Persistence in Brownian motion of an ellipsoidal particle in two dimensions

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We investigate the persistence probability $p(t)$ of the position of a Brownian particle with shape asymmetry in two dimensions. The persistence probability is defined as the probability that a stochastic variable has not changed its sign in the given time interval. We explicitly consider two cases – diffusion of a free particle and that of harmonically trapped particle. The later is particularly relevant in experiments which uses trapping and tracking techniques to measure the displacements. We provide analytical expressions of $p(t)$ for both the scenarios and show that in the absence of the shape asymmetry the results reduce to the case of an isotropic particle. The analytical expressions of $p(t)$ are further validated against numerical simulation of the underlying overdamped dynamics. We also illustrate that $p(t)$ can be a measure to determine the shape asymmetry of a colloid and the translational and rotational diffusivities can be estimated from the measured persistence probability. The advantage of this method is that it does not require the tracking of the orientation of the particle.

I. INTRODUCTION

Particles that exhibit a shape asymmetry are abundant in nature with sizes ranging from few nanometers to few micrometers. Over the last decade, accelerated by the advancement in particle chemistry, a plethora of such particles with enhanced transport properties have been developed in an attempt to mimic nature. These synthetically engineered colloids with multi-functional properties often find wide ranging applications in photonics, nano and biotechnology, drug delivery and other bio-medical uses. Unlike an isotropic particle, the shape asymmetry leads to different transport properties along the symmetry axes of the particle and any real-life application would require the knowledge of these transport properties. Perhaps, the most crucial of these transport properties are the translational and rotational diffusivities that characterizes their stochastic dynamics. For example, the diffusive dynamics of such particles are completely characterized by the mobility matrix. However, the extraction of the diffusivity from the measured mean-square displacement requires the simultaneous measurement of its translational and orientational degrees of freedom, which might not be always feasible.

In this article, we present an alternative approach to measure the diffusivity of shape asymmetric particle from its position coordinates alone. Our approach does not require the measurement of the symmetry axes of the particle. We choose the simplest asymmetric particle – an ellipsoid and look at its two dimensional Brownian motion. Since the dynamics of the translational and the orientational degrees of freedom are stochastic due to the thermal fluctuations from the bath, the position and the orientation are both random variables in time. We use the stochastic nature of the position to calculate the persistence probability $p(t)$ of the particle. The extraction of the diffusion coefficients along the two symmetry axes of the particle as well the rotational diffusion constant follows from the analytical expression of $p(t)$.

The persistence probability $p(t)$ of a stochastic variable is simply the probability that the variable has not changed sign up to time $t$. In physics, the persistence property has been investigated both theoretically\textsuperscript{1-24} and experimentally\textsuperscript{25-32} in spatially extended systems that are out of equilibrium. For a more comprehensive review of the persistence probability in spatially extended systems, we invite the readers to look at the recent review by Bray et al.\textsuperscript{33} and the brief review by Majumdar\textsuperscript{34} on the subject and the references therein. The persistence probability for such systems decays as a power law $p(t) \sim t^{-\theta}$, with $\theta$ being a non-trivial exponent. This algebraic decay of $p(t)$ has been established for a wide class of non-equilibrium systems that includes the classic random walk problem in finite\textsuperscript{12} and infinite medium\textsuperscript{7,17,34}, critical dynamics\textsuperscript{9,13}, diffusion in an infinite medium with\textsuperscript{15} and without advection\textsuperscript{28}, fluctuating interfaces\textsuperscript{1,3,4,6,23}, disordered systems\textsuperscript{14,35,36}, polymer dynamics\textsuperscript{16,37} and granular media\textsuperscript{38,39}. The estimation of the exponent $\theta$ for a general stochastic process is notoriously difficult and the exact form of $p(t)$ exists in very few cases when the process is Gaussian as well Markovian. For a stochastic process $x(t)$ which is Gaussian as well Markovian, the non-stationary process can be mapped into a stationary Ornstein-Uhlenbeck process $\tilde{X}(T)$ via suitable transformations that takes $x \rightarrow \tilde{X}$ and $t \rightarrow T$, with the consequence that the correlator $C(0,t) \equiv \langle \tilde{X}(T)\tilde{X}(0) \rangle$ decay exponentially at all times. Following Slepian\textsuperscript{40}, if the stationary correlator $C(t)$ of a stochastic process decays purely exponentially at all times, the persistence probability $P(X)$ is proportional to $C(t)$ and $p(t)$ can then be reconstructed back by the inverse time transformation applied to $\tilde{X}$. In the case when $C(T)$ does not decay exponentially, the exponent $\theta$ can be extracted using the independent interval approximation (IIA)\textsuperscript{,} provided the density of zero crossings remain finite. In the present scenario, as the calculations reveal, the IIA is not required and suitable transformations space and time takes the non-stationary correlation function into a stationary correlator which then be used to calculate $p(t)$.

The rest of the article is organized as follows. In Section II present the results for the two-time correlation function the position of a free Brownian particle with shape asymmetry. The survival probability is determined from this correlation function. In Section III we carry out a perturbative expansion for the position of an anisotropic Brownian particle trapped in a harmonic potential. The mean-square displacement for the displacements along the two directions and the two-time correlation functions are calculated using the perturbative ex-
pansion. Finally, the persistence probability is constructed from this two-time correlation function. A brief conclusion and the relevance of the work is presented in Section IV.

II. ELLIPSOIDAL PARTICLE IN TWO-DIMENSIONS

We consider an ellipsoidal particle in two dimension with mobilities $\Gamma_x$ and $\Gamma_y$ along the $x$ and $y$ direction respectively and a single rotational mobility $\Gamma_\theta$. The particle is immersed in a bath at a temperature $T$, so that the translational diffusion coefficients along the two directions are given by $D_x = k_BT\Gamma_x$, $D_y = k_BT\Gamma_y$, and the rotational diffusion constant $D_\theta = k_BT\Gamma_\theta$. In a frame fixed to the particle, the translational and the rotational motion of the particle is completely decoupled. However, in the lab-frame, the shape asymmetry of the particle leads to a coupling between the translational and rotational motions of the particle. In the body frame the equations of motion of the particle take the form

$$\Gamma_x^{-1}\frac{\partial \dot{x}}{\partial t} = F_x \cos \theta(t) + F_y \sin \theta(t) + \ddot{\eta}_x(t)$$

$$\Gamma_y^{-1}\frac{\partial \dot{y}}{\partial t} = F_y \cos \theta(t) + F_x \sin \theta(t) + \ddot{\eta}_y(t)$$

$$\Gamma_\theta^{-1}\frac{\partial \dot{\theta}}{\partial t} = \tau + \ddot{\eta}_\theta,$$

where $F_x$ and $F_y$ are the forces acting on the particle along the $x$ and $y$ directions and $\tau$ is the torque acting on the particle. The correlations of the thermal fluctuations in the body frame are given by

$$\langle \ddot{\eta} \rangle = 0 \quad \text{and} \quad \langle \ddot{\eta}_x(t)\ddot{\eta}_x(t') \rangle = 2D_x\delta(t-t')$$

