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Floppy Tethered Networks

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Abstract. — A model for extremely flexible tethered membranes is studied by Monte Carlo simulations and scaling arguments. In contrast to the standard string-and-bead models, no finite-range hard-core repulsion is used to ensure self-avoidance. Instead, the elementary triangles are taken to be impenetrable. Although this leads to an extremely floppy tethered network, the surface is found to be asymptotically flat, with a roughness exponent ζ ≈ 0.7, consistent with the result of self-avoiding string-and-bead models. The orientationally averaged scattering intensity, on the other hand, is found to exhibit a nontrivial scaling behavior characteristic of a crumpled object with an effective fractal dimension df ≈ 2.7. This result is compared with recent experiments on graphite oxide sheets.

1. Introduction.

Tethered surfaces are idealized models of flexible two-dimensional solid sheets which are free to fluctuate in three dimensional space [1, 2]. These surfaces have a finite shear modulus so that the statistical mechanics of tethered networks is controlled by the delicate interplay between the in-plane elastic modes and the out-of-plane undulation modes [3, 4]. Two realizations of these objects which have recently attracted a great deal of attention are exfoliated graphite oxide crystals [5] and the spectrin network of red blood cells [6]. The experiments on graphite oxide membranes [5] suggest that their conformational structure is crumpled, with a fractal dimension df ≈ 2.5. Computer simulations of self-avoiding tethered surfaces, on the other hand, find flat [7-9], compact [10], and folded structures [11], but, with two possible exceptions [12-14], not the crumpled behavior observed in experiment.

Current experimental studies of tethered networks measure the directionally averaged structure factor S(q). In an isotropic crumpled phase, in which the radius of gyration scales like \( R_g \sim L^\nu \), where \( L \) is the (internal) linear dimension of the object, the directionally averaged structure function has the scaling form \( S(q,L) = \Psi(qR_g) \) [5, 10, 15]. For \( q < 2\pi/R_g \), \( S(q) \approx 1 - (qR_g)^2/3 + O(q^4) \), while for \( 2\pi/a > q > 2\pi/R_g \), where \( a \) is on the order of the...
intrinsic thickness of the membrane, one expects

\[ S(q, L) \approx (qR_g)^{-d_f}, \]

with \( d_f = 2/\nu \).

In the experiments described in reference [5] the presence of a scaling region consistent with (1) was interpreted to mean that the membrane was fractal (at the length scales studied). In contrast, for an anisotropic, flat membrane, for which the eigenvalues of the moment of inertia tensor scale as \( \lambda_3 \sim \lambda_2 \sim L^2 \) and \( \lambda_1 \sim L^{2\zeta} \), with \( \zeta < 1 \), one expects \( S(q) \sim q^{-2} \) for \( q \ll 1/\ell \), where \( \ell \) depends on the amplitude of the in-plane and out-of-plane fluctuations [16]. For \( 1/\ell \ll q \ll 1/a \), on the other hand, it has been shown that either \( S(q) \sim q^{-3r} \) (if in-plane fluctuations are either absent or of small amplitude), or \( S(q) \sim q^{-4r} \) (in the case of large in-plane fluctuations) [16]. Since recent simulation studies of tethered networks with free edge boundary conditions indicate that \( \zeta \approx 0.65 \), there should be a crossover from \( q^{-2} \) to either \( q^{-2.35} \) or \( q^{-4.15} \) behavior with increasing \( q \), in disagreement with the experimental result [5] discussed above.

With the exception of the results presented in references [13 and 14], recent simulation studies of two-dimensional self-avoiding tethered networks (without attractive interparticle interactions) all indicate that the membrane is in the anisotropic, flat state described in the last paragraph. Furthermore, it is not clear at present if and when the presence of attractive interactions can lead to a crumpled phase. Abraham and Kardar [11] studied tethered networks in which non-nearest neighbor particles interact through a Lennard-Jones potential truncated at \( 2.5\sigma \) (where \( \sigma \) is the interatomic separation at which the potential goes through zero). Instead of crumpling, they found that the membrane goes through a sequence of "folding" transitions with decreasing temperature which terminate ultimately in a symmetric collapsed state characterized by \( d_f = 3 \). Quite recently, however, Liu and Plischke [12] claim to find a crumpled state characterized by \( d_f \approx 2.5 \) for a range of temperatures for a model tethered membrane in which the particles interact through hard-core repulsion as well as a longer-range attractive square-well potential. More work is needed to clarify which of these scenarios actually occur for a given interaction potential.

