Supplementary Information (SI) for Physical Chemistry Chemical Physics.

Supplementary Information to

The effect of localisation imprecision on quantification of the directionality of motion for single particle tracking applications

Explicit mathematical derivations

Paula Melero Martinez and Christoffer Åberg

Groningen Research Institute of Pharmacy, University of Groningen, A. Deusinglaan 1, 9713 AV Groningen, The Netherlands.

Stationary particles

One dimension

We start by considering only one dimension, as these results will prove useful for the two-dimensional situation. We write the *observed* position of a particle in one dimension, $\tilde{X}(t)$, as

$$\tilde{X}(t) = X(t) + \Xi(t)$$

where $X(t)$ is the *real* position of the particle and $\Xi(t)$ is the error in localisation. Naturally, when considering stationary particles, we assume that the real position of the particle is independent of time, that is, $X(t) = X$. Furthermore, the error in localisation we assume to be a (one-dimensional) normally distributed stochastic variable with 0 mean and a standard deviation equal to the localisation imprecision, σ, *viz.*

$$\Xi(t) \sim \mathcal{N}(0, \sigma^2).$$

Importantly, we assume that the localisation imprecision for different times, t, are independent.

In one dimension, the change in direction corresponding to Equation (1) of the main text is given by

$$\cos\tilde{\Theta} = \frac{\Delta\tilde{X}_{\Delta\tau,0}\Delta\tilde{X}_{\tau+\Delta\tau,\tau}}{|\Delta\tilde{X}_{\Delta\tau,0}||\Delta\tilde{X}_{\tau+\Delta\tau,\tau}|} \tag{1}$$

where

$$\Delta\tilde{X}_{\Delta\tau,0} \equiv \tilde{X}(\Delta\tau) - \tilde{X}(0) \equiv X + \Xi(\Delta\tau) - X - \Xi(0) = \Xi(\Delta\tau) - \Xi(0) \quad (2)$$

and

$$\Delta\tilde{X}_{\tau+\Delta\tau,\tau} \equiv \tilde{X}(\tau+\Delta\tau) - \tilde{X}(\tau) \equiv X + \Xi(\tau+\Delta\tau) - X - \Xi(\tau) = \Xi(\tau+\Delta\tau) - \Xi(\tau). \quad (3)$$

We are interested in three different cases. The first case is the change in direction comparing a displacement from $t = 0$ to $t = \Delta\tau$ with itself. Mathematically this corresponds to $\tau = 0$ (and $\Delta\tau \neq 0$). Clearly the angle in this case always vanishes, that is, $\tilde{\Theta} = 0$ or $\cos\tilde{\Theta} = 1$. This case is trivial and we will not refer to it much.

The second case is the most interesting one. Here we consider the change in direction comparing a displacement from $t = 0$ to $t = \Delta\tau$ with a displacement from $t = \Delta\tau$ to $t = 2\Delta\tau$, that is, a change in direction between successive displacement vectors. Mathematically, this corresponds to $\tau = \Delta\tau$ (and $\Delta\tau \neq 0$).

Finally, we also consider the change in direction comparing a displacement from $t = 0$ to $t = \Delta\tau$ with a later displacement than the successive one. Mathematically, this corresponds to $\tau > \Delta\tau$ (and $\Delta\tau \neq 0$).

Equations (2) and (3) express that $\Delta\tilde{X}_{\Delta\tau,0}$ and $\Delta\tilde{X}_{\tau+\Delta\tau,\tau}$ are both differences of normally distributed variables of mean 0 and variance σ^2 (the localisation imprecision squared). It is well-known that the difference of two normally distributed independent variables is also a normally distributed variable, and specifically, that if the two variables have mean 0 and the same variance, then their difference has mean 0 and double the variance. In our case we consequently have

$$\begin{aligned} \Delta\tilde{X}_{\Delta\tau,0} &\sim \mathcal{N}(0, 2\sigma^2) \equiv \mathcal{N}(0, \sigma_\tau^2) \\ \Delta\tilde{X}_{\tau+\Delta\tau,\tau} &\sim \mathcal{N}(0, 2\sigma^2) \equiv \mathcal{N}(0, \sigma_\tau^2) \end{aligned} \quad (4)$$

(as long as $\Delta\tau \neq 0$). We have here introduced the standard deviation σ_τ as it will prove useful to generalise the presentation to the case of particles moving by Brownian motion.

Distribution of the scalar product

$\cos\tilde{\Theta}$, and $\tilde{\Theta}$ itself even more so, is a complicated stochastic variable as defined from Equation (1). For simplicity, we thus start by considering just the numerator

$$\Delta\tilde{X}_{\Delta\tau,0}\Delta\tilde{X}_{\tau+\Delta\tau,\tau}. \quad (5)$$

which is the one-dimensional equivalent of the scalar product of two displacements in two dimensions. The mean of this quantity is also closely related to the velocity autocorrelation function, C_v, which in our nomenclature is defined as[1]

$$C_{v;\Delta\tau} = \frac{1}{\Delta\tau^2}\langle \Delta\tilde{X}_{\Delta\tau,0}\Delta\tilde{X}_{\tau+\Delta\tau,\tau}\rangle. \tag{6}$$

We will consequently consider also the mean of the scalar product.

As expressed by Equation (5), clearly the scalar product is the product of two differences of normally distributed random variables as expressed in Equation (4). To calculate its distribution we thus need to find the distribution corresponding to a product of normally distributed random variables. The distribution, $f_{R'}$, for the product, R', of two 0 mean unit variance normally distributed variables has been derived by Grishchuk[2] (and earlier by Springer[3] but with a typo). It is given by

$$f_{R'}(r') = \frac{1}{\pi\sqrt{1-\rho^2}}\exp\left(\frac{\rho r'}{1-\rho^2}\right)K_0\left(\frac{|r'|}{1-\rho^2}\right) \tag{7}$$

where K_0 is the 0th order modified Bessel function of the second kind and ρ the correlation coefficient between the two variables. In order to use this result, we consider the stochastic variables $\Delta\tilde{X}_{\Delta\tau,0}/\sigma_\tau$ and $\Delta\tilde{X}_{\tau+\Delta\tau,\tau}/\sigma_\tau$. They clearly have unit variance so Equation (7) applies. Furthermore, the distribution, f_R, of $R = \Delta\tilde{X}_{\Delta\tau,0}\Delta\tilde{X}_{\tau+\Delta\tau,\tau}$, which is what we are actually interested in, is given by

$$f_R(r) = \frac{1}{\sigma_\tau^2}f_{R'}\left(\frac{r}{\sigma_\tau^2}\right) = \frac{1}{\pi\sigma_\tau^2\sqrt{1-\rho^2}}\exp\left(\frac{\rho r}{\sigma_\tau^2(1-\rho^2)}\right)K_0\left(\frac{|r|}{\sigma_\tau^2(1-\rho^2)}\right). \tag{8}$$

The correlation coefficient, ρ, here is the correlation coefficient of the unit variance variables. It is given by

$$\rho \equiv \mathrm{cov}\left(\frac{\Delta\tilde{X}_{\Delta\tau,0}}{\sigma_\tau}, \frac{\Delta\tilde{X}_{\tau+\Delta\tau,\tau}}{\sigma_\tau}\right) = \frac{\mathrm{cov}(\Delta\tilde{\mathrm{X}}_{\Delta\tau,0}, \Delta\tilde{\mathrm{X}}_{\tau+\Delta\tau,\tau})}{\sigma_\tau^2} = \frac{\langle \Delta\tilde{X}_{\Delta\tau,0}\Delta\tilde{X}_{\tau+\Delta\tau,\tau}\rangle - \langle \Delta\tilde{X}_{\Delta\tau,0}\rangle\langle \Delta\tilde{X}_{\tau+\Delta\tau,\tau}\rangle}{\sigma_\tau^2} = \frac{\langle \Delta\tilde{X}_{\Delta\tau,0}\Delta\tilde{X}_{\tau+\Delta\tau,\tau}\rangle}{\sigma_\tau^2} \tag{9}$$

where we have used that $\Delta\tilde{X}_{\Delta\tau,0}$ and $\Delta\tilde{X}_{\tau+\Delta\tau,\tau}$ both have 0 mean. Furthermore, we have that

$$\langle \Delta\tilde{X}_{\Delta\tau,0}\Delta\tilde{X}_{\tau+\Delta\tau,\tau}\rangle = \big\langle \big(\Xi(\Delta\tau) - \Xi(0)\big)\big(\Xi(\tau+\Delta\tau) - \Xi(\tau)\big)\big\rangle = \langle \Xi(\Delta\tau)\Xi(\tau+\Delta\tau)\rangle - \langle \Xi(\Delta\tau)\Xi(\tau)\rangle - \langle \Xi(0)\Xi(\tau+\Delta\tau)\rangle + \langle \Xi(0)\Xi(\tau)\rangle. \tag{10}$$

Let us now consider our two cases. First, for successive displacements, that is, $\tau = \Delta\tau$ (but $\Delta\tau \neq 0$), we find

$$\langle \Delta\tilde{X}_{\Delta\tau,0}\Delta\tilde{X}_{\tau+\Delta\tau,\tau}\rangle = -\langle \Xi(\Delta\tau)\Xi(\tau)\rangle = -\sigma^2 \tag{11}$$

where we have used that $\Xi(t)$ at different times are indepedent and that $\Xi(t)$ has variance σ^2. Using the definition of σ_τ as expressed in Equation (4) we then find

$$\rho = \frac{\langle \Delta\tilde{X}_{\Delta\tau,0}\Delta\tilde{X}_{\tau+\Delta\tau,\tau}\rangle}{\sigma_\tau^2} = -\frac{\sigma^2}{2\sigma^2} = -\tfrac{1}{2}. \tag{12}$$

Inserted into Equation (8), we then find that the distribution of the product $R = \Delta\tilde{X}_{\Delta\tau,0}\Delta\tilde{X}_{\tau+\Delta\tau,\tau}$ is given by

$$f_R(r) = \frac{1}{\sqrt{3}\pi\sigma^2}\exp\left(-\frac{r}{3\sigma^2}\right)K_0\left(\frac{2|r|}{3\sigma^2}\right).$$

