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Anomalous processes with general waiting times: functionals and multi-point structure

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Many transport processes in nature exhibit anomalous diffusive properties with non-trivial scaling of the mean square displacement, e.g., diffusion of cells or of chromosomes inside the cell nucleus, where typically a crossover between different scaling regimes appears over time. Here, we investigate a class of anomalous diffusion processes that is able to capture such complex dynamics by virtue of a general waiting time distribution. We obtain a complete characterization of such generalized anomalous processes, including their functionals and multi-point structure, using a representation in terms of a normal diffusive process plus a stochastic time change. A generalized Feynman-Kac formula is derived, where the non-Markovian features are manifest in a memory kernel that is naturally related to the characteristic functional of the waiting times. In the special case of power law distributed waiting times we recover well-known results from the theory of continuous time random walks. Our results are readily applicable to joint velocity-position data of anomalous diffusive systems, for which a consistent underlying stochastic process can often not be identified among conventional models.

Transport phenomena in biophysical systems are being investigated with more and more interest due to the availability of sophisticated experimental techniques, such as the single-particle tracking [1, 2], the fluorescence correlation spectroscopy [3, 4] or the pulsed-gradient spin-echo NMR measurements [5], which provide an ever growing amount of data of the dynamics of living cells and intracellular organisms. The observed dynamical behaviour is usually classified in terms of the *mean square displacement* (MSD): $MSD(t) = \langle (R(t) - R_0)^2 \rangle$, where $R(t)$ is a time-dependent stochastic vector, either the position or the velocity, and R_0 is its initial value. We distinguish between normal and anomalous [6] diffusive processes for which the MSD scales linearly in time or as a power-law respectively. However, evidence of non linear MSDs was found recently in experiments of cell motility [7–9] and of diffusion of chromosomes inside the nucleus of eukaryotic cells [10], suggesting the need for more general stochastic models of anomalous processes. Here, we investigate a general class of anomalous diffusive processes that can capture such complicated MSD behaviour. In particular, we provide a complete specification of these processes in terms of their multi-point statistics, including associated time-integrated observables. In fact, many models correctly reproduce the observed MSD, but fail with respect to other statistical quantities. For example, in [9] the well known Ornstein-Uhlenbeck process [11], although modelling satisfactorily the experimental MSD, fails to account for the non-Gaussian probability density function (PDF) of the position and the power-law decaying autocorrelation function of the velocity. In [10] the continuous time random walk model (CTRW) [12, 13] with a cut-off waiting time distribution provides the correct anomalous scaling for the MSD, but the data does not exhibit the same aging effects of CTRWs. Crucial information to correctly identify the underlying stochastic

process may be contained in the higher-order correlation functions of the processes and associated time-integrated observables. However, a complete theoretical characterization of the multi-point structure is often a highly challenging task due to the non-Markovian nature of most anomalous processes [14–20].

Without loss of generality, we focus on a 1D process $Y(t)$ as a function of the physical time t and its time-integrated observables, which are naturally defined as functionals [21]:

$$W(t) = \int_0^t U(Y(\tau)) \, \mathrm{d}\tau, \qquad (1)$$

where $U(x)$ is a smooth integrable function. Clearly, if $Y(t)$ is a velocity and $U(x) = x$, $W(t)$ is the corresponding position. We are interested in the joint PDF $P(w, y, t) = \langle \delta(w - W(t)) \delta(y - Y(t)) \rangle$, which is provided by the celebrated Feynman-Kac (FK) formula when $Y(t)$ is a normal diffusion [21]. When $Y(t)$ is anomalous instead, the computation of the joint PDF reveals profound challenges and has so far rarely been studied, the exception being when $Y(t)$ is a CTRW [22–24]. We consider anomalous diffusive processes that incorporate an arbitrary waiting time distribution of the random walk [25–29]. A convenient stochastic representation of such processes starts with a parametrization of the waiting times in terms of a stochastic process $T(\cdot)$ (where $(\cdot)$ denotes the dependence on an arbitrary continuous parameter). We consider $X(\cdot)$ as a normal diffusion and form the time-changed (or subordinated) process [27–29]: $Y(t) = X(S(t))$, where the process $S(t)$ is defined as the inverse of $T(\cdot)$, or more precisely as the collection of first passage times

