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Dynamical fluctuations of spherically closed fluid membranes

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Properties of dynamical shape fluctuations of spherically closed fluid membranes such as vesicles or microemulsion droplets are discussed. As a boundary condition at the interface, we employ the generalized Laplace's formula obtained by Zhong-can and Helfrich. We calculate the oscillation frequencies and the relaxation times of the membranes for a small deformation under the constraint of either constant area or constant volume. Furthermore, the diffusion coefficient of the droplet is estimated from the translational sideways mode. Our result does not depend on the form of the shape energy, in agreement with the recent prediction by Edwards and Schwartz.

1. Introduction

Properties of membranes with high flexibility are of current interest in connection with statistical mechanics of fluctuating surfaces, biophysics of membranes and also high-energy physics [1,2]. Such membranes are typically realized in microemulsion systems being homogeneous mixtures of oil, water and surfactants [3]. Since the surface tension of the membranes is zero or practically zero, the system can contain large internal interfacial areas separating oil and water. Alternatively, the deformation of the surface is governed mainly by the elastic bending energy [4]. Since the associated bending rigidity is known to be the order of $k_B T$, membranes fluctuate due to thermal

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excitations and hence one should handle this problem from the point of view of statistical mechanics.

A similar system is also realized in spherically closed membranes of lipid bilayers called "vesicles" which are typically exemplified by red blood cells. This type of thin-walled fluid vesicles have received great attention as models of cell membranes. Shape transformation among various conformations can be caused by changing, e.g., the osmotic conditions, the temperature or the composition of the lipids [5–7]. These aspects of vesicles might be closely related to the physiological functions of biomembranes. Although vesicles differ from microemulsion droplets by several decades in length scale [8], the ruling physics behind them is expected to be the same.

There have been several dynamical measurements of microemulsion droplets (neutron scattering [9]) and vesicles (fluorescence microscopy [10], video microscopy [11,12], reflection interference contrast microscopy [13]). Along with these experiments, some authors calculated the time correlation function of the out-of-plane displacement for spherically closed fluid membranes. Schneider, Jenkins and Webb were the first ones who obtained this quantity by using the stationary solutions of the Stokes equations which describe the surrounding incompressible fluids [10,14]. They required the so called "stick" boundary conditions and the balance of forces on the surface. Milner and Safran extended their results to the case of non-zero spontaneous curvature [8]. The important assumption in their calculations is that the total area is a conserved quantity as well as the total volume, both for vesicles and microemulsion droplets. In order to incorporate these two constraints simultaneously, they had to introduce the idea of "constant excess area" together with the unknown Lagrange multiplier.

In fact, the appropriate constraint on these systems is still controversial. Van der Linden, Bedeaux and Borkovec insisted that only the area should be kept constant for vesicles, whereas only the volume constraint is necessary for microemulsion droplets since the supply and the loss of surfactants from the bulk phase would take place in a time scale short enough compared to the deformation of droplets [15]. Having focused on the microemulsion droplet case, namely, on the constant volume case, they obtained a dynamical correlation function different from previously mentioned results. Their calculation can be regarded as a generalization of the work by Mellema, Blom and Beekwilder who considered only finite surface tension but took into account the different viscosity inside and outside the droplet [16].

In this paper, we calculate the oscillation frequencies and the relaxation times of a spherically closed fluid membrane whose equilibrium shape is determined by the minimization of the following shape energy [5,7,17,18]:

$$H_{\mathrm{f}} = \tfrac{1}{2}\kappa \oint \mathrm{d}A\,(c_1 + c_2 - c_0)^2 + \Sigma \oint \mathrm{d}A + \Delta P \int \mathrm{d}V\,. \tag{1.1}$$

In the above, $\mathrm{d}A$ and $\mathrm{d}V$ are surface and volume elements, respectively, κ the bending rigidity, c_1 and c_2 are two principal curvatures, c_0 the spontaneous curvature, Σ the surface tension and ΔP the (osmotic) pressure difference

$$\Delta P \equiv P - P'\,, \tag{1.2}$$

measured between outside and inside. (Here and below we shall use the prime in order to distinguish the quantities of the fluid inside from the corresponding quantities of the fluid outside.) The second and third terms either represent the constraints of constant area and volume or actual work.

The first point we would like to stress in this paper is that our results hold regardless of whether area or volume is kept constant. This fact is based on the calculation of shape energy for fluid vesicles by Zhong-can and Helfrich [18]. Hence the present results can be applied both to vesicles and microemulsion droplets in the sense that one of the constraints is incorporated. We consider that simultaneous inclusion of the both constraints needs separate considerations. Secondly, as conditions of balance of forces at the boundary, we employ the generalized Laplace's formula given also by Zhong-can and Helfrich [18]. We believe that this choice is much simpler and straightforward as compared to the previously used boundary conditions [8,10,14,15]. Our calculation differs from that by van der Linden et al. at this stage. Thirdly, we estimate the diffusion coefficient of such a deformable spherically closed membrane according to the translational sideways excitation mode. In contrast to the result by van der Linden et al., our expression does not depend on the surface tension nor on the bending rigidity. This is consistent with the recent papers on the stochastic dynamics of a deformable membrane by Edwards and Schwartz [19,20].

The outline of this article is as follows. First, after some mathematical definitions, the generalized Laplace's formula is extended to the case where the viscous forces are also taken into account. In section 3, we list the expressions of the variations of the shape energy (1.1) up the second order in terms of the out-of-plane displacement. In section 4, simple fluctuating hydrodynamics describing the surrounding fluids are summarized. In section 5, the expression of the oscillation frequencies in the absence of viscous dissipation are derived for small deformations. Although our result is identical to that given by van der Linden et al., we have derived it in a somewhat different manner. In section 6, using the boundary conditions of section 1, we derive the formulae of the relaxation times which are one of the main and new results in this paper. Brownian motion of a droplet is discussed in the last section.

