J. Phys. II France **2** (1992) 1563–1575 AUGUST **1992**, PAGE 1563

Classification
Physics Abstracts
05.40 — 87.22

Fluctuations and stability of polymerized vesicles

Shigeyuki Komura ([1]) and Reinhard Lipowsky ([2])

([1]) Department of Physics, Faculty of Science, Kyoto University, Kyoto 606, Japan
([2]) Institut für Festkörperforschung, Forschungszentrum Jülich, D-5170 Jülich, Germany

(Received 30 April 1992, accepted 13 May 1992)

Abstract. — The shape fluctuations and the stability of polymerized vesicles are investigated within the framework of shell theory. The intrinsic curvature of the vesicle leads to an enhanced coupling between bending and stretching modes which acts to suppress the shape fluctuations on large scales. This effect is explicitly calculated for a cylindrical shape of the vesicle. For a cylinder with radius r_0, the shape fluctuations exhibit the crossover scale $L^* \simeq (r_0^2 \kappa / Y)^{1/4}$, where κ and Y are the bending rigidity and the (2–dimensional) Young modulus of the polymerized membrane. For $L \ll L^*$, the fluctuations are fluid–like but are strongly suppressed for $L \gg L^*$. In addition, the stability of the cylinder with respect to an external pressure is studied, and a sequence of critical pressure values is determined at which the cylinder becomes unstable.

1. Introduction.

Recently, the properties of polymerized membranes have attracted a lot of attention in connection both with the statistical mechanics of random surfaces and with the biophysics of membranes [1, 2]. In these membranes, the molecules form a 2-dimensional network of fixed connectivity. In biomembranes, these networks often consist of semi–flexible polymers and then have a relatively large mesh size. One example is the network of spectrin molecules which is attached to the plasma membrane of erythrocytes; the latter network has a mesh size of 100-200 nm [3]. A polymerized network with a much smaller mesh size is contained in the cell wall of bacterial cells. These networks are composed of peptidoglycan molecules and are capable of resisting great stress since bacteria exhibit an internal excess pressure (the so–called turgor pressure) [4]. Artificial polymerized membranes can be also synthesized from bilayers of polymerizable lipids by irradiating the membranes with ultraviolet light [5]. This technique typically produces network patches whose lateral extension is of the order of 10-20 nm.

Lipid bilayers form closed membranes or vesicles. These vesicles are of current interest as models for cells and organelles which exhibit a large variety of shapes. If the bilayers are fluid, these shapes can be transformed by changing, e.g., the osmotic conditions, the temper-

ature or the composition of the lipids [6]. Similar shape transformations have been found for biomembranes.

So far, the theoretical work on polymerized membranes has focused on membranes which are *flat* in their undeformed state. It has been found that, in spite of their 2-dimensional character, these membranes exhibit a low temperature phase which is rough but not crumpled [7]. The energy of an undulation mode with wave vector $\mathbf{q}$ is expected to scale as $\mathbf{q}^{4-\eta}$ with $(\eta > 0)$. The existence of such an uncrumpled phase has been confirmed by many computer simulations for open polymerized membranes [8-11]. The value of η is still a matter of some controversy [12-14]. Likewise, Monte Carlo simulations of polymerized vesicles showed that flaccid vesicles exhibit uncrumpled configurations and the mean-squared radius of gyration is proportional to the number of monomers in the membrane [15].

In the present paper, we investigate the shape fluctuations of polymerized vesicles (or shells) which are *curved* in the undeformed state. For such a shell, the stretching deformation which accompanies the bending deformation is a first-order effect while it is only a second-order effect for a flat plate. Thus, for a displacement l along the normal direction, the strain tensor is proportional to l and l^2 for shells and plates, respectively. Therefore, one expects that the shape fluctuations of polymerized vesicles will be effectively suppressed.

For mathematical simplicity, we investigate this coupling between bending and stretching modes primarily for the case of cylindrical vesicles. However, one should keep in mind that this coupling is present for arbitrarily curved membranes. We also consider the deformation of polymerized vesicles caused by a uniform osmotic pressure difference between the outside and the inside of the vesicle. For cylindrical vesicles, we obtain a sequence of critical pressures at which the vesicle becomes unstable with respect to infinitesimally small changes in its shape.

It has recently been argued that a polymerized or solid-like membrane with a relatively small shear modulus or a relatively large bending rigidity should exhibit a pronounced crossover between fluid-like behavior on small scales to solid-like behavior on large scales [12]. For membranes which are flat in their undeformed state, this crossover is again a consequence of the nonlinear terms in the strain-tensor. In this paper, we will show that, for curved shells, such a crossover behavior arises already within the linearized theory.

The outline of this article is as follows. First a few formulas from differential geometry are provided in order to define our notation. By regarding the polymerized membrane as an elastic shell, its elastic deformation energy is derived in accordance with classical shell theory [16-18]. In section 3, cylindrical vesicles are considered as the simplest nontrivial examples which exhibit the intrinsic curvature effect. Even in this case, the statistical mechanics of the three-component displacement field is somewhat complicated, see (4.4), but becomes much simpler after the partial trace over the lateral (phonon-like) components of this field have been performed, see (4.9). In this way, the roughness arising from the shape fluctuations of a polymerized cylinder is determined both in the absence of any constraint and for the cases of constant volume or constant area. This calculation reveals a new crossover length for the shape fluctuations, see (4.15) below. Finally, the instabilities induced by an external pressure are discussed in the last section.

