}} \lambda(t), \quad (24)$$

and

$$\begin{aligned} \langle (x(t) - x(0))^2 \rangle = & \frac{2k_B T}{m_{tot}} I(t) + \frac{u_0^2}{m_{tot}^2} \lambda^2(t) - 2x_0 u_0 \omega^2 G(t) I(t) \\ & + \frac{2u_0^2}{m_{tot}} G(t) \lambda(t) - \frac{2x_0 u_0 \omega^2}{m_{tot}} I(t) \lambda(t), \end{aligned} \quad (25)$$

The minimum time value implemented either in Equations (A5)–(A7) or in Equations (23)–(25) should be larger than the ratio of the radius of the particle to the speed of sound in the medium, $t > r/u_s$; otherwise, compressibility effects should be taken into account [64]. Instead, the time lag implemented in Equations (20)–(22) does not suffer from any limitation.

3. Results

The diffusion time scale τ_d and the hydrodynamic time scale τ_v are interrelated with one another. Given that $\tau_d = \frac{m_{tot}}{\gamma_0}$, $\tau_v = r^2 \frac{\rho_f}{\eta}$, and $\gamma_0 = 6\pi\eta r$, one can easily arrive at the relation $\frac{\tau_d}{\tau_v} = \frac{2}{9} \frac{m_p}{m_f} + \frac{1}{9}$. For the discussion and analysis that follows, we set the memory's time scale at $\tau_m = 10^{-7}$ s, which is significantly lower than all other time scales, and the harmonic oscillator's time scale at 10^{-1} s, which is significantly greater than all other time scales. We identify four scenarios for the diffusion and hydrodynamic time scale values: (a) A heavy particle within hydrodynamic fluctuations, $m_p \approx 500m_f$ or $t_d = 10^{-4}$ s and $\tau_v = 10^{-6}$ s. (b) A light particle within hydrodynamic fluctuations, $m_f = 10m_p$, $\tau_d = 10^{-4}$ s, and $\tau_v = 9\tau_d$. (c) The diffusion and hydrodynamic time scales are equal to one another, 10^{-4} s, which leads to the relationship $m_p = 4m_f$. (d) A diffusion time scale of $\tau_d = 10^{-4}$ s and zero hydrodynamic fluctuations. Notably, for all cases examined, the trapping time, $\tau_{tr} = \frac{\tau_h^2}{\tau_d}$, has the value of 10^2 . Moreover, we anticipate that the first and fourth cases should share similarities given that hydrodynamic backflows significantly impact the motion of light particles, with respect to the fluid molecules, rather than the heavy ones.

Figure 2 illustrates the normalized position autocorrelation function versus the lag time, τ , for various values of the parameter α . Over times that are either one scale less than the time scale of the restoring force (cases a and d) or equal to it (cases b and c), the motion is highly persistent. The rate at which the strongly correlated motion transitions to anticorrelated motion increases as the value of the parameter α increases. The exception is in case b for $\alpha = 1.9$, where the highly correlated motion for time scales is twice the time scale of the restoring force, which subsequently starts an oscillating behaviour when the motion enters the trapping time scale. Oscillations are present in case d where hydrodynamic fluctuations are not present, and their amplitude is larger the greater the value of the parameter α ; we employ a reduced scale for data display since the strong oscillatory nature for large values of the parameter α prevents a sharp depiction of the values. The primary effect of hydrodynamic fluctuations on motion, based on the information that a position correlation function can provide, is that it retains highly correlated motion for time scales up to the time scale of the restoring force.

Figure 3 shows the normalized velocity autocorrelation function (NVAf) versus the lag time, τ , for all cases considered here. The last case ($\tau_v = 0$) considers a particle experiencing an oscillating friction during its motion with attenuating amplitude, and it is also subject to a restoring force whose time scale is three times larger than the time scale of friction. Depending on the value of the parameter α , which defines the amplitude of the oscillations, the memory can be extended up to the time scale of the restoring force and then inserted into negative values, starting oscillating and converging slowly to zero. The first case contains all the conditions of the last one, plus it takes into account hydrodynamic fluctuations with a time scale much smaller than the diffusion time scale (heavy particle).

