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The conformation and scaling properties of self-avoiding fluid membranes with an extrinsic bending rigidity \( \kappa \) were studied with the use of Monte Carlo methods. For \( \kappa = 0 \), the results are consistent with branched polymer behavior at large length scales. There is a smooth crossover from a crumpled to an extended state with increasing \( \kappa \), with a peak in the specific heat when the persistence length reaches the system size. The scale-dependent effective bending rigidity is a decreasing function of system size for all bare rigidities. These results indicate that fluid membranes are always crumpled at sufficiently long length scales.

Membranes composed of amphiphilic molecules, such as the monolayers of surfactant molecules at oil-water interfaces in microemulsions, the lipid bilayers that form biological membranes, as well as the layers of surfactant molecules in recently studied lyotropic liquid crystals, are highly flexible (nearly tensionless) surfaces. Membranes play a central role in determining the architecture of biological systems and provide the basic structural element for complex fluids such as microemulsions; an understanding of the statistical mechanics of these self-avoiding surfaces is therefore of considerable importance (1, 2).

In most cases of interest, these membranes are fluid, which means that the molecules can diffuse rapidly within the membrane surface and possess no reference lattice. In the absence of a lateral tension (3, 4), the shape of the membrane is governed by its bending rigidity \( \kappa \). A membrane of linear size \( L \) exhibits transverse fluctuations (5) of extension \( L_x = (kT/\kappa)^{1/2}L \) (where \( k \) is Boltzmann's constant and \( T \) is temperature) on length scales small compared to the persistence length (6) \( \xi_p = \alpha \exp(\kappa/kT) \), where \( \alpha \) is a short-distance cutoff and \( \epsilon = 4\pi/3 \). However, as \( L \) approaches \( \xi_p \), shape fluctuations have been predicted (7, 8) to reduce the bending rigidity and lead to a renormalized rigidity \( \kappa_r = \kappa - (kT/\epsilon) \ln(L/\alpha) \) in the limit of small \( kT/\kappa \). On length scales \( L = 1 \), the membrane should have an effective bending rigidity of the order of \( kT \). At larger length scales \( (L >> \xi_p) \), membranes fluctuating at constant area are expected to have an extremely small bending rigidity and behave as crumpled (9) objects characterized by the absence of long-range orientational order of normals erected perpendicular to the local surface elements. For a fixed topology, the scaling behavior of self-avoiding fluid membranes at these length scales is expected to be the same as that of a branched polymer (2, 10, 11).

Several aspects of this scenario have, however, yet to be verified. Indeed, there is little evidence that self-avoiding fluid membranes with a finite bare bending rigidity really do crumple. It has been speculated (2) that self-avoidance may stabilize the effective bending rigidity at some finite value \( \kappa \sim kT \) and therefore prevent crumpling. Furthermore, there is some controversy concerning the universality classes of the various models used to describe self-avoiding random surfaces (with \( \kappa = 0 \)). The most widely studied models for these surfaces are constructed by taking the elementary 2-cells (plaquettes) on a hypercubic lattice and gluing them together in such a way that each edge is shared by exactly two plaquettes (10). For fixed topology, the long-length-scale behavior of this class of surface has been shown to be that of a branched polymer: the radius of gyration \( R_g \) of a surface of area \( S \) scales as \( R_g \sim S^{\nu} \) (12), with \( \nu = 1 \) in spatial dimension \( d = 3 \) (10). Recently, however, randomly triangulated surfaces of the type we consider here have been investigated for \( \kappa = 0 \) with the use of Monte Carlo techniques (13). A value \( \nu \approx 0.8 \) was reported, implying that such surfaces belong to a different universality class. Given the generality of the entropic mechanism behind branched polymer behavior, this discrepancy is troubling (10, 11).

In this report we present evidence that fluid membranes are crumpled at sufficiently large length scales for any value of the bare bending rigidity and that the crumpled state does indeed exhibit branched polymer behavior. Our conclusions are based on exten-

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The continuum limit of the bending energy is just Helfrich's curvature elastic energy where \( dS \) denotes the surface element and \( c_1 \) is the principal curvatures. The probabilistic decision whether to accept the move is made by comparing the initial and final energies of the system. We chose \( s \) so that \( \approx 1 \) of the updating attempts were accepted. Second, we attempt to flip \( N \) randomly chosen bonds. A bond flip consists of deleting a tether and constructing a new one between the two previously unconnected vertices of the two adjacent triangles. The flip is accepted with the probability given by the Boltzmann factor if all vertices have a minimum of three neighbors. Note that this procedure does not flip boundary tethers, so that the open membranes we simulate have perimeters of constant length. A more detailed discussion of the bond-flipping procedure can be found in (13-17).

