Brownian motion near a liquid-like membrane

Thomas Bickel
CPMOH, Université Bordeaux 1 – CNRS (UMR 5798)
351 cours de la Libération, 33405 Talence, France

The dynamics of a tracer molecule near a fluid membrane is investigated, with particular emphasis given to the interplay between the instantaneous position of the particle and membrane fluctuations. It is found that hydrodynamic interactions create memory effects in the diffusion process. The random motion of the particle is then shown to cross over from a “bulk” to a “surface” diffusive mode, in a way that crucially depends on the elastic properties of the interface.

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I. INTRODUCTION

A largely unsolved problem in soft materials is to understand how the random motion of a colloid is correlated with the viscoelastic properties of the embedding fluid \( \eta \). Since complex fluids store elastic energy, the instantaneous position and velocity of the Brownian particle depend on its prior history. As a signature of this “memory” effect, the friction force experienced by the particle is, in a generalized Langevin description, non-local in time \( \alpha \). To illustrate this point, we consider a system simple enough that allows us to extract the memory kernel. We focus in this paper on the diffusion of nano-particles near a liquid-like membrane. Fluid membranes are soft surfaces, self-assembled from surfactant solutions. In living systems, they divide the cell into distinct compartments and incorporate a large amount of proteins involved in signalling activities. Interactions between nano-sized particles and fluid membranes are ubiquitous in phenomena such as antibiotic delivery, nanoparticle toxicity or virus entry in cells \( \alpha \). This issue is relevant not only from biological or biophysical standpoints, but also in the formulation of many cosmetics and pharmaceutical products \( \alpha \). At a more fundamental level, recent experiments on the dynamics of particles near a membrane have revealed peculiar kinetics effects such as anomalous diffusion \( \alpha \), or hierarchical transport mechanisms \( \alpha \). Besides, anisotropic coherent motion of particles induced by an alternating electric field has also been shown to enhance the smectic order in a lamellar phase of membrane \( \alpha \). From all these perspectives, a detailed investigation of the Brownian motion in the vicinity of a membrane appears to be essential.

Interfacial contributions to transport coefficients have been evaluated long ago for elementary geometries \( \alpha \). Far from the surface, the mobility of a spherical particle of radius \( a \) is \( \mu_0 = 1/(6\pi\eta a) \), with \( \eta \) the shear viscosity of the embedding (simple) fluid. When a horizontal wall is located at distance \( z_0 \) from the particle, the fluid flow resulting from the motion of the bead “reflects” from the interface back to the particle, thus exerting an additional friction force. At leading order, hydrodynamic corrections lead to a position-dependent mobility tensor

\[
\mu_{xx} = \mu_{yy} = \mu_0 \left[ 1 - \frac{a}{z_0} \right], \quad (1a)
\]
\[
\mu_{zz} = \mu_0 \left[ 1 - \frac{a}{z_0} \right], \quad (1b)
\]

and \( \mu_{ij} = 0 \) for \( i \neq j \). Note that within the assumption of point-like particle \( a \ll z_0 \), the exact shape of the colloid is essentially irrelevant \( \alpha \). In Eqs. (1a) and (1b), particle mobility is affected in a manner that depends on the nature of the interface. For a solid surface, the no-slip boundary condition leads to \( \alpha \)

\[
\alpha_{\parallel}^{(s)} = \frac{9}{16}, \quad \text{and} \quad \alpha_{\perp}^{(s)} = \frac{9}{8}. \quad (2)
\]

During the last decade, precise experimental measurements of particle diffusivity near solid surfaces have shown excellent agreement with theory \( \alpha \). If, however, the interface separates two liquids having the same viscosity \( \eta \), the corrections are \( \alpha \)

\[
\alpha_{\parallel}^{(l)} = \frac{3}{32}, \quad \text{and} \quad \alpha_{\perp}^{(l)} = \frac{15}{16}. \quad (3)
\]

More recently, the effects of partial slip \( \alpha \) as well as fluid compressibility \( \alpha \) have also been included in the calculation of the mobility tensor.

