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Dynamics of Rayleigh-like Instability Induced by Laser Tweezers in Tubular Vesicles of Self-Assembled Membranes

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Abstract. — We present a theoretical study of the Rayleigh-like, "pearling", instability recently observed in tubular vesicles of bilayer membranes under the action of optical tweezers. The tweezers are argued to pull the membrane into their operation zone, thereby inducing a tension in it. Consequently, the membrane can respond in a Rayleigh-like instability, in which its area decreases. Our approach is based on a linear stability analysis of the deformations. However, because of the non-linear coupling existing between the suction process and the growth of the unstable modes, the instability develops different characteristics. At long times, the growth of the instability is controlled by the suction rate of the tweezers, assumed constant in time. The wavelength \( \lambda^* \) of the most unstable mode is time-dependent at short times it decreases as \( \sim t^{-1/2} \) at long times it is either constant, of the order of the tube radius \( R \), or increases as \( \sim t^{1/4} \). The amplitude of the most unstable mode increases (at long times) as \( \sim t^{1/2} \). We delineate the different regimes under which these behaviours are predicted, and discuss their possible implications on experiment. We also briefly discuss the possibility for other non-linear effects, such as an induced spontaneous curvature in the membrane, which may be responsible for the appearance of very narrow tubes in the late stages.

1. Introduction

The study of the structure and dynamics of self-assembly systems is a field of growing interest [1–3]. In particular, shape fluctuations and instabilities of bilayer vesicles, under equilibrium or metastability conditions, have been the subject of several recent studies [4–12], particularly with respect to the question of budding [5–7]. Much less attention has been given to the question of the response of vesicles to a strong external perturbation which drives them far from the equilibrium situation. Nevertheless, these questions are of primary importance in trying to understand the behaviour of biological membranes in living tissue.

In this paper, we focus on the response of cylindrical (tubular) vesicles to a local tweezing. Metastable cylindrical vesicles of surfactant bilayers can be formed in the laboratory under flow [13]. Their life-time is sufficiently long to allow for studying their response to an external perturbation. Recently, the effect of using optical (laser) tweezers on such vesicles has been
reported [13]. A durable action of the tweezers on the membrane at a local region defined by the size of the laser beam causes the cylinder to deform with a well-defined wavelength of the order of the tube radius $R$. This reaction, which appears even far away from the place of action, is a reminiscent of the well known Rayleigh instability of cylindrical jets and of cylindrical columns in systems of two immiscible fluids [14–16]. In the latter systems the instability is easily explained by the introduction of surface tension alone: since the interfacial area decreases in a volume conserving deformation, the surface energy is also decreased.

The action of the tweezers has been recently addressed [13,17] (independently of us [18]) in terms of an induced surface tension. This is a reasonable assumption, since surfactant molecules have a dipole moment so that their energy is reduced in the presence of a gradient in the intensity of an electromagnetic field. As a result, the tweezers will tend to pull surfactant molecules into their operation zone. The initial action of the tweezers is thus expected to be a simple pulling of the membrane, which will obviously induce a tension in it. As the tension is raised above a certain “critical” value, it can dominate any bending forces that resist a deformation.

However, just as in the case of the simple Rayleigh instability, the area of the bilayer must decrease in this volume conserving deformation. Hence, if the area is fixed by the membrane itself, the instability cannot develop. For the instability to develop significantly, transport of membrane area out of the unstable region must occur, i.e., molecules must be pulled out from the cylindrical part. The action of the tweezers cannot be therefore explained only in terms of an induced surface tension, as previously suggested [13,17]. The tweezers must also continuously suck molecules into their operation zone.

The way these two effects of induced tension and continuous suction act together, can be explained as follows. As the laser is turned on, there appears a force (per unit length) $\sigma_{\text{ext}}$ towards the center of the operation zone acting on the bilayer. The strength of this force is fixed by the laser power. If, hypothetically, the rest of the cylinder outside the operation zone would not respond to the external perturbation, a mechanical equilibrium could be reached when this force is balanced by the (mostly stretching) elastic forces. In such an equilibrium situation, a fixed tension profile $-\sigma_{\text{ext}}$ would be induced in the membrane (the boundary conditions are zero current at the ends.) However, if this tension is large enough to induce the Rayleigh instability, the membrane will begin to react. The resulting reduction in area will reduce the tension and constrain the instability. This negative feedback mechanism appears as a nonlinear coupling between the induced current in the system and the tension. As a result, though the area instability still exists, it develops different characteristics which we discuss below [19].

To be more explicit, we may estimate the maximum surface tension that can be induced by the tweezers [17] as $\sigma_{\text{ext}} \sim E\delta d$, where $E$ is the energy density of the laser beam in vacuum, $\delta e$ is the difference in dielectric constant of water and surfactant tail, and $d$ is the bilayer half-thickness. Typical values ($E \approx 3 \times 10^4$ erg cm$^{-3}$, $\delta e \approx 0.2$, $d \approx 20$ Å) lead to $\sigma_{\text{ext}} \approx 10^{-3}$ erg cm$^{-2}$. For $R \sim 1$ μm and $\kappa \sim 10k_B T \sim 10^{-12}$ erg, $\sigma_{\text{ext}}$ is indeed well above the critical surface tension $\sigma_c \approx \kappa/k^2 \sim 10^{-4}$ erg cm$^{-2}$ needed to initiate the Rayleigh instability (see below).

Of particular note is the fact that the cylinder is not the minimal bending energy shape. Consider a cylinder of length $L$ and radius $R_0$ which is divided into $N_s$ spheres of radius $R_s$ with the same total volume (but not the same area). The latter implies $N_s = 3R_s^2L/(4R_0^2)$. The bending energy of the cylinder (dropping the Gaussian curvature) is $E_c = \pi \kappa L/R_0$ and that of the spheres $E_s = 6\pi \kappa R_s^3 L/R_0^3$. It can be seen that for $R_s > 6^{1/3} R_0 \approx 1.82 R_0$ one obtains $E_s < E_c$ and the cylinder will be metastable against its breakage into spheres. We may apply the same argument to a “necklace of pearls”, i.e., spheres connected by sharp saddle-type
“necks”, so long as the local radii of curvatures at the saddles obey \( R_1 \simeq -R_2 \), providing a negligible bending energy contribution. This “pearling” instability does not show up in the linear analysis (discussed in Section II) since it is a first-order type, which requires to overcome a bending energy barrier. It is apparently never observed in experiment without the action of the tweezers [13], consistent with the fact that it requires a significant decrease of the area (Otherwise, one could expect that it would be initiated by thermal fluctuations).

Our approach is formulated in terms of a suction rate \( \nu \) which controls the decrease of the surface “coverage” \( \phi \) (or increase of area per molecule), with \( 0 < \phi < 1 \) and \( \phi = 1 \), taken by definition to be the equilibrium coverage. For simplicity, it is assumed (somewhat incorrectly) that the suction, and hence the induced tension, are uniform throughout the cylinder. The rate \( \nu \) may be estimated as follows. With the assumption that the tweezers act on a piece of membrane with a force per unit length \( \sigma_{\text{ext}} \), we can estimate the velocity \( v \) in which that piece of membrane moves under steady state conditions. The (membrane) envelope of the cylinder has to flow with the same velocity \( v \) towards the trap, which is equivalent to the flow of water in the opposite direction with the envelope held fixed. Hence, in the Poiseuille approximation this velocity is

\[
v \simeq \frac{R^2}{\eta} \nabla P \simeq \frac{\sigma_{\text{ext}} R}{\eta L} \quad (1)
\]

where \( \nabla P \simeq \sigma_{\text{ext}}/(RL) \) is the pressure gradient along the cylinder. The amount of stretching of the membrane after a time \( \delta t \) is \( \delta L \simeq v \delta t \), and the surfactant coverage \( \phi \) becomes roughly \( L/(L + \delta L) \). Since \( \delta L \ll L \) we have \( \delta \phi \simeq -\delta L/L \equiv -\nu \delta t \) and we find the desired expression for \( \nu \) [20],

\[
\nu \sim \frac{\sigma_{\text{ext}} R}{\eta L^2} \quad (2)
\]

Together with the estimate for \( \sigma_{\text{ext}} \) given above this yields a rough estimate for \( \nu \). It shows that for very long tubes where \( L \gg R \), \( \nu^{-1} \) becomes extremely long compared to other relevant timescales in the problem. Of course, the suction rate is in fact position dependent so that the above estimate may be regarded as an averaged rate.

