The Shape of Inflated Vesicles

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Abstract
The conformation and scaling properties of self-avoiding fluid vesicles with zero extrinsic bending rigidity subject to an internal pressure increment $\Delta p > 0$ are studied using Monte Carlo methods and scaling arguments. With increasing pressure, there is a first-order transition from a collapsed branched polymer phase to an extended inflated phase. The scaling behavior of the radius of gyration, the asphericities, and several other quantities characterizing the average shape of a vesicle are studied in detail. In the inflated phase, continuously variable fractal shapes are found to be controlled by the scaling variable $x = \Delta p N^{3\nu/2}$ (or equivalently, $y = < V >/N^{3\nu/2}$), where $N$ is the number of monomers in the vesicle and $V$ the enclosed volume. The scaling behavior in the inflated phase is described by a new exponent $\nu = 0.787 \pm 0.02$.

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1. Introduction

In the absence of lateral tension, the shape of a fluid membrane is determined by its elastic bending energy [1,2]. At length scales small compared to the persistence length [3] $\xi_p = a_0 \exp(\frac{4\pi\kappa}{3k_B T})$, where $a_0$ is a microscopic length of the order of the monomer size and $\kappa$ is the bending rigidity, equilibrium configurations minimize the elastic bending energy, and thermal fluctuations can be ignored. At longer length scales, however, fluctuations lead to a decrease in the effective bending rigidity [4] so that membranes of linear dimension $L \gg \xi_p$ are expected to behave as crumpled objects characterized by the absence of long-range orientational order. At these length scales, where the effective rigidity is negligibly small, there is no energy scale and fluctuations determine the equilibrium shape.

Closed membranes or vesicles, usually of spherical topology, are of particular interest. For vesicles of radius $R \ll \xi_p$, it has been shown [5-7] that the intricate shapes observed in experiment correspond to equilibrium configurations which minimize the Helfrich elastic bending energy [5]

$$\mathcal{H} = 2\kappa \int dS (H - H_0)^2$$  \hspace{1cm} (1)

subject to the constraints of constant area and enclosed volume, where $H = \frac{1}{2}(1/R_1 + 1/R_2)$ is the mean curvature (expressed in terms of the principle radii of curvature $R_1$, $R_2$), and $H_0$ is the spontaneous curvature. Here, we are interested in the limit of small bending rigidities [8], where $\xi_p \ll R$. This case has been studied previously in considerable detail in $(d = 2)$-spatial dimensions, where the “vesicle” is a closed ring polymer [9-12]. The shape of these polymer rings can be characterized by the thermal averages of the eigenvalues $\lambda_2 > \lambda_1$ of the moment of inertia tensor. In particular, it has been shown [10] that the value of the shape parameter $\Sigma \equiv \frac{<\lambda_1>}{<\lambda_2>}$ changes continuously with the pressure increment

$$\Delta p = p_{in} - p_{out}.$$  \hspace{1cm} (2)

For $\Delta p = 0$, the polymer ring is a self-avoiding random walk, with $\Sigma \simeq 0.39$. For
\( \Delta p \neq 0 \), \( \Sigma \) is a universal function of the scaling variable

\[
x = pN^{2\nu}, \quad \text{with} \quad p = \Delta p a^2 / k_B T,
\]

where \( N \) is the number of monomers in the ring, \( a \) is a length scale of the order of the interparticle spacing, and \( \nu = 3/4 \) is the self-avoiding random walk exponent in two dimensions which describes the dependence of the radius of gyration, \( R_g \), on the number of monomers via

\[
< R_g^2 > \sim N^{2\nu}.
\]

For \( \Delta p > 0 \), the rings are inflated and approach a circular shape for large \( \Delta p \), so that \( \Sigma(x) \to \Sigma^+ = 1 \) for \( x \to \infty \). For \( \Delta p < 0 \), the rings collapse to a branched polymer shape, with \( \Sigma(x) \to \Sigma^- \simeq 0.23 \) in the limit \( x \to -\infty \).