Second, we consider displacements separated in time from each other, that is, $\tau > \Delta\tau$ (and $\Delta\tau \neq 0$). Again using the independence of $\Xi(t)$ at different times we then find

$$\langle \Delta\tilde{X}_{\Delta\tau,0}\Delta\tilde{X}_{\tau+\Delta\tau,\tau}\rangle = 0 \tag{13}$$

and consequently

$$\rho = 0. \tag{14}$$

From Equation (8) we then have that the distribution of $R = \Delta\tilde{X}_{\Delta\tau,0}\Delta\tilde{X}_{\tau+\Delta\tau,\tau}$ is given by

$$f_R(r) = \frac{1}{2\pi\sigma^2}K_0\left(\frac{|r|}{2\sigma^2}\right).$$

The mean of the scalar product

As already discussed, the mean of the scalar product is very much related to the velocity autocorrelation function [Equation (6)], so it is of interest to also calculate this mean. However, this is something we have largely already done above in terms of Equations (11) and (13) for successive displacements and displacements separated in time from each other, respectively. The only case that remains is the correlation of a displacement with itself, which is simply the variance of the displacement, so

$$\langle \Delta\tilde{X}_{\Delta\tau,0}\Delta\tilde{X}_{\tau+\Delta\tau,\tau}\rangle(\tau = 0) = \sigma_\tau^2 = 2\sigma^2$$

where final equality follows from Equation (4).

Overall, we thus have

$$\langle \Delta\tilde{X}_{\Delta\tau,0}\Delta\tilde{X}_{\tau+\Delta\tau,\tau}\rangle(\tau) = \begin{cases} 2\sigma^2 & \tau = 0 \\ -\sigma^2 & \tau = \Delta\tau \\ 0 & \tau > \Delta\tau. \end{cases} \tag{15}$$

Distribution of the angle between displacements

In one dimension we can actually use the distribution of the scalar product to calculate, in a fairly simple fashion, the distribution of the angle between displacements, $\tilde{\Theta}$, or, equivalently, the cosine of this angle, $\cos\tilde{\Theta}$. In one dimension this angle can only be 0 or π or, equivalently, $\cos\tilde{\Theta}$ can only be 1 or -1. The key observation is then that the denominator of Equation (1) is always positive. Hence it must be the case that the numerator determines whether the whole expression is -1 or 1; more specifically, it must be the case that

$$P[\cos\tilde{\Theta} = -1] = P[\tilde{\Theta} = \pi] = \int_{-\infty}^{0} f_R(r)\mathrm{d}r$$

and

$$P[\cos\tilde{\Theta} = 1] = P[\tilde{\Theta} = 0] = \int_{0}^{\infty} f_R(r)\mathrm{d}r.$$

We now tackle the second integral. According to Equation (8) it reads

$$\int_0^\infty f_R(r)\mathrm{d}r = \frac{1}{\pi\sigma_\tau^2\sqrt{1-\rho^2}}\int_0^\infty \exp\left(\frac{\rho r}{\sigma_\tau^2(1-\rho^2)}\right) K_0\left(\frac{|r|}{\sigma_\tau^2(1-\rho^2)}\right)\mathrm{d}r.$$

A change of variables to

$$u = \frac{r}{\sigma_\tau^2(1-\rho^2)}$$

and the trivial observation that $|r| = r$ when $r > 0$ gives us the integral

$$\frac{1}{\pi\sigma_\tau^2\sqrt{1-\rho^2}}\sigma_\tau^2(1-\rho^2)\int_0^\infty \mathrm{e}^{\rho u}K_0(u)\mathrm{d}u = \frac{\sqrt{1-\rho^2}}{\pi}\frac{\arccos(-\rho)}{\sqrt{1-\rho^2}} = \frac{\arccos(-\rho)}{\pi}$$

where have used Gradshteyn & Ryzhik[4] [Equation (6.611)] to evaluate the integral.

We thus find

$$P[\cos\tilde{\Theta} = 1] = P[\tilde{\Theta} = 0] = \frac{\arccos(-\rho)}{\pi} \tag{16}$$

and

$$P[\cos\tilde{\Theta} = -1] = P[\tilde{\Theta} = \pi] = 1 - P[\tilde{\Theta} = 0] = \frac{\pi - \arccos(-\rho)}{\pi}. \tag{17}$$

For successive displacements, we have $\rho = -\frac{1}{2}$ [Equation (12)] and thus we find

$$P[\cos\tilde{\Theta} = 1] = P[\tilde{\Theta} = 0] = \tfrac{1}{3}$$

and

$$P[\cos\tilde{\Theta} = -1] = P[\tilde{\Theta} = \pi] = \tfrac{2}{3};$$

for displacements separated in time from each other, we instead have $\rho = 0$ [Equation (14)] and thus find simply

$$P[\cos\tilde{\Theta} = 1] = P[\tilde{\Theta} = 0] = \tfrac{1}{2}$$

and

$$P[\cos\tilde{\Theta} = -1] = P[\tilde{\Theta} = \pi] = \tfrac{1}{2}$$

as expected based on the symmetry of the distribution.

Mean cosine of the angle between displacements

From this, we can derive the mean cosine of the angle. Clearly we have

$$\langle\cos\tilde{\Theta}\rangle = (+1)\cdot P[\cos\tilde{\Theta} = 1] + (-1)\cdot P[\cos\tilde{\Theta} = -1] = 2P[\cos\tilde{\Theta} = 1] - 1 = \frac{2}{\pi}\arccos(-\rho) - 1 \quad (18)$$

using Equation (16). For successive displacements, where we have $\rho = -\frac{1}{2}$ [Equation (12)], we then find

$$\langle\cos\tilde{\Theta}\rangle = \frac{2}{\pi}\frac{\pi}{3} - 1 = -\tfrac{1}{3} \quad (19)$$

while for displacements separated in time from each other, where we have $\rho = 0$ [Equation (14)], we find

$$\langle\cos\tilde{\Theta}\rangle = \frac{2}{\pi}\frac{\pi}{2} - 1 = 0. \quad (20)$$

Mean angle between displacements

Likewise, we can derive the mean angle between displacements from

$$\langle\tilde{\Theta}\rangle = 0\cdot P[\tilde{\Theta} = 0] + \pi\cdot P[\tilde{\Theta} = \pi] = \pi P[\tilde{\Theta} = \pi] = \pi - \arccos(-\rho) \quad (21)$$

using Equation (17). For successive displacements, where again we have $\rho = -\frac{1}{2}$ [Equation (12)], we then find

$$\langle\tilde{\Theta}\rangle = \frac{2\pi}{3}; \quad (22)$$

while for displacements separated in time from each other, where we have $\rho = 0$ [Equation (14)], we find

$$\langle\tilde{\Theta}\rangle = \frac{\pi}{2}. \quad (23)$$

Two dimensions

Distribution of the scalar product

We now consider the two-dimensional case, where we are interested in the angle given by

$$\cos\tilde{\Theta} = \frac{\mathbf{\Delta\tilde{R}}_{\Delta\tau,0}\cdot\mathbf{\Delta\tilde{R}}_{\tau+\Delta\tau,\tau}}{|\mathbf{\Delta\tilde{R}}_{\Delta\tau,0}||\mathbf{\Delta\tilde{R}}_{\tau+\Delta\tau,\tau}|}. \tag{24}$$

Again we start by considering only the numerator, that is, the scalar product

$$\mathbf{\Delta\tilde{R}}_{\Delta\tau,0}\cdot\mathbf{\Delta\tilde{R}}_{\tau+\Delta\tau,\tau} = \Delta\tilde{X}_{\Delta\tau,0}\Delta\tilde{X}_{\tau+\Delta\tau,\tau} + \Delta\tilde{Y}_{\Delta\tau,0}\Delta\tilde{Y}_{\tau+\Delta\tau,\tau}. \tag{25}$$

This is the sum of two independent variables whose distributions we know from our above discussion of the one-dimensional case. The sum of two independent variables is a convolution and since the two products have the same distribution, it is going to be a convolution of a function with itself.

Convolutions are typically easier in Fourier space; if we define our Fourier transform as

$$\tilde{f}(k) \equiv \frac{1}{\sqrt{2\pi}}\int_{-\infty}^{\infty} f(x)\mathrm{e}^{ikx}\mathrm{d}x$$

with inverse

$$f(x) = \frac{1}{\sqrt{2\pi}}\int_{-\infty}^{\infty} \tilde{f}(k)\mathrm{e}^{-ikx}\mathrm{d}k$$

then the Fourier transform of the convolution

$$(f*g)(x) \equiv \int_{-\infty}^{\infty} f(\xi)g(x-\xi)\mathrm{d}\xi$$

is

$$\sqrt{2\pi}\tilde{f}\tilde{g}.$$

To use this, we then need to Fourier transform the distribution given by Equation (8). This appears to be a bit complicated to perform, so instead we note that the convolution we are interested in is

$$\begin{aligned}(f_R*f_R)(s) \equiv \int_{-\infty}^{\infty} f_R(r)f_R(s-r)\mathrm{d}r = \\ \frac{1}{\pi^2\sigma_\tau^4(1-\rho^2)}\int_{-\infty}^{\infty}\exp\left(\frac{\rho r}{\sigma_\tau^2(1-\rho^2)}\right)K_0\left(\frac{|r|}{\sigma_\tau^2(1-\rho^2)}\right)\times \\ \exp\left(\frac{\rho(s-r)}{\sigma_\tau^2(1-\rho^2)}\right)K_0\left(\frac{|s-r|}{\sigma_\tau^2(1-\rho^2)}\right)\mathrm{d}r = \\ \frac{1}{\pi^2\sigma_\tau^4(1-\rho^2)}\exp\left(\frac{\rho s}{\sigma_\tau^2(1-\rho^2)}\right)\times \\ \int_{-\infty}^{\infty}K_0\left(\frac{|r|}{\sigma_\tau^2(1-\rho^2)}\right)K_0\left(\frac{|s-r|}{\sigma_\tau^2(1-\rho^2)}\right)\mathrm{d}r \equiv \\ \frac{1}{\pi^2\sigma_\tau^4(1-\rho^2)}\exp\left(\frac{\rho s}{\sigma_\tau^2(1-\rho^2)}\right)(g*g)(s)\end{aligned} \tag{26}$$

where have introduced the function

$$g(r) \equiv K_0\left(\frac{|r|}{\sigma_\tau^2(1-\rho^2)}\right)$$

and identified the convolution of this function with itself.