2. Free Energy Model

Let us assume that the membrane forms a cylindrical shape vesicle of radius \( R_0 \) and length \( L \gg R_0 \) suspended in the solution. The bending free energy associated with deformations of the cylinder can be described by the Helfrich Hamiltonian [21]. Since we ignore topological transformations, we may drop the Gaussian curvature and the Helfrich Hamiltonian is

\[
H_\kappa = \int ds \left\{ \frac{1}{2} \kappa \left( \frac{1}{R_1} + \frac{1}{R_2} \right)^2 \right\}, \quad (3)
\]

where \( R_1 \) and \( R_2 \) are local radii of curvature and \( \kappa \) is the bending modulus. (It is assumed that the membrane has no spontaneous curvature.) In equilibrium the membrane has no tension, but the tweezers induce a surface tension \( \sigma \). In this case the total free energy of the surface is written as

\[
F = H_\sigma + H_\kappa \quad (4)
\]

where

\[
H_\sigma = \int ds \sigma \quad (5)
\]
We consider small axisymmetric deformations of the vesicle from its cylindrical shape conserving the total enclosed volume [15]. We therefore assume that we can describe the deviation from the cylindrical shape by a single-valued function \( u(z) \) which has the property \( \int_0^L u(z) dz = 0 \). The local circular radius \( W(z) \) is written as

\[
W(z) = R + u(z)
\]

so that \( \int_0^L W(z) dz = RL \). Conservation of volume allows us to find \( R \). We have

\[
R^2 L = R^2 L + \int_0^L u(z)^2 dz
\]

Making use of the Fourier transform

\[
u(z) = \sum_q U_q e^{iqz}
\]

we obtain

\[
R^2 = R^2_0 - \sum_q U_q U_{-q}
\]

This result is exact even for large \( u \) so long as it does not exceed \( R \) (in which case the membrane cuts itself).

It will be desirable to calculate the area of the cylinder at a given deformation. For small deformations where \( |\partial u/\partial z| \ll 1 \), the area may be expressed as

\[
S \simeq 2\pi RL + \pi RL \sum_q q^2 U_q U_{-q}
\]

Making use of the volume conservation constraint equation (9) we get

\[
\frac{S}{S_0} = 1 - \tilde{s}
\]

\[
\tilde{s} \simeq \frac{1}{2R_0^2} \sum_q [1 - (qR_0)^2] U_q U_{-q}
\]

Here \( S_0 = 2\pi R_0 L \) is the area of the undeformed cylinder, and so \( \tilde{s} \) is the relative decrease in area.

We can now proceed to calculate the total free-energy for small deformations where \( |\partial u/\partial z| \ll 1 \). Consider first the bending energy, equation (3). In the Appendix we obtain, to order \( U^2 \),

\[
H_\kappa = H_{\kappa 0} + \frac{\pi \kappa L}{R_0^3} \sum_q \left[ \frac{3}{2} - \frac{1}{2} (qR_0)^2 + (qR_0)^4 \right] U_q U_{-q}
\]

where \( H_{\kappa 0} = \frac{\pi \kappa L}{R_0} \) is the bending energy of the undeformed cylinder. The energy associated with surface tension involves only the change in area \( S \) and we obtain the classical (Rayleigh instability) result [15]

\[
H_\sigma = H_{\sigma 0} - \frac{\pi \sigma L}{R_0} \sum_q [1 - (qR_0)^2] U_q U_{-q}
\]
(with \(H_{\sigma_0} = S_0\sigma\)). We define the difference in free-energy densities \(\Delta f\) as

\[
S\Delta f = (H_\sigma - H_{\sigma_0}) + (H_\kappa - H_{\kappa_0})
\]

so that, to order \(U^2\), we finally obtain

\[
\Delta f = \sum_q \left\{ \frac{\kappa}{2R_0^3} \left[ \frac{3}{2} - \frac{1}{2}(qR_0)^2 + (qR_0)^4 \right] - \frac{\sigma}{2R_0^2} [1 - (qR_0)^2] \right\} U_q U_{-q}
\]  

The free-energy (16) implies that fluctuations at all wavelengths are stable if the surface tension is below the critical value

\[
\sigma_c = \frac{3\kappa}{2R_0^2}
\]

If \(\sigma\) is raised above \(\sigma_c\), long wavelength deformations become unstable. For this regime of \(\sigma\) it is convenient to introduce the surface tension difference \(\delta = \sigma - \sigma_c\). In terms of \(\delta\), equation (16) becomes

\[
\Delta f = \sum_q \left\{ \frac{\kappa}{2R_0^3} [(qR_0)^2 + (qR_0)^4] - \frac{\delta}{2R_0^2} [1 - (qR_0)^2] \right\} U_q U_{-q}
\]  

so that small deformations of wavenumber \(q < q_c\), where

\[
q_c^2 = \frac{1}{2R_0^2} \left( -\zeta + \sqrt{\zeta^2 + 4\delta R_0^2/\kappa} \right)
\]

\[
\zeta = 1 + \delta R_0^2/\kappa ,
\]

will grow in time rather than decay (A model for this time dependent growth is discussed in the following sections). For \(\delta << \kappa/R_0^2\) we have

\[
q_c R_0 \simeq \sqrt{\frac{\delta R_0^2}{\kappa}} << 1
\]

while for \(\delta >> \kappa/R_0^2\), \(q_c\) saturates at \(q_c R_0 \simeq 1\). For brevity, from now on we do not distinguish between \(R\) and \(R_0\), and all equations will be expressed in terms of \(R\).

So far we have been concerned solely with the energetic stability of the cylinder. These considerations suggest that the longest wavelength mode (\(\sim L\)) is the most unstable one. However, a different mode may be selected throughout the dynamics, which we consider next.

3. Dynamics

3.1. Hydrodynamic Modes at Constant Surface Tension — Consider the pressure difference between the inside and the outside of the tube. As usual this pressure difference is induced by the elastic forces of the membrane so that [9,15]

\[
\Delta P = P_{in} - P_{out} = \frac{\delta F}{\delta V} = \frac{1}{2\pi WL} \frac{\delta F}{\delta W}
\]

For the undeformed cylinder this equation just leads to the generalized Laplace formula

\[
\Delta P_0 = \frac{\sigma}{R} - \frac{\kappa}{2R^3}
\]
(Note that \( \kappa \) enters with a negative sign.) We denote the pressure difference of the deformed tube as
\[
\Delta P = \Delta P_0 + \delta P
\]
so that, according to equation (22), \( \delta P \) (to linear order in \( u \)) can be obtained from the free-energy density \( \Delta f \) by taking its functional derivative with respect to \( u \),
\[
\delta P(z) = \frac{\delta \Delta f}{\delta u(z)}
\]
In Fourier space this functional derivative simplifies to
\[
(\delta P)_q = \frac{\partial \Delta f}{\partial U_{-q}}
\]
Applying this formula to our free-energy density equation (16) we obtain
\[
(\delta P)_q = -\frac{T(qR)}{R^2} U_q,
\]
where
\[
T(x) = \sigma \left[ 1 - x^2 \right] - \frac{\kappa}{R^2} \left[ \frac{3}{2} - \frac{1}{2} x^2 + x^4 \right]
\]
This result allows us to use previous hydrodynamic calculations for the Rayleigh instability. This involves the following two assumptions. First, we allow for the membrane to be compressible, as done in theories for hydrodynamic relaxation of long wavelength fluctuations in membrane lamellar phases [22]. It is true that the membrane is nearly incompressible on length scales smaller than its persistence length. However, the instability we discuss occurs on wavelengths of the order of microns, and the membrane still thermally undulates on smaller lengths scales (which are not observable in optical microscopy), i.e., at wavenumbers \( q \) larger than \( q_c \). Thus, for the small tensions we consider here, the membrane is effectively quite compressible on the micron scale (see also the discussion following Eq. (43)). Second, we assume that the viscosity associated with the 2D flow of surfactant in the membrane is not too much different from the viscosity of the surrounding fluid, which is also consistent with previous studies [22].

