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# Langevin analysis for time-nonlocal Brownian motion with algebraic memories and delay interactions

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**Abstract.** Starting from a Langevin equation with memory describing the attraction of a particle to a center, we investigate its transport and response properties corresponding to two special forms of the memory: one is algebraic, i.e., power-law, and the other involves a delay. We examine the properties of the Green function of the Langevin equation and encounter Mittag-Leffler and Lambert W-functions well-known in the literature. In the presence of white noise, we study two experimental situations, one involving the motional narrowing of spectral lines and the other the steady-state size of the particle under consideration. By comparing the results to counterparts for a simple exponential memory, we uncover instructive similarities and differences. Perhaps surprisingly, we find that the Balescu-Swenson theorem that states that non-Markoffian equations do not add anything new to the description of *steady-state* or *equilibrium* observables is violated for our system in that the saturation size of the particle in the steady-state depends on the memory function utilized. A natural generalization of the Smoluchowski equation for the time-local case is examined and found to satisfy the Balescu-Swenson theorem and describe accurately the first moment but not the second and higher moments. We also calculate two-time correlation functions for all three cases of the memory, and show how they differ from (tend to) their Markoffian counterparts at small (large) values of the difference between the two times.

## 1 Introduction

The time evolution of the probability density of a particle attracted to a center via harmonic forces while being simultaneously subjected to white Gaussian noise is of interest in a large variety of contexts. The label Ornstein-Uhlenbeck is attached to the system or process under such situations and the governing equation is said to be the Smoluchowski equation. It is ubiquitous in statistical mechanics [1,2] and, in a one-dimensional system takes the form

$$\frac{\partial P(x,t)}{\partial t} = \frac{\partial}{\partial x}\left(\gamma x P(x,t) + D\frac{\partial P(x,t)}{\partial x}\right) \qquad (1)$$

where $\gamma$ measures the rate of attraction to the fixed center and $D$ is the diffusion constant. Its solutions are well-known as being essentially identical to those of the simple diffusion equation (no attractive center) provided the time $t$ itself undergoes a saturation transformation via the Ornstein-Uhlenbeck prescription: $t \to (1 - e^{-2\gamma t})/2\gamma$. The word "essentially" refers here to the fact that initial conditions dictate in this context an extra term not present in the diffusion equation. The term decays at the rate $\gamma$ in a well-known way.

Our interest in the present paper is in systems in which the attraction to the center proceeds via a *time-nonlocal* process. Memory-possessing Langevin equations have come under investigation in various unrelated contexts in the past, but it is appropriate to say that modern work on the topic appears to have begun with Budini and Cáceres [3]. In a report with far-reaching conclusions, they studied generalized Langevin equations producing many interesting results, but restricted their investigations primarily to the exponential memory, focusing on various kinds of non-Gaussian noise including radioactive, Poisson, and Abel noise. They touched upon algebraic memory using fractional derivatives but assumed zero dissipation in their analysis, i.e., $\gamma = 0$. In a more recent study, they analyzed stationary properties of Langevin equations with memories of the algebraic and delay type [4] as did Drozdov [5] using functionals to characterize various noise distributions. There is also an intriguing report in the literature by Fiscina et al. [6]

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of related ideas to the behavior of vibrated granular material: they found that the observed asymptotic spectral density could be well described using a Langevin equation with fractional derivatives. Work exploring the dynamic behavior of a non-Markoffian Langevin equation was reported by Bolivar [7] for arbitrary Gaussian noise correlation functions with a focus on the differentiability of the displacement. A path integral analysis of a Langevin equation with exponential memory has also appeared [8]. Fractional derivatives have been used [9] to introduce what are in essence memory effects into fractional probability density equations. General expositions of the subject that have been highly useful for a number of years are available in various articles [10–13].

As is well-known, stochastic analysis may be undertaken either via Langevin equations or via what has often been called a formalism based on Fokker-Planck equations. The former are ordinary differential equations for stochastic variables. The latter are partial differential equations for the deterministic probability density of those variables, an average of the stochastic variables carried out with the help of the probability density leading to expectation values. In the present paper, except for a discussion leading into the problem as well as an interesting observation at the end of the analysis, we will not use Fokker-Planck equations. A number of subtleties and controversies appear in the discussion of Fokker-Planck equations and we plan to address them in a separate publication. Our focus in the present paper will be *entirely* on the Langevin approach: the ordinary differential equation for the basic stochastic variable under consideration is solved explicitly for the given memory and noise, and the desired function of the variable, for instance an arbitrary power, is averaged taking into account the properties of the noise.

Thus, our general interest is in treating systems in which the Langevin equation for the coordinate $x_i$ of the $i$th particle is

$$\frac{dx_i(t)}{dt} = -\gamma \int_0^t dt'\, \phi(t-t')x_i(t') + \xi_i(t), \qquad (2)$$

where $\xi_i(t)$ denotes the noise and $\phi(t)$ is the memory function. The case when the memory is a $\delta$-function, i.e., when the Langevin equation is time-local, corresponds to equation (1). Our specific interest is in two forms of the memory: algebraic, i.e., power law, and incorporating a delay, as we will explain below.

One example of how non-local (in time) Langevin equations may come about in physical processes is provided by a scenario in which one refrains from making the high-damping approximation in the Langevin equation. Normally, as a result of Newton's law, the latter is a *second* order equation for the time dependence of the particle coordinate $x$. If the damping is very strong, the normal practice is to reduce it to a first order equation by neglecting the inertial term. If the inertial term is kept intact, both variables, the coordinate $x$ and the velocity $v$, are typically treated on an equal footing. An alternative, viable when observables dependent only on the coordinate (and not the velocity) are of interest, is to stick to a one-variable description but to introduce a memory function relating the time derivative of $x$ to an appropriate function of $x$. Such a situation has been discussed in a recent review of the mathematics of animal motion by two of the present authors [14]. The origin of the memory function or of time-nonlocality would be, in this case, the incorporation of inertial terms. A quite different source of time-nonlocality is finiteness of the speed of propagation of signals that are related to the attraction process. Examples may be found in delay formalisms as in a study of Alzheimer walks [15] and a recent analysis of pairwise movement coordination [16] applicable, e.g., to a system of foraging bats [17].

Although we will not use Fokker-Planck equations for our analysis, we will begin our considerations with the Smoluchowski equation (1). We do this because it is easy to introduce our task in that manner and also because there has been a lot of recent activity on that equation. Thus, an analysis has shown [18] how the Smoluchowski equation may be applied to trapping situations and a recent application to the spread of epidemics has been made [19] leading to an interesting description of the transmission of infection in diseases such as the Hantavirus [20].

The rest of the paper is laid out as follows. We first show in the next section why an intuitively natural memory generalization of an equation such as equation (1) does not work. We place our full focus therefore on the Langevin equation with memory. We are generally interested in biological processes far from thermodynamic equilibrium. The near-equilibrium requirement of fluctuation-dissipation relations between the noise and the memory then need not apply. To facilitate calculations we take the noise to be white. Although this restriction is not essential, we limit ourselves to this case on one hand because of its general usefulness in providing insights into a variety of biological systems [21] ranging from physiological [22,23] to ecological scales [24], and on the other hand because of the already rich scenario that we uncover even when the noise is not colored. In Sections 3 and 4 respectively, we obtain explicit usable prescriptions, specifically for algebraic and delay-type memories in the Langevin equation. We find the Green function for the case of an algebraic memory to be the Mittag-Leffler function. In the case of a single delayed delta function, it is associated with the Lambert function. A regime in which there is a monotonic decrease on the one hand and decaying oscillations on the other is found in each of the two Green functions. This is precisely the behavior shown by the simple and well understood case, for instance for a damped harmonic oscillator, when the memory is an exponential.

We discuss in Section 5, two applications of our results in the context of two specific experiments that could be performed on our system in principle. The first is about motional narrowing in the frequency-dependent susceptibility of our system wherein the particle is charged and a time-periodic electric field is applied. The second queries the steady-state size of the system as measured by the mean square displacement of the particle around its fixed

center under the combined action of the time-nonlocal attraction and the diffusion. Comparison of the results for the three memories considered, the algebraic, the single delay, and the simple exponential, brings out interesting similarities and differences. In the discussion which constitutes Section 6, we analyze a natural but incorrect generalization of the Smoluchowski equation yet find the remarkable result that its predictions are correct in the context of the first of the two envisaged experiments.

Contexts in which the analysis presented in this paper should find applicability are, generally speaking, biological systems far from equilibrium. These include movement of animals having a preference to places visited in the past [25] and various versions of what has been called in the mathematical literature 'self-reinforced random walks' as are met in Alzheimer-related investigations [26]. The complexity of the biological nature of the systems involved necessitates a memory description at the Langevin level and the fact that we are not necessarily near equilibrium suggests that a fluctuation-dissipation relationship between the memory and the noise need not apply making our simplification of Gaussian noise a useful first step in the analysis.