2. Polymerized membrane as an elastic sheet.

First, we will collect some formulas from differential geometry which is the most appropriate formalism for the classical theory of elastic shells [18].

One can, in general, parametrize a 2-dimensional membrane in 3-dimensional space by two real inner coordinates $\mathbf{s} = (s^1, s^2)$. The shape of the membrane is then described by a 3-dimensional vector $\mathbf{r} = \mathbf{r}(\mathbf{s})$. At each point on the membrane, there are two tangent vectors

$\mathbf{r}_i = \partial \mathbf{r}/\partial s^i$ with $i = 1, 2$. The outward unit normal vector $\hat{\mathbf{n}}$ is perpendicular to these tangent vectors, i.e., $\hat{\mathbf{n}} = (\mathbf{r}_1 \times \mathbf{r}_2)/|\mathbf{r}_1 \times \mathbf{r}_2|$.

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

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

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

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

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

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

with $\partial_j \mathbf{r}_i = \partial^2 \mathbf{r}/\partial s^i \partial s^j$.

At zero temperature, the membrane is supposed to be in the (undeformed) reference state described by $\mathbf{r} = \mathbf{R}$. Here and below, we shall use capital letters in order to distinguish quantities in the reference state from the corresponding quantities in the deformed state. Thus, $\mathbf{R}_i, \mathbf{N}, G_{ij}, H_{ij}$ represent the tangent and normal vectors, the metric and the curvature tensors in the reference state, respectively.

If the membrane is stretched, the distance between two neighboring points in the membrane is changed. This change can be expressed by the strain tensor u_{ij} defined by

$$u_{ij} \equiv \frac{1}{2}\left[g_{ij} - G_{ij}\right]. \tag{2.4}$$

The mixed strain tensor is obtained by raising one of the indices according to

$$u_i{}^j = u_{ik} g^{kj} \tag{2.5}$$

Here and below, we use Einstein's summation convention and sum over all indices which appear twice. Likewise, the mixed bending tensor

$$b_i{}^j \equiv h_i{}^j - H_i{}^j \tag{2.6}$$

is taken as a measure for the bending deformation. (Note that this choice is not unique; alternative definitions of the mixed bending tensor are possible starting, e.g., from $\tilde{b}_{ij} \equiv h_{ij} - H_{ij}$.)

According to the elasticity theory of thin elastic sheets conventionally known as shell theory, the deformation energy of an isotropic sheet is given by

$$\mathcal{H} = \int \mathrm{d}^2 s \sqrt{G} \left[\frac{1}{2}\lambda \left(u_i{}^i\right)^2 + \mu u_i{}^j u_j{}^i + \frac{1}{2}\kappa \left(b_i{}^i\right)^2 + \gamma \det\left(b_i{}^j\right)\right]. \tag{2.7}$$

The parameters λ and μ are two Lamé coefficients, and κ and γ are two bending moduli ([1]). (By using the relation $\det\left(b_i{}^j\right) = (1/2)[b_i{}^i b_j{}^j - b_i{}^j b_j{}^i]$, one can rewrite the last two terms in (2.7) in the form $a_1 \left(b_i{}^i\right)^2 + a_2 b_i{}^j b_j{}^i$)

([1]) Since $b_i{}^j$ has been defined by the difference between two curvature tensors, it has to satisfy certain compatibility conditions in order to be itself a curvature tensor (in such case κ and γ are called bending rigidity and Gaussian curvature modulus, respectively). Hence, in general, the Gauss-Bonnet theorem will not apply to (2.7). For a planar reference state, however, one has $b_i{}^j = h_i{}^j$ and the Gauss-Bonnet theorem applies, see (3.12) below.

As mentioned, the vector $\mathbf{R}$ is taken to describe the reference state. Any deformed state of the membrane can then be parametrized by

$$\mathbf{r} = \mathbf{R} + u^i \mathbf{R}_i + l\mathbf{N}. \tag{2.8}$$

The variables u^i represent two lateral (in-plane) displacement fields and l represents the transverse (out-of-plane) displacement field. Both strain tensor and bending tensor can be expressed in terms of the components of these displacement fields. Up to first order in the displacement $\mathbf{r} - \mathbf{R}$, the mixed strain tensor turns out to be

$$u_i{}^j \approx \frac{1}{2}\left[D^j u_i + D_i u^j\right] - l H_i{}^j, \tag{2.9}$$

where the covariant derivative D_i is defined by

$$D_i u^j \equiv \partial_i u^j + \Gamma^j_{ik} u^k, \tag{2.10}$$

with the Christoffel symbols

$$\Gamma^j_{ik} = \frac{1}{2} G^{jl}\left[\partial_i G_{kl} + \partial_k G_{il} - \partial_l G_{ik}\right], \tag{2.11}$$

and $\partial_i \equiv \partial/\partial s^i$. In a similar manner, the linear approximation for $b_i{}^j$ leads to

$$b_i{}^j \approx D_i D^j l + l H_i{}^k H_k{}^j + (D_i u^k) H_k{}^j - (D_k u^j) H_i{}^k + u^k (D_k H_i{}^j). \tag{2.12}$$

As mentioned, we have defined the bending tensor in terms of the difference between the mixed components of the curvature tensors in the deformed and the initial state. If we had selected, for instance, the difference of the covariant components, the result would be different. The formalism described in this section is completely general and can be applied to an arbitrary reference state. In the following, we will focus on the case for which the reference state has the shape of a cylinder. This is the simplest example which exhibits the intrinsic curvature effect described in introduction.