2. Shape equation and boundary condition

First, we will collect some formulas from differential geometry. One can, in general, parameterize a 2-dimensional membrane in 3-dimensional space by two real inner coordinates $s = (s^1, s^2)$. The shape of the membrane is then described by a 3-dimensional vector $\boldsymbol{r} = \boldsymbol{r}(s)$. At each point on the membrane, there are two tangent vectors $\boldsymbol{r}_i = \partial \boldsymbol{r}/\partial s^i$ with $i = 1, 2$. The outward unit normal vector $\hat{\mathbf{n}}$ is perpendicular to these tangent vectors, i.e., $\hat{\mathbf{n}} = (\boldsymbol{r}_1 \times \boldsymbol{r}_2)/|\boldsymbol{r}_1 \times \boldsymbol{r}_2|$.

All properties related to the intrinsic geometry of the membrane are expressed in terms of the metric tensor defined by

$$g_{ij} = \boldsymbol{r}_i \cdot \boldsymbol{r}_j \, . \tag{2.1}$$

Two important quantities are the determinant and the inverse of the metric which will be denoted by

$$g = \det(g_{ij}) \quad \text{and} \quad g^{ij} = (g_{ij})^{-1} \, . \tag{2.2}$$

In addition, one has to consider the (extrinsic) curvature tensor given by

$$h_{ij} = \hat{\mathbf{n}} \cdot \boldsymbol{r}_{ij} = -\hat{\mathbf{n}}_i \cdot \boldsymbol{r}_j \, , \tag{2.3}$$

with $\boldsymbol{r}_{ij} = \partial^2 \boldsymbol{r}/\partial s^i \partial s^j$. Similar to (2.2), the determinant and the inverse of the curvature tensor are denoted by

$$h = \det(h_{ij}) \quad \text{and} \quad h^{ij} = (h_{ij})^{-1} \, . \tag{2.4}$$

The mean curvature H and the Gaussian curvature K are calculated according to

$$H = -\tfrac{1}{2}(c_1 + c_2) = \tfrac{1}{2} g^{ij} h_{ij} \, , \tag{2.5}$$

and

$$K = c_1 c_2 = h/g \, , \tag{2.6}$$

respectively.

At zero temperature, the membrane is supposed to be in the (undeformed) reference state described by $\boldsymbol{r} = \boldsymbol{R}$. Any deformed state of the membrane without any overhangs can then be parameterized by using the normal vector $\hat{\mathrm{N}}$ in the reference state, i.e., $\hat{\mathrm{N}} = (\boldsymbol{R}_1 \times \boldsymbol{R}_2)/|\boldsymbol{R}_1 \times \boldsymbol{R}_2|$, in the following way:

$$\boldsymbol{r} = \boldsymbol{R} + l(s^1, s^2, t)\, \hat{\mathrm{N}} . \tag{2.7}$$

Here the variable $l(s^1, s^2, t)$ represents the transverse (out-of-plane) displacement field which can generally depend on time t. (Here we did not include the in-plane displacements which are irrelevant in the bending energy up to the second order in terms of displacement fields. In-plane displacements should be taken into account in describing polymerized elastic membranes [21].)

By requiring that the first variation of the shape energy (1.1) vanishes for any infinitesimal displacement l, Zhong-can and Helfrich obtained the mechanical equilibrium condition of the membrane vesicle such that [18]

$$\Delta P - 2\Sigma H + \kappa(2H + c_0)(2H^2 - 2K - c_0 H) + 2\kappa \nabla^2_{\mathrm{LB}} H = 0 , \tag{2.8}$$

where ∇^2_{LB} is the Laplace–Beltrami operator on the surface defined by

$$\nabla^2_{\mathrm{LB}} = \frac{1}{\sqrt{g}}\, \partial_i (g^{ij} \sqrt{g}\, \partial_j) . \tag{2.9}$$

(2.8) represents the balance of normal forces per unit membrane area and reduces to the well-known Laplace's formula when $\kappa = 0$. (Recently a more general case has been obtained by Onuki and Kawasaki [22].)

In order to construct the boundary condition that must be satisfied at the interface between two viscous fluids in motion, it is natural and straightforward to extend (2.8) in accordance with Landau and Lifshitz [23]. Hence in the presence of both surface tension and bending rigidity, the equality of the forces on the surface of each fluid can be written as

$$(P - P')\mathrm{n}_\alpha - (\sigma_{\alpha\beta} - \sigma'_{\alpha\beta})\mathrm{n}_\beta + [-2\Sigma H + \kappa(2H + c_0)(2H^2 - 2K - c_0 H) + 2\kappa \nabla^2_{\mathrm{LB}} H]\mathrm{n}_\alpha = 0 , \tag{2.10}$$

where n_α are components of the normal vector and $\sigma_{\alpha\beta}$ is the viscous stress tensor given in section 4. (We shall use Greek indices for the range 1, 2 and 3.) (2.10) will be used in sections 5 and 6.

3. Spherically closed fluid membranes

Now, consider a spherically closed fluid membrane of radius r_0 with internal coordinates $(s^1, s^2) = (\theta, \phi)$. As a local basis of 3-dimensional space, we introduce three unit vectors:

$$\hat{e}_r = \begin{pmatrix} \sin\theta\cos\phi \\ \sin\theta\sin\phi \\ \cos\theta \end{pmatrix}, \qquad \hat{e}_\theta = \begin{pmatrix} \cos\theta\cos\phi \\ \cos\theta\sin\phi \\ -\sin\theta \end{pmatrix}, \qquad \hat{e}_\phi = \begin{pmatrix} -\sin\phi \\ \cos\phi \\ 0 \end{pmatrix}. \tag{3.1}$$

With these notations, the reference state is described by

$$\boldsymbol{R} = r_0 \hat{e}_r , \tag{3.2}$$

and a slightly deformed sphere can be represented by (see (2.7))

$$\boldsymbol{r} = \boldsymbol{R} + l(\theta, \phi, t)\,\hat{\mathbf{N}} = [r_0 + l(\theta, \phi, t)]\hat{e}_r . \tag{3.3}$$

The undeformed reference sphere (3.2) is always a solution of the equilibrium shape condition (2.8) if the next relation holds among the parameters [18]:

$$\Delta P\, r_0^3 + 2\Sigma r_0^2 + \kappa c_0 r_0 (c_0 r_0 - 2) = 0 . \tag{3.4}$$

This relation is called the "capillarity condition" [15].

