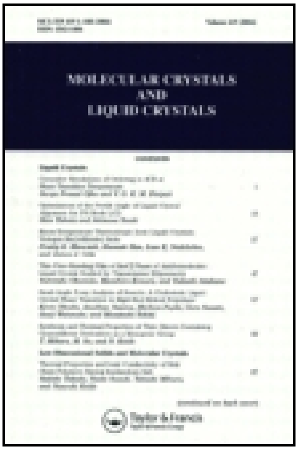

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Shape Dynamics of Nearly Spherical Membrane Bounded Fluid Cells†

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This paper considers the hydrodynamics of low Reynolds number shape fluctuations of incompressible fluid cells with incompressible membrane boundaries. It is assumed that the membrane can be characterized by a shear modulus μ, a bending modulus k_c, an intrinsic mean curvature c_0, and a surface viscosity η_M. The problem is formulated for the general case, and is solved analytically in the spherical limit. The observable hydrodynamics is found to depend sensitively on membrane parameters, and in particular a marked difference between solid and fluid behaviour is apparent.

INTRODUCTION

The characterization of liquid crystalline phases by phenomenological elastic constants and viscosities has been an important part of the overall study of liquid crystals. Biological membranes and their artificial analogues may be, physically speaking, lyotropic liquid crystal thin films, but in liquid crystal terms they are still rather puzzling. The best studied membrane is undoubtedly the red blood cell (RBC) membrane, but two kinds of experiments give contradictory impressions. On the one hand, there are the observable shape fluctuations of RBC's, seeming to indicate a fluid membrane.[1] On the other hand there are the micropipette aspiration experiments, which apparently indicate a solid membrane.[2]

The purpose of this paper is to consider the observable hydrodynamics of membranes in the context of this problem. It is essentially

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an extension of the ideas of Lennon and Brochard, who interpreted the erythrocyte flicker phenomenon as a measure of bending modulus (assuming a fluid membrane). More generally, one should be able to test assumptions about the nature of the membrane by watching how it moves.

The theory of Brownian shape fluctuations of nearly spherical membrane-bounded fluid cells is completely worked out here in the spherical limit. It is found that solid and fluid membranes behave very differently, as one might expect. In addition, the methods used here are sufficiently powerful to yield detailed hydrodynamical predictions even for the more complicated vesicle shapes of actual experimental practice.

Section **I** describes the hydrodynamical model to be solved. Section **II** describes and carries out the spherical limit of the membrane free energy and dissipation functionals. Section **III** solves the hydrodynamics in the spherical limit.

I. HYDRODYNAMIC MODEL

The following model is assumed for a fluid cell undergoing dynamic shape change:

1. the cell is an incompressible Newtonian fluid occupying the (time dependent) region D,
2. the fluid motions are sufficiently slow for the hydrodynamics to be linear, (it will even be quasi-static)
3. the cell membrane M is an incompressible two-dimensional viscoelastic solid or fluid

Assumption 1, that the interior fluid is Newtonian, is satisfied for RBC's and for vesicles in water. In the context of Brownian shape fluctuations assumption 2 needs no additional justification. Assumption 3 is justified because the two dimensional bulk modulus of real biological membranes is large: Brownian area fluctuations are negligible.

Thus the bulk fluids obey the Navier-Stokes equation

$$\rho \frac{\partial \vec{v}}{\partial t} = -\vec{\nabla} P - \eta \vec{\nabla} \times (\vec{\nabla} \times \vec{v}) \tag{1}$$

where ρ, the bulk density, P, the pressure, and η, the shear viscosity, may be different for interior and exterior fluids. In fact, since the

Reynolds number is very small, the left side could be taken as zero, but this result emerges naturally in any case.