It would be expected that these two cases would be much similar, but it is clear that they differ with respect to the oscillating character and the strongly antipersistent character for times larger than that of the restoring force. When hydrodynamic backflows are dominant, as in cases b and c, the rate of decrease of the velocity autocorrelation function is almost identical and independent of the value of the parameter α . The motion gets a memory whose time scale can be even larger than the time scale of the hydrodynamic fluctuations. This memory's time range is closely related to the first vanishing point of the VAF and sets the maximum time window in which the particle moves, on average, in the same direction. It is evident that hydrodynamic fluctuations smooth the changes of the velocity autocorrelation function, retaining its persistent character as the latter converges to zero, even when they have a secondary role in the overall dynamics.

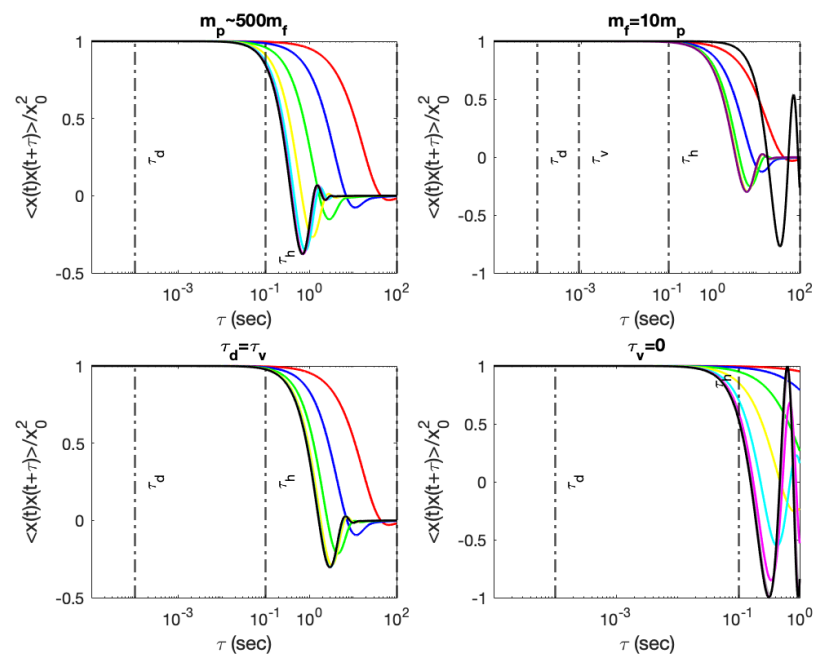


Figure 2. The normalized position autocorrelation function, $\langle x(t)x(t+\tau) \rangle / x_0^2$, versus the lag time, τ , for various values of the parameter α in the range $1.1 \leq \alpha \leq 1.9$ with a step of 0.1 are shown. The color code reads red/blue/green/yellow/cyan/magenta/gray/purple/black for the values of α 1.1/1.2/1.3/1.4/1.5/1.6/1.7/1.8/1.9. The upper left panel shows case a, the upper right panel shows case b, the bottom left panel shows case c, and the bottom right panel shows case d. The trapping time for all cases equals $\tau_{tr} = 10^2$ s.