We can thus use Tomotika’s result [16], who considered a cylindrical thread of one viscous fluid (viscosity \( \eta_1 \)) surrounded by another viscous fluid (viscosity \( \eta_2 \)) with no-slip boundary conditions (i.e., the velocity field is continuous at the surface). In this calculation we may safely assume that inertia can be neglected since the wavelengths of interest are much smaller than the macroscopic length \( (\rho \eta^2 R / \sigma)^{1/2} \sim 10 \text{ cm} \), with \( \rho \) – the fluid density. (More accurately [15], it is self-consistently assumed that \( q \gg \sqrt{\omega(q) \rho / \eta} \)). The rate \( \omega \) associated with the decay or growth of fluctuations, i.e.,
\[
U_q \approx U_q^0 \exp[\omega(q)t]
\]
can be obtained from Tomotika’s work [16] as a special case (\( \eta_1 = \eta_2 = \eta \)). The result can be put in the form
\[
\omega(q) = \frac{T(qR)\Phi(qR)}{2\eta R}
\]
where \( \Phi(x) \) is a monotonically increasing function in the regime \( 0 < x < 1 \). As a result, the function \( (1 - x^2)\Phi(x) \) has a clear maximum at \( x = x^* \approx 0.562 \). This implies that the mode
with \( q = q^* = x^*/R \) will grow faster than any other mode and will dominate the shape at long times. More explicitly we find

\[
\Phi(x) = \frac{x (I_1(x)K_0(x) - I_0(x)K_1(x)) + 2I_1(x)K_1(x)}{x (I_1(x)K_0(x) + I_0(x)K_1(x))}
\]

(31)

For small \( x \), \( \Phi(x) \) adopts the asymptotic expansion

\[
\Phi(x) \approx -(C_1 + \frac{1}{2} \ln x)x^2 + O(x^4)
\]

(32)

where \( C_1 \) is a numerical constant, \( C_1 \approx 0.067 \). Roughly speaking, the hydrodynamics leads to \( \omega \sim q^2 \) for small \( q \), making the long wavelength modes grow very slowly, although, according to energy considerations (as described in the previous section), these should be the most unstable modes. This result differs strongly from the case considered by Rayleigh where \( \eta_2 = 0 \), in which the long wavelength modes remain the most unstable. The same is true for the case of a “hollow” cylinder where \( \eta_1 = 0 \). The difference between the two results may be understood in terms of energy dissipation considerations. Consider the buildup of the longest wavelength mode \( (q = \pi/L, u \sim \sin(\pi z/L)) \) When \( \eta_2 = 0 \) (Rayleigh’s case) the tangential velocity at the surface near the tube ends can be finite and close to the central \((z\text{-axis})\) velocity. Since the outer fluid has to move in the opposite direction in order to fill up space, this means that there are indeed infinitely large velocity gradients in the outer fluid, which can be however tolerated since \( \eta_2 = 0 \). As a result, there are only small velocity gradients in the inner fluid and energy dissipation in total is small. In the case \( \eta_1 = \eta_2 \), on the other hand, the tangential velocity is essentially zero at the surface and one has significant velocity gradients in both regions – outside and inside the cylinder – leading to a large energy dissipation and therefore a suppressed growth rate. (We note that the Poiseuille approximation correctly describes the same effect and also leads to \( [9] \omega \sim q^2 \) for small \( q \).)

3.2. Time-Dependent Surface Tension. — So far we have assumed in the discussion that the surface tension does not depend on time. This however cannot be true if the tension is induced by the tweezers, as explained in Section 1. Let us first generalize our results for an arbitrary time-dependence of the surface tension. In this case the growth rate \( \omega(q, t) \) is also time-dependent, however, in the high viscous regime (neglect of inertia) the following kinetic equation

\[
\frac{dU_q}{dt} = \omega(q, t)U_q
\]

(33)

still holds. Its solution is

\[
U_q(t) = U_q^0 \exp \left[ \int_0^t dt' \omega(q, t') \right]
\]

(34)

or

\[
U_q(t) = U_q^0 \exp \left[ \Omega(x, t) \right]
\]

(35)

where

\[
\Omega(x, t) = \frac{\Phi(x)}{2\eta R} \left[ \Sigma(t) (1 - x^2) - \frac{\kappa}{R^2} t \left( \frac{3}{2} - \frac{1}{2} x^2 + x^4 \right) \right]
\]

(36)

with \( x = qR \) and

\[
\Sigma(t) = \int_0^t dt' \sigma(t')
\]

(37)

Suppose that there is a buildup of the tension with time, starting from zero at \( t = 0 \). There is therefore a critical time \( \tau_c \) for which the instability starts to develop; i.e., for \( t > \tau_c, \sigma(t) \)
is larger than $\sigma_c \approx (3/2(\kappa/R^2))$ and the amplitudes start to grow in time rather than decay. At slightly later times, of order $\tau_c$, the integral over the surface tension $\Sigma(t)$ exceeds $\sigma_c t$. For clarity we shall now focus again only on the instability regime. We shall measure the surface tension relative to its critical value, i.e., using $\bar{\sigma} = \sigma - \sigma_c$, and the time will be reset to zero at $t = \tau_c$, without change of notations however. With these definitions, equation (36) transforms to

$$\Omega(x, t) = \frac{\Phi(x)}{2\eta R} \left[ \bar{\Sigma}(t) \left(1 - x^2\right) - \frac{\kappa}{R^2} t \left(x^2 + x^4\right) \right] - \Omega_0(x)$$

(38)

where $\bar{\Sigma}(t)$ is an integral in time over the surface tension difference $\bar{\sigma}$, and $\Omega_0(x)$ is independent of time and will be absorbed into $U_0^q$. (Later on this contribution to $U^q$ will be neglected since it is important only at very early times of order $\tau_c$.)

From equation (38) we observe that the critical wavenumber describing the range of the unstable band is time-dependent and is given by (neglecting $\Omega_0(x)$)

$$q_c(t)^2 = \frac{1}{2R^2} \left(-\zeta(t) + \sqrt{\zeta(t)^2 + 4\bar{\Sigma}(t)R^2/(\kappa t)}\right)$$

(39)

with

$$\zeta(t) = 1 + \bar{\Sigma}(t)R^2/(\kappa t)$$

(40)

reducing to the two limits

$$q_c \approx \sqrt{\frac{\bar{\Sigma}(t)}{\kappa t}} \ll 1/R$$

(41)

for $\bar{\Sigma}(t) \ll \kappa t/R^2$, and $q_c \approx 1/R$ for $\bar{\Sigma}(t) \gg \kappa t/R^2$.

The wavenumber of the fastest growing mode, $q^*(t)$, is also, in principle, time-dependent. Obviously, a definition of $q^*$ is primarily relevant for the case where $\Omega(q, t) \gg 1$ (for $q \sim q^*$), where mode selection is sharp. Indeed, in this case $q^*(t)$ may be obtained correctly by maximizing $\Omega(q, t)$ instead of the more exact maximization of the product $\omega(q, t)U_q(t)$. For $\bar{\Sigma}(t) \ll \kappa t/R^2$ we may assume $q^*R \ll 1$ and use $\Phi(x)$ to order $x^2$ only. The wavenumber $q^*$ is then obtained as a solution of the equation

$$(q^*)^2 \approx \frac{\bar{\Sigma}(t)}{\kappa t} \frac{1 + 4(C_1 + \ln (q^*R)/2)}{1 + 8(C_1 + \ln (q^*R)/2)}$$

which in the limit $q^*R \to 0$ leads to

$$q^*(t) \approx \sqrt{\frac{\bar{\Sigma}(t)}{2\kappa t}} = \frac{q_c}{\sqrt{2}}$$

(42)

When $\bar{\Sigma}(t) \gg \kappa/R^2$ we obtain again $q^*R \approx 0.56$, independent of time.