Unlike surfaces having a frozen structure, fluid membranes are responsive materials. The Brownian motion of a particle near a membrane is then expected to be dynamically coupled to the conformations of the interface. In Section II we consider the problem of free diffusion near a membrane, with particular emphasis given to hydrodynamic interactions. We show in Section III that the instantaneous solution of the creeping flow equations depends on the whole history of both colloid’s and interface’s motions. Consequences for the diffusion process are then discussed in Section IV. In particular, we find that the random motion of a free colloid crosses over from a “bulk” to a “surface” diffusive mode. Finally, we come

*Electronic address: th.bickel@cpmoh.u-bordeaux1.fr
back to the relationship with experiments and draw some concluding remarks in Section IV the technical details being relegated to the appendices.

II. HYDRODYNAMICS NEAR A FLUID MEMBRANE

A. Physics of membranes

Fluid membranes belong to the general class of soft objects (including polymers, gels . . . ), that deform easily when submitted to external stresses, and undergo thermally excited shape fluctuations to increase their configurational entropy. At length-scales much larger than the bilayer thickness (i.e., micrometers vs. nanometers), a fluid membrane can be described as an infinitely thin liquid layer with essentially no in-plane shear viscosity [18]. The instantaneous conformation of the almost flat membrane is specified through the displacement field \( h(\mathbf{p}, t) \), with \( \mathbf{p} = (x, y) \). For our purpose, it is more convenient to use the (2D) Fourier representation

\[
h_q(t) = \int d^2 \rho \exp[-i \mathbf{q} \cdot \mathbf{h}(\mathbf{p}, t)] , \tag{4}
\]

with \( \mathbf{q} = (q_x, q_y) \). As first explained by Helfrich, the large variety of shapes and topologies assume by membranes is governed by bending energy. For relatively smooth deformations, the statistical mechanics of a single membrane is based on the following Hamiltonian [19]

\[
H_m[h] = \frac{\kappa}{2} \int \frac{d^2 q}{(2\pi)^2} \left( q^4 + \xi_{\parallel}^{-4} \right) |h_q|^2 , \tag{5}
\]

with \( \kappa \) the bending rigidity. In this description, the membrane is confined in a harmonic potential. The in-plane correlation length \( \xi_{\parallel} \) plays the role of an external control parameter setting the mean square displacement of the membrane: \( \langle h(\mathbf{p})^2 \rangle^{1/2} \propto (k_B T/\kappa_{\parallel})^{1/2} \xi_{\parallel} \), with \( T \) the absolute temperature and \( k_B \) the Boltzmann constant.

As recalled in Appendix A, the relaxation rate of a fluctuation with wavevector \( \mathbf{q} \) follows the dispersion relation \( \omega_q = \kappa(q^4 + \xi_{\parallel}^{-4})/(4\eta q) \) [21]. Overdamped surface waves are further characterized by their longest relaxation time

\[
\tau_m = \frac{4\eta}{\kappa \xi_{\parallel}^3} . \tag{6}
\]

For a typical bilayer in water with \( \eta = 10^{-3} \text{ Pa.s} \) and \( \xi_{\parallel} = 0.5 \mu m \), this relaxation time varies between \( \tau_m \approx 10^{-3} \text{ s} \) for \( \kappa = 4.10^{-19} \text{ J} \) and \( \tau_m \approx 0.1 \text{ s} \) for \( \kappa = 4.10^{-21} \text{ J} \).