Figure 4 shows the mean square displacement (MSD) weighted by the thermal energy and the total mass, the mass of the particle plus the mass of the displaced fluid molecules within a volume that equals that of the particle, versus the lag time for each one of the cases examined in this work. This property delivers the fewest insights regarding the role of hydrodynamic fluctuations on the diffusive motion. Three scaling regimes are apparent during the motion; they actually are not really visible for each case and for each value of the parameter α . Ballistic motion is associated with the first regime, where the scaling exponent is 2. It is evident as the slope of the first line of a bilinear function, particularly for low values of the parameter α , such as $\alpha = 1.1$ (red lines in all subplots), where these two concretely different slopes are displayed. It depicts time scales that are slightly greater than the diffusive time scale. The same is true for the remainder of the values of α , but the greater its value, the sooner this slope changes. The second phase describes scaling exponents equal to the value of the parameter α , whereas the last regime represents the trapping when the plateau starts to be constructed. These three regimes are formed in shorter time scales the bigger the value of α is.

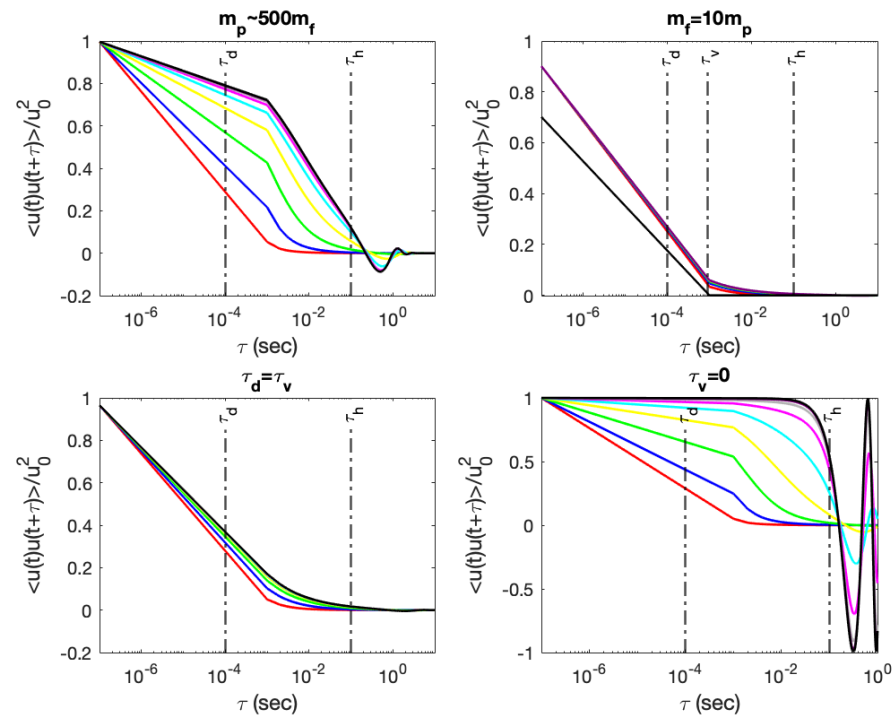


Figure 3. The normalized velocity autocorrelation function, $\langle u(t)u(t+\tau) \rangle / u_0^2$, versus the lag time, τ , for various values of the parameter α in the range $1.1 \leq \alpha \leq 1.9$ with a step of 0.1 is shown. The color code reads red/blue/green/yellow/cyan/magenta/gray/purple/black for the values of α 1.1/1.2/1.3/1.4/1.5/1.6/1.7/1.8/1.9. The upper left panel shows case a, the upper right panel shows case b, the bottom left panel shows case c, and the bottom right panel shows case d.