3.3 A SPECIFIC MODEL. — We now wish to deal with a specific model for the effect of laser tweezers on cylindrical vesicles. We introduce the assumption that the tweezers only suck out surfactant molecules from the membrane. Removing surfactant molecules from the membrane does not correspond exactly to removing part of the membrane itself, since there should be a time delay for the area to respond to a change in the 2D surfactant concentration. It is the osmotic compressibility, namely the dependence of surface tension on concentration, which controls the nature of this response [19].

Given a certain concentration $c_a$ of surfactant molecules in a single monolayer film (of the two films composing the bilayer), we now define the surface “coverage” as $\phi = c_a a_0$ where
$\alpha_0$ is the equilibrium area per molecule. When the membrane is in equilibrium, its (true) surface tension $\sigma$ vanishes and the equilibrium coverage is, by definition, $\phi = 1$. For $\phi < 1$ the surface tension is non-zero (positive). The coverage corresponding to the critical surface tension $\sigma_c = (3/2)\kappa/R^2$ is denoted as $\phi_c$. Assuming (self-consistently, as will be shown later) that $\phi$ remains close to $\phi_c$ at all times, we may linearize the surface tension near $\phi_c$:

$$\bar{\sigma} \simeq \sigma_0(\phi_c - \phi)$$  \hspace{1cm} (43)

Here $\sigma_0$ is essentially the compression modulus at coverage $\phi_c$: denoting $a$ – the area per molecule, and $\Pi$ – the surface pressure, the compression modulus is $B = -\alpha \partial \Pi/\partial a = \phi \partial \Pi/\partial \phi = -\phi \partial \sigma/\partial \phi$, so that $\sigma_0 \phi_c = B(\phi_c)$.

We note in passing that the small surface tensions assumed here imply that they correspond to the thermal undulations regime [3, 13, 23]. When the membrane is pulled out at surface tensions below $\sigma_c$, the thermal “ripples” are being stretched – corresponding to an increase of the projected area per molecule – which decreases the surface entropy and therefore increases the effective tension. For $\sigma > \sigma_c$ the situation is more complicated because the long wavelength axisymmetric modes are unstable, whereas all other modes still experience thermal undulations. We can take this effect into account by redefining $\phi$ as the projected coverage, rather than the real coverage [24].

For simplicity we assume that the surfactant is sucked uniformly from the membrane, and at a constant rate. If $J$ is the suction rate of molecules, then, by definition, the change in the number $N$ of molecules in the membrane per unit time is $dN/dt = -J$, which may be transformed to

$$\frac{d}{dt} \left[ \phi(t) \frac{S(t)}{S_0} \right] = -\nu$$  \hspace{1cm} (44)

where the (coverage) suction rate $\nu$ is $\nu = J\alpha_0/S_0$. (Note that in Section 1 we have made use of a different route and estimated $\nu$ directly.) Integrating equation (44) and using the definition (12) for $\bar{s}(t)$ we obtain

$$\phi(t) = \frac{\phi_c - \nu t}{1 - \bar{s}(t)}$$  \hspace{1cm} (45)

On the other hand, we can evaluate $\bar{s}(t)$ in terms of the surface tension $\bar{\sigma}(t) = \bar{\sigma}[\phi(t)]$

$$\bar{s}(t) \simeq \frac{1}{2R^2} \sum_q [1 - (qR)^2] \langle U_q^0 U_{-q}^0 \rangle \exp \left[ 2 \int_0^t \omega(q, t') dt' \right]$$  \hspace{1cm} (46)

It is a good approximation to assume that, within the time period in which the (true) surface tension increases from zero to its (vanishingly small) critical value, the undulations correlation function does not change from its value at equilibrium. Hence

$$\langle U_q^0 U_{-q}^0 \rangle \simeq \langle U_q U_{-q} \rangle_{eq} = \frac{R^3}{2\pi L} \frac{k_B T}{\kappa \gamma(qR)}$$  \hspace{1cm} (47)

$$\gamma(x) = \frac{3}{2} - \frac{1}{2} x^2 + x^4$$  \hspace{1cm} (48)

This leads to

$$\bar{s}(t) = W_0 \int_0^\infty dx \frac{1 - x^2}{\gamma(x)} \exp \left[ 2\Omega(x, t) \right]$$  \hspace{1cm} (49)

$$W_0 = k_B T/(8\pi^2 \kappa)$$  \hspace{1cm} (50)
Equations (45) and (49) for \( \phi(t) \) and \( \bar{s}(t) \) are coupled via equations (43) for \( \sigma \). Obviously, this should lead to a highly non-linear behaviour, which is quite far from the simple linear analysis given in Section 3.1 [25]. Exact solutions to these coupled equations can be therefore obtained only numerically (Section 3.5). In the next section however we will present a few asymptotic solutions which already bear most of the important physics.

3.4. Asymptotic Solutions

3.4.1. — At short times \( \nu t \ll \phi_c \), we may use \( \bar{s}(t) \ll 1 \) in equation (45) to obtain

\[
\phi \simeq \phi_c - \nu t + \phi_c \bar{s}(t)
\]

(51)

Consider now even shorter times where \( \phi_c \bar{s}(t) \ll \nu t \), for which \( \phi \) is decreasing linearly with time:

\[
\phi \simeq \phi_c - \nu t
\]

(52)

However, by looking at (51), we can expect a crossover behaviour at a time \( t = t_c \) obeying

\[
\bar{s}(t_c) \simeq \nu t_c / \phi_c
\]

(53)

At these times the decrease in area resulting from the deformations starts to dominate and \( \phi \) will increase towards \( \phi_c \) at later times, causing the instability to slow down. Since the instability stops when \( \phi = \phi_c \), it is clear that at all times \( t > 0 \) the inequality \( \phi < \phi_c \) must hold. Together with equation (45), this determines a definite upper bound for \( \bar{s}(t) \):

\[
\bar{s}(t) < \nu t / \phi_c
\]

(54)

This means that \( \bar{s}(t) \) must saturate at its upper bound, i.e.,

\[
\bar{s}(t) \simeq \nu t / \phi_c
\]

(55)

at long times \( t \gg t_c \).

On the other hand, equation (49) still has to be obeyed. Let us consider this equation when \( \Omega(x,t) \) is much larger than unity for (normalized) wavenumbers \( x \) around \( x^* \), which is the situation expected at long times \( t \gg t_c \). The integral in equation (49) is then dominated by these wavenumbers. (Since the condition \( \Omega(x^*) \gg 1 \) corresponds also to \( \Omega(x^*) \gg \ln [(1 - (x^*)^2) / \gamma(x^*)] \), we may safely neglect the effect of the weight \( (1 - x^2) / \gamma(x) \) in determining the dominant value of \( x \).) Equation (49) can therefore be approximated according to the method of steepest descents to give

\[
\bar{s} \simeq W_0 \sqrt{\frac{\pi}{-d^2 \Omega/dx^2|_{x^*}}} \frac{1 - (x^*)^2}{\gamma(x^*)} \exp [2\Omega(x^*,t)]
\]

(56)

Equating the latter equation with \( \nu t / \phi_c \) defines an equation for \( \Sigma(t) \) in the long time regime \( t \gg t_c \). It should be noted, however, that the steepest descents approximation is only marginally valid here because – since \( \bar{s}(t) \) must increase linearly with time in this long time regime – \( \Omega(x^*) \) cannot get typically more than an order of magnitude larger than unity. (This may also give somewhat more significance to the weight in equation (49), which may be however easily taken into account in the steepest descents approximation.) To verify these approximations, a full numerical integration will be used in our numerical analysis in Section 3.5.

