Electronic Supplementary Material (ESI) for Soft Matter.

Supplementary Information for "The effect of active fluctuations on the dynamics of particles, motors and DNA-hairpins"

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We derive a useful relation for Mittag-Leffler functions that is used in the main text. We rewrite the generalized Langevin equation for a particle in a potential surrounded by an active and viscoelastic bath in terms of fractional derivatives. We present the details of our numerical approach and show that it complies with our analytical predictions.

CONTENTS

I. PROPERTY OF MITTAG-LEFFLER FUNCTIONS

We start with the following Mittag-Leffler function

$$E_{\alpha,1}\Big(-h|t-t'|^{\alpha}\Big), \tag{1}$$

with h a constant and t and t' variable. The Laplace transform of this function to both its arguments is

$$\int_0^{\infty} dt \int_0^{\infty} dt'\, e^{-st} e^{-s't'} E_{\alpha,1}\Big(-h|t-t'|^{\alpha}\Big). \tag{2}$$

Because we are dealing with the absolute value of the difference of the two variables, we divide the integral up in the following two parts

$$\int_0^{\infty} dt \int_t^{\infty} dt'\, e^{-st} e^{-s't'} E_{\alpha,1}\Big(-h(t'-t)^{\alpha}\Big) + \int_0^{\infty} dt' \int_{t'}^{\infty} dt\, e^{-st} e^{-s't'} E_{\alpha,1}\Big(-h(t-t')^{\alpha}\Big). \tag{3}$$

Introducing the substitutions $u = t' - t$ and $w = t - t'$ decouples the integrals

$$\int_0^{\infty} dt\, e^{-(s+s')t} \int_0^{\infty} du\, e^{-s'u} E_{\alpha,1}\Big(-hu^{\alpha}\Big) + \int_0^{\infty} dt'\, e^{-(s+s')t'} \int_0^{\infty} dw\, e^{-sw} E_{\alpha,1}\Big(-hw^{\alpha}\Big). \tag{4}$$

The first integral in both terms is easily evaluated, the second is just the Laplace transform of a Mittag-Leffler function which is given by Eq. (13) in the main text. Hence we get

$$\frac{1}{s+s'}\left[s'^{-1}\left(1+hs'^{-\alpha}\right)^{-1} + s^{-1}\left(1+hs^{-\alpha}\right)^{-1}\right]. \tag{5}$$

After some elementary algebra this expression can be written as

$$hs^{-\alpha}s'^{-\alpha}\left(1+hs^{-\alpha}\right)^{-1}\left(1+hs'^{-\alpha}\right)^{-1}\frac{s^{\alpha-1}+s'^{\alpha-1}}{s+s'} + s^{-1}s'^{-1}\left(1+hs^{-\alpha}\right)^{-1}\left(1+hs'^{-\alpha}\right)^{-1}. \tag{6}$$

The second term does not couple the variables s and s' and thus the inverse Laplace transform is readily done, it gives

$$E_{\alpha,1}\Big(-ht^{\alpha}\Big)E_{\alpha,1}\Big(-ht'^{\alpha}\Big). \tag{7}$$

To invert the first term we need the following double Laplace transform

$$\mathcal{L}\left[|t-t'|^{-\alpha}, s, s'\right] = \Gamma(1-\alpha)\frac{s^{\alpha-1}+s'^{\alpha-1}}{s+s'} \tag{8}$$

and we observe that this term is a product of two known Laplace transforms, thus a convolution after inversion to time-space. Combining this with Eq. (1) and Eq. (7), and rearranging some terms we finally have the following relation

$$\frac{h}{\Gamma(1-\alpha)}\int_0^{t} dy \int_0^{t'} dy'\, |t'-t+y-y'|^{-\alpha}\, y^{\alpha-1}y'^{\alpha-1}E_{\alpha,\alpha}\Big(-hy^{\alpha}\Big)E_{\alpha,\alpha}\Big(-hy'^{\alpha}\Big)$$
$$= E_{\alpha,1}\Big(-h|t-t'|^{\alpha}\Big) - E_{\alpha,1}\Big(-ht^{\alpha}\Big)E_{\alpha,1}\Big(-ht'^{\alpha}\Big). \tag{9}$$