In the lab frame, the displacements are related to the body frame as,

$$\delta x = \cos \theta \delta \dot{x} - \sin \theta \delta \dot{y}$$

$$\delta y = \cos \theta \delta \dot{y} + \sin \theta \delta \dot{x}$$

Using Eq. (1), the corresponding Langevin equation in the lab frame is given by,

$$\frac{\partial x_i}{\partial t} = -\Gamma_{ij}\frac{\partial U}{\partial x_j} + \eta_i,$$

where $U(r)$ is the external potential and $\Gamma$ is the mobility tensor given by,

$$\Gamma = \begin{pmatrix} \Gamma + \frac{\Delta \Gamma}{2} \cos 2\theta & \frac{\Delta \Gamma}{2} \sin 2\theta \\
\frac{\Delta \Gamma}{2} \sin 2\theta & \Gamma - \frac{\Delta \Gamma}{2} \cos 2\theta \end{pmatrix}$$

with $\Gamma = (\Gamma_x + \Gamma_y)/2$ and $\Delta \Gamma = \Gamma_y - \Gamma_x$. In the component form, the mobility tensor is given by $\Gamma_{ij} = \Gamma_{0ij} + \frac{\Delta \Gamma}{2} \Delta R_{ij} \delta(t-t')$, where the form of $\Delta \Gamma$ is given by

$$\Delta \Gamma = \begin{pmatrix} \cos 2\theta & \sin 2\theta \\
\sin 2\theta & -\cos 2\theta \end{pmatrix}$$

Using the correlation of the thermal fluctuations from Eq. (2) and Eq. (1), the moments of the stochastic forces are given by,

$$\langle \eta \rangle = 0 \quad \text{and} \quad \langle \eta(t)\eta(t') \rangle = 2k_BT\Gamma \delta(t-t')$$

We first look at the case of a free ellipsoidal particle. Setting the external potential to zero, the formal solution to the equation of motion takes the form

$$x_i(t) = \int_0^t \eta_i(t')dT' + x_i(0)$$

The mean-square displacement of the particle, averaged over the orientational noise can be explicitly calculated from the above equation as,

$$\langle \Delta x_i^2 \rangle_{\eta_0} = 2k_BT \int_0^t \int_0^{t'} d\tau d\tau' \langle \eta_i(t')\eta_i(t'') \rangle$$

$$\quad = 2k_BT \int_0^t d\tau \int_0^{t'} d\tau' \langle \Gamma_{ii}[\theta(t')]\eta_i(t'-\tau') \rangle$$

$$\quad = 2k_BT \int_0^t d\tau' \langle \Gamma_{ii}[\theta(t')]\eta_i \rangle$$

Using the explicit form of $\Gamma_{xx}$ the mean-square displacement along the $x$-direction reads

$$\langle \Delta x_1^2 \rangle_{\eta_0} = 2k_BT \int_0^t d\tau' \left[ \Gamma_1 + \frac{\Delta \Gamma}{2} \cos \theta(t') \right]$$

The ensemble average of $\cos \theta(t)$ over the thermal fluctuations in the orientational degrees of freedom can be done explicitly by noting the fact that $\Delta \theta = \theta(t) - \theta_0$ is a Gaussian random variable and consequently the following identity holds:

$$\langle e^{2\Delta \theta} \rangle_{\eta_0} = e^{-m_d^2t^2}.$$
In order to transform the non-stationary correlation into a stationary correlation we first make the transformation \(X(t) = x(t)/\sqrt{\langle x^2(t) \rangle_{\theta}}\), and the correlation \(\langle X(t_1)X(t_2) \rangle_{\theta}\), reads as
\[
\langle X(t_1)X(t_2) \rangle_{\theta} = \sqrt{\frac{2D t_2}{2D t_1}} \sqrt{\frac{1 + \frac{\Delta t}{2t} \cos \theta_0 \left(\frac{1 - e^{-4D\theta_0}}{4D\theta_0}\right)}{1 + \frac{\Delta t}{2t} \cos \theta_0 \left(\frac{1 - e^{-4D\theta_0}}{4D\theta_0}\right)}}
\]
with \(\Delta t = t_2 - t_1\) and \(\theta_0\) is the initial angle. The first step would be to determine the overall constant \(\mathcal{A}\) in the expression for the persistence probability. This can be fixed by fitting the data with the form of \(p(t)\) given in Eq. (20).

We now define the transformation in time as
\[
e^t = \sqrt{2Dt} \left[ 1 + \frac{\Delta D}{2D} \cos \theta_0 \left(\frac{1 - e^{-4D\theta_0}}{4D\theta_0}\right)\right]
\]
and Eq. (16) takes the simple form of
\[
\langle X(T_1)X(T_2) \rangle_{\theta} = e^{-\langle T_1 - T_2 \rangle/2}
\]
(18)

Following Slepian, if the correlation function of a stochastic variable \(X(T)\) decays exponentially for all times \(C_{XX}(T) = e^{-\lambda T}\), then the persistence probability is given by
\[
P(T) \sim \sin^{-1} e^{-\lambda T}.
\]
Asymptotically, \(P(T)\) takes the form \(P(T) \sim e^{-\lambda T}\). Consequently, looking at Eq. (18) and transforming back in real time \(t\), the persistence probability reads as
\[
p(t) \sim \frac{1}{\sqrt{2Dt}} \left[ 1 + \frac{\Delta D}{2D} \cos \theta_0 \left(\frac{1 - e^{-4D\theta_0}}{4D\theta_0}\right)\right]^{-1/2}.
\]
(20)

In the absence of any asymmetry, the expression for \(p(t)\) correctly reproduces the persistence probability of that of a random walker. Rearranging Eq. (20), the quantity \(t^{1/2}p(t)\) can be recast as
\[
t^{1/2}p(t) \sim \frac{1}{\sqrt{2D}} \left[ 1 + \frac{\Delta D}{D} \cos \theta_0 \left(\frac{1 - e^{-4D\theta_0}}{8\theta_0}\right)\right]^{-1/2}.
\]
(21)

In the limit of \(\Delta D \to 0\), the persistence probability reduces to that of a random walker \(p(t) \sim t^{-1/2}\).

To test Eq. (20), we performed numerical integration of the equations of motion using an Euler scheme for discretization. The initial condition was chosen from a Gaussian distribution with a very small width, so that the sign of \(x(0)\) is clearly defined. The trajectories was evolved in time with an integration time-step of \(\delta t = 0.001\). At every instant the the survival of the particle was checked by looking at the sign of \(x(t)\). Fraction of trajectories for which the position did not change its sign up to time \(t\) gave the survival probability \(p(t)\). A total of \(10^9\) trajectories were used in estimating the survival probability. A comparison of the measured \(p(t)\) with that of the predictions of Eq. (20) is shown Fig. 1 and Fig. 2. The comparison in Fig. 1 clearly shows that the survival probability can pick up the asymmetry in particle shape even when the difference in the diffusivities is as small as 5%.

The process to extract the the diffusion coefficients is as follows. The first step would be to determine the overall constant \(\mathcal{A}\) in the expression for the persistence probability. This can be fixed by fitting the data with the form of \(p(t)\) given in Eq. (20).
of the rotational diffusion coefficient. This is illustrated in Fig. 2. In fact, fitting the data for \( \bar{p} \) with \( \Delta D/\bar{D} \) and \( D_\theta \) as fit parameters yields very good estimates for \( \Delta D/\bar{D} \) and \( D_\theta \). A comparison of these values obtained from the fit with that of the actual values is shown in Table I.

| \( \Delta D/\bar{D} \) | \( D_\theta \) | Estimated \( \Delta D/\bar{D} \) | Estimated \( D_\theta \) |
|-----------------------|-------------|-----------------------------|-----------------------------|
| 2/3                   | 0.01        | 0.6698 ± 0.0018             | 0.0117 ± 0.0002             |
|                       | 0.1         | 0.1146 ± 0.0009             | 0.6799 ± 0.002              |
|                       | 1.0         | 1.076 ± 0.06                | 0.681 ± 0.0295              |

Table I. A comparison of the actual values of \( \Delta D/\bar{D} \) and \( D_\theta \) used in the simulations to those obtained from the fit of the data for \( \mathcal{A}/2\bar{D} p^r(t) \).