Consider now the following situations according to whether the value of \( x^* \) in the relevant time regime is of order unity or not:
i) \( x^* \sim \sqrt{\frac{\Sigma R^2}{\kappa t}} \ll 1 \). Here we find, to leading order,

\[
\ddot{s} \simeq \frac{2\sqrt{\pi}}{3} W_0 \frac{\eta R}{\Sigma(t) \ln \left( \frac{2\kappa t}{\Sigma(t) R^2} \right)} \exp \left\{ \frac{1}{16} \frac{\Sigma(t)^2 R}{\kappa t} \ln \left[ \frac{2\kappa t}{\Sigma(t) R^2} \right] \right\}
\]

(57)

ii) \( x^* \simeq 1 \). Here we have, more precisely, \( x^* \simeq 0.56 \) and we find

\[
\ddot{s} \simeq C W_0 \sqrt{\frac{\eta R}{\Sigma(t)}} \exp \left[ \alpha \frac{\Sigma(t)}{\eta R} \right]
\]

(58)

where \( C = 1.37, \alpha = 0.071 \).

We now equate equations (57) and (58) with \( \nu t/\phi_c \) and solve approximately for \( \Sigma(t) \) (using, for simplicity, only a single iteration of the solution):

i) For \( x^* \sim \sqrt{\frac{\Sigma R^2}{\kappa t}} \ll 1 \) we get \( \dot{\Sigma}(t) \sim \sqrt{\eta \kappa t/R} \) or, more precisely,

\[
\dot{\Sigma}(t) \simeq 4\sqrt{2} \frac{\eta \kappa t}{R} \left[ \ln \left( \frac{\nu t/\phi_c A(t)}{\ln \left( \frac{\kappa t}{4\eta R^3} \right)} \right) \right]^{1/2}
\]

(59)

with

\[
A(t) = \frac{\sqrt{2\pi}}{3} W_0 \left( \frac{\eta R^3}{\kappa t} \right)^{1/4} \left[ \ln \left( \frac{\kappa t}{4\eta R^3} \right) \right]^{-1/2}
\]

Evidently, the self-consistency requirement \( \dot{\Sigma}(t) \ll \kappa t/R^2 \) implies that we deal here with times much longer than the basic hydrodynamic relaxation time of the tube \( \tau_r = \eta R^3/\kappa \).

More accurately, including the numerical prefactors and logarithmic dependences, this self-consistency requires \( t \gg 100\tau_r \) for typical situations \( (W_0 \sim 10^{-2} - 10^{-4}) \).

ii) For \( x^* = 0.56 \) we get

\[
\dot{\Sigma}(t) \simeq \alpha^{-1} \eta R \ln \left[ \frac{\nu \kappa t}{\phi_c W_0} \right]
\]

(60)

where \( \alpha = 2.74 \). Accordingly, the self-consistency requirement \( \dot{\Sigma}(t) \gg \kappa t/R^2 \) implies now that \( t \ll 100\tau_r \).

We see that for \( t \gg t_c \), \( \dot{\Sigma}(t) \) increases with time much more slowly than \( \sim t \), either as a power law \( \sim t^{1/2} \), or logarithmically. This slow increase corresponds to an algebraic decay of the surface tension difference \( \sigma(t) \) towards zero (or of \( \phi \) towards \( \phi_c \)), as obtained from \( \sigma(t) = d\Sigma/dt \). This yields (to leading order)

\[
\sigma \sim \sqrt{\frac{\eta \kappa}{R t}}
\]

(61)

in case (i) where \( x^* \ll 1 \), and

\[
\sigma \simeq \alpha^{-1} \eta R \frac{\nu t}{t}
\]

(62)

in case (ii) where \( x^* \sim 1 \). We may conclude that for sufficiently small suction rates where \( 100\nu \tau_r \ll 1 \), the (true) surface tension \( \sigma \) is essentially equal, in the long time limit \( t \gg 100\tau_r \),
(with $\tau \simeq \eta R^3/\kappa$), to the critical value $\sigma_c \sim \kappa/R^2$. However, for larger rates, $100\nu\tau \gg 1$, the surface tension $\sigma$ at the longest relevant time $t \sim \nu^{-1}$ is still much larger than $\sigma_c$.

Another quantity of interest to us is $< (\nabla u(t))^2 >$, which is expressed as

$$< (\nabla u(t))^2 > = \sum_q q^2 \langle U_q(t)U_{-q}(t) \rangle$$  \hspace{1cm} (63)

It measures the extent of surface corrugation and therefore can be related directly to experiment. Using the same procedure as for $\bar{s}(t)$, equation (63) becomes

$$< (\nabla u(t))^2 > = 2W_0 \int_0^\infty dx \frac{x^2}{\gamma(x)} \exp[2\Omega(x,t)]$$  \hspace{1cm} (64)

For large values of the argument of the exponent, corresponding to $t \gg t_c$, we may again use the method of steepest descents leading to

$$< (\nabla u(t))^2 > \simeq 2W_0 \sqrt{\frac{\pi}{-d^2\Omega/dx^2} |x^*|^2} \frac{(x^*)^2}{\gamma(x^*)} \exp[2\Omega(x^*,t)]$$  \hspace{1cm} (65)

so that

$$< (\nabla u)^2 > \simeq 2 \frac{(x^*)^2}{1 - (x^*)^2} \bar{s}(t)$$  \hspace{1cm} (66)

Again we deal with this result separately for values of $x^* \ll 1$, and for $x^* \sim 1$:

i) For $x^* \sim \sqrt{\Sigma R^2/(\kappa t)} \ll 1$ we find

$$< (\nabla u)^2 > \simeq 2(x^*)^2 \bar{s}(t) \simeq \frac{\bar{\Sigma}(t)R^2\nu}{\kappa \phi_c}$$  \hspace{1cm} (67)

or, using equation (59) for $\bar{\Sigma}(t)$ in this regime (ignoring all numerical factors and logarithmic terms),

$$< (\nabla u)^2 > \sim \nu \left( \frac{\eta R^3}{\kappa} t \right)^{1/2} \sim \nu \sqrt{\tau_t t}$$  \hspace{1cm} (68)

ii) For $x^* \sim 1$ we find

$$< (\nabla u)^2 > \simeq 0.92 \bar{s}(t) \simeq \frac{0.92}{\phi_c} \nu t$$  \hspace{1cm} (69)

Let us now summarize the implications of these results. The discussion henceforth will be divided into two cases, depending on the value of the maximum surface tension $\bar{\sigma}_{\text{max}} = \sigma_c + \bar{\sigma}_{\text{max}}$ that is induced in the membrane during the laser operation, where

$$\bar{\sigma}_{\text{max}} \simeq \sigma_0 \nu t_c,$$  \hspace{1cm} (70)

as compared to the bending energy $\kappa/R^2$. This can be translated into a condition on the suction rate $\nu$, as shown below.
3.4.2. Case 1: $\bar{\sigma}_{\text{max}} \ll \kappa/R^2$. — This is the less relevant case for us where the smallest value of the wavelength $\lambda^*$ that is reached is still much larger than the tube radius $R$. This case corresponds to very small suction rates. To be more specific, we need to find the crossover time $t_c$. For $t \ll t_c$ we have

$$\bar{\Sigma}(t) \approx \frac{1}{2} \sigma_0 \nu t^2$$

The time $t_c$ is obtained from equating this result with the result at long times, equation (59), leading to the scaling

$$t_c \sim \left( \frac{\eta \kappa}{R(\sigma_0 \nu)^2} \right)^{1/3}$$

(ignoring here all logarithmic corrections). Hence the above condition $\bar{\sigma}_{\text{max}} \approx \sigma_0 \nu t_c$ translates into a condition on the suction rate $\nu$,

$$\nu \ll \frac{\kappa^2}{\sigma_0 \eta R^5}$$

or

$$\nu \tau_r \ll \frac{\kappa}{R^2 \sigma_0} \ll 1$$

These are indeed extremely small suction rates relative to the tube relaxation rate $\tau_r^{-1}$. Note that this case corresponds to $\tau_r \ll t_c$, so that the time regime $t > t_c$ corresponds already to $t \gg \tau_r$.