When a spherical membrane deforms slightly without any overhangs according to (3.3), a straightforward calculation up to the first order in terms of the out-of-plane displacement l yields the following expression for the normal vector:

$$\hat{\mathbf{n}} \approx \hat{e}_r - \frac{1}{r_0}\frac{\partial l}{\partial \theta}\hat{e}_\theta - \frac{1}{r_0 \sin\theta}\frac{\partial l}{\partial \phi}\hat{e}_\phi , \tag{3.5}$$

and for the mean curvature H and the Gaussian curvature K:

$$2H \approx -\frac{2}{r_0} + \frac{1}{r_0^2}(2 + \nabla_\perp^2) l(\theta, \phi, t) , \tag{3.6}$$

and

$$K \approx \frac{1}{r_0^2} - \frac{1}{r_0^3}(2 + \nabla_\perp^2) l(\theta, \phi, t) , \tag{3.7}$$

respectively, where

$$\nabla_{\perp}^2 = \frac{1}{\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{\partial}{\partial\theta}\right) + \frac{1}{\sin^2\theta}\frac{\partial^2}{\partial\phi^2}\,. \tag{3.8}$$

It is convenient to expand the function $l(\theta, \phi, t)$ in terms of the spherical harmonics $Y_{nm}(\theta, \phi)$,

$$l(\theta, \phi, t) = \sum_{n,m} l_{nm}(t)\, Y_{nm}(\theta, \phi)\,. \tag{3.9}$$

As usual, we have $l^*_{nm}(t) = (-1)^m l_{n,-m}(t)$ in order to ensure that the displacement field is real (the asterisk denotes the complex conjugate value) and the summation is over $n = 0, 1, 2, \ldots$ and $|m| \leq n$. Hereafter, the well-known relation

$$\nabla_{\perp}^2 Y_{nm}(\theta, \phi) = -n(n+1) Y_{nm}(\theta, \phi) \tag{3.10}$$

will be used frequently.

So far, the variations of the bending energy H_b, the area A and the volume V for spherical parameterization have been calculated by several authors [8,10,18,24,25]. Up to the second order in terms of l_{nm}, they can be summarized as

$$\begin{aligned}\delta H_b \approx{}& \sqrt{4\pi}\,\kappa c_0 r_0 (c_0 r_0 - 2)\frac{l_{00}}{r_0}\\ &+ \tfrac{1}{2}\kappa \sum_{n,m} \{[n(n+1)]^2 - (2 + 2c_0 r_0 - \tfrac{1}{2}c_0^2 r_0^2) n(n+1) + c_0^2 r_0^2\}\frac{|l_{nm}|^2}{r_0^2}\,,\end{aligned} \tag{3.11}$$

$$\delta \oint \mathrm{d}A \approx 2\sqrt{4\pi}\, r_0 l_{00} + \sum_{n,m} [1 + \tfrac{1}{2}n(n+1)]|l_{nm}|^2\,, \tag{3.12}$$

and

$$\delta \int \mathrm{d}V \approx \sqrt{4\pi}\, r_0^2 l_{00} + r_0 \sum_{n,m} |l_{nm}|^2\,, \tag{3.13}$$

respectively. Upon adding all these expressions in accordance with the shape energy (1.1), the total second variation turns out to be

$$\begin{aligned}\delta H_f &= \delta H_b + \Sigma\,\delta \oint \mathrm{d}A + \Delta P\,\delta \int \mathrm{d}V\\ &\approx \tfrac{1}{2} \sum_{n,m} (n-1)(n+2)\{\kappa[n(n+1) - c_0 r_0] - \tfrac{1}{2}\Delta P\, r_0^3\}\frac{|l_{nm}|^2}{r_0^2}\\ &\approx \tfrac{1}{2} \sum_{n,m} (n-1)(n+2)\{\Sigma r_0^2 + \kappa[n(n+1) - 2c_0 r_0 + \tfrac{1}{2}c_0^2 r_0^2]\}\frac{|l_{nm}|^2}{r_0^2}\,.\end{aligned} \tag{3.14}$$

Here the capillarity condition (3.4) has been used to obtain the last identity and all the first order terms have cancelled each other as they should. Since (3.14) depends only on n but not on m, the shape energy has $(2n+1)$-fold degeneracy. Notice that we have not incorporated any constraints at this stage.

Next we consider the case where either the area or the volume is kept constant. The constraint of constant volume can be easily incorporated by using the expression (3.13) for the volume. It then follows from $\delta V=0$ that

$$\sqrt{4\pi}\, l_{00} \approx -\sum_{n,m} \frac{|l_{nm}|^2}{r_0} . \tag{3.15}$$

Hence the constant volume constraint leads to the elimination of the l_{00}-terms. Inserting this into (3.11) and (3.12) and forming the variation

$$\delta H_\mathrm{f} = \delta H_\mathrm{b} + \Sigma\, \delta \oint \mathrm{d}A , \tag{3.16}$$

we obtain

$$\delta H_\mathrm{f} \approx \tfrac{1}{2} \sum_{n,m}{}' (n-1)(n+2)\{\Sigma r_0^2 + \kappa[n(n+1) - 2c_0 r_0 + \tfrac{1}{2}c_0^2 r_0^2]\} \frac{|l_{nm}|^2}{r_0^2} , \tag{3.17}$$

where the only change compared to (3.14) is the prime on the summation indicating that $(n, m) = (0, 0)$ is excluded. This expression is identical to that by van der Linden et al. [15] (our spontaneous curvature differs in sign).

