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Relaxation modes of an adhering bilayer membrane

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Abstract. We determine the relaxational dynamics of the shape fluctuations of a

fluid

membrane in the vicinity of a

substrate. Extending the "classical" description, we include the
coupling between the local shape and the difference of the two monolayer densities as well as
a
lateral tension in the membrane. These extensions introduce additional length scales to the
problem. The asymptotic behavior of the dispersion relation and the correlation functions can be
understood from limiting cases in which either a

free bilayer or a

bound incompressible membrane
is considered. In many cases, however, the relevant length scales do not separate very well, so
that the full dispersion relation will be needed for the interpretation of experiments. It is shown
that in addition to the damping due to bulk viscosity the dissipation due to friction between the
monolayers is observable and indeed dominates the long-time behavior of the dynamical height
correlation function for large wave vectors. As demonstrated with typical sets of parameters,
the transition to this regime will be accessible by optical techniques only for weak adhesion and
strong friction between the monolayers.

1. introduction.

One of the earliest phenomena to receive attention in the study of the physical properties
of biological membranes [1, 2] has been the slow relaxational dynamics of thermally excited
membrane shape fluctuations, such as the "flicker" phenomenon of red blood cells [3]. Fluid
phospholipid membranes, although lacking a cytoskeleton and membrane proteins, serve as
simple model systems for biological membranes, and are experimentally accessible in form
of giant vesicles. These vesicles also exhibit strong shape fluctuations which for free, qua-
sispherical vesicles have been used to determine the bending rigidity, which is the essential
material parameter of lipid membranes [4-6]. In addition, the dynamical correlations of these
fluctuations give information about the main dissipative mechanisms.

The effect of adhesion or interaction with a

substrate on these membranes can be studied in

an experimental set-up recently developed by Ridler and Sackmann [7-9]. They have shown
that one can obtain the static and dynamic height correlation functions of the bound part of
an adhering giant vesicle using reflection interference contrast microscopy.

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In the "classical" model of a

free membrane, the membrane is treated as a single, incom-
pressible fluid sheet, whose energy is solely determined by the resistance to bending [10], and
for which the damping is provided by the surrounding viscous fluid [3]. Three modifications
to this classical treatment of the bending modes of a membrane are necessary to represent the
experimental situation of an adhering membrane.
First of all, the presence of a

substrate modifies the static fluctuation spectrum as well as

the hydrodynamics ill, 12]. Secondly, the membrane will be part of a bound vesicle. For
wave-lengths much smaller than the size of the vesicle (where up to 50 ~lm are accessible),
the geometrical complications of the closed shape should not affect the dynamics. Therefore,
results obtained for a bound planar membrane should apply provided the constraints on vesicle
volume and area are effectively included in the description as a

finite tension in the membrane

[13, 14].
Finally, recent work on the equilibrium shapes of vesicles has demonstrated the relevance
of the coupling between the local curvature and the local density of lipids within the two
monolayers [lS]. For static phenomena such as the equilibrium shapes of vesicles, this effect
adds an additional global energy term, the area-difference-elasticity [16-19]. In the dynamics,
bending is coupled to differences in the local lipid density in the monolayers, because bending
the membrane will compress the inner and stretch the outer monolayer. Since the lipid monc-
layers are free to slide over each other, these density differences may relax [lS, 20]. Thus, the
friction between monolayers will provide an additional dissipative mechanism for the damping
of undulations [21]. In the small-q regime, the density difference relaxation is too fast to have
any effect on the height correlations. For undulations with a sufficiently large wave-vector q,
however, the relaxation rate of the monolayer density difference, which scales as q~, will al-
ways be slower than the relaxation of undulations, which scales as q~ [3]. Due to the coupling
between curvature and density, in this regime undulations will relax by a

fast process only to
the extent that they minimize the energy for given density difference. The slow decay of this
density difference then dominates the long-time behavior of the height correlations.
The aim of this work is to combine these three aspects in order to explore the full dynamical

behavior of a

lipid bilayer membrane near a wall. In particular, we are interested in the
circumstances, under which the density relaxation makes an observable contribution to the
height fluctuations. One therefore needs to know not only the disiersion relation but also the
time-dependent correlation function for the height variable. These quantities inevitably have
a
rich structure due to the presence of four length-scales. Two length-scales follow from the
static correlation function in which the potential, the tension and the bending energy all scale
differently with the wave-vector. As a

third length-scale, the separation from the wall affects
the dynamics. Finally, the fourth length-scale arises from the coupling to the density difference.
Since for a bound membrane, in contrast to the free bilayer, the relaxation rate of the height
correlations can become faster than the density relaxation also for small q, one might first
expect that the double layer aspect of the membrane is then relevant for all q. However, we
will show that the density mode will be observable in the height correlation function only if
the undulation energy is dominated by the resistance to bending. For small tension, I.e. weak
adhesion, the cross-over to this regime is in the visible range and the coupling to the local
density difference should, indeed, have observable effects for a bound membrane.
The paper is organized as follows. In section 2, the general framework of the calculation is
set up. We develop the solution of the full problem starting from limiting cases. In section
3, we discuss the free bilayer, for which we here also calculate the time-dependent correlation
functions. In section 4, we briefly review the relaxation rate for a bound membrane if the bilayer
aspect is ignored. From these two limiting cases, the numerical results for the full problem
can be understood. Rather than discussing the dispersion relation and the corresponding

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correlation function for all possible cases, we present these quantities for three experimentally
motivated examples in section S. Section 6 contains the conclusion. In Appendix A, the lengthy
matrix entries which are the basis for the numerical calculations are given for future reference.
In Appendix B, we justify the low Reynolds number approximation. Appendix C deals with
the modifications which arise for a

typical experimental situation where the fluctuations are

measured within a

linear strip. In Appendix D, we discuss the effect of finite slip at the
substrate as well as the possibility of another slow mode occurring for multilamellar vesicles.
Brief accounts of the dispersion relations for the two limiting cases have been presented in
references ill, 21].