In this paper we consider the corresponding problem for self-avoiding fluid vesicles in \( d = 3 \) spatial dimensions. Our analysis is based on an extensive Monte Carlo study of a simple string-and-bead model for randomly triangulated vesicles. The surface is modeled by a triangular network of \( N \) hard-sphere particles of diameter \( a = 1 \) connected by tethers of maximum extension \( \ell_0 = \sqrt{2} \) [13]. No explicit curvature term is included in the model. The fluid character of the membrane is incorporated by dynamically updating the triangulations. Details concerning the simulation procedure can be found in Refs. 14-16.

Using this model we have recently shown [16-19] that the behavior of fluid vesicles in \( d = 3 \) spatial dimensions is drastically different from that of ring polymers in two-dimensions. For example, already at \( \Delta p = 0 \) a vesicle exhibits a branched polymer conformation [16,17,20-23] characterized by the scaling laws [24]

\[
< R_g^2 > \sim N^{\nu_{bp}}, \\
< V > \sim N^{\nu_{bp}},
\]

with \( \nu_{bp} = 1 \). As \( \Delta p \) increases, there is a slow inflation until at some critical pressure difference \( \Delta p^*(N) > 0 \), there is a first-order transition to a high pressure
inflated phase [18]. Increasing the pressure still further causes the vesicle to slowly inflate until it becomes, ultimately, spherical. In this paper, we characterize the inflated phase in some detail. In Sec. 2, scaling laws for the radius of gyration, the enclosed volume, and the width of the vesicle wall are derived using a generalization of de Gennes’ blob picture. The scaling behavior of the critical pressure difference $\Delta p^*(N)$ at the first-order transition is discussed in Sec. 3, and data for the radius of gyration and enclosed volume in the inflated phase are analyzed. A simple argument is given which indicates that $\Delta p^* \sim N^{-1/2}$. Sec. 4 contains a discussion of the behavior in the two-phase region, and data taken in both the constant pressure and constant volume ensembles are compared. A scaling analysis of various quantities characterizing the vesicle shape in the inflated phase is presented in Sec. 5; the paper closes with a brief summary and discussion.

2. Scaling

The scaling behavior in the inflated phase can be derived using a simple generalization of the blob picture of de Gennes [11,25-27,18]. Consider a $(d - 1)$-dimensional crumpled membrane embedded in $d$-dimensional space. In the blob picture, one envisages the membrane breaking up into a set of $N_b$ weakly stretched blobs of area proportional to $\xi_{\sigma}^{-d-1} = k_BT/\sigma$, where $\xi_{\sigma}$ is the tensile length, under the influence of a uniform extensional tension $\sigma$ applied to the membrane perimeter. Since the blobs themselves are only weakly stretched, it is further assumed that

$$\xi_{\sigma}^{-d-1} \sim M_b^{\nu},$$

where $M_b = N/N_b$ is the number of monomers in a blob. It is then argued that the blobs become independent on length scales much larger than $\xi_{\sigma}$ so that the total projected surface area $< A_b >$ is proportional to $\xi_{\sigma}^{-d-1}(N/M_b) \sim N\sigma^{1/\nu - 1}$. If an inflated vesicle is regarded as a (hyper-) spherical bubble of radius $R$ with a surface tension $\gamma$, one has $\Delta p = 2\gamma/R$. Identifying the surface tension with the
stretching tension $\sigma$, and taking $< A_b > \sim R^{d-1}$, we arrive at the scaling form

$$R_g \sim p^\omega N^{\nu_+}$$

(7)

for the radius of gyration of an inflated vesicle, with $p = \Delta p a^d/k_BT$, and the exponents

$$\omega = \frac{1 - \nu}{d\nu - 1}$$

$$\nu_+ = \frac{\nu}{d\nu - 1}.$$  

(8)

$\nu$ is a new exponent which characterizes the scaling behavior within a blob in the inflated phase.