It should be pointed out, that the values of \( \Delta D/\bar{D} \) and \( D_\theta \) obtained from the fit are sensitive to the value \( \mathcal{A} \) and a careful estimation of \( \mathcal{A} \) is of paramount importance.

### III. Harmonically Trapped Ellipsoidal Particle

In experiments, the tracking of colloidal particles are usually done with laser traps and consequently it is pertinent to discuss the scenario where an ellipsoidal particle is trapped in a harmonic trap. In the following, we assume that the harmonic trap is isotropic and there is no preferential direction of alignment. Further, if we suppose a strong confinement, then at late times the deviations from the mean position of the particle is practically zero. Accordingly, the particle rotates freely so that the angular displacements obey Gaussian statistics. The potential confinement has the form \( U(x, y) = k(x^2 + y^2)/2 \) and the corresponding Langevin equation from Eq. (4) take the form

\[
\frac{\partial x}{\partial t} = -kx \left( \Gamma + \frac{1}{2} \Delta \Gamma \cos \theta(t) \right) - \frac{1}{2}ky\Delta \Gamma \sin \theta(t) + \tilde{\eta}_x(t)
\]

\[
\frac{\partial y}{\partial t} = -\frac{1}{2}kx\Delta \Gamma \sin \theta(t) - ky \left( \Gamma - \frac{1}{2} \Delta \Gamma \cos \theta(t) \right) + \tilde{\eta}_y(t)
\]

\[
\frac{\partial \theta}{\partial t} = \tilde{\eta}_\theta,
\]

(23)

where the correlation of the thermal noise follows Eq. (7).

### A. Perturbative Expansion

Defining the vector \( \mathbf{R} \equiv (x, y)^T \), the equation takes the simple form

\[
\dot{\mathbf{R}} = -k \left[ \Gamma \mathbf{1} + \frac{\Delta \Gamma}{2} \tilde{\mathbf{R}}(t) \right] \mathbf{R}(t) + \eta(t)
\]

(24)

To solve the above equation, we use the perturbative expansion

\[
\mathbf{R}(t) = R_0(t) - \left( \frac{k\Delta \Gamma}{2} \right) \mathbf{R}_1(t) + \left( \frac{k\Delta \Gamma}{2} \right)^2 \mathbf{R}_2(t) + O \left( \frac{k\Delta \Gamma}{2} \right)^3
\]

(25)
Substituting Eq. (25) in Eq. (24) and keeping up to the linear order in $\kappa \Delta \Gamma / 2$ we obtain the equations for $R(t)$ and $R_1(t)$ as

$$
\dot{R}_0 = -\kappa \Gamma R_0(t) + \eta(t)
$$

$$
\dot{R}_1 = -\kappa \Gamma R_1(t) + \overline{R}(t) R_0(t)
$$

$$
\dot{R}_2 = -\kappa \Gamma R_2(t) + \overline{R}(t) R_1(t)
$$

(26)

The solutions for the Eq. (26) together with the initial condition $R(0) = 0$ take the form

$$
R_0(t) = \int_0^t \int_0^\tau e^{-\kappa \tau(\tau-\tau')} \eta(t') d\tau' d\tau
$$

$$
R_1(t) = \int_0^t \int_0^\tau e^{-\kappa \tau(\tau-\tau')} \overline{R}(t) R_0(t') d\tau' d\tau
$$

$$
R_2(t) = \int_0^t \int_0^\tau e^{-\kappa \tau(\tau-\tau')} \overline{R}(t) R_1(t') d\tau' d\tau
$$

(27)

In explicit form, the equal time correlation matrix $R_i(t) R_j(t)$ is then given by

$$
\langle R_i(t) R_j(t) \rangle_{\eta,\theta} = \langle R_0(t) R_0(t) \rangle_{\eta,\theta} - \left( \frac{\kappa \Delta \Gamma}{2} \right) \langle R_0(t) R_1(t) \rangle_{\eta,\theta}
$$

$$
+ \left( \frac{\kappa \Delta \Gamma}{2} \right)^2 \left[ \langle R_1(t) R_1(t) \rangle_{\eta,\theta} + 2 \langle R_0(t) R_2(t) \rangle_{\eta,\theta} \right] + O\left( \frac{\kappa \Delta \Gamma}{2} \right)^3
$$

(28)

where we have used the fact that $\langle R_0 R_1 \rangle = \langle R_0, R_1 \rangle$. Further, note that the thermal noise correlation given in Eq. (7) gives an additional factor of $\kappa \Delta \Gamma / 2$ in the correlation terms $\langle R_{\alpha,i}(t) R_{\beta,j}(t) \rangle$, where $\alpha, \beta$ denotes the order of the perturbation series.

We next proceed to calculate this equal time correlation matrix using the solutions in Eq. (27). The correlation matrix of $R_0(t)$ averaged over the translational and the rotational noise is then given by

$$
\langle R_0(t) R_0(t) \rangle_{\eta,\theta} = \int_0^t \int_0^\tau e^{-\kappa \tau(\tau-\tau')} \eta(t') \eta(t') d\tau' d\tau
$$

(29)

where in correlation of the thermal noise is understood as an outer product of the variable $\eta_i$ and $\eta_j$. Using Eq. (7), the calculation is straight forward and the final form of the correlation matrix is given by

$$
\langle R_0(t) R_0(t) \rangle_{\eta,\theta} = \frac{k_B T}{\kappa} \left[ 1 - e^{-2\kappa \tau} \right] + \Delta D \overline{R}(\theta_0) \left( e^{-4D_\theta t} - e^{-2\kappa \tau} \right)
$$

(30)

More explicitly, the mean-square displacement along the $x$ and $y$ direction are given by

$$
\langle x_0^2(t) \rangle_{\eta,\theta} = \frac{k_B T}{\kappa} \left( 1 - e^{-2\kappa \tau} \right) + \Delta D \cos 2\theta_0 \left( e^{-4D_\theta t} - e^{-2\kappa \tau} \right)
$$

and

$$
\langle y_0^2(t) \rangle_{\eta,\theta} = \frac{k_B T}{\kappa} \left( 1 - e^{-2\kappa \tau} \right) - \Delta D \cos 2\theta_0 \left( e^{-4D_\theta t} - e^{-2\kappa \tau} \right)
$$

(31)

The cross-correlation function $\langle x_0(t)x_0(t) \rangle_{\eta,\theta}$ reads

$$
\langle x_0(t)x_0(t) \rangle_{\eta,\theta} = \Delta D \sin 2\theta_0 \left( e^{-4D_\theta t} - e^{-2\kappa \tau} \right)
$$

(32)

In the limit of $\kappa \to 0$, Eqs. (31) and (32) reproduces the correct result of a free diffusion of an anisotropic particle given in Eqs. (12) and (13). On the other hand, for $\Delta \Gamma \to 0$ Eqs. (31) and (32) yields the correlation matrix for an isotropic Brownian particle in a harmonic trap.