2. Statics and dynamics of a bound bilayer.

In order to calculate the relaxation modes for both height and monolayer density fluctuations,
we first recall the energy of a bilayer. The lipid densities at the neutral surface of each mono-
layer, #+, have an equilibrium value #o for a

flat membrane (in this paper (+) will generally
denote the monolayer opposite to the wall and (-) the monolayer facing the wall). It is useful
to project these densities within each monolayer to a common surface, the bilayer midsurface.
These projected densities, ~b+, will deviate from #+ if the membrane is bent. With H as the
mean curvature of the bilayer and d as the distance between the midsurface of the bilayer and
the neutral surface of a monolayer, this relation reads #+ m ~b+(1+ 2dH) to lowest order in dH.
Expressed in terms of a

scaled density deviation from the equilibrium value, p+ a (~b+ /#o I),
the elastic energy density of each monolayer, f+, is then given by

f+ =

~ ~~

l) =

~

(p+ + 2dH)~ (l + O(dH) + O(p+)),

(1)
2 #o

~

2

where k is the area compression modulus of a monolayer [21].
We consider a membrane bound by a potential V(I) whose minimum is at a distance lo from
a planar wall [22]. Denoting the local displacement of the membrane by h(x, y) + I(x, y) lo,
we can express the mean curvature for small displacements as H

=

)i7~h The continuum
free energy, F, for the entire membrane then is a

functional of the membrane shape, h(x, y),
and the two densities p+(x, y),

F

=

/dx dy (~r(i7~h)~ + E(i7h)~ +

flh~ + k [(p+ + di7~h)~ + (p~ di7~h)~] (2)

2

The first three terms represent the energy of undulations at relaxed densities. This energy

is given by the bending energy of each monolayer with the usual bilayer bending rigidity ~r
[lo], a

tension E, which arises from the constraints on area and volume of the vesicle [13, 14],
and the contribution from the potential, which we expand about its minimum at = lo with
fl = d~V/dl~(i=io. Within the range of wave-lengths accessible by optical methods, these
tensions typically can become as relevant as the bending rigidity (23].
For small deviations from the planar state, we consider a plane wave in the x-direction,
h(x, y) + hqe'~~ and p+(x, y) e (pq+pq)e'~~, written in terms of the difference, p e

(p++p~ /2,

and the average, p e

(p+ p~)/2, of the scaled projected densities of the monolayers. In the
Fourier-transformed free energy,

h~

*

F

"

(hq Pq >q) E(q) Pq (3)
>q

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where we have defined

k q~ + E q~ + fl -2 kdq~ o

E(q) + -2 kdq~ 2 k o

,

(4)

o o 2 k

p decouples from h and p. The star denotes complex conjugation. Here

k

= ~c + 2d~k IS)

is a

renormalized bending rigidity which includes the effects of elastic deformation of the
monolayers. Note that integrating out the density variables, which on the Gaussian level is
equivalent to minimizing with respect to them, one recovers the undulation energy of a bound
incompressible membrane with

Fo e min F

= h~ (~cq~ + Eq~ + fl) h(w hqEo(q)h(. (6)
lPq,Pql 2 2
The static correlation functions can be obtained from the inverse of the energy matrix E,
which leads to

Gajq) ~ Ill h~,p~,p~)*)

=

jE-ijq)
Pq ° ~ ~
1 dq~ °
kBT
dq~ Ehh(q)/2k ~ ~~~
L~Ly E0 (q)
~ o Eo (q)/2 k

where L~ and L~ denote the linear dimensions of the rectangular patch of membrane under
consideration.

For the calculation of the dynamical correlation functions, we have to consider the hydrody-
namics of both the fluid bilayer and the surrounding aqueous solution. The physical properties
of the membrane and its undulation energy will enter the boundary conditions and the force

balance at the membrane.

The hydrodynamics of the incompressible bulk fluid is treated with1il the Stokes approxima-
tion,

V v =

0 and qv~v = Vp for z # lo. (8)

This approximation, neglecting the inertial and advection terms of the Navier-Stokes equation,
is valid for low Reynolds numbers, an assumption which has to be checked afterwards (see
Appendix B).
We again assume plane waves in the z-direction, with velocity v = (uq,~ (z)e~ +uq,z(z)ez)e'~~
and pressure p = pq(z)e~~~ The general solution to the hydrodynamic equations (8) satisfying
the condition of vanishing velocity at the wall and at infinity then is

uj~(z) = [A (sinh(qz) qz cosh(qz)) + Bqz sinh(qz)],

pj (z) = 2qq1-A cosh(qz) + B sinh(qz)],

(9)

u(~(z) =

Ce~~~~~~°~ + Dq(z o)e~~~~~~°~j

,

pt(z) = 2nqDe-~~z-1°),

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where the indices + apply to z § lo, respectively, and u(~(z) =

idzu(~(z)/q. The constants
A, B, C, and D are to be determined by boundary coniitions at th~ membrane, which for
small displacements may be evaluated at z =

lot the normal velocity uz has to be continuous,
whereas assuming a no-slip boundary condition between lipid and water means that VI (lo) has
to coincide with the velocities of the lipid flow within the monolayers. Furthermore, the forces
have to balance in the normal direction at the membrane, leading to