To calculate the integral, we thus calculate the Fourier transform of the latter function

$$\tilde{g}(k) = \frac{1}{\sqrt{2\pi}}\int_{-\infty}^{\infty} g(r)\mathrm{e}^{ikr}\mathrm{d}r = \frac{1}{\sqrt{2\pi}}\int_{-\infty}^{\infty} K_0\left(\frac{|r|}{\sigma_\tau^2(1-\rho^2)}\right)\mathrm{e}^{ikr}\mathrm{d}r =$$
$$\frac{1}{\sqrt{2\pi}}2\int_0^{\infty} K_0\left(\frac{r}{\sigma_\tau^2(1-\rho^2)}\right)\cos(kr)\mathrm{d}r = \frac{1}{\sqrt{2\pi}}2\frac{\pi}{2}\frac{1}{\sqrt{k^2+\frac{1}{\sigma_\tau^4(1-\rho^2)^2}}}$$

using Gradshteyn & Ryzhik[4] [Equation (6.671)] to evaluate the integral. Thus we have that the Fourier transform of $g * g$ is

$$\sqrt{2\pi}\big(\tilde{g}(k)\big)^2 = \sqrt{2\pi}\frac{\pi^2}{2\pi}\frac{1}{k^2+\frac{1}{\sigma_\tau^4(1-\rho^2)^2}} = \sqrt{2\pi}\frac{\pi}{2}\frac{1}{k^2+\frac{1}{\sigma_\tau^4(1-\rho^2)^2}}.$$

Inverting this, we find

$$(g * g)(s) = \frac{1}{\sqrt{2\pi}}\int_{-\infty}^{\infty}\sqrt{2\pi}\frac{\pi}{2}\frac{1}{k^2+\frac{1}{\sigma_\tau^4(1-\rho^2)^2}}\mathrm{e}^{-iks}\mathrm{d}k =$$
$$\frac{\pi}{2}2\int_0^{\infty}\frac{\cos(ks)}{k^2+\frac{1}{\sigma_\tau^4(1-\rho^2)^2}}\mathrm{d}k = \pi\frac{\pi}{2}\sigma_\tau^2(1-\rho^2)\exp\left(\frac{\pm s}{\sigma_\tau^2(1-\rho^2)}\right)\begin{cases} s<0 \\ s\geq 0.\end{cases}$$

where we have again used Gradshteyn & Ryzhik[4] [Equation (3.723)] to evaluate the integral, dealing with the cases $s<0$ and $s\geq 0$ separately.

We then find for the convolution we are actually interested in [Equation (26)]

$$(f_R * f_R)(s) = \frac{1}{\pi^2\sigma_\tau^4(1-\rho^2)}\exp\left(\frac{\rho s}{\sigma_\tau^2(1-\rho^2)}\right)(g * g)(s) =$$
$$\frac{1}{2\sigma_\tau^2}\exp\left(\frac{(\rho\pm 1)s}{\sigma_\tau^2(1-\rho^2)}\right)\begin{cases} s<0 \\ s\geq 0.\end{cases} \tag{27}$$

If we insert the definition of σ_τ as expressed in Equation (4), we then find

$$(f_R * f_R)(s) = \frac{1}{4\sigma^2}\exp\left(\frac{(2\rho\pm 2)s}{4\sigma^2(1-\rho^2)}\right)\begin{cases} s<0 \\ s\geq 0.\end{cases}$$

This is the distribution for the numerator, Equation (25).

For successive displacements, we have $\rho = -\frac{1}{2}$ [Equation (12)] and find

$$\frac{1}{4\sigma^2} \exp\left(\frac{(-1 \pm 2)s}{3\sigma^2}\right) \begin{cases} s < 0 \\ s \geq 0 \end{cases} = \frac{1}{4\sigma^2} \begin{cases} \mathrm{e}^{\frac{1}{3}s/\sigma^2} & s < 0 \\ \mathrm{e}^{-s/\sigma^2} & s \geq 0 \end{cases} \tag{28}$$

while for displacements separated in time from each other, we have $\rho = 0$ [Equation (14)] and find

$$\frac{1}{4\sigma^2}\mathrm{e}^{\pm\frac{1}{2}s/\sigma^2} \begin{cases} s < 0 \\ s \geq 0. \end{cases} \tag{29}$$

The mean of the scalar product

As already noted, the mean of the scalar product is very much related to the velocity autocorrelation function, which in two dimensions is defined as

$$C_{v;\Delta\tau} = \frac{1}{\Delta\tau^2}\langle \Delta\tilde{\mathbf{R}}_{\Delta\tau,0} \cdot \Delta\tilde{\mathbf{R}}_{\tau+\Delta\tau,\tau}\rangle. \tag{30}$$

It is consequently of interest to calculate this mean. However, it follows immediately from Equation (25) and the results in one dimension [Equation (15)] that

$$\langle \Delta\tilde{\mathbf{R}}_{\Delta\tau,0} \cdot \Delta\tilde{\mathbf{R}}_{\tau+\Delta\tau,\tau}\rangle(\tau) = \begin{cases} 4\sigma^2 & \tau = 0 \\ -2\sigma^2 & \tau = \Delta\tau \\ 0 & \tau > \Delta\tau. \end{cases} \tag{31}$$

Distribution of the angle between displacements

In two dimensions we cannot use the same simplifying argument as used in one dimension to calculate the distribution of the angle between displacements from the scalar product Equation (25). Fortunately, it is nevertheless possible to calculate the distribution and we do so by considering $\Delta\tilde{\mathbf{R}}_{\Delta\tau,0}$ and $\Delta\tilde{\mathbf{R}}_{\tau+\Delta\tau,\tau}$ to be complex variables. To this end, we define

$$\Delta\tilde{Z}_{\Delta\tau,0} \equiv \Delta\tilde{X}_{\Delta\tau,0} + i\Delta\tilde{Y}_{\Delta\tau,0} \tag{32}$$

$$\Delta\tilde{Z}_{\tau+\Delta\tau,\tau} \equiv \Delta\tilde{X}_{\tau+\Delta\tau,\tau} + i\Delta\tilde{Y}_{\tau+\Delta\tau,\tau}. \tag{33}$$

The utility of this representation lies in that

$$\Delta\tilde{Z}_{\Delta\tau,0}\Delta\tilde{Z}^*_{\tau+\Delta\tau,\tau} = |\Delta\tilde{Z}_{\Delta\tau,0}||\Delta\tilde{Z}_{\tau+\Delta\tau,\tau}|\big(\cos(\tilde{\Theta}_{\Delta\tau,0} - \tilde{\Theta}_{\tau+\Delta\tau,\tau}) + i\sin(\tilde{\Theta}_{\Delta\tau,0} - \tilde{\Theta}_{\tau+\Delta\tau,\tau})\big)$$

where $\tilde{\Theta}_{\Delta\tau,0}$ and $\tilde{\Theta}_{\tau+\Delta\tau,\tau}$ are the angles corresponding to a polar representation of the complex variables $\Delta\tilde{Z}_{\Delta\tau,0}$ and $\Delta\tilde{Z}_{\tau+\Delta\tau,\tau}$, respectively. The angle we are interested in, $\tilde{\Theta}$, is actually $\tilde{\Theta}_{\Delta\tau,0} - \tilde{\Theta}_{\tau+\Delta\tau,\tau}$ in the polar representation, so we see that if we have the polar representation of $\Delta\tilde{Z}_{\Delta\tau,0}\Delta\tilde{Z}^*_{\tau+\Delta\tau,\tau}$ then, upto the sign and considering that we have thus far only considered angles between 0 and π, we have achieved our goal. Now, by definition $\Delta\tilde{Z}_{\Delta\tau,0}$ and $\Delta\tilde{Z}_{\tau+\Delta\tau,\tau}$ are normally distributed complex stochastic variables and it turns out that there is literature specifically addressing the joint probability distribution of the magnitude and angle of the product of two correlated normally distributed complex stochastic variables.[5] Specifically, for two zero mean normally distributed complex stochastic variables, the joint probability distribution of $R = |\Delta\tilde{Z}_{\Delta\tau,0}||\Delta\tilde{Z}_{\tau+\Delta\tau,\tau}|$ and $\tilde{\Theta} = \tilde{\Theta}_{\Delta\tau,0} - \tilde{\Theta}_{\tau+\Delta\tau,\tau}$ is

$$f_{R,\tilde{\Theta}}(r,\theta) = \frac{2r}{\pi\sigma^2_{z_{\Delta\tau,0}}\sigma^2_{z_{\tau+\Delta\tau,\tau}}\left(1-|\rho|^2\right)} \times \exp\left(\frac{2r\Re\left(\rho \mathrm{e}^{-i\theta}\right)}{\sigma_{z_{\Delta\tau,0}}\sigma_{z_{\tau+\Delta\tau,\tau}}\left(1-|\rho|^2\right)}\right) K_0\left(\frac{2r}{\sigma_{z_{\Delta\tau,0}}\sigma_{z_{\tau+\Delta\tau,\tau}}\left(1-|\rho|^2\right)}\right) \tag{34}$$

where ρ now is the (complex) correlation coefficient between $\Delta\tilde{Z}_{\Delta\tau,0}$ and $\Delta\tilde{Z}_{\tau+\Delta\tau,\tau}$, and, likewise, $\sigma^2_{z_{\Delta\tau,0}}$ and $\sigma^2_{z_{\tau+\Delta\tau,\tau}}$ the (complex) variance of $\Delta\tilde{Z}_{\Delta\tau,0}$ and $\Delta\tilde{Z}_{\tau+\Delta\tau,\Delta\tau}$, respectively.

