

Supplementary Material for 'Confined Brownian motion tracked with motion blur: Estimating diffusion coefficient and size of confining space' by Mortensen et al.

1 CALCULATIONS FOR THE VARIANCE OF A PARTICLE IN A 1D BOX

We insert the expression for $P(x, t|x_0, t_0)$ from Equation (13) in the time-ordered auto-correlation function in Equation (17) and find

$$\begin{aligned}
 & \langle x(t|x_0)x(t'|x_0) \rangle_{0, t' > t} \\
 &= \int_0^L \int_0^L xx' P(x, t|x_0, 0) P(x', t'|x, t) dx dx' \\
 &= \int \int dx dx' \frac{xx'}{L^2} + \frac{2}{L^2} \sum_{q=1}^{\infty} \exp \left[- \left(\frac{q\pi}{2} \right)^2 \frac{t' - t}{\tau} \right] \left[\int dx x \cos \left(\frac{q\pi x}{L} \right) \right]^2 \\
 & \quad + \left[\text{two terms proportional to } \cos \left(\frac{n\pi x_0}{L} \right) \right]
 \end{aligned} \tag{S1}$$

The integrals are

$$\int \int dx dx' \frac{xx'}{L^2} = \frac{L^2}{4}, \tag{S2}$$

$$\int dx x \cos \left(\frac{q\pi x}{L} \right) = \frac{[-1 + (-1)^q] L^2}{\pi^2 q^2}, \text{ i.e. } \frac{-2L^2}{\pi^2 q^2} \text{ for } n \text{ odd, and } 0 \text{ otherwise,} \tag{S3}$$

which we insert in Equation (S1) and get

$$\begin{aligned}
 \langle x(t|x_0)x(t'|x_0) \rangle_{0, t' > t} &= \frac{L^2}{4} + \frac{8L^2}{\pi^4} \sum_{p=0}^{\infty} \exp \left[- \left(\frac{(1+2p)\pi}{2} \right)^2 \frac{t' - t}{\tau} \right] \frac{1}{(1+2p)^4} \\
 & \quad + \left[\text{two terms proportional to } \cos \left(\frac{n\pi x_0}{L} \right) \right].
 \end{aligned} \tag{S4}$$

Inserting this expression in Equation (16) gives

$$\begin{aligned}
 & \langle x_{\text{msr}}(x_0)^2 \rangle_0 \\
 &= \frac{2}{(\Delta t_{\text{open}})^2} \int_{\Delta t - \Delta t_{\text{open}}}^{\Delta t} dt' \int_{\Delta t - \Delta t_{\text{open}}}^{t'} dt \langle x(t|x_0)x(t'|x_0) \rangle_{0,t'>t} \\
 &= \frac{L^2}{4} + \frac{8L^2}{\pi^4} \sum_{p=0}^{\infty} \frac{1}{(1+2p)^4} \left\{ \frac{2}{(\Delta t_{\text{open}})^2} \int_{\Delta t - \Delta t_{\text{open}}}^{\Delta t} dt' \int_{\Delta t - \Delta t_{\text{open}}}^{t'} dt \exp \left[- \left(\frac{(1+2p)\pi}{2} \right)^2 \frac{t' - t}{\tau} \right] \right\} \\
 & \quad + \left[\text{two terms proportional to } \cos \left(\frac{n\pi x_0}{L} \right) \right] .
 \end{aligned} \tag{S5}$$

The double-integral in the curly brackets is

$$\begin{aligned}
 & \frac{2}{(\Delta t_{\text{open}})^2} \int_{\Delta t - \Delta t_{\text{open}}}^{\Delta t} dt' \int_{\Delta t - \Delta t_{\text{open}}}^{t'} dt \exp \left[- \left(\frac{(1+2p)\pi}{2} \right)^2 \frac{t' - t}{\tau} \right] \\
 &= \frac{8}{\pi^2(1+2p)^2} \frac{\tau}{\Delta t_{\text{open}}} + \frac{32}{\pi^4(1+2p)^4} \left\{ \exp \left[- \left(\frac{(1+2p)\pi}{2} \right)^2 \frac{\Delta t_{\text{open}}}{\tau} \right] - 1 \right\} \left(\frac{\tau}{\Delta t_{\text{open}}} \right)^2 ,
 \end{aligned} \tag{S6}$$

which is inserted in Equation (S5),

$$\begin{aligned}
 & \langle x_{\text{msr}}(x_0)^2 \rangle_0 \\
 &= \frac{64L^2}{\pi^6} \sum_{p=0}^{\infty} \frac{1}{(1+2p)^6} \left\{ \frac{\tau}{\Delta t_{\text{open}}} + \frac{4}{\pi^2(1+2p)^2} \left\{ \exp \left[- \left(\frac{(1+2p)\pi}{2} \right)^2 \frac{\Delta t_{\text{open}}}{\tau} \right] - 1 \right\} \left(\frac{\tau}{\Delta t_{\text{open}}} \right)^2 \right\} \\
 & \quad + \frac{L^2}{4} + \left[\text{two terms proportional to } \cos \left(\frac{n\pi x_0}{L} \right) \right] .
 \end{aligned} \tag{S7}$$

