Modeling single-file diffusion with step fractional Brownian motion and a generalized fractional Langevin equation

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Abstract. Single-file diffusion behaves as normal diffusion at small time and as subdiffusion at large time. These properties can be described in terms of fractional Brownian motion with variable Hurst exponent or multifractional Brownian motion. We introduce a new stochastic process called Riemann–Liouville step fractional Brownian motion which can be regarded as a special case of multifractional Brownian motion with a step function type of Hurst exponent tailored for single-file diffusion. Such a step fractional Brownian motion can be obtained as a solution of the fractional Langevin equation with zero damping. Various kinds of fractional Langevin equations and their generalizations are then considered in order to decide whether their solutions provide the correct description of the long and short time behaviors of single-file diffusion. The cases where the dissipative memory kernel is a Dirac delta function, a power-law function and a combination of these functions are studied in detail. In addition to the case where the short time behavior of single-file diffusion behaves as normal diffusion, we also consider the possibility of a process that begins as ballistic motion.

Keywords: stochastic processes (theory), diffusion
1. Introduction

The term single-file diffusion (SFD) refers to the motion of particles in quasi-one-dimensional channels and pores which are so narrow that the particles are unable to pass each other. The exclusion of mutual passage of the diffusing particles means that the sequence of particle labels does not change over time. SFD is encountered in many physical, chemical and biological systems, which include the molecular and atomic motion in zeolites and nanotubes, particle flows in microfluidic devices, ion transport in cell membranes, colloidal motion in narrow tubes, etc [1]–[8].

The main feature of SFD is that for diffusion time $t$ smaller than the typical inter-particle collision time $\tau_c$, the particles diffuse normally and satisfy Fick’s law with its mean square displacement (MSD) $\bar{\Delta}^2(t) := \langle [x(t) - x(0)]^2 \rangle$ given by

$$\lim_{t \ll \tau_c} \bar{\Delta}^2(t) = 2D_0t,$$

with $D_0$ the diffusion coefficient. In other words, for $t \ll \tau_c$, the motion is just ordinary Brownian motion, which is a Markov process. However, for $t \gg \tau_c$,

$$\lim_{t \gg \tau_c} \bar{\Delta}^2(t) = 2F\sqrt{t},$$

where $F$ is the SFD mobility. Recall that diffusion that does not satisfy Fick’s law is known as anomalous diffusion with the MSD satisfying $\bar{\Delta}^2(t) \propto t^\alpha$, $\alpha \neq 1$. It is called superdiffusion when $\alpha > 1$, and subdiffusion when $\alpha < 1$. Thus, the long time behavior of SFD belongs to the subdiffusion regime, which is non-Markovian, indicating that the motion is correlated. Note that SFD displays anomalous diffusion characteristics even when particle–channel interactions are not taken into account.
Another way to characterize SFD is through its probability density function (or propagator) \( P(x, t) \). The probability of finding a particle at position \( x \) at time \( t \), if it is initially at the origin, approaches the Gaussian propagator after a long time:

\[
P(x, t) = \frac{1}{\sqrt{2\pi\Delta^2(t)}} \exp\left(-\frac{x^2}{2\Delta^2(t)}\right).
\]

For \( t \ll \tau_c \), one has

\[
P(x, t) = \frac{1}{\sqrt{4\pi D_0 t}} \exp\left(-\frac{x^2}{4D_0 t}\right),
\]

and for \( t \gg \tau_c \),

\[
P(x, t) = \frac{1}{\sqrt{4\pi F \sqrt{t}}} \exp\left(-\frac{x^2}{4F \sqrt{t}}\right).
\]

The notion of SFD was first introduced by Hodgkin and Keynes [9] who used it to describe the diffusion of ions through narrow channels in biological membranes. Harris was the first to provide a theoretical derivation of (1) and (2) for SFD based on statistical arguments [10]. Subsequently, this result was obtained using various models and methods by several authors including Levitt [11], Fedders [12], van Beijeren et al [13] and Kärger [14]. Recently, there have also been attempts to model SFD based on fractional diffusion equations and fractional Langevin equations [15]–[18]. Despite the numerous theoretical models and numerical simulations, experimental evidence for the occurrence of SFD was only obtained quite recently [19]–[23]. The main reason is that there is a lack of ideal experimentally accessible single-file systems.

The main aim of this paper is to propose some stochastic processes to describe the SFD. We do not address the detailed mechanism of SFD; instead we emphasize more the possibility of finding random processes which have the basic properties of SFD. Various kinds of fractional Langevin equations are considered in order to see whether they yield the stochastic processes which satisfy the basic statistical properties of SFD.

2. Modeling single-file diffusion with step fractional Brownian motion

In this section we introduce a generalization of standard fractional Brownian motion (FBM) [24] called step fractional Brownian motion (SFBM) and show that it can be used to describe the basic statistical properties of SFD. FBM has been widely used to model many areas such as turbulence, Internet traffic, financial time series, biomedical processes, etc. One limitation of the FBM model is that the long time (or low frequency) behavior that exhibits long range dependence, and the short time (or high frequency) behavior that characterizes fractal properties are both described using a single Hurst parameter \( H \). Furthermore, a constant Hurst parameter is too restrictive for many applications. During the past decade different generalizations of FBM have been proposed to address this problem. Among them, the most well known is the multifractional Brownian motion (MBM), which was introduced independently in [25] and [26]. For MBM the Hurst parameter \( H \) is replaced by \( H(t) \), a deterministic function depending on time. MBM was later extended to generalized multifractional Brownian motion (GMBM) in order to model systems which require \( H(t) \) to be a irregular function of time [27,28]. However,
there exist processes and phenomena which exhibit abrupt changes of the Hurst parameter requiring $H(t)$ to be a piecewise constant function of time. For the description of such behavior, Benassi et al [29, 30] introduced the step fractional Brownian motion (SF BM). A similar process known as multiscale fractional Brownian motion with its Hurst parameter varying as a piecewise function of frequencies was also studied by several authors [31, 32].

Recall that the standard FBM $B_H(t)$ is a Gaussian process with mean zero and correlation function given by

$$\langle B_H(t)B_H(s) \rangle = \frac{C_H}{2}(|t|^{2H} + |s|^{2H} - |t - s|^{2H}),$$

where

$$C_H = \frac{\Gamma(1 - 2H)\cos(\pi H)}{\pi H}.$$

$B_H(t)$ is not a stationary process, but its increment process is stationary. FBM is a self-similar process which satisfies, for all $a \in \mathbb{R}_+$,

$$B_H(at) \triangleq a^H B_H(t),$$

where $\triangleq$ denotes equality in all finite distributions. The stationary property of the increments of $B_H(t)$ allows the following harmonizable representation for the process:

$$B_H(t) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} e^{i\omega t} - 1 \frac{\tilde{\eta}(\omega)}{|\omega|^{H+1/2}} d\omega,$$

where $0 < H < 1$, $t \in \mathbb{R}$ and $\tilde{\eta}(\omega)$ is the Fourier transform of $\eta(t)$—the standard white noise defined by

$$\langle \eta(t) \rangle = 0, \quad \langle \eta(t)\eta(s) \rangle = \delta(t - s).$$

For modeling a process that evolves from time $t = 0$, instead of using the usual or standard FBM (which begins at time $t = -\infty$), it will be more appropriate to use an alternative FBM that starts at time zero. This second type of FBM is known as Riemann–Liouville FBM (RL-FBM), which is defined as the RL fractional integral of white noise [33]:

$$W_H(t) = \frac{1}{\Gamma(H + (1/2))} \int_0^t (t - u)^{H-1/2}\eta(u) du, \quad t \in \mathbb{R}_+, H > 0. \quad (3)$$

$W_H(t)$ is a Gaussian process with zero mean $\langle W_H(t) \rangle = 0$ and correlation function given by

$$C_{W_H}(t, s) = \langle W_H(t)W_H(s) \rangle = \frac{t^{H-1/2}s^{H+1/2}}{(H + 1/2)\Gamma(H + 1/2)^2} _2F_1 \left( \frac{1}{2} - H, 1, H + \frac{3}{2}, \frac{s}{t} \right)$$

when $s < t$. Here $_2F_1(a, b, c; z)$ denotes the Gauss hypergeometric function. The variance of the process $W_H(t)$ is

$$\text{Var}(W_H(t)) = \langle W_H(t)^2 \rangle = \frac{t^{2H}}{2H\Gamma(H + 1/2)^2}. \quad (4)$$

Note that for the standard FBM $B_H(t)$, the Hurst parameter $H$ should lie in the range $(0, 1)$, whereas for RL-FBM $W_H(t)$, $H$ takes any positive real value. Both $B_H(t)$ and $W_H(t)$ reduce to ordinary Brownian motion when $H = 1/2$. In contrast to $B_H(t)$
which has stationary increments, \( W_H(t) \) has increments which are non-stationary. Due to the failure of its increments to be stationary, \( W_H(t) \) does not have a harmonizable representation. When \( t \to \infty \), RL-FBM approaches the standard FBM [33].

