FAST TRACK COMMUNICATION

Hydrodynamic coupling between two fluid membranes

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Abstract
The coupled in-plane diffusion dynamics between point-particles embedded in stacked fluid membranes is investigated. We calculate the contributions to the coupling longitudinal and transverse diffusion coefficients due to particle motion within the different as well as the same membranes. The stacked geometry leads to a hydrodynamic coupling between the two membranes.

(Some figures in this article are in colour only in the electronic version)

Biological membranes are fundamental to the existence of life with their ability to separate the ‘in’ and ‘out’ of a cell. Typical membranes are composed of lipid molecules which can spontaneously self-assemble into a fluid bilayer structure [1]. Under normal physiological conditions, finite temperature induces Brownian motion of membrane constituents, resulting in diffusive transport. In the seminal paper by Saffman and Delbrück [2], the diffusion coefficient of a rigid protein in a membrane was calculated. Following this work, there have been theoretical studies on diffusion of a rigid disc on supported membranes [3], a liquid domain in monolayers [4, 5], and a rod on immersed membranes [6, 7] or on Langmuir monolayers [8].

The present work focuses on the coupling of diffusion dynamics between two fluid membranes through an intervening bulk fluid. It was reported that model experimental systems in which two lipid membranes are stacked on a substrate exhibit correlated dynamics [9]. This planar geometry has become favorable to study the membrane dynamics which is otherwise not possible in vesicles [10]. The coupling effect between two membranes can be important in biological systems with a large concentration of cells such as tissues. Other examples are Gram-negative bacteria which enclose a periplasmic space with an approximate width of about 15–20 nm between their inner and outer lipid bilayers [1]. Highly folded membranous organelles such as Golgi apparatus also correspond to a situation in which membranes come in close proximity to each other. In all these cases, it is very relevant to consider the hydrodynamic coupling between two biomembranes.

Hydrodynamic models of membranes exploit their fluid nature, in which the membranes are assumed to be two-dimensional (2D) viscous fluid sheets embedded in a three-dimensional (3D) solvent. One such investigation involved the calculation of the correlated diffusion of proteins embedded in a membrane immersed in an unbounded fluid [11] or a membrane adjacent to a solid support [12]. In this communication, we report on the coupling diffusion coefficients of particles embedded in such a stacked membrane system.

We first establish the governing equations. It is assumed here that the membranes are infinite planar sheets of fluid. The persistence length is defined as the length over which the correlations of the normal vector decay to zero [13]. For a lipid bilayer membrane with bending rigidity of about $10k_B T$, the persistence length turns out to be much larger than the size of a typical cell. Hence we neglect out-of-plane fluctuations of the membrane for mathematical simplicity. The fluid membranes are embedded in a bulk solvent such as water or suitable buffer solution. As shown in figure 1, the membranes are fixed in the $xy$-plane at $z = \pm h/2$. Let $\mathbf{v}^{(i)}(\mathbf{r})$ be the 2D velocity of the membrane fluids. Here the index $i = 1, 2$ represents the two membranes, and the 2D vector $\mathbf{r} = (x, y)$ represents a point in the planes of the membranes. We work in the low-Reynolds number regime of the membrane hydrodynamics so that the
inertial effects can be neglected. This allows us to use the 2D Stokes equations

\[ \eta \nabla^2 \mathbf{v}^{(i)} - \nabla p^{(i)} + \mathbf{f}^{(i)} + \mathbf{F}^{(i)} = 0, \]  
(1)

along with the incompressibility condition

\[ \nabla \cdot \mathbf{v}^{(i)} = 0. \]  
(2)

Here \( \nabla \) is a 2D differential operator, \( \eta \) is the 2D membrane viscosity (same for both the membranes), \( p^{(i)}(\mathbf{r}) \) the 2D in-plane pressure, \( \mathbf{f}^{(i)}(\mathbf{r}) \) the force exerted on the membrane by the surrounding fluid, and \( \mathbf{F}^{(i)}(\mathbf{r}) \) is any other force acting on the membrane.