Eq. (7) is the large extension limit of a more general scaling form. Assuming that the radius of gyration $R_g$ and the tensile strength $\xi_\sigma$ are the only two relevant length scales in the problem, it is natural to expect the projected area $A_b$ to scale as

$$A_b = R_g^{d-1} \Omega_0(\xi_\sigma/R_g) = N^\nu \Omega(\sigma N^\nu),$$

(9)

where the last relation follows from the definition $R_g^{d-1} \sim N^\nu$, and $\xi_\sigma^{d-1} \sim \sigma^{-1}$. For large $\sigma$, $A_b$ should vary linearly with $N$, so that

$$\Omega(z) \sim z^{1/\nu - 1} \text{ for } z \to \infty.$$  

(10)

This implies $A_b \sim N\sigma^{(1-\nu)/\nu}$ in this limit. For an inflated vesicle we have (see discussion preceding Eq.(7)) $\sigma \sim pR_g$ so that

$$A_b \sim R_g^{d-1} \sim N(pR_g)^{(1-\nu)/\nu}$$

(11)

in the large extension limit. Solving this equation for $R_g$, we arrive at (7) with the exponents (8). From (9), we see that the full scaling form in the inflated phase is

$$< R_g^2 > = p^{2\omega} N^{2\nu_+} \Xi_p(pN^{d\nu/(d-1)}).$$

(12)

Similarly, the mean volume can be shown to scale as

$$< V > = p^{d\omega} N^{d\nu_+} \Gamma_p(pN^{d\nu/(d-1)}).$$

(13)
Finally, Eq. (13) suggests that the scaling behavior of the mean square radius of gyration of vesicles fluctuating at fixed enclosed volume (instead of at fixed pressure) scales as

\[ \langle R_g^2 \rangle = V^{2/d} \Xi_V \left( V N^{-d \nu/(d-1)} \right). \]  

(14)

in the single phase inflated regime. Note that for \( d = 2 \), these results are identical with the those of Ref. 11 for polymer rings.

We can go further and calculate the radial extension of the highly convoluted surface of the vesicle. The radial thickness \( \xi \) should scale like the projected area, Eq.(9),

\[ \xi^{d-1} = N^\nu Q(\sigma N^\nu). \]  

(15)

In the large tension limit, \( \xi \) should become independent of \( N \), so that

\[ Q(z) \sim z^{-1} \quad \text{for} \quad z \to \infty. \]  

(16)

This implies that

\[ \xi \sim (pR_g)^{-1/(d-1)} \sim p^{-\nu_p N^{-\nu_p/(d-1)}} \]  

(17)

in this limit. The thickness \( \xi \) described by (17) is just the radius (6) of the blobs in the blob picture. It should therefore be considered to be the intrinsic thickness of the vesicle walls. In particular, it does not take into account the effect of capillary waves on length scales larger than \( \xi \). These excitations can be easily estimated using an effective Hamiltonian for a nearly flat interface of projected area \( A_b \) (with periodic boundary conditions). We introduce a coarse grained variable \( u(r) \) which measures the distance of the membrane from some reference plane. On length scales larger than \( \xi \), the lateral tensions dominates the fluctuations, so that

\[ H_{eff} = \frac{\sigma}{2} \int_{A_b} d^{d-1} r (\nabla u)^2, \]  

(18)

with an implicit short length scale cutoff \( \xi \). The amplitude of the fluctuations is
calculated with the help of the equipartition theorem, as usual, with the result

\[
<u^2> = \int_{R_g^{-1} < q < \xi^{-1}} \frac{d^d q}{(2\pi)^{d-1}} \frac{k_B T}{\sigma q^2}
\]

\[
\sim \begin{cases} 
\frac{k_B T}{\sigma} R_g^{3-d} & \text{for } d < 3 \\
\frac{k_B T}{\sigma} \ln(R_g/\xi) & \text{for } d = 3
\end{cases}
\]

Inserting the results for \(\sigma\) and \(R_g\), we arrive at

\[
\sqrt{\frac{<u^2>}{<R_g^2>}} \sim \begin{cases} 
(pN^{d\nu/(d-1)})^{-1/(d\nu-1)} & \text{for } d < 3 \\
(pN^{3\nu/2})^{-1/(3\nu-1)} \sqrt{\ln(pN^{3\nu/2})} & \text{for } d = 3
\end{cases}
\]

so that capillary wave broadening dominates the thickness of the vesicle walls for all \(d \leq 3\). However, for \(d = 3\), which is the case of interest, the effect of capillary waves is very weak, and can only be observed for very large vesicles.