When the expression for $\langle x_{\text{msr}}(x_0)^2 \rangle_0$ is inserted in Equation (S15) and the integration over initial positions is carried out, we find

$$\begin{aligned}
 & \text{var}(x_{\text{msr}}) \\
 &= \frac{64L^2}{\pi^6} \sum_{p=0}^{\infty} \frac{1}{(1+2p)^6} \left\{ \frac{\tau}{\Delta t_{\text{open}}} + \frac{4}{\pi^2(1+2p)^2} \left\{ \exp \left[- \left(\frac{(1+2p)\pi}{2} \right)^2 \frac{\Delta t_{\text{open}}}{\tau} \right] - 1 \right\} \left(\frac{\tau}{\Delta t} \right)^2 \right\} ,
 \end{aligned} \tag{S8}$$

as the integral over x_0 removes terms proportional to $\cos[n\pi x_0/L]$ and leaves the other terms unchanged. Using that $\sum_{p=0}^{\infty} (1+2p)^{-6} = \pi^6/960$ [Bickel (2007)], we arrive at Equation (18).

For $\Delta t_{\text{open}}/\tau \ll 1$, we Taylor expand the terms in the outer curly brackets in Equation (S8) and find

$$\begin{aligned}
 & \frac{\tau}{\Delta t_{\text{open}}} + \frac{4}{\pi^2(1+2p)^2} \left\{ \exp \left[- \left(\frac{(1+2p)\pi}{2} \right)^2 \frac{\Delta t_{\text{open}}}{\tau} \right] - 1 \right\} \left(\frac{\tau}{\Delta t_{\text{open}}} \right)^2 \\
 &= \frac{\tau}{\Delta t_{\text{open}}} + \frac{4}{\pi^2(1+2p)^2} \left\{ - \left(\frac{(1+2p)\pi}{2} \right)^2 \frac{\Delta t_{\text{open}}}{\tau} + \frac{1}{2} \left(\frac{(1+2p)\pi}{2} \right)^4 \left(\frac{\Delta t_{\text{open}}}{\tau} \right)^2 \right. \\
 & \quad \left. - \frac{1}{6} \left(\frac{(1+2p)\pi}{2} \right)^6 \left(\frac{\Delta t_{\text{open}}}{\tau} \right)^3 + \mathcal{O}[(\Delta t_{\text{open}}/\tau)^4] \right\} \left(\frac{\tau}{\Delta t_{\text{open}}} \right)^2 \\
 &= \frac{(1+2p)^2 \pi^2}{8} - \frac{(1+2p)^4 \pi^4}{96} \frac{\tau}{\Delta t_{\text{open}}} + \mathcal{O}[(\Delta t_{\text{open}}/\tau)^2].
 \end{aligned} \tag{S9}$$

Inserting this in Equation (S8) gives

$$\begin{aligned}
 \text{var}(x_{\text{msr}}) &\simeq \frac{64L^2}{\pi^6} \sum_{p=0}^{\infty} \frac{1}{(1+2p)^6} \left\{ \frac{(1+2p)^2 \pi^2}{8} - \frac{(1+2p)^4 \pi^4}{96} \frac{\tau}{\Delta t_{\text{open}}} \right\} \\
 &= \frac{L^2}{12} \left(1 - \frac{\Delta t_{\text{open}}}{\tau} \right),
 \end{aligned} \tag{S10}$$

where we used that $\sum_{p=0}^{\infty} (1+2p)^{-2} = \pi^2/8$ and $\sum_{p=0}^{\infty} (1+2p)^{-4} = \pi^4/96$ [Bickel (2007)].

2 CALCULATIONS FOR THE MSD FOR A PARTICLE IN A 1D BOX

For the MSD, we have to calculate $\langle x_{\text{msr}, n} x_{\text{msr}, 0} \rangle$ from Equation (21). The calculation is similar to the one for the variance, except that the time points t' and t in the time-ordered autocorrelation belong to different time-lapses [see Equation (S1)]. That is, $t \in [0, \Delta t]$ and $t' \in [n\Delta t, (n+1)\Delta t]$. So the spatial integrals are identical to those in Sec. 1, but the time integrals are different.

