Brownian motion of a torus

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Abstract

A torus is one of the few axisymmetric bodies (solids of revolution), which on account of its peculiar shape, shows interesting characteristics during its motion through a fluid. Specifically, it leads to coupling of the translational and rotational degrees of freedom, resulting in non-trivial contributions of the “cross terms” to the diffusion coefficient and relaxation times. The study of Brownian motion of a torus is important on two counts: most stiff or semiflexible polymers like the DNA miniplasmids can be modeled as tori. Secondly, a torus happens to be one of the simplest objects which can explain a self-propelled microorganism. The length scales in both these problems make the study of Brownian motion important. We calculate in this paper, the translational and rotational diffusion coefficient of a torus and show that the coupling contributes to these coefficients, the effect being a function of the slenderness ratio, $\epsilon$. The effect of the coupling is found to be reinforcing, although moderate. The coupling surprisingly has no effect on the autocorrelation function of the twirling degree of freedom $\psi$, when subject to a harmonic potential. The effect of diffusion on a toroidal swimmer is also calculated and results show that such a swimmer can undergo substantial diffusion before a directional (imposed or self-propelled) motion takes over.

Keywords: Brownian motion; Mobility matrix; Langevin equation; Diffusion coefficient

1. Introduction

Colloidal or submicron particles when dispersed in a solvent show continuous, incessant random movement called Brownian motion even in the absence of any external driving force. Brownian motion is important in colloidal systems like polymers and forms the basis of many instruments like dynamic light scattering, light spectroscopy, etc. [1–3]. The other instance where it is important is the motility of microorganisms. The motion of microorganisms at low Reynolds number is an arduous task, and substantial energy is expended by these organisms in overcoming Brownian motion, caused by thermal fluctuations on account of their small sizes [4]. The accurate estimation of diffusion coefficients of such objects is therefore important.

Interestingly, the Brownian motion of a torus has not been addressed in the literature, although, the problem is significant on at least two counts: firstly, most stiff or semi-flexible ring polymers can be modeled as tori. Secondly, a torus happens to be one of the simplest objects which can explain a self-propelled microorganism. The length scales in both these problems make the study of Brownian motion important.

Brenner [5,6], in a seminal work, demonstrated that in addition to the classical translational and rotational diffusion coefficients, which characterize the intensity of Brownian motion, for certain special objects possessing screw-like geometric properties, a third independent diffusion coefficient exists. The occurrence of this diffusivity is attributed to hydrodynamic coupling between the different degrees of freedom. Similar consequences of hydrodynamic coupling and its effects are observed for heliocoidally isotropic particles [5]. Brenner [6] extended this analysis to motions of rigid particles of arbitrary shape. Such a diffusivity does not exist for particles devoid of screw-like geometric asymmetry. However, there does exist coupling between the translational and rotational diffusion coefficients in objects like ellipsoids. Recently, the first experimental demonstration of Perrin’s [7] analysis of the diffusion of an ellipsoid lead to verification of the diffusion coefficients suggested decades ago [8]. The crossover from anisotropic to isotropic diffusion was experimentally shown to be in agreement with theoretical results. Similar effect is also exhibited in few other instances. The coupled translational and rotational diffusion for a
three-sphere and multi-sphere assembly, studied using Brownian dynamic simulation, suggests that the exact Brownian motion is described if the coupled translational and rotational degrees of freedom are accurately modeled and that the extension of such models to N-sphere system would be the correct way to simulate rigid macromolecules. Similar effect is also observed in the Brownian motion of a structured particle near a structured wall and the friction tensors for such systems have been derived in all the above works, the coupling can be attributed to complicated shapes or to the presence of an external boundary near the object.

A torus is a special geometry which, although not screw-like or connected spheres, on account of its internal degrees of freedom, can undergo translation due to hydrodynamic coupling with the rotational mode. This was first realized by Taylor and later suggested by by Purcell as possible mechanism of locomotion of microorganisms. Recently we demonstrated this fact quantitatively and calculated the propulsion velocity of a force free torus which rotates because of internal degrees of freedom. Thus a torus is probably one of the very few simple geometries which can show coupling of different degrees of freedom, leading to interesting flow characteristics.