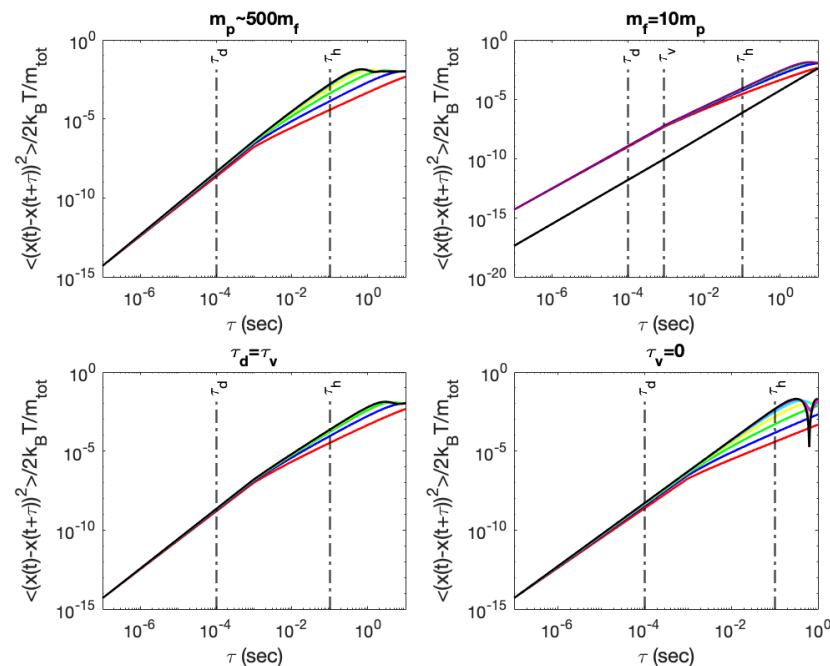


Figure 4. The MSD, $\langle (x(t) - x(t+\tau))^2 \rangle$, weighted by the thermal energy divided by the overall mass, $2k_B T / m_{tot}$, versus the time lag, τ , for various values of the parameter α in the range $1.1 \leq \alpha \leq 1.9$ with a step of 0.1 is shown. The color code reads red/blue/green/yellow/cyan/magenta/gray/purple/black for the values of α 1.1/1.2/1.3/1.4/1.5/1.6/1.7/1.8/1.9. The upper left panel shows case a, the upper right panel shows case b, the bottom left panel shows case c, and the bottom right panel shows case d.

4. Discussion

What is the bare influence of a damped oscillating friction force on the dynamics of a diffusing particle over time? As we already mentioned above, we start from Equation (9), and we eliminate the terms due to hydrodynamic backflows and to restoring force. What remains is the term due to friction, and the response function can easily be expressed in the time domain; see Equation (19).

Figure 5 shows the normalized velocity autocorrelation function, $\frac{\langle u(t)u(t+\tau) \rangle}{u_0^2}$, as it is computed either by numerical inversion of the Laplace transform of the corresponding response function, Equation (10), or by the analytical solution provided by Equation (19) for $\delta = 1$. The agreement between the two is perfect. The motion acquires a memory that spans scales significantly greater than the diffusion time scale (10^{-4} s), regardless of the value of the parameter α . The resulting memory is active for extended periods of time (hundreds of seconds) when α takes large values. In contrast, NVAF converges to zero in the sub-second regime and diminishes linearly at low values of α . In addition, in the long-time limit, by using the expansion $E_{a,b}(z) \sim (-z)^{-1} \sum_{k=0}^{\infty} \frac{\Gamma(k+1)}{\Gamma(b-a(k+1))} \frac{z^{-k}}{k!}$, Equation (19) can be written as $\lim_{t \rightarrow \infty} R_{\delta, Fr} = \frac{\tau_d}{\tau_m^{\alpha-1}} \frac{t^{\alpha-1-\delta}}{\Gamma(2-\delta-\alpha)}$. Substituting the latter into Equations (23)–(25) for $\delta = 0, -1, 1$, we end up with $\langle x(t)x(0) \rangle = x_0^2 - x_0 u_0 \frac{\tau_d}{\tau_m^{\alpha-1}} \frac{t^{\alpha-1}}{\Gamma(2-\alpha)}$, $\langle u(t)u(0) \rangle = u_0^2 \frac{\tau_d}{\tau_m^{\alpha-1}} \frac{t^{\alpha-2}}{\Gamma(1-\alpha)}$, and $\langle (x(t) - x(0))^2 \rangle = \frac{2k_B T}{m} \frac{\tau_d}{\tau_m^{\alpha-1}} \frac{t^{\alpha}}{\Gamma(3-\alpha)}$. The latter demonstrates that the motion is superdiffusive with a scaling exponent equal to the value of the parameter α , and memory that converges to zero as $t^{\alpha-2}$. These results describe the motion of a particle that converts fluctuations to persistent motion: active motion. Such a model can describe a particle that exploits an internal source of energy for moving unidirectionally. However, in this scenario, only the noise term and friction are present. Given that a friction force always goes against the motion, this result leads to rectification, but without the presence of a coupling system. In addition, this model cannot describe a random self-propelling active particle since the fluctuation dissipation relation used in this work does not hold true. This scenario most likely cannot explain a physical system unless it can be used to interpret experimental data, in which case it will need to be re-examined. Conversely, such an active motion can exist in the absence of any internal source of energy when hydrodynamic fluctuations are in place, which go against the motion, and thus the friction force goes against it; see below.