Now consider a small element of the membrane, with unit outer normal $\hat{n}$ and displacement $\vec{u}$ from its equilibrium position. At low Reynolds number its inertia is negligible and the net force on it must vanish, i.e.,

$$\vec{F}_i + \vec{F}_e + \vec{F}_m = \vec{0} \tag{2}$$

Here $\vec{F}_i$ is the force due to the interior fluid, $\vec{F}_e$ is the force due to the exterior fluid, and $\vec{F}_m$ is the force due to adjoining elements of the membrane. In addition the velocities must be continuous at the membrane:

$$\vec{v}_i = \vec{v}_e = \dot{\vec{u}} \tag{3}$$

Eq. (2) may be re-expressed in terms of the stresses $\overleftrightarrow{\sigma}$ in the fluid:

$$\vec{F}_m = (\overleftrightarrow{\sigma}_e - \overleftrightarrow{\sigma}_i) \cdot \hat{n}\, dA \tag{4}$$

It is a boundary condition describing the discontinuity in certain elements of the bulk stress tensor.

$\vec{F}_m$ depends only on properties of the membrane:

$$F_m = -\left(\frac{\delta F}{\delta \vec{u}} + \frac{\delta D}{\delta \dot{\vec{u}}}\right) dA \tag{5}$$

where F and D are the phenomenological free energy and dissipation function of the membrane. Thus the membrane parameters come into the problem only through the boundary condition

$$(\overleftrightarrow{\sigma}_e - \overleftrightarrow{\sigma}_i) \cdot \hat{n} = -\frac{\delta F}{\delta \vec{u}} - \frac{\delta D}{\delta \dot{\vec{u}}} \tag{6}$$

A model of membrane hydrodynamics is completely specified if we specify F and D.

It is widely accepted that

$$F = F_{\text{dilation}} + F_{\text{shear}} + F_{\text{bending}} \tag{7}$$

for real membranes, with a corresponding statement for the dissipation functions. Assumption 3 above, that the membrane is incompressible, eliminates F_{dilation}: its role is taken over by the constraint of incompressibility. Of the remaining two terms, one could imagine that one or the other dominates. This gives two extreme models, which we shall call the pure solid membrane model and the pure fluid membrane model:

$$F = F_{\text{shear}} \qquad \text{(pure solid model)} \tag{8}$$

$$F = F_{\text{bending}} \qquad \text{(pure fluid model)} \tag{9}$$

In either case it is plausible that the membrane dissipation function would be dominated by the shear term:

$$D = D_{\text{shear}} \qquad \text{(solid or fluid model)} \tag{10}$$

The problem is completely specified if we specify the form of F_{shear}, F_{bending}, and D_{shear}. These functionals have been discussed in References 3. For an isotropic membrane M

$$F_{\text{shear}} = \frac{\mu}{2} \int_M S^{ij} S_{ij} \sqrt{g}\, dA \tag{11}$$

where μ is the 2-d shear modulus and

$$S_{ij} = \tfrac{1}{2}(u_{i;j} + u_{j;i}) - \tfrac{1}{2}(\text{div } u)\, g_{ij} + n(C_{ij} - Hg_{ij}). \tag{12}$$

Here i and j label coordinates in the membrane so that u^i is a tangential displacement, and $n = \vec{u} \cdot \hat{n}$ is a normal displacement, from the shear free state. The tensors g_{ij} and C_{ij} are the first and second fundamental forms of the membrane surface, $g = \det g_{ij}$, $H = \frac{1}{2}Tr(C)$ (the mean curvature), and semicolon indicates a covariant derivative with respect to the metric connection on M. D_{shear} has the same form with μ replaced by the 2-d membrane shear viscosity η_M and $\vec{u}$ replaced by $\dot{\vec{u}}$.

A widely accepted form for F_{bending} is the Helfrich curvature free energy[4]

$$F_{\text{bending}} = \frac{1}{2} k_c \int_M (2H - c_0)^2 \sqrt{g}\, dA \tag{13}$$

where k_c is a phenomenological bending modulus and c_0 is a constant tending to bias H, the mean curvature. In fact Eq. (13) is the most general form which is isotropic, homogeneous, and at most quadratic in the principal curvatures. Among the shapes which extremize $F_{bending}$ are shapes startlingly like observed red blood cell shapes (in fact indistinguishable from them), so that this form has a certain measure of experimental support, at least in its hydrostatic consequences.[5,6] Extending the investigation to the hydrodynamics of $F_{bending}$ would therefore appear to be a promising proposition.