## 2 Viable route for the description of memory effects

The temptation to generalize the Smoluchowski equation (1), to incorporate memory effects present in its Langevin equation as in (2), by making (1) *itself* nonlocal in time is natural but meets with the following problem. Let us, in addition to the coordinate $x_i(t)$, define the velocity of the $i$th particle at time $t$ as $v_i(t) = dx_i(t)/dt$, the latter being given as a stated function of $x_i(t)$. The obvious manner of transforming from a particle description to a field description in the space of the field variable $x$ is to begin with the microscopic definitions of the (probability) density $P(x,t)$ and the current density $j(x,t)$,

$$P(x,t) = \sum_i \delta(x - x_i(t)),$$

$$j(x,t) = \sum_i \frac{dx_i(t)}{dt}\delta(x - x_i(t)),$$

and to obtain the continuity equation as a consequence:

$$\frac{\partial P(x,t)}{\partial t} + \frac{\partial j(x,t)}{\partial x} = 0. \tag{3}$$

One then expresses the $x$-derivative of $j(x,t)$ by replacing $x_i(t)$, wherever it occurs, by $x$ given that $\delta(x - x_i(t))$ is present as a multiplying factor. This allows us to combine the continuity equation with the specific relation between $v_i(t)$ and $x_i(t)$ that forms the constitutive relation, and thereby to obtain the Smoluchowski equation. Specifically, for the simple time-local case in the absence of noise,

$$v_i(t) \equiv \frac{dx_i(t)}{dt} = -\gamma x_i(t). \tag{4}$$

Needless to say, this is the high-friction (sometimes referred to as the Aristotelian) limit corresponding to the time-local approximation to the Langevin equation. If standard procedures [27] are now used to derive the diffusion term from the noise in the Langevin equation, one obtains (1). This description may be found for instance in the textbook by van Kampen [28].

However, if the Langevin equation has memory effects as in equation (2), $x_i(t)$ cannot be replaced by $x$ in the $t$-nonlocal velocity equation (2). The connection is to values of $x$ occupied by the $i^{th}$ particle at all times in the past. It is then impossible to replace $x_i(t')$ for all $t'$ by $x$: the delta-functions in $x$ do not allow $x_i(t)$ to be replaced by $x$. This technical failure to arrive at non-Markoffian field equations is a direct consequence, inevitable at least at this level, of the memory nature of the interaction.

A viable route to the description of memory effects lies, however, in refraining from a generalization of equation (1) and in restricting one's attention entirely to the Langevin approach. There are in the literature a number of discussions centered on probability density evolution [29–32]. For instance, for the specific case of Gaussian white noise, a Fokker-Planck like equation of the Smoluchowski form results. San Miguel and Sancho [32] address a system consisting of a Brownian harmonic oscillator with finite inertia. Involved is a second-order linear Langevin equation subject to an additive Gaussian stochastic forcing. Although their derivation is specifically performed for the Brownian harmonic oscillator and results in an exponentially decaying memory kernel,

$$\phi(t) = be^{-bt}, \tag{5}$$

it can be easily generalized to any arbitrary linear memory kernel. The $b$ in equation (5) obeys $\gamma b = \omega^2$, the square of the oscillator frequency.

Let us avoid the probability density treatments and start from equation (2). Since there are no inter-particle interactions, we consider a single particle and drop the label $i$ without loss of generality. Let $\lambda(t)$ be the Green function of the homogenous (without noise) part of equation (2). An immediate consequence of equation (2) is

$$\widetilde{\lambda}(\epsilon) = \frac{1}{\epsilon + \gamma\widetilde{\phi}(\epsilon)} \tag{6}$$

in the Laplace domain: tildes denote Laplace transforms and $\epsilon$ is the Laplace variable. This result leads to the solution of equation (2) in the time domain as

$$x(t) = \lambda(t)x(0) + \int_0^t dt'\lambda(t-t')\xi(t'). \tag{7}$$

From here onwards, we only consider systems in which the noise $\xi(t)$ has zero mean, $\langle\xi(t)\rangle = 0$, and is white, which means that $\langle\xi(t)\xi(s)\rangle = 2D\delta(t-s)$ where the constant $D$ describes the strength of the noise. Expectation values of arbitrary powers of $x$ at a specified time can be calculated explicitly, making the reasonable additional assumption

that the noise is uncorrelated also with the initial value of the observable. The result for the average displacement and average square of the displacement is:

$$\langle x(t)\rangle = x_0\lambda(t), \tag{8}$$

$$\langle \Delta x^2(t)\rangle = \langle x(t)^2\rangle - \langle x(t)\rangle^2 = 2D\int_0^t ds\lambda^2(s), \tag{9}$$

where we take the (localized) initial condition as $x(0) = x_0$. In light of the fact that equation (2) already has $t = 0$ as a special instant at which the memory is initialized, we observe that there are now two, generally different, times 0 and $t_0$, the latter being the time at which $\langle x(t_0)\rangle$ is first measured, e.g., the initial observation time. For the sake of simplicity, we have used $t_0 = 0$; in general the two times may well be different. For non-Markoffian processes, the general case for which the two times are different leads to interesting subtleties which we do not analyze in the present work. In the second line we have displayed the difference of the average of the square of the displacement and the square of its average. We represent it by the symbol $\langle \Delta x^2\rangle$ and refer to it, rather than to $\langle x^2\rangle$, as the mean square displacement (MSD). Expectation values of two-time quantities such as the correlation function $\langle x(t)x(s)\rangle$ can also be obtained straightforwardly:

$$\langle x(t)x(s)\rangle = x_0^2\lambda(t)\lambda(s) + 2D\int_0^s dt'\lambda(t-t')\lambda(s-t'), \tag{10}$$

$$\begin{aligned} f(t,s) &\equiv \langle x(t)x(s)\rangle - \langle x(t)\rangle\langle x(s)\rangle \\ &= 2D\int_0^s dt'\lambda(t-t')\lambda(s-t'). \end{aligned} \tag{11}$$

The above results do *not* require that the noise be Gaussian. If, however, it is known to be Gaussian we can also write, for arbitrary powers of the displacement,

$$\langle x^n(t)\rangle = \begin{cases} \sum_{m=0}^{p} \frac{(2p)!}{(2m)!(p-m)!}\left\langle x_0^{2m}\right\rangle \lambda^{2m}(t) \\ \quad \times \left(D\int_0^t ds\lambda^2(t)\right)^{p-m} & \text{even } n\ \left(p \equiv \frac{n}{2}\right), \\ \sum_{m=0}^{p} \frac{(2p+1)!}{(2m+1)!(p-m)!}\left\langle x_0^{2m+1}\right\rangle \lambda^{2m+1}(t) \\ \quad \times \left(D\int_0^t ds\lambda^2(t)\right)^{p-m} & \text{odd } n\ \left(p \equiv \frac{n-1}{2}\right), \end{cases}$$

and thereby solve the entire problem on the basis of the Gaussian property.

## 3 Algebraic memories in the Langevin equation

The family of algebraic functions provides a useful case study of the class of memories that *cannot* be approximated via a Markoffian procedure. The latter means the replacement of $\phi(t)$ for long times by a delta-function in $t$ of strength $\int_0^\infty dt\phi(t)$. We consider such memories as our starting point for the present section:

$$\phi(t;\nu) = \frac{\alpha(\alpha t)^{\nu-1}}{\Gamma(\nu)}. \tag{12}$$

Here $\alpha$ is a positive constant with units of inverse time and $\Gamma(\nu)$ provides the appropriate normalization. We analyze the Green function $\lambda(t)$.

The Laplace transform[1] of equation (5), and insertion into equation (6), gives the Laplace domain Green function as,

$$\widetilde{\lambda}(\epsilon;\nu) = \frac{1}{\epsilon + \frac{\gamma\alpha^\nu}{\epsilon^\nu}} = \frac{\epsilon^\nu}{\epsilon^{\nu+1} + \gamma\alpha^\nu} = \frac{1}{\epsilon\left(1 + \frac{\gamma\alpha^\nu}{\epsilon^{\nu+1}}\right)}, \tag{13}$$

which is the Laplace-domain representation of the Mittag-Leffler function of one parameter, written in usual notation as $E_{\nu+1}(-\gamma\alpha^\nu t^{\nu+1})$ [33]. In the time domain, setting $\gamma\alpha^\nu \equiv \zeta^{\nu+1}$, this results in the series,

$$\lambda(t;\nu) = \sum_{n=0}^{\infty} \frac{\left[-(\zeta t)^{(1+\nu)}\right]^n}{\Gamma(n(1+\nu)+1)}. \tag{14}$$

This expression is derived through a binomial expansion of the denominator in equation (13). One obtains a formal series in increasing powers of $(\zeta/\epsilon)^{1+\nu}$. A term-by-term inverse Laplace transform of this formal series results in equation (14) for all $\nu > -1$. The resulting series converges for all finite times.

For the parameter range of interest, the Green function we have calculated shows three interesting types of behavior. The first is an overdamped decay, $\nu \in (-1, 0)$, the second is underdamped oscillations, $\nu \in (0, 1)$, and the third unstable oscillations, $\nu \in (1, \infty)$. We depict $\lambda(t)$ for the cases of overdamped decay (left) and underdamped oscillations (right) in Figure 1 over 16 dimensionless time units $\zeta t$. The overdamped regime exhibits sharper initial decays, but longer tails, as the value of $\nu$ is made more negative. Both, the amplitude of oscillation and the time for which they persist, increase for increasing $\nu$ in the underdamped regime. The value of $d\lambda(t)/dt$ at $t = 0$ changes discontinuously when $\nu$ approaches 0 from either direction. For positive values of $\nu$ it vanishes. For negative values it tends to infinity. This behavior is sharply different from that in the case of the simple exponential memory characteristic of the damped harmonic oscillator. For the latter, $d\lambda(t)/dt$ at $t = 0$ always vanishes. Not shown are the unstable oscillations for values of $\nu > 1$. Three special values exist. For $\nu = -1$, we have standard Brownian diffusion, i.e., a Wiener process or an unconfined random walk. For $\nu = 0$, we have the standard Smoluchowski equation, the Ornstein-Uhlenbeck process. For $\nu = 1$, we have pure oscillations with no damping.