In order to describe the dynamic organization of a colloidal suspension near a membrane, \( \tau_m \) has to be compared with the typical time \( \tau_d \) needed for a particle to diffuse over its own radius

\[
\tau_m = \frac{\eta a^3}{k_B T} . \tag{7}
\]

This time-scale ranges from \( \tau_d \approx 10^{-6} \text{ s} \) for a particle with radius of a few nanometers, up to \( \tau_d \approx 1 \text{ s} \) for micrometer beads. Comparison of both time-scales defines two diffusion regimes in a straightforward manner. When \( \tau_d/\tau_m \gg 1 \), i.e. when \( a \gg \xi_{\parallel} \) or \( \kappa \gg k_B T \), the membrane appears essentially flat to the colloid and the system is dominated by the relaxation dynamics of the elastic interface. We call this regime the weak fluctuation (WF) regime. On the other hand when \( \tau_d/\tau_m \ll 1 \), the bead is strongly advected by the random flow caused by thermal undulations of the membrane. We will refer to the strong fluctuation (SF) regime, that will be the particular scope of investigation of this paper.

B. Linear hydrodynamic

For small-amplitude motions, the flow velocity \( \mathbf{u} \) and the local pressure \( p \) are governed by the linearized Navier-Stokes equation

\[
\eta \nabla^2 \mathbf{u} - \nabla p + \mathbf{f} + \Phi = 0 , \tag{8}
\]

together with the incompressibility condition \( \nabla \cdot \mathbf{u} = 0 \). Here, \( \mathbf{f} \) is an external force density causing the fluid motion, and \( \Phi \) the restoring force density exerted by the deformed interface. The latter is always perpendicular to the membrane. Within the assumption of smooth deformations, the force density is given in Fourier space by the functional derivative \( \Phi = (0, 0, -\delta H_m/\delta h_{\parallel}^2) \). The creeping flow Eq. (8) is then solved assuming that the membrane is not permeable to the fluid, a condition quite sensible on experimental time-scales (minutes or hours). Explicitly, this requirement reads

\[
\frac{\partial h}{\partial t} = u_z(\mathbf{p}, h(\mathbf{p}, t), t) . \tag{9}
\]

The hydrodynamic problem defined in this way is both time-dependent and highly non-linear due to the fact that \( h(\mathbf{p}, t) \) is unknown. Although it cannot be solved exactly (excepted by numerical methods), approximate solutions can be found. For instance, iterative methods have proved to be fruitful when the deformation of the interface is asymptotically small [21]. The zeroth-order term usually corresponds to the condition \( u_z(\mathbf{p}, 0, t) = 0 \), thus representing the motion of a spherical bead near a flat surface. The ensuing velocities and stresses can then be used to determine a first nonzero approximation for the deformation of the interface [21]. This strategy is however limited as it only accounts for the first “image” correction to hydrodynamic interactions. Instead, we follow a different route and assume that the fluid motion caused by thermal fluctuations of the membrane is only slightly perturbed by the presence of the colloid. The dynamics of the surface enters the problem through the condition

\[
\frac{\partial h}{\partial t} = u_z(\mathbf{p}, 0, t) . \tag{10}
\]
The consistency of this approach is then ensured as long as non-linear contributions in the deformation field can be neglected. In fact, this condition has the same range of validity as the harmonic description for the membrane Hamiltonian.

III. MOBILITY OF A SPHERE NEAR A FLUID MEMBRANE

A. Green’s function

Standard Green’s function methods are then used to solve Eq. (6). We first evaluate the response function, \( G \), to a point-like force \( f_0 \delta (r - r') \). The fluid flow corresponding to a given force field \( f(r) \) is then obtained from the convolution

\[ u(r, t) = \int dt \int d^3 r' G(r, r', t - t') f(r', t') . \]  

For the hydrodynamic problem defined above, the Oseen tensor splits into two contributions, \( G = G^{(0)} + \Delta G \). The first term, \( G^{(0)} \), is the well-known free space Green’s function and is given in Appendix A. The second contribution, \( \Delta G \), describes the fluid flow caused by the elastic response of the membrane. While the problem is translationally invariant in time and in the directions parallel to the surface, \( \Delta G \) is expected to depend on the perpendicular coordinates \( z \) and \( z' \) separately. Incidentally, we remark that the boundary condition Eq. (16) is more conveniently enforced if we use the set of variables \( \{q, z, \omega\} \), where we define the (time) Fourier transform of any function \( F(t) \) as \( \hat{F}(\omega) = \int_{-\infty}^{+\infty} dt e^{-i\omega t} F(t) \). The main lines of the derivation are given in Appendix B. We find after some algebra

