On Brownian Motion in a Fluid with a Plane Boundary

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0.1 Introduction

Brownian motion was discovered by the botanist Robert Brown in 1827. While studying pollen grains suspended in water under a microscope, Brown observed that particles ejected from the pollen grains executed a jittery motion. After he replaced the pollen grains by inorganic matter, he was able to rule out that the motion was life-related, although its origin was yet to be explained.

In 1905, Einstein[5] explained Brownian motion as the result of bombardment of fluid molecules on the suspended particle. There are two main parts to his paper. First, he finds the following relation of the diffusion coefficient to other physical quantities:

$$D = \frac{k_B T}{6\pi\eta a} = \frac{k_B T}{\zeta}\,,$$

where D is diffusion coefficient, k_B is Boltzmann's constant, a is radius of the particle, η is the dynamic viscosity, and $\zeta = 6\pi\eta a$ is the Stokes drag, which was first calculated by Stokes. Then, Einstein related the diffusion coefficient to the mean square displacement of the particle, $\langle x^2\rangle = 2Dt$, where D is diffusion coefficient. Specifically, Einstein found that the density of the Brownian particles $f(x,t)$ satisfies the heat equation

$$\frac{\partial f}{\partial t} = D\frac{\partial^2 f}{\partial x^2}\,,$$

and after solving the heat equation, he got that the mean square displacement is proportional to time. However, Einstein also noticed when time is short (in ballistic time regime), the mean square displacement should be different, since during very short times individual particles become significant.

In 1908, Langevin[12] used another point of view, he assumed the particles satisfy the Newtonian equation:

$$m\frac{d\mathbf{v}(t)}{dt} = -\zeta\mathbf{v}(t) + \mathbf{X}\,,$$

where $\zeta = 6\pi\mu a$ is the Stokes drag and $\mathbf{X}$ is a random force describing the bombardment by fluid particles. Using this equation, with the assumption that $\langle\mathbf{X}(t)\mathbf{x}(t)\rangle = 0$, $\mathbf{x}(t)$ being the position of particle, Langevin was able to derive the same relation that Einstein derived, i.e.

$$\langle x^2\rangle = \frac{2k_B T}{\zeta}t\,.$$

This is valid when t is large, but when t is small, a particle's inertia becomes significant. In this inertia dominated regime, termed the ballistic regime, the particle's motion is highly correlated. Langevin's approach also applies to the ballistic regime. In the classical Langevin theory, it is assumed that the autocorrelation function of the random force satisfies

$$\langle X(t_1)X(t_2)\rangle = 2\zeta k_B T\delta(t_1 - t_2)\,.$$

This means that the random force acting on the Brownian particle is memoryless. From the Langevin equation, it can be shown that the velocity autocorrelation function has exponentially decay,

$$\Phi(t) := \langle v(t_0)v(t_0+t)\rangle = \langle v(t_0)^2\rangle e^{-\frac{\zeta t}{m}}\,.$$

However, in the 1960s, the famous "tails" of the velocity autocorrelation function were discovered[14]. It was experimentally obeserved that the velocity autocorrelation does not exponential decay, but rather has an algebraic decay. Then, Langevin's approach was generalized to give a more accurate description. Instead of using the δ correlated random force, it was figured out that one should include the memory effect of the fluid.

The long time behavior of velocity autocorrelation function (VACF) and mean square displacement (MSD) have been observed by both experiment and computer simulation for many years. The ballistic regime (where the inertia of Brownian particle will dominate) is hard to observe in experiments, since it requires the position detector equipment to have extraordinary spatial and temporal resolution. Only very recently has Brownian motion in the ballistic regime been observed[9], and the experiment showed excellent agreement with theoretical predictions[4].

There are two main steps that have been taken to derive the VACF and MSD. A deterministic part where one calculates the response function $\zeta(\omega)$ corresponding to a specific frequency. This step is done by solving the linearized Navier-Stokes equations analytically. The second step is a statistical part, where one uses the fluctuation-dissipation theorem to relate correlation functions to the corresponding response functions. The fluctuation-dissipation theorem is a main tool in statistical mechanics to predict behavior of non-equilibrium thermodynamic systems, as widely observed in nature: for example, the Brownian motion seen in the irregular oscillation of a suspended mirror, the thermal noise in resistor, etc.

An important theorem of Nyquist was the first theorem in this area to be proved (to my knowledge). The idea was discovered by J. Johnson and then proved by H. Nyquist. For any network, the square of the voltage during the frequency range $(\nu, \nu + d\nu)$ is given by:

$$E_\nu^2 \, d\nu = 4R_\nu k_B T \, d\nu$$

where E_ν is the electromotive force and R_ν is the real part of the impedance of the network. Using this result, Nyquist proved the following formula that was given in Johnson's paper:

$$I^2 = \frac{2}{\pi} k_B T \int_0^\infty R(\omega)|Y(\omega)| \, d\omega$$

where $Y(\omega)$ is the transfer admittance of any network from the member in which electromotive force in question originates to a member in which the resulting current is measured.