Hydrodynamic coupling between different degrees of freedom and its effect on the diffusivity is also important in macromolecules and bio-polymers. The popular Zimm model in polymer dynamics, takes into account, the hydrodynamic interaction between different monomer beads in a macromolecule. This is known to lead to substantially different results from the predictions of the Rouse model, which does not account for these interactions. The resistance and mobility matrices in these systems are much more complex because of the infinite degrees of freedom and the dependence on the positions of each of these beads. However, macromolecules which show substantial stiffness like the TMV (tobacco mosaic virus) are often modeled as rigid cylindrical rods and the effect of coupled diffusion in such rod like polymers and objects is known since long time. Similar to the TMV, modeled as cylindrical rods, DNA miniplasmids and stiff or semiflexible closed ring polymers can be approximated as a closed torus, and so the understanding of the diffusion coefficient of such objects is crucial to determine their dynamical properties. The results, derived in this paper, should be useful as a starting point for calculation of the dynamics of a dilute solution of DNA miniplasmid (stiff polymer rings).

This paper addresses the issue of Brownian motion of a torus. Two coupled modes are considered, namely unidirectional translation and the twirling mode, which is also called the rotational mode in this work. Such a motion is possible in say a torus (e.g. a miniplasmid) moving along a track. The “translational diffusion coefficient” and the “rotational (twirling) diffusion coefficient” are calculated in the decoupled regime. The effect of hydrodynamic coupling of these two modes on the diffusion coefficients is determined next. The relaxation time for the rotational mode in the presence of a harmonic potential is estimated. Finally, the effect of Brownian motion on the motility of toroidal simmers, and the typical time scales at which the diffusion coefficient can be overcome by such organisms, is analyzed.

2. The theory of generalized Langevin equation

We consider here the Brownian motion of a torus which has two degrees of freedom, namely the twirling mode (rotation) and the translational mode. This situation arises, for example, in a torus translating along a track, in which only unidirectional translation and twirling modes are allowed, while the out of plane rotation and translation in other directions are prevented by topological constraints. The analysis is also applicable when the out of plane (containing the torus) diffusion is much smaller compared to the translational and twirling mode. The twirling mode is a result of the internal degree of freedom of a torus, which is one of the very few objects with a simple geometry, that allows internal motion. This special property was used to demonstrate a nano-machine which can self-propel itself under carefully selected parameters. It was shown that a miniplasmid (which has a geometry like a torus) can be made to twirl in only one direction by rectifying the twirling mode. This can be accomplished by imposing asymmetric non-equilibrium forcing which can be of thermal or potential type. The twirling torus then propels itself similar to the motion of in-viscid rotating smoke rings. The resistance matrix was calculated and it was also shown that the twirling of a force free torus can indeed lead to its translation. Thus the translational and the twirling modes in a torus are of great importance. The hydrodynamic coupling between the translation and the rotational modes, indicates that the rotational and the translational diffusion coefficients of such objects would also be coupled to the “cross effects”, and this forms the basis of this paper. Fig. 2 shows the schematic of a torus and the x and y modes considered in this work. The torus has a smaller radius and a larger radius a. 

Fig. 1. Configuration of the torus: (a) single twirling (rotating) torus and (b) a torus on a track.
The velocities are related to the Brownian variables as $V = \frac{dx}{dt}$ and the angular velocity $\Omega = \frac{d\psi}{dt}$. The mobility matrix $L$ which is the inverse of the symmetric resistance matrix $L^{-1}$ is also symmetric. $\eta_1, \eta_2, \eta_3$ are related to the coefficients $\zeta_1, \zeta_2, \zeta_3$.

2.1. Decoupled translation and rotation

Consider the hypothetical case of decoupled translation and rotation, such that the resistance and the mobility matrices are diagonal and the friction coefficients are given by $\zeta_1 = 1/\eta_1 = 8\pi^2 \mu a (\log(8a/b + 1/2))^{-1}$ and $\zeta_3 = 1/\eta_3 = 8\pi^2 \mu b^2$. In this case, the translational and rotational diffusion coefficients can be easily inferred as (refer Appendix A and B),

$$D_t = \frac{k_B T}{\zeta_1} = \frac{k_B T}{8\pi^2 \mu a} \left( \log \left( \frac{8a}{b} \right) + 1 \right),$$

and

$$D_r = \frac{k_B T}{\zeta_3} = \frac{k_B T}{8\pi^2 \mu ab^2}. \quad \text{(4)}$$

Note the similarity of the expression for $D_r$ for a torus to that of a sphere. We would show that this is indeed the leading order behavior of the diffusion coefficients in the slenderness ratio $\epsilon = b/a$ and that the hydrodynamic interactions result in an $O(\epsilon^3)$ correction. In the analysis of a single self-propelled torus (a DNA miniplasmid as a nanomachine) [13], such a decoupled Brownian motion was assumed and was justified by the slender torus ($\epsilon < 1$) assumption considered in that study.