\[ \Delta \hat{G}_{kl}(q, z, \omega) = \frac{i}{4\eta q} \frac{\omega g}{\omega - i\omega q} \gamma_k(q, z) \gamma_l(q, z') M_{kl} , \]  

where the matrix \( M \) has diagonal elements \( M_{kk} = 1 \), and off-diagonal terms \( M_{xy} = M_{yx} = 1 \), \( M_{xz} = M_{yz} = -i \), and \( M_{zx} = M_{zy} = i \). The functions \( \gamma_k \) are given by

\[ \gamma_x(q, z) = q_x e^{-q|z|} , \] \[ \gamma_y(q, z) = q_y e^{-q|z|} , \] \[ \gamma_z(q, z) = e^{-q|z|} (1 + q|z|) . \]

Remark that these expressions are consistent with the symmetry property of the Green’s functions: indeed, it can be shown from general arguments that \( \delta_{kl}(r, r') = \hat{G}_{kl}(r', r) \). This is re-expressed in terms of our particular choice of variables as \( \hat{G}_{kl}(q, z, z', \omega) = \hat{G}_{kl}(-q, z', z, \omega) \), property that can be checked easily.

In Eq. (12), the prefactor \( (\omega - i\omega q)^{-1} \) is a clear signature of hydrodynamic scattering effects. Indeed, the fluid flow resulting from a displacement of the particle exerts stresses that deform the membrane. Relaxing back to its equilibrium position, the membrane creates a backflow that in turn perturbs the motion of the colloid, and so forth. The infinite sum \( (\omega - i\omega q)^{-1} = i/\omega q \sum_{n=0}^{\infty} (-i)^n (\omega/\omega q)^n \) is the expression of this infinite series of “reflexions” of the original point force on the interface.

B. Mobility tensor

The next step in the derivation consists in identifying the mobility tensor of a sphere. To this aim, we still have to enforce the no-slip boundary condition for the fluid flow on the surface of the colloid. If we note \( v(r_0) \) and \( \Omega \) respectively the translational and rotational velocity of the Brownian particle, \( r_0 \) being the position of its center-of-mass, then the fluid velocity is

\[ u(r_0 + a) = v(r_0) + \Omega \times a , \]  

for any vector \( a \) scanning the bead’s surface. The friction force \( F_H \) exerted by the liquid on the particle is then obtained from

\[ F_H = - \int dV f(r) = - \int dS f(a) , \]  

where the integral is running over the surface of the bead. In this expression, \( f(r) = f(a) \delta[(r - r_0) - a] \) is the force density exerted by the particle on the fluid. Integrating Eq. (17) over the particle’s surface, the angular velocity cancels out and one obtains, together with Eq. (14), a linear relation between the friction force and the velocity of the particle. This relation defines the (frequency-dependent) mobility tensor, \( \mathbf{\tilde{\mu}}(\omega) = -\hat{F}_H(\omega) \). The mobility can be written as the sum of two terms, \( \mu_{kl}(z_0, \omega) = \mu_0 \delta_{kl} + \Delta \tilde{\mu}_{kl}(z_0, \omega) \), with \( \mu_0 = 1/(6\eta a) \) the bulk value. In the limit of a point-like particle \( a \ll z_0 \), we obtain at leading order

\[ \Delta \tilde{\mu}_{kl}(z_0, \omega) = \int \frac{d^2 q}{(2\pi)^2} \Delta \hat{G}_{kl}(q, z_0, z_0, \omega) . \]  

From Eqs. (13a) – (13c), one can easily convince oneself that there are no cross-contributions: \( \Delta \tilde{\mu}_{kl} = 0 \) if \( k \neq l \).