For Brownian motion, the random impact of surrounding molecules has two kinds of effects: first, the molecules act as a random force and second, they give rise to the frictional force. This means the frictional force and random force must be related. This is the essence of the so-called fluctuation-dissipation theorem, which in formulas is given by

$$\zeta(\omega) = \frac{1}{k_B T} \int_0^\infty \langle X(t_0) X(t_0 + t) \rangle e^{i\omega t} \, dt \tag{0.1.1}$$

$$\mu(\omega) = \frac{1}{k_B T} \int_0^\infty \langle v(t_0) v(t_0 + t) \rangle e^{i\omega t} \, dt \, , \tag{0.1.2}$$

where $\zeta(\omega)$ is the response function or friction constant for particular fluid system and $\mu(\omega)$, the admittance, is given by

$$\mu(\omega) = \frac{1}{\zeta(\omega) - im\omega} \, .$$

If we let $s = -i\omega$, then above formulas are just Laplace transformations. Thus, if we want to calculate the autocorrelation function for Brownian particles, we can first calculate the friction constant of the corresponding system, and then use the fluctuation-dissipation theorem to get the final result.

There are several papers[7, 11] that describe the fluctuation-dissipation theorem. Given this theorem, we only need to calculate the response function by solving Navier-Stokes equations. Many people contributed to this area to develop a general theory. The case where Brownian particles are in a viscous, compressible fluid filling the whole of space $\mathbb{R}^3$ has been thoroughly solved[3],[17]. In [4] and [15], the asymptotic behavior of the velocity autocorrelation function(VACF), mean square displacement(MSD), etc. up to higher orders have also been calculated. In this paper, we consider the effect of a plane boundary, i.e., where the fluid occupies only half space, and compute the velocity autocorrelation function for this case.

0.2 Behavior of Brownian particles in different time regimes

The behavior of the VACF and the MSD are different in the different time regimes. On the very short time scale, the inertia of the Brownian particle dominates, while on the longer time scale, the hydrodynamical memory effect plays an important role. There are several characteristic times: $t_p = m/\zeta$, $t_\nu = a^2/\nu$, and $t_c = a/c$, where m is the mass, a is the radius of the Brownian particle, $\zeta = 6\pi\eta a$ is again the Stokes drag, and c is the speed of sound in the fluid. The case $t < t_p$ is the ballistic regime, where as noted above, the inertia of the particle is significant. The case $t \gg t_p$ is the diffusive time regime. When $t \simeq t_\nu$ hydrodynamical effects need to be taken into account, and when $t < t_c$ one must consider compressibility of the fluid. The two graphs of Figs. 1 and 2 show experiment data from [9]. The first graph is the MSD vs. time, while the second one is the VACF vs. time. Evidently, the experiment data fits the theory[4] very well. It worth noting that the model of [4] only considers incompressible fluid dynamics. So if we want to test the behavior of Brownian particle into the compressible regime, we need to both enhance the accuracy of the detector into the nanosecond regime but alter the theory as well.

The mean square displacement in the ballistic regime is given by $\langle x^2 \rangle = t^2 k_B T/m^*$. Here $m^* = m + \frac{1}{2}M$ is the virtual mass of the body and $M = \rho_f 4\pi a^3/3$ is the mass of the displaced fluid. In ballistic time regime, the velocity autocorrelation function is given by $\langle v^2 \rangle = k_B T/m^*$, a result that seems inconsistent with the equipartition theorem $m\langle v^2 \rangle = k_B T$ when $t \to 0$. This discrepancy is explained as the effect of compressibility. When t is smaller than the characteristic time $t \leq t_c = a/c$, the fluid cannot be regarded as incompressible and, in this case, the particle is decoupled from fluid and the effective mass is m. In Zwanzig and Bixon's paper[18], they describe the decrease from $k_B T/m$ to $k_B T/m^*$.

0.3 Brownian Motion in whole space $\mathbb{R}^3$

We consider a spherical particle oscillating in a fluid with velocity $\mathbf{u}_\omega = \mathbf{u}e^{-i\omega t}$. To find the mean square displacement and autocorrelation function, we first to calculate the force $\mathbf{F}(\omega)$

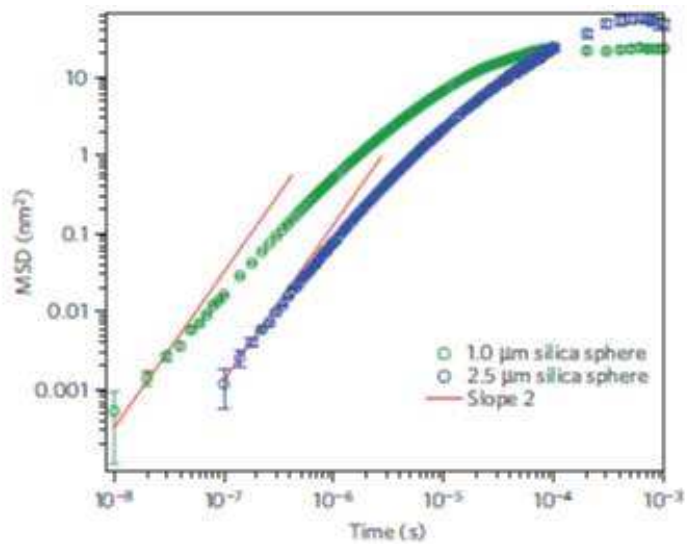

Figure 1: The mean square displacement (MSD) vs. time from the experiments of Ref. [9]