Our next attempt is to look into the correction to the above expression that comes from $R_1(t)$ and $R_2(t)$. For this, we rewrite the solutions for $R_1(t)$ and $R_2(t)$ in explicit form as

$$
R_{1,i}(t) = \int_0^t \int_0^\tau e^{-\kappa \tau(\tau-\tau')} \sum_j R_{ij}(t') R_{0,j}(t') d\tau' d\tau
$$

$$
R_{2,i}(t) = \int_0^t \int_0^\tau e^{-\kappa \tau(\tau-\tau')} \sum_j R_{ij}(t') R_{1,j}(t') d\tau' d\tau
$$

(33)
Using the above relation, the averages of the trigonometric functions over the rotational noise take the form

\[
\begin{align*}
\langle \cos 2[\theta(t') + \theta(t'')] \rangle_\theta &= e^{-4D_\theta \theta(t' + t'')^2} , \\
\langle \sin 2[\theta(t') - \theta(t'')] \rangle_\theta &= 0.
\end{align*}
\]

The final form of the expressions is given by

\[
\langle x(t)x(t) \rangle_{\eta,\theta} = \langle y(t)y(t) \rangle_{\eta,\theta} = \left( \frac{k_B T}{\kappa} \right) \cos 2\theta_0
\] 
\[
\times e^{-4D_\theta \theta(t' + t'')^2} - e^{-2\lambda T} - e^{-2\lambda T + 4D_\theta \theta(t' + t'')} + 2 \left( \frac{k_B T}{\kappa} \right) \frac{\kappa \Delta \Gamma}{2} \left( 1 - e^{-2\lambda T} - e^{-2\lambda T} - e^{-2\lambda T + 4D_\theta \theta(t' + t'')} \right)
\] 

\[\tag{36}\]

In the limit of \( \kappa \to 0 \), both \( \langle \gamma_2^2(t) \rangle = \langle \gamma_1^2(t) \rangle = 0 \).

The final expression for the mean-square displacement along the \( x \) is given by

\[
\langle x^2(t) \rangle_{\eta,\theta} = \left( \frac{k_B T}{\kappa} \right) \left( 1 - e^{-2\lambda T} \right) \cos 2\theta_0
\] 
\[
\times e^{-4D_\theta \theta(t' + t'')^2} - e^{-2\lambda T} - e^{-2\lambda T + 4D_\theta \theta(t' + t'')} + 2 \left( \frac{k_B T}{\kappa} \right) \frac{\kappa \Delta \Gamma}{2} \left( 1 - e^{-4D_\theta \theta(t' + t'')} \right)
\]

\[\tag{39}\]

and that along the \( y \)-direction is given by

\[
\langle y^2(t) \rangle_{\eta,\theta} = \left( \frac{k_B T}{\kappa} \right) \left( 1 - e^{-2\lambda T} \right) \cos 2\theta_0
\] 
\[
\times e^{-4D_\theta \theta(t' + t'')^2} - e^{-2\lambda T} - e^{-2\lambda T + 4D_\theta \theta(t' + t'')} + 2 \left( \frac{k_B T}{\kappa} \right) \frac{\kappa \Delta \Gamma}{2} \left( 1 - e^{-4D_\theta \theta(t' + t'')} \right)
\]

\[\tag{40}\]

B. Mean-square displacement for large rotational diffusion constant

In this section we present an alternate expression for mean-square displacement of an anisotropic particle which is valid for whose rotational diffusion constant is large as compared to the inverse times scales \( \kappa \Delta \Gamma \) and \( \kappa \Delta \Gamma \). In such a scenario, since the particle rotates faster, the mobility of the anisotropic particle is an average mobility over the rotational noise. We start our analysis with Eq. (41), but we set \( R(0) = 0 \).
In terms of the new variable \( u \), the solution for \( \mathbf{R}(t) \) takes the form

\[
\mathbf{R}(t) = t \int_0^1 du \, u e^{-\frac{1}{2} \Delta t u} e^{-\frac{1}{2} \Delta t t} \int_{x_{i=0}} e^r \mathbf{R}([\theta(t)]) \eta[t(1-u)]
\]

(41)

The equal-time correlation is then given by

\[
\langle \mathbf{R}(t) \mathbf{R}(t) \rangle = \int_0^1 du \int_0^1 du' e^{-\frac{1}{2} \Delta t u} e^{-\frac{1}{2} \Delta t t} \int_{x_{i=0}} e^r \mathbf{R}([\theta(t)]) \langle \eta[t(1-u)] \eta[t(1-u')] \rangle
\]

(42)

The correlation of the thermal noise in the transformed variable is

\[
\langle \eta[t(1-u)] \eta[t(1-u')] \rangle = \frac{2k_B T}{t} \delta[u-u']
\]

(43)

Substituting the noise correlation into Eq. (44) and integration over \( u' \) we get

\[
\langle \mathbf{R}(t) \mathbf{R}(t) \rangle = 2k_B T t \int_0^1 du e^{-\frac{1}{2} \Delta t u} e^{-\frac{1}{2} \Delta t t} \int_{x_{i=0}} e^r \mathbf{R}([\theta(t)]) \left[ \Gamma \mathcal{I} + \frac{\Delta \Gamma}{2} \mathbf{R}([\theta(t)]) \right]
\]

(44)

In the asymptotic limit, the integral is dominated by small values of \( u \), the integral in the exponential from \( t(1-u) \) to \( t \) is vanishingly small and can be set to zero. Further, we set \( \mathbf{R}([\theta(t)]) \approx \mathbf{R}([\theta(t)]) \). Consequently, the correlation matrix averaged over the translational noise take the form

\[
\langle \mathbf{R}(t) \mathbf{R}(t) \rangle = 2k_B T T \int_0^1 du e^{-\frac{1}{2} \Delta t t} \left[ \Gamma \mathcal{I} + \frac{\Delta \Gamma}{2} \mathbf{R}([\theta(t)]) \right]
\]

(45)

and performing the average over the rotational noise and the integral over \( u \) we arrive at

\[
\langle \mathbf{R}(t) \mathbf{R}(t) \rangle_{\eta, \beta} = \frac{k_B T}{k} \left( 1 - e^{-2\Delta T} \right) \left( \Gamma \mathcal{I} + \frac{\Delta \Gamma}{2} \mathbf{R}([\theta(t)]) \right).
\]

(46)

C. Persistence Probability

We now turn our attention to the persistence probability of the harmonically trapped ellipsoidal particle. For this, we focus on the two time correlation function \( \langle x(t_1)x(t_2) \rangle_{\eta, \beta} \). Using the perturbation series given in Eq. (25) we have up to order \( O(\kappa \Delta \Gamma / 2) \)

\[
\langle x(t_1)x(t_2) \rangle_{\eta, \beta} = \langle x_0(t_1)x_0(t_2) \rangle_{\eta, \beta} - \left( \frac{\kappa \Delta \Gamma}{2} \right) \left[ \langle x_0(t_1)x_1(t_2) \rangle_{\eta, \beta} + \langle x_0(t_2)x_1(t_1) \rangle_{\eta, \beta} \right]
\]

(52)

where \( t_1 > t_2 \). The correlation functions \( \langle x_0(t_1)x_1(t_2) \rangle_{\eta, \beta} \) and \( \langle x_0(t_2)x_1(t_1) \rangle_{\eta, \beta} \) are equal only in the asymptotic limit, that is for \( t_1 \) and \( t_2 \) large. In this limit, the expression for the two time correlation function takes the form

\[
\langle x(t_1)x(t_2) \rangle_{\eta, \beta} = \langle x_0(t_1)x_0(t_2) \rangle_{\eta, \beta} - \left( \frac{\kappa \Delta \Gamma}{2} \right) \left[ \langle x_0(t_1)x_1(t_2) \rangle_{\eta, \beta} \right]
\]

(53)