1. Static mobility

The integral Eq. (16) can be performed easily when \( \omega = 0 \). As can be noticed from Eq. (12), the static mobility does not depend on the elastic properties of the interface. Indeed, we find

\[ \tilde{\mu}_{xz}(z_0, 0) = \tilde{\mu}_{yz}(z_0, 0) = \mu_0 \left( 1 - \frac{3a}{32z_0} \right) , \] \[ \tilde{\mu}_{zz}(z_0, 0) = \mu_0 \left( 1 - \frac{15a}{16z_0} \right) , \]

We therefore recover the mobility of a sphere near a flat, liquid interface given in Eq. (3).
2. Low-frequency limit

For finite frequency, $\Delta \tilde{\mu}_{kl}$ is a complicated function of the reduced variables $\omega \tau_m$, $a/z_0$ and $\xi_\parallel/z_0$. At this point of the discussion, it is useful to introduce the friction coefficient defined as the inverse of the mobility tensor, $\tilde{\zeta}_{kl} = (\tilde{\mu}^{-1})_{kl}$. Obviously, the friction tensor is also diagonal. In the low-frequency limit $\omega \tau_m \ll 1$, the matrix elements can be expanded and we find, to lowest order,

$$\tilde{\zeta}_{kl}(z_0, \omega) = \zeta_0 \left[ 1 + \alpha^{(l)}_k \left( \frac{a}{z_0} \right) - \beta_k i \omega \tau_m \right], \quad (18)$$

with $\zeta_0 = 1/\mu_0$. The prefactors $\beta_k$ depend on the distance to the membrane. We get $\beta_\parallel = 0$ and $\beta_\perp = 3\pi a/(16\xi_\parallel)$ when $\xi_\parallel \gg z_0$, whereas in the other limit $\xi_\parallel \ll z_0$ we obtain $\beta_\parallel = 9a\xi_\parallel/(64z_0^2)$ and $\beta_\perp = 27a\xi_\parallel/(32z_0^3)$.

Writing the complex friction as $\tilde{\zeta}(\omega) = 6\pi \eta(\omega)a$, it appears that Eq. (18) describes a Maxwell fluid with anisotropic, frequency-dependent shear viscosity $\eta$. The corresponding relaxation time is set by the membrane relaxation time $\tau_m$, multiplied by a geometric prefactor that depends on the distance to the elastic surface. This relaxation time arises from the delay in the response of the membrane to a deformation caused by the fluid flow.

IV. DIFFUSION NEAR A FLUID MEMBRANE

We are now in a position to study the diffusion of a tracer particle in the vicinity of a membrane. Because of hydrodynamic interactions, the friction force experienced by the Brownian particle is non-local in time. The corresponding generalized Langevin equation for a particle of mass $m$ reads

$$m \frac{dv}{dt} = -\int_{-\infty}^t dt' \zeta(t - t')v(t') + F(t), \quad (19)$$

with $F(t)$ the noise term. All the statistical properties of this random walk can, in principle, be calculated given the probability distribution of the random force. However, the problem is still involved because of the non-linear $z$-dependence of the friction term. To avoid this complication, we assume that the particle remains close to its original position during the time of observation — the validity of this assumption being discussed later. According to the fluctuation-dissipation theorem, the random force $F(t)$ can be chosen with zero mean value and correlations given by

$$\langle F_k(\omega) F_l(\omega') \rangle = 2k_BT \text{Re} [\tilde{\mu}_{kl}(\omega)] \times 2\pi \delta(\omega + \omega'). \quad (20)$$

We first focus on the motion in the direction perpendicular to the surface. In the overdamped limit, i.e. for time-scales much longer than $\tau = \zeta_0/m$, inertial terms can be neglected. Defining the mean square displacement (MSD) as $\langle \delta z^2 \rangle = \langle (z(t) - z_0)^2 \rangle$, with $z_0 = z(t = 0)$, we obtain

$$\langle \delta z^2 \rangle = 2D_0 t - \frac{k_BT}{4\pi \eta} \int_0^\infty dq e^{-2qz_0} (1 + qz_0)^2 \times \left\{ t + \frac{e^{-\omega_q t} - 1}{\omega_q} \right\}. \quad (21)$$