Tt + Tz =

- $,

(lo)
q

where we have introduced the liquid stress tensor T,) e -pb~j + q(d;uj + dju,), evaluated
at the upper and lower monolayers. Denoting the monolayer surface pressures with a+, the
membrane surface viscosity with p, and the interlayer friction (whose order of magnitude can
be estimated from experiments as in Refs. [24, 15]) with b, the lateral force balance at the two
monolayers reads

Va+ + Tj + pV~V+ ~ b(V+ V~) = 0. (ll)

The tilde refers to two-dimensional quantities. At this point, we neglect the fact that the
various forces act in different surfaces of the monolayer, e-g- the intermonolayer friction in the
bilayer midplane, and the coupling to bulk water at the lipid headgroups. A more sophisticated
treatment including this effect should then also incorporate orientational degrees of ireedom oi
the lipids within the monolayer, which are excited by the torques produced in this way.
Equations (10) and (11) and the boundary conditions are sufficient to solve ior all oi the
amplitudes A, B, C, and D of both the flow field v(z, z) and the pressure p(z, z) as a

function

of hq, pq and pq. Using the assumption of impermeability of the lipid membrane to water flow
[25], I-e- dhq /dt = uq,z (lo), and the continuity equations for the monolayer densities, dtp) "
-V V(, which are correct when neglecting the slow lipid flip-flop between the monolayers,
we obtain the relaxational dynamics in the form

d

hq hq

fi ~~ ~~~'~°~~~~~ ~~ ~~~~
Pq Pq
The explicit expression for the matrix of kinetic coefficients r, which together with the energy
matrix E contains the full information about the dynamics, is given in Appendix A. Note
that this matrix couples the average density pq to both hq and pq. We can now calculate the
relaxation times as the inverse values of the eigenvalues j, of r E. As shown in Appendix A,
the dynamical correlation iunctions can thus be expressed in the form

3

(hq(t)h((0)) = (hqh()~£A)e~?'~~~~ (13)
1=1
3

(pq(t)pj(o)I = (pqpj)~ £ Aie-?.~~)~,

(14)

1=1

Here, (..)~ are the static correlation iunctions given by equation (7), and the amplitudes
Al'~ sum up to unity, £, At =

£,A[=

I, to ensure the fluctuation-dissipation-theorem.
The explicit expressions for r

are too complex to allow for an analytical calculation of the
eigenvalues and eigenvectors of r E. It turns out, however, that most of the numerical results
for the full problem can be understood in terms of two previously considered limiting cases
[21, 11, 12].

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3. Free bilayer membrane.

The case of a

free bilayer membrane, which formally corresponds to lo - cc, has been inves-
tigated in reference [21]. We will recall the results briefly, and, as a new result, calculate the
correlation functions for this case. Due to the up-down symmetry, the fluctuations of the den-
sity average p decouple from h and the density difference p. The matrix of kinetic coefficients
for h and p is diagonal,

1/4qq 0

r~(q) % q~ ~~~~

~

2(2b + 2qq + pq~

while the energy matrix E~ is just the upper left (2 x 2) submatrix of E

as defined in equation
(4).
The product r~(q)E~(q) has two eigenvalues j(~(q), which yield the dispersion relations
shown in figure la. For simplicity, in this section, wi discuss only the case of vanishing tension.
There are three regimes [21], separated by the crossover wavevectors q(e 2qk/b~ [26] and
q[e @%,

~

3

fi~' q«qi
~

~

2 f

~q2 ~~ ~

'~ ~
2b k~ '

~l < q < q(~f ~

2b '

ql

k

~

~ ~
~

3 f

(16)

j I' q(< q

4q~ '

qi « q.

The wnplitudes At and A(of the contribution of the two modes to the correlation functions
(hq(t)hq(0)) and (pq(t)pq(0)) (see Eqs. (13), (14)), as calculated numerically, are shown in
figure 16.
For q < q(, there is the "classical" bending mode of a

free membrane damped by bulk
viscosity, as given by j(, as well as a second mode due to interlayer friction, which decays fast.
The asymptotic behavior of the correlation functions can be derived as follows. Undulations
relax only by the slow mode, so Al m 1, and Al is negligibly small, since the height variable
is too slow to follow the fast density fluctuations. Density differences between the monolayers,
however, relax by both modes. On time scales longer than 1/j(, the density relaxes as to
minimize the free energy for given undulations. This condition, dF/dp()~ = 0, implies pq =

q

dq~hq, and therefore

~~~~~~~~~°)) * d~q~ (h~(t)hj(o)j ~_

kBTd2 ~~~

~

~~~Y ~~ ~ (PP~)o ~~ e~?'t ~~
From this relation and equation (14) we obtain the coefficient A(m 2kd~ IA for long times
t » 1/j(, for which it is determined solely by the coupling to height fluctuations. Consequently,
A(m 1-A(= ~

IA, so that the ratio of the amplitudes of both modes in the correlation function
is given by the ratio of the two contributions to the renormalized bending rigidity (5).
At q(the two damping rates become comparable, so that for q » q(the density relaxation
is the slowest mode. Density fluctuations will therefore not be induced by the quickly decaying
undulations, so that A(m 1. Fast height fluctuations take place at unrelaxed density difference,

which leads to a

renormalization of the bending rigidity from ~ to k for the faster mode il.
Height fluctuations induced by density differences relax quickly to hq = 2kdpq/kq~ (as can