Likewise, the constraint of constant area can be incorporated by requiring $\delta A = 0$, namely,

$$\sqrt{4\pi}\, r_0 l_{00} \approx -\tfrac{1}{2} \sum_{n,m} [1 + \tfrac{1}{2}n(n+1)]|l_{nm}|^2 . \tag{3.18}$$

By eliminating the l_{00}-terms, one finds

$$\delta H_\mathrm{f} = \delta H_\mathrm{b} + \Delta P\, \delta \int \mathrm{d}V \tag{3.19}$$

is identical to (3.17) again. Therefore (3.17) holds generally regardless of whether volume or area is kept constant. In the following sections, we shall consider the case (3.17) where one of the constraints is included. Although we use the word "microemulsion droplets" in the following, the readers are advised to replace it also by "vesicles".

4. Hydrodynamic equations

In describing the motion of the surrounding fluids, we assumed that both the inside and the outside fluids are incompressible. The basic hydrodynamical equations of mass conservation (continuity equation) and momentum conservation (Navier–Stokes equation) are

$$\operatorname{div} \boldsymbol{v} = 0 , \tag{4.1}$$

$$\rho\left(\frac{\partial \boldsymbol{v}}{\partial t} + (\boldsymbol{v} \cdot \operatorname{grad})\boldsymbol{v} \right) = -\operatorname{grad} P + \eta \nabla^2 \boldsymbol{v} , \tag{4.2}$$

respectively. Here ρ denotes the density of the fluid, $\boldsymbol{v}$ the velocity, η the dynamic viscosity. In (4.2), we have used the fact that the viscous stress tensor for an incompressible fluid is

$$\sigma_{\alpha\beta} = \eta\left(\frac{\partial v_\alpha}{\partial x_\beta} + \frac{\partial v_\beta}{\partial x_\alpha} \right) . \tag{4.3}$$

(4.3) will be used later to apply the boundary condition (2.10).

When we are interested only in the fluctuating properties, the above equations can be linearized by putting

$$\rho = \rho_0 + \delta\rho , \qquad P = P_0 + \delta P , \qquad \boldsymbol{v} = \delta \boldsymbol{v} , \tag{4.4}$$

where ρ_0, P_0 stand for the values in equilibrium and $\delta\rho, \delta P, \delta\boldsymbol{v}$ are their variations. Retaining in (4.2) only up to the first order terms, we obtain

$$\rho_0 \frac{\partial \boldsymbol{v}}{\partial t} = -\operatorname{grad} \delta P + \eta \nabla^2 \boldsymbol{v} . \tag{4.5}$$

In the above equations, it is convenient to decompose the velocity field into irrotational (longitudinal) and rotational (transverse) parts according to

$$\boldsymbol{v} = \boldsymbol{v}^\ell + \boldsymbol{v}^\mathrm{t} , \tag{4.6}$$

$$\operatorname{div} \boldsymbol{v}^\mathrm{t} = 0 , \qquad \operatorname{rot} \boldsymbol{v}^\ell = \boldsymbol{0} . \tag{4.7}$$

Obviously, only the longitudinal part remains in (4.1);

$$\operatorname{div} \boldsymbol{v}^\ell = 0 , \tag{4.8}$$

while (4.5) splits into two equations:

$$\frac{\partial \boldsymbol{v}^{\mathrm{t}}}{\partial t} = \frac{\eta}{\rho_0} \nabla^2 \boldsymbol{v}^{\mathrm{t}} , \tag{4.9}$$

$$\rho_0 \frac{\partial \boldsymbol{v}^{\ell}}{\partial t} = -\mathrm{grad}\, \delta P . \tag{4.10}$$

Hereafter we will be concerned only with the longitudinal part of the velocity $\boldsymbol{v}^{\ell}$ which will be denoted simply by $\boldsymbol{v}$.

Like any vector field having zero rotation, it is advantageous to introduce the velocity potential ψ which provides the velocity $\boldsymbol{v}$ through

$$\boldsymbol{v} = \mathrm{grad}\, \psi . \tag{4.11}$$

Substituting (4.11) into (4.8) and (4.10) yields

$$\nabla^2 \psi = 0 , \tag{4.12}$$

$$\delta P = -\rho_0 \frac{\partial \psi}{\partial t} , \tag{4.13}$$

respectively, where the constant term has been neglected in (4.13). Upon neglecting the inertial term in (4.2), the incompressible condition (4.1) implies that the pressure field also satisfies Laplace's equation

$$\nabla^2 \delta P = 0 . \tag{4.14}$$

5. Undamped oscillations

In this section, we determine the characteristic oscillation frequency of a spherically closed membrane in the absence of any viscous damping effect. The system is regarded as an infinite set of uncoupled harmonic oscillators; each oscillator identified by the set of (n, m). Then the equation of motion of the out-of-plane displacement $l_{nm}(t)$ becomes [15]

$$\ddot{l}_{nm}(t) = -\omega_{nm}^2 l_{nm}(t) , \tag{5.1}$$

where the dot indicates the derivative with respect to time. Our purpose is to express the oscillation frequency ω_{nm} in terms of surface tension and bending rigidity. In view of (5.1), we write $l_{nm}(t) = l_{nm}\, \mathrm{e}^{\mathrm{i}\omega_{nm} t}$ in (3.9), yielding

$$l(\theta, \phi, t) = \sum_{n,m}{}' l_{nm} Y_{nm}(\theta, \phi)\, \mathrm{e}^{\mathrm{i}\omega_{nm} t} . \tag{5.2}$$

(As mentioned in section 3, we exclude $(n, m) = (0, 0)$ due to the presence of the constraint.)