Here, $D_0 = \mu_0 k_BT$ is the bulk diffusion coefficient. This result presents some interesting features. At short times, the MSD behaves like

$$\langle \delta z^2 \rangle \sim 2D_0 t, \quad (22)$$

for $t \ll \tau_m$. In this limit, the particle does not “feel” the membrane. On the other hand, one retrieves for long time-scales the mobility of a particle near a non-deformable liquid interface

$$\langle \delta z^2 \rangle \sim 2D_0 \left( 1 - \frac{15a}{16z_0} \right) t = 2D_\perp t. \quad (23)$$

for $t \gg \tau_m$. In this limit, the MSD is thus independent of the elastic properties of the surface $\kappa$. Both the unexpected short-time and the long-time behaviours can however be explained from the definition of $\tau_m$. As can be noticed from Eq. (19), the limit $t \gg \tau_m$ actually coincides with the limit of an infinitely rigid interface $\kappa \to \infty$. Therefore, one ultimately expects to recover the result for a flat interface when $t \to \infty$. On the other hand, the limit $t \ll \tau_m$ corresponds to a membrane with vanishing bending rigidity $\kappa \to 0$. One can easily check that all the corrections discussed so far actually die out in that.

\footnote{In fact, the long-time diffusion coefficient could also have been obtained from the general relation $D_\perp = k_BT \tilde{\mu}_\perp(\omega = 0)$, compare with Eq. (17).}
limit, so that we recover the bulk diffusion coefficient when \( t \to 0 \).

For intermediate time-scales, one crosses over continuously from the fast to the slow diffusion mode. Defining \( \Delta(t) = (\delta z^2) - 2D_1 t \), we compute numerically and plot in Fig. 1 the excess MSD \( \Delta(t)/\Delta(t \to \infty) \) for different values of the ratio \( d = z_0/\xi_\parallel \). When the particle is far away from the membrane, surface deformations and fluctuations are essentially irrelevant and the particle reaches the “long-time” diffusive regime on a time-scale much shorter than \( \tau_m \). However, as the particle approaches the membrane, the strength of the hydrodynamic coupling between the motion of the particle and the fluctuations of the membrane increases. The effect of the friction kernel on the Brownian motion of the colloids can be seen on much longer time-scales, and the cross-over time increases up to \( \sim \tau_m \) for \( z_0 \sim \xi_\parallel \). As expected, hydrodynamic effects are all the more relevant as the bending rigidity (or the confinement potential) of the membrane are small, corresponding to large values of \( \tau_m \).

As far as the \( x \) and \( y \) directions are concerned, one just has to replace the factor \((1 + qz_0)^2 \) by \( qz_0/2 \) in Eq. (21) to get the corresponding MSD. The results are essentially the same as for the \( z \) direction. In particular, one recovers the diffusion coefficient \( D_\parallel = k_B T \mu_0 (1 - 3a/(32z_0)) \) in the long-time limit for the same reasons as stated above.

Finally, we still have to comment on our approximation that neglects the \( z \)-dependence of the friction coefficient. This assumption is valid as long as \( (\delta z) \ll z_0^2 \), that is for \( t \ll \tau_0 = a z_0^2 \eta/(k_B T) \). Therefore, the results described above can be observed provided that \( \tau_0 > \tau_m \), so that we reach the condition

\[
\frac{k_B T}{\kappa} < \frac{a z_0^2}{\xi_\parallel^3}.
\]

For an initial height \( z_0 = 10 \xi_\parallel \) and a typical bending rigidity \( \kappa = 10 k_B T \), this condition is readily satisfied.

V. DISCUSSION AND CONCLUSION

To summarize, we have shown that the random motion of a colloid near a soft membrane is a non-Markovian process. Indeed, the delay in the response of the elastic interface to hydrodynamic stresses induce memory effects that are relevant over almost three decades in time (see Fig. 1). Consequently, the Brownian motion of a colloidal particle might locally appear anomalous with MSD given by \( (\delta r^2) \sim t^\gamma \). But as explained above, hydrodynamic interactions are not expected to lead to “true” subdiffusive or superdiffusive behaviours since the MSD is always linear in time when \( t \to \infty \).