Carrying out the time integrals gives

$$\begin{aligned}
 & \int_0^{\Delta t} dt \int_{n\Delta t}^{(n+1)\Delta t} dt' \exp \left[- \left(\frac{(1+2p)\pi}{2} \right)^2 \frac{t' - t}{\tau} \right] s_1(t) s_{n+1}(t') \\
 &= \frac{1}{(\Delta t_{\text{open}})^2} \int_{\Delta t - \Delta t_{\text{open}}}^{\Delta t} dt \int_{(n+1)\Delta t - \Delta t_{\text{open}}}^{(n+1)\Delta t} dt' \exp \left[- \left(\frac{(1+2p)\pi}{2} \right)^2 \frac{t' - t}{\tau} \right] \\
 &= \left(\frac{\tau}{\Delta t_{\text{open}}} \right)^2 \left(\frac{2}{(1+2p)\pi} \right)^4 \exp \left[- \left(\frac{(1+2p)\pi}{2} \right)^2 \frac{n\Delta t + \Delta t_{\text{open}}}{\tau} \right] \\
 & \quad \times \left(1 - e^{\left(\frac{(1+2p)\pi}{2} \right)^2 \Delta t_{\text{open}}/\tau} \right)^2.
 \end{aligned} \tag{S11}$$

This is inserted in the expression for $\langle x_{\text{msr}, n} x_{\text{msr}, 0} \rangle$ and we arrive at Equation (22) in a similar way as when calculating the variance.

3 CALCULATIONS FOR THE VARIANCE AND MSD FOR A PARTICLE ON A 2D CIRCULAR DISC

The calculation follows the same steps as for the 1D case. All radial integrals below are from 0 to a , and all angular integrals are from 0 to 2π .

Due to symmetry it holds that $\langle \mathbf{r}_{\text{msr}} \rangle = (0, 0)$, so

$$\text{var}(\mathbf{r}_{\text{msr}}) = \langle \mathbf{r}_{\text{msr}}^2 \rangle = \frac{1}{\pi a^2} \int d\phi_0 \int dr_0 r_0 \langle \mathbf{r}_{\text{msr}}^2(r_0, \phi_0) \rangle_0. \quad (\text{S12})$$

We insert the definition of the measured position [see Equation (1)] and find

$$\langle \mathbf{r}_{\text{msr}}(x_0, y_0)^2 \rangle_0 = \frac{2}{(\Delta t_{\text{open}})^2} \int_{\Delta t - \Delta t_{\text{open}}}^{\Delta t} dt' \int_{\Delta t - \Delta t_{\text{open}}}^{t'} dt \langle \mathbf{r}(t|x_0, y_0) \cdot \mathbf{r}(t'|x_0, y_0) \rangle_{0, t' > t}, \quad (\text{S13})$$

and then write the time-ordered autocorrelation function as [recall that $\mathbf{r} = (x, y) = (r \cos \phi, r \sin \phi)$]

$$\begin{aligned} & \langle \mathbf{r}(t|x_0, y_0) \cdot \mathbf{r}(t'|x_0, y_0) \rangle_{0, t' > t} \\ &= \int r dr \int r' dr' \int d\phi \int d\phi' r r' (\cos \phi \cos \phi' + \sin \phi \sin \phi') \\ & \quad \times P(r, \phi, t|r_0, \phi_0, t_0) P(r', \phi', t'|r, \phi, t). \end{aligned} \quad (\text{S14})$$

Using Equation (27), the product of propagators are

$$\begin{aligned} & P(r, \phi, t|r_0, \phi_0, t_0) P(r', \phi', t'|r, \phi, t) \\ &= \left(\frac{1}{\pi a^2} \right)^2 + \left(\frac{1}{\pi a^2} \right)^2 \sum_{n'} \cos[n'(\phi' - \phi)] \sum_{m'}(n') + \left(\frac{1}{\pi a^2} \right)^2 \sum_n \cos[n(\phi - \phi_0)] \sum_m(n), \\ & \quad + \left(\frac{1}{\pi a^2} \right)^2 \sum_n \sum_{n'} \cos[n(\phi - \phi_0)] \cos[n'(\phi' - \phi)] \sum_m(n) \sum_{m'}(n'), \end{aligned} \quad (\text{S15})$$

with the short-hand notation

$$\sum_m(n) = \sum_{m=1}^{\infty} \frac{\alpha_{nm}^2}{\alpha_{nm}^2 - n^2} \exp \left[-\alpha_{nm}^2 \frac{t}{\tau} \right] \frac{J_n(\alpha_{nm} \frac{r}{a}) J_n(\alpha_{nm} \frac{r_0}{a})}{J_n(\alpha_{nm})^2}, \quad (\text{S16})$$

$$\sum_{m'}(n') = \sum_{m'=1}^{\infty} \frac{\alpha_{n'm'}^2}{\alpha_{n'm'}^2 - n'^2} \exp \left[-\alpha_{n'm'}^2 \frac{t'}{\tau} \right] \frac{J_{n'}(\alpha_{n'm'} \frac{r'}{a}) J_{n'}(\alpha_{n'm'} \frac{r}{a})}{J_{n'}(\alpha_{n'm'})^2}. \quad (\text{S17})$$