II. SPHERICAL LIMIT

A sphere of fixed volume and surface area has no shape degrees of freedom—it cannot exhibit shape fluctuations at all, because of the constraints. Thus in describing shape fluctuations one is denied the luxury of spherical geometry, at least initially, and must proceed somewhat indirectly as follows:

(1) Find the equilibrium shape of a *nearly* spherical cell by minimizing the appropriate membrane free energy at fixed V and A.

(2) Expand the membrane free energy to second order about this non-spherical minimum configuration, keeping volume and surface area constant. In this way the free energy becomes a quadratic form in the deformation parameters.

(3) Take the limit of the quadratic form in (2) as the equilibrium shape approaches a sphere.

As we shall show, the limit referred to in (3) exists, makes sense, and can be found analytically as a quadratic form in deformation parameters. Even though the scale of allowed deformations goes to zero in this limit, the relaxation eigenmodes and relaxation times smoothly approach finite and sensible limits. (This is possible because the hydrodynamics is linear, i.e., homogeneous of degree one: the scale on which it occurs is irrelevant.) This spherical limit is one of our main results.

Steps (1), (2), and (3) above may seem needlessly difficult. One is trying to describe a sphere, yet step (2) requires essentially that one should be able to deal with an arbitrary shape as a necessary intermediate step! This complicates things, but it is not clear that there is a simpler way to do it. The spherical limit is, in the end, very simple and satisfying, but it only makes sense as a limit.

One interesting consequence of the spherical limit is the existence of a hydrodynamic Goldstone mode of a nearly spherical fluid membrane, described below, a new result. Upon reflection one realizes that such a mode must in fact be there. This gives one confidence that the next sections, although intricate, do ultimately yield insight into a problem of unexpected subtlety.

A. Fluid model membrane

In this section we carry out steps (1), (2), and (3) of the previous section for F_{bending} (Eq. (13)), which describes a pure fluid membrane. Each step is done in the corresponding subsection.

1. Equilibrium shape

Parametrize shapes near the sphere $r = a$ as slightly distorted spheres

$$r = a + n'(\theta, \phi) \tag{14}$$

The function $n'(\theta, \phi)$ gives the distortion as a radial displacement. The effect of this distortion on the volume V and area A, to second order in n', is

$$\delta V = \int n' a^2 \, d\Omega + \int n'^2 a \, d\Omega \tag{15}$$

$$\delta A = 2\int n' a \, d\Omega + \int \left[\frac{1}{2} \left| \vec{\nabla} n' \right|^2 + \left(\frac{n'}{a} \right)^2 \right] a^2 \, d\Omega \tag{16}$$

Since we are interested in parametrizing shapes which have slightly less volume than the sphere of the same surface area, n' is chosen to keep A fixed. Then V can only decrease. To second order this is done by representing n' as

$$n' = n'_1 + n'_2 \tag{17}$$

where

$$\int n'_1 \, a^2 d\Omega = 0 \tag{18}$$

so that $\delta A = \delta V = 0$ to first order, and then choose $n'_2 = \mathcal{O}(n_1'^2)$ so that $\delta A = 0$ also in second order. That is,

$$n'_2 = -\frac{1}{2} \int \left[\frac{1}{2} \left| \vec{\nabla} n'_1 \right|^2 + \left(\frac{n'_1}{a} \right)^2 \right] a \, d\Omega \tag{19}$$

and then

$$\delta V = \int \left[\frac{1}{2} n_1'^2 a - \frac{1}{4} \left| \vec{\nabla} n_1' \right|^2 a^3 \right] d\Omega \tag{20}$$

For

$$n_1' = \epsilon' Y_{lm} \tag{21}$$

one has

$$\delta V_l = \frac{a\epsilon'^2}{4} [2 - l(l + 1)] \tag{22}$$

The case $l = 0$ violates the conditions Eq. (18) and $l = 1$ is trivially a rigid translation, but for $l \geq 2$ Eq. (22) gives δV as a diagonalized quadratic form in the space of area-preserving distortions.