2.2. Coupled translational and rotational mode

Consider the Brownian motion of the torus when the coupling between the translational and rotational mode cannot be ignored. Brenner [5,6] has discussed, in details, the restriction on the diffusion coefficients in the case of helicoidal and other geometries, in which the mobility matrix is dense with non-zero non-diagonal elements. Using the mobility relation (2) and considering conservative external forces and torques (expressed as gradients of a potential) and fluctuating forces and torques, the governing Langevin equations can be written as

$$\left( \begin{array}{c} \frac{dx}{dt} \\ \frac{d\psi}{dt} \end{array} \right) = \left( \begin{array}{cc} \eta_1 & \eta_2 \\ \eta_2 & \eta_3 \end{array} \right) \left( \begin{array}{c} \frac{\partial U}{\partial x} + f \\ \frac{\partial U}{\partial \psi} + \gamma \end{array} \right),$$

and

$$2b^2 a$$

where $f$ and $\gamma$ are fluctuating force and torque in the two variables $x$ and $\psi$ satisfying the fluctuation dissipation theorem given by

$$\langle f(t)f(t') \rangle = 2k_B T \zeta_1 \delta(t - t').$$

Fig. 2. Local coordinate system for the torus.
\[ \langle f(t) T(t') \rangle = 2k_B T \zeta_2 \delta(t - t'). \]  
\[ \langle T(t) f(t') \rangle = 2k_B T \zeta_3 \delta(t - t'). \]  
\[ \langle T(t) T(t') \rangle = 2k_B T \zeta_3 \delta(t - t'). \]

The solution of the Langevin equations is analyzed under two conditions: one in which the translational and rotational modes occur in the absence of any external potential, and the other, in which, the rotational mode is subjected to a potential. Interestingly, in the nanomachine considered in [13], it was shown that for a DNA miniplasmid, the potential can be internal and a function of the intrinsic properties of the DNA strand.

2.2.1. Free translational and rotational diffusion

We first consider the case in which there is no external/internal potential acting on the torus. The solution to the Langevin equations can then be written by integrating Eq. (1) as

\[ x(t) = x_0 + \int_0^t \eta_1 f(t') \, dt' + \int_0^t \eta_2 T(t') \, dt', \]
\[ \psi(t) = \psi_0 + \int_0^t \eta_2 f(t') \, dt' + \int_0^t \eta_3 T(t') \, dt', \]

where \( x_0 \) and \( \psi_0 \) are the ensemble averaged initial position of the torus. The reason why problems with hydrodynamic interactions become complicated (example, the Zimm model in polymer dynamics) is the dependence of the elements of the mobility matrix on the relative positions of the different fluctuating modes. However, the mobility matrix of a single torus is a constant (\( \eta_1, \eta_2, \eta_3 \) are constants), and so the calculations are expected to be less complicated.

The mean square translational and angular displacements are given by

\[ \langle (x(t) - x_0)^2 \rangle = 2D_r t \]
\[ = \eta_1^2 \int_0^t \int_0^t \, dt' \, \int_0^t \, dt'' \langle f(t') f(t'') \rangle + \eta_1 \eta_2 \int_0^t \, dt' \int_0^t \int_0^t \, dt'' \langle f(t') T(t'') \rangle + \eta_2 \int_0^t \, dt' \int_0^t \int_0^t \, dt'' \langle T(t') f(t'') \rangle + \eta_2^2 \int_0^t \, dt' \int_0^t \int_0^t \, dt'' \langle T(t') T(t'') \rangle. \]

Substituting the variances from the fluctuation dissipation theorem, we get,

\[ \langle (x(t) - x_0)^2 \rangle = 2D_r t = 2(\eta_1^2 \zeta_1 + 2\eta_1 \eta_2 \zeta_2 + \eta_2^2 \zeta_3)k_B T \zeta_r. \]