As explained, in the regime $t \ll t_c$ all amplitudes remain small ($\Omega(x) \ll 1$) and mode selection is not sharp. Nevertheless, it is of interest to obtain the wavenumber of the fastest growing mode, $q^*$, which – in this regime – is the one which maximizes $\omega(q, t)U_q^0$. It increases from zero (actually, from $\sim 1/L$) as $q^* \sim t^{1/2}$, or, more explicitly, as

$$q^* \approx \sqrt{\frac{\sigma_0 \nu t}{\kappa}}$$

until it reaches a maximum value $(q^*)_{\text{max}}$. This value is given by $(x^* = q^* R)$

$$(x^*)_{\text{max}} \sim \frac{\bar{\Sigma}(t_c) R^2}{\kappa t_c} \sim \left( \frac{\eta \sigma_0 \nu R^3}{\kappa^2} \right)^{1/6} \sim \left( \frac{\tau_r R^2 \sigma_0}{\kappa} \right)^{1/6}$$

In the regime $t \gg t_c$ we have $\bar{s}(t) \approx \nu t/\phi_c$ and the most unstable wavenumber decays back to zero as

$$x^* \sim \left( \frac{\eta R^3}{\kappa t} \right)^{1/4}$$

(ignoring logarithmic dependencies) showing a very slow increase of the wavelength as $\lambda^* \sim t^{1/4}$.

Since the wavelength $\lambda^*$ of the most unstable mode is much larger than the radius $R$ at all times, the instability is likely to stay invisible. We may confirm this by looking at the time dependence of $<(\nabla u)^2>$; after the "exponential" increase up to the time $t_c$, it increases as $\sim t^{1/2}$ (following Eq. (68)), indeed a very slow increase. Moreover, since $\nu \tau_r \ll 1$, one can expect very small suction rates corresponding to $100\nu \tau_r \ll 1$, and it follows from equation (68) that $<(\nabla u)^2>$ is also vanishingly small even at the longest relevant times $\nu t \sim 1$, where almost all the membrane of the initial tube has been sucked out.
3.4.3 Case 2: $\tilde{\sigma}_{\text{max}} \gg \kappa/R^2$. — Here the wavelength becomes of the order of the tube radius $R$ at a relatively short time $t^*$, $t^* \ll t_c$. For $t \ll t^*$, $q^*$ increases initially as $\sim t^{1/2}$ following equation (74). (Recall that we refer here to a non-sharp mode selection.) The time $t^*$, defined by $x^*(t^*) \simeq 1$, is therefore

$$t^* \simeq \frac{2\kappa}{R^2\sigma_0\nu}$$

In the wide regime $t^* \ll t \ll 100\tau$, we find that $q^* \simeq 1/R$ and is not very sensitive to the time $t_c$. Importantly, however, the time $t_c$ signifies that mode selection becomes sharp. For $t \gg 100\tau$, we have again $\tilde{\sigma}(t) \ll \kappa t/R^2$ so that $q^*$ decays back to zero primarily as $\sim t^{-1/4}$ following equation (76).

The crossover time $t_c$ from the early "exponential" growth of $\tilde{s}(t)$ to a linear growth is found, in a similar way to Case 1, to scale as

$$t_c \sim \left(\frac{\eta R}{\sigma_0
nu}\right)^{1/2}$$

(77)

(Numerically, this is an underestimate of $t_c$ by roughly an order of magnitude.) This means that we consider in this case large suction rates which obey

$$\nu\tau \gg 10^{-2}\frac{\kappa}{R^2\sigma_0}$$

(78)

or $100\tau \gg t^*$. Note that the right hand side of (78) is usually much smaller than unity for tubes of radius $R \sim 1\mu m$. (It is $\sim 10^{-4}$ for the regime where surface tension is governed by thermal undulations [24], taking $\kappa \sim 10k_B T$ and $\sigma_0 \sim 5 \times 10^{-5}k_B T/\Lambda^2$). Therefore even very small suction rates compared to $\tau^{-1}$ are likely to fall in this category. More explicitly, we may use our estimates for $\nu$ (Eq. (2)) and $\sigma_{\text{ext}}$ (Section 1) to conclude that — for the experiment reported in reference [13], taking $R \sim 1\mu m$ — the above condition is safely obeyed for $L \ll 1\text{mm}$, which is indeed the experimental situation.

As for the evolution of $\langle (\nabla u)^2 \rangle$, there is a wide regime $t_c \ll t \ll 100\tau$, in which the increase is close to linear following equation (69). Then, the condition (78) allows us to distinguish between two situations: i) If $100\nu\tau \geq 1$, $\langle (\nabla u)^2 \rangle$ can become of order unity. This will occur at time $t \sim \nu^{-1}$. ii) If $\kappa/(R^2\sigma_0) \ll 100\nu\tau$, we have a crossover to a $\sim t^{1/2}$ behaviour (Eq. (68)) at $t \sim 100\tau$. When $t \sim \nu^{-1}$, corresponding to almost complete suction of the membrane, $\langle (\nabla u)^2 \rangle$ is still very small, $\langle (\nabla u)^2 \rangle \sim (\nu\tau)^{1/2} \ll 1$. This is an important result: since visible instabilities correspond to $\langle (\nabla u)^2 \rangle \sim 1$, the suction rate has to be at least as large as $(10^{-2} \nu)$ the tube relaxation rate for the instability to be clearly visible. Our results also suggest that even for such suction rates, one needs to wait until a significant fraction of the initial tube has been sucked out.

3.5. Numerical Results. — In order to verify our asymptotic results we have solved numerically equations (45) and (49), which are coupled via equation (43). Typical results are presented in Figures 1 and 2. In Figure 1 we have chosen parameters corresponding to Case 1 discussed above, and similarly in Figure 2 (Case 2). The quantities discussed above were found to follow well the asymptotic solutions obtained above. We can clearly see how $\tilde{s}(t)$ saturates at a linear growth at $t \gg t_c$ (Figs. 1a and 2a). For $t \gg t_c$ it has been confirmed that the behaviours $\sigma \sim t^{-1}$ (Fig. 2a) and $\sim t^{-1/2}$ (Fig. 1a) is approached quite accurately. This is found to be correlated with the behaviour of $q^*$: When $q^*$ remains almost constant ($= 0.56/R$) at $t > t_c$ (Figs. 2b-c), corresponding to Case 2, the behaviour $\sigma \sim t^{-1}$ is observed,
Fig 1 — Numerical solutions of equations (45), (49) and (43), using the following parameters: \( \nu = 10^{-4} \), \( \eta = 10^{-2} \), \( R = 20 \), \( \phi_c = 0.97 \), \( W_0 = 10^{-5} \), \( \kappa = 50 \), and \( \sigma_0 = 1 \). a) Surface tension difference \( \tilde{\sigma}(t) \) and sucked area \( \tilde{s}(t) \) against time \( t \). The decay of \( \tilde{\sigma} \) at \( t > 1600 \) can be well fitted with a power law \( \sim t^{\zeta} \) with \( \zeta = 0.527 \); b) Wavenumber of the fastest growing mode \( q^* \), and a measure of the "average deformation" \( \langle (\nabla u)^2 \rangle \), against time \( t \). The behaviours at \( t > 1600 \) can be well fitted with a power law \( \sim t^{\mu} \), with \( \mu = -0.223 \) for \( q^* \), and \( \mu = 0.61 \) for \( \langle (\nabla u)^2 \rangle \) (the fit of \( \langle (\nabla u)^3 \rangle \) is less good); c) Same as b), but on a Log-Log plot.
Fig. 2. — Same as Figure 1 but for $R = 100$ and $\kappa = 1$ (other parameters are the same). In a) the decay of $\bar{\sigma}$ at $t > 2500$ can be well fitted to a power law $\sim t^{-\zeta}$ with $\zeta = 1.01$. 
while \( \sigma \sim t^{-1/2} \) is observed when \( q^* \) falls off immediately at \( t > t_c \) (Figs. 1b-c), in which case it approaches \( q^* \sim t^{-1/4} \) (Case 1).