Now for fixed δV we want to minimize $\delta F_{\text{bending}}$ in that same space in order to find the nearly spherical equilibrium shape. Here F_{bending} is given in Eq. (13), the Helfrich curvature free energy. Its expression as a quadratic form in n was given in Reference 3, Eqs. (36)–(43). In the basis of Eq. (21), with the sphere as reference configuration, it too is diagonal, with diagonal elements

$$F_1 = -2\, k_c\, [l(l + 1) - c_0 a]\, \delta V_l / a^3 \tag{23}$$

It is now elementary to choose n_1' to minimize δF_l for fixed δV. The minimum is achieved for $l = 2$, i.e., the distortion is quadrupolar, as one might have guessed. One could take, for example,

$$n_1' = \epsilon' Y_{20}. \tag{24}$$

Indeed this result is already known in the case of azimuthally symmetric distortions, from the work of Helfrich and Deuling,[5] and also Jensen.[6] The above result is more general, however, in an important respect. The restriction of azimuthal symmetry is removed. The eigenspace corresponding to the equilibrium configuration is degenerate: *any* $l = 2$ distortion will do in Eq. (24), not simply Y_{20}. In choosing one $l = 2$ configuration over another the system spontaneously breaks a continuous symmetry. This is our first hint of a Goldstone mode in the system, a hint which is confirmed in the hydrodynamics calculation.

2. Expand F_{bending}

The second step in finding the spherical limit of F_{bending} is to expand F_{bending} to second order in deformation parameters for deformations which preserve the volume and surface area of the non-spherical shape M. Take coordinates (x^1, x^2) in M and parametrize the deformation by $n(x^1, x^2)$, the displacement along the local normal to M which transforms it into the deformed surface. (The idea is similar to what was done in Eq. (14) but more general. The local normal direction is now not necessarily the radial direction in spherical polar coordinates. The equilibrium surface is not a sphere but the surface of M defined in Eq. (14) with some definite choice for n'. The roles played by n' in the last section and n in this one, though similar in appearance, are conceptually very different.)

In order to preserve volume and area, n must obey

$$n = n_1 + n_2 \tag{25}$$

where

$$\int n_1 \sqrt{g}\, dA = 0 \tag{26}$$

$$\int n_1 H \sqrt{g}\, dA = 0 \tag{27}$$

and $n_2 = \mathcal{O}(n_1^2)$ is chosen to preserve volume and area in second order. For consistency of the expansion one should require that n_2 is in fact a small correction to n_1. For M the surface defined by Eqs. (14) and (24) this turns out to mean

$$\max n_1 \ll \epsilon' \tag{28}$$

This very reasonably says that ϵ', a measure of the distance of M from a sphere, puts a kinematic bound on the amplitude of allowable shape fluctuations.

The expansion of F_{bending} under these conditions was done in Reference 3, Eqs. (65)–(72). It is

$$\delta F_{\text{bending}} = \frac{1}{2} k_c \int_M [(\nabla^2 n_1)^2 + A^{ij} n_{,i} n_{,j} + B\, n_1^2] \sqrt{g}\, dA \tag{29}$$

where

$$A^{ij} = -2(2H - c_0)(C^{ij} - Hg^{ij}) + g^{ij}[-6H^2 + 4K - 2c_0H - (Q - R\langle H\rangle)/4S] \quad (30)$$

$$B = 16H^4 - 20H^2K + 4K^2 + 2\nabla^2(2H^2 - K) + 4[(C^{ij} - Hg^{ij})H_{,i}]_{;j} + Q(\langle H\rangle H - K/2)/S + R(\langle H^2\rangle H - \langle H\rangle K/2)/S \quad (31)$$

$$Q = \langle 4|\vec{\nabla} H|^2 - 8H^4 + 4HK(2H - c_0)\rangle \quad (32)$$

$$R = \langle 8H^3 - 4K(2H - c_0)\rangle \quad (33)$$

$$S = \langle H^2\rangle - \langle H\rangle^2 \quad (34)$$

$$H = (c_1 + c_2)/2 \quad (35)$$

$$K = c_1c_2 \quad (36)$$

Here c_1 and c_2 are the principle curvatures, and $\langle\ \rangle$ indicates average over the surface.