The solvent regions are denoted by the region \( j = i-, ii-i, \) and \( iii-ii \). The velocities and pressures in these regions are written as \( \mathbf{v}^{(i)}(\mathbf{r}, z) \) and \( p^{(i)}(\mathbf{r}, z) \), respectively. We assume that the solvent in the three regions have the same 3D viscosity denoted by \( \eta_s \). The solvent inertia is neglected and hence it also obeys the 3D Stokes equations

\[ \eta_s \nabla^2 \mathbf{v}^{(i)} - \nabla p^{(i)} + \mathbf{f}^{(i)} + \mathbf{F}^{(i)} = 0, \]  
(3)

where \( \nabla \) represents a 3D differential operator. Similar to the fluid membrane, the solvents in all the regions are considered to be incompressible

\[ \nabla \cdot \mathbf{v}^{(i)} = 0. \]  
(4)

The presence of the surrounding solvent is important because it exerts force on the liquid membranes. The force on membrane 1, indicated as \( \mathbf{f}^{(i)} \) in (1), is given by the projection of \( (\mathbf{\sigma}^{(i)} - \mathbf{\sigma}^{(ii)})_{z=h/2} \cdot \hat{\mathbf{e}}_z \) onto the \( xy \)-plane of the membrane. Here \( \hat{\mathbf{e}}_z \) is the unit vector along the \( z \)-axis, and \( \mathbf{\sigma}^{(i)} \) are the stress tensors

\[ \mathbf{\sigma}^{(i)} = -p^{(i)} \mathbf{I} + \eta_s \left[ \nabla \mathbf{v}^{(i)} + (\nabla \mathbf{v}^{(i)})^T \right]. \]  
(5)

In the above, \( \mathbf{I} \) is the identity tensor and the superscript ‘T’ indicates the transpose. Similarly, the force on membrane 2 is given by the projection of \( (\mathbf{\sigma}^{(ii)} - \mathbf{\sigma}^{(iii)})_{z=-h/2} \cdot \hat{\mathbf{e}}_z \) on the \( xy \)-plane. The general procedure is to first resolve (1) into components along \( \mathbf{k} \) and perpendicular to it, where \( \mathbf{k} \) is the 2D wavevector in Fourier space. We then solve the resulting differential equations for the velocities. Stick boundary conditions at the membrane–solvent interfaces are imposed. It is also assumed that the solvent velocities decay to zero at sufficiently large distances from the membranes. A similar calculation for a single membrane confined between two walls has been previously performed [14, 15].

Owing to the linearity of the governing Stokes equations, the in-plane velocity in membrane 1 can be obtained in Fourier space as

\[ v_a^{(1)}(\mathbf{k}) = G_a^{(11)}[\mathbf{k}] F_\beta^{(1)}[\mathbf{k}] + G_a^{(12)}[\mathbf{k}] F_\beta^{(2)}[\mathbf{k}], \]  
(6)

Here \( G_a^{(11)} \) and \( G_a^{(12)} \) are the mobility tensors given by

\[ G_a^{(11)}[\mathbf{k}] = \frac{1}{\eta v^2} \frac{1 + 2 K + \coth(K H)}{K g(K, H)} \left( \delta_{a\beta} - \frac{k_a k_\beta}{k^2} \right), \]  
(7)

\[ G_a^{(12)}[\mathbf{k}] = \frac{1}{\eta v^2} \frac{1}{K g(K, H)} \left( \delta_{a\beta} - \frac{k_a k_\beta}{k^2} \right), \]  
(8)

with

\[ g(K, H) = 1 + 2 K \left( 1 + K \right) \left( 1 + 2 K \right) \coth(K H), \]  
(9)

and \( v = 2 \eta_s / \eta \). Notice that \( v^{-1} \) represents the hydrodynamic screening length. We have also used the definitions \( H = h v \) and \( K = k / \nu \) with \( k = |\mathbf{k}| \). By symmetry, a similar set of expressions can be written down for the membrane 2 also. The above mobility tensors represent the hydrodynamic coupling between the two membranes.

Consider a pair of particles embedded in the membrane undergoing Brownian motion separated by \( r \). For sufficiently large times, the particle displacements obey \( \Delta r = 2 D_{ab} \Delta \tau \), where \( \Delta r \) represents the displacement of the first particle and \( \Delta \tau \) represents that of the second particle. In the above, the diffusion tensor for over-damped dynamics is given by the Einstein relation \( D_{ab} = k_B T G_a^{(1)} \), where \( k_B \) is the Boltzmann constant and \( T \) the temperature. We now apply these definitions to a double-membrane system. The line of centers connecting any two particles in the membranes after projection onto the 2D plane can be taken to be along the \( x \)-axis without loss of generality. Then we obtain the coupling longitudinal diffusion coefficients as \( D_L^{(11)}(\mathbf{r}) = k_B T G_L^{(11)}(\mathbf{r} \mathbf{e}_z) \) for two particles within the same membrane, and \( D_L^{(12)}(\mathbf{r}) = k_B T G_L^{(12)}(\mathbf{r} \mathbf{e}_z) \) for two particles in different membranes. The coupling transverse diffusion coefficients are \( D_T^{(11)}(\mathbf{r}) = k_B T G_T^{(11)}(\mathbf{r} \mathbf{e}_z) \) and \( D_T^{(12)}(\mathbf{r}) = k_B T G_T^{(12)}(\mathbf{r} \mathbf{e}_z) \). The longitudinal coupling diffusion coefficient is associated with Brownian motion along the line of centers, while the transverse one is associated with motion perpendicular to the line of centers [11, 12, 16]. In the following, we discuss the above four diffusion coefficients sequentially.
The real-space expressions for the mobility tensors can be obtained by inverse Fourier transform. Using the notation \( R = R v \), we obtain the longitudinal coupling diffusion coefficient within the same membrane as