The variance of $\Delta\tilde{Z}_{\Delta\tau,0}$ is given by[6]

$$\sigma^2_{z_{\Delta\tau,0}} = \mathrm{Var}\left[\Delta\tilde{Z}_{\Delta\tau,0}\right] = \mathrm{Var}\left[\Delta\tilde{X}_{\Delta\tau,0}\right] + \mathrm{Var}\left[\Delta\tilde{Y}_{\Delta\tau,0}\right] = \sigma^2_\tau + \sigma^2_\tau = 2\sigma^2_\tau$$

where we have inserted the known variances $\Delta\tilde{X}_{\Delta\tau,0}$ and $\Delta\tilde{Y}_{\Delta\tau,0}$. In the same way we find

$$\sigma^2_{z_{\tau+\Delta\tau,\tau}} = \mathrm{Var}\left[\Delta\tilde{Z}_{\tau+\Delta\tau,\tau}\right] = 2\sigma^2_\tau.$$

To calculate the correlation coefficient, we start with the covariance which is given by[6]

$$\begin{aligned}
&\mathrm{Covar}\left[\Delta\tilde{Z}_{\Delta\tau,0}, \Delta\tilde{Z}_{\tau+\Delta\tau,\tau}\right] = \\
&\quad\Big(\mathrm{Covar}\left[\Delta\tilde{X}_{\Delta\tau,0}, \Delta\tilde{X}_{\tau+\Delta\tau,\tau}\right] + \mathrm{Covar}\left[\Delta\tilde{Y}_{\Delta\tau,0}, \Delta\tilde{Y}_{\tau+\Delta\tau,\tau}\right]\Big) + \\
&\quad i\Big(\mathrm{Covar}\left[\Delta\tilde{Y}_{\Delta\tau,0}, \Delta\tilde{X}_{\tau+\Delta\tau,\tau}\right] - \mathrm{Covar}\left[\Delta\tilde{X}_{\Delta\tau,0}, \Delta\tilde{Y}_{\tau+\Delta\tau,\tau}\right]\Big) = \\
&= \Big(\mathrm{Covar}\left[\Delta\tilde{X}_{\Delta\tau,0}, \Delta\tilde{X}_{\tau+\Delta\tau,\tau}\right] + \mathrm{Covar}\left[\Delta\tilde{Y}_{\Delta\tau,0}, \Delta\tilde{Y}_{\tau+\Delta\tau,\tau}\right]\Big) + i\big(0-0\big) = \\
&\quad = \mathrm{Covar}\left[\Delta\tilde{X}_{\Delta\tau,0}, \Delta\tilde{X}_{\tau+\Delta\tau,\tau}\right] + \mathrm{Covar}\left[\Delta\tilde{Y}_{\Delta\tau,0}, \Delta\tilde{Y}_{\tau+\Delta\tau,\tau}\right]
\end{aligned}$$

where we have used that the x and y directions are independent.

This expression can be evaluated in the same way as as for the one-dimensional case, Equation (10); for successive displacements, we then have

$$\text{Covar}\left[\Delta\tilde{Z}_{\Delta\tau,0}, \Delta\tilde{Z}_{\tau+\Delta\tau,\tau}\right] = -\sigma^2 - \sigma^2 = -2\sigma^2,$$

while for displacements separated in time from each other everything vanishes

$$\text{Covar}\left[\Delta\tilde{Z}_{\Delta\tau,0}, \Delta\tilde{Z}_{\tau+\tau,\tau}\right] = 0.$$

The correlation coefficient now follows from the covariance[6]

$$\rho \equiv \frac{\text{Covar}\left[\Delta\tilde{Z}_{\tau,0}, \Delta\tilde{Z}_{\tau+\tau,\tau}\right]}{\sqrt{\text{Var}\left[\Delta\tilde{Z}_{\tau,0}\right]\text{Var}\left[\Delta\tilde{Z}_{\tau+\tau,\tau}\right]}} = \frac{\text{Covar}\left[\Delta\tilde{Z}_{\tau,0}, \Delta\tilde{Z}_{\tau+\tau,\tau}\right]}{\sigma_{z_{\tau,0}}\sigma_{z_{\tau+\tau,\tau}}} = \frac{\text{Covar}\left[\Delta\tilde{Z}_{\tau,0}, \Delta\tilde{Z}_{\tau+\tau,\tau}\right]}{2\sigma_\tau^2}$$

using the variances we calculated above. For successive displacements, we then find

$$\rho = \frac{-2\sigma^2}{2\sigma_\tau^2} = -\frac{\sigma^2}{2\sigma^2} = -\tfrac{1}{2} \tag{35}$$

and for displacements separated in time from each other

$$\rho = 0 \tag{36}$$

just like in real space [Equations (12) and (14), respectively].

Inserting the variances into Equation (34) and using the fact that for our case the correlation coefficient is real so that

$$\Re\left(\rho e^{-i\theta}\right) = \rho\cos\theta$$

and

$$|\rho|^2 = \rho^2$$

we find

$$f_{R,\tilde{\Theta}}(r,\theta) = \frac{r}{2\pi\sigma_\tau^4\left(1-\rho^2\right)}\exp\left(\frac{\rho r\cos\theta}{\sigma_\tau^2\left(1-\rho^2\right)}\right)K_0\left(\frac{r}{\sigma_\tau^2\left(1-\rho^2\right)}\right).$$

The distribution of the angle, $f_{\tilde{\Theta}}(\theta)$ then follows by integrating over r

$$f_{\tilde{\Theta}}(\theta) = \int_0^\infty 2f_{R,\tilde{\Theta}}(r,\theta)\mathrm{d}r$$

where the extra factor of 2 comes from the fact that we have only considered angles between 0 and π. Inserting, we find

$$f_{\tilde{\Theta}}(\theta) = \int_0^\infty 2 f_{R,\tilde{\Theta}}(r,\theta)\mathrm{d}r = \frac{2}{2\pi\sigma_\tau^4\left(1-\rho^2\right)}\int_0^\infty r\exp\left(\frac{\rho r\cos\theta}{\sigma_\tau^2\left(1-\rho^2\right)}\right)K_0\left(\frac{r}{\sigma_\tau^2\left(1-\rho^2\right)}\right)\mathrm{d}r.$$

In Gradshteyn & Ryzhik[4] we find [Equation (6.624)]

$$\int_0^\infty x\mathrm{e}^{-\alpha x}K_0(\beta x)\mathrm{d}x = \frac{1}{\alpha^2-\beta^2}\left(\frac{\alpha}{\sqrt{\alpha^2-\beta^2}}\ln\left(\alpha/\beta+\sqrt{(\alpha/\beta)^2-1}\right)-1\right).$$

However, we have

$$\ln\left(z+\sqrt{z^2-1}\right) = i\arccos z$$

and, furthermore, we may write

$$\sqrt{\alpha^2-\beta^2} = i\sqrt{\beta^2-\alpha^2},$$

so we can rewrite this expression to

$$\int_0^\infty x\mathrm{e}^{-\alpha x}K_0(\beta x)\mathrm{d}x = -\frac{1}{\beta^2-\alpha^2}\left(\frac{\alpha}{i\sqrt{\beta^2-\alpha^2}}i\arccos(\alpha/\beta)-1\right) = \frac{1}{\beta^2-\alpha^2}\left(1-\frac{\alpha}{\sqrt{\beta^2-\alpha^2}}\arccos\left(\alpha/\beta\right)\right).$$

Using this, we find for the distribution we are interested in

$$f_{\tilde{\Theta}}(\theta) = \frac{1}{\pi\sigma_\tau^4\left(1-\rho^2\right)}\int_0^\infty r\exp\left(\frac{\rho r\cos\theta}{\sigma_\tau^2\left(1-\rho^2\right)}\right)K_0\left(\frac{r}{\sigma_\tau^2\left(1-\rho^2\right)}\right)\mathrm{d}r = \frac{1}{\pi\sigma_\tau^4\left(1-\rho^2\right)}\frac{\sigma_\tau^4\left(1-\rho^2\right)^2}{1-\rho^2\cos^2\theta}\times \left(1+\frac{\rho\cos\theta}{\sqrt{1-\rho^2\cos^2\theta}}\arccos\left(-\rho\cos\theta\right)\right) = \frac{1}{\pi}\frac{1-\rho^2}{1-\rho^2\cos^2\theta}\left(1+\frac{\rho\cos\theta\arccos\left(-\rho\cos\theta\right)}{\sqrt{1-\rho^2\cos^2\theta}}\right) \quad (37)$$

where we have used that

$$\sigma_\tau^2\left(1-\rho^2\right) > 0.$$

This is the distribution of the angle in two dimensions.