The behaviour of \( <(\nabla u)^2 > \) has some interesting features which were not discussed in Section 3.4. At very short times \( t < t_c \) it first decreases from its non-zero equilibrium value at \( t = 0 \) (Figs. 1c and 2c). The latter corresponds to the thermal ripples (undulations) of the membrane, so the decrease may be understood as the “stretching” of these ripples. The exponential increase which follows is due to the Rayleigh instability, in which the thermal undulations are suppressed. The regime which follows the exponential increase can also be correlated with the behaviour of \( q^* \). If \( q^* \) saturates at \( \sim 1/R \) for \( t > t_c \) (corresponding to Case 2) then \( <(\nabla u)^2 > \) increases almost linearly at \( t > t_c \) (Fig. 2b), following quite closely equation (69). If \( q^* \) starts to fall off immediately after reaching its maximum value (Case 1), the increase of \( <(\nabla u)^2 > \) is clearly sublinear, and approaches \( a \sim t^{1/2} \) behaviour as expected (Fig. 1b).

4. Discussion

4.1. Experimental Implications. — From the linear theory we obtain the result that there is a critical laser intensity, corresponding to a critical surface tension \( \sigma_c \), below which the instability will not occur at all. This intensity is determined by combining equation (17) for \( \sigma_c \) with the estimated relation [17] between the laser intensity and the induced tension described in Section 1. It should be interesting to verify this prediction more quantitatively by varying either the intensity or the critical intensity, which latter is achieved by changing the radius of the tube.

The relevance of the different time regimes discussed in Section 3 (up to the times \( t \sim \tau_t \)) to the phenomena observed in the experiment [13] is evident. However, it could be hard to verify these predictions quantitatively by using only the video microscopy technique. Nevertheless, the predictions for the average gradient of \( u(x) \) appear to be compatible with experiment. The experiment seems to suggest that this quantity does not evolve exponentially with time, but rather in a slower, quasi-linear, evolution. This is qualitatively consistent with our analysis of equations (45) and (49) as discussed above.

Furthermore, we have found that, roughly speaking, \( 100\nu \tau_t \geq 1 \) is the condition needed for the instability to be observed after a typical time \( t \sim \nu^{-1} \) of tweezer action. Using our estimates for the suction rate \( \nu \) (Eq. (2)) and the value of induced tension \( \sigma_{ext} \) given in Section 1, we are able to give explicit predictions for the experimental systems of reference [13]. Let us first define \( \sigma_{ext} = \sigma_{ext} R^2/\kappa \) (i.e., the ratio of the external tension to the bending tension), which is dimensionless, typically of order 10. For cylindrical vesicles whose length \( L \) is of order \( 10\sqrt{\sigma_{ext} R} \) or less, the peeling instability will be pronounced after a sufficiently “long” operation time. However, for longer cylinders of length \( L \gg 10\sqrt{\sigma_{ext} R} \), \( |\nabla u| \) will not exceed the value \( \sim (\sigma_{ext} R^2/L^2)^{1/4} \) and the instability will be less pronounced. In addition, our results imply that by increasing \( R \) at constant \( L \) it is possible to pass from cylinders which never show a significant peeling instability (\( 100\nu \tau_t \ll 1 \)) to cylinders which do show it (\( 100\nu \tau_t \geq 1 \)).

The regime \( t \gg 100\nu \tau_t \), when relevant, corresponds to a slow increase of the wavelength with time as \( t^{1/4} \). (This should also be the case if the suction is suddenly stopped at any time.) Possibly, this prediction is related to the experimentally observed coarsening, e.g., the coalescence of two small, nearly spherical, pearls to a single larger spherical pearl [13]. Of course, this process occurs in a highly non-linear regime and therefore this interpretation may be questioned.
4.2. Non-Linear Regime – Induced Spontaneous Curvature. — The discussion so far has indeed built on the assumption of small deformations (i.e., a linear stability analysis) and therefore cannot be used to explain the observed experimental shapes in the nonlinear (large deformation amplitude) regime. In addition, we have assumed that the exchange of surfactant molecules between the two monolayers building up the bilayer (the “flip-flop” process [3]) is slow enough to be neglected. This process may not be neglected however at very long times, and may strongly influence the shape as shown below.

The coalescence of pearls leads eventually to the formation of very narrow and long tubes connecting the pearls [13], whose stability (or metastability) is quite peculiar. If no spontaneous curvature is present, these cylinders present a large bending energy cost and therefore should be quite unfavorable. We may exclude the possibility that long range van-der-Waals interactions play a role here, as discussed in general by Bruinsma [9]. These interactions contribute an energy $\sim A L d^2 / R^3$, where $A \sim k_B T$ is the Hamaker constant and $d$ is the bilayer half-thickness. It is easily shown that a wide cylinder will not collapse to a narrow one (with $R \sim d$) if $\kappa \gg A$, as in our case. (Allowing for surface tension does not alter this conclusion.)

Possibly, the stability of these long, narrow, tubes is due to a spontaneous curvature effect, which latter is induced by uneven exchange of surfactant molecules between the two sides (monolayers) of the bilayer [26]. To illustrate this idea more clearly, we now discuss a simple model. We assume that bending energy and surface tension provide a driving force for a surfactant current between the two sides of the bilayer, which changes the spontaneous curvature towards the local curvature. To estimate this effect, we start with a kinetic equation for the surfactant density

$$\frac{d}{dt} \delta \phi = -\Lambda \frac{\partial f}{\partial \delta \phi} \tag{79}$$

where $\delta \phi$ is the difference in surfactant coverage between the two sides, $\Lambda$ is an Onsager coefficient, and $f$ is the free-energy density. Importantly, in $f$ we include the compressibility (with modulus $B$), which can oppose any deviation of surfactant density from the optimal one. To first order in $\delta \phi$, the spontaneous curvature is given by $C_0 = \alpha \delta \phi$. The free-energy density of the tube is thus

$$f = \sigma + \frac{1}{2} \kappa \left( \frac{1}{R} - \alpha \delta \phi \right)^2 + \frac{1}{2} B \delta \phi^2 \tag{80}$$

Using this in equation (79), and transforming into an equation for the spontaneous curvature, we obtain

$$\frac{dC_0}{dt} = \Lambda \alpha \kappa \left( \frac{1}{R} - C_0 \right) - \frac{\Lambda B}{\alpha} C_0 \tag{81}$$

Consider now the dynamics of the tube radius $R$, assuming that the cylindrical shape is preserved. Since it is still quite complicated and strongly model dependent (concerning the exchange of membrane area between different parts of the system), we consider here the steady state limit $dR/dt = 0$. The latter corresponds to a vanishing net force acting on the membrane ($\Delta P = 0$), i.e.,

$$\frac{\sigma}{R} - \frac{\kappa}{2R} \left( \frac{1}{R^2} - C_0^2 \right) = 0 \tag{82}$$

This leads to

$$\frac{1}{R} = \left( C_0^2 + \frac{2\sigma}{\kappa} \right)^{1/2} \tag{83}$$
Using this in equation (81) we obtain a differential equation for $C_0$, whose steady state solution, $dC_0/dt = 0$, is

$$
C_0^{-2} = \frac{\kappa}{2\sigma} \left( \frac{B^2}{\kappa^2 \alpha^4} + \frac{2B}{\kappa \alpha^2} \right) \quad (84)
$$

While values of $B$ can be estimated from experiment [6], it is quite hard to obtain a reliable value for $\alpha$ without a detailed molecular calculation [2]. A rough estimate [27] is $\alpha \sim 1/d$, where $d$ is the bilayer half-thickness. Together with the rough estimate $B \sim \kappa T/a \sim \kappa/d^2$ (with $a$, the area per molecule), this implies $C_0 \simeq \sqrt{\sigma/\kappa}$, leading to a radius $R_0$ only a few times smaller than the initial radius $R_0$ (taking $\sigma \sim 10\kappa/R_0^2$). However, values of $B$ smaller than $0.1\kappa^2$, which are still reasonable [28], lead to values of $R \sim 1/C_0$ in the range of those observed [13] in experiment (0.1 μm or less). Note that the spontaneous curvature vanishes for $\sigma = 0$, demonstrating the crucial role of the optical tweezers in this effect.