The integrals over the angles in Equation (S14) are

$$\int d\phi \int d\phi' \cos[n'(\phi' - \phi)] \cos \phi \cos \phi' = \pi^2 \delta_{n', \pm 1}, \quad (\text{S18})$$

$$\int d\phi \int d\phi' \cos[n(\phi - \phi_0)] \cos \phi \cos \phi' = 0, \quad (\text{S19})$$

$$\int d\phi \int d\phi' \cos[n(\phi - \phi_0)] \cos[n'(\phi' - \phi)] \cos \phi \cos \phi' = \pi^2 \delta_{n,0} \delta_{n', \pm 1} \quad (\text{S20})$$

$$\text{or } \frac{\pi^2}{2} \cos[2x_0] \delta_{n,2} \delta_{n', \pm 1},$$

and

$$\int d\phi \int d\phi' \cos[n'(\phi' - \phi)] \sin \phi \sin \phi' = \pi^2 \delta_{n', \pm 1}, \quad (\text{S21})$$

$$\int d\phi \int d\phi' \cos[n(\phi - \phi_0)] \sin \phi \sin \phi' = 0, \quad (\text{S22})$$

$$\int d\phi \int d\phi' \cos[n(\phi - \phi_0)] \cos[n'(\phi' - \phi)] \sin \phi \sin \phi' = \pi^2 \delta_{n,0} \delta_{n', \pm 1} \quad (\text{S23})$$

$$\text{or } \frac{-\pi^2}{2} \cos[2x_0] \delta_{n,2} \delta_{n', \pm 1}.$$

The integrals over $1/(\pi a^2)$ vanish for both cos and sin integrals.

This gives

$$\int d\phi \int d\phi' \int dr r^2 \int dr' r'^2 (\cos \phi \cos \phi' + \sin \phi \sin \phi') \times P(r, \phi, t | r_0, \phi_0, t_0) P(r', \phi', t' | r, \phi, t) \quad (\text{S24})$$

$$= \frac{4}{a^4} \left\{ \int dr \int dr' r^2 r'^2 \sum_{m'} (n' = 1) + \int dr r^2 \int dr' r'^2 \sum_m (n = 0) \sum_{m'} (n' = 1) \right\}.$$

where we used that $J_n(x) = (-1)^n J_{-n}(x)$, so

$$J_{-n} \left(\alpha_{nm} \frac{r}{a} \right) J_{-n} \left(\alpha_{nm} \frac{r'}{a} \right) \frac{\alpha_{nm}^2}{\alpha_{nm}^2 - (-n)^2} = J_n \left(\alpha_{nm} \frac{r}{a} \right) J_n \left(\alpha_{nm} \frac{r'}{a} \right) \frac{\alpha_{nm}^2}{\alpha_{nm}^2 - n^2}. \quad (\text{S25})$$

We need the following relations for the Bessel functions [Riseborough and Hänggi (1982)]

$$\int_0^a r^q J_n(\lambda r) dr = \left[\frac{r^q}{\lambda} J_{n+1}(\lambda r) \right]_0^a + \frac{n+1-q}{\lambda} \int_0^a r^{q-1} J_{n+1}(\lambda r) dr, \quad (\text{S26})$$

$$J_{n+1}(r) = \frac{n}{r} J_n(r) - J'_n(r), \quad (\text{S27})$$

$$J'_{n+1}(r) = \frac{n+1}{r} J'_n(r) + \left(1 - \frac{n(n+1)}{r^2} J_n(r) \right), \quad (\text{S28})$$

to calculate integrals on the form

$$\begin{aligned}
 \int_0^a dr r^2 J_1\left(\alpha_{1m} \frac{r}{a}\right) &= \left[\frac{r^2 a}{\alpha_{1m}} J_2\left(\alpha_{1m} \frac{r}{a}\right) \right]_0^a + \frac{1+1-2}{\alpha_{1m}/a} \int_0^a r J_{2+1}(r\alpha_{1m}/a) dr \\
 &= \frac{a^3}{\alpha_{1m}} [J_2(\alpha_{1m}) - J_2(0)] \\
 &= \frac{a^3}{\alpha_{1m}} \left[\frac{1}{\alpha_{1m}} J_1(\alpha_{1m}) - J_1'(\alpha_{1m}) \right] \\
 &= \frac{a^3}{\alpha_{1m}^2} J_1(\alpha_{1m}),
 \end{aligned} \tag{S29}$$

where we used Equation (S27), $J_2(0) = 0$, and $J_1'(\alpha_{1m}) = 0$.

Returning to Equation (S24), we write the last term as

$$\begin{aligned}
 &\int dr \int dr' r^2 r'^2 \sum_m (n=0) \sum_{m'} (n'=1) \\
 &= \int dr \int dr' r^2 r'^2 \left(\sum_{m=1}^{\infty} \frac{\alpha_{0m}^2}{\alpha_{0m}^2 - 0^2} \exp\left[-\alpha_{0m}^2 \frac{t-t_0}{\tau}\right] \frac{J_0(\alpha_{0m} \frac{r}{a}) J_0(\alpha_{0m} \frac{r_0}{a})}{J_0(\alpha_{0m})^2} \right) \\
 &\quad \times \left(\sum_{m'=1}^{\infty} \frac{\alpha_{1m'}^2}{\alpha_{1m'}^2 - 1^2} \exp\left[-\alpha_{1m'}^2 \frac{t'-t_0}{\tau}\right] \frac{J_1(\alpha_{1m'} \frac{r'}{a}) J_1(\alpha_{1m'} \frac{r}{a})}{J_1(\alpha_{1m'})^2} \right).
 \end{aligned} \tag{S30}$$

