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Molecular dynamics simulations of the structure of closed tethered membranes

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Abstract. — The equilibrium structure of closed self-avoiding tethered vesicles are investigated by molecular dynamics simulations. To allow for local flexibility, the vesicles are constructed by connecting linear chains of \( n \) monomers to form a closed membrane. For all \( n \) studied, \( 0 \leq n \leq 16 \), we find that the membranes remain flat. The height fluctuations \( \langle h^2 \rangle \sim N^\zeta \), where \( \zeta = 0.55 \pm 0.02 \) and \( N \) is the total number of monomers in the vesicle. This result for \( \zeta \) is significantly lower than earlier estimates for open membranes with a free perimeter but is in agreement with recent estimates of Abraham for membranes without a free perimeter. Results for the static structure factor, \( S(q) \), are calculated and compared to recent data on red blood cell skeletons.

1. Introduction.

Recently, there has been an extensive amount of work, both theoretical and numerical, investigating the properties of \( D \) dimensional self-avoiding tethered surfaces embedded into \( d \) dimensions [1, 2]. While linear polymers (\( D = 1 \)) is a well-known example of this type of system, recent interest has focused on 2-dimensional tethered surfaces (\( D = 2 \)). Kantor et al. [3] suggested, based on a simple Flory-level theory, that in the presence of only excluded volume interactions, a tethered membrane would crumple and that its size \( R_g \) would scale as \( R_g^{df} \sim N \), where \( N \) is the number of monomers in the membrane and \( df \) is the fractal dimension. For \( D = 2 \) and \( d = 3 \), Flory theory predicts that \( df = 2.5 \) [3]. This conclusion was supported by renormalization group calculations [3-6] which suggested that the flat phase was unstable in \( d = 3 \) and by early Monte Carlo simulations [3]. However, more extensive computer simulations [7-15] on large system has shown convincingly that in fact tethered membranes do not crumple but remain flat (\( R_g^2 \sim N \) if one includes only excluded volume interactions. This lack of a crumpling transition in \( d = 3 \) has been explained [11] in terms of an implicit bending}

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rigidity which is induced by the self-avoidance requirement even when no such term is present in the microscopic Hamiltonian. Then, if bending rigidity is relevant, one cannot expect the Flory theory to work. An alternative explanation by Goulian [16] based on a generalization of Edward’s model which uses a Gaussian variational approximation and by Guitter and Palmeri [17] and Le Doussal [18] based on expansion in large embedding space dimension \( d \) suggests that the flat phase is stable for \( d = 3 \) and tethered membranes crumple only for \( d > 4 \). This is in agreement with the recent molecular dynamics simulations of Grest [15]. However, in the presence of attractive interactions, Liu and Plischke [19] find that the membranes undergo a phase transition from the high temperature flat phase to a low temperature crumpled phase. This crumpled phase seems to exist over a range of temperatures and is characterized by a fractal dimension \( d_f \sim 2.5 \).

While some understanding of why self-avoiding membranes do not crumple is beginning to emerge, there remain a number of unsettled issues. Important unresolved issues include the precise value for the exponent \( \zeta \) that describes the size of the height fluctuations \( \langle h^2 \rangle \sim N^{\zeta} \) and the interpretation of several recent scattering experiments on tethered surfaces. Lipowsky and Girardet [20] used a scaling analysis to show that \( \zeta \geq 1/2 \) and suggested from a Monte Carlo simulation for a continuum model of a solid-like elastic sheet that \( \zeta = 1/2 \). This result agreed with a simple one-loop self-consistent theory of Nelson and Peliti [21] which predicted \( \zeta = 1/2 \). If this were true, then the membrane would have a finite shear modulus on large scales, since shear modulus scales as \( \sim N^{\sigma_{\mu}/2} \) and \( \eta_{\mu} = 4\zeta - 2 \). This result is in contrast with all previous simulations on bead-spring models which gave \( \zeta = 0.64 \pm 0.04 \). However, Abraham [22] has pointed out that this larger value for \( \zeta \) may be due to finite size effects since all these bead-spring type simulations were performed on open membranes with a free perimeter. Abraham replaced the free-perimeter with a periodic boundary condition (pbc) by using a computational box which was allowed to vary in size using a constant pressure molecular dynamics technique and found \( \zeta = 0.53 \pm 0.03 \), consistent with Lipowsky and Girardet [20]. Recently Le Doussal and Radzihovsky [23] carried out a self-consistent screening approximation which improved on the Nelson-Peliti theory by allowing non-trivial renormalization of the elastic moduli. They found \( \zeta = 0.590 \), in between the previous estimates. In this paper, we present the results of extensive molecular dynamics simulations on closed vesicles in which we find \( \zeta = 0.55 \pm 0.02 \), slightly larger than, but consistent with Abraham’s result [22]. This finding gives further evidence to the suggestion that the larger values of \( \zeta \) obtained earlier for open membranes are probably due to finite size effects due to the free perimeter. (See note added in proof.)