The correlation functions \( \langle x_0(t_1)x_0(t_2) \rangle_{\eta, \beta} \) and \( \langle x_0(t_1)x_1(t_2) \rangle_{\eta, \beta} \) are derived in in Appendix A (see Eq. (B.1) ) and in Appendix D (see Eq. (D.4)), respectively. For completeness, we quote the main results here.

\[
\langle x_0(t_1)x_0(t_2) \rangle_{\eta, \beta} = \frac{k_B T}{k} \left[ e^{2\Delta T t_1} - e^{4\Delta T t_1 t_2} \right] + \left( \frac{k_B T}{k} \right) \kappa \Delta \Gamma \cos 2\theta_0 e^{4\Delta T t_1} \left[ \frac{e^{4\Delta T t_1 t_2} - e^{-4\Delta T t_1 t_2}}{2k\Gamma - 2D_0} \right]
\]

(54)

\[
\langle x_0(t_1)x_1(t_2) \rangle_{\eta, \beta} = \left( \frac{k_B T}{k} \right) \cos 2\theta_0 \left[ e^{4\Delta T t_1 t_2} - e^{-4\Delta T t_1 t_2} \right] + \left( \frac{k_B T}{k} \right) \left( \frac{\Delta \Gamma}{2} \right) e^{4\Delta T t_1} \left[ \frac{e^{4\Delta T t_1 t_2} - e^{-4\Delta T t_1 t_2}}{2k\Gamma + 4D_0} \right]
\]

(55)
Note that in calculating the two time correlation function up to an order $O(\kappa \Delta \Gamma)$, we will use only the first term appearing in Eq. (55). Looking at Eq. (53) and Eqs. (54) and (55) it is clear that the first term contained in the parenthesis in Eq. (55) cancels with the term proportional to $\Delta \Gamma$ in Eq. (54). The final expression for $\langle x(t_1) x(t_2) \rangle_{\eta, \theta}$ reads

$$\langle x(t_1) x(t_2) \rangle_{\eta, \theta} = \left( \frac{2k_B T}{\kappa} \right) e^{-\kappa \Gamma t_1} \left[ \sinh \kappa \Gamma t_2 + \frac{\kappa \Delta \Gamma}{2} \cos 2\theta_0 e^{-\kappa \Gamma t_2} \left( 1 - \frac{e^{-4D_{\theta} t_2}}{4D_{\theta}} \right) \right]^{1/2}$$

As before, defining the variable $X(t) = x(t)/\sqrt{\langle x^2 \rangle_{\eta, \theta}}$, the correlation function of $\langle X(t_1)X(t_2)\rangle_{\eta, \theta}$ is given by

$$\langle X(t_1)X(t_2)\rangle_{\eta, \theta} = \frac{e^{-\kappa \Gamma t_1/2}}{e^{-\kappa \Gamma t_2/2}} \left[ \sinh \kappa \Gamma t_2 + \frac{\kappa \Delta \Gamma}{2} \cos 2\theta_0 e^{-\kappa \Gamma t_2} \left( 1 - \frac{e^{-4D_{\theta} t_2}}{4D_{\theta}} \right) \right]^{1/2}$$

Using the transformation $\tilde{X} = e^{\kappa \Gamma t} \left[ \sinh \kappa \Gamma t + \frac{\kappa \Delta \Gamma}{2} \cos 2\theta_0 e^{\kappa \Gamma t} \left( 1 - \frac{e^{-4D_{\theta} t_2}}{4D_{\theta}} \right) \right]^{1/2}$ for an imaginary time variable $T$, the correlation function $\langle X(T_1)X(T_2)\rangle_{\eta, \theta}$ becomes a stationary correlator $\langle X(T_1)X(T_2)\rangle_{\eta, \theta} = e^{-\kappa \Gamma T_1/2}$ and the corresponding persistence probability is given by

$$p(t) \sim \frac{\sqrt{e^{-\kappa \Gamma t/2}}}{\left[ \sinh \kappa \Gamma t + \frac{\kappa \Delta \Gamma}{2} \cos 2\theta_0 e^{-\kappa \Gamma t} \left( 1 - \frac{e^{-\eta_{\theta} t}}{4D_{\theta}} \right) \right]^{1/2}}$$

In the limit of $\Delta \Gamma \to 0$ the equation correctly reproduces the persistence of probability of an isotropic particle in the presence of a harmonic trap. The other limit of $\kappa \to 0$ reproduces the persistence probability of a free anisotropic particle derived in Eq. (20).

**IV. CONCLUSION**

In summary, we have determined the persistence probability of an anisotropic particle in two spatial dimensions, in the presence as well as in the absence of a confining harmonic potential. The two time correlation functions of the position of the particle has been calculated in both cases. In the case of a harmonically confined particle, a perturbative solution has been provided for the correlation functions. The persistence probability is computed from the two-time correlation function using suitable transformations in space and time. The determination of the rotational and the translational diffusion coefficients have been explicitly carried out for an anisotropic particle that undergoes free Brownian motion. Additionally, the analytical results have been confirmed by numerical simulation of the underlying stochastic dynamics.

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Appendices

Appendix A  CALCULATION OF $\langle R_{0i}(t)R_{0j}(t) \rangle$.

$\langle R_{0i}(t)R_{0j}(t) \rangle_{\eta,\beta} = \int_0^t dt' \int_0^t dt'' e^{-2\bar{\Gamma}(t-t')} e^{-2\bar{\Gamma}(t''-t')} \left[ \bar{\Gamma} I + \frac{\Delta \Gamma}{2} \bar{R}(t') \right] \delta(t' - t'') \tag{A.1}$

$\langle R_{0i}(t)R_{0j}(t) \rangle_{\eta,\beta} = 2k_B T e^{-2\bar{\Gamma}t} \int_0^t dt' e^{2\bar{\Gamma}t'} \left[ \bar{\Gamma} I + \frac{\Delta \Gamma}{2} \bar{R}(t') \right] \eta_{\eta,\beta} \tag{A.2}$

$\langle R_{0i}(t)R_{0j}(t) \rangle_{\eta,\beta} = \frac{k_B T}{k} \bar{I} \left( 1 - e^{-2\bar{\Gamma}t} \right) + 2k_B T e^{-2\bar{\Gamma}t} \int_0^t dt' e^{2\bar{\Gamma}t'} \frac{\Delta \Gamma}{2} \bar{R}(t_0) e^{-4\bar{\Gamma}t} \tag{A.3}$

$\langle R_{0i}(t)R_{0j}(t) \rangle_{\eta,\beta} = \frac{k_B T}{k} \bar{I} \left( 1 - e^{-2\bar{\Gamma}t} \right) + \Delta D \bar{R}(t_0) e^{-2\bar{\Gamma}t} \left( \frac{e^{(2\bar{\Gamma}T-4\bar{\Gamma}t_0)} - 1}{2\bar{\Gamma} - 4\bar{\Gamma}} \right) \tag{A.4}$

$\langle R_{0i}(t)R_{0j}(t) \rangle_{\eta,\beta} = \frac{k_B T}{k} \bar{I} \left( 1 - e^{-2\bar{\Gamma}t} \right) + \Delta D \bar{R}(t_0) \left( \frac{e^{-4\bar{\Gamma}t_0} - e^{-2\bar{\Gamma}t}}{2\bar{\Gamma} - 4\bar{\Gamma}} \right) \tag{A.5}$

Appendix B  CALCULATION OF $\langle R_{0i}(t)R_{1,j}(t) \rangle$.

Calculation of $\langle R_{0i}(t)R_{1,j}(t) \rangle$.