Now we want to consider the step fractional Brownian motion (SFBM). Such a generalization of FBM was first introduced for standard FBM using the harmonizable representation as follows [29]:

\[
B_{H(t)}(t) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} \frac{e^{i\omega t} - 1}{|\omega|^{H(t)+1/2}} \eta(\omega) \, d\omega,
\]

where

\[
H(t) = \sum_{i=1}^{N} 1_{[\tau_{i-1}, \tau_i)} H_i,
\]

with \( \tau_0 = -\infty \) and \( \tau_N = \infty \), \( H_i \in (0, 1) \), \( 1_I(t) = 1 \) if \( t \in I \) and \( 1_I(t) = 0 \) if \( t \notin I \). This is an adaptation of MBM, which is defined for a time-dependent Hurst parameter \( H(t) \). Due to the absence of such a representation in RL-FBM, we generalize \( W_H(t) \) to RL-SFBM on the basis of the moving average representation (3):

\[
W_{H(t)}(t) = \frac{1}{\Gamma(H(t) + 1/2)} \int_0^t (t-u)^{H(t)-1/2} \eta(u) \, du,
\]

with \( H(t) \) the piecewise function given by (5), except that in this case \( \tau_0 = 0 \), and \( H_i \in (0, \infty) \). Its covariance is given by

\[
C_{W_H(t)}(t, s) = \langle W_{H(t)}(t)W_{H(s)}(s) \rangle = \frac{2}{\Gamma(H(s) + 3/2) \Gamma(H(t) + 1/2)} \binom{1}{2} - H(t), 1, H(s) + \frac{3}{2}, \frac{s}{t} \rbinom{1}{2} \binom{1}{2} \binom{1}{2} \binom{1}{2} \binom{1}{2} \binom{1}{2} \binom{1}{2} \binom{1}{2}
\]

if \( s < t \), and its variance \( \langle W_{H(t)}(t)^2 \rangle \) is given by (4) with \( H \) replaced by \( H(t) \).

The property of global self-similarity does not apply to either MBM or SFBM. In the case of MBM, the notion of self-similarity is replaced by the local asymptotic self-similarity [26], which is also satisfied by SFBM and RL-SFBM with some modification. Suppose \( W_{H(t)}(t) \) is a RL-SFBM with scaling function \( H(t) \) defined above. For all \( t \in (\tau_{i-1}, \tau_i) \),

\[
\lim_{\varepsilon \to 0} \left\{ \frac{W_{H(t)}(t+\varepsilon u) - W_{H(t)}(t)}{\varepsilon^{H_i}} \right\}_{u \in \mathbb{R}_+} \triangleq \{ B_{H_i}(u) \}_{u \in \mathbb{R}_+}.
\]

The convergence is in the sense of distributions. In other words, the tangent process of RL-SFBM for each scale \( H_i \) is \( B_{H_i} \), a FBM indexed by \( H_i \).

For modeling SFD, we use RL-SFBM with a single change of scale, that is the process (6) with \( H(t) = H_1 \mathbf{1}_{[0, \tau]} + H_2 \mathbf{1}_{[\tau, \infty)} \). To be more specific, we denote this process by \( W_{H_1, H_2}(t) \), the two-scale RL-SFBM indexed by \( H_1 > 0 \) and \( H_2 > 0 \). For this simple case, we can write

\[
W_{H_1, H_2}(t) = \frac{1_{[0, \tau]}(t)}{\Gamma(H_1 + 1/2)} \int_0^t (t-u)^{H_1-1/2} \eta(u) \, du + \frac{1_{[\tau, \infty]}(t)}{\Gamma(H_2 + 1/2)} \int_0^t (t-u)^{H_2-1/2} \eta(u) \, du.
\]
Figure 1. Two-step RL-SFBM $W_{H_1,H_2}(t)$ with $H_1 = 0.5$, $H_2 = 0.25$, $\tau = 5$, $\chi_{H_1} = 1$ and $\chi_{H_2}^2 = \sqrt{5\Gamma(0.75)^2/2}$. The smaller window shows the MSD of the process.

From the above definition, one sees that $W_{H_1,H_2}(t)$ is a Gaussian process with zero mean and correlation function

$$\langle W_{H_1,H_2}(t)W_{H_1,H_2}(s) \rangle = 1_{[0,\tau)}(t)1_{[0,\tau)}(s)C_{H_1,H_1}(t, s) + 1_{[\tau,\infty)}(t)1_{[0,\tau)}(s)C_{H_1,H_2}(t, s) + 1_{[\tau,\infty)}(t)1_{[\tau,\infty)}(s)C_{H_2,H_2}(t, s)$$

when $s < t$, and $C_{H_i,H_j}(t, s)$, $i, j = 1, 2$, is given by (7) with $H(t) = H_i$ and $H(s) = H_j$. Similarly, its variance $\langle W_{H_1,H_2}(t)^2 \rangle$ is given by (4) with $H$ replaced by $H(t)$. Thus, $W_{H_1,H_2}(t)$, the RL-SFBM with two scales, behaves like $W_{H_1}(t)$ for $t \in [0, \tau)$, and behaves like $W_{H_2}(t)$ when $t \in [\tau, \infty)$ (see figure 1). $W_{H_1,H_2}(t)$ is piecewise self-similar; it is self-similar of order $H_1$ in the time interval $[0, \tau)$, and of order $H_2$ in the time interval $[\tau, \infty)$.

In order to use RL-SFBM for modeling SFD, it is necessary to carry out some minor modifications to the definition of $W_{H_1,H_2}(t)$. Equations (6) and (8) are to be replaced by

$$W_H(t) = \frac{\chi_H}{\Gamma(H(t) + 1/2)} \int_0^t (t - u)^{H(t) - 1/2} \eta(u) \, du,$$

and

$$W_{H_1,H_2}(t) = \frac{\chi_{H_1}1_{[0,\tau)}(t)}{\Gamma(H_1 + 1/2)} \int_0^t (t - u)^{H_1 - 1/2} \eta(u) \, du + \frac{\chi_{H_2}1_{[\tau,\infty)}(t)}{\Gamma(H_2 + 1/2)} \int_0^t (t - u)^{H_2 - 1/2} \eta(u) \, du,$$

where $\chi_{H_i}$, $i = 1, 2$, are positive constants which are introduced for the purpose of obtaining the correct coefficients for the MSD of the diffusing particles. Note that by definition, $\langle W_{H^0}(t) \rangle = 0$ and $W_{H^0}(0) = 0$. Therefore the variance of $W_{H^0}(t)$ is equal to the MSD.

Now we want to see how the RL-SFBM with two scales can be used to describe the basic properties of SFD. By letting $H_1 = 1/2$, $H_2 = 1/4$, and $\chi_{H_i}$ in terms of the diffusion

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coefficient $D_0$ and SFD mobility $F$ with $\chi_{H_1} = 2D_0$, $\chi_{H_2} = \Gamma(0.75)^2F$, then the MSD of $W_{H_1,H_2}(t)$ is equal to

$$\langle W_{H_1,H_2}(t)^2 \rangle = \langle [W_{H_1,H_2}(t) - W_{H_1,H_2}(0)]^2 \rangle = \begin{cases} 2D_0t, & t \in [0, \tau), \\ 2F\sqrt{t}, & t \in [\tau, \infty), \end{cases}$$

which is the same as the MSD of SFD. The value of $\tau$ is set to equal to $\sqrt{F/D_0}$ so that the MSD is continuous. A sample path of the process $W_{H_1,H_2}(t)$ is shown in figure 1.

Note that the sample paths of RL-SFBM are discontinuous at the point of discontinuity of the Hurst function $H(t)$. In fact, it has been shown that when the Hurst function $H(t)$ is discontinuous, then the sample paths of MBM are discontinuous at points of discontinuity of $H(t)$ [34].