\[
D_L^{(11)}(R, H) = 2 \int_0^\infty dK \frac{1}{2} + 2K + \coth(KH) J_n(KR) \frac{g(K, H)}{KR},
\]

where \( J_n(t) \) are the Bessel functions of the first kind of order \( n \). Hereafter, all the diffusion coefficients are scaled by a factor \( k_B T/(4\pi \eta) \) in order to make them dimensionless. In figure 2, we plot \( D_L^{(11)}(R, H) \) as a function of \( R \) for various \( H \) following numerical integration. It is a monotonically decreasing function of \( R \). The red circles (\( H = 0.1 \)), blue squares (\( H = 1 \)) and green triangles (\( H = 10 \)) represent the intermediate membrane separations respectively. These symbols represent the same \( H \) values in the rest of the figures in this communication.

For \( H \gg 1 \), we can approximate \( \coth(KH) \approx 1 \) so that the \( H \)-dependence drops out. In this limit, the integral can be analytically performed to yield

\[
D_L^{(11)}(R) = \frac{2}{R^2} + \frac{\pi}{R}[H_n(R) - Y_n(R)],
\]

where \( H_n(t) \) are the Struve functions and \( Y_n(t) \) are the Neumann functions. Since the membranes are effectively isolated from each other for large \( H \), (11) coincides with the analytical expression obtained for a single membrane geometry [11]. For \( R \ll 1 \), (11) has a logarithmic behavior, i.e., \( D_L^{(11)}(R \ll 1) \approx \ln(2/R) - \gamma + 0.5 \), where \( \gamma \approx 0.5772 \ldots \) is Euler’s constant. In this regime, the outer solvent surrounding the membrane is unimportant and the membrane behaves effectively as a pure 2D fluid. In the opposite \( R \gg 1 \) limit, (11) decays algebraically \( D_L^{(11)}(R \gg 1) \approx 2/R \).

On recasting in dimensional form, we see that this limiting expression is independent of the membrane viscosity \( \eta \), and dependent only on the solvent viscosity \( \eta_s \). When \( H \ll 1 \), on the other hand, (10) becomes \( D_L^{(11)}(R/2)/2 \). The vanishing thickness of the solvent region results in a rescaling of \( v^{-1} \) by a factor of two, and hence the resultant expression. The solid and dashed lines in figure 2 represent the above limiting cases of large and small \( H \) limits, respectively. It is thus observed that the presence of the second membrane also has a finite contribution to the longitudinal coupling diffusion coefficient within the same membrane.

Following the same argument, the transverse coupling diffusion coefficient between two particles within the same membrane is calculated according to

\[
D_T^{(11)}(R, H) = 2 \int_0^\infty dK \frac{1}{2} + 2K + \coth(KH) \frac{g(K, H)}{KR} \times \left[ J_0(KR) - \frac{J_1(KR)}{KR} \right].
\]

In figure 3, the variation of \( D_T^{(11)}(R, H) \) as a function of \( R \) for various \( H \) is plotted using numerical integration. Similar to the longitudinal case, it is also a monotonically decreasing function of \( R \). For \( H \gg 1 \), (12) can analytically treated to have the form

\[
D_T^{(11)}(R) = \frac{2}{R^2} - \frac{\pi}{R}[H_n(R) - Y_n(R)] + \pi[H_0(R) - Y_0(R)],
\]

which also coincides with the expression for the transverse coupling diffusion coefficient in a single membrane [11]. In this case, the asymptotic behaviors for small and large \( R \) are \( D_T^{(11)}(R \ll 1) \approx \ln(2/R) - \gamma + 0.5 \) and \( D_T^{(11)}(R \gg 1) \approx 2/R^2 \), respectively. In the opposite limit of \( H \ll 1 \), (12) becomes \( D_T^{(11)}(R/2)/2 \). As before, the solid and dashed lines
As seen by the green triangles (the above integral vanishes for large inter-membrane distances $H$ values of $H$ correspond to the exponential decay of the cosech $H$ for various $H$ values of $H$ when $H$ is plotted. The above integral also vanishes when $H \gg 1$, as expected for the decoupled membranes. In the opposite limit of $H \ll 1$, (15) results in $D^{(12)}_T(R)/2$ as plotted by the dashed line in figure 5.