In this paper we present results for the conformation and scaling behavior of two-dimensional tethered networks obtained using a model introduced in references [13 and 14] for extremely flexible triangulated surfaces. In this model, self-avoidance is guaranteed by requiring that the elementary surface triangles do not intersect. In this way it is possible to avoid the use of finite-range repulsive potentials between vertices; this results in a very flexible surface which can fold in on itself without any cost in energy. The resulting surface is therefore much rougher than the commonly studied string-and-bead models. Nevertheless, we show (in disagreement with references [13 and 14]) that the surface is asymptotically flat. The directionally averaged structure factor, on the other hand, exhibits a nontrivial scaling behavior reminiscent of (1), with an effective fractal dimension \( d_f \approx 2.7 \). The outline of this paper is as follows. In section 2 we describe the model and simulation procedure. Section 3 contains an analysis of the scaling behavior of the eigenvalues of the moment of inertia tensor as well as the structure factor \( S(q) \). Results are also presented and compared with theory for the behavior of the smallest eigenvalue of the moment of inertia tensor (which measures the width of the surface) averaged over subdomains of increasing diameter. We show that although the network is asymptotically flat, with \( \zeta \approx 0.7 \), the directionally averaged \( S(q) \) scales as \( q^{-2.7} \) over the whole scaling region \( 2\pi/R_g < q < 2\pi/a \), at least for the system sizes we were able to study. The paper closes with a brief discussion in section 4.

The model we consider consists of an open triangular network containing \( N \) vertices with free edge boundary conditions. The network is hexagonal in shape, with a diameter \( L \), so that \( N = (3L^2 + 1)/4 \). The vertices are connected by tethers of maximum extension \( \ell_0 = \sqrt{2} \). This is the only length scale in the problem. Our Monte Carlo procedure consists of sequentially updating the vertex positions by a random increment in the cube \([-\delta, \delta]^3\). In contrast to the commonly used string-and-bead models for tethered surfaces, we do not utilize a finite-range hard-core repulsion of vertex monomers to ensure self-avoidance. Rather, an update is accepted if no vector connecting tethered vertices intersects an elementary triangle of the surface. This guarantees that there is no intersection of elementary surface triangles. In this way, the model resembles that studied in references [13 and 14]. Note, however, that we do not impose any constraint on the minimum separation between vertices. The resulting network is therefore extremely "floppy" in that it can fold back on itself with negligible excluded volume effects. Because of this we have found that the models sweeps out phase space very quickly, so that, for a given system size, relaxation times are typically significantly shorter than for the standard string-and-bead models [13]. An advantage of the model we employ compared to the giant-hole membranes studied in reference [17] is the large reduction in the number of degrees of freedom for a given number of vertices. In our simulations, we have studied membranes sizes from \( L = 7 \) (\( N = 37 \)) to \( L = 25 \) (\( N = 469 \)). Averages are taken typically over \( 10^6 \) Monte Carlo steps per vertex (MCS), which, because of the short relaxation times [13], is sufficient to give good statistical accuracy.

3. Results.

One of the simplest ways to characterize the conformation of the tethered network is to examine the eigenvalues \( \lambda_1 \leq \lambda_2 \leq \lambda_3 \) of the moment of inertia tensor

\[
\mathbf{T}_{\alpha,\beta} = \frac{1}{N} \sum_i (r_{i\alpha} r_{i\beta} - \bar{r}_\alpha \bar{r}_\beta),
\]