3. Take the spherical limit

In the quadratic form above the coefficients A^{ij} and B are known functions of ϵ'. It is now straightforward, though tedious, to take the limit as ϵ' goes to zero. The spherical limit is diagonal in the basis

$$n_1 = \epsilon Y_{lm} \quad (37)$$

and has diagonal elements

$$(\delta F_{\text{bending}})_l = \frac{1}{2} k_c \left(\frac{\epsilon}{a}\right)^2 [l(l + 1) - 2][l(l + 1) - 6] \quad (38)$$

which is the result we were aiming at.

Several features of this result are worth a comment.

(1) It is independent of c_0. This seems a little strange.

(2) $l = 0$ is, of course, not allowed (it violates Eq. (26)).

(3) $l = 1$ labels the translation modes of the sphere. These are not distortions of the shape and do not cost curvature energy.

(4) $l = 2$ distortions *also* do not cost curvature energy! This is the Goldstone phenomenon. The system can wander freely in a multidimensional space of energetically equivalent configurations.

(5) In the spherical limit the constraint equations (26) and (27) become identical—i.e., one constraint is lost. This is potentially misleading, since it overestimates the number of allowed $l = 2$ modes by one. In fact, from Eq. (27), we must impose

$$\int_M n_1 n_1' \sqrt{g}\, dA = 0 \tag{39}$$

when M is close to a sphere and hence also in the spherical limit. Since n_1' is $l = 2$, there are only four $l = 2$ modes available to n_1 instead of five. Of these four modes, two come from the rigid rotations of (non-spherical) M, and are in a sense trivially Goldstone modes, in the same way that the $l = 1$ modes are. That leaves two non-trivial Goldstone modes, and these are, in effect, a surprise and potentially interesting. They should correspond to anomalously large amplitude, slowly relaxing hydrodynamic modes of nearly spherical vesicles, as we see in more detail below.

(6) The existence of non-trivial $l = 2$ Goldstone modes, costing nearly zero energy even for non-spherical systems, suggests that the non-spherical equilibrium configurations may become infinitesimally unstable to $l = 2$ perturbations. This inference has been verified in Reference 7, where it is shown that the azimuthally symmetric red blood cell shape becomes infinitesimally unstable to the $l = 2$, $m = 2$ perturbation when c_0 is sufficiently positive.

B. Solid model membranes

In this section we find the spherical limit of F_{shear}, the pure solid model. The general outline of the argument is that of the previous section, but the results are quite different. In particular, there is no Goldstone mode.

1. Equilibrium shape

The shear strain S_{ij} of Eq. (12) implicitly refers to a pre-defined shape in which $S_{ij} = 0$. Thus there is considerable arbitrariness in F_{shear}.

One could, for example, take any nearly spherical shape, as given in Eq. (14), and assume that for this shape $S_{ij} = 0$. It would therefore be the equilibrium shape: the shape would be "built in." This will be our assumption, in fact: that in the equilibrium shape all shear strain has relaxed.

Another possibility is that because of constraints the equilibrium shape has a built in shear strain which cannot relax. This case is more difficult and more arbitrary. One case of this kind is closely related to the spherical limit and is interesting in its own right, so we consider it here.