There are some further generalizations of RL-SFBM which can also be used to model SFD. For example, one can use a piecewise linear function $H(t)$ in (9) such that $H(t) = 1/2$ for $t \in [0, \tau_1)$, $H(t) = 1/4$ for $t \in [\tau_2, \infty)$ and $H(t)$ is a linear function interpolating the points $(\tau_1, 1/2)$ and $(\tau_2, 1/4)$ in the interval $[\tau_1, \tau_2]$. If $\chi_{H(t)} = 2D_0$ for $t \in [0, \tau_1)$, $\chi_{H(t)} = \Gamma(0.75)^2F$ for $t \in [\tau_2, \infty)$, and $\chi_{H(t)}$ is a linear function of $t$ in the interval $[\tau_1, \tau_2]$ and so $\chi_{H(t)}$ is continuous, then the process $W_{H(t)}(t)$ is a special case of RL-MBM, which provides a model for SFD that has continuous sample paths (see figure 2).

As remarked by Käger [2], the random walk model can only be regarded as an approximation to the real SFD systems. At short times, such systems first undergo ballistic motion with the MSD $\Delta(t) \sim t^2$. In other words, there is the possibility of a direct transition from the ballistic regime to the single-file regime. Such a tendency becomes more prominent with increasing concentration; and it has been demonstrated by molecular dynamical simulations [35]. For modeling SFD that is ballistic at small $t$, we can use RL-SFBM with three scales. Here we consider the case of a SFD process with
Figure 3. Three-step RL-SFBM $W_{H_1,H_2,H_3}(t)$ with $H_1 = 1$, $H_2 = 0.5$, $H_3 = 0.25$, $\tau_1 = 1$, $\tau_2 = 5$, $\chi_{H_1} = 1$, $\chi_{H_2}^2 = 1/(2\Gamma(1.5)^2)$ and $\chi_{H_3}^2 = \sqrt{5}\Gamma(0.75)^2/(4\Gamma(1.5)^2)$. The smaller window shows the MSD of the process.

three regimes: an initial ballistic regime followed by the normal diffusion, and finally the single-file diffusion region. For such a process, we let the time-dependent Hurst exponent in (9) be

$$H(t) = H_1 1_{[0,\tau_1]}(t) + H_2 1_{[\tau_1,\tau_2]}(t) + H_3 1_{[\tau_2,\infty]}(t),$$

where $H_1 = 1$, $H_2 = 1/2$ and $H_3 = 1/4$. An example of such a process is shown in figure 3.

3. Modeling single-file diffusion with fractional Langevin equations

In this section we want to examine whether it is possible to describe the basic characteristics of SFD on the basis of the various kinds of fractional Langevin equations. We shall first show that how RL-SFBM can be obtained as the solution to the ‘free fractional Langevin equation’ (fractional Langevin equation without damping). This will be followed by discussion on the fractional generalized Langevin equation and its extensions.

3.1. The fractional Langevin equation

First note that the definition of RL-FBM (3) can be written as

$$W_H(t) = a I_t^{H+1/2} \eta(t),$$

where the Riemann–Liouville (RL) fractional integral $a I_t^\alpha$ is given by [36]–[40]

$$(a I_t^\alpha f)(t) = \frac{1}{\Gamma(\alpha)} \int_0^t (t-u)^{\alpha-1} f(u) \, du.$$  

The Riemann–Liouville fractional derivative $a D_t^\alpha$ is defined as

$$a D_t^\alpha := D_t^n a I_t^{n-\alpha}.$$  

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for \( n - 1 \leq \alpha < n \). In view of the property that
\[
_0I^\alpha_t \, _0D^\alpha_0 f(t) = f(t) - \sum_{k=1}^{n} \frac{t^{\alpha-k}}{\Gamma(\alpha-k+1)} [0D^\alpha f](0),
\]
we can consider (11) as the solution of the ‘free’ fractional Langevin equation
\[
_0D^{H+1/2}_t W_H(t) = \eta(t),
\]
subject to the initial condition \((_0D^{H-1/2}_t W_H)(0) = 0\). In a similar way, we see that (8) can be re-expressed as
\[
_W_{H_1,H_2}(t) = _0I_t^{H(t)+1/2} \eta(t) = [\mathbf{1}_{[0,\tau]}(t)_0I_t^{H_1+1/2} + \mathbf{1}_{[\tau,\infty]}(t)_0I_t^{H_2+1/2}] \eta(t),
\]
and \( W_{H_1,H_2}(t) \) can be regarded as the solution of
\[
_0D^{H(t)+1/2}_t W_{H(t)}(t) = \eta(t),
\]
with \( H(t) = H_1 \mathbf{1}_{[0,\tau]} + H_2 \mathbf{1}_{[\tau,\infty]} \), and subject to the initial condition \((_0D^{H(t)-1/2}_t W_{H(t)})(0) = 0\).

Recall that Brownian motion can also be regarded as the position process associated with the solution (velocity process) of the usual Langevin equation. It is natural to ask whether a similar link exists for \( RL-FBM \) and \( RL-SFBM \) with fractional Langevin equations. For this purpose we first consider the following general type of fractional Langevin equation with two different fractional orders \( \alpha \) and \( \gamma \) [41]:
\[
(_0D_0^\alpha + \lambda)^\gamma v_{\alpha,\gamma}(t) = \eta(t), \quad 0 < \alpha < 1, \quad \gamma > 0,
\]
where \( \lambda > 0 \) is the dissipative parameter, and \( \eta(t) \) is the standard white noise. Here we remark that \((_0D_0^\alpha + \lambda)^\gamma \) can be regarded as a ‘shifted’ fractional derivative as compared with the unshifted one \(_0D_0^\alpha \). By using binomial expansion, the shifted fractional derivative can be formally expressed as
\[
(_0D_0^\alpha + \lambda)^\gamma = \sum_{j=0}^{\infty} \left( \begin{array}{c} \gamma \\ j \end{array} \right) \lambda^j _0D_0^{\alpha(\gamma-j)}.
\]
A special case of (13) with \( \alpha > 0, \gamma = 1 \) has been considered previously in [42]–[45]; and the solutions of the fractional Langevin equation with \( \alpha = 1, \gamma > 0 \) have also been studied in [46,47]. It is found that for the general case, if one considers the solution \( v_{\alpha,\gamma}(t) \) to (13) as the velocity process, then the corresponding position process \( x_{\alpha,\gamma}(t) \) depends only on the differential relation between \( v_{\alpha,\gamma}(t) \) and \( x_{\alpha,\gamma}(t) \) [48]. If the usual velocity–position relation is used, that is velocity is the ordinary derivative of position, then the variance of the position process does not depend on the long time behavior of the correlation of the velocity process. One always gets that the variance of \( x_{\alpha,\gamma}(t) \) behaves like \( \text{Var}(x_{\alpha,\gamma}(t)) \sim t \), just like for the case of normal diffusion. The long time dependence of the covariance of \( v_{\alpha,\gamma}(t) \) which varies as \( t^{-\alpha-1} \) does not enter in the leading term of the variance of \( x_{\alpha,\gamma}(t) \); it only appears as the second-leading term [48].

On the other hand, if one assumes
\[
v_{\alpha,\gamma}(t) = _0D_t^\beta x_{\alpha,\gamma;\beta}(t), \quad (_0D_t^{\beta-1} x_{\alpha,\gamma;\beta})(0) = 0,
\]
where \( 0 < \beta \leq 1 \), such that
\[
x_{\alpha,\gamma;\beta}(t) = _0I_t^\beta v_{\alpha,\gamma}(t),
\]
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then we have shown in [48] that \( \text{Var}(x_{\alpha,\gamma}(t)) \sim t^{2\beta-1} \). Therefore for \( \beta = 3/4 \), one gets the correct long time behavior for the MSD of SFD. Here we have used the fact that \( x_{\alpha,\gamma}(0) = 0 \). As for the short time behavior, it can be shown that \( \text{Var}(x_{\alpha,\gamma}(t)) \sim t^{2\alpha+2\beta-1} \) [48]. Therefore, in order to obtain the correct short time behavior for the MSD of SFD, we require \( \alpha \gamma = 1 - \beta = 1/4 \). We notice that for the characterization of the mean square displacements of SFD, the parameters \( \alpha \) and \( \gamma \) appear in the combination \( \alpha \gamma \). Therefore, we can restrict ourselves to the case \( \gamma = 1 \), for then the process \( x_{\alpha,1}(t) \) satisfies the following fractional Langevin equation:

\[
0D^\beta_\alpha x_{\alpha,1}(t) = v_{\alpha,1}(t) \quad 0D^\alpha_\alpha v_{\alpha,1}(t) + \lambda v_{\alpha,1}(t) = \eta(t), \tag{14}
\]

with initial conditions \( (0D^\beta_\alpha x_{\alpha,1})(0) = 0 \) and \( (0D^\alpha_\alpha v_{\alpha,1})(0) = 0 \). Setting \( \alpha = 1/4 \) and \( \beta = 3/4 \) recovers the basic properties for SFD (see figure 4). On the other hand, setting \( \alpha = \beta = 3/4 \) gives a process \( x_{a,1;\beta} \) that is ballistic (i.e., its MSD is \( t^2 \)) at small \( t \), and subdiffusive with exponent \( 1/2 \) when \( t \) is large (see figure 5).