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(al

'm

j"~
~~

~

~

qi q2

4 5 6 7 8 9 10

'°g q / m~~l

(b)

"

0.1
h
,
,
,
,
,

~~

~, A~
,
,
,
,

~l ~2

,
,

4 5 6 7 8 9 lo

log q / m~~)

Fig. 1. (a) Dispersion relations for a

free bilayer for ~ =

10~~~ J, k

= 0.07 J/m~, d

= 1

nm,
q =

10~~ J s/m~, p =

10~~° J s/m~, and b =

10~ J s/m~; the dashed lines indicate the asymptotic
behavior ii * ~q~/4q and j2 * kq~/2b, for small q; j2 * kq~/4q and ii * kq~~/2bk, for large
q, respectively. (b) Amplitudes of the dynamic correlation functions for a

free bilayer as defined in
equations (13, 14). The ratio ~

Ii ci 0A17 determines the asymptotic behavior of both the amplitudes
A[for small q

and Al for large q.

be calculated from dF/dh() = 0), which yields At m 2d~k/k, similar to the calculation

Pq

displayed in equation (17).
Finally, for q » q(interlayer friction is replaced by monolayer surface viscosity as the
dominant in-membrane damping mechanism in j([27]. This crossover, however, does not
affect the amplitudes in the correlation function.
An important consequence of these results is that any experiment measuring (hq(t)hq(0))
will also pick up a contribution from the slow density mode for q > q(, provided it is sensitive
to the time-scale I/j(. In particular, the slowest time-scale in this regime will scale as q~~
rather than as q~~ In fact, recent measurements of the undulation modes on lipid multilayers
using a neutron spin-echo method can be interpreted in this way [28]. For q < q(, however,
the fast membrane-damped mode is not visible in the height correlations.

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4. Bound membrane without internal modes.

Neglecting the bilayer aspect and the in-plane density modes one finds the relaxational behavior
of a bound incompressible membrane as calculated in references [11, 12]. This limit can also be
obtained analytically from the full solution as obtained with the formalism of section 2. The
damping rate, is, of the single mode can be split in a product of a dynamical part r~ and the energy Eo (cf. Eq. (6)),

i~(q,io,fz,f~l =

rS(q,iolEo(q,fz,f~l =

r(q,iol L [f2~q~ + q~ + fi~] (18j

For wave-vectors below q =

fj~ a @fl the dominant contribution to the undulation
energy arises from the potential. Above q =

fj~ tension dominates the potential, until bending
energy dominates above q =

f~~ +

fi. If the tension is too small to produce a

tension-

dominated regime, I.e. for L <

/&, there is only one crossover at q =

f~~ with f

+
(fl/~)~H.
The hydrodynamical flow fields fall of as q~~ perpendicular to the membrane, cf. equation
(9). Consequently, for q < l~~ the presence of the wall modifies the damping. This crossover
shows up in the kinetic coefficient

~~~~'~°~ (q
sinh~(qlo)