For successive displacements, we have $\rho = -\frac{1}{2}$ [Equation (35)] and hence

$$f_{\tilde{\Theta}}(\theta) = \frac{1}{\pi}\frac{1-\frac{1}{4}}{1-\frac{1}{4}\cos^2\theta}\left(1-\frac{1}{2}\frac{\cos\theta\arccos\left(\frac{1}{2}\cos\theta\right)}{\sqrt{1-\frac{1}{4}\cos^2\theta}}\right) =$$

$$\frac{1}{\pi}\frac{3}{4-\cos^2\theta}\left(1-\frac{\cos\theta\arccos\left(\frac{1}{2}\cos\theta\right)}{\sqrt{4-\cos^2\theta}}\right) =$$

$$\frac{1}{\pi}\frac{3}{3+\sin^2\theta}\left(1-\frac{\cos\theta\arccos\left(\frac{1}{2}\cos\theta\right)}{\sqrt{3+\sin^2\theta}}\right),$$

while for displacements separated in time from each other, we have $\rho = 0$ [Equation (36)] and hence

$$f_{\tilde{\Theta}}(\theta) = \frac{1}{\pi}.$$

Though it is not apparent, the former equation is equivalent to the expression previously derived by Hurford,[7] as we will now show. We have that

$$f_{\tilde{\Theta}}(\theta) = \frac{1}{\pi}\frac{3}{4-\cos^2\theta}\left(1-\frac{\cos\theta\arccos\left(\frac{1}{2}\cos\theta\right)}{\sqrt{4-\cos^2\theta}}\right) =$$

$$\frac{1}{\pi}\frac{3}{4-\cos^2\theta}\left(\frac{4-\cos^2\theta}{4-\cos^2\theta}-\frac{\sqrt{4-\cos^2\theta}\cos\theta\arccos\left(\frac{1}{2}\cos\theta\right)}{4-\cos^2\theta}\right) =$$

$$\frac{6}{2\pi}\frac{4-\cos^2\theta-\sqrt{4-\cos^2\theta}\cos\theta\arccos\left(\frac{1}{2}\cos\theta\right)}{(4-\cos^2\theta)^2} =$$

$$\frac{6}{2\pi}\frac{4-\cos\theta\left(\cos\theta+\sqrt{4-\cos^2\theta}\arccos\left(\frac{1}{2}\cos\theta\right)\right)}{(4-\cos^2\theta)^2} =$$

$$\frac{24-3\cos\theta\left(2\cos\theta+\sqrt{4-\cos^2\theta}\cdot 2\arccos\left(\frac{1}{2}\cos\theta\right)\right)}{2\pi(4-\cos^2\theta)^2}.$$

From $\tan(x) = \sin(x)/\cos(x)$ and the trigonometric identity it follows that for $x \in (0, 1]$ we have

$$\arccos(x) = \arctan\frac{\sqrt{1-x^2}}{x}$$

and hence that

$$2\arccos\left(\tfrac{1}{2}\cos\theta\right) = 2\arctan\frac{\sqrt{1-\frac{1}{4}\cos^2\theta}}{\frac{1}{2}\cos\theta} = 2\arctan\frac{\sqrt{4-\cos^2\theta}}{\cos\theta}$$

Furthermore, for $x > 0$ we have

$$\arctan\frac{1}{x} = \frac{\pi}{2} - \arctan x = \frac{\pi}{2} + \arctan(-x)$$

where we have used that arctan is odd. Hence we find

$$2\arccos\left(\tfrac{1}{2}\cos\theta\right) = 2\left(\frac{\pi}{2} + \arctan\frac{-\cos\theta}{\sqrt{4-\cos^2\theta}}\right) = \pi + 2\arctan\frac{-\cos\theta}{\sqrt{4-\cos^2\theta}}.$$

We then finally have

$$f_{\tilde{\Theta}}(\theta) = \frac{24 - 3\cos\theta\left(2\cos\theta + \sqrt{4-\cos^2\theta}\left(\pi + 2\arctan\frac{-\cos\theta}{\sqrt{4-\cos^2\theta}}\right)\right)}{2\pi(4-\cos^2\theta)^2} \qquad (38)$$

which is the same result as derived by Hurford,[7] if we take into account that we have only considered angles between 0 and π and hence Hurford's expression, Equation (6), should be multiplied by a factor of 2.

Mean cosine of the angle between displacements

We can use the distribution of the angle to calculate $\langle\cos\tilde{\Theta}\rangle$, which of course is given by

$$\langle\cos\tilde{\Theta}\rangle = \int_0^\pi f_{\tilde{\Theta}}(\theta)\cos\theta\,\mathrm{d}\theta.$$

Making a change of variables to $u = -\cos\theta$ changes the integral to

$$\begin{aligned}\langle\cos\tilde{\Theta}\rangle &= \int_{-1}^{1}\frac{1}{\pi}\frac{1-\rho^2}{1-\rho^2u^2}\left(1 + \frac{-\rho u\arccos(\rho u)}{\sqrt{1-\rho^2u^2}}\right)(-u)\frac{\mathrm{d}u}{\sqrt{1-u^2}} = \\ &= -\frac{1}{\pi}(1-\rho^2)\int_{-1}^{1}\left(1 + \frac{-\rho u\arccos(\rho u)}{\sqrt{1-\rho^2u^2}}\right)\frac{u\mathrm{d}u}{(1-\rho^2u^2)\sqrt{1-u^2}}.\end{aligned}$$

The first term is odd, so when integrated over the symmetric interval it vanishes. We are thus left with the second term

$$\langle\cos\tilde{\Theta}\rangle = \frac{1}{\pi}(1-\rho^2)\int_{-1}^{1}\frac{\rho u^2\arccos(\rho u)}{(1-\rho^2u^2)^{3/2}}\frac{\mathrm{d}u}{\sqrt{1-u^2}} \equiv \frac{1}{\pi}(1-\rho^2)\int_{-1}^{1}g(u)\mathrm{d}u$$

where we have defined the integrand $g(u)$ in the last step. In general we have

$$g(u) = \tfrac{1}{2}\big(g(u) + g(-u)\big) + \tfrac{1}{2}\big(g(u) - g(-u)\big)$$

where the first term is even and the second odd. The odd term will disappear when integrated over the symmetric interval, so we may ignore it. For the even term we have, however

$$\tfrac{1}{2}\big(g(u) + g(-u)\big) = \frac{\rho}{2}\frac{u^2}{(1-\rho^2u^2)^{3/2}\sqrt{1-u^2}}\big(\arccos(\rho u) + \arccos(-\rho u)\big).$$

But we have the identity

$$\arccos(-\rho u) = \pi - \arccos(\rho u)$$

and thus

$$\tfrac{1}{2}\big(g(u)+g(-u)\big) = \frac{\rho}{2}\frac{u^2}{(1-\rho^2u^2)^{3/2}\sqrt{1-u^2}}\big(\pi+\arccos(\rho u)-\arccos(\rho u)\big) = \frac{\pi\rho}{2}\frac{u^2}{(1-\rho^2u^2)^{3/2}\sqrt{1-u^2}}.$$

We then find

$$\langle\cos\tilde{\Theta}\rangle = \frac{1}{\pi}(1-\rho^2)2\int_0^1 \tfrac{1}{2}\big(g(u)+g(-u)\big)\mathrm{d}u = \frac{1}{\pi}(1-\rho^2)2\int_0^1 \frac{\pi\rho}{2}\frac{u^2}{(1-\rho^2u^2)^{3/2}\sqrt{1-u^2}}\mathrm{d}u = (1-\rho^2)\rho\int_0^1 \frac{u^2}{(1-\rho^2u^2)^{3/2}\sqrt{1-u^2}}\mathrm{d}u.$$

To solve this integral we note that the complete elliptic integral of the first kind is given by[8]

$$\mathbf{K}(k) = \int_0^1 \frac{\mathrm{d}x}{\sqrt{1-x^2}\sqrt{1-k^2x^2}} = \int_0^1 \frac{\mathrm{d}x}{\sqrt{1-k^2x^2-x^2+k^2x^4}},$$

which implies that

$$\frac{\mathrm{d}\mathbf{K}}{\mathrm{d}k} = \int_0^1 -\tfrac{1}{2}\frac{-2kx^2+2kx^4}{\big((1-x^2)(1-k^2x^2)\big)^{3/2}}\mathrm{d}x = k\int_0^1 \frac{x^2-x^4}{\big((1-x^2)(1-k^2x^2)\big)^{3/2}}\mathrm{d}x = k\int_0^1 \frac{x^2(1-x^2)}{\big((1-x^2)(1-k^2x^2)\big)^{3/2}}\mathrm{d}x = k\int_0^1 \frac{x^2\mathrm{d}x}{\sqrt{1-x^2}(1-k^2x^2)^{3/2}}.$$

Thus, we have

$$\int_0^1 \frac{x^2\mathrm{d}x}{\sqrt{1-x^2}(1-k^2x^2)^{3/2}} = \frac{1}{k}\frac{\mathrm{d}\mathbf{K}}{\mathrm{d}k}.$$

However, according to Gradshteyn & Ryzhik[4] [Equation (8.123)] we also have

$$\frac{\mathrm{d}\mathbf{K}}{\mathrm{d}k} = \frac{\mathbf{E}(k)}{kk'^2} - \frac{\mathbf{K}(k)}{k} = \frac{\mathbf{E}(k)-k'^2\mathbf{K}(k)}{kk'^2}$$

where $k'^2 = 1 - k^2$ and $\mathbf{E}(k)$ is the complete elliptic integral of the second kind,[8] so

$$\int_0^1 \frac{x^2 \mathrm{d}x}{\sqrt{1-x^2}(1-k^2x^2)^{3/2}} = \frac{1}{k}\frac{\mathrm{d}\mathbf{K}}{\mathrm{d}k} = \frac{\mathbf{E}(k) - (1-k^2)\mathbf{K}(k)}{k^2(1-k^2)}.$$

We thus find for the mean cosine of the angle

$$\langle \cos\tilde{\Theta}\rangle = (1-\rho^2)\rho \int_0^1 \frac{u^2}{(1-\rho^2u^2)^{3/2}\sqrt{1-u^2}}\mathrm{d}u = (1-\rho^2)\rho\frac{\mathbf{E}(\rho) - (1-\rho^2)\mathbf{K}(\rho)}{\rho^2(1-\rho^2)} = \frac{\mathbf{E}(\rho) - (1-\rho^2)\mathbf{K}(\rho)}{\rho}. \tag{39}$$

For successive displacements, when $\rho = -\frac{1}{2}$ [Equation (35)], this expression is close to -0.406, while for displacements separated in time from each other, when $\rho = 0$ [Equation (36)], this expression reduces to 0 using known properties of elliptic integrals.[8]

Mean angle between displacements

Finally, we calculate the mean angle, which naturally is given by

$$\langle\tilde{\Theta}\rangle = \int_0^\pi f_{\tilde{\Theta}}(\theta)\theta\mathrm{d}\theta.$$

Again making the change of variables $u = -\cos\theta$, we find

$$\begin{aligned}\langle\tilde{\Theta}\rangle &= \int_{-1}^1 \frac{1}{\pi}\frac{1-\rho^2}{1-\rho^2u^2}\left(1 + \frac{-\rho u \arccos(\rho u)}{\sqrt{1-\rho^2u^2}}\right)\arccos(-u)\frac{\mathrm{d}u}{\sqrt{1-u^2}} = \\ &= \frac{1}{\pi}(1-\rho^2)\int_{-1}^1\left(1 - \frac{\rho u\arccos(\rho u)}{\sqrt{1-\rho^2u^2}}\right)\frac{\arccos(-u)\mathrm{d}u}{(1-\rho^2u^2)\sqrt{1-u^2}} \\ &\equiv \frac{1}{\pi}(1-\rho^2)\int_{-1}^1 g(u)\mathrm{d}u\end{aligned}$$

where we have introduced the function $g(u)$ for ease of notation.