Similarly for the variable \( \psi \), we get,

\[ \langle (\psi(t) - \psi_0)^2 \rangle = 2D_r t \]
\[ = \eta_2^2 \int_0^t \, dt' \int_0^t \int_0^t \, dt'' \langle f(t') f(t'') \rangle + \eta_2 \eta_3 \int_0^t \, dt' \int_0^t \int_0^t \, dt'' \langle f(t') T(t'') \rangle + \eta_3 \eta_2 \int_0^t \, dt' \int_0^t \int_0^t \, dt'' \langle T(t') f(t'') \rangle + \eta_3^2 \int_0^t \, dt' \int_0^t \int_0^t \, dt'' \langle T(t') T(t'') \rangle. \]

Substituting the variances from the fluctuation dissipation theorem, we get,

\[ \langle (\psi(t) - \psi_0)^2 \rangle = 2D_r t = 2(\eta_2^2 \zeta_1 + 2\eta_2 \eta_3 \zeta_2 + \eta_3^2 \zeta_3)k_B T \zeta_r. \]

Note that for the case of a diagonal resistance matrix, \( \zeta_2 = \eta_2 = 0 \), we get \( D_t = k_B T / \zeta_1 \) and \( D_r = k_B T / \zeta_3 \). We express \( \zeta_1 = \zeta_1^0 \), \( \zeta_2 = \zeta_2^0 \) \( \zeta_3 = \zeta_3^0 \). Substituting in the expression for \( D_t \) and \( D_r \), and expanding the diffusion coefficients in \( b \), we get,

\[ D_t = k_B T \left( \frac{1}{\zeta_1^0} + b^2 \left( \frac{\zeta_2^0}{\zeta_1^0} \right)^2 \right). \]
\[ D_r = k_B T \left( \frac{1}{\zeta_3^0} + b^2 \left( \frac{\zeta_2^0}{\zeta_3^0} \right)^2 \right). \]

One can re-write the above as

\[ D_t = D_t^0 + b^2 \left( \frac{\zeta_2^0}{\zeta_1^0} \right)^2 \]
\[ D_r = D_r^0 + b^2 \left( \frac{\zeta_2^0}{\zeta_3^0} \right)^2. \]

Note that the coefficients are given by

\[ \zeta_1 = \zeta_1^0 = \frac{8\pi^2 \mu a}{\log(8/\epsilon) + (1/2)}, \]
\[ \zeta_2 = \zeta_2^0 = b^2 (4\pi \mu) \log(8/\epsilon) - (1/2), \]
\[ \zeta_3 = \zeta_3^0 = b^2 (8\pi^2 \mu a). \]

The appropriately non-dimensionalised diffusivities show that \( D_t = D_r \), and the equality is really a consequence of symmetric nature of the resistance and mobility matrices. When the expressions for \( \zeta_1^0, \zeta_2^0 \) and \( \zeta_3^0 \) are substituted, we get the correction to the diffusion coefficient in terms of the slenderness ratio \( \epsilon \) as

\[ D = D_t = D_r = 1 + \frac{c^2}{8} \left( \frac{1 - 2\log(8/\epsilon)^2}{1 + 2\log(8/\epsilon)} \right) + O(\epsilon^4). \]

The expressions for non-dimensional translational and rotational diffusion coefficient, correct to \( O(\epsilon^6) \), indicate that the enhancement of the diffusion coefficients because of hydrodynamic coupling, is the same for the two degrees of freedom in agreement with earlier results [5,6] and are functions only of the slenderness ratio \( \epsilon \). Fig. 3 shows the variation of the non-dimensional diffusion coefficients as a function of \( \epsilon \). The figure indicates an enhancement of around 10% for \( \epsilon = 0.5 \) and around 20% for \( \epsilon = 0.8 \).

The use of analytical expressions, derived for a slender torus, in extrapolating the calculation of diffusion coefficients to large enough \( \epsilon \), is justified by our recent study on the hydrodynamics of a rotating torus [20]. Fig. 4 compares the analytical and
numerical propulsion velocity (non dimensionalized by \( \omega a \)) of a force free torus, moving with a angular velocity \( \omega \). The numerical results were obtained by boundary integral calculations. The results show a surprisingly good match between the analytical and numerical values for slenderness ratio as high as \( \epsilon = 0.5 \).