In figure 5, the variation of $D^{(12)}_T(R, H)$ as a function of $R$ for various $H$ is plotted. This is due to the exponential decay of the cosech($KH$) term. Up to $H = 1$, the proximity to the second membrane leads to additional contributions to the coupling diffusion coefficients. This is the main result of this work. Using typical values for the solvent (water) and membrane (lipid bilayer) viscosities, the hydrodynamic screening length $\nu^{-1}$ can be estimated to be of the order of micrometers. This implies that an adjacent membrane within this distance ($H < 1$) can have a strong bearing on the diffusion dynamics, such as for typical stacked supported membrane experiments [9]. Qualitatively, the presence of the second membrane can enhance the effective coupling diffusion.

Even though the model presented in this communication captures the essential physics, it looks somewhat simplistic in several respects. We have neglected the finite size effects of the membrane inclusions which are known to modify the membrane response at small inter-particle distances [11, 12]. At distances much larger than the inclusion size, however, these effects become unimportant. The curvature effects of the membrane are significant when the radius of curvature is of the order of the hydrodynamic screening length $\nu^{-1}$ [17, 18]. The extension of our work to membranes in spherical configuration is possible. The out-of-plane fluctuations of the membrane lead to a reduction in the diffusion coefficient of proteins in single membranes [19, 20]. However, it is known that the presence of a substrate or a second membrane would suppress the out-of-plane membrane fluctuations [21–23]. Overall, we expect that fluctuations and the presence of a substrate will not qualitatively affect our results.

To summarize, we have calculated the longitudinal as well as transverse coupling diffusion coefficients of particles undergoing Brownian motion in a stacked double-membrane geometry. We obtained the additional contribution to the coupling diffusion coming from the proximity to adjacent membranes. As in biological systems such as tissues the cells are rarely isolated, our results also imply that the proximity to neighboring cells can affect the diffusion of objects in cell membranes.

\[ D^{(12)}_L(R, H) = 2 \int_0^\infty dK \frac{\cosh(KH) J_1(KR)}{g(K, H) KR}. \] (14)

The functional dependence of $D^{(12)}_L(R, H)$ on $R$ for various $H$ is shown in figure 4. Since cosech$(KH) \approx 0$ for $H \gg 1$, the above integral vanishes for large inter-membrane distances as seen by the green triangles ($H = 10$) in figure 4. This is a reasonable result as the membranes are effectively independent of each other. In the limit of $H \ll 1$, (14) results in $D^{(11)}_L(R)/2$ which is plotted by the dashed line in figure 4. As mentioned earlier, this function has an initial logarithmic behavior followed by an asymptotic $1/R$-decay.

The transverse coupling diffusion coefficient between two particles in different membranes is given by

\[ D^{(12)}_T(R, H) = 2 \int_0^\infty dK \frac{\cosh(KH) J_1(KR)}{g(K, H) KR} \times \left[ J_0(KR) - \frac{J_1(KR)}{KR} \right]. \] (15)

In figure 5, the variation of $D^{(12)}_T(R, H)$ as a function of $R$ for various $H$ is plotted. The above integral also vanishes when $H \gg 1$, as expected for the decoupled membranes. In the opposite limit of $H \ll 1$, (15) results in $D^{(11)}_T(R)/2$ as plotted by the dashed line in figure 5.

From figures 4 and 5, it can be observed that for large $H$, the coupling diffusion coefficients vanish. This is due to the exponential decay of the cosech($KH$) term. Up to $H = 1$, the proximity to the second membrane leads to additional contributions to the coupling diffusion coefficients. This is the main result of this work. Using typical values for the solvent (water) and membrane (lipid bilayer) viscosities, the hydrodynamic screening length $\nu^{-1}$ can be estimated to be of the order of micrometers. This implies that an adjacent membrane within this distance ($H < 1$) can have a strong bearing on the diffusion dynamics, such as for typical stacked supported membrane experiments [9]. Qualitatively, the presence of the second membrane can enhance the effective coupling diffusion.

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