Actually, anomalous diffusion has been observed in recent fluorescence correlation spectroscopy experiments close to a vesicle [6]. It has been shown that the intensity autocorrelation function is the sum of two contributions. The short time-scale correlations arise from the (regular) diffusion of the particles near an impenetrable wall, whereas the long time-scale correlations come from fluctuations of the membrane itself. The latter create in turn intensity fluctuations by modulating the number of detected particles. The results are interpreted by considering that diffusion is anomalous due to the collisions of the particles with the membrane [6, 26]. This mechanism is however not incompatible with our results: indeed, collisions have been neglected so far as they are not easily included within a Langevin formalism. In order to get a more accurate description of the system, both collisions and hydrodynamic interactions should therefore be taken into account. Note also that the hydrodynamic coupling described in this work is expected to be most pronounced for \( a \sim z_0 \sim \xi_\parallel \). A more careful study, presumably numerical, of these various points is therefore required.

In conclusion, we have investigated the motion of a Brownian particle in the vicinity of a liquid-like membrane. The formalism developed in this work allowed us to accurately account for membrane-induced hydrodynamic interactions. Obviously, this study might be relevant from a biological perspective as diffusive properties of nano-particles near a membrane have been shown to crucially depend on the elastic properties — or equivalently the composition — of the bilayer. In particular, it would be interesting to see whether the predicted memory effects can influence the various kinetic processes taking place near a membrane.

**APPENDIX A: FLUCTUATING HYDRODYNAMICS NEAR A FLUID MEMBRANE**

Thermal undulations of the membrane actually originate from hydrodynamic fluctuations of the embedding fluid. In this appendix, we re-derive the well-known equation of motion for the membrane \[19, 20\] directly from fluctuating hydrodynamic. In Fourier representation, the creeping flow equation satisfied by the flow velocity \( u(k, t) = \int d^2r e^{-i(k \cdot r)}u(r, t) \) is

\[
u(k, t) = \frac{1}{\eta k^2} (I - \hat{k}k) \Phi(k, t) + g(k, t),
\]

with \( I \) the identity tensor, \( \hat{k}k \) the dyadic product, and \( \hat{k} = k/k \). The restoring force density \( \Phi = (0, 0, \Phi_z) \) is given, in Fourier representation, by \( \Phi_z(k, \omega) = -\delta H_m/\delta h^z = -E_q \tilde{h}_q(\omega) \), with the energy density \( E_q = \kappa(q^4 + \xi_q^4) \) and \( q = (k_x, k_y) \). In Eq. (A1), the field \( g(k, t) \) is a stochastic variable characterizing thermal fluctuations in the liquid. Following Ref. 28, it can be shown that the random noise has a Gaussian distribution with zero mean value, \( \langle g(k, t) \rangle = 0 \), and correlations

\[
\langle g_i(k, t)g_j(k', t') \rangle = \frac{2k_B T}{\eta k^2} \delta_{ij} \delta(k - k') (2\pi)^3 \delta(k - k')
\]

(A2)
where the square brackets denote average over an equilibrium ensemble.

In order to derive the equation of motion for the membrane, we still need to express the impermeability condition

$$\frac{\partial h_q}{\partial t} = u_z(q, z = 0, t) = \int_{-\infty}^{\infty} \frac{dk_z}{2\pi} u_z(k, t), \quad (A3)$$

where the velocity $u_z(k, t)$ is given in Eq. (A1). Performing the latter integral, we obtain the Langevin equation describing the relaxation dynamics of a membrane

$$\frac{\partial h_q}{\partial t} - \omega_q h_q + f_q, \quad (A4)$$

with $\omega_q = E_q/(4\eta q)$. The white noise $f_q$ is related to the stochastic variable $g(k, t)$ through $f_q(t) = (2\pi)^{-1} \int dk_z g_z(q, k_z, t)$. Integration of Eq. (A2) shows that its distribution is still Gaussian with zero mean value, $\langle f_q(t) \rangle = 0$, and correlations given by

$$\langle f_q(t) f_{q'}(t') \rangle = \frac{k_B T}{2\eta q} \delta(t - t')(2\pi)^2 \delta(q + q') \quad (A5)$$