$$S(t) = \inf_{s > 0} \{s : T(s) > t\}. \qquad (2)$$

The dynamics of X and T can be written down explicitly

in terms of Langevin equations in the operational time s,

$$\dot{X}(s) = F(X(s)) + \sigma(X(s)) \cdot \xi(s), \quad (3a)$$

$$\dot{T}(s) = \eta(s), \quad (3b)$$

where we take $\xi(s)$ and $\eta(s)$ as two independent noises such that X and T are statistically independent processes. The functions $F(x)$, $\sigma(x)$ satisfy standard conditions [30] and we adopt the Itô convention for the multiplicative term of Eq. (3a). For X to be a normal diffusion, we require $\xi(s)$ to represent white Gaussian noise with $\langle \xi(s) \rangle = 0$ and $\langle \xi(s_1)\xi(s_2) \rangle = \delta(s_2 - s_1)$. On the other hand, we specify $\eta(s)$ as a one-sided increasing Lévy noise with finite variation [31]. More specifically, we characterize $\eta(s)$ and thus the waiting time process $T(s)$ with the characteristic functional:

$$G[k(s)] = \left\langle e^{-\int_0^{+\infty} k(s)\eta(s)\,\mathrm{d}s} \right\rangle = e^{-\int_0^{+\infty} \Phi(k(s))\,\mathrm{d}s}, \quad (4)$$

where $\Phi(k(s))$ is a non negative function with $\Phi(0) = 0$ and strictly monotone first derivative. One can easily show that the function $\Phi(s)$ is the Laplace exponent of the integrated process $T(s)$. By choosing Φ in a suitable way, a large variety of different waiting time statistics can be captured. If we choose, e.g., a power law $\Phi(\lambda) = \lambda^\alpha$ with $0 < \alpha \le 1$, $\eta(s)$ represents one sided stable Lévy noise of order α, with $0 < \alpha \le 1$ [32]. Consequently, $T(s)$ is a stable process of order α and $Y(t)$ describes a CTRW [33]. If instead $T(s) = s$, or equivalently $\Phi(\lambda) = \lambda$, we recover $Y(t)$ as a normal diffusion, which thus represents the Brownian limit. We consider a more complicated example of Φ below.

The monotonicity of $T(s)$ and $S(t)$ implies [14]:

$$\Theta(s - S(t)) = 1 - \Theta(t - T(s)), \quad (5)$$

which, together with the continuity of the paths of $S(t)$ and the corresponding Itô formula, provides the relation:

$$\delta(t - T(s)) = \delta(s - S(t))\dot{S}(t), \quad (6)$$

Formally, this equation and the following ones in which derivatives of $S(t)$ appear are to be interpreted in their corresponding integral forms, with $\dot{S}(t) = \lim_{\Delta t \to 0} \frac{S(t+\Delta t) - S(t)}{\Delta t}$ being a shorthand notation for the stochastic integrals with respect to the time-change. We can then define the time-changed process $Y(t)$ and its functional $W(t)$ as in Eqs. (1)–(2), which are equivalent to the set of time-changed Langevin equations [34]:

$$\dot{Y}(t) = F(Y(t))\dot{S}(t) + \sigma(Y(t)) \cdot \xi(S(t))\dot{S}(t), \quad (7a)$$

$$\dot{W}(t) = U(Y(t)) \quad (7b)$$

Let us now derive the generalized FK formula for the joint PDF of $Y(t)$ and $W(t)$. The FK formula describes the time evolution of the Fourier transform of $P(w, y, t)$:

$\widehat{P}(p, y, t) = \left\langle e^{i\,p\,W(t)}\delta(y - Y(t)) \right\rangle$ [21]. In the following, $\widehat{g}(k) = \int_0^{+\infty} e^{ikx}g(x)\,\mathrm{d}x$ denotes the Fourier transform of $g(x)$, whereas $\widetilde{f}(\lambda) = \int_0^{+\infty} e^{-\lambda t}f(t)\,\mathrm{d}t$ the Laplace transform of $f(t)$. Our derivation begins with the Itô formula for the joint process $Z(t) = (Y(t), W(t))$:

$$f(Z(t)) = f(Z_0) + \frac{1}{2}\int_0^t \frac{\partial^2 f}{\partial y\,\partial w}(Z(t))\,\mathrm{d}[Y, W]_t \quad (8)$$

$$+ \int_0^t \frac{\partial}{\partial y}f(Z(t))\,\mathrm{d}Y(t) + \int_0^t \frac{\partial}{\partial w}f(Z(t))\,\mathrm{d}W(t)$$

$$+ \frac{1}{2}\int_0^t \frac{\partial^2}{\partial y^2}f(Z(t))\,\mathrm{d}[Y, Y]_t + \frac{1}{2}\int_0^t \frac{\partial^2}{\partial w^2}f(Z(t))\,\mathrm{d}[W, W]_t.$$

where we denote with square brackets the quadratic variation of two processes [30]. By using the exact relation $[Y, Y]_t = \int_0^t \sigma^2(Y(\tau))\dot{S}(\tau)\,\mathrm{d}\tau$ [35, 36] and the fact that $[W, W]_t = 0 = [Y, W]_t$, together with Eqs. (7), we obtain:

$$f(Z(t)) = f(Z_0) + \int_0^t \frac{\partial}{\partial w}f(Z(\tau))U(Y(\tau))\,\mathrm{d}\tau$$

$$+ \int_0^t \frac{\partial}{\partial y}f(Z(\tau))F(Y(\tau))\dot{S}(\tau)\,\mathrm{d}\tau$$

$$+ \frac{1}{2}\int_0^t \frac{\partial^2}{\partial y^2}f(Z(\tau))\sigma^2(Y(\tau))\dot{S}(\tau)\,\mathrm{d}\tau$$

$$+ \int_0^t \frac{\partial}{\partial y}f(Z(\tau))\sigma(Y(\tau))\xi(S(\tau))\dot{S}(\tau)\,\mathrm{d}\tau. \quad (9)$$

If we now evaluate Eq. (9) for $f(Z(t)) = e^{i\,k\,Y(t)+i\,p\,W(t)}$ and take its ensemble average, we derive an equation for $\widehat{\widehat{P}}(p, k, t)$. We remark that the last integral in the rhs of Eq. (9) disappears due to the independence of the increments of $\xi(s)$. Indeed, if we make the inverse transform and recall the Fokker-Planck operator associated with Eq. (3a): $\mathcal{L}_{FP}(y) = \frac{\partial}{\partial y}F(y) + \frac{1}{2}\frac{\partial^2}{\partial y^2}\sigma^2(y)$, we obtain the equation:

$$\frac{\partial}{\partial t}\widehat{P}(p, y, t) = i\,p\,U(y)\,\widehat{P}(p, y, t)$$

$$+ \mathcal{L}_{FP}(y)\frac{\partial}{\partial t}\left\langle \int_0^t e^{i\,p\,W(\tau)}\delta(y - Y(\tau))\dot{S}(\tau)\,\mathrm{d}\tau \right\rangle. \quad (10)$$

However, we still need to relate the stochastic integral in Eq. (10) to $\widehat{P}(p, y, t)$. Starting from its definition, we first change variables in the exponential term, i.e. $e^{ip\int_0^t U(Y(\tau))\,\mathrm{d}\tau} = e^{ip\int_0^{S(t)} U(X(\tau))\eta(\tau)\,\mathrm{d}\tau}$, then we use the relation: $\delta(y - Y(t)) = \int_0^{+\infty} \delta(y - X(s))\,\delta(s - S(t))\,\mathrm{d}s$ and finally Eq. (5). Thus, we obtain for the Fourier-Laplace transform $\widetilde{\widehat{P}}(p, y, \lambda)$:

$$\widetilde{\widehat{P}}(p, y, \lambda) = \quad (11)$$

$$\int_0^{+\infty} \left\langle e^{-\int_0^s \eta(\tau)(\lambda - i\,p\,U(X(\tau)))\,\mathrm{d}\tau}\eta(s)\delta(y - X(s)) \right\rangle \mathrm{d}s,$$

which contains the derivative of $G[k(l)]$ in Eq. (4) with $k(l) = (\lambda - i\,p\,U(X(l)))\,\Theta(s - l)$. We then derive by performing the average with respect to $\eta(s)$ first:

$$\widehat{\widetilde{P}}(p, y, \lambda) = \frac{\Phi\left[\lambda - ipU(y)\right]}{\lambda - ipU(y)} \times$$
$$\times \int_0^{+\infty} \left\langle e^{-\lambda T(s) + ipW(s)} \delta(y - X(s)) \right\rangle \mathrm{d}s, \quad (12)$$

If we recall Eq. (6), we can verify that the stochastic integral in Eq. (10) and the one in Eq. (12) coincide in Laplace space. Consequently, we can use Eq. (12) in Eq. (10) to write the generalized FK formula:

$$\frac{\partial}{\partial t} \widehat{P}(p, y, t) = i\,p\,U(y)\,\widehat{P}(p, y, t) + \mathcal{L}_{FP}(y) \times \qquad (13)$$
$$\times \left[\frac{\partial}{\partial t} - i\,p\,U(y) \right] \int_0^t K(t - \tau)\,e^{i\,p\,U(y)(t-\tau)}\,\widehat{P}(p, y, \tau)\,\mathrm{d}\tau,$$

where the memory kernel is related to $G[k(s)]$ by:

$$\widetilde{K}(\lambda) = \frac{1}{\Phi(\lambda)}. \qquad (14)$$

Eq. (13) with the fundamental connection Eq. (14) between the memory kernel and the Laplace exponent of the waiting times is our first main result. We remark that other results are derived from Eq. (13) in a straightforward way. Indeed, if we set $p = 0$, we obtain a generalized Fokker-Planck equation for $P(y, t) = \langle \delta(y - Y(t)) \rangle$:

$$\frac{\partial}{\partial t} P(y, t) = \mathcal{L}_{FP}(y) \frac{\partial}{\partial t} \int_0^t K(t - \tau) P(y, \tau)\,\mathrm{d}\tau. \qquad (15)$$

In the special case where $W(t)$ corresponds to the position, $U(x) = x$, Eq. (13) yields a generalized Klein-Kramers equation exhibiting retardation effects. The Brownian limit is achieved for $K(t) = 1$, where Eqs. (13,15) reproduce the standard FK formula [21], as well as the Fokker-Planck and Klein-Kramers equations [11], respectively. Moreover, in the CTRW case the kernel is $K(t) = \frac{1}{\Gamma(\alpha)} t^{\alpha-1}$, so that the integral operator in Eq. (13) coincides with the fractional substantial derivative [22] or with the familiar Riemann-Liouville operator, if we choose $p = 0$. Thus, Eqs. (13,15) turn into the fractional FK formula [23], the fractional Fokker-Planck equation [37, 38] and the fractional Klein-Kramers equation [22, 24], respectively. We emphasize that our derivation confirms that the descriptions of CTRW functionals in terms of fractional equations on the one hand and in terms of subordinated Langevin equations on the other, are indeed equivalent. A proof of this equivalence has long been an open problem [38, 39].