Expanding the product of the sums, we see that all terms are proportional to $J_0(\alpha_{0m} \frac{r_0}{a})$. Recall that in Equation (S12) we have an integral of the form $\int_0^a dr_0 r_0 [\dots]$, and as

$$\begin{aligned}
 \int_0^a dr_0 r_0 J_0\left(\alpha_{0m} \frac{r_0}{a}\right) &= \left[\frac{r_0 a}{\alpha_{0m}} J_1\left(\alpha_{0m} \frac{r_0}{a}\right) \right]_0^a + \frac{(0+1-1)a}{\alpha_{0m}} \int_0^a dr_0 J_1\left(\alpha_{1m} \frac{r_0}{a}\right) \\
 &= \frac{a^2}{\alpha_{0m}} J_1(\alpha_{0m}) \\
 &= \frac{a^2}{\alpha_{0m}} \left[\frac{0}{\alpha_{0m}} J_0(\alpha_{0m}) - J_0'(\alpha_{0m}) \right] \\
 &= 0,
 \end{aligned} \tag{S31}$$

we can neglect the last term in Equation (S24). The integral $\int_0^a dr_0 r_0$ over the first term in Equation (S24) simply gives πa^2 , which cancels the prefactor in Equation (S12).

Putting the pieces together, we find that Equation (S14) is

$$\begin{aligned}
 & \langle \mathbf{r}(t|x_0, y_0) \cdot \mathbf{r}(t'|x_0, y_0) \rangle_{0, t' > t} \\
 &= \frac{4}{a^4} \int dr \int dr' r^2 r'^2 \sum_{m=1}^{\infty} \frac{\alpha_{1m}^2}{\alpha_{1m}^2 - 1} \exp \left[-\alpha_{1m}^2 \frac{t' - t}{\tau} \right] \frac{J_1(\alpha_{1m} \frac{r}{a}) J_1(\alpha_{1m} \frac{r'}{a})}{J_1^2(\alpha_{1m})} + f(r_0) \\
 &= \frac{4}{a^4} \sum_{m=1}^{\infty} \frac{\alpha_{1m}^2}{\alpha_{1m}^2 - 1} \exp \left[-\alpha_{1m}^2 \frac{t' - t}{\tau} \right] \frac{\left(\frac{a^3}{\alpha_{1m}^2} \right)^2 J_1^2(\alpha_{1m})}{J_1^2(\alpha_{1m})} + f(r_0) \\
 &= 4a^2 \sum_{m=1}^{\infty} \frac{1}{\alpha_{1m}^2 (\alpha_{1m}^2 - 1)} \exp \left[-\alpha_{1m}^2 \frac{t' - t}{\tau} \right] + f(r_0)
 \end{aligned} \tag{S32}$$

where $f(r_0)$ represents terms that depend on r_0 .

We then insert the expression in Equation (S13) and carry out the time integrals [see, e.g., Equation (S6)], and the result is inserted in Equation (S4). The integral over initial positions removes terms with r_0 and leaves the others unchanged. So we finally arrive at the expression for the variance in Equation (28).

If we expand the exponential in Equation (28) in powers of $\Delta t_{\text{open}}/\tau$, we find that the variance becomes

$$\begin{aligned}
 & \text{var}(\mathbf{r}_{\text{msr}}) \\
 & \simeq \frac{8a^2}{(\Delta t_{\text{open}})^2} \sum_{m=1}^{\infty} \left\{ \frac{\tau \Delta t_{\text{open}}}{\alpha_{1m}^6 - \alpha_{1m}^4} - \frac{\tau^2}{\alpha_{1m}^8 - \alpha_{1m}^6} \right. \\
 & \quad \times \left(1 - \left[1 - \alpha_{1m}^2 \frac{\Delta t_{\text{open}}}{\tau} + \frac{1}{2} \alpha_{1m}^4 \frac{(\Delta t_{\text{open}})^2}{\tau^2} - \frac{1}{6} \alpha_{1m}^6 \frac{(\Delta t_{\text{open}})^3}{\tau^3} + \dots \right] \right) \left. \right\} \tag{S33} \\
 & \simeq 4a^2 \sum_{m=1}^{\infty} \frac{1}{\alpha_{1m}^2 (\alpha_{1m}^2 - 1)} - \frac{4a^2}{3} \sum_{m=1}^{\infty} \frac{1}{\alpha_{1m}^2 (\alpha_{1m}^2 - 1)} \\
 & = \frac{a^2}{2} \left(1 - \frac{4}{3} \frac{\Delta t_{\text{open}}}{\tau} \right),
 \end{aligned}$$

where the expressions for the sums may be found in [Bickel (2007)].