Take the *spherical* shape to be shear free ($S_{ij} = 0$) and now decrease the volume as in section II.A.1. The expansion of the shear free energy is given in Reference 3 and also below. The result is that δF is diagonal in the basis of Eq. (21) with diagonal elements

$$(\delta F_{\text{shear}})_l = \mu\epsilon'^2 \left[1 - \frac{2}{l(l+1)}\right] = \frac{-\mu(\delta V)_l}{4al(l+1)} \qquad (40)$$

Now we seek to minimize $(\delta F_{\text{shear}})_l$ for fixed δV in order to determine the non-spherical equilibrium shape—but no minimum exists! Since $\Delta V < 0$, it is true that $(\delta F)_l > 0$, but the increase in free energy can be made arbitrarily small by taking l large enough. The physical reason for this singular result is probably that the surface can pucker or wrinkle without shearing very much. Of course wrinkles cost bending energy, but in the pure solid model the bending energy was omitted. For a realistic membrane the bending energy would have to become important in determining the equilibrium shape. The pure solid model is somewhat pathological!

2. Expand F_{shear}

Let M be a nearly spherical surface as in Eq. (14) free of shear strain (and hence the equilibrium configuration of the pure solid model). Parametrize deformations of M by the normal displacement n, exactly as in Section II.A.2. The expansion of F_{shear} was found in Reference 3, Eqs. (46)–(55). The result is that one must solve the linear equations on M

$$\nabla^2\alpha = -2nH \qquad (41)$$

$$\nabla^4\beta + 2(K\beta_{,i})_{;j}g^{ij} + 2\alpha_{,i}K_{,j}e^{ij} + [n(C^{ij} - Hg^{ij})]_{;i;k}e_j^k = 0 \qquad (42)$$

where $e^{ij} = \epsilon^{ij}g$ is the antisymmetric tensor on M and $\epsilon^{12} = -\epsilon^{21} = 1$. Then F_{shear} is found by evaluating Eq. (11) with S_{ij} given by Eq. (12) and

$$u_i = \alpha_{,i} + e_i^j\beta_{,j} \tag{43}$$

3. Take the spherical limit

Choose $n_1 = \epsilon Y_{lm}$ with $l \geq 2$ as a basis for the deformations of M in the spherical limit, just as in Eq. (37) and following. Then we can solve Eqs. (41) and (42):

$$\alpha_l = \frac{2a\epsilon Y_{lm}}{l(l+1)}[1 + \mathcal{O}(n')] \tag{44}$$

$$\beta_1 = \mathcal{O}(n'n) \tag{45}$$

Putting these expressions into Eqs. (43), (12), and (11) and taking the limit as n' goes to zero yields

$$(\delta F_{\text{shear}})_l = \mu\epsilon^2\left[1 - \frac{2}{l(l+1)}\right] \qquad (l \geq 2) \tag{46}$$

which is the spherical limit of F_{shear}, a diagonal quadratic form in this basis.

D_{shear} then has the same form in the spherical limit, with $\vec{u}$ replaced by $\dot{\vec{u}}$ and μ replaced by η_M.

III. HYDRODYNAMICS

Eqs. (1), (3), and (6), together with Eqs. (7), (10), (38), and (46) define the hydrodynamics of shape fluctuations in the spherical limit. It is straightforward to work out the consequences. We choose to work with the one-sided (Laplace) transform

$$\vec{v}(r, z) = \int_0^\infty \vec{v}(\vec{r}, t)\, e^{izt}\, dt \tag{47}$$

for the bulk fluid velocities. In particular we find the hydrodynamic eigenmodes corresponding to distortions $n_l = \epsilon Y_{lm}$.

A useful basis for the space of divergenceless flows is the eigenfunctions of $\vec{\nabla} \times \vec{\nabla} \times$ with eigenvalues $k^2 > 0$, namely

$$\vec{v}_l^{(1)} = -\sqrt{l}\, j_{l+1}(kr)\vec{Y}_{ll+1\,m} + \sqrt{l+1}\, j_{l-1}(kr)\, \vec{Y}_{ll-1\,m} \qquad (48)$$

$$\vec{v}_l^{(2)} = j_l(kr)\, \vec{Y}_{llm} \qquad (49)$$

together with the gradients of harmonic functions

$$\vec{v}_l^{(3)} = R\vec{\nabla}\left[\left(\frac{r}{R}\right)^l Y_{lm}\right] \qquad (50)$$

where $R = a$ is the radius of the spherical cell. Here the $\vec{Y}_{Jlm}$ are vector spherical harmonics, and the j_l are spherical Bessel functions. (For the interior bulk flow the j_l must be of the first kind in order to be non-singular at $r = 0$. For the exterior bulk flow they are not so restricted.)