~~~~~)l~h(~~~~~sh(qlo)
+ (qlo)

~~~~

~_

l(q~ /12q, q < 1/lo

~~
'~

l/4qq, q » I /lo,

where the q~-dependence for small q can be understood as the effect of conserving the enclosed
fluid volume.

Depending on the relative size of the crossover length-scales lo, ix, and f~, different cases have
to be distinguished for the q-dependence of the damping rate j~. For q < min(l~~, f£~, fj~),
one finds
~ ~

j~ m

~~°~ (21)
12q

The asymptotic behavior for large q is that of a

free membrane with j~ m ~q~/4~. In the
intermediate regime, the damping rate depends on the relative magnitude of lo, ix and f~.
The asymptotic behavior in the various regimes can then be obtained by multiplication of the
respective dominant terms of P and Eo.

5. Results for the bound bilayer.

In the iull problem, the presence oi the wall breaks the symmetry between p+ and p~ There-
fore, the dynamics couples both variables p

and p to the fluctuations of h, which leads to three
coupled modes. As will be discussed in Appendix B, however, the coupling to the >mode is
typically weak, which allows to discuss the relaxation times and correlation functions for h
and p as if they were decoupled from p. The remaining two modes are those already discussed
above: a hydrodynamic mode arising from the damping of membrane undulations by bulk
viscosity and a density mode damped by in-membrane friction.
Due to the number of length-scales involved, there are now many different scenarii for the
relaxation rates. Instead of an exhaustive discussion of all possible cases we will discuss the
general behavior in terms of the mechanisms explained in the preceding sections for the two
limit cases, and present the dispersion relations for three exemplary sets of parameters.

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A useful distinction arises from the criterion whether or not there is a crossover vector qi
separating a small-q regime, where the density adjusts to the height fluctuations, from a

large-q
regime, where the density cannot follow the fast height fluctuations, whereas the height adjusts
to density fluctuations. In analogy to the free bilayer discussed in section 3, such a crossover
vector qi would be given by the implicit equation

~~2 7~(qi) " #, (22)

Since j~ (cf. Eq. (18)) follows regimes not present for the free bilayer, qi will in general differ
from the value q(. A crossover vector qi exists, if the criterion

fll(< 6kq/b (23)

is fulfilled, as follows from the small-q behavior of is (21).
For any given potential V(I), this criterion can, in principle, be checked. However, it is known

from renormalization group calculations on the interaction of membranes with substrates that

fluctuations can renormalize such a potential [1, 22], which makes the direct evaluation of
criterion (23) difficult. A crude estimate can be obtained as follows. The free parameters on
the right side of this criterion are independent of the potential. Typical values are k

=
0.07

J/m2, q =

10~~ J s/m~, b

=

108 J s/m4. Since fll(amounts to an adhesion energy Vo,
relation (23) becomes %lo < 10~~~ J/m. With typical adhesion energies below lo~~ J/m2,
and substrate distances below 100 nm [29, 9] this criterion will thus be met in most physical
situations.

We will now discuss the dispersion relations and the correlation functions for three parameter
sets. For the first two, the criterion (23) is fulfilled; in the third example we present a case

where it is violated.

S-I FIRST EXAMPLE: SMALL TENSION. A first example is shown in figure 2: parameters
for this case are chosen to follow an experimental example [9]. We consider a giant vesicle in
weak adhesion with the fluctuating part facing the wall in an intermediate distance lo Cf 50 nm.
Tension in such a case is small. The curvature of the potential can be estimated via fl =

nf~4
(or fl = Lf~~) from the measured real-space correlation length of height fluctuations, which
is of the order 0.5 pm. The remaining material parameters as given in the figure caption are
chosen as for the free bilayer case.
For q < qi, the density difference relaxes quickly and thus follows the undulations. The
damping rate for the undulations, as given by ii, runs with increasing q

through the crossovers
fj~ and f~~ already present for a single layer near a

substrate, discussed in section 4. Due
to the coupling of height and density, the undulations which relax slowly with damping rate
ii induce density correlations. These can be calculated analogously to equation (17) with the
result

(24) 2kd~q~ Aim (q « qi)

j 4 ~ ~q2 +Q

So, ior q « f~~, A[effectively vanishes, and A[m 1, whereas at wave vectors (~~ < q < qi,
one observes the low-q behavior of the free bilayer without tension.
The value of qi m

(6kq/b~l~)~/4 is close to l~~ in this example. Thus for q » qi one recovers
the behavior at q » q(of the free bilayer as in section 3. In particular, the relaxation of
density differences is too slow to follow the undulations and therefore the bending rigidity is

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(a)
I

~

Yl
~ 4

~

,," j-i j-i

,' K

5 6 7 8

log q / m~~l

(b)
i

,

',,
j~P"
2 ',
,
,

1
1
1
1
1

/
i I

j j

' j~i~ J

'~l

5 6 7 8

log q / m~~)

Fig. 2. (a) Dispersion relations for a bound bilayer; case of weak adhesion and low tension. ~ =
0.5 x

10~~~ J, k

=

0.1 J/m~, d

= 1

nm, q =

10~~ J s/m~, p =

10~~° J s/m~, b

=

5 x

10~ J s/m~,
Q

=
4 x

10~ J, E

=

10~~ J /m~, and lo #

50 nm; The dashed lines indicate the asymptotic behavior in
the various regimes, I-e- kq~/2b and kq~/4q for12; Ql~q~/12q, El~q~/12q, ~l~q~/12q,

and kq~~/2bk
for ii Evaluation of qi using the full damping rate from equation (18) gives qi "

1.45 x

10~ m~~.

The relaxation mode j3 of the average monolayer density p decays much faster. (b) Amplitudes of the
dynamic correlation functions for a bound bilayer as defined in equations (13, 14). Parameters used
here give ~/k ci 0.2 for the large-q behavior of Al-

renormalized. As above, it is possible to calculate the asymptotics of the amplitudes of the
dynamic correlation function in this regime, giving A[m 1,

~~

~

kq~

~~~

+ Q

~~ ~ ~~

'

~~~~

~~ ~ ~j ~z i Al For q > (~ ~>

~~~ ~~P~~~~~~ ~~ ~~~~~~~ ~~ ~~~~~~

5.2 SECOND EXAMPLE: LARGE TENSION. A second example, also motivated by an ex-

perimental situation, is presented in figure 3. Here, we consider a pancake-like vesicle which
is strongly bound to the substrate. The upper side of such a vesicle is flickering, while the