Notice that for \( n - 1 \leq \alpha < n \), the Laplace transform of the Riemann–Liouville fractional derivative \( 0D^\alpha_t \) is given by [38]

\[
\tilde{0D^\alpha_\alpha f}(s) = s^\alpha \tilde{f}(s) - \sum_{k=0}^{n-1} s^k [0D^\alpha_t - k s^{\alpha-k-1}] f(0), \tag{15}
\]

where \( \tilde{f}(s) \) denotes the Laplace transform of \( f(t) \). In solving the fractional Langevin equation (13), we have assumed that \( (0D^\alpha_\alpha v)(0) = 0 \) and so the Laplace transform of the solution \( \tilde{v}_{\alpha,\gamma}(s) \) satisfies

\[
\tilde{v}_{\alpha,\gamma}(s) = \frac{\tilde{\eta}(s)}{(s^\alpha + \lambda)^\gamma},
\]

and the solution \( v_{\alpha,\gamma}(t) \) is obtained by taking the inverse Laplace transform of this equation. From the practical point of view, the applicability of the Riemann–Liouville
fractional derivative is limited by the absence of a physical interpretation of the initial condition of the type \( (0D_t^{\alpha-1}v_{\alpha,\gamma})(0) = v_0 \), when \( \alpha \) is not an integer. There is another definition of a fractional derivative called the Caputo fractional derivative which is defined as

\[
C_0^\alpha D_t^\alpha f(t) := 0_I^{n-\alpha} D_t^n f(t)
\]

when \( n-1 < \alpha \leq n \). The difference between the Caputo fractional derivative and the Riemann–Liouville fractional derivative (12) lies in the order of taking differentiation and integration. In contrast to (15), the Laplace transform of Caputo fractional derivative is given by [38]

\[
\tilde{C}_0^\alpha D_t^\alpha f(s) = s^\alpha \tilde{f}(s) - \sum_{k=0}^{n-1} s^{\alpha-k-1} f^{(k)}(0)
\]  

when \( n-1 < \alpha \leq n \). The initial conditions that need to be specified now are the values of the ordinary derivatives of \( f \) at \( t = 0 \), which have natural physical interpretations. For the system (14), we have assumed that \( (0D_t^\beta x_{\alpha,1;\beta})(0) = 0 \) and \( (0D_t^{\alpha} v_{\alpha,1})(0) = 0 \), which give the relations

\[
s^\beta \tilde{x}_{\alpha,1;\beta}(s) = \tilde{v}_{\alpha,1}(s), \quad s^\alpha \tilde{v}_{\alpha,1}(s) + \lambda \tilde{v}_{\alpha,1}(s) = \tilde{\eta}(s)
\]

for the Laplace transforms of \( x_{\alpha,1;\beta}(t) \) and \( v_{\alpha,1}(t) \). Comparing the Laplace transforms of the Riemann–Liouville fractional derivative and Caputo fractional derivative (15) and (16), we find that we can also interpret our solution to (14) as satisfying

\[
C_0^\alpha D_t^\beta x_{\alpha,1;\beta}(t) = v_{\alpha,1}(t), \quad C_0^\alpha D_t^\alpha v_{\alpha,1}(t) + \lambda v_{\alpha,1}(t) = \eta(t),
\]

with initial conditions \( x_{\alpha,1;\beta}(0) = 0 \) and \( v_{\alpha,1}(0) = 0 \), where now the Caputo fractional derivative is used. In the following, we are only going to use the Caputo fractional derivative. Therefore, we are going to use the symbol \( _0D_t^\alpha \) instead of the symbol \( C_0^\alpha D_t^\alpha \) for

\[\text{doi:10.1088/1742-5468/2009/08/P08015} \]
the Caputo fractional derivative and this should not cause any confusion. The short time behavior of MSD is sensitive to the initial conditions. For example, the stochastic process satisfying (17) with \( \alpha = \beta = 1 \) and initial conditions \( v(0) \neq 0 \) has MSD behaving like \( \sim t^2 \) as \( t \to 0 \), whereas for \( v(0) = 0 \), the MSD behaves like \( \sim t^3 \) as \( t \to 0 \). We will consider the more general case where \( x(0) \neq 0 \) and \( v(0) \neq 0 \) in the subsequent discussions.

### 3.2. The fractional generalized Langevin equation

We consider the generalized Langevin equation [49]

\[
D_\alpha x(t) = v(t), \quad D_\alpha v(t) + \int_0^t \gamma(t - u)v(u) \, du = F(t), \tag{18}
\]

where \( \gamma(t) \) is the dissipative memory kernel, and \( F(t) \) is a Gaussian random force with zero mean and correlation

\[
\langle F(t)F(s) \rangle = C_F(|t - s|). \tag{19}
\]

When \( \gamma(t) = \lambda \delta(t) \) and \( C_F(|t|) = \delta(t) \), (18) reduces to the ordinary Langevin equation. It has been shown that it is possible to obtain the position process \( x(t) \) as an anomalous diffusion if \( F(t) \) is considered as internal noise with long tailed correlation [50]. More precisely, if \( \gamma(t) = \lambda t^{-\kappa} \), where \( 0 < \kappa \leq 1 \), and the fluctuation-dissipation theorem holds with \( C_F(t) = k_B T \gamma(t) \), one then has \( \langle [x(t) - x(0)]^2 \rangle \sim t^\theta \) as \( t \to \infty \). On the other hand, if \( F(t) \) is an external noise with \( C_F(t) = c_0 |t|^{-\theta} \), \( 1 < \theta < 1 \), one has \( \langle x(t)^2 \rangle \sim t^{2 \kappa - \theta} \) as \( t \to \infty \) if \( 2 \kappa > \theta \) [51, 52]. We thus have subdiffusion when \( 0 < 2 \kappa - \theta < 1 \), superdiffusion when \( 2 \kappa - \theta > 1 \), and normal diffusion for \( 2 \kappa - \theta = 1 \). However, the short time behavior of \( x(t) \) is always ballistic [50].

In this subsection we want to examine whether it is possible to describe the basic characteristics of SFD on the basis of the fractional Langevin equation in the following general setting:

\[
0 D_\alpha^\beta x(t) = v(t), \quad 0 < \beta \leq 1, \quad 0 D_\alpha^\alpha v(t) + \int_0^t \gamma(t - u)v(u) \, du = F(t), \quad 0 < \alpha \leq 1, \tag{20}
\]

where now \( 0 D_\alpha^\alpha \) and \( 0 D_\alpha^\beta \) are Caputo fractional derivatives, \( \gamma(t) \) is the frictional kernel and \( F(t) \) is a Gaussian noise with zero mean and the following correlation:

\[
\langle F(t)F(s) \rangle = C_F(t - s) = c_0 |t - s|^{-\theta}.
\]

When \( \beta = 1 \), we have ordinary velocity, and for \( \beta \neq 1 \), the velocity is a fractional velocity.

If the dissipative memory kernel is given by \( \gamma(t) = \lambda t^{-\kappa} \), \( 0 < \kappa \leq 1 \), the second equation in (20) can be written in a more compact form:

\[
0 D_\alpha^\alpha v(t) + \chi_0 \mathcal{I}_\alpha^\zeta v(t) = F(t), \tag{21}
\]

where \( \zeta = 1 - \kappa \) and \( \chi = \Gamma(\zeta) \lambda \). Formally, when \( \zeta = 0 \) and \( F(t) = \eta(t) \), (21) reduces to the ordinary Langevin equation. Laplace transforming (20) and (21) gives

\[
s^\beta \hat{x}(s) - s^{\beta-1} \hat{x}_0 = \hat{v}(s), \quad s^\alpha \hat{v}(s) - s^{\alpha-1} \hat{v}_0 + \chi s^{-\zeta} \hat{v}(s) = \hat{F}(s), \tag{22}
\]

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where \( x_0 = x(0) \) and \( v_0 = v(0) \). We assume that \( v_0 \neq 0 \). From (22) one gets
\[
\hat{v}(s) = \frac{\tilde{F}(s)}{s^\alpha(1 + \chi s^{-\zeta - \alpha})} + \frac{v_0}{s(1 + \chi s^{-\zeta - \alpha})},
\]
\[
\hat{x}(s) = \frac{x_0}{s} + \frac{\tilde{F}(s)}{s^{\alpha + \beta}(1 + \chi s^{-\zeta - \alpha})} + \frac{v_0}{s^{\beta + 1}(1 + \chi s^{-\zeta - \alpha})}.
\]
(23)