We now focus on deriving the multi-point statistics of $Y(t)$ and $W(t)$, which are necessary to fully characterize these two non-Markovian processes. We note that the multi-point PDF is not accessible from Eq. (13) due to its single-point nature. However, our formulation of the generalized anomalous process in terms of a stochastic time change reveals a common structure that allows for a full solution of this problem for general Φ. Since Eqs. (3) are not coupled, we can directly compute any average over a single point function of $Y(t)$ as:

$$f(y, t) = \langle f(Y(t)) \rangle = \int_0^{+\infty} \langle f(X(s)) \rangle\,h(s, t)\,\mathrm{d}s, \qquad (16)$$

where we factorize the average over the noises $\xi(s)$ and $\eta(s)$ and define the single point PDF of the process $S(t)$: $h(s, t) = \langle \delta(s - S(t)) \rangle$, which can be computed with Eq. (5): $h(s, t) = -\frac{\partial}{\partial s} \langle \Theta(t - T(s)) \rangle$. By recalling that $\int_0^{+\infty} e^{-\lambda t} \langle \Theta(t - T(s)) \rangle\,\mathrm{d}t = \langle e^{-\lambda T(s)} \rangle$ and by using Eq. (4), we obtain in Laplace space [29]:

$$\widetilde{h}(s, \lambda) = -\frac{1}{\lambda} \frac{\partial}{\partial s} \left\langle e^{-\lambda T(s)} \right\rangle = \frac{\Phi(\lambda)}{\lambda} e^{-s\Phi(\lambda)}. \qquad (17)$$

Thus, if we know $\langle f(X(s)) \rangle$ for $X(s)$, Eqs. (16), (17) yield the corresponding average $\langle f(Y(t)) \rangle$.

If we now consider an averaged two-point function of $Y(t)$ [14], we derive:

$$f(y_2, t_2; y_1, t_1) = \langle f(Y(t_1), Y(t_2)) \rangle \qquad (18)$$
$$= \int_0^{+\infty} \int_0^{+\infty} \langle f(X(s_2), X(s_1)) \rangle\,h(s_2, t_2; s_1, t_1)\,\mathrm{d}s_2\,\mathrm{d}s_1$$

where we can use Eq. (5) to write the two-point PDF of $S(t)$ as: $h(s_2, t_2; s_1, t_1) = \langle \delta(s_2 - S(t_2))\delta(s_1 - S(t_1)) \rangle = \frac{\partial^2}{\partial s_2 \partial s_1} \langle \Theta(t_2 - T(s_2))\Theta(t_1 - T(s_1)) \rangle$. Therefore, the Laplace transform of h is related to the two-point characteristic function $Z(\lambda_2, s_2; \lambda_1, s_1) = \left\langle e^{-\lambda_2 T(s_2)} e^{-\lambda_1 T(s_1)} \right\rangle$:

$$\widetilde{h}(s_2, \lambda_2; s_1, \lambda_1) = \frac{1}{\lambda_1 \lambda_2} \frac{\partial^2}{\partial s_2 \partial s_1} Z(\lambda_2, s_2; \lambda_1, s_1), \qquad (19)$$

whose computation follows straightforwardly if we distinguish the two cases $t_2 > t_1$ and $t_2 < t_1$ and we recall the independence of the increments of $T(s)$:

$$Z(\lambda_2, s_2; \lambda_1, s_1) = \Theta(s_2 - s_1) e^{-s_1\,\Phi(\lambda_1 + \lambda_2)} e^{-(s_2 - s_1)\,\Phi(\lambda_2)}$$
$$+ \Theta(s_1 - s_2) e^{-s_2\,\Phi(\lambda_1 + \lambda_2)} e^{-(s_1 - s_2)\,\Phi(\lambda_1)}. \qquad (20)$$

This result can then be substituted in Eq. (19) to derive:

$$\widetilde{h}(s_2, \lambda_2; s_1, \lambda_1) =$$
$$\delta(s_2 - s_1) \frac{\Phi(\lambda_1) - \Phi(\lambda_1 + \lambda_2) + \Phi(\lambda_2)}{\lambda_1 \lambda_2} e^{-s_1 \Phi(\lambda_1 + \lambda_2)}$$
$$+ \Theta(s_2 - s_1) \frac{\Phi(\lambda_2)\left[\Phi(\lambda_1 + \lambda_2) - \Phi(\lambda_2)\right]}{\lambda_1 \lambda_2} \times$$
$$\times e^{-s_1 \Phi(\lambda_1 + \lambda_2)} e^{-(s_2 - s_1)\Phi(\lambda_2)}$$
$$+ \Theta(s_1 - s_2) \frac{\Phi(\lambda_1)\left[\Phi(\lambda_1 + \lambda_2) - \Phi(\lambda_1)\right]}{\lambda_1 \lambda_2} \times$$
$$\times e^{-s_2 \Phi(\lambda_1 + \lambda_2)} e^{-(s_1 - s_2)\Phi(\lambda_1)}. \qquad (21)$$