A second, important, unresolved issue concerns the interpretation of recent scattering experiments on experimental realizations of polymerized membranes. A laser scattering measurement [24] of the static structure function \( S(q) \) for graphite oxide crystalline membranes suggests that they are crumpled and not flat as indicated by computer simulations. For a flat membrane, one expects that for large enough systems, \( S(q) \sim q^{-2} \) while for a crumpled membrane \( S(q) \sim q^{-2.5} \). While Wen et al. [24] do observe a power law decay with a slope of 2.54, one should not interpret this as convincing evidence for a crumpled phase, since these experiments are on randomly oriented membranes. Abraham and Goulain [25] have shown that for a wide range of \( q \), the isotropically averaged \( S(q) \sim q^{-2.35} \) for open, tethered membranes containing as many as 29420 monomers. They found no evidence for a \( q^{-2} \) regime even though these membranes are definitely flat. The expected \( q^{-2} \) regime which must be present for large enough samples is apparently suppressed and only the rough regime [26] in which \( S(q) \sim q^{-3+\epsilon} \) is observed. This interpretation is consistent with the computer simulations of Abraham and Goulian [25] on open membranes where \( \zeta \approx 0.64 \). It also agrees with recent X-ray and light scattering experiments [27, 28] on isolated red blood cell (RBC) skeletons in high salt in which a \( q^{-2} \) regime at low \( q \) was followed by a \( q^{-2.35} \) regime for larger \( q \), consistent
with the behavior expected for a flat membrane. While it is fairly certain that the interaction in the red blood cell skeletons is purely repulsive, it is possible that attractive interactions [19] are important for the graphite oxide crystalline membranes and they are in fact crumpled. Additional experiments are needed to clarify whether attractive interactions play a role in this case.

![Diagram of spectrin/actin membrane skeleton of RBC](image)

**Fig. 1.** — Illustration of the spectrin/actin membrane skeleton of RBC (from Ref. [27]) showing the main components and the triangulated structure. For clarity, the skeleton is drawn in an expanded state. *In vivo*, the average node distance is about 1/3 of the contour length of spectrin tetramers (2000 Å).