In order to shed light on the differences of the cases studied in this work, Figure 6 shows the NVAF vs. the time lag, τ , for values of the parameter α in the range [1.1–1.9] with a step of 0.1. In the case of ($m_p \gg m_f$), the hydrodynamic backflow can be ignored. This is in line with the results displayed in Figure 4, where the black and red lines, which describe motion in the absence of hydrodynamic fluctuations, coincide for all values of the parameter α . In contrast, the NVAF value is always smaller for the blue and green lines, which represent either light particles or particles with a mass similar to the fluid molecules. It is evident from cases a, b, and c that the antagonism between friction and hydrodynamic fluctuations produces a memory whose time scale is longer than the diffusional and the hydrodynamic time scale and causes the particle to actively move before the latter inserts in the trapping time scale.

This work is a first attempt to understand the dynamics of diffusing particles subject to (i) hydrodynamic fluctuations, (ii) restoring force, and (iii) oscillating friction force with attenuating amplitude. Future research can compute energy balances using the results presented here. On the other hand, experimental data can provide feedback on the range of applicability of the present model. This model may be used to evaluate the trajectories of colloids moving in complex surroundings, since such situations are conducive to the generation of memory effects. Moreover, the model makes it possible to test experimental

results based on methods such as optical tweezers, atomic force microscopy, and even liquid cell transmission electron microscopy investigations under specific conditions, particularly those pertaining to the beam rate. In each type of experiment, the key point is that the mass of the probe particle should be comparable to the mass of the solute molecules in order for the hydrodynamic backflows to be in operation.