3. Polymerized cylinder.

Now, consider a polymerized cylinder of radius r_0 and length L with internal coordinates $(s^1, s^2) = (\phi, z)$ where ϕ and z are the azimuthal angle and coordinate along the axis of rotational symmetry, respectively. As a local basis of 3-dimensional space, we employ the corresponding unit vectors, $\hat{\mathbf{e}}_r = (\cos\phi, \sin\phi, 0)$, $\hat{\mathbf{e}}_\phi = (-\sin\phi, \cos\phi, 0)$ and $\hat{\mathbf{e}}_z = (0, 0, 1)$. The reference state is described by

$$\mathbf{R} = r_0 \hat{\mathbf{e}}_r + z\hat{\mathbf{e}}_z, \tag{3.1}$$

and a slightly deformed cylinder is represented by

$$\begin{aligned} \mathbf{r} &= \mathbf{R} + u^1 \mathbf{R}_1 + u^2 \mathbf{R}_2 + l\mathbf{N} \\ &= (r_0 + l)\hat{\mathbf{e}}_r + u^1 r_0 \hat{\mathbf{e}}_\phi + (z + u^2)\hat{\mathbf{e}}_z. \end{aligned} \tag{3.2}$$

Within the linear approximation, the components of the mixed strain tensor are given by (2.9). A simple calculation then leads to

$$u_1{}^1 \approx \frac{l}{r_0} + \frac{\partial u^1}{\partial \phi}, \tag{3.3}$$

$$u_1{}^2 \approx \frac{1}{2}r_0^2\frac{\partial u^1}{\partial z} + \frac{1}{2}\frac{\partial u^2}{\partial \phi}, \tag{3.4}$$

$$u_2{}^1 \approx \frac{u_1{}^2}{r_0^2}, \tag{3.5}$$

$$u_2{}^2 \approx \frac{\partial u^2}{\partial z} \tag{3.6}$$

Likewise, it follows from the expression (2.12) for the mixed bending tensor that

$$b_1{}^1 \approx \frac{1}{r_0^2}\frac{\partial^2 l}{\partial \phi^2} + \frac{l}{r_0^2}, \tag{3.7}$$

$$b_1{}^2 \approx \frac{\partial^2 l}{\partial \phi \partial z} + \frac{1}{r_0}\frac{\partial u^2}{\partial \phi}, \tag{3.8}$$

$$b_2{}^1 \approx \frac{1}{r_0^2}\frac{\partial^2 l}{\partial \phi \partial z} - \frac{1}{r_0}\frac{\partial u^1}{\partial z}, \tag{3.9}$$

$$b_2{}^2 \approx \frac{\partial^2 l}{\partial z^2}. \tag{3.10}$$

For the polymerized cylinder, the last term in the elastic free energy (2.7) can be written as

$$\det\left(b_i{}^j\right) = \det\left(h_i{}^j\right) + \frac{1}{r_0}\frac{\partial^2 l}{\partial z^2}. \tag{3.11}$$

If the cylinder satisfies periodic boundary conditions along the z-direction, i.e., for $\mathbf{r}(\phi, z) = \mathbf{r}(\phi, z+L)$, the second term in (3.11) drops out after the integration over the membrane surface. Then the Gauss-Bonnet theorem tells us that the integral

$$\int \mathrm{d}^2 s\sqrt{G}\det\left(h_i{}^j\right) \approx \int \mathrm{d}^2 s\sqrt{g}\det\left(h_i{}^j\right), \tag{3.12}$$

always leads to a constant that does not depend on the size of the membrane but only on its topology or, more precisely, on its Euler characteristic, χ. For an open cylinders as considered here, the Euler characteristic $\chi = 0$ and the integral in (3.12) vanishes. Hence the remaining contribution which arises from the curvature tensor is only the third term in the deformation energy (2.7),

$$\left(b_i{}^i\right)^2 \approx \left[\frac{1}{r_0^2}\left(\frac{\partial^2 l}{\partial \phi^2} + l\right) + \frac{\partial^2 l}{\partial z^2}\right]^2, \tag{3.13}$$

which depends only on l but not on u^1 and u^2. Consequently, the effective Hamiltonian of the polymerized cylinder takes the form

$$\mathcal{H}\left\{u^1, u^2, l\right\} = \int_0^{2\pi}\mathrm{d}\phi\int_0^L \mathrm{d}z\, r_0\left[\frac{1}{2}\lambda\left(u_i{}^i\right)^2 + \mu u_i{}^j u_j{}^i + \frac{1}{2}\kappa\left(b_i{}^i\right)^2\right]. \tag{3.14}$$

In the following sections, we investigate the shape fluctuations governed by this effective Hamiltonian under the constraint of constant volume or constant area and the effect of an external pressure. For these purposes, it is necessary to express the volume V and the area A of a deformed cylinder in terms of the displacement fields. As shown in Appendix A, the volume is given by

$$\begin{aligned} V &\approx \int_0^{2\pi}\mathrm{d}\phi\int_0^L \mathrm{d}z\left[\frac{1}{2}\left(r_0^2 + 2r_0 l + l^2\right) + r_0\left(-u^1\frac{\partial l}{\partial \phi} - u^2\frac{\partial l}{\partial z} + \frac{1}{2}r_0\left(u^1\right)^2\right)\right] \\ &\approx V_0 + \int_0^{2\pi}\mathrm{d}\phi\int_0^L \mathrm{d}z\, r_0\left[l + \frac{l^2}{2r_0} - u^1\frac{\partial l}{\partial \phi} - u^2\frac{\partial l}{\partial z} + \frac{1}{2}r_0\left(u^1\right)^2\right], \end{aligned} \tag{3.15}$$