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Y3

7'~ ,,"' Y2 (~)
fl ,"' ,,'
~ ,,"
~

,,"" Vi

~

qi

log q / m~~)

~~~

A(

A(

A~

I
I
I
' ,,--,

' , ',
' '

,

' '
,' ',
' '

' ,

', ' ',

~P" ,' '
2 ', ,'
',, ,'

5 7

log q / m~~)

Fig. 3. (a) Dispersion relations for a bound bilayer; case of large tension. Parameters as in figure 2,
except for E =

2

x
10~~ J/m~, lo " 400 nm, Q

= 0. The dashed lines indicate the asymptotic behavior
in the various regimes as discussed in the text: the in-membrane damping rate changes from kq~/2b to
kq~~/2bk, while the bulk damping passes the asymptotical behaviors El~q~/12q, Eq/4q, and kq~/4q.
(b) Amplitudes of the dynamic correlation functions for a

bound bilayer as defined in equations (13,
14). The ratio ~/k is as in figure 2, whereas Al is determined by the coupling to density and follows
equation (25) for q » qi

lower side is essentially fixed by the strong adhesion. We assume that the no-slip boundary
condition at the wall, which now is coated by the fixed bilayer, is still valid (for a modification
see Appendix D). Due to the large distance from the wall, the fluctuating membrane will not
be held in position by a potential, but rather by the constraints on area and volume of the
vesicle. Thus fl may be neglected. Because of the strong adhesion, tension is large, but should

be still one order of magnitude below the lysis tension, which is of the order fi [30].

With parameters accordingly chosen as given in the figure caption, qi =

6k~/bLl] turns
out to be very small, and the length scales qi, lp~, and f~~ are well separated. For q < qi,
At ci A[m I, since the coupling of density correlations to the slowly decaying undulations
follows equation (24) and thus effectively vanishes for q « (£~
For q » qi there is a

large tension-dominated regime, where the damping changes from

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j~ m Ll~q4 /12~ to j2 G3 Lq/4~ at q =

l~~ The height correlations induced by the slow density
mode follow equation (25), and thus are negligible for q < (£~ Only above q =

/fi does
the (for all q > qi renormalized) bending rigidity dominate the undulation energy. From the
form of A), (Eq. (25)), it can be seen, that only in this regime the height correlations are
influenced by the slow density mode, whose damping rate ii is then shifted by a

factor ~/k.
Thus, in this example observable effects of the renormalization of bending rigidity set in at a
larger value of q

than where the interchange of slow and fast mode takes place, contrary to the
free bilayer case.

5.3 THIRD EXAMPLE: sLow DENSITY RELAXATION FOR ALL q. If the condition (23) is
violated, the density relaxation is always slow and there is no crossover wave-vector qi Density
differences cannot be induced by fast height fluctuations, resulting in a negligible Al. The
height correlations induced by the coupling to the slowly decaying density mode, as expressed
in A), are now given by equation (25) for all q. As we have seen in the second example,
however, this turns out to be an observable effect in the height correlation function, I.e. A) is
of order unity, only for q »

/fi.
As an example, we consider a charged membrane pushed by a

linear potential (e.g. arising
from an osmotic pressure) towards the substrate. In weak electrolytes, where the screening
length is large compared to lo, fluctuations beyond the harmonic level can safely be ignored.
The potential then reads V(I) =

irkBT/(21B ii + pi, where lB Cf 0.7 nm is the Bjerrum length in
water. For this potential, fll(=

irkBT/lB Cf 1.8 x

lo~~~ J/m is independent of lo and indeed
violates the criterion (23).

For the relaxation rates, a

large fl means that there will be no tension-dominated regime.
The two modes follow the small-q-behavior of is m fll(q2 /12~ (21) and j[m kq2 /2b (16), until
at q >/ ((~~ +

(fl/~)(~/4), l~~) the large-q behavior of the free bilayer is recovered again.

6. Conclusion.

We have calculated the relaxation modes for a bound membrane with additional degrees of
freedom due to monolayer density fluctuations employing linearized hydrodynamics. The pres-
ence of the wall and the undulation energy contributions by potential and tension give rise to
a number oi cross-over length-scales and affect not only the damping oi undulations, but also
the relevance oi the coupling of bending and density modes.
The coupling between density and height variables becomes relevant to the height correlation
function beyond a cross-over q-vector given by the maximum of qi (as defined in Eq.(22)), 1If
and I /f~ (as defined in Sect. 4). Since this cross-over typically is larger than the corresponding
value of a

free bilayer membrane, the coupling between shape and density will be more difficult
to detect for a bound membrane. For typical values, as used in the examples discussed above,
this cross-over corresponds to a wave-length which is at the lower end of the optically accessible
range (compare Fig. 2) or even smaller as in figure 3.
If the coupling between shape and density can be neglected, the correlation function for the
height variable is given by

kBT ~-,s(q)t (hq (till] (°I) (26) "

L~Ly(~q4 + Lq~ + ill '

with j~(q) defined in equation (18). Since the tension in the membrane can cause a crossover
in the optical range as it happens for weak adhesion shown in figure 2, the relaxation rate does
not obey a simple power law for a

large q-range. In such a case, a

fit against the expression (26)
is necessary. This expression contains in js(q) two more parameters, namely the viscosity ~

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and the separation lo, than the corresponding static correlation functions, I.e. the mean square
amplitudes as given by the preexponential factor in (26). Since the viscosity is known, the
dynamical measurements could be used to obtain the separation lo from these data. Likewise,
if this separation is measured independently, the dynamical data provide a much better basis
for a determination of the curvature fl of the adhesion potential and the effective tension L.
In any precise determination of these quantities from experimental data, one has to be aware
of additional modifications some of which we discuss in Appendices C and D. In Appendix C,
we present the correlation functions as appropriate if data have been obtained only for a
linear strip. As shown in Appendix D, the relaxation rate will also be slightly modified if the
substrate is coated with another bilayer. There we also point out that for multilamellar vesicles
a peristaltic mode causes another slow time-scale. As a consequence of all these complications,
a quantitative comparison with experimental data available so far [9] has not yet been achieved.
The full analysis presented in this paper will be mandatory for the interpretation of any ex-
periment in the cross-over regime where the height correlation function will show bi-exponential
behavior. In the two examples presented here, the two time-scales are well separated even in
the cross-over region but different parameter sets may lead to a closer gap between the two

modes.

There are, in principle, two possibilities to reach this cross-over regime where the coupling
between shape and density becomes relevant. For weak adhesion, which implies weak tension,
as shown in figure 2, the cross-over is dominated by qi, I.e. the value of the friction coefficient
b. If the friction is increased, the cross-over would shift towards longer wave-length. One
may speculate whether close to the main transition this friction becomes larger due to the
freezing of the chains. Likewise in mixtures of lipids with different chain lengths, this friction
may be enhanced. Secondly, one could contemplate grazing incidence neutron reflection or,
at the present stage even more speculatively, X-ray microscopy, to gain access to wave-vectors
beyond 10~/m. In fact, the recent analysis of a neutron spin echo experiment [28] on a stack
of membranes has given strong indication for the relevance of the coupling between height and
density difference for q ci 108 log/m.

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and H. Strey.

Appendix A.

Explicit expression for r and calculation of modes.

The matrix £

as defined in equation (12) has been calculated analytically using the computer
algebra system Maple [31]. The result for the matrix elements then reads