For $\Delta t_{\text{open}} \gg \tau$, we get

$$\text{var}(\mathbf{r}_{\text{msr}}) \simeq \frac{8a^2 \tau}{\Delta t_{\text{open}}} \sum_{m=1}^{\infty} \frac{1}{\alpha_{1m}^4 (\alpha_{1m}^2 - 1)} = \frac{7a^2 \tau}{24 \Delta t_{\text{open}}}. \tag{S34}$$

Here we used that the sum equals $7/192$, which is seen by writing the fraction in the sum as $[x^4(x^2-1)]^{-1} = (x^2-1)^{-1} - x^{-4} - x^{-2}$ and using the method described in App. C in Riseborough and Hänggi (1982).

As for the case of a particle in a 1D box, the calculation for the mean-squared displacement for the 2D case is similar to the calculation of the variance, except that the time integrals are different [see Sec. 2].

4 CALCULATIONS FOR THE VARIANCE AND MSD FOR A PARTICLE CONFINED IN A 3D SPHERE

The calculation is similar to the 1D and 2D cases. All radial integrals over r are from 0 to a , all integrals over θ are from 0 to π , and all integrals over ϕ are from 0 to 2π . Due to symmetry it holds that $\langle \mathbf{r}_{\text{msr}} \rangle = (0, 0, 0)$, so

$$\text{var}(\mathbf{r}_{\text{msr}}) = \langle \mathbf{r}_{\text{msr}}^2 \rangle = \frac{3}{4\pi a^3} \int_0^\pi d\theta \sin \theta \int_0^\pi d\phi \int_0^a dr_0 r_0^2 \langle \mathbf{r}_{\text{msr}}^2(r_0, \theta_0, \phi_0) \rangle_0. \quad (\text{S35})$$

We insert the definition of the measured position [see Equation (1)] and find

$$\langle \mathbf{r}_{\text{msr}}^2(r_0, \theta_0, \phi_0) \rangle_0 = \frac{2}{(\Delta t_{\text{open}})^2} \int_{\Delta t - \Delta t_{\text{open}}}^{\Delta t} dt' \int_{\Delta t - \Delta t_{\text{open}}}^{t'} dt \langle \mathbf{r}(t|r_0, \theta_0, \phi_0) \cdot \mathbf{r}(t'|r_0, \theta_0, \phi_0) \rangle_{0, t' > t}, \quad (\text{S36})$$

and then write the time-ordered autocorrelation function as

$$\begin{aligned} & \langle \mathbf{r}(t|r_0, \theta_0, \phi_0) \cdot \mathbf{r}(t'|r, \theta, \phi) \rangle_{0, t' > t} \\ &= \int dr r^2 \int dr' r'^2 \int d\theta \sin \theta \int d\theta' \sin \theta' \int d\phi \int d\phi' \\ & \quad \times rr' \cos \gamma P(r', \theta', \phi', t'|r, \theta, \phi, t) P(r, \theta, \phi, t|r_0, \theta_0, \phi_0, t_0). \end{aligned} \quad (\text{S37})$$

Here we recall that $\langle \mathbf{r}(t|r_0, \theta_0, \phi_0) \cdot \mathbf{r}(t'|r_0, \theta_0, \phi_0) \rangle_{0, t' > t} = \langle \mathbf{r}(t|r_0, \theta_0, \phi_0) \cdot \mathbf{r}(t'|r, \theta, \phi) \rangle_{0, t' > t}$ as the points are on the same trajectory and define $\gamma = \gamma(\theta', \phi', \theta, \phi)$ to be the angle between $\mathbf{r}(t|r, \theta, \phi)$ and $\mathbf{r}(t'|r', \theta', \phi')$. Following Bickel (2007), we express this angle in terms of spherical harmonics,

$$\cos \gamma = \frac{4\pi}{3} \sum_{m=-1}^{m=1} Y_{1m}^*(\theta', \phi') Y_{1m}(\theta, \phi). \quad (\text{S38})$$

The product of the propagators [see Equation (34)] and the angle is

$$\begin{aligned} & \cos \gamma P(r', \theta', \phi', t'|r, \theta, \phi, t) P(r, \theta, \phi, t|r_0, \theta_0, \phi_0, t_0) \\ &= \left[\frac{3}{4\pi a^3} + \frac{2}{a^3} \sum_{\ell=0}^{\infty} \sum_{m=-\ell}^{\ell} Y_{\ell m}(\theta', \phi') Y_{\ell m}^*(\theta, \phi) R_{\ell}(r', r, t', t) \right] \\ & \quad \times \left[\frac{3}{4\pi a^3} + \frac{2}{a^3} \sum_{\ell'=0}^{\infty} \sum_{m'=-\ell'}^{\ell'} Y_{\ell' m'}(\theta, \phi) Y_{\ell' m'}^*(\theta_0, \phi_0) R_{\ell'}(r, r_0, t, t_0) \right] \\ & \quad \times \frac{4\pi}{3} \sum_{m''=-1}^1 Y_{1m''}^*(\theta', \phi') Y_{1m''}(\theta, \phi), \end{aligned} \quad (\text{S39})$$