While the graphite oxide crystalline membranes [24] are open membranes with a large local bending rigidity, induced by the close packing of monomers, the RBC skeleton is a closed vesicle, roughly spherical, made up of flexible worm-like chains in a triangulated network. The RBC skeleton (Fig. 1) is attached to the cytoplasmic side of the liquid lipid cell membrane and can be isolated by detergent treatment. The spectrin tetramers which made up the chains have a contour length of \( \approx 2000 \) Å and a persistence length of 100 – 200 Å in high ionic strength buffer. Mammalian RBC skeletons contain \( \sim 70,000 \) triangular meshes. Obviously, this system is too complex to simulate directly on the computer. However, the essential properties of the membrane on intermediate length scales can be obtained from a study of a coarse-grained bead-spring model which has the same topology. Following Abraham’s study of open membranes of linear flexible chains connected at tri-functional vertices [12], we study a triangular array of linear chains containing \( n \) monomers connected to form a closed vesicle as shown in figure 2. All but 12 of the vertices are 6-fold coordinated. The remaining 12 vertices are 5-fold coordinated, as is necessary to close the vesicle. In high salt, the actin oligomers have a mean spacing of 400 – 500 Å, or about 1/5 – 1/4 of their fully extended length. Using this as a means of mapping to our bead spring model, we estimate that each linear chain should contain about 50-60 monomers for the model studied here. Since the largest system we can equilibrate and study in a few hundred hours of Cray time is about 25,000 monomers, our initial studies were limited to \( n \leq 16 \) in order to increase the number of triangles in the system. Larger systems will have to wait for the next generation of supercomputers. However, in spite of the fact that
our largest samples are about 50 times smaller than the experimental systems and contain only excluded volume interactions, the cross sections of the simulated membranes compare very favorably with the shape of RBC skeletons. Results for $S(q)$ are also in agreement with recent X-ray and light scattering studies of Schmidt et al. [28].

![Fig. 2. Illustration of the initial state for a membrane of size $N = 9002$ with $n = 8$ additional monomers between each vertex. This system contains 1080 edges and 720 triangular faces. Note that the bond lengths are not exactly the same as the points have been projected onto the surface of a sphere. If all bond lengths would be equal, the membrane would have the shape of an icosahedra.](image)

The outline of the paper is as follows. In section 2, we describe the model and the molecular dynamics technique used. In section 3, we present our results for the eigenvalues of the moment of inertia tensor, the radius of gyration and the static structure function for membranes containing from $252 - 25002$ monomers with $0 \leq n \leq 16$ monomer per linear chain. All membranes were connected to form an icosahedra. In section 4, we discuss the relaxation of the membranes and then briefly summarize our results in section 5.

2. Model and method.

Each membrane consists of $N$ monomers of mass $m$ connected by anharmonic springs. The monomers interact through a shifted Lennard-Jones potential given by

$$U^0(r) = \begin{cases} 
4\varepsilon \left[ \left( \frac{\sigma}{r} \right)^{12} - \left( \frac{\sigma}{r} \right)^6 + \frac{1}{4} \right] & \text{if } r \leq r_c; \\
0 & \text{if } r > r_c,
\end{cases}$$

with $r_c = 2^{1/6}\sigma$. This purely repulsive potential represents the excluded volume interactions that are dominant in the case of monomers immersed in a good solvent. For monomers which
are tethered (nearest neighbors) there is an additional attractive interaction potential as described elsewhere [14, 15]. No explicit bending terms are included.

Denoting the total potential of monomer \( i \) by \( U_i \), the equation of motion for monomer \( i \) is given by

\[
m \frac{d^2 r_i}{dt^2} = - \nabla U_i - m \Gamma \frac{dr_i}{dt} + W_i(t).
\]

(2)

Here \( \Gamma \) is the bead friction which acts to couple the monomers to the heat bath. \( W_i(t) \) describes the random force acting on each bead. It can be written as a white noise term with

\[
\langle W_i(t) \cdot W_j(t') \rangle = \delta_{ij} \delta(t - t') 6 k_B T \eta,
\]

(3)

where \( T \) is the temperature and \( k_B \) is the Boltzmann constant. We have used \( \Gamma = \tau^{-1} \) and \( k_B T = 1.2 \epsilon \). Here \( \tau = \sigma (m/\epsilon)^{1/2} \). The equations of motion are then solved using a velocity-Verlet algorithm [29] with a time step \( \Delta t = 0.012 \tau \) [15]. With this choice of parameters, the average bond length between nearest neighbors that are tethered is 0.97\( \sigma \). The program was vectorized for a supercomputer following the procedure described by Grest et al. [30] except that the periodic boundary conditions were removed and additional interactions between monomers which are tethered was added. For a tethered membrane of size \( N = 16002 \), 1 million time steps took about 80 hours of cpu time on our Cray XMP 14/s. Further details of the method can be found elsewhere [31].