It is clear in Eq. (1) that $\nabla^2 P = 0$, so take

$$P = P_l\left(\frac{r}{R}\right)^l Y_{lm} \qquad (51)$$

where P_l is a constant.

Substituting the form

$$\vec{v}_l = A_{1l}\,\vec{v}_l^{(1)} + A_{2l}\,\vec{v}_l^{(2)} + A_{3l}\,\vec{v}_l^{(3)} \qquad (52)$$

into the Navier-Stokes equation yields a solution provided that

$$iz\rho = -\eta k^2 \qquad (53)$$

$$-iz\rho\, RA_{3l} = P_l \qquad (54)$$

with A_{1l} and A_{2l} still arbitrary. [This remark applies twice: once to the interior and once to the exterior flow.] Note that kR is the square root of the Reynolds number, and hence small.

In the spherical limit many simplifications occur in the boundary conditions to make the problem tractable. One simplification is that $\vec{v}_l^{(2)}$ (see Eq. (52)) decouples from shape fluctuations. The tangential flow in the membrane induced by the condition of incompressibility is $\vec{\nabla} Y_{lm}$, agreeing with $\vec{v}_l^{(1)}$ and $\vec{v}_l^{(3)}$. The tangential behavior of $v_l^{(2)}$, however, is $\vec{r} \times \vec{\nabla} Y_{lm}$. In addition the tangential stress on the membrane due to the bulk flows $\vec{v}_l^{(1)}$ and $\vec{v}_l^{(3)}$, and also the internal shear stress due to the induced membrane flow alluded to above are all of the form to be balanced by the gradient of a 2-d isotropic stress in the membrane. Since the membrane is incompressible, this 2-d isotropic stress can be whatever it needs to be, and the balance of tangential stresses is guaranteed (at least for modes in which $\vec{v}_l^{(2)}$ does not participate).

Thus the hydrodynamic problem splits into two decoupled problems: one, with which we shall be concerned, in which coupled normal and tangential flows of the membrane must be matched to bulk flows of the form of $\vec{v}_l^{(1)}$ and $\vec{v}_l^{(3)}$; and another, with which we shall not be concerned, in which purely tangential flows of the membrane must be matched to bulk flows of the form $\vec{v}_l^{(2)}$. The latter modes, since they are independent of the shape fluctuations, would be more difficult to observe directly.

By the above remarks the boundary conditions Eqs. (3) and (6) reduce to five homogeneous linear equations (continuity of normal and tangential velocities, both interior and exterior, and continuity of normal stress) in five unknowns $A_{1l}^{(\text{int})}$, $A_{3l}^{(\text{int})}$, $A_{1l}^{(\text{ext})}$, $A_{3l}^{(\text{ext})}$, and n_l. To describe thermal shape fluctuations it is advantageous to add an inhomogeneous stochastic driving term, a fluctuating pressure $\pi_l Y_{lm}$. Then we can find the response in the observable n_l.

In the limit of low Reynolds number, the five equations are

$$\begin{pmatrix} -iz & l(l+1) & l & 0 & 0 \\ \dfrac{-2iz}{l(l+1)} & l+3 & 1 & 0 & 0 \\ -iz & 0 & 0 & l(l+1) & -(l+1) \\ \dfrac{-2iz}{l(l+1)} & 0 & 0 & -(l-2) & 1 \\ \lambda & \nu_i & 0 & \nu_e & 0 \end{pmatrix} \begin{pmatrix} n_l \\ A^{(\text{int})}{}_{1l} \\ A^{(\text{int})}{}_{3l} \\ A^{(\text{ext})}{}_{1l} \\ A^{(\text{ext})}{}_{3l} \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 0 \\ -\pi_{lm} \end{pmatrix} \quad (55)$$

where

$$\begin{aligned} \lambda &= k_c[l(l+1)-2][l(l+1)-6]/a^2 \\ &\quad + 2(\mu + iz\eta_M)[1 - 2/l(l+1)] \\ \nu_i &= (l+1)(2l+3)\eta_i/R \\ \nu_e &= -l(2l-1)\eta_e/R \end{aligned} \quad (56)$$