where we introduced the notation for the radial and temporal part of the propagator

$$R_{\ell}(r', r, t', t) = \sum_{n=1}^{\infty} \frac{\beta_{\ell n}^2}{\beta_{\ell n}^2 - \ell(\ell+1)} \exp \left[-\beta_{\ell n}^2 \frac{t' - t}{\tau} \right] \frac{j_{\ell} \left(\beta_{\ell n} \frac{r'}{a} \right) j_{\ell} \left(\beta_{\ell n} \frac{r}{a} \right)}{j_{\ell}(\beta_{\ell n})^2}. \quad (\text{S40})$$

Due to the orthogonality relation of spherical harmonics,

$$\int_0^\pi d\theta \int_0^{2\pi} d\phi Y_{\ell m}^*(\theta, \phi) Y_{\ell' m'}(\theta, \phi) \sin \theta = \delta_{\ell, \ell'} \delta_{m, m'}, \quad (\text{S41})$$

terms with only $Y_{1m''}^*(\theta, \phi)$ or $Y_{1m''}(\theta', \phi')$ vanish when inserting Equation (S39) in Equation (S37) and integrating over (θ, ϕ) or (θ', ϕ') , as the constant term is proportional to Y_{00} . So, with the use of Equations (S39) and (S41), Equation (S37) becomes

$$\begin{aligned} & \langle \mathbf{r}(t|r_0, \theta_0, \phi_0) \cdot \mathbf{r}(t'|r, \theta, \phi) \rangle_{0, t' > t} \\ &= \int dr r^2 \int dr' r'^2 \int d\theta \sin \theta \int d\theta' \sin \theta' \int d\phi \int d\phi' \\ & \quad \times \left[r r' \frac{3}{4\pi a^3} \times \frac{2}{a^3} \sum_{\ell=0}^{\infty} \sum_{m=-\ell}^{\ell} Y_{\ell m}(\theta', \phi') Y_{\ell m}^*(\theta, \phi) R_{\ell}(r', r, t', t) \right. \\ & \quad \left. \times \frac{4\pi}{3} \sum_{m''=-1}^1 Y_{1m''}^*(\theta', \phi') Y_{1m''}(\theta, \phi) \right] + [\text{terms with } Y_{\ell m}^*(\theta_0, \phi_0)] \quad (\text{S42}) \\ &= \frac{6}{a^6} \int_0^a dr' r'^3 \int_0^a dr r^3 r(r', r, t', t) + [\text{terms with } Y_{\ell m}^*(\theta_0, \phi_0)] \\ &= \frac{6}{a^6} \int_0^a dr' r'^3 \int_0^a dr r^3 \sum_{n=1}^{\infty} \frac{\beta_{1n}^2}{\beta_{1n}^2 - 2} \exp \left[-\beta_{1n}^2 \frac{t' - t}{\tau} \right] \frac{j_1 \left(\beta_{1n} \frac{r'}{a} \right) j_1 \left(\beta_{1n} \frac{r}{a} \right)}{j_1(\beta_{1n})^2} \\ & \quad + [\text{terms with } Y_{\ell m}^*(\theta_0, \phi_0)]. \end{aligned}$$

The radial integrals are

$$\begin{aligned} \int_0^a dr r^3 j_1 \left(\beta_{1n} \frac{r}{a} \right) &= \int_0^a dr r^3 \left\{ \frac{\sin \left(\beta_{1n} \frac{r}{a} \right)}{\left(\beta_{1n} \frac{r}{a} \right)^2} - \frac{\cos \left(\beta_{1n} \frac{r}{a} \right)}{\beta_{1n} \frac{r}{a}} \right\} \\ &= \frac{a^4}{\beta_{1n}} \left\{ \frac{3 \sin \beta_{1n}}{\beta_{1n}^3} - \frac{3 \cos \beta_{1n}}{\beta_{1n}^2} - \frac{\sin \beta_{1n}}{\beta_{1n}} \right\} \quad (\text{S43}) \\ &= \frac{a^4}{\beta_{1n}} \left\{ \frac{1}{\beta_{1n}} j_1(\beta_{1n}) - j_1'(\beta_{1n}) \right\} \\ &= \frac{a^4}{\beta_{1n}^2} j_1(\beta_{1n}), \end{aligned}$$

where we used that $j_1(x) = \sin(x)/x^2 - \cos(x)/x$, $j_1'(x) = 2 \cos(x)/x^2 - 2 \sin(x)/x^3 + \sin x/x$, and $j_1'(\beta_{1n}) = 0$ per definition of β_{1n} . So Equation (S42) becomes

$$\begin{aligned} & \langle \mathbf{r}(t|r_0, \theta_0, \phi_0) \cdot \mathbf{r}(t'|r, \theta, \phi) \rangle_{0, t' > t} \\ &= 6a^2 \sum_{n=1}^{\infty} \frac{1}{\beta_{1n}^2 (\beta_{1n}^2 - 2)} \exp \left[-\beta_{1n}^2 \frac{t - t'}{\tau} \right] + [\text{terms with } Y_{\ell m}^*(\theta_0, \phi_0)]. \quad (\text{S44}) \end{aligned}$$

We insert this in Equation (S36) and carry out the integrals over time. The result is inserted in Equation (S35), and when integrating over all initial positions, the terms with $Y_{\ell m}^*(\theta_0, \phi_0)$ vanish and we get the final result for the variance of the measured positions [Equation (35)].