The simulations were performed on two-dimensional triangular arrays of \( n \) monomers connected to form a closed vesicle. This was done by construction of an icosahedron with \( n_V \) monomers per edge. An additional \( n \) monomers connected linearly were then added between each node. The total number of monomers \( N = 10 n_V^2 (1 + 3 n) + 2 \) with \( 30 n_V^2 \) edges of length \( L_o = n_V (n + 1) \) and \( 20 n_V^2 \) faces. As seen in table I, for \( n = 0 \) we studied systems up to 8412 monomers corresponding to \( n_V = 29 \), about twice as large as that considered by Komura and Baumgärtner [32], though their closed vesicles did not have the icosahedron symmetry. Our largest system, \( N = 25002 \), \( n = 8 \) is comparable to the largest open membrane with free perimeter studied by Abraham [12]. Typically, \( 0.5 \times 1.0 \times 10^6 \) time steps were needed to equilibrate the system. This equilibration was checked by monitoring the autocorrelation function of the mean square radius of gyration as discussed in section 4.

As mentioned in the introduction, the average distance between nodes in the RBC skeleton is about \( 1/4 - 1/5 \) of its fully extended length. With only excluded volume interactions as studied here, the average distance between nodes for \( n \geq 8 \) approximately satisfies the relation \( \langle R_o^2 \rangle = a (n + 1)^{2 \nu} \), where \( \nu = 0.59 \) is the excluded volume exponent and \( a \approx 1.8 \), though the value of \( a \) for a free chain under the same conditions is somewhat smaller, approximately 1.5. Thus \( n \approx 50 - 60 \) for \( \langle R_o \rangle /(n + 1) \approx 1/4 \). An alternative mapping based on comparing the persistence length for the spectrin tetramers with that of the bead-spring model would suggest a small value of \( n \). However, from our experience on polymer melts, comparing the persistence length is not as meaningful since there is some arbitrariness in the definition of a statistical segment. Due to the slow equilibration of these membranes, the largest sample we can handle at present is the order of 25 000 monomers. Therefore, we limited \( n \) to 16, for which \( \langle R_o \rangle /(n + 1) = 0.43 \), in order to have a reasonable number of triangles in our system.

To analyze our results we calculated several quantities. Every 500 steps the inertia matrix, its eigenvalues \( \lambda_i \) (\( \lambda_1 < \lambda_2 < \lambda_3 \)) and the radius of gyration squared, \( R_g^2 \), given by their sum were calculated. These quantities were also used to check the equilibration of the system through their autocorrelation. For closed membranes in the flat phase, all three eigenvalues should scale as \( N \). Results for total duration of the run, \( \langle R_g^2 \rangle \) and \( \langle h^2 \rangle \) are given in table I. If we treat the data saved every 500 time steps as statistically independent, then the error in
Table I. — The number of monomers per edge \( n_V \) in the original icosahedron, number of monomers \( n \) between each node, total number of monomers \( N \) and total duration of the run, \( T_t/\tau \), after equilibration for the closed vesicles studied here. Also shown is the average mean square radius of gyration \( \langle R_g^2 \rangle \) and the mean square height fluctuation \( \langle h^2 \rangle \) for each membrane.

<table>
<thead>
<tr>
<th>( n_V )</th>
<th>( n )</th>
<th>( N )</th>
<th>( T_t/\tau )</th>
<th>( \langle R_g^2 \rangle )</th>
<th>( \langle h^2 \rangle )</th>
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<td>0.36</td>
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<tr>
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<td>12000</td>
<td>114.1</td>
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<tr>
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<td>232.9</td>
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<tr>
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<td>489.6</td>
<td>1.15</td>
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<tr>
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<td>4</td>
<td>2082</td>
<td>18000</td>
<td>106.9</td>
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<tr>
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<td>130.5</td>
<td>3.80</td>
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<td>12252</td>
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</table>