Since both inside and outside fluids are assumed to be incompressible, the respective velocity potentials satisfy Laplace's equation (4.12). Hence, as given by Lamb [15, 26], they can be expanded in terms of the solid spherical harmonics:

$$\psi'(r, t) = \sum_{n,m}{}' \psi'_{nm}\left(\frac{r}{r_0}\right)^n Y_{nm}(\theta, \phi)\, \mathrm{e}^{\mathrm{i}\omega_{nm}t}\,, \tag{5.3}$$

$$\psi(r, t) = \sum_{n,m}{}' \psi_{nm}\left(\frac{r_0}{r}\right)^{n+1} Y_{nm}(\theta, \phi)\, \mathrm{e}^{\mathrm{i}\omega_{nm}t}\,. \tag{5.4}$$

In the absence of a dissipation mechanism due to the viscous damping effect, the so called "slip" boundary condition is employed here [15]. This condition requires the continuity of the radial component of the velocity, i.e., $v_r = \partial\psi/\partial r = \partial l/\partial t$ at the boundary. Since the velocity field is linear in the displacement amplitude l, one can impose the boundary condition at $r = r_0$. Accordingly, the first boundary condition follows from (5.3) and (5.4) as

$$\begin{aligned} v_r(r = r_0) &= \frac{1}{r_0} \sum_{n,m}{}' n\psi'_{nm} Y_{nm}(\theta, \phi)\, \mathrm{e}^{\mathrm{i}\omega_{nm}t} \\ &= -\frac{1}{r_0} \sum_{n,m}{}' (n+1)\psi_{nm} Y_{nm}(\theta, \phi)\, \mathrm{e}^{\mathrm{i}\omega_{nm}t} \\ &= \sum_{n,m}{}' \mathrm{i}\omega_{nm} l_{nm} Y_{nm}(\theta, \phi)\, \mathrm{e}^{\mathrm{i}\omega_{nm}t}\,, \end{aligned} \tag{5.5}$$

where we took the time derivative of (5.2) to obtain the last equation. By comparing the coefficients of the spherical harmonics, one finds

$$n\psi'_{nm} = -(n+1)\psi_{nm} = \mathrm{i} r_0 \omega_{nm} l_{nm}\,. \tag{5.6}$$

The second boundary conditions stems from the balance of forces on the surface. In the absence of viscous forces, the boundary condition (2.10) reduces to the generalized Laplace's formula (2.8). By noticing that the Laplace–Beltrami operator is now the usual Laplacian operator on the sphere

$$\nabla^2_{\mathrm{LB}} = \frac{\nabla^2_{\perp}}{r_0^2}\,, \tag{5.7}$$

and substituting (3.6) and (3.7) into (2.8), the force balance for the radial part up to the first order in terms of $l(\theta, \phi, t)$ turns out to be

$$\delta P - \delta P' - \frac{1}{r_0^4}\,[\Sigma r_0^2 - \kappa(\nabla_\perp^2 + 2c_0 r_0 - \tfrac{1}{2}c_0^2 r_0^2)](2 + \nabla_\perp^2)l(\theta, \phi, t) = 0\,, \quad (5.8)$$

at $r = r_0$. In the above, we used the fact that $n_r \approx 1$ and all the constant terms have cancelled each other because of the capillarity condition (3.4). If we replace δP and $\delta P'$ with the use of (4.13), one obtains

$$\rho_0 \frac{\partial \psi}{\partial t} - \rho_0' \frac{\partial \psi'}{\partial t} + \frac{1}{r_0^4}\,[\Sigma r_0^2 - \kappa(\nabla_\perp^2 + 2c_0 r_0 - \tfrac{1}{2}c_0^2 r_0^2)](2 + \nabla_\perp^2)l(\theta, \phi, t) = 0\,, \quad (5.9)$$

at $r = r_0$. By substituting (5.2)–(5.4) into (5.9) and comparing the coefficients of the spherical harmonics as before, we have

$$\begin{aligned} &\mathrm{i}\rho_0 \omega_{nm} \psi_{nm} - \mathrm{i}\rho_0' \omega_{nm} \psi'_{nm} \\ &\quad = \frac{1}{r_0^4}\,(n-1)(n+2)\{\Sigma r_0^2 + \kappa[n(n+1) - 2c_0 r_0 + \tfrac{1}{2}c_0^2 r_0^2]\}\,l_{nm}\,. \end{aligned} \quad (5.10)$$

(5.6) and (5.10) are sufficient to obtain the oscillation frequency ω_{nm} which is

$$\omega_{nm}^2 = \frac{1}{\rho_n r_0^5}\,(n-1)(n+2)\{\Sigma r_0^2 + \kappa[n(n+1) - 2c_0 r_0 + \tfrac{1}{2}c_0^2 r_0^2]\}\,, \quad (5.11)$$

with

$$\rho_n = \frac{\rho_0'}{n} + \frac{\rho_0}{n+1}\,. \quad (5.12)$$

When $\kappa = 0$, (5.11) reduces to the old expression first given by Rayleigh a long time ago [27]. (5.11) is also identical to the result by van der Linden, Bedeaux and Borkovec [15].

Since the expression (5.11) vanishes for $n = 1$, the smallest possible frequency of oscillations corresponds to $n = 2$, and is

$$\omega_{\min} = \sqrt{\frac{4}{(\rho_0'/2 + \rho_0/3)r_0^5}\,[\Sigma r_0^2 + \kappa(6 - 2c_0 r_0 + \tfrac{1}{2}c_0^2 r_0^2)]}\,. \quad (5.13)$$

Furthermore, the total variation of the shape energy (3.17) can be simply expressed in terms of ω_{nm} as

$$\delta H_{\mathrm{f}} = \frac{1}{2} \sum_{n,m}{}' \rho_n r_0^3 \omega_{nm}^2 |l_{nm}|^2 . \tag{5.14}$$

With the use of equipartition theorem (or straightforward Gaussian integrations), the fluctuation amplitudes are easily estimated:

$$\begin{aligned}\langle |l_{nm}|^2 \rangle &= \frac{k_{\mathrm{B}} T}{\rho_n r_0^3 \omega_{nm}^2} \\ &= \frac{k_{\mathrm{B}} T r_0^2}{(n-1)(n+2)\{\Sigma r_0^2 + \kappa[n(n+1) - 2c_0 r_0 + c_0^2 r_0^2/2]\}} ,\end{aligned} \tag{5.15}$$

where k_{B} is the Boltzmann constant and T is the temperature. It is important to realize that (5.15) is valid only for $n \geqslant 2$, since $n = 1$ corresponds to the simple translational sideways displacement of the droplet as a whole requiring no energy. This mode is essentially related to the Brownian motion of the droplet and will be discussed in detail in the last section.