[1] The Laplace transform of (12) only exists for $\nu > 0$. However, the form of the Laplace-domain noiseless Green function, (13), suggests extending the domain of validity to $\nu \geq -1$. In this range, the Green function is unity when $t = 0$ (except for $\nu = -1$ where $\lambda(t) = 1/2$).

$$\lambda(t;\nu) = -\frac{\sin\nu\pi}{\pi}\int_0^\infty dr e^{-r\zeta t}\frac{r^\nu}{r^{2(\nu+1)} - 2r^{\nu+1}\cos\nu\pi + 1} + \begin{cases} 0 & -1 < \nu \le 0 \\ \frac{2}{\nu+1} e^{-\zeta t\cos\frac{\nu\pi}{\nu+1}}\cos(\zeta t\sin\frac{\nu\pi}{\nu+1}) & 0 < \nu \le 2 \\ \frac{2}{\nu+1}\left[e^{-\zeta t\cos\frac{\nu\pi}{\nu+1}}\cos(\zeta t\sin\frac{\nu\pi}{\nu+1}) + e^{-\zeta t\cos\frac{3\nu\pi}{\nu+1}}\cos(\zeta t\sin\frac{3\nu\pi}{\nu+1})\right] & 2 < \nu \le 4 \end{cases} \quad (15)$$

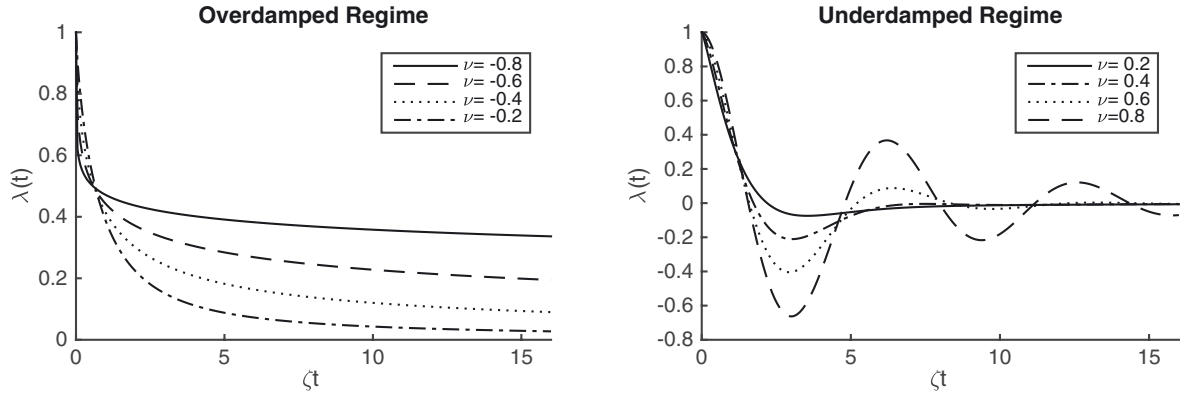


**Fig. 1.** The noiseless Green function, $\lambda(t)$, for varying $\nu$ in equation (14) over the overdamped (left) and underdamped (right) regimes in the range $[-0.8, -0.2]$ and $[0.2, 0.8]$ in steps of 0.2. Time is plotted in units of $1/\zeta$. As $\nu$ is made more negative, $\lambda(t)$ approaches 0 more slowly. As $\nu$ is made more positive, the amplitude of the oscillations increases and they last longer.

A second representation of $\lambda(t)$ is found by explicitly closing the Bromwich contour. For all non-integer values of $\nu$, (13) has at least two singularities of interest: branch points at zero and infinity, connected by a branch cut chosen to be along the negative real axis. This choice of branch cut is made to restrict the domain of $\epsilon$ to the Riemann sheet with $|\arg(\epsilon)| < \pi$. Additionally, simple poles exist for all relevant values of $\nu$ at the points $\ln(\epsilon) = \pm i(1+2m)\pi/(\nu+1)$ where $m$ is an integer greater than zero. However, not all fall on the relevant Riemann sheet. When $\nu < 0$, there are no additional simple poles. As $\nu$ passes through each successive even integer, two additional poles move on to the relevant Riemann sheet. Therefore, the Bromwich integral of equation (13) can be performed to quadrature and results in

*see equation (15) above*

where $\lambda(0;\nu)$ equals 1 for all $\nu$. The integral in equation (15) is the Laplace transform of a positive definite function and, for non-integer values of $\nu$, is therefore non-negative at all times. As mentioned previously, additional exponential terms become relevant as $\nu$ is increased further.

We have given two separate representations of $\lambda(t)$ in the time domain: the series, (14), and the integral expression, (15). These are compared in Figure 2, over a range of approximately 30 dimensionless time units for 2 values of $\nu$ in each regime, $\pm 0.1$ and $\pm 0.9$. The two representations match up well over shorter time periods. However, at longer times, the numerical implementation of the series leads to divergent results as a consequence of round-off errors.

The integral in equation (15) is not reducible in terms of known functions for arbitrary $\nu$. For the particular case of $\nu = m + 1/2$, where m is an integer, a transform of $u = r^{1/2}$ simplifies the integrand to

$$(-1)^{m+1}\frac{2}{\pi}\int_0^\infty du e^{-u^2\zeta t}\frac{u^{2(m+1)}}{u^{2(2m+3)}+1}.$$

The denominator of this integral is easily factorable into the $(2m+3)$th roots of 1. This results in the standard integral representation of the Faddeeva function, $w(iz) = \mathrm{erfcx}(z)$, which corresponds to the scaled error functions with complex arguments [34]. We give here the $\nu = -1/2$ $(m = -1)$ and $\nu = 1/2$ $(m = 0)$ cases:

$$\lambda\left(t;-\frac{1}{2}\right) = e^{\zeta t}\mathrm{erfc}\left((\zeta t)^{\frac{1}{2}}\right), \quad (16)$$

$$\lambda\left(t;\frac{1}{2}\right) = e^{-\frac{\zeta t}{2}}\cos\frac{3^{\frac{1}{2}}\zeta t}{2} + w\left(i(\zeta t)^{\frac{1}{2}}\right) - w\left(i\left(\zeta t e^{\frac{i2\pi}{3}}\right)^{\frac{1}{2}}\right) - w\left(i\left(\zeta t e^{-\frac{i2\pi}{3}}\right)^{\frac{1}{2}}\right). \quad (17)$$

The long-time behavior of the Mittag-Leffler function, valid for non-integer values of $\nu$ in the region $(-1, 1)$, is well-known [33],

$$\lambda(t\to\infty;\nu) = -\sum_{n=1}^{p}\frac{1}{\Gamma[1-n(\nu+1)]}\left[\frac{-1}{(\zeta t)^{(\nu+1)}}\right]^n + O\left(t^{-p(\nu+1)}\right). \quad (18)$$

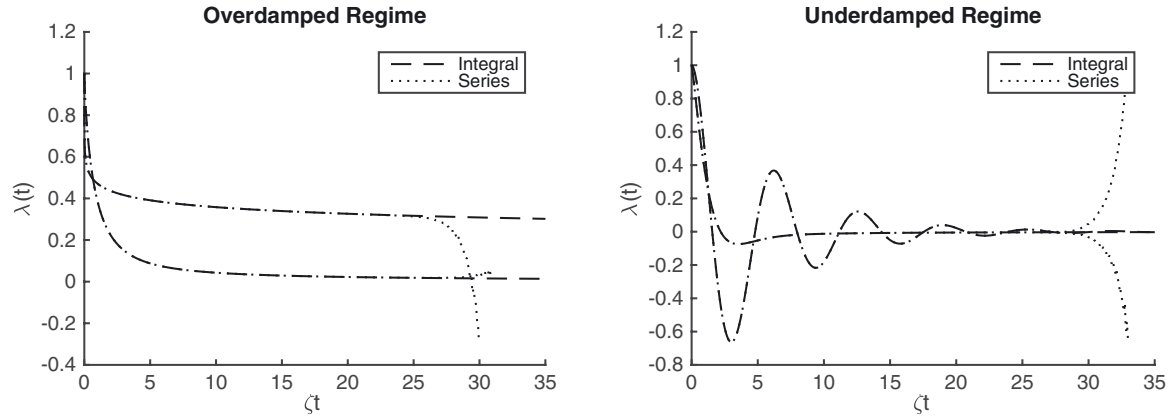


**Fig. 2.** Depicts the integral representation and the series representation of the Green function, $\lambda(t)$, over short times for the overdamped regime (left) and the underdamped regime (right) for $\nu = \pm 0.2, \pm 0.8$. The series representation is indicated by dots and the integral representation by solid lines. At approximately 30 dimensionless time units $\zeta t$, the series begins to diverge as a result of numerical round-off results.

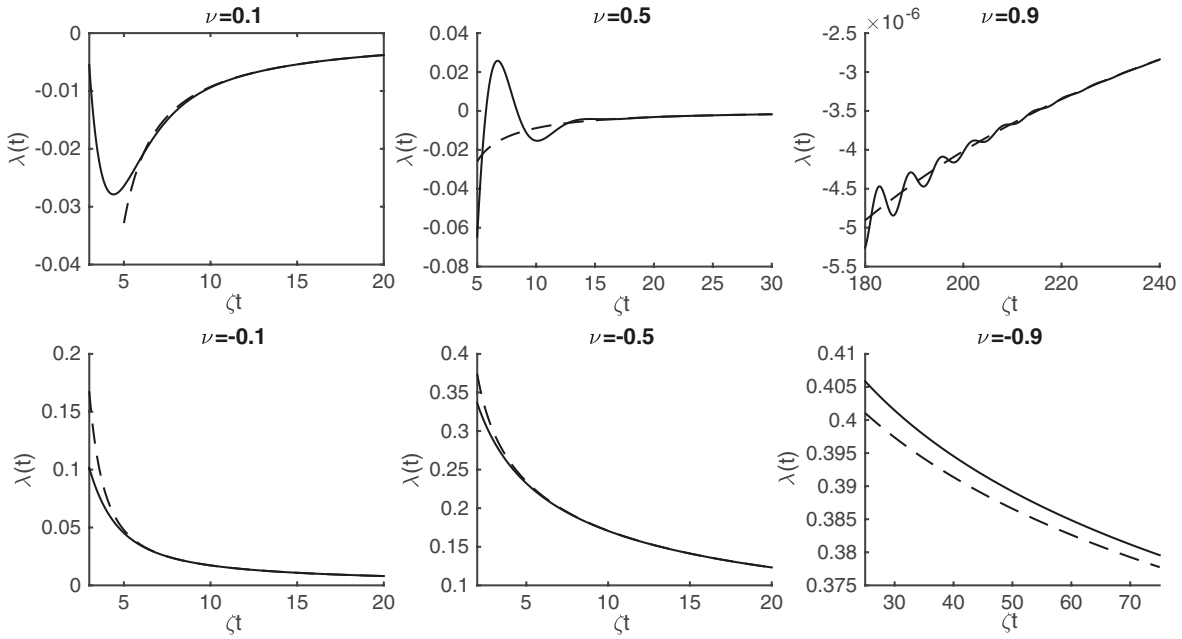


**Fig. 3.** Comparison at long times of the integral expression, equation (15), of the noiseless Green function (solid line) with its series approximation (dashed line), equation (18). The latter consists of the first 5 terms for 3 values of $\nu$, 0.1 (left), 0.5 (center), and 0.9 (right) over differing time ranges.