II. FRACTIONAL INTEGRAL AND DERIVATIVE

Equation (5) in the main text can be rewritten in a more compact form using the formalism of fractional derivatives [1]. The (left) Riemann-Liouville fractional integral $\mathcal{I}^\mu$ of order $\mu > 0$ is defined as

$$\mathcal{I}^\mu x(t) = \frac{1}{\Gamma(\mu)} \int_0^t d\tau \, (t-\tau)^{\mu-1} x(\tau), \tag{10}$$

with $\Gamma(\cdot)$ the gamma-function. This fractional integral is, in fact, a generalization of the Cauchy formula for n-time repeated integration to real values of n. Using this definition one can construct a fractional derivative $\mathcal{D}^\alpha$ of order α. For physical processes the most commonly used fractional derivative is the one in the Caputo sense

$${}_c\mathcal{D}^\alpha x(t) = \mathcal{I}^{m-\alpha} x^{(m)}(t), \tag{11}$$

where the superscript (m) stands for the ordinary m^{th} derivative with m the value of α rounded up to the nearest integer, thus $m = \lceil \alpha \rceil = 1$ in our model. The friction term in the main text, i.e. the left-hand side of Eq. (5), can thus be written as

$$\eta_\alpha \, {}_c\mathcal{D}^\alpha x(t). \tag{12}$$

Here we introduced $\eta_\alpha = \gamma\Gamma(3-\alpha)$ for simplicity. We can now write the full equation of motion for a particle in a potential $V(x)$, surrounded by an active viscoelastic bath, it reads

$$\eta_\alpha \, {}_c\mathcal{D}^\alpha x(t) = -\frac{dV}{dx} + \xi_T(t) + \xi_A(t)\Theta(t). \tag{13}$$

III. NUMERICAL ALGORITHM

The methods used for solving the standard Langevin equation (Eq. (1) in the main text) numerically are well known and, in general, straightforward [4]. By integrating the equation discretely one can find an iterative expression from which $x(t+\Delta t)$ is calculated using $x(t)$, where $\Delta t > 0$ is a small time difference. This time step is chosen small for large accuracy but not so small as to severely slow the algorithm down. Due to the stochastic nature of an equation that includes Gaussian white-noise, uncorrelated random numbers need to be generated. Modern computers can provide these fast and accurately. Finding a numerical solution for Eq. (13), however, is not such a trivial problem. The standard techniques used to simulate the ordinary Langevin equation are inadequate. The difficulty here is twofold. First there are two stochastic terms whose correlation is a function of time, meaning that the generated noise depends on previously generated noise. Second, due to the non-local nature of a fractional differential equation the numerical integration of it should be done with care. The solution to both difficulties will be addressed here.

A. Coloured noise

The active and the thermal noise, represented collectively by $\xi(t)$ without subscript, in our model are Gaussian distributed with zero mean and a correlation that is a function of the absolute difference in time. So in general

$$\langle \xi(t)\xi(t') \rangle = f(|t-t'|). \tag{14}$$

The algorithm should generate a string of random numbers that comply with this correlation and that are Δt separated in time. We denote the i^{th} random number as $\xi[i] = \xi(i\Delta t)$. The different correlations require different techniques to be generated.

1. Power-law correlated noise

We first show that the power-law correlated thermal noise of our model can be identified with fractional Gaussian noise [5, 6]. This result is not new but we include it for completeness. Fractional Gaussian noise can be derived from a process called fractional Brownian motion. Reintroduced by Mandelbrot and van Ness [6], fractional Brownian motion $B_H(t)$ is a generalization of Brownian motion where the increments are not independent of each other. This stochastic process is defined by the following criteria

- $B_H(t)$ has stationary increments
- $B_H(0) = 0$ and $\langle B_H(t)\rangle = 0$ for $t \geq 0$
- $\langle B_H^2(t)\rangle = t^{2H}$ for $t \geq 0$
- $B_H(t)$ has a Gaussian distribution with

$$\langle B_H(t)B_H(t')\rangle = \frac{1}{2}\Big(t^{2H} + t'^{2H} - |t-t'|^{2H}\Big) \tag{15}$$

The Hurst exponent $H \in [0,1]$ determines the raggedness of the motion, larger values for H make smoother motions. When $H < 1/2$, the increments are negatively correlated while for $H > 1/2$ they are positively correlated. For $H = 1/2$ we recover classical Brownian motion, the so-called Wiener process. We can define fractional Gaussian noise $X_H(t)$ as the ratio of the increment difference and the time step over which this increment occurred

$$X_H(t) = \frac{B_H(t+\Delta t) - B_H(t)}{\Delta t}. \tag{16}$$

Using the second property, one immediately finds that the mean of this noise is zero, i.e. $\langle X_H(t)\rangle = 0$. Since the motion is Gaussian distributed, the noise is also a Gaussian variable. Thus if we also calculate its second moment, it is fully described

$$\begin{aligned}\langle X_H(t)X_H(t')\rangle = \frac{1}{\Delta t^2}\Big(&\langle B_H(t)B_H(t')\rangle - \langle B_H(t)B_H(t'+\Delta t)\rangle \\ &- \langle B_H(t')B_H(t+\Delta t)\rangle + \langle B_H(t+\Delta t)B_H(t'+\Delta t)\rangle\Big).\end{aligned} \tag{17}$$