In general we have

$$g(u) = \tfrac{1}{2}\big(g(u)+g(-u)\big) + \tfrac{1}{2}\big(g(u)-g(-u)\big)$$

where the first term is even and the second odd. The odd term will disappear when integrated over the symmetric interval, so we may ignore it. The even term, instead, will contribute the same amount for either part of the symmetric interval so we may write

$$\langle\tilde{\Theta}\rangle = \frac{1}{\pi}(1-\rho^2)\int_{-1}^1 g(u)\mathrm{d}u = \frac{1}{\pi}(1-\rho^2)2\int_0^1 \tfrac{1}{2}\big(g(u)+g(-u)\big)\mathrm{d}u = \frac{1}{\pi}(1-\rho^2)\int_0^1\big(g(u)+g(-u)\big)\mathrm{d}u.$$

What is the integrand in this expression? We have

$$g(u)+g(-u)=\frac{1}{(1-\rho^2u^2)\sqrt{1-u^2}}\Bigg(\arccos(-u)-$$
$$\frac{\rho u\arccos(\rho u)\arccos(-u)}{\sqrt{1-\rho^2u^2}}+\arccos(u)+\frac{\rho u\arccos(-\rho u)\arccos(u)}{\sqrt{1-\rho^2u^2}}\Bigg)$$

But

$$\arccos(-u)+\arccos(u)=\pi-\arccos(u)+\arccos(u)=\pi$$

and

$$-\rho u\arccos(\rho u)\arccos(-u)+\rho u\arccos(-\rho u)\arccos(u)=$$
$$\rho u\big(-\arccos(\rho u)\arccos(-u)+\arccos(-\rho u)\arccos(u)\big)=$$
$$\rho u\Big(-\arccos(\rho u)\big(\pi-\arccos(u)\big)+\big(\pi-\arccos(\rho u)\big)\arccos(u)\Big)=$$
$$\rho u\pi\big(\arccos(u)-\arccos(\rho u)\big)$$

so

$$g(u)+g(-u)=\frac{\pi}{(1-\rho^2u^2)\sqrt{1-u^2}}\left(1+\frac{\rho u\big(\arccos(u)-\arccos(\rho u)\big)}{\sqrt{1-\rho^2u^2}}\right)$$

and

$$\langle\tilde{\Theta}\rangle=(1-\rho^2)\times$$
$$\int_0^1\frac{1}{(1-\rho^2u^2)\sqrt{1-u^2}}\left(1+\frac{\rho u\big(\arccos(u)-\arccos(\rho u)\big)}{\sqrt{1-\rho^2u^2}}\right)\mathrm{d}u. \quad (40)$$

We start by looking at the second term

$$(1-\rho^2)\rho\int_0^1\frac{u}{(1-\rho^2u^2)^{3/2}\sqrt{1-u^2}}\big(\arccos(u)-\arccos(\rho u)\big)\mathrm{d}u.$$

We observe that

$$\frac{\mathrm{d}}{\mathrm{d}u}\left(\frac{\sqrt{1-u^2}}{\sqrt{1-\rho^2u^2}}\right)=-u\frac{1-\rho^2}{(1-\rho^2u^2)^{3/2}\sqrt{1-u^2}}$$

so we can write the second term

$$-\rho\int_0^1\frac{\mathrm{d}}{\mathrm{d}u}\left(\frac{\sqrt{1-u^2}}{\sqrt{1-\rho^2u^2}}\right)\big(\arccos(u)-\arccos(\rho u)\big)\mathrm{d}u=$$
$$-\rho\frac{\sqrt{1-u^2}}{\sqrt{1-\rho^2u^2}}\big(\arccos(u)-\arccos(\rho u)\big)\Bigg|_0^1+$$
$$\rho\int_0^1\frac{\sqrt{1-u^2}}{\sqrt{1-\rho^2u^2}}\frac{\mathrm{d}}{\mathrm{d}u}\big(\arccos(u)-\arccos(\rho u)\big)\mathrm{d}u$$

where we have performed a partial integration. The first term vanishes, while for the second term we have

$$\frac{\mathrm{d}}{\mathrm{d}u}\big(\arccos(u)-\arccos(\rho u)\big)=-\frac{1}{\sqrt{1-u^2}}+\frac{\rho}{\sqrt{1-\rho^2u^2}}$$

so we get

$$(1-\rho^2)\rho\int_0^1\frac{u}{(1-\rho^2u^2)^{3/2}\sqrt{1-u^2}}\big(\arccos(u)-\arccos(\rho u)\big)\mathrm{d}u=$$
$$\rho\int_0^1\frac{\sqrt{1-u^2}}{\sqrt{1-\rho^2u^2}}\left(\frac{\rho}{\sqrt{1-\rho^2u^2}}-\frac{1}{\sqrt{1-u^2}}\right)\mathrm{d}u=$$
$$\rho^2\int_0^1\frac{\sqrt{1-u^2}}{1-\rho^2u^2}\mathrm{d}u-\int_0^1\frac{\rho}{\sqrt{1-\rho^2u^2}}\mathrm{d}u$$

where

$$\int_0^1\frac{\rho}{\sqrt{1-\rho^2u^2}}\mathrm{d}u=\arccos(-\rho)-\frac{\pi}{2}.$$

This was just the second term in our expression, Equation (40), for the mean angle. Inserting this into the full expression, we then find

$$\langle\tilde{\Theta}\rangle=$$
$$(1-\rho^2)\int_0^1\frac{1}{(1-\rho^2u^2)\sqrt{1-u^2}}\left(1+\frac{\rho u\big(\arccos(u)-\arccos(\rho u)\big)}{\sqrt{1-\rho^2u^2}}\right)\mathrm{d}u=$$
$$(1-\rho^2)\int_0^1\frac{1}{(1-\rho^2u^2)\sqrt{1-u^2}}\mathrm{d}u+\rho^2\int_0^1\frac{\sqrt{1-u^2}}{1-\rho^2u^2}\mathrm{d}u-\left(\arccos(-\rho)-\frac{\pi}{2}\right)=$$
$$\int_0^1\frac{1}{(1-\rho^2u^2)}\left(\frac{1-\rho^2}{\sqrt{1-u^2}}+\rho^2\sqrt{1-u^2}\right)\mathrm{d}u+\frac{\pi}{2}-\arccos(-\rho)=$$
$$\int_0^1\frac{1}{(1-\rho^2u^2)}\frac{1-\rho^2+\rho^2(1-u^2)}{\sqrt{1-u^2}}\mathrm{d}u+\frac{\pi}{2}-\arccos(-\rho)=$$
$$\int_0^1\frac{1}{\sqrt{1-u^2}}\mathrm{d}u+\frac{\pi}{2}-\arccos(-\rho)=$$
$$\frac{\pi}{2}+\frac{\pi}{2}-\arccos(-\rho)=\pi-\arccos(-\rho). \quad (41)$$

For successive displacements, we have $\rho=-\frac{1}{2}$ [Equation (35)] and consequently

$$\langle\tilde{\Theta}\rangle=\pi-\arccos(-\rho)=\frac{2\pi}{3} \quad (42)$$

while for displacements separated in time from each other, we have $\rho=0$ [Equation (36)] and thus

$$\langle\tilde{\Theta}\rangle=\pi-\arccos(-\rho)=\frac{\pi}{2}. \quad (43)$$

Incidentally, this is exactly the same as the results for one dimension (in contrast to the mean cosine of the angle between displacements).

Particles moving by Brownian motion

One dimension

For particles moving by Brownian motion we now write the observed position of a particle as

$$\tilde{X}(t) = X_{\mathrm{BM}}(t) + \Xi(t)$$

where $X_{\mathrm{BM}}(t)$ is the real position of the particle undergoing Brownian motion. We now have that

$$\Delta\tilde{X}_{\Delta\tau,0} \equiv \tilde{X}(\Delta\tau) - \tilde{X}(0) \equiv X_{\mathrm{BM}}(\Delta\tau) + \Xi(\Delta\tau) - X_{\mathrm{BM}}(0) - \Xi(0) = \\ X_{\mathrm{BM}}(\Delta\tau) - X_{\mathrm{BM}}(0) + \Xi(\Delta\tau) - \Xi(0).$$

As for the case of stationary particles, we have that

$$\Xi(\Delta\tau) - \Xi(0) \sim \mathcal{N}(0, 2\sigma^2).$$

Furthermore, we have

$$X_{\mathrm{BM}}(\Delta\tau) - X_{\mathrm{BM}}(0) \sim \mathcal{N}(0, 2D\Delta\tau)$$

where D is the diffusion coefficient. This expression reflects the fact that particles moving by Brownian motion will distribute according to a normal distribution where the variance (the mean square displacement) is proportional to $2D\Delta\tau$ (in one dimension). Both terms above are normally distributed random variables, and their sum is also a normally distributed random variable. More precisely, we have

$$\Delta\tilde{X}_{\Delta\tau,0} \sim \mathcal{N}(0, 2D\Delta\tau + 2\sigma^2) \equiv \mathcal{N}(0, \sigma_\tau^2) \tag{44}$$

Similarly, we have

$$\Delta\tilde{X}_{\tau+\Delta\tau,\tau} \sim \mathcal{N}(0, 2D\Delta\tau + 2\sigma^2) \equiv \mathcal{N}(0, \sigma_\tau^2). \tag{45}$$

We observe that if we let $D = 0$ in these expressions, we regain our description of stationary particles, Equation (4), as expected.