From now on, we concentrate just on the solution to \( x(t) \). Inverse Laplace transforming (23) gives
\[
x(t) = x_0 + v_0 t^\beta E_{\alpha + \zeta, \beta + 1}(-\chi t^{\alpha + \zeta}) + \int_0^t (t-u)^{\alpha + \beta - 1} E_{\alpha + \zeta, \alpha + \beta}(-\chi(t-u)^{\alpha + \zeta}) F(u) \, du,
\]
where
\[
E_{\mu, \nu}(z) = \sum_{k=0}^{\infty} \frac{z^k}{\Gamma(\mu k + \nu)}
\]
is the two-parameter Mittag–Leffler function [53]. Taking the expectation value using (19), we have
\[
\langle x(t) \rangle = x_0 + v_0 t^\beta E_{\alpha + \zeta, \beta + 1}(-\chi t^{\alpha + \zeta}),
\]
and
\[
\sigma_{xx}^2(t) = \langle (x(t) - \langle x(t) \rangle)^2 \rangle = 2c_0 \Gamma(1 - \theta) \int_0^t u^{2\alpha + 2\beta - \theta - 1}
\times E_{\alpha + \zeta, \alpha + \beta}(-\chi u^{\alpha + \zeta}) E_{\alpha + \zeta, \beta + 1}(-\chi u^{\alpha + \zeta}) \, du,
\]
where we have used the following identities [38]: for \( \nu > 0 \) and \( \theta \leq 1 \),
\[
\int_0^t (t-u)^{-\theta} u^{\nu - 1} E_{\mu, \nu}(-\chi u^{\mu}) \, du = \Gamma(1 - \theta) t^{\nu - \theta} E_{\mu, \nu - \theta}(-\chi t^{\mu}).
\]
The MSD is given by
\[
\tilde{\Delta}^2(t) = \sigma_{xx}^2(t) + (\langle x(t) \rangle - x_0)^2 = \sigma_{xx}^2(t) + v_0 t^{2\beta} E_{\alpha + \zeta, \beta + 1}(-\chi t^{\alpha + \zeta})^2.
\]
Now by using the asymptotic properties of the Mittag–Leffler function [53]:
\[
E_{\mu, \nu}(-z) = \sum_{k=1}^{N} \frac{(-1)^k z^{-k}}{\Gamma(\nu - \mu k)} + O(z^{-1-N}), \quad z \to \infty,
\]
\[
E_{\mu, \nu}(-z) = \frac{1}{\Gamma(\nu)} + O(z), \quad z \to 0,
\]
one gets for \( t \to 0 \),
\[
\sigma_{xx}^2(t) \sim t^{2\alpha + 2\beta - \theta},
\]
\[
\tilde{\Delta}^2(t) \sim \begin{cases} t^{2\beta}, & \text{if } 2\alpha \geq \theta, \\ t^{2\alpha + 2\beta - \theta}, & \text{if } 2\alpha < \theta, \end{cases}
\]
with the assumption that \( 2\alpha + 2\beta > \theta \) and so \( \sigma_{xx}^2(t) \) has a finite limit as \( t \to 0 \).

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For large time asymptotic behaviors, we have generically
\[ \sigma^2_{xx}(t) \sim \begin{cases} \frac{t^{2\beta - 2\zeta}}{\ln t}, & \text{if } 2\beta - 2\zeta > \theta, \\ \text{constant}, & \text{if } 2\beta - 2\zeta < \theta. \end{cases} \]

Therefore, for the MSD, if \( 2\alpha \geq \theta \), the large time asymptotic behavior of \( \tilde{\Delta}^2(t) \) is the same as that of \( \sigma^2_{xx}(t) \). However, if \( 2\alpha < \theta \), then as \( t \to \infty \),
\[ \tilde{\Delta}^2(t) \sim \begin{cases} \frac{t^{2\beta - 2\zeta}}{\ln t}, & \text{if } 2\beta - 2\zeta > 2\alpha, \\ \text{constant}, & \text{if } 2\beta - 2\zeta < 2\alpha. \end{cases} \]

Now consider the general case with \( 0 \leq \zeta < 1 \). The fluctuation-dissipation theorem requires
\[ \langle F(t)F(s) \rangle = c_\theta |t-s|^{-\theta} = k_B T \chi \frac{|t-s|^{-\frac{\zeta - 1}{\Gamma(\zeta)}}}{\Gamma(\zeta)}, \]
which gives \( \theta = 1 - \zeta \). In order to ensure that the MSD satisfies the properties of SFD, we require its asymptotic behavior to be \( \sim t \) when \( t \to 0 \) and \( \sim \sqrt{t} \) when \( t \to \infty \). This gives two possibilities:

Case I \( 2\alpha \geq 1 - \zeta \), \( \beta = \frac{1}{2}, \ 2\beta - \zeta - 1 = \frac{1}{2} \),
Case II \( 2\alpha < 1 - \zeta \), \( 2\alpha + 2\beta + \zeta - 1 = 1, \ 2\beta - 2\zeta - 2\alpha = \frac{1}{2} \).

Case I implies that \( \zeta = -1/2 \), which is a contradiction to \( \zeta \in [0,1) \). For Case II, we find that
\[ \alpha = \frac{3}{8} - \frac{3\zeta}{4}, \quad \beta = \frac{\zeta}{4} + \frac{5}{8}. \] (24)

The conditions \( \alpha \in (0,1], \beta \in (0,1] \) and \( 2\alpha < 1 - \zeta \) imply that \( \zeta \in [0,1/2) \). In other words, for any \( \zeta \in [0,1/2) \), define \( \alpha \) and \( \beta \) by (24). Then the process \( x(t) \) gives a correct description of SFD.

If we assume that \( x(t) \) is ballistic instead of normally diffusive at small \( t \), then the possibilities are

Case I \( 2\alpha \geq 1 - \zeta \), \( \beta = 1, \ 2\beta - \zeta - 1 = \frac{1}{2} \),
Case II \( 2\alpha < 1 - \zeta \), \( 2\alpha + 2\beta + \zeta - 1 = 2, \ 2\beta - 2\zeta - 2\alpha = \frac{1}{2} \).

Case I gives \( \beta = 1, \zeta = 1/2 \) and \( \alpha \geq 1/4 \). For Case II, we find that
\[ \alpha = \frac{5}{8} - \frac{3\zeta}{4}, \quad \beta = \frac{\zeta}{4} + \frac{7}{8}. \]

The condition \( \beta \leq 1 \) leads to \( \zeta \leq 1/2 \). However, the condition \( 2\alpha < 1 - \zeta \) gives \( \zeta > 1/2 \), which is a contradiction. Therefore, in order to ensure that (20) gives a suitable model for \( x(t) \) which is ballistic at small \( t \) and subdiffusive of exponent 1/2 at large \( t \), we need to set \( \beta = 1, \zeta = 1/2 \) and \( \alpha \) can be any number between 1/4 and 1. When \( \alpha = 1 \), this reduces to the model (18) at the beginning of this section.

Finally, let us remark on the case where \( v_0 = 0 \). In this case, \( \tilde{\Delta}(t)^2 = \sigma^2_{xx}(t) \). We then find that the properties of SFD are satisfied if
\[ \alpha = \frac{1}{4} - \zeta, \quad \beta = \frac{\zeta}{2} + \frac{3}{4}. \]
for $\zeta \in [0, 1/4)$. When $\zeta = 0$, this gives $\alpha = 1/4$ and $\beta = 3/4$, which agrees with the result in section 3.1. On the other hand, if we require the process to be ballistic at small $t$, then

$$\alpha = \frac{3}{4} - \zeta, \quad \beta = \frac{\zeta}{2} + \frac{3}{4}$$

for $\zeta \in [0, 3/4)$. $\zeta = 0$ gives $\alpha = \beta = 3/4$, which again agrees with the result in section 3.1.