The fourth property, Eq. (15), is used to find

$$\langle X_H(t)X_H(t')\rangle = \frac{1}{2\Delta t^2}\Big(|t-t'+\Delta t|^{2H} + |t-t'-\Delta t|^{2H} - 2|t-t'|^{2H}\Big). \tag{18}$$

Now we let the time step Δt approach zero, $\Delta t \to 0$. The first and second term in Eq. (18) can be expanded using Taylor series

$$|t-t' \pm \Delta t|^{2H} = |t-t'|^{2H}\left(1 \pm 2H\frac{\Delta t}{|t-t'|} + H(2H-1)\frac{\Delta t^2}{|t-t'|^2} + \mathcal{O}(\Delta t^3)\right). \tag{19}$$

Putting this in Eq. (18), we find

$$\langle X_H(t)X_H(t')\rangle \approx H(2H-1)|t-t'|^{2H-2}. \tag{20}$$

If we now multiply this by $2\gamma k_B T$ and take $H = (2-\alpha)/2$, we recover the correlation of our model's thermal noise.

To generate this fractional Gaussian noise we use the algorithm conceived by Hosking in 1984 [7]. The Hosking algorithm produces a collection of numbers from a stationary process with a normal marginal distribution and correlation function $\rho[i] = \rho(i\Delta t)$. For our purpose this correlation function is $\rho[i] = \langle X_H(0)X_H(i\Delta t)\rangle$. Using Eq. (18) brings us to

$$\rho[i] = \frac{1}{2}\Big(|i+1|^{2-\alpha} + |i-1|^{2-\alpha} - 2|i|^{2-\alpha}\Big). \tag{21}$$

The scheme below produces fractional Gaussian noise $X[i]$ with Δt unity. It requires the production of Gaussian white noise $N(\mu, \sigma^2)$ with mean μ and variance σ^2.

1. Set $v = 1$.
2. Generate a starting value $X[0] = N(0, v)$.

3. For every i^{th} time step do

$$\begin{aligned}
\psi_k &= \phi_k \\
\phi_i &= v^{-1}\rho[i] - v^{-1}\sum_{j=1}^{i-1}\rho[i-j]\psi_j \\
v &= (1-\phi_i^2)v \\
\\
\phi_k &= \psi_k - \phi_i\psi_{i-k} \\
m &= \sum_{j=1}^{i} X[i-j]\phi_j \\
X[i] &= N(m,v)
\end{aligned}$$

In every step, the indices k is always taken from 1 to $i-1$. For the scheme presented here we chose clarity above efficiency. For example, if several realisations of the same process are being performed, one could consider to calculate all needed values of ϕ beforehand. To rescale this process to one where $\Delta t \neq 1$, all $X[i]$ need to be multiplied by $\Delta t^{-\alpha/2}$. Then to make the transition from fractional Gaussian noise to the thermal noise $\xi_T(t)$, we should multiply it by $\sqrt{2\gamma k_B T}$, thus we have

$$\xi_T[i] = \sqrt{2\gamma k_B T \Delta t^{-\alpha}}\, X[i]. \tag{22}$$

2. Exponentially correlated noise

The Hosking algorithm can surely be used to generate the active noise, which would have the following correlation function in the algorithm

$$\rho[i] = e^{-i\Delta t/\tau_A}. \tag{23}$$

The generated numbers should be multiplied by $\sqrt{C}$. But exponentially correlated noise can also be generated by a linear damping equation driven by Gaussian white noise. This method is much more time and memory efficient than the Hosking method. An integral algorithm for exponentially correlated noise was proposed by Fox *et al.* [8]. Its scheme is as follows, where again $N(\mu,\sigma^2)$ stands for Gaussian white noise with mean μ and variance σ^2

1. Set $E = e^{-\Delta t/\tau_A}$.
2. Generate a starting value $\xi_A[0] = N(0,C)$.
3. For every i^{th} time step do

$$\xi_A[i] = E\,\xi_A[i-1] + N(0,(1-E^2)C). \tag{24}$$

B. Fractional differential equation

Here we seek the numerical integration of a differential equation of fractional order, in our work it always has the form of the following initial value problem

$${}_c\mathcal{D}^{\alpha}x(t) = g(t,x(t)), \quad x(0) = x_0, \tag{25}$$

with $0 < \alpha \leq 1$. The function g could be any nonlinear function, in our specific case, using Eq. (13), we have

$$g(t,x(t)) = \frac{1}{\eta_\alpha}\left(-\frac{dV(x)}{dx} + \xi_T(t) + \xi_A(t)\right), \tag{26}$$

for $t > 0$. In order to solve Eq. (25) one can use the fact that this initial value problem is equivalent to the following Volterra integral [9]

$$x(t) = x_0 + \frac{1}{\Gamma(\alpha)}\int_0^t dy\,(t-y)^{\alpha-1}g(y,x(y)). \tag{27}$$