4.3. COMPARISON WITH THE THEORY OF NELSON, POWERS AND SEIFERT. — The theory of Nelson et al. [17] addresses the same phenomena discussed in the present study and described in reference [13], but differs from ours in several respects, some of which are fundamental. First we note some minor differences for completeness.

In order to compute the dispersion relation $\omega(q)$ (equivalent to Eqs.(30-31)), Nelson et al. take into account two effects which we have omitted in the present study: i) membrane incompressibility, and ii) the friction associated with the 2-dimensional surfactant flow of one monolayer against the other. As explained in detail in Section 3.1, we believe that these effects need not be included here. We also note that the result of Nelson et al. for $\omega(q)$ is only slightly different from the result of Tomotika which we use in our work (for the curvatures we consider here). The main point (in both results) is that one obtains a finite value of $q^*$, which is of order $1/R$ for the case of large surface tension.

Much more important are the differences regarding the evolution of the unstable modes in time. In the model of Nelson et al. the tension $\sigma_{ext} \gg \sigma_c$ is applied instantaneously to the tube. The amplitude then grows exponentially with time as long as $(\nabla u)^2 < 1$ (required for the linear stability analysis to work). We have shown however that this cannot be the case for realistic suction rates, and so, in our model, the growth of the amplitudes is algebraic for $t > t_c$, corresponding still to $(\nabla u)^2 < 1$. Whereas the surface tension is constant in time in the model of Nelson et al., it is strongly time-dependent in our model, showing a large overshoot at $t \approx t_c$ and a slow algebraic decay at $t \gg t_c$. At these “long” times the value of the surface tension is not sensitive anymore to the laser power. We believe that these are clear cut differences which could be detected in an appropriately designed experiment. For example, by using Fourier transform of the shape it might be possible to measure the deformation amplitude $u$ as a function of time when it is still much smaller than $R$.

5. Conclusions

In this paper we presented a theoretical study of the effect of optical tweezers on tubular, surfactant bilayer, vesicles. We adopted a simple model for the action of the tweezers, based a uniform suction of surfactant from the bilayer. The latter leads to an induced surface tension which initializes the Rayleigh instability. The instability continues to develop due to the continuous suction. At long times the extent of the instability becomes entirely controlled by the suction rate.

Evidently, our assumption of a uniform surface tension cannot possibly hold since the surfactant current must always vanish at the ends. Surface tension gradients imply that there
are shear forces acting on the membrane, known as Marangoni effects [29]. They have been neglected in this paper because of the large complication they involve. Their inclusion demands solving the hydrodynamics of the two dimensional surface and that of the solvent, under dynamically changing boundary conditions and spatial anisotropy. The study of the role of surface tension gradients in the observed phenomena, such as the limited range of the instability and the flow of pearls along the cylinder towards the operation zone [13], is left for a future publication.

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Appendix A

Bending Energy of a Deformed Cylinder

Here we calculate the bending energy of a deformed cylinder, whose volume is conserved, to quadratic order in \( u(z) \). In general the curvature can be expressed in terms of the normal unit vector to the surface \( \hat{n} \)

\[
\frac{1}{R_1} + \frac{1}{R_2} = \nabla \hat{n}
\] (A.1)

In our case \( \hat{n} \) is given by

\[
\hat{n} = \frac{\hat{r} - \frac{\partial u}{\partial z} \hat{z}}{\left[ 1 + \left( \frac{\partial u}{\partial z} \right)^2 \right]^{1/2}}
\] (A.2)

Taking the divergence of \( \hat{n} \) in cylindrical coordinates we obtain

\[
\frac{1}{R_1} + \frac{1}{R_2} = \frac{1}{(R + u) \left[ 1 + \left( \frac{\partial u}{\partial z} \right)^2 \right]^{1/2}} \frac{\partial u}{\partial z} \left[ 1 + \left( \frac{\partial u}{\partial z} \right)^2 \right]^{1/2}
\] (A.3)

Dropping higher than quadratic order terms in \( u \) we obtain

\[
H_\kappa = \pi \kappa \int dz (R + u) \left[ 1 + \left( \frac{\partial u}{\partial z} \right)^2 \right]^{1/2} \left( \frac{1}{R_1} + \frac{1}{R_2} \right)^2
\]

\[
\simeq \pi \kappa \int dz \left\{ \frac{1}{(R + u) \left[ 1 + \left( \frac{\partial u}{\partial z} \right)^2 \right]^{1/2}} + R \left( \frac{\partial^2 u}{\partial z^2} \right)^2 \right\}
\]

\[
\simeq \pi \kappa \int dz \left\{ \frac{1}{R} \left[ 1 - \frac{1}{2} \left( \frac{\partial u}{\partial z} \right)^2 \right] + \frac{u^2}{R^3} + R \left( \frac{\partial^2 u}{\partial z^2} \right)^2 \right\}
\] (A.4)
(The linear terms contribute a constant which depends on the boundary conditions, and is omitted here). In Fourier space this leads to

\[ H_\kappa = \frac{\pi \kappa L}{R} + \frac{\pi \kappa L}{R^3} \sum_q \left[ 1 - \frac{1}{2} (qR)^2 + (qR)^4 \right] U_q U_{-q} \]  

(A.5)

Inserting the relation between \( R \) and \( R_0 \), equation (9), we finally obtain (to order \( U^2 \))

\[ H_\kappa = H_{\kappa_0} + \frac{\pi \kappa L}{R_0^3} \sum_q \left[ \frac{3}{2} - \frac{1}{2} (qR_0)^2 + (qR_0)^4 \right] U_q U_{-q} \]  

(A.6)

where \( H_{\kappa_0} = \frac{\pi \kappa L}{R_0} \) is the bending energy of the undeformed cylinder.

References

[19] A similar approach has been recently proposed to describe the dynamics of spontaneous emulsification, see Granek R., Ball R.C. and Cates M E., J Phys II France 3 (1992) 829.
A more accurate estimate, which takes into account the response of the cylinder at time $t$ via its surface tension $\sigma(t)$, is

$$\nu \approx \frac{2\pi R (\sigma_{\text{ext}} - \sigma(t))}{\eta L^2}$$

For $\sigma_{\text{ext}} < \sigma_c$ (Eq (17)) this expression leads to an increase of $\sigma(t)$ with time towards $\sigma_{\text{ext}}$. As shown in Section 3, for the unstable case $\sigma_{\text{ext}} \gg \sigma_c$ we may neglect $\sigma(t)$ in this expression, which reduces to equation (2).

[24] For a free surface of linear size $l \gg \sqrt{\kappa/\sigma}$ and thickness $\delta$, the relation (Refs. [3, 23])

$$\frac{\Delta S}{S_0} \approx \frac{k_B T}{8\pi \kappa} \ln \left( \frac{\kappa}{\sigma \delta^2} \right)$$

describing the ratio of excess area $\Delta S$ to projected area $S_0$, resulting from the thermal undulations, can be used. We thus find for the compression parameter $\sigma_0$ appearing in equation (43)

$$\sigma_0 \approx \frac{8\pi \kappa}{k_B T} \sigma_c$$

It should be noted however that the length scale obtained from $\sqrt{\kappa/\sigma}$ is of the order of the tube radius $R$, and so essentially is in the crossover regime between two-dimensional and (effectively) one-dimensional undulations. This is therefore only a rough estimate for $\sigma_0$.

[25] We self-consistently assume that, because of the very small suction rates involved, the effect of this non-linear coupling is exemplified already for $|\nabla u| \ll 1$, in which case the free-energy expansion equation (16) is still valid.

[26] Ideas in similar spirit have been described by Seifert et al., reference [5].
[27] This estimate is obtained using equation (6 21) in reference [1], which is the result of a simple, molecular-type, model.
[28] The experimental value obtained for DMPC, which is used in reference [13], is $B = 3.5 \kappa/d^2$. One needs $\alpha$ as small as $0.2/d$ to make $B \approx 0.1 \kappa \alpha^2$