We see that an algebraic memory results in an algebraic time-dependence of $\lambda(t)$ at long times. The dominant term in the series, proportional to $1/(\zeta t)^{1+\nu}$, leads to a decay which is stronger when $\nu$ is larger. In the underdamped regime, the leading term of (18) is negative. Therefore, at long times, $\lambda(t)$ approaches zero from below, confirming the existence of at least one minimum. This term is positive for the overdamped regime. The correspondence between the long-time approximation, equation (18) with 5 terms, and the full propagator is depicted in Figure 3 for 6 values of $\nu$: $\pm 0.1$, $\pm 0.5$, $\pm 0.9$. The long-time approximation does not lead to oscillations, rather to an overall decay towards 0. For smaller values of $|\nu|$, the approximation becomes valid at earlier times.

Thus, the algebraic memory leads to a noiseless Green function that corresponds exactly to the Mittag-Leffler function. Three separate regimes emerge: overdamped decay, $-1 < \nu \leq 0$, underdamped oscillations, $0 < \nu < 1$, and unstable oscillations, $\nu \geq 1$.

## 4 Memories that represent delay processes

We now focus on the particular case of a memory which selects only one time $\tau$ in the past via a Dirac-delta centered at $\tau$. In other words,

$$\phi(t) = \delta(t - \tau). \tag{19}$$

To avoid dealing with a piece-wise process, i.e. Wiener dynamics for $0 \leq t < \tau$, and delayed dynamics for $t \geq \tau$, we extend the range of integration in the Langevin equation (2), back in time to $-\tau < 0$ and consider,

$$\frac{dx(t)}{dt} = -\gamma \int_{-\tau}^{t} ds\, \phi(t-s)x(s) + \xi(t), \qquad (20)$$

such that at time $t = 0$ the evolution of $x$ is now dependent on the history of $x$ between $t = -\tau$ and $t = 0$. After inserting $\phi(t) = \delta(t-\tau)$ in equation (20) one recognizes that the system is governed by the stochastic delay Langevin equation,

$$\begin{aligned} \frac{dx(t)}{dt} &= -\gamma x(t-\tau) + \xi(t), \qquad t > 0 \\ x(t) &= \Phi(t), \qquad -\tau \leq t < 0, \end{aligned} \qquad (21)$$

where $\Phi(t)$ is a given deterministic history function. To solve this stochastic delay differential equation we use the Laplace transform and obtain [35]

$$\begin{aligned} x(t) = x(0)\lambda(t) + \int_0^t ds\, \lambda(t-s)\xi(s) \\ - \gamma \int_{-\tau}^{0} ds\, \lambda(t-s-\tau)\Phi(s), \end{aligned} \qquad (22)$$

which is formally equivalent to equation (7), with the additional term representing the history-dependence. In equation (22) the Laplace transform of the Green function $\lambda(t)$ is given by,

$$\tilde{\lambda}(\epsilon) = \frac{1}{\epsilon + \gamma e^{-\epsilon\tau}}. \qquad (23)$$

We now turn to the analysis of single delay processes.

The analytic calculation of $\lambda(t)$ from equation (23) has been performed independently or repeated by a number of authors [15,16,36–38] following different methods. Using Cauchy's residue theorem one can write $\lambda(t)$ as:

$$\lambda(t) = \sum_{\text{Res}} \frac{e^{\epsilon t}}{\epsilon + \gamma e^{\epsilon\tau}}, \qquad (24)$$

where the summation is over the residues. The poles of $\tilde{\lambda}(\epsilon)$ are the roots, $\eta$, of the characteristic equation, $\eta + \gamma e^{-\eta\tau} = 0$, which can be written in the form $\eta\tau e^{\eta\tau} = -\gamma\tau$. This latter transcendental equation corresponds exactly with the inverse relationship, $W(z)e^{W(z)} = z$, defining the so-called Lambert function, $W(z)$ [36,39]. By direct comparison we see that the roots of the characteristic equation for the single delay process are given by the multivalued Lambert function,

$$\eta = \frac{1}{\tau} W(-\gamma\tau). \qquad (25)$$

Given the well documented computational procedures for evaluating the Lambert function [40], $\lambda(t)$ can be computed with sufficiently high precision. The roots $\eta$ of the characteristic equation provide information about the evolution and stability of the system at long times. It is known [35] that the system decays monotonically for $0 \leq \gamma\tau < e^{-1}$, undergoes oscillatory decay for $e^{-1} \leq \gamma\tau < \pi/2$, and performs unstable oscillations for $\gamma\tau > \pi/2$. At long times the eigenvalue with the largest real part, corresponding to the principal branch of the Lambert function $W_0$, dominates the behavior of the system. In the monotonic regime ($\gamma\tau < e^{-1}$), this principal branch can actually be evaluated analytically [40] via $W_0(-\gamma\tau) = -\sum_{n=1}^{\infty} n^{n-1}(\gamma\tau)^n/n!$.

Whilst the expression for $\lambda(t)$ in equation (24) is exact and provides insights into the long time behavior of the system, it is of limited use for studying the dynamics of the system at shorter times because of the large number of eigenvalues required. An alternative expression can be derived for $\lambda(t)$ by expanding (23) as a power series and performing the Laplace inversion to give the following expression [15,16,37,38],

$$\lambda(t) = \sum_{k=0}^{\infty} \frac{(-\gamma)^k}{k!} (t - k\tau)^k \Theta(t - k\tau), \qquad (26)$$

where $\Theta$ represents the Heaviside step function.

The alternative exact expression (26) requires only a finite number of terms for any finite time $t$ with a functional dependence given by a polynomial of degree $k$ in each interval $k\tau \leq t \leq (k+1)\tau$. Expansion of the sum in equation (26) shows in fact explicitly that $\lambda(t) = 1$ for $t \in [0, \tau]$, and $= 1 - \gamma(t-\tau)$ for $t \in [\tau, 2\tau]$, and $= 1 - \gamma(t-\tau) + \alpha^2(t-2\tau)^2/2$ for $t \in [2\tau, 3\tau]$, and so on. From these polynomials we can study properties of the Green function, such as the time at which it first reaches zero, denoted $t_0$, when the system is in the oscillatory regime. The Green function is constant on the first interval and thus clearly $t_0 > \tau$. The Green function on the second interval, when $\tau < t_0 \leq 2\tau$, has one root at $t_0 = \tau + 1/\gamma$, which corresponds with the first zero crossing when $\gamma\tau \geq 1$. This implies that the first zero is found in the second interval for the entirety of the unstable regime ($\gamma\tau \geq \pi/2$), and roughly half of the oscillating regime ($\gamma\tau \geq e^{-1}$). For other parameter values in most cases one has to find the roots of the higher order polynomials numerically. Examples of the different regimes for $\lambda(t)$ are shown in Figure 4. Note also that, as $\tau \to 0$, we recover the Ornstein-Uhlenbeck process (no memory effects in the attraction to the center) and, as $\gamma \to 0$, we have the Wiener process, i.e., a standard walk with no confinement.

## 5 Application to experiments and comparison of consequences of different forms of the memory

With an application of our formalism in mind, to two experiments possible in principle, one on the motional narrowing of spectral lines and the other on the spatial extension of the particle in the steady-state, we now compare

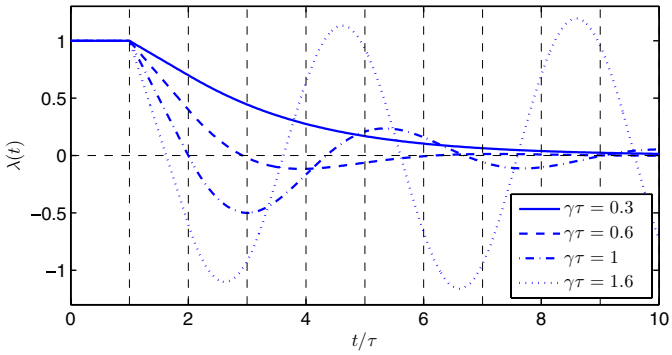


**Fig. 4.** The noiseless Green function $\lambda(t)$ for the single delay process. Four choices of the parameter $\gamma\tau$ are considered, representing the stable non-oscillatory, stable oscillatory and unstable oscillatory regimes. The first zero crossings of the oscillatory curves occur in the second interval for $\gamma\tau \geq 1$, specifically at $t_0/\tau = 1.625$ and 2 for $\gamma\tau = 1.6$ and 1.0, respectively. In contrast, the oscillatory curve with $\gamma\tau = 0.6$ first crosses the zero in the third interval.

our predictions for the three cases of algebraic, single delay, and exponential memory. The latter, given by equation (5), has appeared not only in the damped harmonic oscillator treated by the authors of reference [32], but in numerous contexts, some to describe transport with an arbitrary degree of quantum mechanical coherence [41–43]. Although its introduction in the latter references has occurred in the context of site-to-site motion of a quasiparticle, rather than of attraction towards a center as in the present paper, many expressions obtained earlier, and much of the intuition, can be ported over here. The Green function $\lambda(t)$ is well-known to be given by the simple expression

$$\lambda(t) = e^{-bt/2}\left[\cos \Omega t + (b/2\Omega)\sin \Omega t\right], \qquad (27)$$

where $\Omega = \sqrt{\omega^2 - b^2/4}$. Standard features well-known from other fields of study, ranging from quantum quasiparticle transport to RLC circuits in electrical networks, are easily noticed in equation (27): the original undamped oscillator frequency $\omega$, its reduction to $\Omega$ when damping is introduced via the damping exponent $b/2$, the passage from the damped oscillatory regime to the overdamped regime when $b/2 > \omega$ when the trigonometric functions change into their hyperbolic counterparts leading to familiar phenomena such as motional narrowing of spectral lines.