### 3.3. The extended fractional generalized Langevin equation

In this section, we stretch our Langevin approach to an even more general setting which includes the cases discussed in sections 3.1 and 3.2. Consider the following extended version of the fractional generalized Langevin equation:

$$\partial^\beta_0 x(t) = v(t),$$

$$\partial^\beta_0 v(t) + \lambda v(t) + \frac{\chi}{\Gamma(\zeta)} \int_0^t (t-u)^{\zeta-1} v(u) \, du = F(t),$$

where now the dissipative memory kernel $\gamma(t)$ is given by

$$\gamma(t) = 2\lambda \delta(t) + \frac{\chi}{\Gamma(\zeta)} t^{\zeta-1},$$

with $0 \leq \zeta < 1$. When $\alpha = \beta = 1$ and $\zeta = 1/2$, this system was proposed as a model for describing SFD for which the MSD is ballistic (i.e. $\sim t^2$) when $t$ is small. When $\lambda = 0$ or $\chi = 0$, the equations (25) reduce to the equations (20) considered in section 3.2. Therefore here we assume that $\lambda \neq 0$ and $\chi \neq 0$. As in section 3, taking Laplace transforms gives

$$\tilde{x}(s) = \frac{x_0}{s} + \frac{\tilde{F}(s)}{s^{\alpha+\beta}(1 + \lambda s^{-\alpha} + \chi s^{-\zeta-\alpha})} + \frac{v_0}{s^{\beta+1}(1 + \lambda s^{-\alpha} + \chi s^{-\zeta-\alpha})}.$$  

In the following, we assume that $v_0 \neq 0$. Let $K_1(t)$ and $K_2(t)$ be respectively the inverse Laplace transforms of

$$\frac{1}{s^{\alpha+\beta}(1 + \lambda s^{-\alpha} + \chi s^{-\zeta-\alpha})} \quad \text{and} \quad \frac{1}{s^{\beta+1}(1 + \lambda s^{-\alpha} + \chi s^{-\zeta-\alpha})}.$$  

Then the inverse Laplace transform of (27) gives

$$x(t) = x_0 + \int_0^t K_1(t-u) F(u) \, du + v_0 K_2(t).$$  

(28)

From this, we find that the variance of $x(t)$ and the MSD are given respectively by

$$\sigma^2_{xx}(t) = 2 \int_0^t \int_0^u K_1(v) C_F(u-v) K_1(u) \, dv \, du,$$

$$\Delta^2(t) = 2 \int_0^t \int_0^u K_1(v) C_F(u-v) K_1(u) \, dv \, du + v_0^2 K_2(t)^2.$$  

(29)

The asymptotic behaviors of the functions $K_1(t)$ and $K_2(t)$ at small and large $t$ are studied in the appendix. The result is: as $t \to 0$,

$$K_1(t) \sim t^{\alpha+\beta-1}, \quad K_2(t) \sim t^\beta.$$  

(30)

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As $t \to \infty$,
\[ K_1(t) \sim t^{\beta - \zeta - 1}, \quad K_2(t) \sim t^{\beta - \zeta - \alpha}. \]  
(31)

As in section 3.2, we assume the following generalized fluctuation-dissipation theorem [54]:
\[ C_F(t) = k_B T \left( 2\lambda \delta(t) + \frac{X}{\Gamma(\zeta)} t^{\zeta - 1} \right). \]  
(32)

Then the MSD (29) can be rewritten as
\[ \bar{\Delta}^2(t) = v_0^2 K_2(t)^2 + 2k_B T \lambda \int_0^t K_1(u)^2 \, du + 2k_B T \frac{X}{\Gamma(\zeta)} \int_0^t \int_0^u K_1(v)(u - v)^{\zeta - 1} K_1(u) \, dv \, du. \]  
(33)

Equation (30) then implies that its small time asymptotic behavior is
\[ \bar{\Delta}^2(t) \sim \begin{cases} t^{2\beta}, & \text{if } \alpha \geq 1/2, \\ t^{2\alpha + 2\beta - 1}, & \text{if } \alpha < 1/2. \end{cases} \]  
(34)

For the large time asymptotic behavior of the MSD, (31) and (33) give
\[ \bar{\Delta}^2(t) \sim \begin{cases} t^{2\beta - \zeta - 1}, & \text{if } 2\alpha + \zeta \geq 1 \text{ and } 2\beta - \zeta > 1, \\ \ln t, & \text{if } 2\alpha + \zeta \geq 1 \text{ and } 2\beta - \zeta = 1, \\ \text{constant}, & \text{if } 2\alpha + \zeta \geq 1 \text{ and } 2\beta - \zeta < 1, \\ t^{2\beta - 2\zeta - 2\alpha}, & \text{if } 2\alpha + \zeta < 1 \text{ and } \beta - \zeta \geq \alpha, \\ \text{constant}, & \text{if } 2\alpha + \zeta < 1 \text{ and } \beta - \zeta < \alpha. \end{cases} \]  
(35)

The short time asymptotic behavior of the MSD is governed by the term $\delta(t)$ in the dissipative memory kernel and the long time asymptotic behavior is governed by the term $t^{\zeta - 1}$. This gives the general form of dissipative memory kernel (26) the advantage of being able to interpolate between the two particular cases with $\lambda = 0$ and $\chi = 0$ considered in the previous subsections. Now to satisfy the characteristics of SFD, there are a few possibilities:

Case I $\alpha \geq \frac{1}{2}$, $\beta = \frac{1}{2}$, $2\beta - \zeta - 1 = \frac{1}{2}$,
Case II $\alpha < \frac{1}{2}$, $2\alpha + 2\beta - 1 = 1$, $2\alpha + \zeta \geq 1$, $2\beta - \zeta - 1 = \frac{1}{2}$,
Case III $\alpha < \frac{1}{2}$, $2\alpha + 2\beta - 1 = 1$, $2\alpha + \zeta < 1$, $2\beta - \zeta - 2\alpha = \frac{1}{2}$.

We have used the fact that $\alpha \geq 1/2$ implies $2\alpha \geq 1 > \zeta$. Case I gives $\zeta = -1/2$, which is a contradiction. The two equalities in Case II imply that $2\alpha + \zeta = 1/2$, which contradicts $2\alpha + \zeta \geq 1$. For Case III, the two equalities lead to
\[ \alpha = \frac{3}{8} - \frac{\zeta}{2}, \quad \beta = \frac{5}{8} + \frac{\zeta}{2}. \]  
(35)

The condition $2\alpha + \zeta < 1$ is then automatically satisfied. The conditions $0 < \alpha < 1/2$ and $0 < \beta \leq 1$ then imply that $0 \leq \zeta < 3/4$. In other words, for any $\zeta \in [0, 3/4]$, define $\alpha$ and $\beta$ by (35). Then we obtain a solution $x(t)$ to (25) which has the characteristics of SFD.

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Comparing to the solution in section 3.2 which only allows \( \zeta \) to lie in the range \([0, 1/2)\), we find that the extended fractional generalized Langevin equation (25) can be used to describe SFD in a larger range of \( \zeta \). In both cases, the maximum value of \( \alpha \) is 3/8.

Next we consider the case addressed in the paper [54], where the MSD is ballistic for small \( t \), and become subdiffusive \( \sim \sqrt{t} \) when \( t \) is large enough. For this to happen, there are a few possibilities:

Case I \( \alpha \geq \frac{1}{2}, \quad \beta = 1, \quad 2\beta - \zeta - 1 = \frac{1}{2}, \)
Case II \( \alpha < \frac{1}{2}, \quad 2\alpha + 2\beta - 1 = 2, \quad 2\alpha + \zeta \geq 1, \quad 2\beta - \zeta - 1 = \frac{1}{2}, \)
Case III \( \alpha < \frac{1}{2}, \quad 2\alpha + 2\beta - 1 = 2, \quad 2\alpha + \zeta < 1, \quad 2\beta - 2\zeta - 2\alpha = \frac{1}{2}. \)

For the first case, we find that \( \beta = 1, \zeta = 1/2 \) and the only restriction on \( \alpha \) is \( \alpha \geq 1/2 \). When \( \alpha = 1, \) this is the case considered in [54]. Case II and Case III imply that \( \alpha = 3/2 - \beta \geq 1/2 \) since \( \beta \leq 1 \). But this violates the condition \( \alpha < 1/2 \). In summary, to characterize the behavior of a system which is ballistic at small \( t \) and subdiffusive at large \( t \), one can use the extended generalized fractional Langevin model (25) with \( \beta = 1 \) (where the velocity is the normal derivative of position), \( \zeta = 1/2 \) and \( \alpha \) any values between 1/2 and 1. Comparing to the result of section 3.2, we find that when \( \lambda = 0 \), the conditions \( \zeta = 1/2 \) and \( \beta = 1 \) are the same, but the range of \( \alpha \) is from 1/4 to 1, which is a larger range of values as compared to the case when \( \lambda \neq 0 \).