2.2.2. Rotational diffusion under a potential

In a recent work [13], an exciting possibility was demonstrated for a miniplasmid, modeled as a torus, to run as a nanomachine using the ratchet effect. It was shown that an intrinsic potential can act on the twirling degree of freedom if the properties of the miniplasmid (base pair sequence) is suitably chosen. The Brownian fluctuations of the twirling mode can then be rectified to obtain uni-directional twirling which propels the ring in translational motion. It is therefore interesting, to study the effect of potential on the Brownian motion of the twirling mode and the resulting relaxation time and autocorrelation function. The translational mode is unaffected by this potential and is not considered here. The Langevin equation for the twirling mode is given by (refer Appendix C)

\[
\frac{d\psi}{dt} = -\eta_3 \frac{dU}{d\psi} + \eta_2 f + \eta_3 \mathcal{Y}.
\]  

(21)

The asymmetric potential considered in [13] is non-quadratic and so renders analytical solutions difficult. We consider instead, a quadratic potential \( U = (1/2)k\psi^2 \), such that the Langevin equation is given by

\[
\frac{d\psi}{dt} = -\eta_3 k\psi + \eta_2 f + \eta_3 \mathcal{Y}.
\]  

(22)

The relaxation time can be identified as \( \tau = 1/\eta_3 k \), where \( 1/\eta_3 = b^2 \zeta_3 \), and the solution to the Langevin equation can be written as

\[
\psi(t) = \frac{1}{\eta_2} \int_{-\infty}^{t'} \frac{d't}{t'} e^{-(t-t')/\tau} f(t') + \frac{1}{\eta_3} \int_{-\infty}^{t'} \frac{d't}{t'} e^{-(t-t')/\tau} \mathcal{Y}(t'),
\]

so that,

\[
\langle \psi(t)\psi(0) \rangle = \frac{1}{\eta_2^2} \int_{-\infty}^{t} \int_{-\infty}^{t'} \frac{d't}{t'} \frac{d't''}{t''} e^{-(t-t'')/\tau} \langle f(t')f(t'') \rangle + \frac{1}{\eta_3^2} \int_{-\infty}^{t} \int_{-\infty}^{t'} \frac{d't}{t'} \frac{d't''}{t''} e^{-(t-t'')/\tau} \langle \mathcal{Y}(t')\mathcal{Y}(t'') \rangle + \frac{2}{\eta_2\eta_3} \int_{-\infty}^{t} \int_{-\infty}^{t'} \frac{d't}{t'} \frac{d't''}{t''} e^{-(t-t'')/\tau} \langle f(t')\mathcal{Y}(t'') \rangle.
\]

(23)

Simplifying,

\[
\langle \psi(t)\psi(0) \rangle = \frac{2k_B T \zeta_1}{\eta_2} \int_{-\infty}^{0} \frac{d't}{t'} e^{-(t-2t'')/\tau} + \frac{2k_B T \zeta_3}{\eta_3} \int_{-\infty}^{0} \frac{d't}{t'} e^{-(t-2t'')/\tau} + \frac{4k_B T \zeta_2}{\eta_2\eta_3} \int_{-\infty}^{0} \frac{d't}{t'} e^{-(t-2t'')/\tau},
\]

(24)

such that,

\[
\langle \psi(t)\psi(0) \rangle = k_B T \zeta_1 \eta_2^2 \tau e^{-t/\tau} + k_B T \zeta_3 \eta_3^2 \tau e^{-t/\tau} + 2k_B T \zeta_2 \eta_2\eta_3 \tau e^{-t/\tau},
\]

(25)

and,

\[
\langle \psi(t)\psi(0) \rangle = \frac{k_B T}{k} e^{-t/\tau}.
\]

(26)

Interestingly, all the other terms cancel off to give \( k_B T/k \). Thus the amplitude of the angular displacement as well as the relaxation time, are unaffected by the hydrodynamic coupling between the two modes in the harmonic potential approximation.