### 5.1 Motional narrowing of spectral lines

We envisage two experiments that could be performed, in principle, on our system. In one, our particles are charged (but noninteracting among themselves), we apply a time-varying electric field and measure the polarization and therefore the susceptibility. Thus, equation (2), with the label $i$ suppressed, is augmented by an appropriate term to give

$$\frac{dx(t)}{dt} = -\gamma \int_0^t dt'\, \phi(t-t')x(t') + \mathcal{E}(t) + \xi(t). \qquad (28)$$

Our interest lies in measuring the frequency-dependent susceptibility which is the ratio of the Fourier transforms of the polarization and the applied electric field. The term $\mathcal{E}(t)$ is essentially the electric field and absorbs unimportant proportionality constants. The spectral line at frequency $f$ is proportional to $\frac{\langle \hat{x}(f)\rangle}{\hat{\mathcal{E}}(f)}$ where the circumflexes denote Fourier transforms and $f$ is the frequency. We use $f$ rather than the more usual $\omega$ to distinguish it from the oscillator frequency that we have already used in our treatment.

A well-known phenomenon known in systems with a simple exponential memory, as in the case of a damped harmonic oscillator, is motional narrowing: spectral lines, sharp if the damping in the system is vanishing or small, broaden as the damping is increased but, after a critical value of the damping is exceeded, separate lines coalesce and increased damping *narrows* the line. The question we ask here is whether this motional narrowing occurs also for our algebraic and delay cases. One sees from equation (28) that what is required is the one-sided Fourier transform of the Green function $\lambda(t)$: the frequency-dependent susceptibility is proportional to $\frac{\langle \hat{x}(f)\rangle}{\hat{\mathcal{E}}(f)}$. From equation (27) for the exponential memory and equations (14) and (26) for the algebraic and single delay memories, respectively, we have:

$$\hat{\lambda}(f) = \begin{cases} \dfrac{\sqrt{f^2+b^2}}{\sqrt{(f^2-\omega^2)^2+f^2b^2}} & \text{Exponential,} \quad (29a) \\ \dfrac{1}{\sqrt{f^2-2f\tau\mu^2\sin f\tau+\tau^2\mu^4}} & \text{Single Delay,} \quad (29b) \\ \dfrac{(|f|)^\nu}{\sqrt{|f|^{2(\nu+1)}-2(|f|\zeta)^{\nu+1}\sin\frac{\pi\nu}{2}+\zeta^{2(\nu+1)}}} & \text{Algebraic,} \quad (29c) \end{cases}$$

where we have given the absolute value of the Fourier transforms. In equation (29b) we have introduced the coherence parameter $\mu$ for the delay case. It is analogous to $\omega$ in the exponential case and $\zeta$ in the algebraic case and is specifically defined via $\mu = \sqrt{\gamma/\tau}$.

We show the results of equation (29) in Figure 5 for the exponential (left), single delay (center) and algebraic (right) memories. Units along the vertical axis are arbitrary. The left panel depicts motional narrowing for the exponential memory: two peaks broaden and then coalesce into a single peak as the damping is increased. Similar transitions are seen for both the single delay memory (coherence measured by the value $\mu\tau$) and the algebraic memory (coherence measured by the value $\nu$). The single delay process is quite similar with the exception that it develops additional symmetric peaks, indicated by the arrows. The algebraic process, while also similar in the overall aspects, exhibits sharp differences for small values of $f$. The source of this peculiar behavior is the fact that the integral of $\lambda(t)$ over all time changes drastically as one crosses from the oscillatory to the monotonic region. The integral is 0 for positive $\nu$ and infinite for negative values of $\nu$.

The motional narrowing phenomenon [44] we have discussed above is ubiquitous and appears in magnetic resonance observations [45], neutron scattering

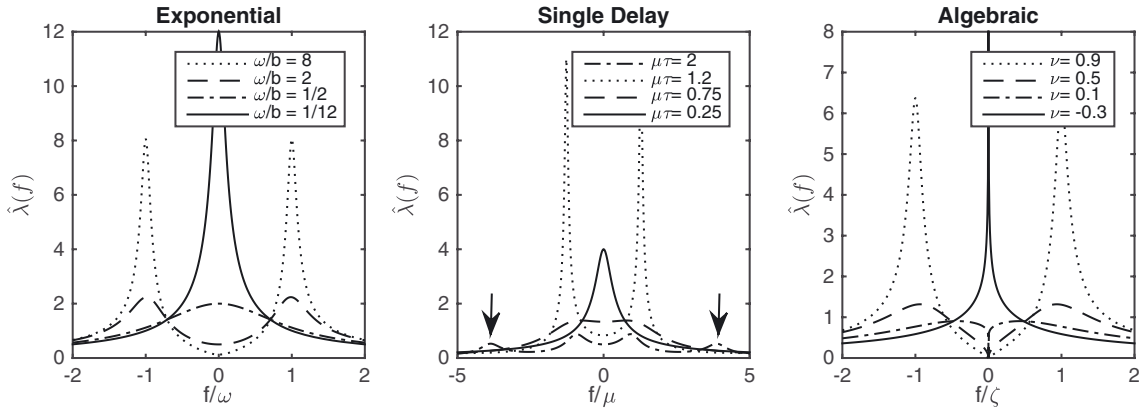


**Fig. 5.** Motional narrowing in the dependence of the a.c. susceptibility on the frequency, $f$, of the applied electric field. All three cases, exponential (left), single delay (center), and algebraic (right), display two peaks in the coherent limit which, as damping is increased, initially broaden more and more and move towards each other (to the center, i.e., the region $f = 0$). On increasing the damping beyond a critical value in each case, however, the line *narrows* rather than broadens as the damping is increased. Frequency is plotted on the horizontal axis in units of the coherent parameter $\omega$, $\mu$ and $\zeta$ for the three respective cases. Units along the vertical axis are arbitrary. Two arrows in the central panel locate the additional peaks that develop for the single delay process. The sharp transition at $f = 0$ for the algebraic memory, which is a jump from a vanishing to an infinite value, can be seen when comparing the $\nu = -0.3$ case with the $\nu = 0.1$ case.

experiments [46–51], and in numerous other contexts such as optical absorption whenever there is underlying dynamics of a system undergoing spectral diffusion. Such spectral diffusion can arise not only from thermal motion in an inhomogeneous medium as has been sometimes mentioned in the past but from a variety of sources including changes in the bath fluctuation rate [52]. The simple exponential memory case we have mentioned for the sake of comparison above can be seen to arise explicitly in the magnetic resonance context. This can be noted clearly in Appendix F of the text *Principles of Magnetic Resonances* by Slichter [45] where the exponential nature of the memory emerges simply on elimination of separate quantities $M_{\pm}$, summing them to get the total magnetization $M$. The original theory of motional narrowing was developed by Kubo [44], a number of cases for neutron scattering were treated for simple memories by other authors such as Brown and Kenkre [47–51] and a recent lineshape theory was given by Jung et al. [52] directed at the important observation of power-law statistics in spectral diffusion.

### 5.2 Spatial extent in the steady state

What is the spatial extent of the particle in the steady-state when the diffusive tendency to enhance it and the attraction to the center to reduce it have balanced themselves? A number of experimental techniques could be devised in principle to measure the spatial extent or size. The size in the steady-state is given by the saturation value of the MSD $\langle \Delta x^2 \rangle$. The time-dependent MSD for the exponential memory is given by,

$$\langle \Delta x^2 \rangle (t) a = \frac{D}{\omega}\left[\frac{\omega}{b} + \frac{b}{\omega} - \frac{be^{-bt}}{\omega}\left(\frac{\omega^4}{b^2\Omega^2} + \frac{4\Omega^2 - \omega^2}{4\Omega^2} \times \cos 2\Omega t + \frac{3\omega^2 - 4\Omega^2}{2b\Omega}\sin 2\Omega t\right)\right]. \quad (30)$$

Although any two of the three parameters $\omega$, $b$ and $\Omega$ uniquely determine the third, we have used all three here and elsewhere to avoid cumbersome square roots in the display. In the overdamped limit, i.e., when $b > 2\omega$, the above trigonometric functions turn into hyperbolic functions as $\Omega$ becomes imaginary.

The MSD for the case of the single delay is:

$$\langle \Delta x^2 \rangle (t) = 2D\left(\Theta(k)\sum_{l=0}^{k-1}\int_{l\tau}^{(l+1)\tau} ds\, g_l(\gamma s) + \int_{k\tau}^{t} ds\, g_k(\gamma s)\right), \quad (31)$$

where $g_k(t)$ is defined as:

$$g_k(\gamma t) = \sum_{m=0}^{k}\sum_{n=0}^{k}\frac{(-1)^{(m+n)}}{m!n!}(\gamma t - m\gamma\tau)^m(\gamma t - n\gamma\tau)^n,$$

in any interval, $k\tau \le t \le (k+1)\tau$.