Note that these correlations have been obtained after direct integration of the hydrodynamic equations, in the same way as the Langevin equation of a Brownian particle can be found from fluctuating hydrodynamics. In particular, the relaxation of a membrane undulation is characterized by $\langle |h_q(t)|^2 \rangle = \langle |h_q(0)|^2 \rangle \exp[-\omega_q t]$, with equilibrium fluctuation amplitude

$$\langle |h_q(0)|^2 \rangle = \frac{k_B T}{\kappa (q^4 + \xi^4)}, \quad (A6)$$

which is expected from the fluctuation-dissipation theorem.

### APPENDIX B: EVALUATION OF THE GREEN’S FUNCTION

In this appendix, we want to derive the correction to the Oseen tensor coming from hydrodynamic interactions with the membrane. Because the Navier-Stokes equation is linear, the general solution to an arbitrary force field $f$ is given by Eq. (11). In this problem, the embedding medium is anisotropic so that the Oseen tensor $G$ is not translationally invariant in space. The Green function is obtained as the response to a point-force applied at point $r'$ at instant $t$, $f(r, t) = f(t)\delta(r - r')$. After Fourier analysis, the solution of the Navier-Stokes equation together with the incompressibility condition reads

$$\tilde{u}(k, \omega) = \frac{1}{\eta k^2} (\mathbf{I} - \hat{k}\hat{k}) \left\{ \tilde{f}(\omega) e^{-i\mathbf{k}\mathbf{r}'} + \Phi(k, \omega) \right\} \quad (B1)$$

with the same notation as in Appendix A. Yet, the deformation field is coupled to the fluid velocity through the impermeability condition Eq. (10). This condition is more conveniently expressed as $i\omega_h_q(\omega) = \tilde{u}_z(q, 0, \omega)$, where we used the set of variables $\{q, z, \omega\}$. Finally, coming back to the variables $\{k, \omega\}$, we find the following expression for the restoring force density

$$\tilde{\Phi}_z(k, \omega) = i\frac{E_q}{\omega} \int_{-\infty}^{\infty} \frac{dk_z}{2\pi} \tilde{u}_z(k, \omega) \quad (B2)$$

Obviously, this result together with Eq. (B1) gives an integral relation for the velocity field. This can be solved by writing explicitly the equation satisfied by the $z$-component of the velocity. Integrating both side of the equation over $k_z$, we get

$$\int_{-\infty}^{\infty} \frac{dk_z}{2\pi} \tilde{u}_z(k, \omega) = \frac{\exp[-i\mathbf{q}\mathbf{\rho}]}{4\eta q} \frac{\omega}{\omega - i\omega_q} \left[ \left( k_z \tilde{f}_x + k_y \tilde{f}_y \right) i\zeta^z e^{-|\mathbf{q}|^2} + \tilde{f}_z e^{-|\mathbf{q}||z'|} (1 + |q||z'|) \right], \quad (B3)$$

with $\omega_q = E_q/(4\eta q)$ and $\mathbf{\rho}' = (x', y')$. Finally, bringing together the results Eqs. (B1), (B2) and (B3) shows that the velocity field is proportional to the force, $\tilde{u}_z(k, \omega) = \sum_j \tilde{G}_{ij}(k, \omega) \tilde{f}_j(\omega)$. The Oseen tensor can then be written as $\tilde{G} = \tilde{G}_0 + \Delta \tilde{G}$, with the isotropic contribution $\tilde{G}_0(k, \omega) = (\mathbf{I} - \hat{k}\hat{k})/(\eta k^2)$. The explicit expression for the second contribution is given in Eq. (12) in terms of the variables $\{q, z, \omega\}$.

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