Although the results are derived in the limiting case of a slender torus, the analysis is more general. Thus, the Brownian motion of any two degrees of freedom \((\xi_1, \xi_2)\) which are characterized by a mobility matrix \( \mathbf{L} \) and satisfying the Langevin
equation and Fluctuation dissipation defined respectively by Eqs. (1) and (2), and
\[
\begin{pmatrix}
\zeta_1 & \zeta_2 \\
\zeta_2 & \zeta_3
\end{pmatrix} = L^{-1},
\]
the scaled diffusion coefficients are given by
\[
\begin{align*}
\tilde{D}_1 &= \frac{D_1}{k_B T/\zeta_1} = \frac{\zeta_1 \zeta_3}{\zeta_1 \zeta_3 - \zeta_2^2}, \\
\tilde{D}_2 &= \frac{D_2}{k_B T/\zeta_1} = \frac{\zeta_1 \zeta_3}{\zeta_1 \zeta_3 - \zeta_2^2}.
\end{align*}
\]
It is also easy to show, that if \( \zeta_2 \) is subjected to a harmonic potential the auto-correlation in the variable \( \zeta_2 \) is given by
\[
\langle \zeta_2(t)\zeta_2(0) \rangle = \frac{k_B T}{k} e^{-t/\tau}.
\]

3. Effect of Brownian motion on the motion of a toroidal swimmer

Self propelled organisms have always interested researchers, and mechanisms like flagellar motors, screw-type motion, etc have been suggested [4]. In this pursuit, there have been several attempts to describe a low Reynolds number swimmer which has a simple geometry and an easy mechanism for self-propulsion. A rotating (twirling) torus [4,12] was suggested as the simplest example of a self-propelled micro-organism, although, apart from few works [13,24], there have been very few investigations on quantitative studies of such a swimmer. We consider here the effect of Brownian motion on the motion of such a self-propelled organism. Out of plane (of the torus) rotational diffusion is expected to substantially reduce the diffusion in such a swimmer. The typical relaxation time scale of out of plane rotational diffusion is of the order \( \eta R^2/(k_B T) \) (up to logarithmic corrections [25]) which for a ring with diameter \( d = 1.0 \mu m \) leads to 0.03 s, while the translational diffusion occurs over a time scale of 0.4 s. This means that a single twirling ring in solution will not perform any noticeable translational drift. A possible solution to the problem is to put the swimmer on a “track”, e.g. to thread it on a straight DNA chain. Such a swimmer, moving with a speed of \( u = 1 \mu m/s \) will then overcome dispersion due to translational diffusion after \( t = u^2/D = 1 \) s.

4. Conclusions

The translational and rotational diffusion coefficient of a torus is determined. In this work, we bring out the interesting fact that the rotational and translational diffusion coefficients of a torus are coupled, due to a dense mobility matrix, and the effect of the cross terms on the coefficients is reinforcing, although moderate. It is shown that the translational and the rotational modes are coupled and the coupling is a function of the slenderness ratio \( \epsilon \). The effect of the coupling on the resultant rotational and translational diffusion coefficients is calculated and it is found that for a torus with slenderness ratio of \( \epsilon = 0.5 \) the change in the diffusion coefficient is around 10% and the coupling effect is reinforcing. Interestingly, the correction to the non-dimensional translational and rotational diffusion coefficient, because of the hydrodynamic coupling, is the same. This may be attributed to the symmetric nature of the mobility matrix. The relaxation time is then estimated for a torus subjected to a harmonic potential in the rotational degree of freedom. It is found that the amplitude of the correlation is unaffected by the coupling. Finally the effect of diffusion on a toroidal swimmer is determined and results show that such a swimmer can undergo substantial diffusion before the free streaming velocity can take over. We believe that the analysis presented here should form the basis for a detailed investigation which takes into account the out of plane rotation of the torus and thereby affects the motion of an active particle. Moreover, the formalism introduced here for the calculation of diffusion of a torus, together with our recently suggested work [26] on the hydrodynamic interaction between two tori, should be the first step in the solution of Brownian dynamics of a dilute solution of DNA miniplasmids, especially in the Rouse like limit.