These two quantities as well as the moments of inertia are much less than 1%. However, this surely underestimates the error as the configurations are surely not independent. Averaging subsets of the data suggests that a reasonable estimate of the statistical error is about 1% for \( \langle R_g^2 \rangle \) and about 2-3% for \( \langle h^2 \rangle \). We also measured the spherically averaged structure factor, \( S(q) \), given by

\[
S(q) = \frac{1}{N} \langle \sum_{i,j} e^{i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)} \rangle. \tag{4}
\]

The angle brackets represent a configurational average typically taken every 5000 time steps. For each \( q = |\mathbf{q}| \), we averaged over 20 random orientations.

3. Results.

Figure 3 shows typical results for three vesicles with approximately the same number of monomers for \( n = 0, 8 \) and 16. For \( n = 0 \), the membrane retains the shape characteristic of an icosahedron. This system has a relatively large local bending rigidity due to the high connectivity of each monomer. The magnitude of the height fluctuations is relatively small as seen in table I. Only for much larger systems would one expect this system to look less faceted. For \( n = 8 \) and 16, however, the local bending rigidity is significantly reduced and there is little evidence of the icosahedra topology even for systems as small as a few thousand monomers. In figure 4 we show a typical configuration for 16002 and compare its cross-sections through the center with RBC skeletons in high salt. Note the similarity between the RBC skeletons and the simulated membranes even though the simulated membranes are significantly smaller.
Fig. 3. — Typical configuration for closed self-avoiding tethered membranes for: a) $N = 4002$ ($n = 0$); b) $N = 4002$ ($n = 8$) and c) $N = 4412$ ($n = 16$).

Fig. 4. — a) Isolated red blood cell skeletons in 2.5$nM$ monovalent salt viewed under video enhanced differential interference contrast microscopy. The mean diameter is $5.3 \pm 0.4 \mu m$ [28]. Typical configuration for the membrane of size $N = 16002$ ($n = 8$) shown for b) entire network and c) three cross sections through the center of the network. Note similarity to shape of the experimental and simulated system.
The mean squared radius of gyration $\langle R_g^2 \rangle$ versus $N$ is shown in figure 5 for the membranes studied. Least squared fits to the data for each $n$ show that $\langle R_g^2 \rangle \sim n_V^{2\nu_3}$ with $\nu_3 = 0.99 \pm 0.02$ for $n = 0$, $0.95 \pm 0.03$ for $n = 8$ and $0.89 \pm 0.04$ for $n = 16$. All of these results are consistent with a flat phase as one expects from previous simulations on open membranes as well as Komura and Baumg"artner's simulations [32] for closed membranes with $n = 0$. One interesting feature of this data is that to first approximation $\langle R_g^2 \rangle$ depends on $n$ and $n_V$ only through $N \sim n_V n$. This can be understood if one considers the elementary triangles that make up the membrane. Since the distance between nodes $\langle R_e \rangle \sim (n + 1)^\nu$, the surface area per triangle scales as $n^{2\nu}$. Since the number of triangles is $20n_V^2$ and the membrane is approximately spherical, one would expect that $\langle R_g^2 \rangle$ would scale as the surface area, which is proportional to $n_V^2 R_e^2 \sim n_V^2 n^{1.18} \sim N n^{0.18}$. Therefore, the dominant contribution to the size of the membrane is due to $N$, the total number of monomers. The number of monomers between each node enters only weakly via a power $n^{0.18}$ for fixed $N$. Similar argument at the theta point where $R_e^2 \sim N$ suggests that $\langle R_g^2 \rangle$ would depend only on $N$ and would be essentially independent of the number of monomers between each node for membranes of the topology studied here.