~~~

lllllllll~lllllllllllllll~~~~~~+~~~~jjj~

F12 " F21 "

(~ l/ + /L ~) ~~ l~ / l~ ~~~~
r13 =

r31 = (2 b + 2

q q + p q~) q~l((cosh(loq) sinh(loq)) /Y2 (29)

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F22 " ~~~ (~/L10~~ SlDll(10~) /L l~~~ COSII(10q)~ + /L10 ~~ C°S~I(~0~) +

/L i~ q~ SID~I(10~)

+
~l COSII(10~) + /L ~ SlDll(10~) +

~l SID~I(~0~)) / ~2 (30)
£~3 = £32 = n q~ (1+ 21(q~ 2 loq) (cosh(loq) sinh(loq)) /Y2 (31)
~33 " ~~ ((~l ~ + /L l~~~ + /L ~~ + 2 l~~~ + 2 ~ ~10~ /L10~~) SID~I(10~)
+ (~ q + p lo q~ 2 b I(q~ p I(q~ + 2 b lo q) cosh (lo q)) / Y2 (32)

where we have defined
Yi = 8~2 lo q~ + 4

~ p q~ 8 ~~ l(q~ + 4p b I(q~ + 4

p b

q 4p q

cosh~ (lo q) 4p b loq~
-2 p~ loq~ + 2 p~l(q~ 8 b

~ cosh~(loq) 8 ~~q cosh~ (lo q) 2 p~q~ cosh(loq) sinh(loq)
- 8 ~ b cosh(loq sinh(loq) 2 p~ q~ cosh~ (lo q) + 2 p~q~ 4

p b

q cosh(lo q) sinh(loo)
- 8 ~ /1q~ cosh(loq) sinh(loq) + 4

~ b 8 ~~q sinh(loq) cosh(loq)
- 8 ~ p q~ cosh~(ioqj,

(33j
Y2 = (2 b

q + 4 q2 lo q~ 4 q~ I(q~ + p~ q~ + 2

q p q~ + 2

p b I(q~ 2

p b lo q~

+ 2 pbq + p~l(q~ p~loq~) 2 sinh(loq)
+ (2 b

n 4 n2 to q2 + 4 n2 q + 4 n21(q3 + 2

n p q2 2

p b

11 q3 + 2

p b to q~

/12 1(q~ + /1~loq~) 2 cosh(loq) (34)
Having obtained the matrix of kinetic coefficients, we can now calculate the dynamical corre-
lation functions. A formal solution to the relaxational dynamics, equation (12), is (hq, pq, pq (t)
= exp (-£ E t) (hq, pq, pq) (0). After diagonalization, one obtains the relaxation times oi the
various modes as the inverse values of the eigenvalues j, of £ E.
The time-dependent correlation functions can be obtained from the corresponding eigenvec-

tors g,. The matrix U =

[gi g2 g3) with matrix elements U,j diagonalizes £ E. The dynamical
correlation matrix G(t) is defined by

hq 11 0 0
G(t) e pq (t) (hq, pq, pq)* (0) =

e~~'~ ~Ga = U exp 0

j2 0 t U~~ Go
fig ° °

3~

(35)

In order to obtain the static correlation functions for t

- 0

we have used the fluctuation

dissipation theorem G(t = 0) = Go =

(kBT/L~L~) E~~ For the matrix elements of G(t),
equation (35) reads

Gjk(t) = £e~?'~Uj,£(U~~),m(Go)mk (36)

e j~e~?'~A(~(~o)jk (37)

where the condition G(t = 0) = Go is equivalent to £,A(~ =

l. Here, the indices (1,2,3)
correspond to (h,p,p), I.e (hq(t)h((0)) = Gii(t), and so on. For convenience, we use At
instead of A)~, and A[for A]2.

Appendix B.

Reynolds numbers and coupling to p.

The Reynolds number, Re, is a measure for the relevance of inertial and advective terms in the
Navier-Stokes equation as compared to the friction term. For decaying plane waves it can be

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estimated by replacing the spacial derivatives by the wave-vector q, and the time derivative by
the relaxation rate j, leading to Re = pi /qq~. Calculating the Reynolds number for the two
slowest modes ji,2 gives Rei,2 < 1, thus confirming the validity of the Stokes approximation,
(Eq. (811.
The coupling of h and p to p, expressed in the amplitudes of the correlation function A(
and A(, turns out to be negligible for all reasonable parameter values. These amplitudes are
always less than 10~5 for the examples discussed in section 5. We conclude that the error we
made by treating also this mode within the Stokes approximation should not be harmful to
our results. For the >mode, we find j3 G3 2klq2 In for small q, and j3 G3 kq/(2q + pq), the
result obtainable analytically for the free bilayer as well, for large q. However, these results are
non-physical, since the Reynolds number for this mode, estimated as Re3 = p73/nq~, turns out
to be bigger that 1. A proper treatment of this mode would then reveal oscillatory "sound"
modes within the membrane [21].

Appendix C.

Measurement of correlations in a

linear strip.