$\langle R_{0i}(t_1)R_{0j}(t_2) \rangle_{\eta} = \int_0^{t_1} dt' \int_0^{t_2} dt'' e^{-2\bar{\Gamma}(t_1-t')} e^{-2\bar{\Gamma}(t_2-t'')} \left( \eta(t_1)\eta(t_2) \right)_{\eta} \tag{B.1}$

$\langle R_{0i}(t_1)R_{0j}(t_2) \rangle_{\eta} = 2k_B T e^{-2\bar{\Gamma}(t_1+t_2)} \int_0^{t_1} dt' \int_0^{t_2} dt'' e^{2\bar{\Gamma}(t_1+t_2)} \left[ \bar{\Gamma} \delta_{ij} + \frac{\Delta \Gamma}{2} R_{ij}(t_1) \right] \delta(t_1 - t_2) + 2k_B T \frac{\Delta \Gamma}{2} e^{-2\bar{\Gamma}(t_1+t_2)} \int_0^{t_1} dt' \int_0^{t_2} dt'' e^{2\bar{\Gamma}(t_1+t_2)} R_{ij}(t_1) \delta(t_1 - t_2) \tag{B.1}$

$\langle R_{0i}(t_1)R_{0j}(t_2) \rangle_{\eta} = 2k_B T \bar{\Gamma} \delta_{ij} e^{-2\bar{\Gamma}(t_1+t_2)} \int_0^{\min(t_1,t_2)} dt_1' e^{2\bar{\Gamma}t_1'} + 2k_B T \frac{\Delta \Gamma}{2} e^{-2\bar{\Gamma}(t_1+t_2)} \int_0^{\min(t_1,t_2)} dt_1' e^{2\bar{\Gamma}t_1'} R_{ij}(t_1') \tag{B.1}$

$\langle R_{0i}(t_1)R_{0j}(t_2) \rangle_{\eta} = \frac{k_B T}{\bar{\Gamma}} \delta_{ij} \left[ e^{-2\bar{\Gamma}t_1} - e^{-2\bar{\Gamma}t_2} \right] + k_B T \Delta \bar{\Gamma} e^{-2\bar{\Gamma}(t_1+t_2)} \int_0^{\min(t_1,t_2)} dt_1' e^{2\bar{\Gamma}t_1'} R_{ij}(t_1') \tag{B.1}$

$\langle R_{0i}(t)R_{1,j}(t) \rangle_{\eta,\beta} = \left\langle R_{0j}(t) \int_0^t dt' e^{-2\bar{\Gamma}(t-t')} \sum_k R_{ik}(t') R_{0k}(t') \right\rangle_{\eta,\beta} \tag{B.2}$

$\langle R_{0i}(t)R_{1,j}(t) \rangle_{\eta,\beta} = \left\langle \int_0^t dt' e^{-2\bar{\Gamma}(t-t')} \sum_k R_{ik}(t') \left\langle R_{0j}(t)R_{0k}(t') \right\rangle_{\eta} \right\rangle_{\eta} \tag{B.2}$

Using the final form of $\langle R_{0i}(t_1)R_{0j}(t_2) \rangle$ from Eq. (B.1) and identifying $t_1 \equiv t$, $t' \equiv t_2$ with $t' < t$ we get

$\langle R_{0i}(t)R_{0j}(t) \rangle_{\eta} = \frac{k_B T}{\bar{\Gamma}} \delta_{ij} \left[ e^{-2\bar{\Gamma}t} - e^{-2\bar{\Gamma}t} \right] + k_B T \Delta \bar{\Gamma} e^{-2\bar{\Gamma}t} \int_0^t dt' e^{2\bar{\Gamma}t'} R_{ij}(t_1') \tag{B.3}$
Substituting Eq. (B.3) in Eq. (B.2) we get

\[ \langle R_{0,1}(t) R_{1,1}(t) \rangle_{\eta, \theta} = \int_0^t dt' e^{-i (t-t')} \left( k_B T \delta_{ik} (e^{-i T(t-t')} - e^{-i T(t')}) + k_B T (2 \alpha \Gamma e^{-2 i T(t-t')} \int_0^t dt'' e^{2 i T(t-t'')} R_{ik}(t'') \right) \right)_{\eta, \theta} \]

\[ \langle R_{0,1}(t) R_{1,1}(t) \rangle_{\eta, \theta} = \left( \frac{k_B T}{k} \right) \int_0^t dt' e^{-i (t-t')} \left( k_B T \delta_{ik} (e^{-i T(t-t')} - e^{-i T(t')}) + k_B T (2 \alpha \Gamma e^{-2 i T(t-t')} \int_0^t dt'' e^{2 i T(t-t'')} \sum_k R_{ik}(t'') \delta_{ik} \right) \right)_{\eta, \theta} \]

\[ \langle R_{0,1}(t) R_{1,1}(t) \rangle_{\eta, \theta} = \left( \frac{k_B T}{k} \right) \int_0^t dt' e^{-i (t-t')} \left( k_B T \delta_{ik} (e^{-i T(t-t')} - e^{-i T(t')}) + k_B T (2 \alpha \Gamma e^{-2 i T(t-t')} \int_0^t dt'' e^{2 i T(t-t'')} \sum_k R_{ik}(t'') \right) \right)_{\eta, \theta} \]

\[ (B.4) \]

For the mean-square displacement along the \( x \) and the \( y \) direction, the second term in the last line of Eq. (B.4) yields

\[ \left\langle \sum_k R_{ik}(t') R_{ik}(t') \right\rangle_{\eta} = \cos 2 \theta(t') \cos 2 \theta(t') + \sin 2 \theta(t') \sin 2 \theta(t') = \cos 2 (\theta(t') - \theta(t')) \]

\[ (B.5) \]

On the other hand for \( i \neq j \), the term \( \left\langle \sum_k R_{ik}(t') R_{jk}(t') \right\rangle_{\eta} = 0 \). Using Eq. (B.5) the contribution to the mean-square displacement along the \( x \)-direction becomes

\[ \left\langle \chi_0(t) \chi_1(t) \right\rangle_{\eta, \theta} = \left( \frac{k_B T}{k} \right) \cos 2 \theta_0 e^{-2 i T(t-t')} \left( \frac{(2 \alpha \Gamma + 4 D_\theta)}{2(2 \alpha \Gamma + 4 D_\theta)} - \frac{1 - e^{-4 D_\theta t'}}{4 D_\theta} \right) + k_B T \Delta \Gamma e^{-2 i T(t-t')} \int_0^t dt'' e^{2 i T(t-t'')} \left( \frac{1 - e^{-4 D_\theta t''}}{4 D_\theta} \right) \]

\[ (B.6) \]

and that along the \( y \)-direction takes the form

\[ \left\langle \gamma_0(t) \gamma_1(t) \right\rangle_{\eta, \theta} = - \left( \frac{k_B T}{k} \right) \cos 2 \theta_0 e^{-2 i T(t-t')} \left( \frac{(2 \alpha \Gamma + 4 D_\theta)}{2(2 \alpha \Gamma + 4 D_\theta)} - \frac{1 - e^{-4 D_\theta t'}}{4 D_\theta} \right) + k_B T \Delta \Gamma \left( \frac{1 - e^{-2 i T(t-t')}}{2(2 \alpha \Gamma + 4 D_\theta)} - \frac{e^{-2 i T(t-t')} - e^{-2 i T(t-t')}}{4 D_\theta} \right) \]

\[ (B.7) \]