up to second order in the displacement fields with $V_0 = \pi r_0^2 L$, whereas the area is found to be

$$\begin{aligned} A &= \int \mathrm{d}^2 s \sqrt{g} \\ &\approx A_0 + \int_0^{2\pi} \mathrm{d}\phi \int_0^L \mathrm{d}z\, r_0 \left[\frac{l}{r_0} + \frac{\partial u^1}{\partial \phi} + \frac{\partial u^2}{\partial z} + \frac{1}{2r_0^2}\left(\frac{\partial l}{\partial \phi}\right)^2 + \frac{1}{2}\left(\frac{\partial l}{\partial z}\right)^2 \right. \\ &\quad \left. + \frac{\partial u^1}{\partial \phi}\frac{\partial u^2}{\partial z} - \frac{\partial u^1}{\partial z}\frac{\partial u^2}{\partial \phi} - \frac{u^1}{r_0}\frac{\partial l}{\partial \phi} + \frac{l}{r_0}\frac{\partial u^2}{\partial z} + \frac{1}{2}\left(u^1\right)^2 \right], \end{aligned} \tag{3.16}$$

with $A_0 = 2\pi r_0 L$.

4. Thermally-excited shape fluctuations.

4.1 WITHOUT CONSTRAINT. — In order to decompose the displacements in terms of appropriate eigenmodes, we use the following convention for the Fourier transformation of any field $f(\phi, z)$ defined on the cylinder:

$$f(\phi, z) = \sum_{m,n} f_{mn} \exp\left\{ i \left[m\phi + n\left(\frac{2\pi z}{L}\right)\right]\right\}, \tag{4.1}$$

$$f_{mn} = \frac{1}{2\pi L}\int_0^{2\pi} \mathrm{d}\phi \int_0^L \mathrm{d}z\, f(\phi, z) \exp\left\{ -i \left[m\phi + n\left(\frac{2\pi z}{L}\right)\right]\right\}. \tag{4.2}$$

As usual, one has $f^*_{mn} = f_{-m,-n}$ in order to ensure that the displacement fields are real (the asterisk denotes the complex conjugate value).

It is convenient to introduce the parameter

$$q \equiv \frac{2\pi r_0}{L}, \tag{4.3}$$

which represents the aspect ratio of the cylinder or, more precisely, of the rectangular membrane surface which is obtained by cutting the cylinder parallel to its rotation axis.

If the elastic free energy as given by (3.14) is expressed in terms of the displacement modes u^1_{mn}, u^2_{mn} and l_{mn}, one obtains

$$\begin{aligned} \mathcal{H}\{u^1, u^2, l\} &= \frac{2\pi L}{r_0}\sum_{m,n}\frac{\lambda}{2}\left[m^2 r_0^2 |u^1_{mn}|^2 + n^2 q^2 |u^2_{mn}|^2 + |l_{mn}|^2 \right. \\ &\quad \left. + 2mnqr_0 u^{1*}_{mn} u^2_{mn} + 2iml^*_{mn} r_0 u^1_{mn} + 2inql^*_{mn} u^2_{mn}\right] \\ &+ \frac{2\pi L}{r_0}\sum_{m,n}\frac{\mu}{2}\left[(2m^2 + n^2q^2) r_0^2 |u^1_{mn}|^2 + (m^2 + 2n^2q^2)|u^2_{mn}|^2 + 2|l_{mn}|^2 \right. \\ &\quad \left. + 2mnqr_0 u^{1*}_{mn} u^2_{mn} + 4iml^*_{mn} r_0 u^1_{mn}\right] \\ &+ \frac{2\pi L}{r_0}\sum_{m,n}\frac{\kappa}{2r_0^2}(m^2 + n^2q^2 - 1)^2 |l_{mn}|^2. \end{aligned} \tag{4.4}$$

In this expression, the displacement modes u^1_{00} and u^2_{00} which correspond to a rigid rotation around the z-axis and to a rigid translation parallel to the z-axis do not enter since the corresponding energy is identically zero (in field-theoretic language, these modes are called "zero modes"). This implies that the l_{00}-mode is completely decoupled from all other modes.

The effective Hamiltonian as given by (4.4) is quadratic in the phonon-like fields u^i. Performing the Gaussian functional integrations over all (m,n)-modes of u^2 and u^1 with $(m,n) \neq (0,0)$, one obtains

$$\int \mathcal{D}\{u^1\}\,\mathcal{D}\{u^2\}\,\mathrm{e}^{-\mathcal{H}\{u^1,u^2,l\}/T} \equiv \mathrm{e}^{-\mathcal{H}_{\mathrm{eff}}\{l\}/T}, \tag{4.5}$$

where the new effective configuration energy $\mathcal{H}_{\mathrm{eff}}$ now depends only on the transverse mode l:

$$\begin{aligned}\mathcal{H}_{\mathrm{eff}}\{l\} &= \frac{2\pi L}{r_0}\left\{\frac{1}{2}(\lambda+2\mu)|l_{00}|^2\right. \\ &\left. +{\sum_{m,n}}' \frac{1}{2}\left[Y\left(\frac{n^2q^2}{m^2+n^2q^2}\right)^2 + \frac{\kappa}{r_0^2}(m^2+n^2q^2-1)^2\right]|l_{mn}|^2\right\},\end{aligned} \tag{4.6}$$

where the prime indicates that the l_{00}-term is not included in the summation. The parameter

$$Y = \frac{4\mu(\lambda+\mu)}{\lambda+2\mu}, \tag{4.7}$$

is the 2-dimensional Young modulus. This modulus describes the elastic response of the 2-dimensional sheet when subjected to an uniaxial tension. It is interesting to note that the same modulus is also relevant if one considers a *flat* reference state and includes the leading *non-linear* term in the strain tensor [7].