![Fig. 5. — Mean square radius of gyration $\langle R_g^2 \rangle$ versus $N$ for closed self-avoiding membranes with $n = 0$ (o), 4 (x), 8 (o) and 16 (Δ).](image)

In figure 6, results for the eigenvalues of the moment of inertia tensor $\lambda_i$ versus $N$ are presented for $n = 0$ and 8. Least square fits to the data suggest that all of these eigenvalues satisfy a scaling relaxation $\lambda_i \sim N^{\nu_i}$. For $n = 0$, the three values of $\nu_i$ are essentially 1, with the ratio $\lambda_1/\lambda_3$ increasing slightly with increasing $N$. However, for $n = 8$, the effective exponents $\nu_i$ are very different and the ratio $\lambda_1/\lambda_3$ increases slightly as $N$ increases. Best fits to the data give $\nu_1 = 0.99$, $\nu_2 = 0.95$ and $\nu_3 = 0.91$ for $n = 8$. At first these results seem a little surprising until one notes that in the limit of large $N$, all three eigenvalues must be the same. Differences in the $\lambda_i$'s for finite $N$ are a result of fluctuations in the height $h$ which vanish in comparison to the radius of the membrane as $N \to \infty$ since $\zeta < 1$. This can be seen very simply by considering the energy cost to produce a fluctuation in the membrane for large
Fig. 6. — Eigenvalues of the moment of inertia tensor $\lambda_1$ (a), $\lambda_2$ (Δ) and $\lambda_3$ (x) versus $N$ for (a) $n = 0$ and (b) $n = 8$.

$N$. Excursions from the mean radius $R_0$ of the membrane can be written as

$$R = R_0 + n P_l(\Omega)$$

(5)

where $P_l(\Omega)$ is the Legendre polynomial of order $l$. The lowest order excitation which dominates $\langle h^2 \rangle$ is for $l = 2$ and the energy of the excitation has the form

$$E \sim \int dA (\nabla^2 h)^2$$

(6)

which has the scaling form

$$E = \kappa (h/R)^2.$$ 

(7)

Here $\kappa$ is the renormalized bending rigidity which scales as $R^\eta$, where $\eta = 2 - 2\zeta$ for $D = 2$. Therefore, for $E$ of order $k_B T$, $\langle h^2 / R^2 \rangle \sim R^{-\eta}$ which vanishes as $R \to \infty$. The disparity in the values of $\nu_i$ is due to finite size corrections. Thus the ratio $\lambda_1 / \lambda_3$ must increase with $N$.
Fig. 7. — Height fluctuations $\langle h^2 \rangle$ versus $N$ for $n = 0$ and 8.

giving rise to values of $\lambda_i$ which have different effective exponent for the range of $N$ studied here. A similar argument also applies to the largest 2 eigenvalues for open membranes which also must be equal in the asymptotic limit. This point has apparently been overlooked in a number of previous simulations, apparently because for membranes with no holes ($n = 0$), the dependence of $\lambda_1/\lambda_3$ on $N$ is very weak. Only for perforated membranes as those studied here is this correction to scaling more apparent.

As discussed in the introduction, the actual value for the exponent $\zeta$ is not well determined. For open membranes, an issue has been raised as to whether the free perimeter can give rise to finite size corrections which lead to an overestimation of $\zeta$ for the system size studied to date. Abraham [22] used pbc and a constant pressure simulation to eliminate the free perimeter. His simulations gave a significantly lower estimate for $\zeta$. An alternative method for eliminating the free perimeter is to study closed vesicles as we have done here. In this case, the height fluctuations $\langle h^2 \rangle$ can be easily measured from the fluctuations of the mean squared radius of gyration,

$$\langle h^2 \rangle \sim [(\delta R_g^2)^2]^{1/2} \equiv [(R_g^4) - \langle R_g^2 \rangle^2]^{1/2}$$

In figure 7, we present results for $\langle h^2 \rangle$ versus $N$ for $n = 0$ and 8. We find $\zeta = 0.55 \pm 0.02$ for $n = 0$ and $\zeta = 0.56 \pm 0.02$ for $n = 8$, only slightly larger than Abrahams [22] results for membranes with pbc $\zeta = 0.53 \pm 0.03$. Our results are significantly lower than those observed for open membranes with free perimeters, $\zeta = 0.64 \pm 0.03$, as well as earlier results of Komura and Baumgärtner [32] for a closed vesicle with $n = 0$, $\zeta = 0.645 \pm 0.08$. Our results are close to those obtained from a self-consistent screening approximation by Le Doussal and Radzihovsky [23] who find $\zeta = 0.59$.