Appendix A. The Langevin equation for translational degree of freedom

The diffusion coefficient for a sphere undergoing 1 – \( D \) Brownian motion is derived here. The derivation can be found in several texts [22,1,2]. The x-directional force balance for a torus is given by
\[
m \frac{d^2 x}{dt^2} = -\zeta \frac{dx}{dt} - \frac{dU}{dx} + f(t).
\]
\[
(A.1)
\]
The term on the left-hand side is the acceleration of the Brownian particle of mass \( m \) and size \( a \), \( x \) is the position of the particle, the first term on the right-hand side is the Stokes drag, where \( \zeta = 6 \pi \mu a \), and \( \mu \) is the viscosity of the fluid. The second term is the force due to an external potential and \( f(t) \) is the stochastic force on the particle such that \( \langle f(t) \rangle = 0 \). This is understandable so because the average force does not have any directional bias. The random force is uncorrelated in time and is expressed as \( \langle f(t)f(t') \rangle = A \delta(t - t') \). Any physical quantity \( M \) is expressed as \( \langle M \rangle \) and is defined as the ensemble average over all possible realizations of the stochastic force. In the following analysis, we assume for simplicity that there is no external potential (and hence force) acting. The Langevin equation can be written in terms of velocity as
\[
m \frac{dv}{dt} = -\zeta v + f(t).
\]
\[
(A.2)
\]
This has a solution which is given by
\[
v(t) = v(0)e^{-\zeta/mt} + e^{-\zeta/mt} \int_0^t \frac{f(\tau)}{m} e^{\zeta/mt} d\tau,
\]
\[
(A.3)
\]
\[
\langle v(t)^2 \rangle = \langle v(0)^2 \rangle e^{-2\zeta/mt} + e^{-2\zeta/mt} \int_0^t d\tau_1 \int_0^\tau d\tau_2 \frac{\langle f(\tau_1) f(\tau_2) \rangle}{m^2} e^{\zeta/mt(\tau_1 + \tau_2)}.
\]
\[
(A.4)
As \( t \to \infty \), the system should reach equilibrium, and equipartition of energy should be satisfied which is mathematically expressed as \( \langle m(v(t)^2) \rangle / 2 = (k_B T/2) \).

\[
\langle v(t)^2 \rangle = A e^{-2\zeta/m} \int_0^t dt_1 e^{2\zeta/m(t_1)} = A e^{-2\zeta/m} m \frac{1}{2\zeta} e^{2\zeta/m(t)} \bigg|_{t=0} = \frac{A}{2m\zeta}. \tag{A.5}
\]

This gives the strength of the noise \( A = 2k_B T \zeta \), so that \( \langle f(t)f(t') \rangle = 2k_B T \zeta \delta(t-t') \). This is one form of the fluctuation dissipation theorem. Thus the strength of the noise is determined by the requirement that equipartition is satisfied at long times (Equilibrium). We can now calculate the dynamics of the Brownian particle. We note that in the inertial Langevin Eq. (B.1), the mass is much smaller so that the inertial term can be neglected. This is called as the over-damped Langevin equation, which is given by

\[
\frac{dx}{dt} = -\frac{dU}{dx} + f(t). \tag{A.6}
\]

In the absence of any external potential, we find that the position is given by

\[
x(t) = x_0 + \int_0^t \frac{f(t)}{\zeta} dt. \tag{A.7}
\]

The expression for \( x(t) \), shows that it is an integral (sum) of \( f(t) \), which are uncorrelated random forces. The average position is an ensemble average of the above equation is

\[
\langle x(t) \rangle = x_0 + \int_0^t \langle \frac{f(t)}{\zeta} \rangle dt = x_0, \tag{A.8}
\]

and for the Brownian particle the variance can be calculated as

\[
\langle (x(t) - x_0)^2 \rangle = \int_0^t dt_1 \int_0^t dt_2 \frac{\langle f(t_1)f(t_2) \rangle}{\zeta^2} = 2k_B T \zeta t = 2D_t t. \tag{A.9}
\]

Giving the Stokes Einstein relation for self diffusion coefficient. This can be easily checked for a Brownian sphere (\( \zeta = 6\pi \mu a \)) as \( D_t = k_B T/6\pi \mu a \).

Appendix B. The Langevin equation for twirling degree of freedom

The diffusion coefficient for a sphere undergoing rotational Brownian motion is derived here. The \( \psi \)-directional torque balance for a torus is given by

\[
mb^2 \frac{d^2 \psi}{dr^2} = -\zeta \frac{d\psi}{dr} - \frac{dU}{dx} + \Upsilon(t). \tag{B.1}
\]