3. Radius of Gyration and Volume

The behavior of the average volume \(<V>\) of a vesicle as a function of \(\Delta p\) is shown in Fig. 1 for three different system sizes, \(N = 47, 127,\) and \(247\). Each data point corresponds to an equilibrium average over 20–40 million Monte Carlo steps per monomer (MCS). The evolution of a discontinuity in \(<V>\) with increasing \(N\) is clearly visible. Since the transition pressure is difficult to determine from \(<V(p)>\) itself, we have also calculated the probability distribution \(P(V)\). For \(N = 47\), the distribution has a single sharp peak for low and high pressures, but broadens and exhibits a weakly bimodal structure near \(\Delta p^* = 1.05 \pm 0.05\), which we identify with the location of the transition. The distribution for \(N = 127\) is shown in Fig. 2 for various pressures near the transition. For \(\Delta p = 0.50\), a bimodal distribution is found, with a somewhat larger peak at large volumes. We estimate that the transition occurs at \(\Delta p^* = 0.47 \pm 0.02\) in this case. For \(N = 247\) it is more difficult to determine the location of the transition, since the vesicle jumps back and forth between the inflated and the branched polymer states so rarely that the equilibrium probability distribution for \(V\) could not be determined.
with sufficient accuracy. However, by studying the time dependence of \( V \) for \( \Delta p = 0.30 \) and \( \Delta p = 0.35 \), see Fig. 3, we conclude that the transition occurs at \( \Delta p^* = 0.34 \pm 0.01 \) for \( N = 247 \). From the three vesicles sizes we have studied, it is difficult to determine the \( N \)-dependence of \( \Delta p^* \). Nevertheless, we present a simple argument at the end of this Section which suggests that \( \Delta p^* \sim N^{-1/2} \). This is in excellent agreement with our estimates for the transition pressures quoted above for \( N = 127 \) and \( N = 247 \).

As already shown in Ref. 18, the data for \( < V > \) in the inflated phase \((\Delta p > \Delta p^*)\) scale according to (13) with an exponent \( \nu = 0.787 \pm 0.020 \). In fact, all our data for both the volume and the mean square radius of gyration in the inflated regime scale for this value of \( \nu \). The scaling function \( F \) for the volume,

\[
F(pN^{3\nu/2}) = < V > / (p^{3\nu} N^{3\nu}),
\]

and the scaling function \( \Xi_V(< V > / N^{3\nu/2}) \) for the mean square radius of gyration, see Eq. (14), are shown in Fig. 4. At very large inflations, scaling breaks down. The deviation of the last (large \( \Delta p \)) data point for each vesicle size in Fig. 4a is due to this effect and thus delimits the scaling region. For large scaling argument, \( F(y) \) must approach a constant value. It is interesting to note that this occurs rather rapidly, so that significant deviations can be observed only for small pressures and vesicle sizes. In fact, for \( N \to \infty \), this asymptotic limit is is attained in all of the inflated phase, since as argued below, \( \Delta p^* \) scales as \( N^{-1/2} \) for large \( N \) so that \( \Delta p N^{3\nu/2} \to \infty \) for all \( \Delta p > \Delta p^* \).

In Fig. 4b, as well as in Fig. 7 below, we have replaced \( N \) by \( (N - N_0) \) in the scaling function in order to hasten convergence when \( N \to \infty \). This parameterization incorporates the leading corrections to scaling.