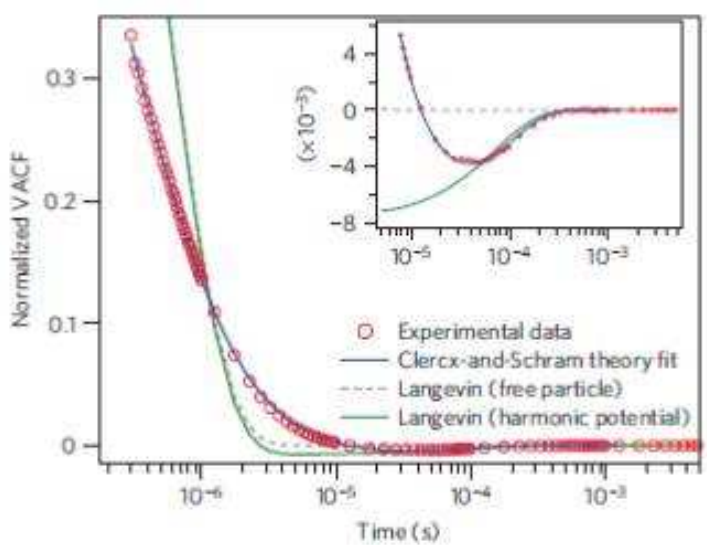

Figure 2: The velocity autocorrellation function (VACF) vs. time from the experiments of Ref. [9]

of the fluid acting on the sphere as $\mathbf{F}(\omega) = -\zeta(\omega)\mathbf{u}_\omega$. This is done by solving the linearized Navier-Stokes equations. After we obtain the solution, we use the fluctuation-dissipation theorem to find the MSD and the VACF of the particle. We first consider the case where the fluid fills all of $\mathbb{R}^3$, and then use the result to find an approximate solution when the fluid is bounded by a plane and occupies half space.

To solve the NS equations in domain $\mathbb{R}^3$, we can fix the sphere to the origin for simplicity. Then, in this frame, the velocity field of the fluid $\tilde{\mathbf{v}}$ changes with time, and satisfies the non-slip boundary condition at the surface of the sphere, i.e., $\tilde{\mathbf{v}} = 0$ on the sphere and $\tilde{\mathbf{v}} = -\mathbf{u}e^{-i\omega t}$ at infinity. Next we decompose the velocity field as $\tilde{\mathbf{v}} = \mathbf{v} - \mathbf{u}e^{-i\omega t}$, where $\mathbf{v}$ does not oscillate with time, and apply the boundary condition $\mathbf{v} = \mathbf{u}e^{-i\omega t}$ at the sphere boundary (non-slip) and $\mathbf{v} = \mathbf{u}e^{-i\omega t}$ at infinity. For fluid motion $\mathbf{v}$ satisfies the following Navier-Stokes (NS) equations:

$$\rho\left(\frac{\partial \mathbf{v}}{\partial t} + (\mathbf{v}\cdot\nabla)\mathbf{v}\right) = -\nabla p + \eta\nabla^2\mathbf{v} + \left(\frac{\eta}{3} + \mu\right)\nabla(\nabla\cdot\mathbf{v}) \tag{0.3.1}$$

$$\frac{\partial \rho}{\partial t} + \nabla\cdot(\rho\mathbf{v}) = 0\,, \tag{0.3.2}$$

with boundary condition $\mathbf{v} = \mathbf{u}e^{-i\omega t}$ at the sphere and $\mathbf{v} = 0$ at infinity. For Brownian motion, the Reynolds is number is very small, so we can consider the linearized NS equations instead

$$\rho_0\frac{\partial \mathbf{v}}{\partial t} = -\nabla p + \eta\nabla^2\mathbf{v} + \left(\frac{\eta}{3} + \mu\right)\nabla(\nabla\cdot\mathbf{v}) \tag{0.3.3}$$

$$\frac{\partial \rho}{\partial t} = -\rho_0\nabla\cdot\mathbf{v}\,, \tag{0.3.4}$$

with the same boundary conditions as above. After solving these equations, we can calculate the drag of the fluid acting on the sphere for arbitrary motion by Fourier decomposing as

$$\mathbf{v}(t) = \int_{-\infty}^{\infty}\mathbf{u}_\omega e^{-i\omega t}\,d\omega \qquad \text{and} \qquad \mathbf{F}(t) = \int_{-\infty}^{\infty}\mathbf{F}_\omega e^{-i\omega t}\,d\omega\,.$$

Because of the linearity assumption, the Fourier component of force is proportional to Fourier component of velocity, $\mathbf{F}_\omega = -\zeta(\omega)\mathbf{u}_\omega$. This means we can find the drag for arbitrary motion if we know each Fourier component.