The term on the left-hand side is the net torque acting on the Brownian torus of mass \( m \) and the outer and inner diameters, \( a \) and \( b \) respectively. \( \psi \) is the twirling angle about the centerline, the first term on the right-hand side is the Stokes torque, where \( \zeta = 6\pi \mu a \), and \( \mu \) is the viscosity of the fluid. The second term is the force due to an external potential and \( \Upsilon(t) \) is the stochastic torque on the particle such that \( \langle \Upsilon(t) \rangle = 0 \). This is understandably so because the average torque does not have any directional bias. The random torque is uncorrelated in time and is expressed as \( \langle \Upsilon(t) \Upsilon(t') \rangle = A \delta(t-t') \). In the following analysis, we assume for simplicity that there is no external potential (and hence force) acting. The Langevin equation can be written in terms of angular velocity \( \Omega = d\psi/dr \) as

\[
\frac{d\Omega}{dt} = -C \Omega + \frac{\Upsilon(t)}{D}, \tag{B.2}
\]

where \( C = \zeta/m b^2 \) and \( D = mb^2 \). This has a solution which is given by

\[
\Omega(t) = \Omega(0)e^{-Ct} + \frac{1}{D} \int_0^t \Upsilon(t) e^{Ct} dt, \tag{B.3}
\]

so that

\[
\langle \Omega(t)^2 \rangle = e^{-2Ct} \int_0^t dt_1 \int_0^t dt_2 \frac{\langle \Upsilon(t_1)\Upsilon(t_2) \rangle}{D^2} e^{C(t_1+t_2)}. \tag{B.4}
\]

As \( t \to \infty \), the system should reach equilibrium, and equipartition of energy should be satisfied which is mathematically expressed as \( (mb^2/2\Omega(t)^2) / 2 = (k_B T/2) \).

\[
\langle \Omega(t)^2 \rangle = A e^{-2Ct} \int_0^t dt_1 \int_0^t dt_2 \frac{\langle \Upsilon(t_1) \Upsilon(t_2) \rangle}{D^2} e^{C(t_1+t_2)} = \frac{A}{2D^2 C}. \tag{B.5}
\]

This gives the strength of the noise \( A = 2k_B T \zeta \), so that \( \langle \Upsilon(t) \Upsilon(t') \rangle = 2k_B T \zeta \delta(t-t') \). This is one form of the fluctuation dissipation theorem. Thus the strength of the noise is determined by the requirement that equipartition is satisfied at long times (Equilibrium). We can now calculate the dynamics of the Brownian particle. We note that in the inertial Langevin Eq. (B.1), the mass is much smaller so that the inertial term can be neglected. This is called as the over-damped Langevin equation, which is given by

\[
\frac{d\psi}{dt} = -\frac{dU}{dr} + \Upsilon(t). \tag{B.6}
\]

In the absence of any external potential, we find that the position is given by

\[
\psi(t) = \psi_0 + \int_0^t \frac{\Upsilon(t)}{\zeta} dt. \tag{B.7}
\]

The expression for \( \psi(t) \), shows that it is an integral (sum) of \( \Upsilon(t) \), which are uncorrelated random forces. The average position is an ensemble average of the above equation is

\[
\langle \psi(t) \rangle = \psi_0 + \int_0^t \langle \frac{\Upsilon(t)}{\zeta} \rangle dt = \psi_0. \tag{B.8}
\]

and for the Brownian particle the variance can be calculated as

\[
\langle (\psi(t) - \psi_0)^2 \rangle = \int_0^t dt_1 \int_0^t dt_2 \frac{\langle \Upsilon(t_1) \Upsilon(t_2) \rangle}{\zeta^2} = \frac{2k_B T}{\zeta} t = 2D_t t. \tag{B.9}
\]
This can be easily checked for the case of a rotating sphere, for which \( \zeta = 8\pi \mu a^3 \), as \( D_r = k_B T/(8\pi \mu a^3) \).

Appendix C. Brownian particle under a potential

The relaxation of the auto-correlation, for a sphere undergoing \( 1 - D \) Brownian motion under a harmonic potential, is derived here. The derivation can be found in several texts [22,1,2]. Consider a Brownian particle under potential \( U = (1/2)k x^2 \), the Langevin equation can be written as

\[
\frac{dx}{dt} = -kx + f(t),
\]

(C.1)

Solving this equation in a similar manner, we get the following results for a Brownian particle.

\[
x(t) = x_0 e^{-t/\tau} + \frac{1}{\zeta} \int_0^t e^{-(t-t')/\tau} f(t') dt',
\]

(C.2)

and the auto-correlation is given by

\[
\langle x(t)x(0) \rangle = k_B T \frac{e^{-t/\tau}}{k},
\]

(C.3)

where \( \tau = \zeta/k \), and indicates an exponential decay of the correlation function, with a time constant which depends on the spring constant.

References