6. Overdamped oscillations

In the previous section, the frictional forces due to the viscous damping effect have been neglected. The inclusion of viscous forces introduces the damping term in the equation of motion (5.1) in such a way that [15]

$$\ddot{l}_{nm}(t) = -\gamma_{nm} \dot{l}_{nm}(t) - \omega_{nm}^2 l_{nm}(t) . \tag{6.1}$$

In this section, we consider the case where the motion of the membrane surface is overdamped. Then the restoring forces of the membrane balance with the viscous resistance forces due to the surrounding fluid after a short initial period of motion. In such a case, (6.1) reduces to

$$\dot{l}_{nm}(t) = -\frac{1}{\tau_{nm}} l_{nm}(t) , \tag{6.2}$$

where

$$\frac{1}{\tau_{nm}} = \frac{\omega_{nm}^2}{\gamma_{nm}} . \tag{6.3}$$

From (6.2), the time dependence of $l_{nm}(t)$ is now given by $l_{nm}(t) = l_{nm}\, e^{-t/\tau_{nm}}$. This leads to the expansion of $l(\theta, \phi, t)$ in the form of

$$l(\theta, \phi, t) = \sum_{n,m}{}' l_{nm} Y_{nm}(\theta, \phi)\, e^{-t/\tau_{nm}} . \tag{6.4}$$

By using the results of the fluctuation amplitudes (5.15), the time correlation function is simply provided by

$$\begin{aligned}\langle l_{nm}(t)\, l_{nm}(0)^* \rangle &= \frac{k_B T}{\rho_n r_0^3 \omega_{nm}^2}\, e^{-t/\tau_{nm}} \\ &= \frac{k_B T r_0^2\, e^{-t/\tau_{nm}}}{(n-1)(n+2)\{\Sigma r_0^2 + \kappa[n(n+1) - 2c_0 r_0 + c_0^2 r_0^2/2]\}} .\end{aligned} \tag{6.5}$$

Upon taking the viscous effect into account, the motion of the surrounding fluid is described by the "creeping flow" neglecting the inertial term at low Reynolds number (Stokes equation),

$$\eta \nabla^2 \boldsymbol{v} = \operatorname{grad} P , \tag{6.6}$$

together with the incompressibility condition

$$\operatorname{div} \boldsymbol{v} = 0 . \tag{6.7}$$

As has been already discussed by several authors [8,10,15], we are interested in the irrotational solutions, which simplifies the problem to some extent. The solution of (6.6) and (6.7) in spherical coordinates can be constructed by the two scalar functions describing inside the fluid, $\psi'(\boldsymbol{r}, t)$, $P'(\boldsymbol{r}, t)$, and by those outside the fluid, $\psi(\boldsymbol{r}, t)$, $P(\boldsymbol{r}, t)$. Since all of these functions satisfy Laplace's equation (see (4.12) and (4.14)), they can be expanded in terms of solid spherical harmonics as before:

$$\psi'(\boldsymbol{r}, t) = \sum_{n,m}{}' \psi'_{nm} \left(\frac{r}{r_0}\right)^n Y_{nm}(\theta, \phi)\, e^{-t/\tau_{nm}} , \tag{6.8}$$

$$P'(\boldsymbol{r}, t) = \sum_{n,m} P'_{nm} \left(\frac{r}{r_0}\right)^n Y_{nm}(\theta, \phi)\, e^{-t/\tau_{nm}} , \tag{6.9}$$

and

$$\psi(\boldsymbol{r}, t) = \sum_{n,m}{}' \psi_{nm} \left(\frac{r_0}{r}\right)^{n+1} Y_{nm}(\theta, \phi)\, e^{-t/\tau_{nm}} , \tag{6.10}$$

$$P(r, t) = \sum_{n,m} P_{nm}\left(\frac{r_0}{r}\right)^{n+1} Y_{nm}(\theta, \phi)\, \mathrm{e}^{-t/\tau_{nm}} . \tag{6.11}$$

(For the pressure fields P' and P, we include $(n, m) = (0, 0)$ terms because of the presence of the non-fluctuating hydrostatic pressure.)

According to the general stationary solution of the Stokes equation given by Lamb [26], the velocity field inside of the droplet is written as [28]

$$\begin{aligned} \boldsymbol{v}'(\boldsymbol{r}, t) = {\sum_{n,m}}' \bigg(\psi'_{nm} \operatorname{grad} &+ \frac{n+3}{2\eta'(n+1)(2n+3)} P'_{nm} r^2 \operatorname{grad} \\ &- \frac{n}{\eta'(n+1)(2n+3)} P'_{nm} \boldsymbol{r}\bigg)\left(\frac{r}{r_0}\right)^n Y_{nm}(\theta, \phi)\, \mathrm{e}^{-t/\tau_{nm}} , \end{aligned} \tag{6.12}$$

whereas the corresponding solution for the outside is provided by

$$\begin{aligned} \boldsymbol{v}(\boldsymbol{r}, t) = {\sum_{n,m}}' \bigg(\psi_{nm} \operatorname{grad} &- \frac{n-2}{2\eta n(2n-1)} P_{nm} r^2 \operatorname{grad} \\ &+ \frac{n+1}{\eta n(2n-1)} P_{nm} \boldsymbol{r}\bigg)\left(\frac{r_0}{r}\right)^{n+1} Y_{nm}(\theta, \phi)\, \mathrm{e}^{-t/\tau_{nm}} . \end{aligned} \tag{6.13}$$