Finally we remark on the case where \( v_0 = 0 \). In this case, \( \bar{\Delta}^2(t) = \sigma^2_x(t) \). Therefore, the small and large time asymptotic behaviors of the MSD for SFD are obtained if

\[
\alpha = \frac{1}{4} - \frac{\zeta}{2}, \quad \beta = \frac{\zeta}{2} + \frac{3}{4}, \quad 0 \leq \zeta < \frac{1}{2},
\]

and the MSD is ballistic at small \( t \) if

\[
\alpha = \frac{3}{4} - \frac{\zeta}{2}, \quad \beta = \frac{\zeta}{2} + \frac{3}{4}, \quad 0 \leq \zeta < 1.
\]

3.4. Further generalizations

The results above can be easily generalized to a fractional Langevin equation with more general (nonlocal) dissipative memory kernel \( \gamma(t) \) which behaves like \( \sim t^{\kappa - 1}, \kappa \in [0, 1) \), when \( t \to 0 \) and behaves like \( \sim t^{\zeta - 1}, \zeta \in [0, 1) \), when \( t \to \infty \). We note that \( \zeta = \kappa = 0 \) is the special case of the Dirac delta function kernel. The fractional generalized Langevin equation considered in section 3.2 corresponds to \( \kappa = \zeta \), whereas the extended fractional generalized Langevin equation considered in section 3.3 corresponds to \( \kappa = 0 \). For the Laplace transform of the dissipative memory kernel \( \tilde{\gamma}(s) \), one finds that \( \tilde{\gamma}(s) \sim s^{-\zeta} \) when \( s \to 0 \) and \( \tilde{\gamma}(s) \sim s^{-\kappa} \) when \( s \to \infty \). The solution \( x(t) \) can be written as (28), where the small \( t \) and large \( t \) asymptotic behaviors of the functions \( K_1(t) \) and \( K_2(t) \) are still given by (30) and (31). The generalized fluctuation-dissipation theorem (32) then implies that like \( \gamma(t), C_F(t) \sim t^{\kappa - 1} \) when \( t \to 0 \) and \( C_F(t) \sim t^{\kappa - 1} \) when \( t \to \infty \). One can then deduce that when \( t \to 0 \), the MSD (20) behaves like

\[
\bar{\Delta}^2(t) \sim \begin{cases} t^{2\beta}, & \text{if } 2\alpha + \kappa \geq 1, \\ t^{2\alpha + 2\beta + \kappa - 1}, & \text{if } 2\alpha + \kappa < 1. \end{cases}
\]
The large time asymptotic behavior of the MSD is still given by (34). After some analysis, we find that the short and long time properties of SFD are obtained if $(\zeta, \kappa)$ satisfies (see the first graph in figure 6)

$$\zeta \geq 0, \quad \kappa - 2\zeta \geq \frac{1}{2}, \quad \kappa < 1,$$

and the values of $\alpha$ and $\beta$ are

$$\alpha = \frac{1}{4} - \zeta, \quad \beta = \frac{1}{2};$$

or $(\zeta, \kappa)$ satisfies (see the second graph in figure 6)

$$\zeta \geq 0, \quad \kappa \geq 0, \quad \kappa - 2\zeta < \frac{1}{2}, \quad \kappa + 2\zeta < \frac{3}{2},$$

(36)

and the values of $\alpha$ and $\beta$ are

$$\alpha = \frac{3}{8} - \frac{\kappa}{4} - \frac{\zeta}{2}, \quad \beta = \frac{5}{8} - \frac{\kappa}{4} + \frac{\zeta}{2}.$$

On the other hand, the MSD is $\sim t^2$ (ballistic) at small $t$ if $\kappa \in [0, 1)$, $\beta = 1$, $\zeta = 1/2$ and

$$\alpha \geq \max \left\{ \frac{1}{4}, \frac{1 - \kappa}{2} \right\};$$

or $(\zeta, \kappa)$ satisfies (see figure 7)

$$\frac{1}{2} < \zeta < \frac{3}{4}, \quad \kappa - 2\zeta \geq -1/2, \quad \kappa < 1,$$

(37)

and

$$\alpha = \frac{3}{4} - \zeta, \quad \beta = 1.$$

Finally we would like to comment that if the dissipative memory kernel is a finite sum of power-law functions, then $\kappa \leq \zeta$, and the situations (36) and (37) would not appear.
4. Concluding remarks

We have proposed some stochastic processes that can describe the basic properties of SFD. In the first case, we find that the replacement of the Hurst exponent in fractional Brownian motion by a two-valued step function can be used to model SFD that is normally diffusive at small time and subdiffusive at large time. Since fractional Brownian motion can be considered as a solution of a special case of the fractional Langevin equation, it is natural to consider modeling SFD with a fractional Langevin equation. We have discussed in detail the cases where the dissipative memory kernel is a Dirac delta function or a power-law function or a combination of these. For each of these cases, we find that there is a range of the parameters where the corresponding fractional Langevin equation can be used to model a process whose MSD has $\sim t$ behavior when $t$ is small and $\sim \sqrt{t}$ behavior when $t$ is large. This range has been determined explicitly. The corresponding range of the parameters for which the MSD of the process has $\sim t^2$ behavior instead of $\sim t$ behavior when $t$ is small is also determined.

Here we would like to comment on the first-passage-time problem for SFD, looking at whether consideration of such a problem can help to illuminate the theory of SFD. If we ignore the ballistic motion which occurs only for a very short time at the beginning, then we can consider SFD as a normal diffusion followed by subdiffusion. Since the first-passage problem for Brownian motion can be solved analytically [56], one can calculate the transition time or switch-over time for going from normal diffusion to subdiffusion. This may provide a means for comparing the theoretical results with the experimental data. Recently indications have been found that single-file diffusion actually becomes normal diffusion again after a very long time [2,57]. If that is the case, and if we regard the subsequent subdiffusion as represented by fractional Brownian motion, then the estimation of the time interval for the second transition will require finding the first-passage time of the fractional Brownian motion; how to do this is still an unsolved problem. For Levy type anomalous diffusion, the corresponding fractional Fokker–Planck equation can be derived [58], and the first-passage time associated with such a process can be obtained analytically [59]. However, until now there has been no successful derivation of the correct.
Fokker–Planck equation for fractional Brownian motion; only an effective Fokker–Planck equation exists. Such an effective Fokker–Planck equation does not describe the fractional Brownian motion uniquely [2]. There exist various methods for determining the large time limit of the first-passage-time distribution of FBM such as ones based on the level crossing and first-return time [60,61], distribution of the maximum of the FBM [62]–[64], etc.

One weakness of using SFBM to model SFD is that the change of scaling exponent is not continuous, so the transition from the normal diffusion to the subdiffusion occurs abruptly. However, SFBM remains a Gaussian process even at the point of transition or switch-over from one Hurst index to another. Note that SFBM (or multifractional Brownian motion in general) can be defined at each point and can be expressed as the sum of a Gaussian wavelet series (see [27,30]). In fact, step fractional Brownian motion is a random wavelet series of independent and identically distributed (i.i.d.) random variables. But it is technically more complicated to deal with wavelets in our modeling. For a more complete description of SFD, it is necessary to take into account the boundary conditions at the ends of the single-file system, as these would become relevant if the system was of finite extent [55]. Furthermore, one may also have to consider the interaction between the diffusion particles and the wall of the single-file system. Thus, the evolution of the particles in the single-file system during the intermediate time interval becomes relevant. We hope to incorporate the physical mechanism of SFD in order to obtain a more realistic model of SFD in our future work.

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Appendix. The functions $K_1(t), K_2(t)$ and their asymptotic properties

In this appendix, we give the details of the analysis of the asymptotic properties of the functions $K_1(t)$ and $K_2(t)$ defined as the inverse Laplace transforms of

$$\frac{1}{s^{\alpha+\beta}(1 + \lambda s^{-\alpha} + \chi s^{-\zeta+\alpha})} \quad \text{and} \quad \frac{1}{s^{\beta+1}(1 + \lambda s^{-\alpha} + \chi s^{-\zeta+\alpha})}.$$ 

By taking Laplace transforms, it is easy to check that

$$K_1(t) = \sum_{k=0}^{\infty} (-1)^k \sum_{j=0}^{k} \binom{k}{j} \lambda^{k-j} \Gamma\left(\frac{t^{\alpha k+\zeta j+\alpha+\beta-1}}{\Gamma(\alpha k + \zeta j + \alpha + \beta)}\right),$$

$$K_2(t) = \sum_{k=0}^{\infty} (-1)^k \sum_{j=0}^{k} \binom{k}{j} \lambda^{k-j} \Gamma\left(\frac{t^{\alpha k+\zeta j+\beta}}{\Gamma(\alpha k + \zeta j + \beta + 1)}\right).$$