4.2 CONSTANT VOLUME OR CONSTANT AREA. — We can easily incorporate the constraint of constant volume using the expression (3.15) for the volume. It then follows from $V - V_0 = 0$ that

$$2\pi L r_0 l_{00} = -\frac{2\pi L}{r_0}{\sum_{m,n}}' \frac{r_0}{2}\left[r_0^2|u^1_{mn}|^2 + |l_{mn}|^2 + 2iml^*_{mn}r_0u^1_{mn} + 2inql^*_{mn}u^2_{mn}\right]. \tag{4.8}$$

The constant volume constraint leads to the elimination of the l_{00}-terms in the effective Hamiltonian (4.6). Since all terms in (4.6) are quadratic in the modes, elimination of the l_{00}-mode *via* (4.8) generates third and fourth order contributions which can be ignored for the quadratic approximation considered here. Hence the only change in (4.6) is that

$$\mathcal{H}_{\mathrm{eff}}\{l\} = \frac{2\pi L}{r_0}{\sum_{m,n}}' \frac{1}{2}\left[Y\left(\frac{n^2q^2}{m^2+n^2q^2}\right)^2 + \frac{\kappa}{r_0^2}(m^2+n^2q^2-1)^2\right]|l_{mn}|^2, \tag{4.9}$$

which governs all modes with $(m,n) \neq (0,0)$. The properties of the l_{00} are then determined, *via* (4.8), in terms of these modes.

The constraint of constant area can be imposed in a similar manner. Using the relation (3.16), it follows from $A - A_0 = 0$ that

$$\begin{aligned}2\pi L l_{00} &= -\frac{2\pi L}{r_0}{\sum_{m,n}}' \frac{1}{2}\left[r_0^2|u^1_{mn}|^2 + (m^2+n^2q^2)|l_{mn}|^2\right. \\ &\left. +2iml^*_{mn}r_0u^1_{mn} + 2inql^*_{mn}u^2_{mn}\right].\end{aligned} \tag{4.10}$$

We see again that all other modes remain unaffected at the harmonic level, namely, up to second order in the displacements. Hence the effective Hamiltonian is again identical to (4.9).

4.3 ROUGHNESS OF FLICKERING MEMBRANE. — The cases in section 4.1 and in section 4.2 are identical as far as the behavior of the l_{mn}-modes with $(m,n) \neq (0,0)$ are concerned. In addition, we do not include the l_{10}-term since it corresponds to a simple translation of the cylinder perpendicular to its rotation axis requiring no energy. With the use of the equipartition theorem (or the explicit Gaussian integrations), the mean-squared mode amplitude for both cases is

$$\langle |l_{mn}|^2 \rangle = \frac{T r_0^3}{2\pi\kappa L} \left[\bar{Y} \left(\frac{n^2 q^2}{m^2 + n^2 q^2} \right)^2 + (m^2 + n^2 q^2 - 1)^2 \right]^{-1}, \tag{4.11}$$

with the dimensionless parameter

$$\bar{Y} \equiv Y r_0^2 / \kappa. \tag{4.12}$$

The case of zero shear modulus or $\overline{Y} = 0$ corresponds to fluid membranes as studied previously [19] provided the spontaneous curvature $C_0 = 1/r_0$. It follows from (4.11) that the presence of a finite shear modulus or $\overline{Y} > 0$ reduces the amplitude of all shape fluctuations as expected. This reduction is most effective for $m \ll nq$, i.e., for "anisotropic" modes which exhibit many humps along the cylinder (parallel to the z–axis) but only a few humps in the azimuthal direction. In this case, one has

$$\langle |\ell_{mn}|^2 \rangle \lesssim [\overline{Y} + (nq)^4]^{-1} \tag{4.13}$$

and these modes are dominated by $\overline{Y}$ for wavenumbers

$$n 2\pi/L \lesssim p_z^* \equiv (Y/\kappa r_0^2)^{1/4} \tag{4.14}$$

Thus, one has the crossover length

$$L^* \equiv c(r_0^2 \kappa / Y)^{1/4} \tag{4.15}$$

where c is a dimensionless coefficient of order one.

Such a crossover is also present for "isotropic" modes with $m = nq$. In the latter case, one obtains from (4.11) the behavior

$$\langle |\ell_{mn}|^2 \rangle \sim \left[\frac{1}{4} \overline{Y} + 4m^4 \right]^{-1} \tag{4.16}$$

which implies that modes with wavenumber $n\, 2\pi/L = m/r_0 \lesssim \frac{1}{2}(Y/\kappa r_0^2)^{1/4}$ are dominated by $\overline{Y}$. Thus, apart from a prefactor of order one, one again obtains the crossover length L^* as given by (4.15).