Given these results, there is a simple argument which indicates that the transition between the collapsed and inflated phases occurs at a critical pressure difference \( \Delta p^* \) which scales as

\[
\Delta p^* \sim N^{-1/2},
\]
independent of the value of $\nu$. For $\Delta p = 0$, the vesicle exhibits a branched polymer conformation. To leading order, the configurational entropy $S(N)$ of a branched polymer of length $N$ is given $S(N) = N \ln(z) - \theta \ln(N)$, where $z$ is a nonuniversal quantity independent of $N$ and $\theta = 3/2$ [24]. The free energy in the deflated phase therefore scales as

$$\beta F_- = -N \ln(z) - c \Delta p N$$

(23)

to leading order in $N$ and $\Delta p$, where $c$ is a constant of order unity. In writing (23) we have included the leading pressure dependent contribution [18] $\Delta p < V > \sim \Delta p N$. In the inflated phase, on the other hand, we know that $< V > = -\partial \beta F / \partial \Delta p$ scales as $V_0^+ \Delta p^{3\omega} N^{3\nu_+}$ [28]. The leading contribution to the free energy in the inflated phase is, therefore,

$$\beta F_+ = -\frac{V_0^+}{3\omega + 1} (\Delta p)^{3\omega+1} N^{3\nu_+}.$$  

(24)

The transition occurs when the free energies of the deflated and inflated phases become equal, i.e. for

$$N \sim (\Delta p^*)^{3\omega+1} N^{3\nu_+},$$

(25)

or, equivalently, $\Delta p^* \sim N^{-1/2}$, independent of $\nu$.

4. Two-Phase Region

We have performed simulations in both the constant-pressure and constant-volume ensembles. Our motivation is twofold. First, we want to check the equivalence of the two ensembles for large vesicles. This is important because experiments are usually performed at constant volume [7,29]. Second, we want to see what a vesicle looks like in the ‘two-phase’ region. Namely, is there two-phase coexistence between a branched polymer region and an inflated part of the vesicle?

A few typical configurations for $N = 247$ and $V = 120$ are shown in Fig. 5. They demonstrate that the vesicle does indeed exhibit two-phase coexistence, with a tree-like part attached to a quasi-spherical region. Also, we find that $N = 247$ is large enough to give essentially identical results in the inflated phase for the two
ensembles. This can be seen in Fig. 4b, where the scaling function of the mean square radius of gyration is plotted for a range of volumes covering the inflated phase, the two-phase region, as well as the collapsed, branched polymer phase. Data obtained in both ensembles are included in this figure. Note that the data appear to scale over the whole range of volumes. We will see this remarkable behavior for other scaling functions in Section 5. We want to emphasize, however, that no true scaling can occur in the two-phase region.

For fully extended spherical vesicles, the volume and the radius of gyration are related by \( V = \frac{4}{3} \pi R_g^3 \). Thus,

\[
\tilde{\pi} \equiv \frac{3}{4} < V > < R_g^2 >^{-3/2}
\]  
(26)

is a measure for the deviation of the shape of the vesicle from an ideal sphere. Our results for \( \tilde{\pi}(< V > N^{-3\nu/2}) \) are shown in Fig. 6. For \( V \) large at fixed \( N \), \( \tilde{\pi} \rightarrow \pi \), as expected. Fig.6 shows again that the data appear to scale not only in the inflated phase, but in the two-phase region as well.

5. Mean Vesicle Shapes in Inflated Phase

In order to characterize the shape of the vesicles, we have studied the eigenvalues \( \lambda_1 < \lambda_2 < \lambda_3 \) of the moment of inertia tensor,

\[
\mathcal{T}_{\alpha,\beta} = \frac{1}{2N^2} \sum_{i,j} \frac{q_i q_j}{6} [r_i^\alpha - r_j^\alpha][r_i^\beta - r_j^\beta],
\]  
(27)

where \( r_i \) is the position of monomer \( i \), and \( q_i \) is its coordination number. In particular, we have analysed the asphericities, \( \Gamma_1 = < \lambda_1/\lambda_3 > \), \( \Gamma_2 = < \lambda_2/\lambda_3 > \), as well as the quantities \([17,30,31]\)

\[
\Delta_d = \frac{d}{d-1} \frac{< \text{Tr} \mathcal{T}^2 >}{< (\text{Tr} \mathcal{T})^2 >},
\]  
(28)