We remark that Eq. (21) is equal to the result of [14] in the special case of CTRWs. As a consequence, if $\langle f(X(s_2), X(s_1))\rangle$ is known, Eqs. (18, 21) provide the corresponding average $\langle f(Y(t_2), Y(t_1))\rangle$ in Laplace space. Moreover, also correlation functions of $W(t)$ can be easily derived using:

$$\left\langle \widetilde{W}(\lambda_1)\widetilde{W}(\lambda_2)\right\rangle = \frac{1}{\lambda_1\lambda_2}\times \tag{22}$$
$$\times \int_0^{+\infty}\int_0^{+\infty}\langle U(X(s_2)), U(X(s_1))\rangle\widetilde{h}(s_2,\lambda_2;s_1,\lambda_1)\mathrm{d}s_2\mathrm{d}s_1.$$

Exact results in physical time are obtained by solving Eqs. (18–22) in Laplace space first and then calculating the inverse Laplace transform. We remark that the corresponding formulas for higher orders can be derived in a similar way, thus providing full access to the complete multi-point structure of $Y(t)$ and $W(t)$.

As specific example we consider $\eta(s)$ as a tempered Lévy-stable noise with tempering index μ and stability index $0 < \alpha \leq 1$ [40]. This implies that $\Phi(\lambda) = (\mu + \lambda)^\alpha - \mu^\alpha$ and thus $K(t) = e^{-\mu t}t^{\alpha-1}E_{\alpha,\alpha}((\mu t)^\alpha)$ [41]. When $X(s)$ is normal diffusion the MSD exhibits crossover scaling between subdiffusive and normal diffusive regimes (see Fig. 1), as often observed in realistic systems [10, 42]. The purely subdiffusive CTRW case is recovered for $\mu = 0$, whereas the Brownian limit is obtained for $\mu \to \infty$. We derive the asymptotic scaling both for small and large times of the MSDs, by applying the Tauberian theorems to the exact results given by Eqs. (13,15). We find that $\langle Y^2(t)\rangle \sim \frac{\sigma^2}{\Gamma(1+\alpha)}t^\alpha$ and $\left\langle \overline{W}^2(t)\right\rangle \sim \frac{2\sigma^2}{\Gamma(3+\alpha)}t^\alpha$ for small times and $\langle Y^2(t)\rangle \sim \frac{\sigma^2}{\alpha\mu^{\alpha-1}}t$ and $\left\langle \overline{W}^2(t)\right\rangle \sim \frac{\sigma^2}{3\alpha\mu^{\alpha-1}}t$ for large times. When $X(s)$ is given as an Ornstein-Uhlenbeck process, we obtain likewise an interesting hybrid model, where $Y(t)$ intermediates between a CTRW and a normal diffusive oscillator. Fig. 2a shows the MSD of the time-averaged $Y(t)$-process as a function of time. The CTRW limit ($\mu = 0$) exhibits a α-dependent plateau for $t \to \infty$ highlighting the entropy breaking of the process (blue curve in Fig. 2a) [23]. For $\mu \neq 0$ we see that the MSD shows the CTRW scaling for short times, but converges to zero for $t \to \infty$ as in the Brownian limit (black curve), confirming the ergodic nature of this anomalous process. This highlights that the MSD needs to be observed for a sufficiently long time to properly assess ergodicity breaking. We also obtain the associated two-point correlation functions. For $\mu = 0$ this generalizes the simple MSD result obtained in Ref. [23] (Fig. 2b). The effect of $\mu \neq 0$ on both the Y-correlations and the correlations of its time-average are clearly visible (Fig. 2c,d), which allow to distinguish between a purely power-law waiting time distribution (CTRW) and waiting times distributed according to a tempered Lévy-stable law. Clearly, many more complicated forms of Φ can be taken into account in