In experiments using video microscopy, membrane displacements are frequently measured in
a
finite strip of length L parallel to z = o, averaging over the width B of the strip [32, 9]. B
will most often be determined by the optical resolution of the microscope. Thus the observed
quantity is
1 B/2
H(z) =

/

h(z,y)dy (38)
B
-B/2

Defining the Fourier transform as H(z) = £~ Hq~ exp(iq~z), one obtains from equation (38)

~~~x £

(~)qx,qv)

~ ~~~j~~~~~ ~ ~'~x,~v) ~~

~~~~
'

~~~~

q~ qv

for the static height correlation function, where the correction factor K~(q~) arises from the
Fourier-transform of the finite strip. For q~ < B~~, this factor reduces to unity. Projection
on the strip is then equivalent to integration over all possible values of the component of the
wave-vector perpendicular to the strip.
For an evaluation, we replace the sum by an integral, £~~ -

(L~/2ir) fZ~ dq~. Using

equation (7) for (h)~ ~ ~) we obtain in the limit B

-

0
x> v

kBT (~~/~ Sin)

arCCOS(C)) ~

2

~~ ~~ ~ ~ ~~~ ~ ~~~~~~~~ ~

~~ ~ ~~~

~qx 40)

kBT /~ /l) ~

L~ 2L2/~-8fl D+

~

D-

'~ ~~~~'

with
L + 2

~ q~~

~ (41)
2@jfl+Lq~~+~q~~'

and

D~ e L + 2

~ q~~ + L2 4

~

fl (42)

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In the special case without a potential, fl = 0, this expression has been previously derived by

Mutz and Helfrich [32]. For small q~, the function (H(is constant. For fj~ < q~ < (j~ it
behaves as (aq~)~~, and for q » (£~ it behaves as (~q))~~
The expression for the dynamical correlation function in the same geometry yields

(Hq~(tiHi~ (oil =

)

/dq~ (h(q~,q~)(tihiq~,q~) (oil =

)

/day (hlq~,q~))~ LAte-?.~~~ ~v~~

1

(43)
which has to be evaluated numerically. Thus, for every particular wave-vector q~, a superpo-

sition of relaxation rates contributes to the correlation function.

As an example, we have performed the q~-integration of the dynamical correlation func-
tion, equation (43) for the "classical" case of a

free membrane without internal degrees of
freedom. For ~t w ~q]t/4q < 1, the result is governed by the static correlations on the strip,
(Hq~(t)H((0)) +~ te~"/4q~t =

e~"/(~q]). However, for ~t » I, we obtain (Hq~(t)H(~(0)) ~
te~"/4q~t~)2 =

2q~/2e~"/t~/2(~q])~/2, I-e- the correlations on the strip fall off faster with q~
than in (h(q~, q~ = 0, t)h(q~, q~ = 0, 0)).

Appendix D.

Extensions of the model.

In this section, we want to discuss briefly some simple extensions of the model applying to
somewhat modified physical situations. So far, we have assumed a

smooth solid wall with

no-slip boundary condition. If the interface between substrate and adjacent fluid is coated by
another lipid monolayer or bilayer, as is also the case for strongly adhering "pancake" vesicles
(see example 2), it may be necessary to allow for a

finite slip at z =

0. This is done by
introducing a

friction term c~ [u~(z =0) with a phenomenological friction coefficient in the
force-balance at the substrate that replaces the condition u~(z=0) =

o. It turns out that, for
small q, this leads to a correction factor (4 +

[lo/n)/(I +

[lo/n) in the damping rate (18) of
the undulation mode. The q2 dependency of the damping, however, remains valid, since the
slip does not change the conservation of volume between membrane and substrate. For coating
with a bilayer, we may use [

m b

=

lo8 J s/m~, which leads for lo Ci 50 nm to [lo In Ci 5 x

lo~.

In this case, the correction due to the slip is negligible. In the low-friction regime, that is for
b < lo In, the correction can reach a

factor of 4.

In experiments, very often multilamellar vesicles are observed. In the case of adhesion,
they may be described as a

stack of membranes near a wall separated by small amounts of
water [33]. Thus, for small q, the flow fields of the membranes affect each other leading
to a modification of, the damping rates. As a simple example, we consider two membranes
without internal degrees of freedom as discussed in section 4. Their distances to the wall will
be denoted by li and 12, respectively, with a mean intermembrane distance Al < (li,12).
We can expand the membrane-membrane interaction potential Vm(12 li) about Al, with
flm a d2Vm /d(12 li)~(i~-ii=m For q < (£~, the two modes for two independent membranes
with j m fll)q~ /12q (see Eq. (21); I

= 1, 2) are replaced by two collective modes. The faster
mode has essentially twice the undulation energy of a single membrane, while the second one,
the "peristaltic" mode [3], can become very slow with

a ~i~3 ~2
11 "

~'

~~ ,

144)

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since the length I, is replaced by Al. Both modes lead to appreciable height fluctuations and
should be visible in optical experiments.

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lo~~

m/s [34]. Converting the hydrostatic pressures to the concentration difference of a

solute with

the same osmotic pressure, we obtain a net flux of solvent through the membrane dQ/dt =
P6p/RT m

10~~~ kg /m~s. This is equivalent to a

relative velocity of membrane and solvent of
10~~~ m/s which is totally negligible in comparison with typical membrane velocities of the order
jh m

10~~ m/s. (A similar argument can be found in Youhei F., Pl~ysica A 203 (1994) 214) If
transient pores are created dynamically by shear forces, one has to replace equation (10) by two
seperate force balances for the membrane and the solvent at lo- In the simplest approximation,
this gives rise to a q-independent term in is. Only for much higher permeabilities than measured
statically one expects measurable effects.

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factor ~

Ii for the
sake of better consistency with the respective definition for the case of a

bound bilayer.
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