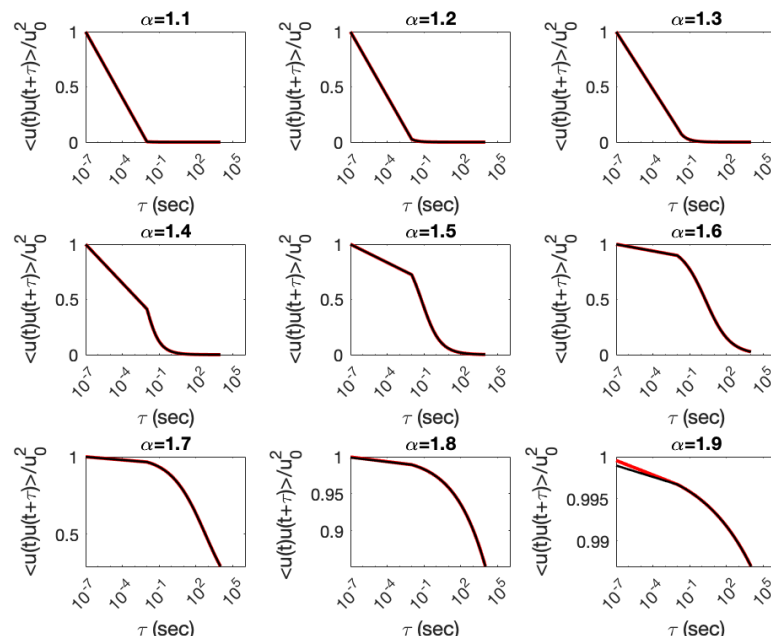


Figure 5. The normalized velocity autocorrelation function, $\langle u(t)u(t+\tau) \rangle / u_0^2$, versus the lag time, τ , for various values of the parameter α in the range $1.1 \leq \alpha \leq 1.9$ with a step of 0.1 is shown. Red lines stand for the numerical inversion of Equation (10), and the black ones for the analytical solution given by Equation (19) for $t > \tau_m$. Note that the values of $\tau_m = 10^{-7}$ and $\tau_d = 10^{-4}$ have been used for the calculations.

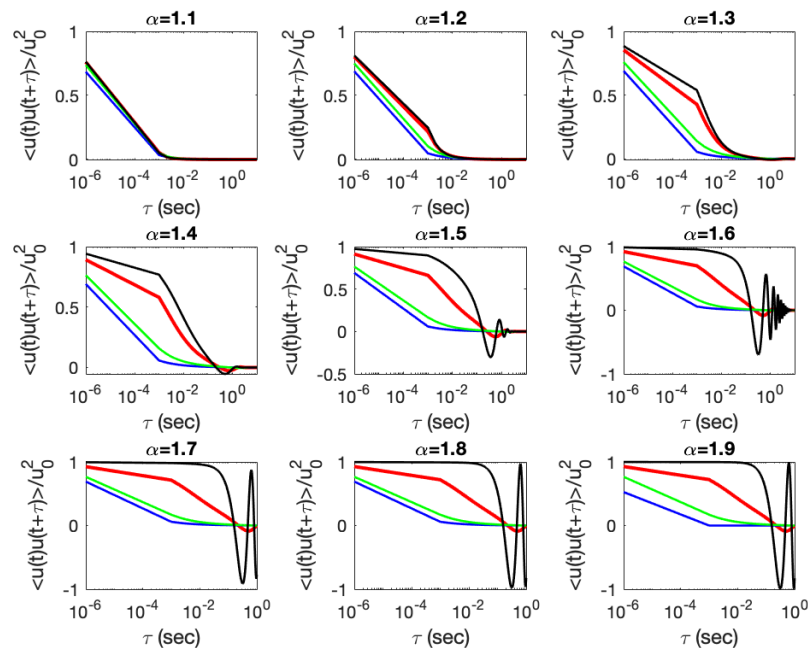


Figure 6. The normalized velocity autocorrelation function, $\langle u(t)u(t+\tau) \rangle / u_0^2$, versus the lag time, τ , for various values of the parameter α in the range $1.1 \leq \alpha \leq 1.9$ with a step of 0.1 is shown for the different cases studied in this work. The color code is red for case a ($m_p \gg m_f$), blue for case b ($m_f = 10m_p$), green for case c ($m_p = 4m_f$), and black for case d (hydrodynamic fluctuations are not present).

5. Conclusions

This work investigates the effect of hydrodynamic fluctuations when a restoring force is present and the friction term corresponds to a damped oscillation function defined by a one-parameter ML function with the parameter $1 < \alpha < 2$. Solving the fractional Langevin equation, the generalized response function in Laplace space is derived, and its inversion yields precise analytical forms for the MSD, the PAF, and the VAF which are valid over extended time periods. In the long-time limit, MSD converges to a constant value, highlighting trapping due to restoring force; the PAF goes to zero, as does the VAF. PAF and MSD do not offer much information in distinguishing among different cases considered in this work. On the contrary, VAF provides valuable insights on the role of each force. When a restoring force, hydrodynamic fluctuations, and a damped oscillatory friction force are in operation, then the memory's time span can reach, in some cases, even the time scale of the restoring force. The created memory, the result of the antagonism between friction and hydrodynamic backflows, supports a persistent motion and can be a good model for explaining the superdiffusivity of a probe particle. In the time window defined by the time scale of the restoring force and the trapping time, the VAF would change sign consecutively, thus tracing periods between persistent and antipersistent motion if hydrodynamic backflow were to be turned off. Conversely, its presence leads to a significant reduction in the amplitude of the fluctuations in this regime.