The MSD for the algebraic case is given by,

*see equations (32a) and (32b) next page*

with

$$A = \frac{2}{\nu+1}, \qquad C(r) = \frac{r^\nu \sin\nu\pi}{r^{2(\nu+1)} - 2r^{(\nu+1)}\cos\nu\pi + 1},$$
$$\beta = \cos\left(\frac{\nu\pi}{\nu+1}\right), \qquad \kappa = \sin\left(\frac{\nu\pi}{\nu+1}\right).$$

Our interest lies in the steady-state size of the particle given by these expressions in the limit $t \to \infty$. Calling the saturation value of the MSD as the particle size $S$ (in units

$$\langle \Delta x^2 \rangle(t) = 2D \int_0^t ds \begin{cases} \left[ \int_0^\infty \frac{dr}{\pi} e^{-r\zeta s} C(r) \right]^2 & -1 < \nu \leq 0, \quad (32a) \\ \left[ A e^{-\beta\zeta s} \cos \kappa\zeta s - \int_0^\infty \frac{dr}{\pi} e^{-r\zeta s} C(r) \right]^2 & 0 < \nu \leq 2, \quad (32b) \end{cases}$$

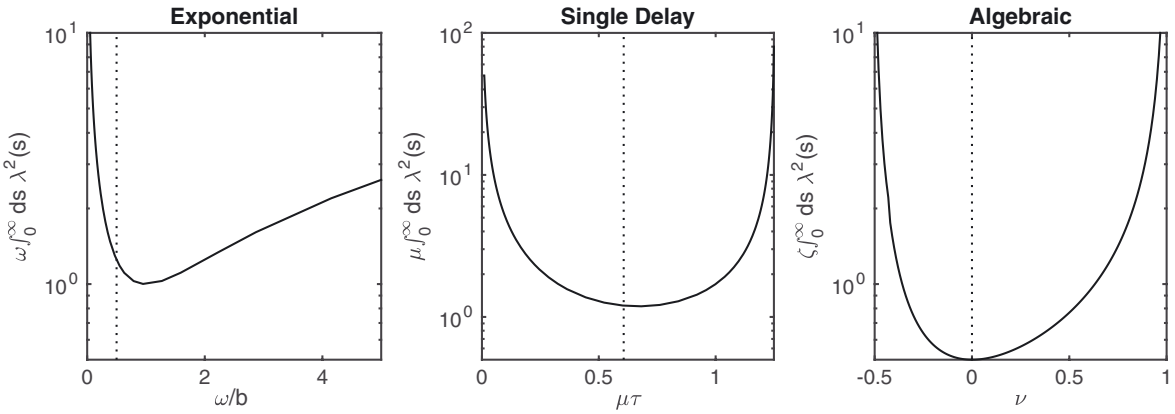


**Fig. 6.** Spatial extent of the particle in the steady-state, measured by the saturation value of the MSD at long times for: the exponential memory (left), the single delay memory (center) and the algebraic memory (right). On the vertical axis, the size is normalized to the steady-state size for the time-local case ($\phi(t) = \delta(t)$) with $\gamma$ equal to the respective coherent parameter: $\omega$ for the exponential memory, $\mu$ for the single delay memory and $\zeta$ for the algebraic memory. Dotted lines indicate the location of the transition from monotonic (overdamped) to oscillatory (underdamped) regimes.

of area for our 1-dimensional system), we have

$$S = \begin{cases} \frac{D}{\omega}\left(\frac{\omega}{b} + \frac{b}{\omega}\right), & (33a) \\ \frac{D}{\mu}\left(\frac{1+\sin\mu^2\tau^2}{\mu\tau\cos\mu^2\tau^2}\right), & (33b) \\ \frac{D}{\zeta}\int_0^\infty\int_0^\infty \frac{drdq}{\pi^2}\frac{2C(r)C(g)}{r+q}, & (33c) \\ \frac{D}{\zeta}\left[\frac{A^2}{2}\left(\beta+\frac{1}{\beta}\right) - 4A\int_0^\infty \frac{dr}{\pi}\frac{C(r)(\beta+r)}{r^2+2\beta r+1}\right. \\ \left. + \int_0^\infty\int_0^\infty \frac{drdq}{\pi^2}\frac{2C(r)C(q)}{r+q}\right], & (33d) \end{cases}$$

where (33c) is for $-0.5 < \nu \leq 0$ and (33d) is for $0 < \nu \leq 1$. For the parameter range $\nu \leq -0.5$, the MSD diverges as,

$$\lim_{t\to\infty} \langle \Delta x^2 \rangle(t) \propto \begin{cases} \ln t & \nu = -0.5, \\ t^{2|\nu|-1} & -1 \leq \nu < -0.5. \end{cases}$$

The MSD for the single delay memory, equation (33b), is known to have the analytic expression shown above. It is obtained by solving [38] the differential equation governing the evolution of the covariance at long times.

Plots of the long-time expressions in equation (33) are shown in Figure 6 for the exponential (left), single delay (center) and algebraic (right) memories, respectively. The MSD for all three memories are normalized using the respective coherent parameters: $\omega$, $\mu$ and $\zeta$. In all three cases, an increase in $D$ leads to a monotonic increase in the MSD. The exponential memory process has a symmetric dependence on $\omega$ and $b$, with a minimum for $b = \omega$. For the single delay, only in the stable regime, $\mu^2\tau^2 < \pi/2$, is the expression valid and, as expected, the MSD diverges as one approaches the unstable limit. Both for the exponential and the single delay memories, the saturation value of the MSD diverges as $\omega/b$ and $\mu\tau$, respectively, approach zero. In both these cases, the tendency to confinement around the attractive center disappears since $\gamma$ vanishes.

At long times, the MSD for the algebraic case diverges when $\nu \to -0.5$ from the right and when $\nu \to 1$ from the left. The divergence at the lower limit occurs due to the long-time algebraic dependence of the Green functions, equation (18). The value of $\nu$ for which the steady-state size is minimum depends on the ratio of time constants, $\gamma/\alpha$. When this ratio is equal to 1, the minimum is at $\nu = 0$. This is obvious in Figure 6. For an arbitrary $\gamma/\alpha$, the value of $\nu$ at the minimum is given by the transcendental equation,

$$\ln\frac{\gamma}{\alpha} = -(\nu_{\min}+1)^2 \frac{d}{d\nu}\ln F(\nu_{\min}), \quad (34)$$

where $F(\nu)$ is the functional form of the curve plotted in the right panel of Figure 6. An increase in the ratio of $\gamma/\alpha$ shifts the minimum rightwards, a decrease shifts it to the left.

A fruitful comparison of the MSD dynamics can be done by setting the parameters such that, $\varrho \int_0^\infty ds\, \lambda_d^2(s) = V$, where $\varrho$ is the appropriate coherent parameter, is identical for all three cases. The saturation value of the three memories are not comparable over the entire parameter space. The single-delay process has a minimum MSD saturation when $2\mu^2\tau^2 = \cos(\mu^2\tau^2)$, i.e., a value $V = 1.19$,

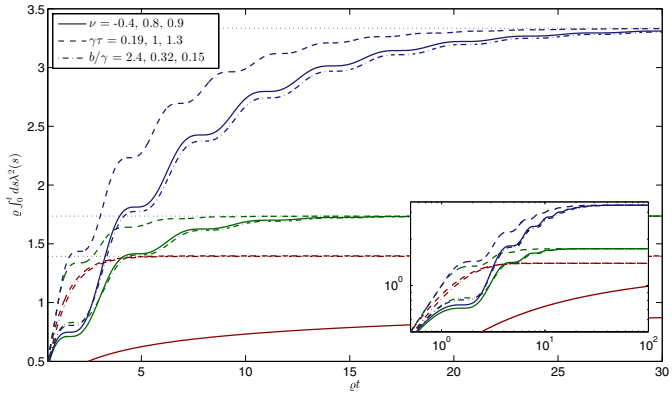


**Fig. 7.** Comparing the dynamics of the MSD for the algebraic (*straight line*), exponential memory (*dot-dashed line*) and single delay (*dashed line*) memories when their MSD have a mutual saturation value. We set $\varrho$ equal to the coherent parameter for each of the memories divided by the diffusion constant, i.e., $\omega$ for exponential, $\mu$ for the single delay, and $\zeta$ for the algebraic, respectively. Two curves for each memory depict dynamics in the underdamped regime and one for each memory in the overdamped regime. The inset plot shows that same figure with logarithmic axes.

while the exponential memory process has a minimum value, $V = 1$ exactly when $b/\gamma = 1$. For these parameter values, the algebraic memory has a minimum value of $V = 1/2$ located at $\nu = 0$.

We select values of $V > 1.19$ from the algebraic long-time MSD, equations (33c) and (33d), and solve the exponential and single delay memory expressions in equations (30) and (31) to obtain

$$\omega/b = V \pm \sqrt{V^2 - 1}$$

and

$$\sin(\mu^2\tau^2) = \left[(2V\mu\tau)^2 - 1\right] / \left[(2V\mu\tau)^2 + 1\right],$$

respectively. For the exponential memory we choose the negative branch, $\omega/b = V - \sqrt{V^2 - 1}$, for the monotonic regime and the positive branch, $\omega/b = V + \sqrt{V^2 - 1}$, in the oscillatory regime.

Using these values of $V$, $\omega/b$ and $\mu\tau$ we plot in Figure 7 the dynamics of the three memories. In the oscillatory regime, all three memories exhibit the apparent saturation behavior associated with the oscillations of their respective Green functions. The single delay memory clearly saturates the fastest. The exponential and algebraic memories have very similar MSD, with the exponential memory initially larger. In the overdamped regime, the algebraic memory saturates much slower due to its heavy tail.