In (6.12) and (6.13), the gradient operators also act on the solid spherical harmonics outside the square bracket. For the purpose of writing down the boundary conditions explicitly, we should keep in mind that the gradient operator in spherical coordinates takes the form

$$\operatorname{grad} = \hat{\boldsymbol{e}}_r \frac{\partial}{\partial r} + \hat{\boldsymbol{e}}_\theta \frac{1}{r} \frac{\partial}{\partial \theta} + \hat{\boldsymbol{e}}_\phi \frac{1}{r \sin\theta} \frac{\partial}{\partial \phi} . \tag{6.14}$$

In contrast to the previous section, the so called "stick" boundary condition is used here as in van der Linden et al. [15]. This condition requires that both the velocity of the membrane and the velocity of the fluid on each side of the membrane are equal to $\partial r/\partial t$ (see (3.3)). The first two boundary conditions come from the continuity of the velocity field. In the $\hat{e}_r$-direction, this is written as

$$\begin{aligned} v_r(r = r_0) &= \frac{1}{r_0} {\sum_{n,m}}' \left(n\psi'_{nm} + \frac{n}{2\eta'(2n+3)} r_0^2 P'_{nm}\right) Y_{nm}(\theta, \phi)\, \mathrm{e}^{-t/\tau_{nm}} \\ &= -\frac{1}{r_0} {\sum_{n,m}}' \left((n+1)\psi_{nm} - \frac{n+1}{2\eta(2n-1)} r_0^2 P_{nm}\right) Y_{nm}(\theta, \phi)\, \mathrm{e}^{-t/\tau_{nm}} \\ &= {\sum_{n,m}}' \frac{-1}{\tau_{nm}} l_{nm} Y_{nm}(\theta, \phi)\, \mathrm{e}^{-t/\tau_{nm}} , \end{aligned} \tag{6.15}$$

where the last equation has been obtained by taking the time derivative of (6.4). Likewise, the continuity of the velocity in the $\hat{e}_\theta$-direction yields

$$v_\theta(r=r_0) = \frac{1}{r_0} \sum_{n,m}' \left(\psi'_{nm} + \frac{n+3}{2\eta'(n+1)(2n+3)} r_0^2 P'_{nm} \right) \frac{\partial Y_{nm}}{\partial \theta} \mathrm{e}^{-t/\tau_{nm}}$$

$$= \frac{1}{r_0} \sum_{n,m}' \left(\psi_{nm} - \frac{n-2}{2\eta n(2n-1)} r_0^2 P_{nm} \right) \frac{\partial Y_{nm}}{\partial \theta} \mathrm{e}^{-t/\tau_{nm}} . \tag{6.16}$$

The continuity condition in the $\hat{e}_\phi$-direction results in the same condition as (6.16). From (6.15) and (6.16), we obtain the following relations among coefficients of the spherical harmonics:

$$n\psi'_{nm} + \frac{n}{2\eta'(2n+3)} r_0^2 P'_{nm} = -(n+1)\psi_{nm} + \frac{n+1}{2\eta(2n-1)} r_0^2 P_{nm}$$

$$= -\frac{r_0}{\tau_{nm}} l_{nm} \tag{6.17}$$

and

$$\psi'_{nm} + \frac{n+3}{2\eta'(n+1)(2n+3)} r_0^2 P'_{nm} = \psi_{nm} - \frac{n-2}{2\eta n(2n-1)} r_0^2 P_{nm} , \tag{6.18}$$

at $r = r_0$.

Additional sets of boundary conditions follow from the balance of forces on the membrane. Up to the first order in terms of the amplitudes l, the force balance in the $\hat{e}_r$-direction is given by

$$(\delta P - \delta P') - \left(2\eta \frac{\partial v_r}{\partial r} - 2\eta' \frac{\partial v'_r}{\partial r} \right)$$

$$- \frac{1}{r_0^4} [\Sigma r_0^2 - \kappa(\nabla_\perp^2 + 2c_0 r_0 - \tfrac{1}{2} c_0^2 r_0^2)](2 + \nabla_\perp^2) l(\theta, \phi, t) = 0 , \tag{6.19}$$

at $r = r_0$. Like in eq. (5.8), the fact that $\mathrm{n}_r \approx 1$ and the capillarity condition (3.4) has been used. In fact, (6.19) differs from (5.8) only due to the viscous terms. Likewise, the force balance in the $\hat{e}_\theta$-direction yields

$$(P_0 - P'_0)\mathrm{n}_\theta - \left[\eta \left(\frac{1}{r} \frac{\partial v_r}{\partial \theta} + \frac{\partial v_\theta}{\partial r} - \frac{v_\theta}{r} \right) - \eta' \left(\frac{1}{r} \frac{\partial v'_r}{\partial \theta} + \frac{\partial v'_\theta}{\partial r} - \frac{v'_\theta}{r} \right) \right]$$

$$+ \left(\frac{2\Sigma}{r_0} - \frac{\kappa c_0}{r_0^2} (2 - c_0 r_0) \right) \mathrm{n}_\theta = 0 , \tag{6.20}$$

at $r = r_0$. Notice that $\mathrm{n}_\theta \approx -(1/r_0)(\partial l/\partial \theta)$ which is linear in l. Again the terms proportional to n_θ cancel each other because of the capillarity condition (3.4). After substituting the expressions for the velocity and the pressure fields into (6.19) and (6.20), the relations between the coefficients for $n \geqslant 1$ are given by

$$P'_{nm} - \left(2\eta' n(n-1)\frac{\psi'_{nm}}{r_0^2} + \frac{n(n+1)}{2n+3}P'_{nm}\right)$$
$$- P_{nm} + \left(2\eta(n+1)(n+2)\frac{\psi_{nm}}{r_0^2} - \frac{n(n+1)}{2n-1}P_{nm}\right)$$
$$= \frac{1}{r_0^4}(n-1)(n+2)\{\Sigma r_0^2 + \kappa[n(n+1) - 2c_0 r_0 + \tfrac{1}{2}c_0^2 r_0^2]\} l_{nm}\,, \tag{6.21}$$

and

$$2\eta'(n-1)\frac{\psi'_{nm}}{r_0^2} + \frac{n(n+2)}{(n+1)(2n+3)}P'_{nm}$$
$$+ 2\eta(n+2)\frac{\psi_{nm}}{r_0^2} - \frac{(n-1)(n+1)}{n(2n-1)}P_{nm} = 0\,. \tag{6.22}$$