and

\[
S_d = \frac{d^2}{(d-1)(d-2)} \frac{< \text{Tr} \mathcal{T}^3 >}{< (\text{Tr} \mathcal{T})^3 >},
\]  
(29)
where $\hat{T}_{\alpha\beta} = T_{\alpha\beta} - \bar{\lambda} \delta_{\alpha\beta}$, and $\bar{\lambda} = \frac{1}{d} \sum_{i=1}^{d} \lambda_i$. $0 \leq \Delta_d \leq 1$ is a normalized measure of the anisotropy. The sign of $-\frac{1}{(d-1)^2} \leq S_d \leq 1$ determines whether the object is oblate ($S_d < 0$), or prolate ($S_d > 0$); its magnitude is a measure of the strength of the anisotropy. For $d = 3$, one has

$$\Delta_3 = \frac{\lambda_1^2 + \lambda_2^2 + \lambda_3^2 - (\lambda_1 \lambda_2 + \lambda_2 \lambda_3 + \lambda_3 \lambda_1)}{\lambda_1 + \lambda_2 + \lambda_3}$$

(30) and

$$S_3 = \frac{(\lambda_1 - \bar{\lambda})(\lambda_2 - \bar{\lambda})(\lambda_3 - \bar{\lambda})}{2 \lambda_3^2}.$$  

(31)

Let us first consider the averaged eigenvalues $<\lambda_i>$, $i = 1, 2, 3$, of the moment of intertia tensor. In the inflated phase, the deviation of the vesicle from a completely spherical shape is determined by the fluctuations studied in Section 2. To leading order, we therefore expect

$$<\lambda_i> \simeq \frac{1}{3} \left[ \sqrt{<R_g^2>} + \frac{1}{2} c_i \sqrt{<u^2>^2} \right].$$

(32)

so that $3 <\lambda_i> / <\lambda_1 + \lambda_2 + \lambda_3>$

$$= 1 + c_i y^{1/(3-3\nu)},$$

(33)

with $y = <V> N^{-3\nu/2}$. Since $\sum_i <\lambda_i> = <R_g^2>$, one has $\sum_i c_i = 0$. With the critical exponent $\nu = 0.787$ determined above, we therefore expect $[3 <\lambda_i> / <R_g^2> - 1]$ to decay asymptotically as $y^{-1.565}$. The Monte Carlo data are shown in Fig. 7. The eigenvalues $<\lambda_1>$ and $<\lambda_3>$ exhibit the expected asymptotic power law behavior with an exponent which is very close to the value quoted above. $[1 - 3 <\lambda_2> / <R_g^2>],$ on the other hand, decays much faster for large $y$, approximately as $y^{-3.05}$. Indeed, the data are consistent with $c_2 \simeq 0$. Since we expect, in general, that corrections to spherical shape in the large inflation limit can be expressed as a power series in $\sqrt{<u^2> / <R_g^2>}$, the asymptotic behavior in this case should be given by the next to leading term, which is $y^{-2/(3-3\nu)} = y^{-3.13}$, in good agreement with our numerical result. From Fig. 7, the constants $c_i$ in Eq. (33) are found to be

$$c_1 = -0.012, \quad c_2 = 0, \quad \text{and} \quad c_3 = 0.013,$$

(34)
consistent with the requirement \( \sum_i c_i = 0 \).

The asphericities can be analysed in a similar way. Under the assumption that \( \Gamma_{1,2} = \langle \lambda_{1,2} / \lambda_3 \rangle \geq \langle \lambda_{1,2} \rangle / \langle \lambda_3 \rangle = \Sigma_{1,2} \), we obtain [32]

\[
1 - \Gamma_{1,2}(y) = -(c_{1,2} - c_3)y^{-1/(3-3\nu)}
\]

with \( y = \langle V \rangle N^{-3\nu/2} \). It has been shown in Ref. 17 that for \( \Delta p = 0 \) the \( \Gamma_i \) and \( \Sigma_i \) differ by very little, so that this assumption seems justified. The data shown in Fig. 8 are in reasonable agreement with the scaling form (35), with an amplitude which is indeed close to the difference of the \( c_i \)’s. Note, however, that Eq. (35) is only an approximation, and that in general, one would expect \( 1 - \Gamma_i \) to scale with an independent exponent. In fact, it appears that the asymptotic slope of \( 1 - \Gamma_1 \) does differ somewhat from (35).