In order to insure reparametrization invariance we use a discretized version of the invariant measure \( \delta D[\vec{r}] = \prod_i \delta dr(\xi_1,\xi_2,\xi_3) \) in our simulations, where \( r(\xi) \) is the three-dimensional coordinate vector of a point on the surface with internal coordinate \( \xi \) and \( g \) is determinant of the metric tensor (21). We also choose a metric in which all internal lengths are equal, that is, all triangles on the surface are equilateral and of area \( \text{constant} \). The volume \( \omega_i \) of the dual image of vertex \( i \) (which is the discrete analog of the invariant volume \( f d^3 \xi \sqrt{g} \)) is proportional to the coordination number \( q_i \) of this vertex: \( \omega_i = q_i / 3 \). On the triangulated surface the curvature is concentrated at the vertices; with the present normalization the scalar curvature \( R = \pi (6 - q_i) / q_i \) and the discretization of the measure is \( \Omega D[\vec{r}] = \omega_i^{1/2} \) (14-17).

Simulations were performed with both \( \epsilon_o = \sqrt{2} / 0.0 \) and \( \sqrt{2.8} \). In the former case we chose \( s = 0.1 \) and in the latter \( s = 0.15 \). This choice of parameters ensures self-avoidance when updating the monomer coordinates. However, there is only weak self-avoidance during bond flipping for \( \epsilon_o = \sqrt{2.8} \). Indeed, it is easy to show that implementing complete self-avoidance would require taking \( \epsilon_o \) at least less than \( \sqrt{8.3} \) in this case. We have in fact observed vesicle inversion for \( \epsilon_o = \sqrt{2.8} \) when \( \lambda = 0 \). Nevertheless, data for the mean-squared radius of gyration \( R^2 \) for zero bending energy indicate that this does not influence the asymptotic fractal dimension of the crumpled phase. Furthermore, it does not qualitatively affect the behavior for finite \( \lambda \). The primary advantage of using a larger tethering length is that the diffusivity of the monomers in the membrane surface is substantially enhanced: the bond-flip acceptance rate is a factor of 2 greater for \( \epsilon_o = \sqrt{2.8} \) than for \( \sqrt{2.0} \) when \( \lambda = 0 \); the effect is greater for larger values of \( \lambda \).

In Fig. 1 we plot data for \( R^2 \) versus the number of monomers for the case of zero bending rigidity, \( \lambda = 0 \). Averages were taken over \( 1 \times 10^4 \) to \( 2 \times 10^3 \) Monte Carlo steps per monomer. Data are shown both for vesicles with \( \epsilon_o = \sqrt{2.8} \) and planar membranes with \( \epsilon_o = \sqrt{2.0} \). For both topologies, the data are consistent with \( R^2 \) scaling with \( N \) (12) with \( v = 1 \). Furthermore, typical configurations, such as the one illustrated in Fig. 2, indicate a "branched polymer" structure in which there are long arms extending out in various directions. This result suggests an entropic mechanism that favors treelike ramified objects, which is in agreement with results obtained for random surfaces constructed of elementary plaquettes on a cubic lattice (10). For vesicles, our results are consistent with \( S \sim R^2 \) and \( V \sim R^2 \) (where \( S \) and \( V \) are the surface area and...
The renormalized bending rigidity can be expressed in terms of the mean curvature susceptibility as 
\[
\langle \kappa \rangle / \kappa_{\text{eff}} = \int d^d \xi \sqrt{g(\xi)} \left[ d^d \xi \right]^{(d-1)/2d}
\]