As shown in reference [12], the crossover length for plates arising from the nonlinear terms of the strain tensor depends on temperature. If the critical exponent $\eta = 1$ as concluded from the Monte Carlo simulations in reference [12], the latter crossover length is given by $L^* \simeq \kappa/(TY)^{1/2}$. In contrast, the crossover length for shells as given by (4.15) is independent of temperature but depends explicitly on the curvature radius r_0.

On the other hand, the modes with $n = 0$ are not suppressed at all by the presence of the shear modulus (within the harmonic approximation used here). These modes have no deformations in the z–direction and, thus, are effectively one–dimensional. In general, all "anisotropic" modes with $m \gg nq$ are essentially unaffected by the polymerization since one then has

$$\langle |\ell_{mn}|^2 \rangle \sim [\overline{Y}(nq/m)^4 + m^4]^{-1} \sim m^{-4} \tag{4.17}$$

as for fluid membranes.

4.4 EXAMPLES FOR POLYMERIZED MEMBRANES. — First, consider a polymerized membrane consisting of a thin solid–like sheet. In this case, the elastic moduli of the membrane can be estimated starting from the elastic properties of the bulk material. For an isotropic material in three dimensions, one has two Lamé coefficients λ_3 and μ_3. For a membrane of thickness a, one finds that the Lamé coefficients $\lambda = a\,2\lambda_3\mu_3/(\lambda_3+2\mu_3)$ and $\mu = a\mu_3$ and that its bending rigidity κ is given by $\kappa = 4\mu_3[(\lambda_3+\mu_3)/(\lambda_3+2\mu_3)]a^3/12 = (\lambda+2\mu)a^2/12$. This implies that $\kappa/Y \propto a^2$ and thus

$$\overline{Y} \propto (r_0/a)^2 \quad \text{and} \quad L^* \propto (r_0 a)^{1/2} \tag{4.18}$$

These estimates should apply, for example, to the cell wall of bacteria. If the radius of the cylinder is $r_0 \simeq 1\ \mu$m and the thickness of the membrane is $a \simeq 5$ nm, one has $(r_0 a)^{1/2} \simeq 70$ nm which sets the scale for the crossover length L^*.

Next, consider the tethered membranes which have recently been studied in many computer simulations. For example, the networks studied in reference [8] are characterized by the values $Ya^2/T \simeq 20$ and $\kappa/T \simeq 1$, where a is the mesh size of the network, which implies $Y/\kappa \simeq 20/a^2$ and thus

$$\overline{Y} \simeq 20(r_0/a)^2 \quad \text{and} \quad L^* \simeq \frac{1}{2}(r_0 a)^{1/2} \tag{4.19}$$

Thus, for the accessible sizes of networks with $r_0 \simeq 3a - 6a$, the crossover scale L^* is of the order of a, and all fluctuations will be suppressed by the polymerization.

Finally, it is instructive to consider the plasma membrane of red blood cells. The elastic moduli of this membrane are estimated to be $\kappa \simeq 3 \times 10^{-20}$ J and $Y \simeq 2 \times 10^{-5}$ Jm^{-2} [20-22]. This leads to $Y/\kappa \simeq 0.7 \times 10^3/\mu\text{m}^2$. Using an effective radius $r_0 = 1\ \mu$m, one obtains $\overline{Y} \simeq 7 \times 10^2$ and the crossover length $L^* \simeq 0.2\ \mu$m. The latter length scale is comparable to the mesh size of the spectrin network, and somewhat smaller than the crossover length arising from the nonlinear terms of the strain tensor as estimated in reference [12].

Very recently, Sackmann and coworkers have made a detailed comparison between experiment and theory for the flickering of red blood cells which have the shape of discocytes [23, 24]. Somewhat surprisingly, they conclude that the experimentally observed flickering shows no effect of the small but finite shear modulus $\propto Y$ arising from the spectrin network. This is difficult to understand especially because the discocyte shape itself should be determined by this network.

5. Pressure-induced instabilities.

In this section, we investigate the case where the polymerized vesicle is subjected to the (osmotic) pressure difference P

$$P \equiv P_{\text{ex}} - P_{\text{in}}, \tag{5.1}$$

measured between the outside and the inside of the vesicle. This leads to a stretching (or compression) of the membrane and, thus, to a lateral tension. For a polymerized membrane, this tension is, in general, anisotropic. Since the length L of the cylinder parallel to the z–axis is not allowed to change here, the tension, Σ_2, in the z–direction must vanish. Therefore, the pressure is balanced by the tension Σ_1, which acts in the "hoop" or ϕ–direction.

In thermal equilibrium, the probability for the realization of a certain configuration of the vesicle is now governed by the Boltzmann weight $e^{-\mathcal{H}'/T}$ with

$$\mathcal{H}'\{u^1, u^2, l\} = \mathcal{H}\{u^1, u^2, l\} + \Sigma_1 A + PV, \tag{5.2}$$

where $\mathcal{H}$ is given by (3.14). According to the expressions (3.15) and (3.16) for the volume V and for the area A, the sum of the last two terms is

$$\begin{aligned}\Sigma_1 A + PV &= \Sigma_1 A_0 + PV_0 + 2\pi L(\Sigma_1 + Pr_0) l_{00} \\ &+ \frac{2\pi L}{r_0} \sum_{m,n} \frac{1}{2} \left[(\Sigma_1 + Pr_0) \left(r_0^2 |u^1_{mn}|^2 + 2iml^*_{mn} r_0 u^1_{mn} + 2inql^*_{mn} u^2_{mn} \right) \right. \\ &+ \left. \left\{ \Sigma_1 (m^2 + n^2 q^2) + Pr_0 \right\} |l_{mn}|^2 \right] . \end{aligned} \tag{5.3}$$