our formalism, paving the way to a refined identification of the stochastic process underlying anomalous diffusion with complicated internal dynamics, as observed, e.g., in cell motility experiments [9].
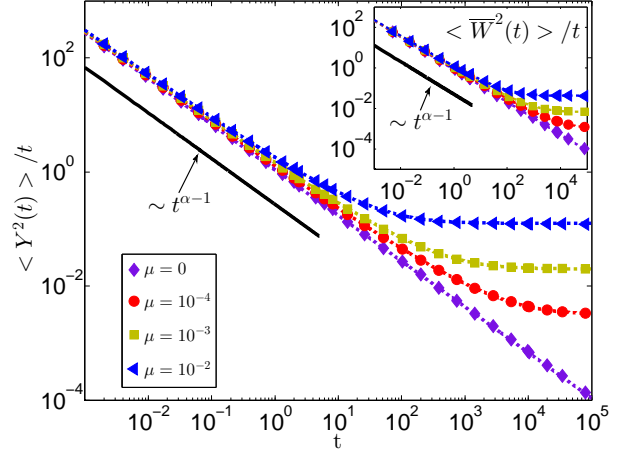

Figure 1. (Colors online) MSDs of Y(t) (main plot) and $\overline{W}(t) = W(t)/t$ (inset) normalized with the expected scaling at large times. Here, $\eta(s)$ is specified as a tempered Lévy-stable noise, $X(s)$ is normal diffusion ($F(x) = 0$, $\sigma(x) = \sigma$), and $U(x) = x$. We show our theoretical results (dotted lines) together with results from direct simulations of the Eqs. (3) (symbols) for $\alpha = 0.2$. Here, ensembles of 10^5 trajectories are simulated with the algorithms of [40, 43].
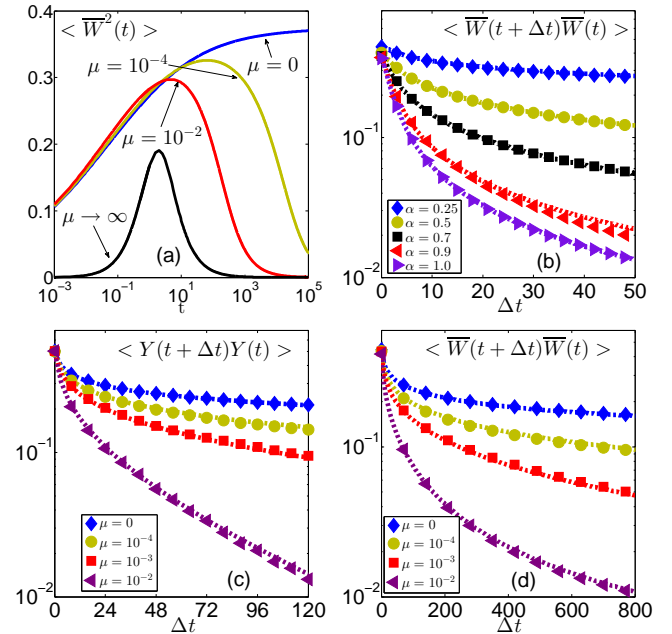

Figure 2. (Colors online) Here, $X(s)$ is given as an Ornstein-Uhlenbeck process ($F(x) = -\gamma x$, $\sigma(x) = \sigma$) and $U(x) = x$. (a) The MSD of $\overline{W}(t) = W(t)/t$ (for initial position $x_0 = 0$ and $\alpha = 0.25$). We also show the associated two-point correlation functions (for x_0 at equilibrium): (b) The special case $\mu = 0$ (CTRW).(c,d) $\mu \neq 0$ with $\alpha = 0.25$. Analytical results utilize a numerical inverse Laplace transform algorithm [44].

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