Now suppose $\mathbf{v}(x,y,z,t) = \mathbf{v}_\omega(x,y,z)e^{-i\omega t}$. To avoid clutter in what follows, we denote $\mathbf{v}_\omega$ by $\mathbf{v}$, and similarly for other quantities. Thus we have

$$-i\omega\rho_0\mathbf{v} = -\nabla p + \eta\nabla^2\mathbf{v} + (\frac{1}{3}\eta + \mu)\nabla(\nabla\cdot\mathbf{v}) \tag{0.3.5}$$

$$-i\omega\rho = -\rho_0\nabla\cdot\mathbf{v}\,, \tag{0.3.6}$$

with the pressure and density related by $\nabla P = C^2\nabla\rho$. Combining (0.3.5) and (0.3.6) gives

$$\omega^2\mathbf{v} + C_l^2\nabla\nabla\cdot\mathbf{v} - C_t^2\nabla\times\nabla\times\mathbf{v} = 0\,, \tag{0.3.7}$$

where $C_l^2 = C^2 - i\omega\nu_l$, $C_t^2 = -i\omega\nu_t$, and $\nu_t = \eta/\rho_0$, $\nu_l = (4\eta/3 + \mu)/\rho_0$. The boundary conditions are now given by $\mathbf{v} = \mathbf{u}$ at the sphere and $\mathbf{v} = \mathbf{0}$ at infinity, where $\mathbf{u}e^{-i\omega t}$ is the

velocity of the sphere in lab frame. Next we decompose as $\mathbf{v} = \nabla\phi + \nabla \times \mathbf{A}$, and obtain the following equations:

$$\nabla^2\phi + \beta^2\phi = 0 \qquad \text{and} \qquad \nabla \times \nabla \times \mathbf{A} - \alpha^2\mathbf{A} = 0\,, \tag{0.3.8}$$

where $\alpha^2 = i\omega\rho_0/\eta$, $\beta^2 = \omega^2/C_l^2$, and boundary condition $\nabla\phi + \nabla \times \mathbf{A} = \mathbf{u}$ on the sphere. The exact solution of this problem is given in [3]:

$$v_r = \left[2A\left(-\frac{1}{r^3} + \frac{i\alpha}{r^2}\right)e^{i\alpha r} + B\left(-\frac{2}{r^3} + \frac{2i\beta}{r^2} + \frac{\beta^2}{r}\right)e^{i\beta r}\right]u\cos\theta \tag{0.3.9}$$

$$v_\theta = \left[A\left(-\frac{1}{r^3} + \frac{i\alpha}{r^2} + \frac{\alpha^2}{r}\right)e^{i\alpha r} + B\left(-\frac{1}{r^3} + \frac{i\beta}{r^2}\right)e^{i\beta r}\right]u\sin\theta \tag{0.3.10}$$

where $x = i\alpha a$, $y = i\beta a$, $\Delta = 2x^2(3 - 3y + y^2) + y^2(3 - 3x + x^2)$,

$$P = \frac{3}{\Delta}(3 - 3y + y^2)\,, \qquad Q = -\frac{3}{\Delta}(3 - 3x + x^2)$$

and

$$A = Pa^3e^{-i\alpha a}\,, \qquad B = Qa^3e^{-i\beta a}\,.$$

The force of fluid acting on the sphere is given by integrating as follows:

$$\begin{aligned}
\mathbf{F} &= \oint da\big[-p\cos\theta + 2\eta e_{rr}\cos\theta - 2\eta e_{r\theta}\sin\theta + (\mu - 2\eta/3)\,(\nabla\cdot\mathbf{v})\cos\theta\big] \\
&= \oint da\big[(\mu - 2\eta/3 + iC^2\rho_0/\omega)(\nabla\cdot\mathbf{v})\cos\theta + 2\eta e_{rr}\cos\theta - 2\eta e_{r\theta}\sin\theta\big] \\
&= 4\pi\eta a x^2\mathbf{u}\big[(1-y)Q + 2(x-1)P\big]/3\,.
\end{aligned}$$

0.4 Brownian Motion in the Half Space $\mathbb{R}^+ \times \mathbb{R}^2$

Now we suppose the sphere is moving in a fluid that occupies a region bounded by a plane. The perpendicular distance from the center of the sphere to the plane is given by l and the radius of the sphere is given by a. We assume the velocity of sphere is given by $\mathbf{u}e^{-i\omega t}$, as before, and again use the frame of sphere instead of the lab frame, i.e., we let sphere be fixed and the fluid have velocity $\tilde{\mathbf{v}}$. Then again we decompose as $\tilde{\mathbf{v}} = \mathbf{v} - \mathbf{u}e^{-i\omega t}$. The linearized Navier-Stokes equations are the same as Eqs. (0.3.3) and (0.3.4) that we used for the $\mathbb{R}^3$ case, with boundary condition $\mathbf{v} = \mathbf{u}e^{-i\omega t}$ at the sphere and $\mathbf{v} = 0$ at the plane and at infinity.