When the cylinder represents an equilibrium shape, the first variation of the configurational energy should vanish for any infinitesimal displacement; in other words, the forces should be balanced within the membrane. Therefore, the term proportional to l_{00} in (5.3) leads to the equilibrium condition

$$\Sigma_1 + Pr_0 = 0. \tag{5.4}$$

This equation has to be distinguished from the well-known Laplace equation for fluid droplets because of the anisotropy of the tension. In the geometry considered here, $\Sigma_1 = -Pr_0$ while $\Sigma_2 = 0$ as mentioned. In contrast, if one introduces two membrane caps at $z = 0$ and $z = L$ which close the cylinder, the pressure onto these caps leads to $\Sigma_2 = -\frac{1}{2} Pr_0$. [17]

By eliminating Σ_1 *via* (5.4), one can further simplify (5.3) and then obtains

$$\Sigma_1 A + PV = \Sigma_1 A_0 + PV_0 - \frac{2\pi L}{r_0} \sum_{m,n} \frac{Pr_0}{2} (m^2 + n^2 q^2 - 1) |l_{mn}|^2, \tag{5.5}$$

which does not depend on u^1 or u^2. Hence by adding (5.5) to (4.6), we end up with the effective Hamiltonian

$$\begin{aligned}\mathcal{H}'_{\rm eff}\{l\} &= \frac{2\pi L}{r_0} \left\{ \frac{1}{2} (\lambda + 2\mu + Pr_0) |l_{00}|^2 + \sum_{m,n}{}' \frac{1}{2} \left[Y \left(\frac{n^2 q^2}{m^2 + n^2 q^2} \right)^2 \right. \right. \\ &+ \left. \left. \frac{\kappa}{r_0^2} (m^2 + n^2 q^2 - 1)^2 - Pr_0 (m^2 + n^2 q^2 - 1) \right] |l_{mn}|^2 \right\} . \end{aligned} \tag{5.6}$$

For constant volume, inserting (4.8) into

$$\mathcal{H}''_{\rm eff}\{l\} \equiv \mathcal{H}_{\rm eff}\{l\} + \Sigma_1 A, \tag{5.7}$$

and using the relation $\Sigma_1 = -Pr_0$ as in (5.4), one finds that $\mathcal{H}''_{\rm eff}$ is identical to (5.6) for all modes with $(m,n) \neq (0,0)$. For constant area, on the other hand, by using (4.10) in a similar way one finds that

$$\mathcal{H}'''_{\rm eff}\{l\} \equiv \mathcal{H}_{\rm eff}\{l\} + PV, \tag{5.8}$$

to be again identical to (5.6) for all modes with $(m,n) \neq (0,0)$.

In view of these results, (5.6) provides the basis for analyzing the stability of polymerized cylinders subject to an external pressure. For the modes with $(m,n) \neq (0,0)$, the coefficients of $|l_{mn}|^2$ will become negative as soon as P exceeds the critical value

$$P_c(m,n) = \frac{Y}{r_0} \frac{1}{m^2 + n^2 q^2 - 1} \left(\frac{n^2 q^2}{m^2 + n^2 q^2} \right)^2 + \frac{\kappa}{r_0^3} (m^2 + n^2 q^2 - 1). \tag{5.9}$$

For $P = P_c(m,n)$, the (m,n)-mode becomes unstable under an infinitesimal perturbation of the shape.

In the case of rotational symmetry, i.e., for $m = 0$, the effective Hamiltonian (5.6) simplifies and becomes

$$\mathcal{H}'_{\rm eff}\{l\} = \frac{2\pi L}{r_0} \sum_n{}' \frac{\kappa}{2r_0^2} \left[(n^2q^2)^2 - (\bar{P}+2)n^2q^2 + \bar{P} + \bar{Y} + 1\right] |l_{0n}|^2, \tag{5.10}$$

with

$$\bar{P} \equiv Pr_0^3/\kappa. \tag{5.11}$$

The necessary condition for unstable $(0,n)$–modes is the relation

$$(\bar{P}+2)^2 - 4(\bar{P}+\bar{Y}+1) \geq 0. \tag{5.12}$$

If this relation is satisfied, all modes within the n–band as given by

$$(\bar{P}/2) + 1 - W \leq n^2q^2 \leq (\bar{P}/2) + 1 + W \tag{5.13}$$

with

$$W \equiv \sqrt{\left(\frac{\bar{P}}{2}+1\right)^2 - (\bar{P}+\bar{Y}+1)}\,. \tag{5.14}$$

are unstable. The least stable mode within this band is given by

$$n = \sqrt{\bar{P}+2}/\sqrt{2}q\,. \tag{5.15}$$

More precisely, n is that integer which is closest to $\sqrt{\bar{P}+2}/\sqrt{2}q$. On the other hand, the $(0,n_*)$–mode which exhibits the smallest critical pressure, P_c, is characterized by the condition that W as given by (5.14) vanishes. This implies that n_* is given by

$$n_* = [\sqrt{\bar{Y}}+1]^{1/2}/q \tag{5.16}$$

and that the corresponding critical pressure is $\bar{P}_c = 2\sqrt{\bar{Y}}$ or

$$P_c = 2\sqrt{Y\kappa}/r_0^2\,. \tag{5.17}$$

Acknowledgments.