Distribution of the scalar product

Like for stationary particles, we start by considering the product $\Delta\tilde{X}_{\Delta\tau,0}\Delta\tilde{X}_{\tau+\Delta\tau,\tau}$. The derivation can proceed in the same way as for the case of stationary particles and hence we have according to Equation (8) that the distribution of $R = \Delta\tilde{X}_{\Delta\tau,0}\Delta\tilde{X}_{\tau+\Delta\tau,\tau}$ is given by

$$f_R(r) = \frac{1}{\pi\sigma_\tau^2\sqrt{1-\rho^2}} \exp\left(\frac{\rho r}{\sigma_\tau^2(1-\rho^2)}\right) K_0\left(\frac{|r|}{\sigma_\tau^2(1-\rho^2)}\right). \tag{46}$$

The correlation coefficient, ρ, however, is different for particles moving by Brownian motion. Indeed, according to Equation (9) we have

$$\rho = \frac{\langle \Delta\tilde{X}_{\Delta\tau,0}\Delta\tilde{X}_{\tau+\Delta\tau,\tau}\rangle}{\sigma_\tau^2}$$

where

$$\langle \Delta\tilde{X}_{\Delta\tau,0}\Delta\tilde{X}_{\tau+\Delta\tau,\tau}\rangle = \Big\langle \big(X_{\mathrm{BM}}(\Delta\tau) - X_{\mathrm{BM}}(0) + \Xi(\Delta\tau) - \Xi(0)\big)\times \\ \big(X_{\mathrm{BM}}(\tau+\Delta\tau) - X_{\mathrm{BM}}(\tau) + \Xi(\tau+\Delta\tau) - \Xi(\tau)\big)\Big\rangle\,.$$

The position of a particle and the localisation error are independent from each other, so all terms containing a combination of $X_{\mathrm{BM}}(t)$ and $\Xi(t)$ vanish. Furthermore, we have that

$$\Big\langle \big(X_{\mathrm{BM}}(\Delta\tau) - X_{\mathrm{BM}}(0)\big)\big(X_{\mathrm{BM}}(\tau+\Delta\tau) - X_{\mathrm{BM}}(\tau)\big)\Big\rangle = 0$$

We then end up with the same expression, Equation (10) as for stationary particles

$$\langle \Delta\tilde{X}_{\Delta\tau,0}\Delta\tilde{X}_{\tau+\Delta\tau,\tau}\rangle = \\ \langle \Xi(\Delta\tau)\Xi(\tau+\Delta\tau)\rangle - \langle \Xi(\Delta\tau)\Xi(\tau)\rangle - \langle \Xi(0)\Xi(\tau+\Delta\tau)\rangle + \langle \Xi(0)\Xi(\tau)\rangle.$$

For successive displacements, we then have

$$\langle \Delta\tilde{X}_{\Delta\tau,0}\Delta\tilde{X}_{\tau+\Delta\tau,\tau}\rangle = -\langle \Xi(\Delta\tau)\Xi(\Delta\tau)\rangle = -\sigma^2 \tag{47}$$

using that $\Xi(t)$ at different times are indepedent and that $\Xi(t)$ has variance σ^2. Using the definition of σ_τ as expressed in Equations (44) and (45) we then find

$$\rho = \frac{\langle \Delta\tilde{X}_{\Delta\tau,0}\Delta\tilde{X}_{\tau+\Delta\tau,\tau}\rangle}{\sigma_\tau^2} = -\frac{\sigma^2}{2D\Delta\tau + 2\sigma^2} = -\frac{1}{2 + 2D\Delta\tau/\sigma^2}. \tag{48}$$

If we let $D = 0$ in this expression, it reduces to the expression for stationary particles, Equation (12), as expected. Conversely, for displacements separated in time from each other we have

$$\langle \Delta\tilde{X}_{\Delta\tau,0}\Delta\tilde{X}_{\tau+\Delta\tau,\tau}\rangle = 0, \tag{49}$$

using that $\Xi(t)$ at different times are indepedent from each other, and hence simply

$$\rho = 0. \tag{50}$$

For successive displacements, the distribution of $\Delta\tilde{X}_{\Delta\tau,0}\Delta\tilde{X}_{\tau+\Delta\tau,\tau}$ is then given by Equation (46) inserting ρ from Equation (48). For displacements separated in time from each other, where $\rho = 0$ [Equation (50)], the distribution simplifies to

$$f_R(r) = \frac{1}{\pi\sigma_\tau^2} K_0\left(\frac{|r|}{\sigma_\tau^2}\right).$$

In both cases, the distribution reduces to the one for stationary particles if we let $D = 0$, as expected.

The mean of the scalar product

As mentioned several times, the mean of the scalar product is very much related to the velocity autocorrelation function [Equation (6)], so it is of interest to also calculate this mean. This is something we have already done in terms of, respectively, Equation (47) and (49) for successive displacements and displacements separated in time from each other. Furthermore, for the correlation of a displacement with itself, this is simply the variance of the displacement, so

$$\langle\Delta\tilde{X}_{\Delta\tau,0}\Delta\tilde{X}_{\tau+\Delta\tau,\tau}\rangle(\tau = 0) = \sigma_\tau^2 = 2D\Delta\tau + 2\sigma^2$$

where final equality follows from Equation (44).

Overall, we thus have

$$\langle\Delta\tilde{X}_{\Delta\tau,0}\Delta\tilde{X}_{\tau+\Delta\tau,\tau}\rangle(\tau) = \begin{cases} 2D\Delta\tau + 2\sigma^2 & \tau = 0 \\ -\sigma^2 & \tau = \Delta\tau \\ 0 & \tau > \Delta\tau. \end{cases} \quad (51)$$

Distribution of the angle between displacements

We can derive the distribution of the angle in the same way as for stationary particles, just reinterpreting the value of the correlation coefficient, ρ. More specifically, we have according to Equation (16)

$$P[\cos\tilde{\Theta} = 1] = P[\tilde{\Theta} = 0] = \frac{\arccos(-\rho)}{\pi}$$

and according to Equation (17)

$$P[\cos\tilde{\Theta} = -1] = P[\tilde{\Theta} = \pi] = \frac{\pi - \arccos(-\rho)}{\pi}.$$

If we let $D = 0$, these expressions trivially reduce to the expressions we previously derived for stationary particles, as they only depend upon ρ, which we know reduces as expected. For successive displacements, we should insert ρ from Equation (48); for displacements separated in time from each other, the expressions simplifies to the same values as for stationary particles, *viz.*

$$P[\cos\tilde{\Theta} = 1] = P[\tilde{\Theta} = 0] = \tfrac{1}{2}$$

and

$$P[\cos\tilde{\Theta} = -1] = P[\tilde{\Theta} = \pi] = \tfrac{1}{2}.$$

Mean cosine of the angle betwen displacements

Similarly, the mean cosine of the angle between displacements follows from the results for stationary particles [Equation (18)]

$$\langle\cos\tilde{\Theta}\rangle = \frac{2}{\pi}\arccos(-\rho) - 1.$$

For successive displacements, we should insert ρ from Equation (48) into this expression, which results in

$$\langle\cos\tilde{\Theta}\rangle = \frac{2}{\pi}\arccos\left(\frac{1}{2 + 2D\Delta\tau/\sigma^2}\right) - 1$$

while for displacements separated in time from each other, we have $\rho = 0$ [Equation (50)] and the expression simplifies to

$$\langle\cos\tilde{\Theta}\rangle = 0.$$

If we let $D = 0$, both of these expressions reduce to the expressions for stationary particles [Equations (19) and (20)] as expected.

Let us now consider the time dependence of the expression for successive displacements. For successive displacements $\tau = \Delta\tau$, so we can formulate it in either τ or $\Delta\tau$; we choose τ to connect to previous approaches. We then have

$$\langle\cos\tilde{\Theta}\rangle(\tau = 0) = \frac{2}{\pi}\arccos\left(\tfrac{1}{2}\right) - 1 = \frac{2}{\pi}\frac{\pi}{3} - 1 = -\tfrac{1}{3}$$

as well as

$$\lim_{\tau\to\infty}\langle\cos\tilde{\Theta}\rangle = \frac{2}{\pi}\arccos(0) - 1 = \frac{2}{\pi}\frac{\pi}{2} - 1 = 0.$$

Thus $\langle\cos\tilde{\Theta}\rangle(\tau)$ starts at $-\frac{1}{3}$ and tends to 0 for large τ.

Mean angle betwen displacements

Finally, from the results for stationary particles [Equation (21)] the mean angle between displacements is

$$\langle\tilde{\Theta}\rangle = \pi - \arccos(-\rho).$$

For successive displacements, we should insert ρ from Equation (48) into this expression, which results in

$$\langle\tilde{\Theta}\rangle = \pi - \arccos\left(\frac{1}{2 + 2D\Delta\tau/\sigma^2}\right),$$

while for displacements separated in time from each other, we have $\rho = 0$ (Equation (50)) and the expression simplifies to

$$\langle\tilde{\Theta}\rangle = \frac{\pi}{2}.$$

Again, both of these expressions reduce to the expressions for stationary particles (Equations (22) and (23)) if we let $D = 0$, as expected.

If we consider the time-dependence of the expression for successive displacements, then we have

$$\langle\tilde{\Theta}\rangle(\tau = 0) = \pi - \arccos(\tfrac{1}{2}) = \pi - \frac{\pi}{3} = \frac{2\pi}{3}$$

and

$$\lim_{\tau\to\infty}\langle\tilde{\Theta}\rangle = \pi - \arccos(0) = \pi - \frac{\pi}{2} = \frac{\pi}{2}$$

so $\langle\tilde{\Theta}\rangle(\tau)$ starts at $2\pi/3$ and tends to $\pi/2$ for large τ.