If the sphere oscillates in an arbitrary direction, we lose symmetry, and the problem becomes hard to solve. So we first consider the case where sphere oscillates perpendicular to the plane. It seems that the most appropriate coordinate system to use to solve the above equations with the sphere-plane boundary conditions is the bipolar coordinate system. Actually, Brenner and several other authors [10],[2],[8] used bipolar coordinate to solve for the drag force when a sphere is approaching a plane. For their problem this was equivalent to solving a Laplace-type equation in bipolar coordinates. In our case, for simplicity, we

consider an incompressible fluid, so we can use a Stokes stream function Ψ. Defining the Laplace-type operator L^2 as

$$L^2 = \frac{\sin^2\theta}{r}\left\{\frac{\partial}{\partial\xi}\left(\frac{1}{r}\frac{\partial}{\partial\xi}\right) + \frac{\partial}{\partial\theta}\left(\frac{1}{r}\frac{\partial}{\partial\theta}\right)\right\},$$

where (ξ, θ) are the bipolar coordinates with

$$r := \frac{c\sin\theta}{\cosh\xi - \cos\theta},$$

and $(0, \pm c)$ being the foci of the bipolar coordinate system. For our problem the stream function satisfies

$$L^4\Psi + \alpha^2 L^2\Psi = 0,$$

where $\alpha^2 = i\omega\rho_0/\eta$. Defining $\Pi = L^2\Psi$, we see Π satisfies a Helmholtz equation of the form

$$L^2\Pi + \alpha^2\Pi = 0.$$

If we let $\Pi = f \cdot \sqrt{r}$, then we can simplify this equation as follows:

$$\frac{\partial^2 f}{\partial\xi^2} + \frac{\partial^2 f}{\partial\theta^2} - \frac{3}{4\sin^2\theta} f + \frac{\alpha^2 r^2}{\sin^2\theta} f = 0.$$

Unfortunately, in bipolar coordinate this kind of Helmholtz equation is not separable[13]. So instead, we resort to approximation.

Our approximation is based on the image method[8]. We first use this method to obtain an approximate solution and then use the fluctuation-dissipation theorem to calculate the VACF for the Brownian particle. As we will see later on, we don't need to restrict to the incompressible case or to perpendicular oscillation. However, the image method of approximation makes the velocity field only satisfy the Neumann boundary condition at the plane. So we have to assume slip boundary condition on the plane instead of the more physical no-slip boundary condition. Using the image method, we can find approximate solution up to some order of $\frac{a}{l}$, which will be a good approximation if the distance of the sphere to the plane l is much larger than radius of the sphere a. Suppose the vector field that satisfies equations (0.3.9) and (0.3.10) is $\mathbf{v}_1$, and $\mathbf{v}_2$ is the image velocity field of $\mathbf{v}_1$ obtained by reflecting through the plane (imagining the plane as a mirror). Then, suppose $\mathbf{v}_3$ is the velocity field satisfy $\mathbf{v}_3 = -\mathbf{v}_2$ at the sphere and vanishing at infinity, $\mathbf{v}_4$ is image velocity field of $\mathbf{v}_3$, etc. We suppose that $\mathbf{v} = \mathbf{v}_1 + \mathbf{v}_2 + \ldots$ would be the actual solution (with Neumann boundary condition). However, even with this assumption the $\mathbf{v}_i, i = 3, 5, \ldots$ are not easy to obtain. So, instead we seek $\mathbf{v}_3 = -\mathbf{v}_2(2l, \gamma)$ at the sphere, where γ depends on the direction of oscillation of the sphere. Solving for $\mathbf{v}_3$ is the same as solving for $\mathbf{v}_1$, so this simplifies matters. Finally, we suppose that summation of $\mathbf{v}_i$ will converge to actual solution, but this eventually needs to be shown.

In the following, we only consider $\mathbf{v} = \mathbf{v}_1 + \mathbf{v}_2 + \mathbf{v}_3 + \mathbf{v}_4$, which will be a good approximation up to order $O(a/l)$ or $O((a/l)^3)$, depending on whether the frequency is high or low. Let $\mathbf{F}_i$ be the drag force of $\mathbf{v}_i$ acting on sphere, and suppose the angle between the normal component of plane and the velocity of sphere is γ. We calculate the drag force of