We thank Dr. A. Baumgärtner, Prof. T. Izuyama, Prof. K. Kitahara and Dr. S. Adachi for their interest. One of the authors (S.K.) is also grateful for the hospitality of the Institut für Festkörperforschung at the Forschungszentrum Jülich.

Appendix A.

Volume of deformed cylinder.

In this Appendix, we derive the expression for the volume of a deformed cylinder up to second order in the displacement fields. It is convenient to reparametrize the cylindrical shape in terms of the radial distance from the z-axis, i.e., $r = \rho(\phi, z)$ with which the total volume is represented as

$$V = \int_0^{2\pi} {\rm d}\phi \int_0^L {\rm d}z\, \frac{1}{2}\rho^2(\phi, z). \tag{A1}$$

The next step is to obtain the relation between ρ and the displacement fields u^1, u^2, and l. Suppose some particular point on the membrane undergoes a certain deformation and moves to another point parametrized by $(\phi, z) = (\theta + \delta, d + u^2)$. The radial distance of this point after the deformation is

$$\rho(\theta + \delta, d + u^2) \approx r_0 + l(\theta, d) + \frac{1}{2} r_0 \left[u^1(\theta, d)\right]^2 \tag{A2}$$

Using the definitions $\delta \equiv \Delta\phi$ and $u^2 \equiv \Delta z$, (A2) can be rewritten as

$$\rho(\phi, z) \approx r_0 + l(\phi - \Delta\phi, z - \Delta z) + \frac{1}{2} r_0 \left[u^1(\phi - \Delta\phi, z - \Delta z)\right]^2 \tag{A3}$$

In addition to the obvious relation $\Delta z = u^2(\phi, z)$, we need $\Delta\phi$ up to first order in the displacement fields;

$$\begin{aligned} \Delta\phi &= \arctan\left[\frac{u^1(\phi - \Delta\phi, z - \Delta z)}{1 + l/r_0}\right] \\ &\approx u^1(\phi, z). \end{aligned} \tag{A4}$$

Substituting these results into (A3), one finally obtains

$$\rho(\phi, z) \approx r_0 + l(\phi, z) - u^1 \frac{\partial l}{\partial \phi} - u^2 \frac{\partial l}{\partial z} + \frac{1}{2} r_0 (u^1)^2. \tag{A5}$$

Insertion of (A5) into (A1) leads to (3.15).

References

[1] Proceedings of the Fifth Jerusalem Winter School, "Statistical Mechanics of Membranes and Surfaces, D. R. Nelson, T. Piran and S. Weinberg Eds, (World Scientific, Singapore, 1989).

[2] Lipowsky R., *Nature* **349** (1991) 475.

[3] Darnell J., Lodish H. and Baltimore D., Molecular Cell Biology (Freeman, New York, 1990)

[4] See, e.g., Koch A.L., *Am. Sci.* **78** (1990) 327.

[5] See, e.g., Sackmann E., Eggl P., Fahn C., Bader H., Ringsdorf H. and Schollmeier M., *Ber. Bunsenges. Phys. Chem.* **89** (1985) 1198.

[6] Berndl K., Käs J., Lipowsky R., Sackmann E. and Seifert U., *Europhys. Lett.* **13** (1990) 659.

[7] Nelson D.R. and Peliti L., *J. Phys. France* **48** (1987) 1085.

[8] Kantor Y. and Nelson D.R., *Phys. Rev. Lett.* **58** (1987) 2774; *Phys. Rev. A* **36** (1987) 4020.

[9] Plischke M. and Boal D., *Phys. Rev. A* **38** (1988) 4943.

[10] Abraham F.F., Rudge W.E. and Plischke M., *Phys. Rev. Lett.* **62** (1989) 1757.

[11] Ho J.-S. and Baumgärtner A., *Phys. Rev. Lett.* **63** (1989) 1324.

[12] Lipowsky R. and Girardet M., *Phys. Rev. Lett.* **65** (1990) 2893; *Phys. Rev. Lett.* **67** (1991) 1670.

[13] Gompper G. and Kroll D.M., *Europhys. Lett.* **15** (1991) 783.

[14] Abraham F.F., *Phys. Rev. Lett.* **67** (1991) 1669.

[15] Komura S. and Baumgärtner A., *J. Phys. France* **51** (1990) 2395; *Phys. Rev. A* **44** (1991) 3511.

[16] Landau L.D. and Lifshitz E.M., "Theory of Elasticity", (Wiley, New York, 1970).

[17] Flügge W., Stresses in Shells (Springer Verlag, Berlin, 1973).

[18] Niordson F.I., "Shell Theory" , (North Holland, New York, 1985).

[19] Ou-Yang Zhong-can and Helfrich W., *Phys. Rev. A* **39** (1989) 5280.

[20] Zilker A., Engelhardt H. and Sackmann E., *J. Phys. France* **48** (1987) 2139.

[21] Waugh R. and Evans E.A., *Biophys. J.* **26** (1979) 115.

[22] Engelhardt H. and Sackmann E., *Biophys. J.* **54** (1988) 495.

[23] Zilker A., Strey H. and Sackmann E., The structure and conformation of amphiphilic membranes, R. Lipowsky, D. Richter and K. Kremer Eds. (Springer, Heidelberg, 1992).

[24] Peterson M.A., Strey H. and Sackmann E., *J. Phys. II France* (in press).

[25] Duwe H.P., Käs J. and Sackmann E., *J. Phys. France* **51** (1990) 945.

[26] Mutz M. and Helfrich W., *J. Phys. France* **51** (1990) 991.