**Appendix C** **CALCULATION OF** \( \langle R_{1,1}(t) R_{1,1}(t) \rangle \)

The correlation matrix now takes the form

\[ \langle R_{1,1}(t) R_{1,1}(t) \rangle_{\eta, \theta} = \int_0^t dt' \int_0^t dt'' e^{-i (t-t')} e^{-i (t-t'')} \right) \left\langle \sum_k R_{ik}(t'') \sum_l R_{lk}(t') \right\rangle_{\eta, \theta} \]

\[ (C.1) \]

Rearranging and averaging first over the translational noise we get,

\[ \langle R_{1,1}(t) R_{1,1}(t) \rangle_{\eta, \theta} = \int_0^t dt' \int_0^t dt'' e^{-i (t-t')} e^{-i (t-t'')} \right) \left\langle \sum_k R_{ik}(t'') \sum_l R_{lk}(t') \right\rangle_{\eta, \theta} \]

\[ (C.2) \]

\[ \langle R_{1,1}(t) R_{1,1}(t) \rangle_{\eta, \theta} = 2 k_B T \int_0^t dt' \int_0^t dt'' e^{-i (t-t')} e^{-i (t-t'')} \right) \left\langle \sum_k R_{ik}(t'') \sum_l R_{lk}(t') \right\rangle_{\eta, \theta} \]

\[ (C.3) \]
Integrating over the delta function and ignoring the term proportional to $\Delta \Gamma$ we get

$$
\langle R_1(t)R_1(t)\rangle_{\eta,\theta} = 2k_B T e^{-2\beta\Gamma} \int_0^t dt' \int_0^{t'} dt'' \int_0^{\min(t',t'')} \int_0^{\min(t',t'')} dt' e^{2\beta \Gamma t' + e^{-2\beta \Gamma t'} e^{-2\beta \Gamma t''} e^{-2\beta \Gamma t''} + e^{-2\beta \Gamma t'} - e^{-2\beta \Gamma t''} - e^{-2\beta \Gamma t'''}} \left( e^{e^{2\beta \Gamma t'} - 1} e^{-4D\theta(t' - t')} \right)
$$

(C.4)

In order to proceed further, we look at $\langle \chi_5^2(t) \rangle_{\eta,\theta}$ and $\langle \chi_4^2(t) \rangle_{\eta,\theta}$ by setting $i = j$ and subsequently using Eq. (B.5)

$$
\langle \chi_5^2(t) \rangle_{\eta,\theta} = 2k_B T e^{-2\beta\Gamma} \int_0^t dt' \int_0^{t'} dt'' \int_0^{t''} dt'' e^{2\beta \Gamma t'} \left( e^{2\beta \Gamma t'} - 1 \right) e^{-4D\theta(t' - t'')} \left( e^{2\beta \Gamma t'} - 1 \right) e^{-4D\theta(t'' - t')}
$$

(C.5)

Substituting for $\langle \cos 2[\theta(t') - \theta(t'')] \rangle_{\theta}$ from Eq. (35) we get

$$
\langle \chi_5^2(t) \rangle_{\eta,\theta} = 2k_B T e^{-2\beta\Gamma} \int_0^t dt' \int_0^{t'} dt'' \int_0^{t''} dt'' e^{2\beta \Gamma t'} e^{-4D\theta(t' + t' - 2\min(t',t''))}
$$

(C.6)

$$
\langle \chi_4^2(t) \rangle_{\eta,\theta} = 2k_B T e^{-2\beta\Gamma} \int_0^t dt' \int_0^{t'} dt'' \int_0^{t''} dt'' e^{2\beta \Gamma t'} e^{-4D\theta(t' + t' - 2\min(t',t''))} - \frac{1}{2\beta} \int_0^t dt' \int_0^{t'} dt'' \int_0^{t''} dt'' e^{2\beta \Gamma t'} e^{-4D\theta(t' + t' - 2\min(t',t''))}
$$

(C.7)

$$
\langle \chi_4^2(t) \rangle_{\eta,\theta} = 2k_B T e^{-2\beta\Gamma} \int_0^t dt' \int_0^{t'} dt'' \int_0^{t''} dt'' e^{2\beta \Gamma t'} e^{-4D\theta(t' + t' - 2\min(t',t''))}
$$

(C.8)

$$
\langle \chi_4^2(t) \rangle_{\eta,\theta} = 2k_B T e^{-2\beta\Gamma} \int_0^t dt' \int_0^{t'} dt'' \int_0^{t''} dt'' e^{2\beta \Gamma t'} \left( e^{2\beta \Gamma t' + e^{-4D\theta(t' - t'')} - e^{-4D\theta(t' - t'')} - e^{4D\theta(t' - t'')} \right) + e^{4D\theta(t' - t'')} \left( e^{4D\theta(t' - t'')} - e^{-4D\theta(t' - t'')} \right)
$$

(C.9)

$$
\langle \chi_4^2(t) \rangle_{\eta,\theta} = 2k_B T e^{-2\beta\Gamma} \int_0^t dt' \int_0^{t'} dt'' \int_0^{t''} dt'' e^{2\beta \Gamma t'} \left( e^{2\beta \Gamma t' + e^{-4D\theta(t' - t'')} - e^{-4D\theta(t' - t'')} - e^{4D\theta(t' - t'')} \right) + e^{4D\theta(t' - t'')} \left( e^{4D\theta(t' - t'')} - e^{-4D\theta(t' - t'')} \right)
$$

(C.10)

$$
\langle \chi_4^2(t) \rangle_{\eta,\theta} = 2k_B T e^{-2\beta\Gamma} \int_0^t dt' \int_0^{t'} dt'' \int_0^{t''} dt'' e^{2\beta \Gamma t'} \left( e^{2\beta \Gamma t' + e^{-4D\theta(t' - t'')} - e^{-4D\theta(t' - t'')} - e^{4D\theta(t' - t'')} \right) + e^{4D\theta(t' - t'')} \left( e^{4D\theta(t' - t'')} - e^{-4D\theta(t' - t'')} \right)
$$

(C.11)

$$
\langle \chi_4^2(t) \rangle_{\eta,\theta} = 2k_B T e^{-2\beta\Gamma} \int_0^t dt' \int_0^{t'} dt'' \int_0^{t''} dt'' e^{2\beta \Gamma t'} \left( e^{2\beta \Gamma t' + e^{-4D\theta(t' - t'')} - e^{-4D\theta(t' - t'')} - e^{4D\theta(t' - t'')} \right) + e^{4D\theta(t' - t'')} \left( e^{4D\theta(t' - t'')} - e^{-4D\theta(t' - t'')} \right)
$$

(C.12)

$$
\langle \chi_4^2(t) \rangle_{\eta,\theta} = 2k_B T e^{-2\beta\Gamma} \int_0^t dt' \int_0^{t'} dt'' \int_0^{t''} dt'' e^{2\beta \Gamma t'} \left( e^{2\beta \Gamma t' + e^{-4D\theta(t' - t'')} - e^{-4D\theta(t' - t'')} - e^{4D\theta(t' - t'')} \right) + e^{4D\theta(t' - t'')} \left( e^{4D\theta(t' - t'')} - e^{-4D\theta(t' - t'')} \right)
$$

(C.13)

$$
\langle \chi_4^2(t) \rangle_{\eta,\theta} = 2k_B T e^{-2\beta\Gamma} \int_0^t dt' \int_0^{t'} dt'' \int_0^{t''} dt'' e^{2\beta \Gamma t'} \left( e^{2\beta \Gamma t' + e^{-4D\theta(t' - t'')} - e^{-4D\theta(t' - t'')} - e^{4D\theta(t' - t'')} \right) + e^{4D\theta(t' - t'')} \left( e^{4D\theta(t' - t'')} - e^{-4D\theta(t' - t'')} \right)
$$