Combining (6.17), (6.18), (6.21) and (6.22), one can express ψ'_{nm}, P'_{nm}, ψ_{nm} and P_{nm} in terms of l_{nm}. After some calculations, the relaxation time τ_{nm} in (6.2) turns out to be

$$\frac{1}{\tau_{nm}} = \frac{1}{\eta r_0^3}\,\frac{(n-1)n(n+1)(n+2)(2n+1)(E+1)}{[(2n^2+4n+3)E + 2n(n+2)][2(n^2-1)E + 2n^2+1]}$$
$$\times \{\Sigma r_0^2 + \kappa[n(n+1) - 2c_0 r_0 + \tfrac{1}{2}c_0^2 r_0^2]\}\,, \tag{6.23}$$

with $E \equiv \eta'/\eta$. For $\kappa = 0$, the result (6.23) reduces to that given by Mellma, Blom and Beekwilder [16]. This relaxation time is quite similar to that obtained by Lisy [29], at least concerning the dependence on n, but it still differs in the prefactor.

Since ω_{nm} has already been obtained in the previous section, one can immediately obtain the frictional coefficient γ_{nm} via (6.3) as

$$\gamma_{nm} = \tau_{nm}\omega_{nm}^2$$
$$= \frac{\eta}{\rho_n r_0^2}\,\frac{[(2n^2+4n+3)E + 2n(n+2)][2(n^2-1)E + 2n^2+1]}{n(n+1)(2n+1)(E+1)}\,. \tag{6.24}$$

It is important to note that our result for the frictional coefficient depends only on the properties of the surrounding fluids but not on the membrane properties, such as the surface tension or the bending rigidity. This point is in sharp contrast to the result by van der Linden et al. [15].

7. Diffusion coefficient

In this section, we estimate the diffusion coefficient of a deformable droplet. The simplest way is to use the expression of the frictional coefficient with respect to the translational motion identified with the $n = 1$ mode. Since (6.24) corresponds to the frictional coefficient per unit mass, the total frictional coefficient Γ can be effectively obtained by [15]

$$\Gamma = (\tfrac{4}{3}\pi\rho_{n=1}r_0^3)\gamma_{n=1,m} = 2\pi\eta r_0 \frac{3E+2}{E+1} . \tag{7.1}$$

Hence the diffusion coefficient D can be calculated through Einstein's relation as

$$D = \frac{k_{\mathrm{B}}T}{\Gamma} = \frac{k_{\mathrm{B}}T}{2\pi\eta r_0} \frac{\eta + \eta'}{2\eta + 3\eta'} , \tag{7.2}$$

which coincides with the old result by Hadamard [30] and Rybczynski [31], who did not take into account the deformation of the droplet. When $\eta' = \eta$, (7.2) reduces to

$$D = \frac{k_{\mathrm{B}}T}{5\pi\eta r_0} . \tag{7.3}$$

As discussed by Landau and Lifshitz [23], in the case of $\eta' \to \infty$ (corresponding to a solid sphere), (7.2) becomes the well-known Stokes formula

$$D = \frac{k_{\mathrm{B}}T}{6\pi\eta r_0} . \tag{7.4}$$

In the opposite limit $\eta' \to 0$ (corresponding to a gas bubble), (7.2) reduces to

$$D = \frac{k_{\mathrm{B}}T}{4\pi\eta r_0} . \tag{7.5}$$

In view of (7.2), one can conclude that the form of the shape energy is irrelevant to the diffusion coefficient as far as the deformation of the droplet is small enough. A more general case has been recently discussed by Edwards and Schwartz in their theory of stochastic dynamics of a deformable membrane

[19,20]. In fact, (7.2) can be also derived by following their arguments. In the absence of thermal agitation, the equation for $\dot{l}_{nm}$ should be of the following form:

$$\dot{l}_{nm} = -K_n \frac{\partial \delta H_f}{\partial l_{n,-m}}$$
$$= -K_n(n-1)(n+2)\{\Sigma r_0^2 + \kappa[n(n+1) - 2c_0 r_0 + \tfrac{1}{2}c_0^2 r_0^2]\} \frac{l_{nm}}{r_0^2} . \tag{7.6}$$

Combining this result with our result of τ_{nm} (see (6.23)), one concludes that

$$K_n = \frac{1}{\eta r_0} \frac{n(n+1)(2n+1)(E+1)}{[(2n^2+4n+3)E + 2n(n+2)][2(n^2-1)E + 2n^2 + 1]} . \tag{7.7}$$

When $E = 1$ ($\eta' = \eta$), (7.7) reduces to

$$K_n = \frac{2}{\eta r_0} \frac{n(n+1)}{(2n-1)(2n+1)(2n+3)} , \tag{7.8}$$

as given by Edwards and Schwartz [20] who considered only the surface tension in the shape energy. One of their important results is that the diffusion coefficient of the deformable droplet is identified with the $n = 1$ mode as

$$D = k_B T\left(\frac{3}{4\pi} K_{n=1}\right) . \tag{7.9}$$

Substituting (7.7) into (7.9) yields (7.2).

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