An expansion to lowest orders in $\Delta t_{\text{open}}/\tau$ gives

$$\begin{aligned}
 & \text{var}(\mathbf{r}_{\text{msr}}) \\
 & \simeq \frac{12a^2}{(\Delta t_{\text{open}})^2} \sum_{n=1}^{\infty} \left[\frac{\tau \Delta t_{\text{open}}}{\beta_{1n}^4 (\beta_{1n}^2 - 2)} - \frac{\tau^2}{\beta_{1n}^6 (\beta_{1n}^2 - 2)} \left(\beta_{1n}^2 \frac{\Delta t_{\text{open}}}{\tau} - \frac{1}{2} \left[\beta_{1n}^2 \frac{\Delta t_{\text{open}}}{\tau} \right]^2 + \frac{1}{6} \left[\beta_{1n}^2 \frac{\Delta t_{\text{open}}}{\tau} \right]^3 \right) \right] \\
 & = \frac{12a^2}{(\Delta t_{\text{open}})^2} \sum_{n=1}^{\infty} \left[\frac{1}{2} \frac{\Delta t_{\text{open}}^2}{\beta_{1n}^2 (\beta_{1n}^2 - 2)} - \frac{1}{6} \frac{1}{\beta_{1n}^2 - 2} \frac{(\Delta t_{\text{open}})^3}{\tau} \right] \\
 & = \left(6 \sum_{n=1}^{\infty} \frac{1}{\beta_{1n}^2 (\beta_{1n}^2 - 2)} - \frac{2\Delta t_{\text{open}}}{\tau} \sum_{n=1}^{\infty} \frac{1}{\beta_{1n}^2 - 2} \right) a^2 \\
 & = \frac{3a^2}{5} \left(1 - \frac{5}{3} \frac{\Delta t_{\text{open}}}{\tau} \right),
 \end{aligned} \tag{S45}$$

where we in the last step used that the first and second sums may be found to equal, respectively, $(1 - 3/5)/4 = 1/10$ and $1/2$ [Bickel (2007)].

As for a particle in a 1D box or on a 2D disc, the calculation for the mean-squared displacement for the 3D case is similar to the calculation of the variance, except that the time integrals are different [see Sec. 2].

5 PLOTS OF THE VARIANCE FOR 1D DIFFUSION

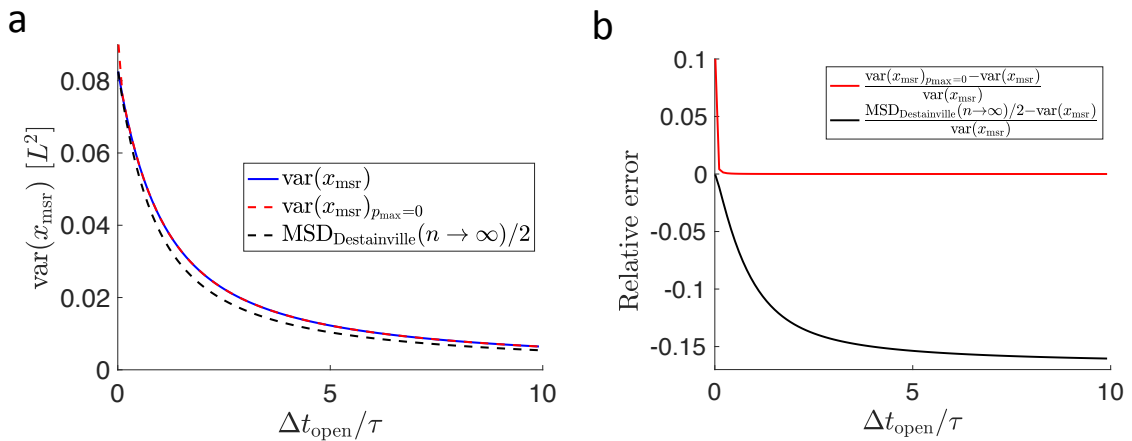


Figure S1. (a) The variance of measured positions as function of $\Delta t_{\text{open}}/\tau$ for the exact expression in Equation (18) (blue full line) [same as in Figure 2d], the expression in Equation (18) but keeping only the term with $p = 0$ in the sum (dashed, red line), and the variance obtained as half the value of the expression for the MSD in Equation (25) for $n \rightarrow \infty$ (dashed, black line). (b) Relative differences between the approximate and exact results.

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