Therefore, as $t \to 0$,

$$K_1(t) \sim t^{\alpha+\beta-1} \quad \text{and} \quad K_2(t) \sim t^\beta.$$
This behavior is independent of the coefficients $\lambda$ and $\chi$. For the large $t$ asymptotic behaviors, notice that

\begin{align}
K_1(t) &= \frac{1}{\Gamma(\alpha + \beta)} \int_0^t (t - u)^{\alpha + \beta - 1} K_0(u) \, du, \\
K_2(t) &= \frac{1}{\Gamma(\beta + 1)} \int_0^t (t - u)^\beta K_0(u) \, du,
\end{align}

where $K_0(t)$ is the inverse Laplace transform of $1/(1 + \lambda s^{-\alpha} + \chi s^{-\zeta - \alpha})$, which has an integral representation

\begin{align}
K_0(t) &= \frac{1}{2\pi i} \int_{-i\infty}^{i\infty} \frac{e^{st}}{1 + \lambda s^{-\alpha} + \chi s^{-\zeta - \alpha}} \, ds \\
&= \frac{1}{2\pi} \int_0^\infty \frac{e^{ist}}{1 + \lambda(id) s^{-\alpha} + \chi(id)s^{-\zeta} - s^{-\zeta}} \, ds \\
& \quad + \frac{1}{2\pi} \int_0^\infty \frac{e^{-ist}}{1 + \lambda(-id)s^{-\alpha} + \chi(-id)s^{-\zeta} - s^{-\zeta}} \, ds \\
&= \text{Im} \left[ \frac{1}{\pi} \int_0^\infty \frac{e^{-st}}{s^{\zeta + \alpha} + \lambda e^{\pi i}s^{-\alpha} + \chi e^{\pi i}s^{-\zeta}} \, ds \right].
\end{align}

Now for the large $t$ asymptotic behavior of $K_0(t)$, we have

\begin{align}
K_0(t) &= t^{-\zeta - \alpha} \text{Im} \left[ \frac{1}{\pi} \int_0^\infty \frac{e^{-st}}{(s/t)^{\zeta + \alpha} + \lambda e^{\pi i}(s/t)^{-\alpha} + \chi e^{\pi i}(\zeta + \alpha)} \, ds \right] \\
&= t^{-\zeta - \alpha} \text{Im} \left[ \frac{1}{\pi} \int_0^\infty \sum_{k=0}^{\infty} \left\{ \frac{(-1)^k}{\chi^{k+1}} \sum_{j=0}^{k} \binom{k}{j} \lambda^{k-j} e^{-i\pi(\zeta k + \alpha j + \zeta + \alpha)} \right\} \right] \\
& \quad \times t^{-\zeta - \alpha} \chi^{k+j+\zeta + \alpha} e^{-s} \, ds \sim -\frac{t^{-1-\zeta-\alpha}}{\pi} \sum_{k=0}^{\infty} \left\{ \frac{(-1)^k}{\chi^{k+1}} \sum_{j=0}^{k} \binom{k}{j} \lambda^{k-j} \sin(\pi(\zeta k + \alpha j + \zeta + \alpha)) \right\} \\
& \quad \times \Gamma(\zeta k + \alpha j + \zeta + \alpha + 1) t^{-\zeta - \alpha}.
\end{align}

This implies that, generically,

$$K_0(t) \sim t^{-1-\zeta-\alpha}, \quad t \to \infty.$$
We then obtain from (A.1) that as $t \to \infty$,
\[ K_1(t) \sim t^{\beta - \zeta - 1}, \quad K_2(t) \sim t^{\beta - \zeta - \alpha}. \]

References

[1] Kärgér J and Ruthven D M, 2008 
Diffusion in Zeolites and Other Microporous Solids
2nd edn (New York: Wiley)

[2] Kärgér J, Single-file diffusion in zeolites, 2008
Molecular Sieves—Science and Technology, Adsorption and Diffusion ed H G Karge and J Weitkamp (New York: Springer) p 329

[3] Roque-Malherbe R M A, 2007
Adsorption and Diffusion in Nanoporous Materials
(Boca Raton, FL: Taylor and Francis)

[4] Cheng C Y and Bowen C R, 2007
ChemPhysChem 8 2077

[5] Strook A D, Weck M, Chiu D T, Huck W T S, Kenis P J A, Ismagilov R F and Whitesides G M, 2000
Phys. Rev. Lett. 84 3314

[6] Aidley D J and Standfield P R, 1996
Ion Channels: Molecules in Action (New York: Cambridge University Press)

[7] Alberts B, Johnson A, Lewis J, Raff M, Roberts K and Walter P, 2008
Molecular Biology of the Cell
5th edn (New York: Garland)

[8] Li G W, Berg O G and Elf J, 2009
Nat. Phys. 5 294

[9] Hodgkin A L and Keynes R D, 1955
J. Physiol. 128 61

[10] Harris T E, 1965
J. Appl. Probab. 2 323

[11] Levitt D G, 1973
Phys. Rev. A 8 3050

[12] Mandelbrot B B and van Ness J W, 1968
SIAM Rev. 10 422

[13] Podlubny I, 1999
Fractional Differential Equations
(San Diego, CA: Academic)

[14] West B J, Bologna M and Grigolini P, 2003
Physics of Fractal Operators
(New York: Springer)

[15] Kilbas A A, Srivastava H M and Trujillo J J, 2006
Theory and Applications of Fractional Differential Equations
(Amsterdam: Elsevier)

[16] Melzak Z A, 1967
An Invitation to Combinatorial Mathematics (New York: Wiley)

[17] Arnold V I, 1988
Mathematical Methods of Classical Mechanics
(New York: Springer)

[18] Podlubny I, 1999
Fractional Differential Equations
(Amsterdam: Gordon and Breach)

[19] West B J, Bologna M and Grigolini P, 2003
Physics of Fractal Operators
(New York: Springer)

[20] Lim S C, Li M and Teo L P, 2008
Phys. Lett. A 372 6309

[21] Podlubny I, 1999
Fractional Differential Equations
(Amsterdam: Gordon and Breach)

[22] West B J, Bologna M and Grigolini P, 2003
Physics of Fractal Operators
(New York: Springer)

[23] Kilbas A A, Srivastava H M and Trujillo J J, 2006
Theory and Applications of Fractional Differential Equations
(Amsterdam: Elsevier)

[24] Lim S C, Li M and Teo L P, 2008
Phys. Lett. A 372 6309

[25] Podlubny I, 1999
Fractional Differential Equations
(Amsterdam: Gordon and Breach)

[26] West B J, Bologna M and Grigolini P, 2003
Physics of Fractal Operators
(New York: Springer)

[27] Kilbas A A, Srivastava H M and Trujillo J J, 2006
Theory and Applications of Fractional Differential Equations
(Amsterdam: Elsevier)

[28] Lim S C, Li M and Teo L P, 2008
Phys. Lett. A 372 6309

[29] Podlubny I, 1999
Fractional Differential Equations
(Amsterdam: Gordon and Breach)

[30] West B J, Bologna M and Grigolini P, 2003
Physics of Fractal Operators
(New York: Springer)
[44] West B J and Picozzi S, 2002 Phys. Rev. E 65 037106
[45] Picozzi S and West B J, 2002 Phys. Rev. E 66 046118
[46] Lim S C and Eab C H, 2006 Phys. Lett. A 335 87
[47] Lim S C, Li M and Teo L P, 2007 Fluct. Noise Lett. 7 L169
[48] Lim S C and Teo L P, 2009 J. Phys. A: Math. Theor. 42 065208
[49] Lutz E, 2001 Phys. Rev. E 64 051106
[50] Wang K G, 1991 Phys. Lett. A 151 119
[51] Fa K S, 2006 Phys. Rev. E 73 061104
[52] Fa K S, 2007 Eur. Phys. J. E 24 139
[53] Erdelyi A, Magnus W, Oberhettinger F and Tricomi F G, 1953 Higher Transcendental Functions vol 3 (New York: McGraw-Hill)
[54] Taloni A and Lomholt M, 2008 Phys. Rev. E 78 051116
[55] Barkai E and Silbey R, Theory of single file diffusion in a force field, 2009 Phys. Rev. Lett. 102 050602
[56] Feller W, 1957 An Introduction to Probability and its Applications vol 2 (New York: Wiley)
[57] Nelson P H and Auerbach S M, 1999 J. Chem. Phys. 110 9235
[58] Metzler R, Barkai E and Klafter J, 1999 Phys. Rev. Lett. 82 3563
[59] Rangarajan G and Ding M, 2000 Phys. Rev. E 62 120
[60] Ding M and Yang M, 1995 Phys. Lett. A 273 322
[61] Krug J, 1998 Markov Processes Related Fields 4 509
[62] Sinai Ya G, 1997 Russ. Math. Surveys 52 359
[63] Molchan G M, 1999 Commun. Math. Phys. 205 97
[64] Michna Z, 1999 Math. Methods Oper. Res. 49 335