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Abbreviations

The following abbreviations are used in this manuscript:

BB	Boussinesq–Basset
ML	Mittag-Leffler
PAF	Position Autocorrelation Function
VAF	Velocity Autocorrelation Function
MSD	Mean Square Displacement
NVAF	Normalized Velocity Autocorrelation Function

Appendix A

Starting from Equation (5) of the main text and by using the Laplace pair $L\{f(t)\}(s) = \int_0^\infty e^{-st} f(t) dt$, we write $m_{tot}(s^2 x(s) - sx_0 - u_0) + \frac{\gamma_0}{\tau_m} \frac{s^{\alpha-1}}{s^\alpha + \tau_m^{-1}} (sx(s) - x_0) + \gamma_0 \sqrt{\tau_v} s^{-1/2} (s^2 x(s) - sx_0 - u_0) + kx(s) = \zeta(s)$. Rearranging the terms of the latter with respect to the $x(s)$, we end up with Equation (6) of the main text, where

$$G(s) = \frac{1}{s^2 + \frac{\gamma_0}{\tau_m m_{tot}} \frac{s^\alpha}{s^\alpha + \tau_m^{-1}} + \frac{\gamma_0 \sqrt{\tau_v} s^{3/2}}{m_{tot}} + \frac{k}{m_{tot}}}. \quad (A1)$$

The response functions $I(s)$ and $g(s)$ are defined through Equation (A1) as follows: $I(s) = G(s)/s$, $g(s) = sG(s)$. Instead of computing each one of the response functions $I(s)$, $G(s)$, and $g(s)$ in the time domain, we define the generalized response function $R_\delta(s)$ [38], which returns $I(s)$ for $\delta = -1$, $G(s)$ for $\delta = 0$, and $g(s)$ for $\delta = 1$. Using Equations (11) and (12), the generalized response function for $1 < \alpha < 2$ reads as follows:

(i) for $\alpha < 3/2$

$$R_\delta(t) = t^{1-\delta} E_{(2+a, 2, \frac{1}{2}+a, a, \frac{1}{2}), 2-\delta}(-\nu_4 t^{2+a}, -\nu_3 t^2, -\nu_1 \nu_2 t^{\frac{1}{2}+a}, -\nu_2 t^a, -\nu_1 t^{\frac{1}{2}}) \\ + t^{1+a-\delta} E_{(2+a, 2, \frac{1}{2}+a, a, \frac{1}{2}), 2+a-\delta}(-\nu_4 t^{2+a}, -\nu_3 t^2, -\nu_1 \nu_2 t^{\frac{1}{2}+a}, -\nu_2 t^a, -\nu_1 t^{\frac{1}{2}}), \quad (\text{A2})$$

(ii) for $\alpha = 3/2$

$$R_\delta(t) = t^{1-\delta} E_{(\frac{7}{2}, 2, \frac{3}{2}, \frac{1}{2}), 2-\delta}(-\nu_4 t^{\frac{7}{2}}, -(\nu_1 \nu_2 + \nu_3) t^2, -\nu_2 t^{\frac{3}{2}}, -\nu_1 t^{\frac{1}{2}}) \\ + t^{\frac{5}{2}-\delta} E_{(\frac{7}{2}, 2, \frac{3}{2}, \frac{1}{2}), \frac{7}{2}-\delta}(-\nu_4 t^{2+a}, -(\nu_1 \nu_2 + \nu_3) t^2, -\nu_2 t^{\frac{3}{2}}, -\nu_1 t^{\frac{1}{2}}), \quad (\text{A3})$$