The algorithm developed to solve this integral is a generalization of the classical Adams-Bashforth-Moulton integrator for first-order problems [10, 11]. The generalization was done by Diethelm *et al.* [12] and optimized by Daftardar-Gejji *et al.* [13]. It will produce $x_{n+1} = x(t_{n+1})$ (with $t_i = i\Delta t$) a time step Δt further than x_n. Note that the integral in Eq. (27) goes from 0 to t which is a consequence of the non-locality of fractional differential equations. Therefore the iterative process of the algorithm will use all calculated solutions x_i instead of only the previous solution x_n as is done in the integration of the ordinary Langevin equation. The algorithm uses the following predictor-corrector protocol where P^1_{n+1} and P^2_{n+1} are the predictors and x_{n+1} the corrector

$$P^1_{n+1} = x_0 + \frac{\Delta t^\alpha}{\Gamma(2+\alpha)} \sum_{i=0}^{n} a_{i,n}\, g(t_i, x_i), \tag{28}$$

$$P^2_{n+1} = \frac{\Delta t^\alpha}{\Gamma(2+\alpha)}\, g(t_{n+1}, P^1_{n+1}), \tag{29}$$

$$x_{n+1} = P^1_{n+1} + \frac{\Delta t^\alpha}{\Gamma(2+\alpha)}\, g(t_{n+1}, P^1_{n+1} + P^2_{n+1}). \tag{30}$$

The weight $a_{i,n}$ (which ultimately originates from the memory kernel, Eq. (3) in the main text) occuring in the previous solutions has the following form

$$a_{i,n} = \begin{cases} n^{\alpha+1} - (n-\alpha)(n+1)^\alpha & \text{if} \quad i = 0 \\ (n-i+2)^{\alpha+1} + (n-i)^{\alpha+1} - 2(n-i+1)^{\alpha+1} & \text{if} \quad 1 \le i \le n \end{cases} \tag{31}$$

The algorithm is now fully described. Note that for large n it will be increasingly intensive to calculate the next value of x_i due to the summation in Eq. (28). One also has to store all calculated values of $g(t_i, x_i)$ in the memory which could overflow for large n. We only presented the algorithm for one particle in one dimension, the extension to many (interacting) particles in three dimensions is relatively straightforward and not given here.

In Fig. 1, we show both the analytic as the numeric results of the mean-squared distance travelled by a particle in a harmonic potential, surrounded by an active viscoelastic bath. Initially, the particle is in equilibrium in the potential. The analytic data (shown as black lines) is an evaluation of Eq. (24) from the main text. The simulated data (represented by coloured dots) is produced by the algorithm explained above. From this figure it is clear that our numerical scheme performs satisfactory since it generates data that lies perfectly on top of the analytic results for a large range of time-scales.

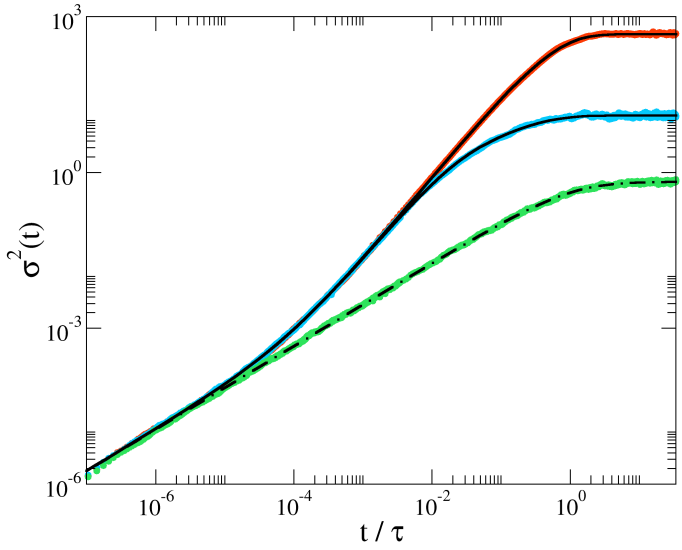

FIG. 1. Log-log plot of the mean-squared displacement of a harmonic oscillator ($k = 3$) as a function of t/τ in a viscoelastic medium ($\alpha = 0.8$, $k_BT = \gamma = 1$) in the presence of active forces with $C = 10^4$ and $\tau_A/\tau = 1,\ 10^{-2}$ (full lines, top to bottom) compared to that without active forces (dashed-dotted line). The coloured dots are the simulated data (averaged over 10^3 histories) with green ($C = 0$), blue ($\tau_A/\tau = 10^{-2}$) and orange ($\tau_A/\tau = 1$).

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