(C.14)

$$
\langle \chi_4^2(t) \rangle_{\eta,\theta} = 2k_B T e^{-2\beta\Gamma} \int_0^t dt' \int_0^{t'} dt'' \int_0^{t''} dt'' e^{2\beta \Gamma t'} \left( e^{2\beta \Gamma t' + e^{-4D\theta(t' - t'')} - e^{-4D\theta(t' - t'')} - e^{4D\theta(t' - t'')} \right) + e^{4D\theta(t' - t'')} \left( e^{4D\theta(t' - t'')} - e^{-4D\theta(t' - t'')} \right)
$$

(C.15)
Appendix D  CALCULATION OF \( \langle R_0(t) R_z(t) \rangle_\eta \)

\[
\langle R_0(t_1) R_1(t_2) \rangle_\eta = \left( R_0(t_1) \int_0^{t_2} dt'_2 e^{-\tau(t_2-t'_2)} \sum_k \mathcal{R}_{jk}(t'_2) R_{0,k}(t'_2) \right)_\eta
\]

\[
\langle R_0(t_1) R_1(t_2) \rangle_\eta = \int_0^{t_2} dt'_2 e^{-\tau(t_2-t'_2)} \sum_k \mathcal{R}_{jk}(t'_2) \left( \langle R_0(t_1) \rangle_{R_0,k}(t'_2) \right)_\eta
\]

\[
\langle R_0(t_1) R_1(t_2) \rangle_\eta = \left( \frac{k_B T}{k} \right) \int_0^{t_2} dt'_2 e^{\tau(t_2-t'_2)} \sum_k \mathcal{R}_{jk}(t'_2) \left[ \frac{k_B T}{\kappa} \delta_{jk} \left( e^{-\tau(t_1-t'_2)} - e^{-\Gamma(t_1-t'_2)} \right) + k_B T \Delta \Gamma e^{-\tau(t_1-t'_2)} \right] \int_0^{\min(t_1,t'_2)} dt'' e^{2 \Delta \Gamma t''} \sum_k \mathcal{R}_{jk}(t'_2) R_{0,k}(t''') \]

\[
\langle R_0(t_1) R_1(t_2) \rangle_\eta = \left( \frac{k_B T}{k} \right) e^{-\tau(t_1+t_2)} \int_0^{t_2} dt'_2 e^{\tau(t_1-t'_2)} \left( e^{\tau t'_2} - e^{-\tau t'_2} \right) + k_B T \Delta \Gamma e^{-\tau(t_1+t_2)} \int_0^{\min(t_1,t'_2)} dt'' e^{2 \Delta \Gamma t''} \sum_k \mathcal{R}_{jk}(t'_2) R_{0,k}(t''') \right)
\]

\[
(D.1)
\]

\[
\langle R_0(t_1) R_1(t_2) \rangle_{\eta,\theta} = \left( \frac{k_B T}{k} \right) e^{-\tau(t_1+t_2)} \int_0^{t_2} dt'_2 e^{\tau t'_2} \left( \langle R_0(t_1) \rangle_{R_0,k}(t'_2) \right)_\theta \left( e^{\tau t'_2} - e^{-\tau t'_2} \right) + k_B T \Delta \Gamma e^{-\tau(t_1+t_2)} \int_0^{\min(t_1,t'_2)} dt'' e^{2 \Delta \Gamma t''} \sum_k \langle R_{jk}(t'_2) \rangle R_{0,k}(t''') \right)_\theta
\]

\[
(D.2)
\]

\[
\langle \chi_0(t_1) \chi_1(t_2) \rangle_{\eta,\theta} = \left( \frac{k_B T}{k} \right) \cos 2 \theta_0 e^{-\tau(t_1+t_2)} \int_0^{t_2} dt'_2 \left( e^{2 \Delta \Gamma t'_2} - e^{-4 D \Delta t'_2} \right) + k_B T \Delta \Gamma e^{-\tau(t_1+t_2)} \int_0^{t_2} dt'_2 \int_0^{t_2} dt'' e^{2 \Delta \Gamma t'} \sum_k \left( \langle R_{jk}(t'_2) \rangle R_{0,k}(t''') \right)_\theta
\]

\[
(D.3)
\]
\[ \langle x_0(t_1) x_1(t_2) \rangle_{\eta, \theta} = \left( \frac{k_B T}{\kappa} \right) \cos 2\theta_0 \ e^{-\Delta T} \left( \frac{e^{(\kappa-4D_\theta)t_2} - e^{-\Delta T}}{2\kappa \Delta - 4D_\theta} - \frac{e^{-\Delta T} - e^{-(4D_\theta+\kappa)t_2}}{4D_\theta} \right) \]

\[ + \left( \frac{k_B T}{\kappa} \right) \left( \frac{\Delta T}{2\kappa} \right) e^{-\Delta T} \left[ \frac{e^{\Delta T} - e^{-\Delta T}}{2\kappa T + 4D_\theta} - \frac{2\alpha T}{\kappa T} \frac{e^{\Delta T} - e^{-\Delta (2\alpha T+4D_\theta)t_2}}{\kappa T + 4D_\theta} \right] \]

(D.4)

\[ \langle R_{0,i}(t) R_{2,j}(t) \rangle_{\eta, \theta} = \int_0^t dt' e^{-\alpha T(t-t')} \sum_k R_{ij}(t') R_{1,k}(t') \left( \frac{k_B T}{\kappa} \right) e^{-\alpha T(t-t')} \int_0^t dt' \ e^{\alpha T(t-t')} \sum_k R_{jk}(t') R_{1,k}(t') \eta, \theta \]

(D.5)

\[ \langle R_{0,i}(t) R_{1,k}(t') \rangle_{\eta, \theta} = \left( \frac{k_B T}{\kappa} \right) e^{-\alpha T(t-t')} \int_0^t dt' \int_0^t dt' \ e^{-\alpha T(t-t')} \ e^{\alpha T(t-t')} \left( \frac{k_B T}{\kappa} \right) e^{-\alpha T(t-t')} \int_0^t dt' \ e^{\alpha T(t-t')} \int_0^t dt' \ e^{2\alpha T(t-t')} \sum_k R_{1,k}(t') R_{1,k}(t') \eta, \theta \]

(D.6)

Neglecting the second term in Eq. (D.6), we have

\[ \langle R_{0,i}(t) R_{2,j}(t) \rangle_{\eta, \theta} = \left( \frac{k_B T}{\kappa} \right) \int_0^t dt' \ e^{-\alpha T(t-t')} \sum_k R_{jk}(t') \left( \frac{k_B T}{\kappa} \right) e^{-\alpha T(t-t')} \int_0^t dt' \ e^{\alpha T(t-t')} \sum_k R_{jk}(t') R_{1,k}(t') \eta, \theta \]

(D.7)

For the mean-square displacement along x and y direction, setting \( j = i \) and using Eq. (B.5) we get

\[ \langle x_0(t_1) x_2(t_2) \rangle_{\eta, \theta} = \langle y_0(t_1) y_2(t_2) \rangle_{\eta, \theta} = \left( \frac{k_B T}{\kappa} \right) e^{-2\alpha T} \int_0^t dt' \int_0^t dt' \left( e^{2\alpha T} - 1 \right) \cos 2 \left( \theta(t') - \theta(t'') \right) \]

(D.8)