Finally, the shape parameters \( \Delta_3 \) and \( S_3 \) are shown in Fig. 9. The data again fall onto a single curve when the scaling variable \( y = \langle V \rangle N^{-3\nu/2} \) is used. The asymptotic behavior in this case is found to be

\[
\begin{align*}
\Delta_3(y) & \sim y^{-\gamma_\Delta} \\
S_3(y) & \sim y^{-\gamma_S},
\end{align*}
\]

with \( \gamma_\Delta \simeq 3.31 \), and \( \gamma_S \simeq 6.22 \). Since these quantities are also averages of the eigenvalues \( \lambda_i \), it is tempting to conjecture that

\[
\gamma_\Delta \simeq 2/(3 - 3\nu) \quad \text{and} \quad \gamma_S \simeq 4/(3 - 3\nu).
\]

Note that \( S_3 > 0 \), but very small, so that the vesicles are prolate on average.

6. Summary and Discussion

The scaling behavior of fluid vesicles subject to an internal pressure increment \( \Delta p > 0 \) has been studied. Clear evidence has been presented for a phase transition from a low-pressure branched-polymer phase to a high-pressure inflated phase. Several quantities characterizing the vesicle shape, such as the average
eigenvalues of the moment of inertia tensor and the asphericities, have been determined using Monte Carlo methods. In the inflated phase, all quantities are found to be universal functions of the scaling variable \( x = \Delta p N^{3\nu/2} \) (or equivalently, \( y = \langle V \rangle N^{-3\nu/2} \)), with \( \nu = 0.787 \pm 0.02 \). The asymptotic scaling behavior for large inflation can be understood using simple scaling arguments.

Much still needs to be learned regarding the properties of the inflated phase, however. Although we feel that the nature of the transition between the branched polymer phase and the inflated phase is well established, the relatively modest system sizes which can currently be studied make it difficult to determine \( \nu \) precisely, or even exclude the possibility that the behavior we observe in the inflated phase is dominated by crossover effects. Nevertheless, the fact that arguments based on the blob picture of de Gennes yield such a concise and consistent description of the observed scaling behavior makes us believe that the scenario described above is basically correct. In any case, a better theoretical understanding of the nature of the inflated phase is necessary if we are to appreciate the implications of these results.

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REFERENCES

[1] *Statistical Mechanics of Membranes and Surfaces*, edited by D.R. Nelson, T. Piran, and S. Weinberg (World Scientific, 1989).

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

[3] P.G. de Gennes and C. Taupin, J. Chem. Phys. **86**, 2294 (1982).

[4] L. Peliti and S. Leibler, Phys. Rev. Lett. **54**, 1690 (1985).

[5] W. Helfrich, Z. Naturforsch. **28c**, 693 (1973); H.J. Deuling and W. Helfrich, J. Phys. (Paris) **37**, 1335 (1976).

[6] S. Svetina and B. Zeks, Eur. Biophys. J. **17**, 101 (1989).

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

[8] Synthetic lipid bilayer vesicles containing a small percentage of bola lipid are known to have bending rigidities of the order of $k_B T$, or less, see H.P. Duwe, J. Käs, E. Sackmann, J. Phys. (Paris) **51**, 945 (1990).

[9] S. Leibler, R.R.P. Singh, and M.E. Fisher, Phys. Rev. Lett. **59**, 1989 (1987).

[10] C.J. Camacho and M.E. Fisher, Phys. Rev. Lett. **65**, 9 (1990).

[11] A.C. Maggs, S. Leibler, M.E. Fisher, and C.J. Camacho, Phys. Rev. A**42**, 691 (1990).

[12] C.J. Camacho, M.E. Fisher, and R.R.P. Singh, J. Chem. Phys. **94**, 5693 (1991).

[13] This value of $\ell_0$ is small enough to ensure self-avoidance while still yielding bond-flip acceptance rates ($\sim 8\%$ for $\Delta p = 0$ and $\sim 2 - 3\%$ for large $\Delta p$) which are large enough to allow the monomers to diffuse in the membrane surface.