$\mathbf{v}$ in two cases: first, when the sphere oscillates perpendicular to the plane surface of fluid, i.e., $\gamma = 0$, and then when the sphere oscillates parallel to the plane, when $\gamma = \pi/2$. The general case is a linear combination of the two. Notice that $|\mathbf{F}_2|/|\mathbf{F}_3| = O(a/l)$ and $\mathbf{F}_4$ is even smaller than $\mathbf{F}_2$; therefore, we can just consider the contributions of $\mathbf{F}_1$ and $\mathbf{F}_3$, i.e.,

$$\mathbf{F} \simeq \mathbf{F}_1 + \mathbf{F}_3 \,.$$

The drag force $\mathbf{F}_1$ was given in the previous section as

$$\mathbf{F}_1 = -\zeta(\omega)\mathbf{u}$$

where

$$\zeta(\omega) := -4\pi\eta a x^2 \big[(1-y)Q + 2(x-1)P\big]/3 \,.$$

So, to next order

$$\mathbf{F} \simeq \mathbf{F}_1 + \mathbf{F}_3 = -\zeta(\omega)\mathbf{u} + \zeta(\omega)\mathbf{v}_2(2l,\gamma) \,.$$

When the sphere is moving perpendicular to plane

$$\mathbf{v}_2(2l,0) = -v_r(2l,0)\,\frac{\mathbf{u}}{|\mathbf{u}|} \,,$$

while when the sphere is moving parallel to plane

$$\mathbf{v}_2(2l,\pi/2) = v_\theta(2l,\pi/2)\,\frac{\mathbf{u}}{|\mathbf{u}|} \,.$$

For the case of arbitrary motion, supposing the angle between normal component of plane and the velocity of sphere is γ, we obtain

$$\mathbf{v}_2(2l,\gamma) = -\mathbf{n}\, v_r(2l,0) u \cos\gamma + \mathbf{t}\, v_\theta(2l,\pi/2) u \sin\gamma \,,$$

where $\mathbf{n}$ and $\mathbf{t}$ are unit vectors normal and tangent to the plane, respectively.

Although in principle, we can use the fluctuation-dissipation theorem to find the VACF, the complicated formula of the response function ζ makes it hard to find exact an expression for the VACF. So, we simplify the response function by considering high frequency and low frequency regimes. Let $s = -i\omega$. Define $t_\nu = a^2\rho_0/\eta, t_{\nu'} = a^2\rho_0/(4\eta/3+\mu), t_c = a/C$ to be three characteristic time scales. In order for $\sqrt{s}$ to make sense, we place a branch cut along negative real axis, making the square root well defined. Below we make several simplifications, the reasons for which will become clear later.

<u>Low Frequency Case, $s << 1$:</u>

$$\begin{aligned}
x^2 &= -\alpha^2 a^2 = s t_\nu \,, \qquad x = -\sqrt{s t_\nu} \\
y^2 &= \frac{s^2 t_c^2}{1 + s\left(\frac{t_c}{t_{\nu'}}\right) t_c} \simeq s^2 t_c^2 \left(1 - s\,(t_c/t_{\nu'})\, t_c\right) \quad \Rightarrow \quad y \simeq -t_c s \\
\triangle &= 2x^2(3-3y+y^2) + y^2(3-3x+x^2) \simeq 6 s t_\nu \\
P &\simeq (3 + 3 s t_c + s^2 t_c^2)/2 s t_\nu \,, \qquad Q \simeq -(3 + 3\sqrt{s t_\nu} + s t_\nu)/2 s t_\nu
\end{aligned}$$

$$\zeta(s) \simeq 6\pi\eta a(1+\sqrt{st_\nu})$$

$$\begin{aligned}
v_r(2\theta,0) &\simeq \left[-\frac{2P}{(2l)^3}\,a^3e^{i\alpha(2l-a)} - \frac{2Q}{(2l)^3}\,a^3e^{i\beta(2l-a)}\right]u \\
&\simeq -\frac{2}{8}\left(\frac{a}{l}\right)^3\left[\left(\frac{3}{2st_\nu}+\frac{3t_c}{2t_\nu}\right)-\left(\frac{3}{2st_\nu}+\frac{3}{2\sqrt{st_\nu}}\right)\right]u \\
&\simeq \frac{3}{8}\left(\frac{a}{l}\right)^3\frac{u}{\sqrt{st_\nu}} \\
v_\theta(2\theta,\pi/2) &\simeq \left[-\frac{P}{(2l)^3}\,a^3e^{i\alpha(2l-a)} - \frac{Q}{(2l)^3}\,a^3e^{i\beta(2l-a)}\right]u \\
&\simeq \frac{3}{16}\left(\frac{a}{l}\right)^3\frac{u}{\sqrt{st_\nu}}\,.
\end{aligned}$$