## 6 Discussion

The focus of this paper has been the investigation of the dynamics of a Smoluchowski system whose Langevin equation (see, e.g. Eq. (2)) describes the attraction, to a fixed center, of a particle via a *memory function* corresponding to a time-nonlocal process. The case when the memory function, denoted by the symbol $\phi$ in the paper, is a simple delta function in time (time-local case), is textbook material and leads to solutions of the corresponding Smoluchowski equation via the Ornstein-Uhlenbeck time transformation. We have investigated here the case of two time-nonlocal memories of interest to certain current applications in biological systems: the algebraic memory, equation (12), and a delay case, equation (19), and compared them with a standard exponential memory, equation (5), whose consequences are typically known in the literature. We find similarities across the three cases as well as some distinguishing characteristics of each. The connection to the time-local case represented by $\phi(t) = \delta(t)$ is trivial in the case of the delay and the exponential cases because, in both instances, the integral of the memory over all time exists and equals 1. For the algebraic case this is not true and leads to some peculiar behavior. Such behavior and its analysis given in the present paper will no doubt be of importance when parameters controlling the non-Markoffian dynamics are strong. Even when they are weak, the algebraic nature of the memory will ensure that they must be generally taken into account.

### 6.1 Similarities and dissimilarities in the consequences of the three memories

In all three cases, $\gamma\phi(t)$ is convolved with the displacement $x(t)$ in the starting Langevin equation. The strength of the confinement to the attractive center may be said to be described by $\gamma$ and the specific (memory) manner in which the confining is done may be ascribed to $\phi(t)$. The loss of coherence occurs at the rate $b$ in the exponential case, at the rate $1/\tau$ equal to the reciprocal of the delay time in the single delay case, and at $\alpha$ in the algebraic case. The coherence parameter is $\omega$ in the exponential case, $\mu = \sqrt{\gamma/\tau}$ for the delay case and $\zeta = \sqrt{\gamma\alpha}$ in the algebraic case. The three respective memories are found in equations (5), (19) and (12).

The basic quantity that determines the behavior of the system is the Green function $\lambda(t)$. It is given generally in the Laplace domain by equation (6). In the time domain it takes the form given in equation (27) for the exponential memory, equation (26) for the delay memory, and equation (14) for the algebraic case. One of the additional results of this paper is an alternate form, equation (15) that we have derived. We use it along with its asymptotic form, equation (18), which is well-known in the literature [33]. Both provide considerable computational convenience. While Figure 1 shows the Green function for the algebraic case in the underdamped and overdamped regimes, Figures 2 and 3 display the usefulness of the alternate forms we provide for the computations.

Noteworthy is the fact that the single parameter $\nu$ determines in the algebraic case whether oscillatory (positive $\nu$) or monotonically decreasing (negative $\nu$) time variation obtains. Whereas for the other two memories, with the coherence parameter ($\omega$ or $\mu$) held constant, variation

of the damping ($b$ or $1/\tau$ respectively) transitions the system from the oscillatory to the overdamped regimes, variation of $\alpha$ with $\zeta$ held constant does not do anything similar in the algebraic case. As a result of the scaling behavior of the power-law dependence, the damping parameter $\alpha$ introduced in the definition (12) of the algebraic memory in a manner analogous to the exponential case (5), completely drops out of the picture in the Green function (see Eq. (14)). The coherence parameter merely serves to scale time and $\nu$ alone determines the oscillatory-decaying transition. This remarkable feature of the algebraic memory stems from its scale-free nature.

The time-integral of the Green function from zero to infinity, i.e., $\int_0^\infty \lambda(t)dt$, is of direct relevance for several observables. Equation (6) shows that it is given by $1/\gamma\tilde{\phi}(0)$. For the exponential and the delay memories, this presents no problems but for the algebraic case one runs into the peculiarity that $\tilde{\phi}(0)$ may not exist. Indeed, $\int_0^\infty \lambda(t)dt$ vanishes for positive $\nu$ but becomes infinite for negative $\nu$ provided $\nu \in [-1, 0)$. One of the direct consequences of this feature is the drastic jump from 0 to $\infty$ observed in the spectral line at zero frequency $f$ noted in Figure 5 which depicts motional narrowing of the a.c. susceptibility.

For algebraic and delay memories there is also the regime of unbounded oscillations that occurs for $|\nu| > 1$ and $\gamma\tau > \pi/2$, respectively. Because the regime is seldom physical, we have shown it only passingly in Figure 4 and only for the delay case, with an effect in the central panel of Figure 5 where additional peaks in the spectral line result.

### 6.2 Non-local attractive term in the Smoluchowski equation

Although our focus in this paper is entirely on an analysis of the Langevin equation (with time non-locality), let us recall that there are in the literature, already, a number of discussions centered on probability density evolution [29–32]. These approaches aim to construct an effective Fokker-Planck equation corresponding to the non-Markoffian Langevin equation (2), in what has been termed the *bona-fide* Fokker Planck *description* [30]. Given the memory in the Langevin equation, what is required is a Fokker-Planck equation for the *conditional* probability distribution rather than for the single-time probability distribution, as would be appropriate in the absence of the memory. Unfortunately, available in the literature as a Fokker-Planck equation corresponding to equation (2), with Gaussian noise, is only an equation for a *single*-time probability distribution [10,32]. Also, the practical utility of that equation appears not to have been tested. It is known to be undefined (drift and diffusion coefficients blow up) when the mean of the distribution crosses zero [53]. Crossings of this kind occur whenever the Langevin dynamics is oscillatory. It is because of these uncertainties surrounding Fokker-Planck treatments of such non-Markoffian cases that we have restricted our analysis here to consequences of a Langevin equation. Let us, nevertheless, return briefly to the issue about the generalization of the Smoluchowski equation. In our introductory comments in Section 2, we mentioned that, while one could be easily tempted into generalizing the standard Smoluchowski equation by incorporating a memory function in its attraction term to describe the non-Markoffian pull towards the center, such a procedure would be incorrect.

Such a generalization of the time-local Smoluchowski equation (1) brought about by introducing a memory into the attraction term, would result in

$$\frac{\partial P(x,t)}{\partial t} = \frac{\partial}{\partial x}\left(\gamma \int_0^t dt'\, \phi(t-t') x P(x,t')\right) + D\frac{\partial^2 P(x,t)}{\partial x^2}. \tag{35}$$

Although we have seen in Section 2 that it is not possible to deduce (35) from the Langevin equation with memory (Eq. (2)), it is interesting to ask what consequences (35) leads to, since it has the appearance of a natural generalization of equation (1) to incorporate a memory process in the attraction. Through an integration by parts, and using the physical expectation that $P(x,t)$ vanishes sufficiently rapidly at $x = \pm\infty$, we obtain equations for the moments of equation (35) by multiplying it by $x^n$ and integrating over $x$ from $-\infty$ to $+\infty$. For the first moment, $(n = 1)$, we get:

$$\frac{d\langle x(t)\rangle}{dt} + \gamma \int_0^t dt'\, \phi(t-t')\langle x(t')\rangle = 0. \tag{36}$$

How does this result for the average displacement $\langle x(t)\rangle$ compare with one obtained from the *correct* generalized Langevin equation (Eq. (2))? Differentiation of equation (9) followed by the use of equation (6) show explicitly that the evolution of the mean displacement as predicted by the exact (2) is precisely that given by equation (36)! While the first moment is given correctly by the inappropriate generalization (35), higher moments are not. For $n > 1$, the moment evolution from equation (35) is:

$$\frac{d\langle x^n(t)\rangle}{dt} + n\gamma \int_0^t dt'\, \phi(t-t')\langle x^n(t')\rangle = \frac{n!}{(n-2)!} D\langle x^{n-2}\rangle. \tag{37}$$

To see explicitly that higher moments predicted by the incorrectly generalized Smoluchowski equation are inaccurate, consider the $n = 2$ solution of equation (37). We obtain

$$\langle x^2\rangle(t) = \lambda_{\gamma\to 2\gamma}(t) x_0^2 + 2D \int_0^t dt'\, \lambda_{\gamma\to 2\gamma}(t'). \tag{38}$$

Here, by $\lambda_{\gamma\to 2\gamma}$ is meant what is obtained by replacing $\gamma$ by $2\gamma$ in the expression for the Green function $\lambda(t)$. Comparison to the correct moment (Eq. (14)), shows that the higher moment prediction of the inappropriate generalization is always inaccurate except for the time-local case

when $\phi(t) = \delta(t)$ leading to an exponential $\lambda(t)$ and to the accidental correctness of the relation $\lambda_{\gamma\to 2\gamma}(t) = \lambda^2(t)$.

Situations of this kind, wherein lower moments are accurately reproduced by an approximate description but higher moments are not, are frequently encountered in transport theory [54]. In the next subsection we will see that, as a consequence of the above, although the inappropriate generalization of the Smoluchowski equation accurately describes the motional narrowing phenomenon (given that the latter depends completely on the first moment), it fails to reproduce correctly the size of the particle, time-dependent or steady-state. The size is determined by the *second* moment.

### 6.3 Apparent violation of the Balescu-Swenson theorem

There is a theorem in non-equilibrium statistical mechanics, named after Balescu [55] and Swenson [56], that states that, while non-Markoffian equations that are the consequence of microscopic dynamics describe more accurately the approach to equilibrium or the steady-state than their Markoffian counterparts, their use is unnecessary for describing observables in the steady-state. This is so because the latter are reproduced with perfect accuracy by the Markoffian equations. The reasoning behind the theorem can be understood by realizing that the Markoffian approximation involves the replacement by zero of the Laplace variable in the Laplace transforms of memories; and that asymptotic results also require such replacement in the implementation of Tauberian theorems. Steady state results are asymptotic results valid at long times.