One of the original motivations for studying large membranes with holes was to investigate whether reducing the local binding rigidity would cause the membranes to crumple. Abraham [12] showed very conclusively in his study of perforated open membranes that the order parameter [33], defined as the ratio of the the mean square radius of gyration over the membrane size, vanishes only as the mass fraction of the perforated membrane vanishes. In figure 8, we present similar evidence for our closed membrane. Here we plot $\langle R_g^2 \rangle/L_o^2$, where $L_o = n_V(n + 1)$ is the length of an edge in the initial configuration of the membrane and is a measure of the
membrane's size. In agreement with Abraham, we find that the membranes remain flat and do not crumple, even when the holes are quite large and the mass fraction is quite low.

Experimentally x-ray and light scattering have been used to study randomly oriented membranes [24, 27, 28]. In figure 9, we present our results for the scaled \( S(q)/N \) for \( n = 8 \). Unlike open membranes, there is extra structure in \( S(q) \) because the membrane is closed. Since the vesicles are to first approximation spherical shells of diameter \( 2R \sim N^{1/2} \), it is not surprising that the first few minima scale as \( qN^{1/2} \). These oscillations are more pronounced than one finds in the experimental data due both to the ideal monodispersity and relatively low value of \( n \). For \( n = 0 \), these oscillations persist over the entire \( q \) range. However, for \( n = 8 \), there is also a high \( q \) regime which scales as \( qN^{6/2} \) with \( \delta \approx 0.85 \pm 0.05 \), consistent with the isotropically averaged \( S(q) \) for open membranes [25, 26]. Because of the oscillations due to the monodispersity of the sample at small \( q \), this scaling regime is relatively small and extends only from \( 3 < \ln qN^{5/2} < 5 \). This value of \( \delta \) corresponding to roughness exponent \( \zeta = 3 - 2/\delta \approx 0.65 \pm 0.05 \). While this value is slightly larger, it is not particular reliable compared to that determined directly from the height fluctuations, due to the limited scaling regime in \( S(q) \).

4. Dynamics.

To determine if the membranes studied have equilibrated, it is useful to measure the time dependent correlation function for the mean square radius of gyration,

\[
\phi(t) = \frac{\langle R_g^2(t)R_g^2(0) \rangle - \langle R_g^2 \rangle^2}{\langle R_g^2 \rangle - \langle R_g^2 \rangle^2}
\]  

(9)

Since the present simulations do not include solvent molecules explicitly and the membranes are weakly coupled to a heat bath, the membranes should have a Rouse-like relaxation. Kantor et al. [3] suggested, following the analysis for a linear polymer chain [34], that the longest relaxation time \( \tau_R \sim N^{1+\nu} \). For a flat membrane, \( \nu = 1 \) and the analogy would predict that \( \tau_R \sim N^2 \). However as seen in figure 10, our data scale as \( t/N \), not as \( t/N^2 \). This is in agreement with earlier data of Komura and Baumgärtner for closed vesicles with \( n = 0 \). They suggested one possible explanation for this faster relaxation observed is that for flat membranes, the relaxation has both an in-plane and an out-of-plane component. The in-plane longitudinal relaxation time scales as \( N \), similar to the case of permanent crosslinked networks [35], whereas the out-of-plane relaxation time scales as \( N^{1+\zeta} \). This explanation is consistent with both our data and that of Komura and Baumgärtner if one assumes that the amplitude of the in-plane contribution to \( \phi(t) \) is much larger than the out-of-plane amplitude. Unfortunately, neither our data nor theirs is accurate enough to tell whether this second relaxation mode is present.