where \(\langle \cdot \rangle\) is the average membrane area, \(\sqrt{g(\xi)}\), denotes the cumulant average, and \(H(\xi)\) is the mean curvature at \(\xi\). In the continuum limit, the extrinsic curvature tensor is proportional to the unit normal vector. The discretization (23) of \(f d^d \xi \sqrt{g(\xi)} H(\xi)\) we use is therefore \(\sum_i [\Sigma_{ij}(\sigma_{ij})^j](r_i - r_j)\), where the \(i\)-sum runs over monomers, the \(j\)-sum runs over the neighbors of \(i\), and \(z_i\) is the sign of \(\delta_i \Sigma_{ij}(r_i - r_j)\), with \(\delta_i\) the surface normal at monomer \(i\) obtained by averaging over the normals of all triangles that have \(i\) as a corner, \(l_p\) is the distance between the two neighboring monomers \(i\) and \(j\), and \(\sigma_{ij}\) is the length of a bond in the dual lattice. For the metric we have chosen, \(\sigma_{ij}/l_p = 1/\sqrt{3}\) so that the right-hand side of Eq. 4.1 becomes

\[
\frac{1}{3} \sum_{i} \left| \sum_{j} (r_i - r_j) \right| \sum_{i} \left| \sum_{k} (r_k - r_m) \right|.
\]

For vesicles, the amplitude of the average in Eq. 4.1 depends strongly on the size of the region over which one averages. In particular, in the continuum limit, one finds that \(\kappa_{\text{eff}} = \kappa\) when averaging over one-half of a sphere when fluctuations are ignored. Our results for \(\kappa = \kappa_{\text{eff}}\) obtained by averaging over \(N = (N-1)/2\) contiguous sites on the vesicle surface, are shown in Fig. 3B (24); \(\kappa_{\text{eff}}\) is a decreasing function of \(N\) for all \(\lambda\). There does not appear to be any qualitative difference in the behavior of \(\kappa_{\text{eff}}\) for large and small \(\lambda\).

We have not attempted to make a quantitative comparison with theory in the large \(\lambda\) regime, because the expected logarithmic renormalization of \(\kappa\) is completely masked by averaging over a finite portion of the membrane surface (25). Nevertheless, the essential qualitative feature that \(\kappa_{\text{eff}}\) is a decreasing function of length scale in both the large and small \(\lambda\) regimes supports the interpretation that there is no phase transition as a function of \(\lambda\).

Self-avoiding fluid membranes are therefore very likely always crumpled at large length scales, independent of the value of the microscopic bending rigidity \(\kappa\). Furthermore, the crumpled state of the model we considered is a treelike ramified object characterized by \(R_s^2 \sim N\), in agreement with results obtained for hypercubic plaquette models (10) of random surfaces. A more detailed analysis, however, is required to determine the quantitative scale dependence of the renormalized bending rigidity.

Fluid membranes with a wide range of bare bending rigidities have been studied experimentally in considerable detail recently. For phospholipid bilayers at room temperature, \(\kappa/kT = 10\) to 20 [see (26-28), which contain many additional references], so that the persistence length is usually much greater than the size of the membranes and the reduction of the bending rigidity by shape fluctuations is negligible. An exceptionally small value for \(\kappa_{\text{eff}}\) however, is obtained for lecithin membranes containing a small amount of bipolar lipid (26); this may also be the case for surfactant bilayers that form lamellar and bicontinuous phases (29). These systems are therefore well suited for experimental studies of flexible fluid membranes.

We have shown that simulations performed using simple string-and-bead models for randomly triangulated surfaces (with a wide range of finite bare bending rigidities) can yield detailed information on both the large- and small-scale conformations as well as the scale dependence of elastic constants such as the bending rigidity. Further numerical studies of this model and its generalizations should permit a more detailed comparison with experiment and could provide new insights into many phenomena involving membranes and other self-assembling systems.
18. This result can be obtained by covering a sphere with equilateral triangles of side $\ell$ and taking the limit $\ell \to 0$. A similar procedure was followed in (19) for a cylindrical surface; in that case it was found that $\kappa = V \sqrt{3}/2$.
20. Unlike (13) (in which the maximum number of nearest-neighbor bonds was limited to eight), we do not explicitly restrict the maximum number of neighbors. We find, for example, that the average maximum connectivity of a monomer is $-12$ for the largest systems studied (for $\lambda = 0$). We have checked, however, that restricting the maximum number of nearest neighbors as in (13) does not prevent the observed collapse to branched polymer behavior.
24. In evaluating $\bar{z}$ we have used $\langle z \rangle = 2N, 1 - 2N \rangle$ ($\bar{z}$), with $\langle z \rangle > 0.78$, where $\langle z \rangle$ is the average area of an elementary triangle.
25. See (22) for a partial treatment of this problem for tethered membranes.
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