Two dimensions

We now turn to particles moving by Brownian motion in two dimensions. Overall, the derivation we made for stationary particles carries through in the same way, aside from that we need to re-interpret σ_τ [now given by Equations (44) and (45)] as well as the correlation coefficient ρ [now given by Equation (48) for successive displacements or by Equation (50) for displacements separated in time from each other].

Distribution of the scalar product

Using the results from stationary particles, Equation (27), we find

$$(f_R * f_R)(s) = \frac{1}{2\sigma_\tau^2}\exp\left(\frac{(\rho \pm 1)s}{\sigma_\tau^2(1-\rho^2)}\right)\begin{cases} s < 0 \\ s \geq 0. \end{cases}$$

For successive displacements, we should insert ρ from Equation (48) into this expression, while for displacements separated in time from each other, $\rho = 0$ [Equation (50)] and the expression simplifies to

$$(f_R * f_R)(s) = \frac{1}{2\sigma_\tau^2}\exp\left(\frac{\pm s}{\sigma_\tau^2}\right)\begin{cases} s < 0 \\ s \geq 0. \end{cases}$$

Both of these expressions reduce to the results for stationary particles when inserting $D = 0$, as expected.

The mean of the scalar product

The mean of the scalar product is intimately related to the velocity autocorrelation function, which in two dimensions is defined by Equation (30). Again, it follows immediately from Equation (25) and the results in one dimension [Equation (51)] that

$$\langle \Delta\tilde{\mathbf{R}}_{\Delta\tau,0} \cdot \Delta\tilde{\mathbf{R}}_{\tau+\Delta\tau,\tau} \rangle(\tau) = \begin{cases} 4D\Delta\tau + 4\sigma^2 & \tau = 0 \\ -2\sigma^2 & \tau = \Delta\tau \\ 0 & \tau > \Delta\tau. \end{cases} \tag{52}$$

The results for $\tau = 0$ and $\tau = \Delta\tau$ correspond to earlier results derived by Weber *et al.*[9] if one takes into account our assumptions.

Distribution of the angle between displacements

To calculate the distribution of the angle between displacement, we need to redo the introduction of the complex variables $\Delta\tilde{Z}_{\Delta\tau,0}$ and $\Delta\tilde{Z}_{\tau+\Delta\tau,\tau}$ [Equation (33)], now with the displacements corresponding to particles moving by Brownian motion. This causes no difficulties, and just like for stationary particles we find

$$\sigma^2_{z_{\Delta\tau,0}} = 2\sigma^2_\tau$$
$$\sigma^2_{z_{\tau+\Delta\tau,\tau}} = 2\sigma^2_\tau.$$

We also find

$$\text{Covar}\left[\Delta\tilde{Z}_{\Delta\tau,0}, \Delta\tilde{Z}_{\tau+\Delta\tau,\tau}\right] = -2\sigma^2$$

for successive displacements, so that the correlation coefficient, ρ, is given by the same expression as in one dimension, Equation (48); for displacements separated in time from each other we instead find

$$\text{Covar}\left[\Delta\tilde{Z}_{\Delta\tau,0}, \Delta\tilde{Z}_{\tau+\Delta\tau,\tau}\right] = 0$$

so that the correlation cofficient, $\rho = 0$, just like in one dimension [Equation (50)]. We can also show that it still holds that

$$\sigma^2_\tau \left(1 - \rho^2\right) > 0.$$

The rest of the derivation then follows through, so we may finally conclude that for particles moving by Brownian motion, the distribution of the angle is given by

$$f_{\tilde{\Theta}}(\theta) = \frac{1}{\pi} \frac{1 - \rho^2}{1 - \rho^2 \cos^2\theta} \left(1 + \frac{\rho\cos\theta \arccos\left(-\rho\cos\theta\right)}{\sqrt{1 - \rho^2\cos^2\theta}}\right)$$

with ρ given by Equation (48) for successive displacements, while for displacements separated in time from each other, $\rho = 0$ [Equation (50)] and the expression simplifies to

$$f_{\tilde{\Theta}}(\theta) = \frac{1}{\pi}.$$

Inserting $D = 0$ we find the same expressions as for stationary particles.

Mean cosine of the angle between displacements

The derivation of the mean cosine of the angle between displacements that we performed for stationary particles only depends on the expression for the distribution of the angle, so we can immediately conclude that the mean cosine of the angle between displacements is given by Equation (39), *viz.*

$$\langle \cos \tilde{\Theta} \rangle = \frac{\mathbf{E}\,(\rho) - (1 - \rho^2)\mathbf{K}\,(\rho)}{\rho}.$$

For successive displacements, we should insert ρ given by Equation (48) into this expression, while for displacements separated in time, $\rho = 0$ [Equation (50)] and the expression simplifies to

$$\langle \cos \tilde{\Theta} \rangle = 0.$$

As expected, we find the same expression as for stationary particles by inserting $D = 0$.

Let us now we consider the time-dependence of the expression for successive displacements. For $\tau = 0$, Equation (48) shows that ρ just becomes $-\frac{1}{2}$ and consequently that

$$\langle \cos \tilde{\Theta} \rangle (\tau = 0) = \frac{\mathbf{E}\left(-\frac{1}{2}\right) - (1 - \frac{1}{4})\mathbf{K}\left(-\frac{1}{2}\right)}{-\frac{1}{2}} = \tfrac{3}{2}\mathbf{K}\left(-\tfrac{1}{2}\right) - 2\mathbf{E}\left(-\tfrac{1}{2}\right) = \tfrac{3}{2}\mathbf{K}\left(\tfrac{1}{2}\right) - 2\mathbf{E}\left(\tfrac{1}{2}\right) \approx -0.406$$

using that the elliptic integrals are even. Conversely, for $\tau \to \infty$ we have from Equation (48) that $\rho \to 0$. Gradshteyn & Ryzhik[4] [Equations (8.113) and (8.114)] gives the expansions

$$\mathbf{K}\,(k) = \frac{\pi}{2}\left(1 + \tfrac{1}{4}k^2 + \cdots\right)$$
$$\mathbf{E}\,(k) = \frac{\pi}{2}\left(1 - \tfrac{1}{4}k^2 + \cdots\right)$$

so we find

$$\lim_{\tau\to\infty}\langle\cos\tilde{\Theta}\rangle = \lim_{\rho\to 0}\frac{\mathbf{E}\left(\rho\right)-(1-\rho^2)\mathbf{K}\left(\rho\right)}{\rho} =$$
$$\lim_{\rho\to 0}\frac{\pi}{2}\frac{1+\frac{1}{4}\rho^2+\cdots-(1-\rho^2)\big(1-\frac{1}{4}\rho^2+\cdots\big)}{\rho} =$$
$$\lim_{\rho\to 0}\frac{\pi}{2}\frac{\frac{1}{4}\rho^2+\frac{1}{4}\rho^2+\rho^2+\cdots}{\rho} = 0.$$

Overall, thus $\langle\cos\tilde{\Theta}\rangle(\tau)$ starts at $\frac{3}{2}\mathbf{K}\left(\frac{1}{2}\right)-2\mathbf{E}\left(\frac{1}{2}\right)\approx -0.406$ and tends to 0 for large τ.

Mean angle between displacements

Similarly, the derivation of the mean angle between displacements that we performed for stationary particles also only depends on the expression for the distribution of the angle, so we have that mean angle is given by Equation (41)

$$\langle\tilde{\Theta}\rangle = \pi - \arccos(-\rho).$$

As usual, for successive displacements we should insert ρ given by Equation (48) into this expression which results in

$$\langle\tilde{\Theta}\rangle = \pi - \arccos\left(\frac{1}{2+2D\Delta\tau/\sigma^2}\right),$$

while for displacements separated in time, we have $\rho = 0$ [Equation (50)] and the expression simplifies to

$$\langle\tilde{\Theta}\rangle = \frac{\pi}{2}.$$

Again, both of these expressions reduce to the expressions for stationary particles (Equations (43) and (42)) if we let $D = 0$, as expected.

If we consider the time-dependence of the expression for successive displacements, then we have

$$\langle\tilde{\Theta}\rangle(\tau=0) = \pi - \arccos(\tfrac{1}{2}) = \pi - \frac{\pi}{3} = \frac{2\pi}{3}$$

and

$$\lim_{\tau\to\infty}\langle\tilde{\Theta}\rangle = \pi - \arccos(0) = \pi - \frac{\pi}{2} = \frac{\pi}{2}$$

so $\langle\tilde{\Theta}\rangle(\tau)$ starts at $2\pi/3$ and tends to $\pi/2$ for large τ.

References

[1] R. Metzler, J.-H. Jeon, A. G. Cherstvy and E. Barkai, *Physical Chemistry Chemical Physics*, 2014, **16**, 24128–24164.

[2] L. P. Grishchuk, *Physical Review D*, 1996, **53**, 6784–6795.

[3] M. D. Springer, *The algebra of random variables*, John Wiley & Sons Inc, New York, NY, 1979.

[4] I. S. Gradshteyn and I. M. Ryzhik, *Table of integrals, series, and products*, Academic Press, Boston, MA, 7th edn., 2007.

[5] Y. Li, Q. He and R. S. Blum, *IEEE Signal Processing Letters*, 2020, **27**, 16–20.

[6] K. I. Park, Chapter 5. Characterization of random variables, in *Fundamentals of probability and stochastic processes with applications to communications*, Springer International Publishing, Cham, Switzerland, 2018, pp. 109–133.

[7] A. Hurford, *PLOS ONE*, 2009, **4**, e5632.

[8] I. S. Gradshteyn and I. M. Ryzhik, Chapter 8–9 Special functions, in *Table of integrals, series, and products*, Academic Press, Boston, 7th edn., 2007, pp. 859–1048.

[9] S. C. Weber, M. A. Thompson, W. E. Moerner, A. J. Spakowitz and J. A. Theriot, *Biophysical Journal*, 2012, **102**, 2443–2450.