<u>High Frequency Case, $s >> 1$:</u>

$$\begin{aligned}
x &= -\sqrt{s\,t_\nu}\,, \qquad y \simeq -\sqrt{st_{\nu'}}\,, \qquad \Delta \simeq 3st_\nu st_{\nu'} \\
P &\simeq \frac{3s^{-3/2}}{t_\nu\sqrt{t_{\nu'}}}+\frac{s^{-1}}{t_\nu}\,, \qquad Q \simeq -\frac{3\,s^{-3/2}}{t_{\nu'}\sqrt{t_\nu}}-\frac{s^{-1}}{t_{\nu'}}
\end{aligned}$$

$$\zeta(s) \simeq 2\zeta\left[(t_\nu/t_{\nu'}+2)+(\sqrt{t_\nu/t_{\nu'}}+2)\sqrt{st_\nu}\right]/9$$

$$\begin{aligned}
v_r(2l,0) &\simeq \left[-\frac{1}{2}\left(\frac{a}{l}\right)^2 e^{-\sqrt{st_\nu}2l/a}\frac{1}{\sqrt{st_\nu}}+\frac{a}{2l}e^{-\sqrt{st_{\nu'}}2l/a}\right]u \\
v_\theta(2l,\frac{\pi}{2}) &\simeq \left[-\frac{a}{2l}e^{-\sqrt{st_\nu}2l/a}+\frac{1}{4}\left(\frac{a}{l}\right)^2 e^{-\sqrt{st_{\nu'}}\frac{2l}{a}}\frac{1}{\sqrt{st_{\nu'}}}\right]u\,.
\end{aligned}$$

Now we use the above expressions to calculate the velocity autocorrelation function, which is given by

$$\Phi(t) = \langle v_i(0)v_i(t)\rangle = \frac{k_BT}{\pi}\int_{-\infty}^{\infty} d\omega e^{-i\omega t}Re\frac{1}{-i\omega m+(1+c)\zeta(\omega)} \tag{0.4.1}$$

$$= \frac{k_BT}{2\pi i}\int_{\epsilon-i\infty}^{\epsilon+i\infty} ds\,e^{st}\frac{1}{sm+(1+c)\zeta(s)}\,, \tag{0.4.2}$$

where $s=-i\omega$ and c is a correction term that depends on γ.

Let us first calculate the velocity autocorrelation in the low frequency case, $s << 1$, without the plane ($c=0$), which will we will use to compare with the plane case. We obtain

$$\frac{\Phi(t)}{k_BT} \simeq \mathcal{L}^{-1}\left(\frac{1}{6\pi\eta a+6\pi\eta a\sqrt{t_\nu s}}\right) \tag{0.4.3}$$

$$= \frac{1}{6\pi\eta a\sqrt{t_\nu}}\mathcal{L}^{-1}\left(\frac{1}{\sqrt{s}+1/\sqrt{t_\nu}}\right)\,. \tag{0.4.4}$$

From any table of inverse Laplace transforms, we obtain

$$\mathcal{L}^{-1}\left(\frac{1}{\sqrt{s}+a}\right) = \frac{1}{\sqrt{\pi t}} - ae^{a^2 t}\mathrm{erfc}(a\sqrt{t}) \tag{0.4.5}$$

where the erfc function is defined by

$$\mathrm{erfc}(x) = \frac{e^{-x^2}}{x\sqrt{\pi}} \sum_{n=0}^{\infty} (-1)^n \frac{(2n-1)!!}{(2x^2)^n} \,. \tag{0.4.6}$$

From (0.4.5) we find for the case without the plane, that the asymptotic behavior of velocity autocorrelation as $t \to \infty$ is given by

$$\Phi(t) = \frac{k_B T}{6\pi\eta a\sqrt{t_\nu}} \frac{t_\nu}{2\sqrt{\pi}} t^{-\frac{3}{2}} = \frac{k_B T}{\zeta} \frac{\sqrt{t_\nu}}{2\sqrt{\pi}} t^{-\frac{3}{2}} \,, \tag{0.4.7}$$

where $\zeta := 6\pi\eta a$, for simplicity.

Now, when the fluid is bounded by the plane, I couldn't find a formula in any Laplace inverse transformation table. Thus, we needed to directly calculate it. The following formula will be useful for the case $s << 1$. Suppose $R(\sqrt{s}) = a_0 + a_1\sqrt{s} + a_2 s + a_3 s^{3/2}$. When t is large, we obtain

$$\Phi(t) = \frac{k_B T}{2\pi i} \int_{\epsilon - i\infty}^{\epsilon + i\infty} ds\, e^{st} \frac{\sqrt{s}}{R(\sqrt{s})} = -\frac{k_B T}{2a_0\sqrt{\pi}} t^{-3/2} \,. \tag{0.4.8}$$

The above follows because

$$\begin{aligned}
\Phi(t) &= \frac{k_B T}{2\pi i} \int_{\epsilon - i\infty}^{\epsilon + i\infty} ds\, e^{st} \frac{\sqrt{s}}{R(\sqrt{s})} \\
&\simeq -\frac{k_B T}{2\pi i} \int_0^\infty dr \left[\frac{e^{-tr}\sqrt{r}i}{R(\sqrt{r}i)} + \frac{e^{-tr}\sqrt{r}i}{R(-\sqrt{r}i)}\right] \\
&= -\frac{2k_B T}{\pi} \int_0^\infty d(s/\sqrt{t}) \frac{e^{-s^2} s^2/t(a_0 - a_2 s^2/t)}{(a_0 - a_2 s^2/t)^2 + (a_3 s^3/t^{3/2} - a_1 s/\sqrt{t})^2} \\
&\simeq -\frac{2k_B T}{\pi a_0} t^{-3/2} \int_0^\infty ds\, e^{-s^2} s^2 \\
&= -\frac{k_B T}{2a_0\sqrt{\pi}} t^{-3/2} \,.
\end{aligned}$$