The result, which takes the same form for both fluid and solid models, is

$$n_l = \chi_l(z)\pi_{lm} \tag{57}$$

where the complex response function is

$$\chi_l = [B_l(1 + iz/\Gamma_l)]^{-1} \tag{58}$$

and

$$B_l = \frac{k_c}{R^4}[l(l+1) - 2][l(l+1) - 6] + \frac{2\mu}{R^2}[1 - 2/l(l+1)] \tag{59}$$

Also

$$B_l/\Gamma_l = 2\eta_M\left[1 - \frac{2}{l(l+1)}\right] + \frac{(l-1)(2l+3)}{2la}\eta_i + \frac{(l+2)(2l-1)}{2(l+1)a}\eta_e \tag{60}$$

Here η_i and η_e refer to the bulk shear viscosities (interior and exterior) while η_M refers to the 2-d membrane shear viscosity.

By the fluctuation-dissipation theorem, the power spectrum for a model labeled by l is

$$S_l(\omega) = -2k_BT\frac{Im\chi_l(\omega)}{\omega} = \frac{2k_BT\,\Gamma_l}{B_l(1 + \omega^2/\Gamma_l^2)} \tag{61}$$

Then, by the Wiener Khintchine theorem, the autocorrelation function for $n_l(t)$ is the cosine transform of $S_l(\omega)$ (divided by R^2)

$$C^l_{\text{shape}}(\tau) = \langle n_l^*(t)\, n_l(t + \tau)\rangle = (k_BT/R^2B_l)\, e^{-\Gamma_l\tau} \tag{62}$$

Evidently Γ_l^{-1} can be interpreted as a sum of relaxation times, one for the interior fluid, one for the exterior fluid, and one for the membrane itself.

The spectrum of relaxation rates Γ_l is quite different for the pure fluid and pure solid models. Thus the hydrodynamics of shape fluctuations provides a straightforward method for distinguishing experimentally between solid and fluid membranes.

One can distinguish special cases of the models depending upon whether the viscous dissipation takes place predominantly in the bulk or in the membrane itself. Roughly speaking, one has (1) fluid membrane, dissipation predominantly in bulk, $\Gamma_l \sim l^3$, (2) fluid membrane, dissipation predominantly in membrane, $\Gamma_l \sim l^4$, (3) solid membrane, dissipation predominantly in bulk $\Gamma_l \sim l^{-1}$, (4) solid membrane, dissipation predominantly in membrane, Γ_l = constant (independent of l).

Case (4) above, in which all hydrodynamic modes relax at the same rate for a pure solid membrane if dissipation is predominantly within the membrane, is a particularly powerful prediction, because it is true generally and not just in the spherical limit. Such behavior, if it were actually exhibited, would be immediately noticed. We can be quite sure, for example, that red blood cells are not described by case (4), since the erythrocyte flicker phenomenon has been known for a long time to have a broad spectrum of relaxation times.[1]

IV. SUMMARY

This paper has given a complete account of vesicle shape fluctuations in the spherical limit. In order to do so, techniques were developed which are powerful enough to handle even the problem of realistic vesicle shapes. It is likely, however, that the spherical limit already contains the essential features of the problem. One learns that the spectrum of relaxation rates is sensitive to the viscoelastic parameters of the membrane, and that even qualitative information about the spectrum may have immediate implications for the physical state of the membrane. A detailed measurement of the relaxation spectrum, together with accurate computations using the techniques of this paper would be a powerful tool in determining the still puzzling physical properties of membranes.

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