While the Balescu-Swenson theorem is largely appropriate and highly useful, its non-judicious and blind application can lead to misleading expectations. The present analysis can provide an interesting example. In order to understand the context, the usefulness, and the apparent violation of the theorem here, notice that the theorem appeared in the work, first of Balescu [55] and then of Swenson [56], about the time that the generalized master equation (GME) made its important appearance. The GME arose in the fundamental work of various investigators [57–62]. By dint of the memory function it possessed, the GME was able to access and describe short-time behavior of systems not accessible to the Master equation. The latter is essentially the GME with a delta-function memory. An object well-known before the advent of memories, it was the central entity of non-equilibrium statistical mechanics and capable of predicting irreversibility, approach to equilibrium and the second law of thermodynamics.

Because the GME unravelled numerous features of a system additional to those described by the (memoryless) Master equation, it was of particular importance to know, a priori, what descriptions made it essential to use the GME in preference to the Master equation and for what properties the Master equation was sufficient. The Balescu-Swenson theorem performed this important task. The theorem showed that, although frequency-dependent quantities require the GME for their correct description, dc transport coefficients do not: the latter are accurately described by the Master equation without memory. Although the theorem had been presented in detailed derivations in the original enunciations [55,56], the physical understanding and origin of the theorem can be made clear quite simply. Let us assume, e.g. in the context of Ohm's law, that one has solved for the time-dependent current and thence for a transport coefficient such as the ac conductivity. If the calculation uses a GME with memory $\phi(t)$, the expression for the current in the Laplace domain will have inside it $\widetilde{\phi}(\epsilon)$, the Laplace transform of $\phi(t)$. The Master equation (which is the GME without memory) would replace $\widetilde{\phi}(\epsilon)$ by $\widetilde{\phi}(0)$ and thus predict a wrong current in general.

However, the Balescu-Swenson theorem pointed out that in calculating a dc transport coefficient such as the dc conductivity, equivalently the $t \to \infty$ limit of the relevant observable such as the steady-state current, one would take the limit $\epsilon \to 0$ as part of a Tauberian argument. One would then get nothing different from the prediction of the memoryless Master equation: in the latter $\widetilde{\phi}(\epsilon)$ is replaced by $\widetilde{\phi}(0)$ from the very beginning. This applies to various steady-state transport coefficients such as the dc susceptibility, dc conductivity, viscosity, and $\omega = 0$ values of lineshapes.

Instances where the Balescu-Swenson theorem does not apply have been pointed out earlier [42,63]. Physically, the apparent violation has been shown [42] to occur when the particle under consideration has a finite lifetime as is the case with an excitation (e.g. a Frenkel exciton): the Balescu-Swenson theorem requires that motion over the entire lifetime (or at least over much of it) be played out.

It was surprising for us to find that the theorem fails in spite of the fact that we have not assumed a finite lifetime. The saturation value of the MSD in our system, in other words the steady-state size of the particle, is an asymptotic quantity. Yet we find that its value is dependent on the memory function and does not reduce to that predicted by the Markoffian approximation (when such is possible). This is clear from equation (33).

In order to clarify this further, we have constructed Figure 8 to display the time-dependence and the saturation value of the mean square displacement for the simple case of exponential memory, with $\gamma$ held constant, for three values of the damping parameter $b/\gamma = 0.5$, 2.5 and 12.5. The exact results are shown by dashed lines and we see that the saturation size increases as $b$ is decreased even though $\gamma$ does not change. This is certainly in conflict with the Balescu-Swenson statement if we assume the saturation value of the particle extent to be a valid steady-state observable in the language of the theorem. Figure 8 also shows the interesting result that the incorrectly generalized Smoluchowski equation whose predictions are shown in solid line for the corresponding values of the memory parameters do follow the Balescu-Swenson

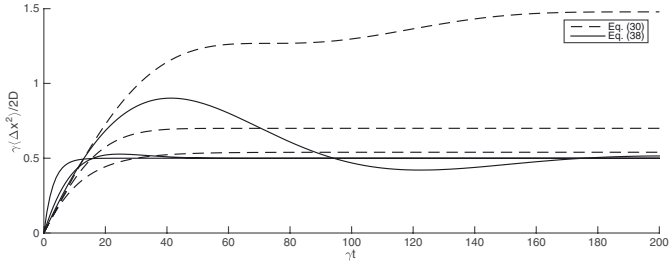


**Fig. 8.** Time-dependence of the extent of the particle (as measured by the value of the MSD) and apparent violation of the Balescu-Swenson theorem. The MSD for the exponential memory is plotted for a constant $\gamma$ and three values of the damping parameter $b/\gamma$: 0.5, 2.5 and 12.5. The correct results as given by our theory, (30), correspond to the dashed lines. Their saturation values are seen to increase as $b/\gamma$ is increased. As all cases have the same $\gamma$, that increase signifies a violation of the Balescu-Swenson theorem. By contrast, all three solid lines which are predictions of the incorrect generalization, (38), of the Smoluchowski equation (see text) saturate to the same single value (as given by the time-local equation) even though the corresponding values of $b/\gamma$ are different (as in the respective dashed line cases).

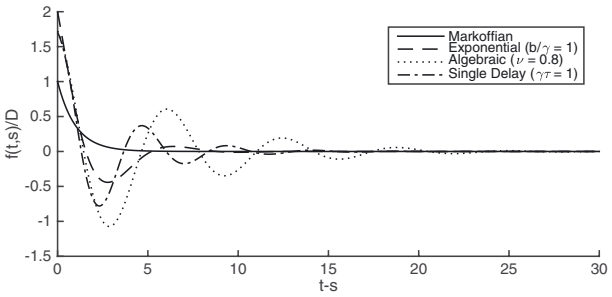


**Fig. 9.** The two-time auto-correlation function plotted as a function of the difference of the times $t$ and $s$ for all three cases of the memory function, along with the limit of no memory. We depict $f(t,s)/D$ on the $y$-axis and $t-s$ on the $x$-axis, both in units of $1/\gamma$. The sum of the times is held constant at a reasonably large value, $\gamma(t+s)=30$ while the difference $\gamma(t-s)$ is varied over the range $[0,30]$. Parameters are chosen so that the same Markoffian limit is obtained for all three memories, exponential (*dashed line*), algebraic (*dotted line*) and single delay (*dot-dashed line*). We show only the underdamped case.

theorem. The solid lines[2] all go to the same saturation value of the size that would have been obtained for the time-local situation.

Needless to say, the original context of the Balescu-Swenson theorem did not include systems such as the one treated in the present paper. What precise feature of our system makes the theorem inapplicable? The simple answer to this question is that, whereas the theorem deals with situations in which the memory describes the *entire* process, the memory we consider here refers only to a *part of the* process. It appears only in the systematic term of the Langevin equation (2).

### 6.4 A comment on two-time correlation functions

Two-time correlation functions, such as $\langle x(t)x(s)\rangle$ and their antisymmetrized combination $f(t,s)$ which is termed in some quarters as their *covariance*, are of importance in physical situations in which, in addition to the initial time, two more times rather than a single one are of importance. As a practical example, consider an electric current placed initially in a conductor in some manner. The current is disappearing as the system relaxes to equilibrium. At a certain instant $s$ before equilibrium is reached, a time-dependent electric field is switched on. At a later time $t$ the time-dependent current is measured. The analysis of such an experimental set-up clearly requires $\langle v(t)v(s)\rangle$, the two-time autocorrelation of the velocity (instead of $x$) [64]. Studies of two-time correlations have also been given in the context of aging [65,66]. We have provided general expressions for $\langle x(t)x(s)\rangle$ and $f(t,s)$ in equation (10) and (11), respectively in terms of the Green function $\lambda(t)$. For instance, for the case of an exponential memory, we have

$$f(t,s)=\frac{D}{\omega}\Bigg\{e^{-\frac{b}{2}(t-s)}\left(\left[\frac{b}{\omega}+\frac{\omega}{b}\right]\cos\left[\Omega(t-s)\right]+\frac{b^2-\omega^2}{2\Omega\omega}\sin\left[\Omega(t-s)\right]\right)-\frac{be^{-\frac{b}{2}(t+s)}}{\omega}\left(\frac{\omega^4}{\Omega^2b^2}\cos\left[\Omega(t-s)\right]+\frac{4\Omega^2-\omega^2}{4\Omega^2}\cos[\Omega(t+s)]+\frac{b^2-\omega^2}{2\Omega b}\sin\left[\Omega(t+s)\right]\right)\Bigg\}. \tag{39}$$

The Markoffian counterpart, obtained by taking the limit $b\to\infty$, $\omega\to\infty$, such that $\omega^2/b=\gamma$, is:

$$f(t,s)=\frac{D}{\gamma}\left(e^{-\gamma(t-s)}-e^{-\gamma(t+s)}\right). \tag{40}$$

Neither of these quantities, needless to say, is a function of merely the difference $t-s$. As expected, the non-Markoffian two-time correlation function differs from its Markoffian counterpart at short values of the difference $t-s$ but tends to the latter for large values. We depict this behavior for the underdamped case graphically in Figure 9 for all three cases of the memory by fixing the sum of the two times and varying the difference. Two features are clear, one at short and one at long $t-s$. As expected, all cases tend to the Markoffian limit at large values of $t-s$. At short times, on the other hand, they all are different. This is so because there they depict the (dimensionless) mean square displacements which we have seen indeed differ, representing the violation of the Balescu-Swenson theorem.

[2] Care must be taken not to take these predictions of the incorrect generalization of the Smoluchowski equation too seriously. There are physical parameter ranges in which the solid lines can dip below zero. The negativity is simply a consequence of the fact that equation (35) is not fully accurate.

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