We find that the longest relaxation time \( \tau_R \) depends strongly on both the system size \( N \) and the number of monomers \( n \) between each node, unlike \( \langle R_g^2 \rangle \). This result can be understood by recalling that for a linear polymer in a good solvent, the longest relaxation time scales as \( n^{1+2\nu} \), where \( \nu = 0.59 \) [34]. Therefore, if we assume that \( \tau_R \) scales as \( N \sim n^2 \), \( \tau_R \sim Nn^{1.18} \). This extra factor of \( n^{1.18} \) accounts, within the accuracy of our data, for the difference in \( \phi(t) \) we observe for \( 4 \leq n \leq 16 \).
Fig. 8. — Mean square radius of gyration $\langle R_g^2 \rangle / L_0^2$ versus mass fraction for $0 \leq n \leq 16$. Here, $L_0 = n\nu(n + 1)$ is length of each edge in the initial icosahedra configuration.

Fig. 9. — Scaling plots for isotropically averaged $S(q)$ for the 5 closed self-avoiding membranes with $n = 8$. 
5. Conclusions.

In this paper we have presented simulation results for closed tethered membranes. To model the red blood cell skeleton, the vesicles are constructed by connecting the ends of a linear chain of \( n \) monomers into a predominately triangular array which is closed in the shape of an icosahedra. For all \( n \) studied, the membranes are flat and the mean square radius of gyration of the membranes grows as \( N \) as expected from previous simulations on open membranes. However the introduction of holes in the membranes give rise to a local flexibility. For membranes without hole, \( n = 0 \), for the sizes studied here \( (N \leq 8412) \), the equilibrated membranes retain the shape characteristic of an icosahedra due to the relatively large local bending rigidity which results from the excluded volume interactions. For \( n \) as small as 8, the local flexibility of the membrane is significantly enhanced and the membranes appear more rounded. Cross sections through the center of the simulated membranes are very similar to red blood cell skeletons, even though the membranes studied here are considerably smaller.

It is now clear from the large number of simulations on both open and closed tethered membranes, that in the presence of only excluded volume interactions alone membranes do not crumple but remain flat. This is presumably due to an implicit bending rigidity which is induced by the self-avoidance requirement. The open question remains: does the crumpled phase exist? One of us (G. S. G.) has showed that in higher embedding dimension \( (d > 4) \) that tethered membranes do crumple even when only excluded volume interactions are present. It now appears that attractive interactions may play a role in overcoming this implicit bending rigidity induced by the self-avoidance requirement. In \( d = 3 \), Liu and Plischke [19] have shown that the introduction of a long range attractive interaction appears to produce an intermediate temperature regime in which the membrane has a fractal dimension \( d_\ell \sim 2.5 \). Their simulations were carried out for open membranes with no holes \( (n = 0) \) for fairly small systems. It is clearly important to carry more detailed simulations, on larger systems, to study the transition between the high temperature flat phase and the intermediate crumpled phase, should it exist. Since the membranes studied here have no free perimeter and will not be able to fold [36] as open membranes can, they appear to be good candidates for investigating the effect of longer range attractive interactions. We plan to carry out such a study in the future.
Note added in proof: Zhang, Davies and Kroll [37] have recently shown that for closed membranes, there is a linear coupling between the out-of-plane undulation modes and the in-plane phonon modes, which suppresses the out-of-plane fluctuations at long length scales. This leads to a new scaling relation for what we identified as the height fluctuations $<h^2>$ in equation (8). As defined in equation (8), Zhang et al. point out that $<h^2> \sim N^{\zeta_1}$, where $\zeta_1 = (2 - \eta/2)/(2 + \eta)$, instead of $N^\zeta$, with $\zeta = (1 - \eta/2)$. Reinterpreting our results in terms of $\zeta_1$ gives $\eta = 0.85 \pm 0.04$ and $\zeta = 0.58 \pm 0.02$, which agrees with the theoretical prediction of Le Doussal and Radzihovsky [23].

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References