We can switch the order of the limit and the integration in above calculation as long as $a_0 \neq 0$ and $a_0/a_2 \neq a_1/a_3$. The physics of the problem indicates that Φ should approach zero when $t \to \infty$, which means the poles of above integrand are located in the region $Re(s) < 0$. These poles will contribute to exponential decay to $\Phi(t)$, which is much smaller than the contribution from the branch point $s = 0$, which gives algebraic decay. Thus, if we want to know the long time behavior of $\Phi(t)$, we can assume $s << 1$ to simplify our calculation, because the contributions from a_1, a_2, a_3 are much smaller than that from a_0.

When $a_0 = 0$, which corresponds to no plane case ($l = \infty$), we cannot switch the order of limit and integration. We should first set $a_0 = 0$ in the integral, then take limit in t, giving

$$\Phi(t) = \frac{k_B T}{2\sqrt{\pi}} \frac{a_2}{a_1^2} t^{-\frac{3}{2}} \,. \tag{0.4.9}$$

Notice, by plugging in the a_i, this gives the same formula as we derived above, $\Phi(t) = k_B T\sqrt{t_\nu}\, t^{-3/2}/(2\zeta\sqrt{\pi})$.

Now, consider the long time behavior of $\Phi(t)$ when there is a plane and $\gamma = 0$, i.e., the sphere is moving perpendicular to the plane. Again letting $\zeta = 6\pi\eta a$, we have

$$\begin{aligned}\Phi(t) &= k_B T \mathcal{L}^{-1}\left(\frac{1}{ms + \zeta\left[\sqrt{st_\nu} + (1 - 3(a/l)^3/8) - 3(a/l)^3/(8\sqrt{st_\nu})\right]}\right) \\ &= \frac{k_B T}{2\pi i}\int_\Gamma ds\, \frac{e^{ts}\sqrt{s}}{ms^{3/2} + \zeta\sqrt{t_\nu}s + \zeta\left[1 - 3(a/l)^3/8\right]\sqrt{s} - 3\zeta(a/l)^3/(8\sqrt{t_\nu})}\,,\end{aligned}$$

where Γ is a contour from $-\infty$ to 0 and 0 to $-\infty$ and $a_0 = -3\zeta(a/l)^3/(8\sqrt{t_\nu})$. So, when $t \to \infty$ we obtain

$$\Phi(t) = \frac{4k_B T\sqrt{t_\nu}}{3\sqrt{\pi}\zeta}\left(\frac{a}{l}\right)^{-3} t^{-\frac{3}{2}}\,. \tag{0.4.10}$$

For $\gamma = \frac{\pi}{2}$, i.e., when the sphere is moving parallel to the plane, $a_0 = 3\zeta(a/l)^3/(16\sqrt{t_\nu})$ and

$$\Phi(t) = -\frac{8k_B T\sqrt{t_\nu}}{3\sqrt{\pi}\zeta}\left(\frac{a}{l}\right)^{-3} t^{-\frac{3}{2}}\,. \tag{0.4.11}$$

For general case, $\mathbf{u} = u[\mathbf{n}\cos\gamma + \mathbf{t}\sin\gamma]$ and

$$\mathbf{F} = -u\,\zeta(\omega)\left[\mathbf{n}\,(1 + v_r(2l, 0))\cos\gamma + \mathbf{t}\,(1 - v_\theta(2l, \pi/2))\sin\gamma\,\right]\,.$$

Notice, for general case, the direction of $\mathbf{F}$ is no longer parallel to the direction of $\mathbf{u}$. So must first specify a direction, then calculate correlation function along that direction.

For the short time behavior of $\Phi(t)$, we can perform calculations similar to those above to find the approximate behavior. The branch point $s = 0$ contributes $b_0\sqrt{t}$, while the poles contribute $\sum_i b_i e^{tp_i}$, where p_i denotes the locations of the poles of $R(\sqrt{s})$. So, $\Phi(t) = b_0\sqrt{t} + \sum_i b_i e^{tpi}$. To find exact values of b_i and p_i is very tedious, which we do not do here.

For the future work, the most natural goal would be to find the exact solution for the linearized Navier-Stokes equation, at least for the case $\gamma = 0$. But, this is a difficult problem due to the non-separability of Helmholtz equation in bipolar coordinate. If we try to use another coordinate system, for example spherical coordinates, to find the coefficients of the eigenfunctions that match the boundary condition at the sphere, then matching at the plane will not be likely possible. So, instead of finding an exact solution, it is reasonable to seek a better approximate solution than that provided by the image method.

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