[14] A. Billoire and F. David, Nucl. Phys. B**275**, 617 (1986); D.V. Boulatov, V.A. Kazakov, I.K. Kostov, and A.A. Migdal, Nucl. Phys. B**275**, 641 (1986).
As can be seen from Fig. 4a, this is a good approximation only for the two larger system sizes we studied. For $N = 47$ there appear to be finite $\Delta p N^{3\nu/2}$ corrections (see Eq. (13)). We will neglect this effect in the current discussion.
our simulations, such corrections cannot be distinguished from simple power laws.
FIGURE CAPTIONS

1.) The average volume $<V>$ as a function of the pressure increment $p$, for $N = 47 (\Diamond), N = 127 (\times)$, and $N = 247 (\bigodot)$. $p = \Delta p a^3/k_B T$, where the constant $a$ is of the order of the interparticle spacing; for convenience we set $a^3/k_B T = 1$ here and in the following.

2.) Probability distribution $P(V)$ for $N = 127$, at $p = 0.40$ (dashed line), $p = 0.50$ (full line), and $p = 0.60$ (dotted line).

3.) Volume $V$ vs. number of Monte Carlo steps per monomer, for $N = 247$, $p = 0.30$ and $p = 0.35$. In the case $p = 0.30$, the data are shifted by $\Delta V = -40$ in order to separate the two curves.

4.) Scaling functions for (a) the volume, $F = <V> p^{3\omega} N^{-3\nu}$, as a function of the scaled pressure $p N^{3\nu/2}$, and (b) the mean square radius of gyration, $\Xi_V = <R_g^2> <V>^{-2/3}$, as a function of the scaled volume $<V> (N - N_0)^{-3\nu/2}$, with $N_0 = 13$. Data for both the constant-pressure ensemble with $N = 47 (\Diamond), N = 127 (\times)$, and $N = 247 (\bigodot)$, as well as the constant volume ensemble with $N = 247 (\bigodot)$ are plotted.

5.) Configurations of a vesicle in the two-phase region with $N = 247$ monomers and volume $V = 120$, after 4, 8, 12, 16 and 20 million MCS.

6.) The volume to area ratio $\tilde{\pi}(<V> N^{-3\nu/2})$ of an inflated vesicle. Data for both the constant-pressure ensemble with $N = 47 (\Diamond), N = 127 (\times)$, and $N = 247 (\bigodot)$, as well as the constant volume ensemble with $N = 247 (\bigodot)$ are plotted.

7.) Scaling plot of the eigenvalue ratios $3 < \lambda_i > / < \lambda_1 + \lambda_2 + \lambda_3 >$ of the moment of inertia tensor as a function of $<V> (N - N_0)^{-3\nu/2}$, with $N_0 = 5$. The solid lines in (a) and (c) indicate the asymptotic scaling behavior (33) with the amplitudes $c_3 = 0.013$ and $c_1 = -0.012$, respectively. Data for both the constant-pressure ensemble with $N = 47 (\Diamond), N = 127 (\times)$, and $N = 247 (\bigodot)$, as well as the constant volume ensemble with $N = 247 (\bigodot)$ are plotted.
8.) Scaling plot of the asphericities $\Gamma_1$ and $\Gamma_2$ as a function of $< V > N^{-3\nu/2}$.

The solid lines indicate the scaling behavior (35) with the amplitudes $(c_3 - c_1) = 0.025$ and $(c_3 - c_2) = c_3 = 0.013$. Data for both the constant-pressure ensemble with $N = 47$ (⋄), $N = 127$ (×), and $N = 247$ (□), as well as the constant volume ensemble with $N = 247$ (○) are plotted.

9.) Scaling plot of $\Delta_3$ and $S_3$ as a function of $< V > N^{-3\nu/2}$. Data for both the constant-pressure ensemble with $N = 47$ (○), $N = 127$ (×), and $N = 247$ (□), as well as the constant volume ensemble with $N = 247$ (○) are plotted.