(iii) and for $3/2 < \alpha < 2$

$$R_\delta(t) = t^{1-\delta} E_{(2+a, \frac{1}{2}+a, 2, a, \frac{1}{2}), 2-\delta}(-\nu_4 t^{2+a}, -\nu_1 \nu_2 t^{\frac{1}{2}+a}, -\nu_3 t^2, -\nu_2 t^a, -\nu_1 t^{\frac{1}{2}}) \\ + t^{1+a-\delta} E_{(2+a, \frac{1}{2}+a, 2, a, \frac{1}{2}), 2+a-\delta}(-\nu_4 t^{2+a}, -\nu_1 \nu_2 t^{\frac{1}{2}+a}, -\nu_3 t^2, -\nu_2 t^a, -\nu_1 t^{\frac{1}{2}}). \quad (\text{A4})$$

Using Equations (6) and (7), we construct the correlations $x(s)x(s')$, $x(s)u(s)$, and $u(s)u(s')$. The inversion of them in the time domain yields Equations (A5)–(A7). These equations are valid under the assumption that both t and t' take large values and the difference $|t - t'|$ is finite.

$$\langle x(t)x(t') \rangle = x_0^2 - \frac{k_B T}{m_{tot}} I(|t - t'|) + (\frac{k_B T}{m_{tot}} - x_0^2 \omega^2)(I(t) + I(t')) \\ + x_0 u_0 (G(t) + G(t')) + (u_0^2 - \frac{k_B T}{m_{tot}}) G(t) G(t') \\ + (x_0^2 \omega^4 - \frac{k_B T}{m_{tot}} \omega^2) I(t) I(t') - x_0 u_0 \omega^2 (G(t) I(t') + G(t') I(t)) \\ + \frac{x_0 u_0}{m_{tot}} (\lambda(t) + \lambda(t')) - \frac{x_0 u_0 \omega^2}{M} (I(t) \lambda(t') + I(t') \lambda(t)) \\ + \frac{u_0^2}{m_{tot}} (G(t) \lambda(t') + G(t') \lambda(t)) + \frac{u_0^2}{m_{tot}^2} \lambda(t) \lambda(t'), \quad (\text{A5})$$

$$\langle u(t)u(t') \rangle = \frac{k_B T}{m_{tot}} g(|t - t'|) + (u_0^2 - \frac{k_B T}{m_{tot}}) g(t) g(t') \\ + (x_0^2 \omega^4 - \frac{k_B T}{m_{tot}} \omega^2) G(t) G(t') - x_0 u_0 \omega^2 (G(t) g(t') + G(t') g(t)) \\ + \frac{u_0^2}{m_{tot}} (g(t) \lambda(t') + g(t') \lambda(t)) - \frac{x_0 u_0 \omega^2}{m_{tot}} (G(t) \lambda(t') + G(t') \lambda(t)) \\ + \frac{u_0^2}{m_{tot}^2} \lambda(t) \lambda(t') \quad (\text{A6})$$

$$\langle (x(t) - x(t'))^2 \rangle = \frac{2k_B T}{m_{tot}} I(|t - t'|) + (u_0^2 - \frac{k_B T}{m_{tot}}) (G(t) - G(t'))^2 \\ + \omega^2 (\omega^2 x_0^2 - \frac{k_B T}{m_{tot}}) (I(t) - I(t'))^2 + \frac{u_0^2}{m_{tot}^2} (\lambda(t) - \lambda(t'))^2 \\ - 2x_0 u_0 \omega^2 (G(t) - G(t')) (I(t) - I(t')) \\ + \frac{2u_0^2}{m_{tot}} (G(t) - G(t')) (\lambda(t) - \lambda(t')) \\ - \frac{2\omega^2 x_0 u_0}{m_{tot}} (I(t) - I(t')) (\lambda(t) - \lambda(t')), \quad (\text{A7})$$

where $\lambda(t)$ has an analytical form given from the generalized response function for $\delta = \frac{3}{2}$.

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