where \( \alpha, \beta \in \{x, y, z\} \), and the sum runs over all vertices of a given configuration; \( \bar{r}_\alpha \) is the \( \alpha \) component of the center of mass for that configuration. In the flat phase, the average eigenvalues are expected to scale as \( \langle \lambda_1 \rangle \sim N^5 \), and \( \langle \lambda_3 \rangle \sim \langle \lambda_2 \rangle \sim N \) [7]. Our results for \( \langle \lambda_1 \rangle \) and \( \langle R_g^2 \rangle \sim \sum_j < \lambda_j > \) as a function of the system size \( N \) are plotted in figure 1. The solid line is a plot of \( \langle R_g^2 \rangle \sim N^{0.87} \), and the dashed line corresponds to \( \langle \lambda_1 \rangle \sim N^{0.63} \), or \( \zeta = 0.63 \). Both \( \langle \lambda_2 \rangle \) and \( \langle \lambda_3 \rangle \) clearly scale with a larger exponent than \( \langle \lambda_1 \rangle \). Note, however, that there is very little curvature in the data for \( \langle R_g^2 \rangle \) so that considerably larger system sizes will be required before the true asymptotic behavior \( \langle R_g^2 \rangle \sim N \) is attained.

Figure 2 contains results for \( \lambda_1(N_s) \) obtained by taking averages on subdomains containing \( N_e \) vertices for a membrane of size \( N = 469 \). Although there is no simple powerlaw relationship between \( \lambda_1(N_s) \) and \( N_e \), the average slope of the log-log plot is approximately 0.71.

The scaling functions, which are indicated by the solid and dashed lines in figure 2 have been evaluated as follows. Restricting attention to the out-of-plane motion, the leading order contribution to the elastic bending energy of a thin, flat plate from its \( z = 0 \) zero temperature reference state is [18]

\[
\frac{\beta H}{\kappa} = \int d^2r \left\{ \frac{\kappa}{2} \nabla^2 z(r)^2 + \bar{\kappa} \det[\partial_i \partial_j z(r)] \right\},
\]

(3)
Fig. 1. — Scaling plot of the mean square radius of gyration \(< R_{g}^{2} >\) and expectation value \(< \lambda_{1} >\) of the smallest eigenvalue of the moment of inertia tensor for membranes containing \(N = 37, 127, 271,\) and 469 vertices. The solid line has slope 0.87, and the dashed line is a plot of \(< \lambda_{1} > \sim N^{0.63}\)

where, for a plate of thickness \(h\) composed of an isotropic elastic material with three-dimensional Young’s modulus \(E\) and Poisson’s ratio \(\sigma\), \(\kappa = Eh^{3}/[12(1-\sigma^{2})]\) and \(\bar{k} = (1-\sigma)\kappa\) [18].

The Hamiltonian (3) can be diagonalized by expanding \(z(r)\) in eigenfunctions of the equation

\[
\nabla^{4}z = \lambda^{4}z, \tag{4}
\]

where for a circular disc of radius \(R\) with free edges, the boundary conditions are

\[
\nabla^{2}z|_{r=R} - (1-\sigma) \left[ \frac{1}{r^{2}} \frac{\partial^{2}z}{\partial \theta^{2}} + \frac{1}{r} \frac{\partial z}{\partial r} \right]_{r=R} = 0 \tag{5.a}
\]

and

\[
\frac{\partial}{\partial r} \nabla^{2}z \bigg|_{r=R} + (1-\sigma) \left[ \frac{1}{r} \frac{\partial}{\partial r} \left( \frac{1}{r} \frac{\partial^{2}z}{\partial r \partial \theta} - \frac{1}{r^{2}} \frac{\partial z}{\partial \theta} \right) - \frac{1}{r^{2}} \frac{\partial z}{\partial \theta} \right]_{r=R} = 0. \tag{5.b}
\]

The solutions of (4) are

\[
\Psi_{nI}(r, \phi) = c_{nI} \left[ J_{n}(\lambda_{nI} r) + B_{nI} I_{n}(\lambda_{nI} r) \right] \left\{ \begin{array}{c} \sin(n\phi) \\ \cos(n\phi) \end{array} \right\}, \tag{6}
\]

where \(J_{n}\) and \(I_{n}\) are Bessel functions. There is a doubly infinite set of eigenvalues \(\lambda_{nI}\), with \(n = 0, 1, 2, \ldots\) and \(l = 1, 2, \ldots\) enumerating the eigenvalues for each order \(n\). The eigenvalues (as well as the \(B_{nI}\)) are determined by the boundary conditions (5); \(c_{nI}\) is a normalization constant.

Because of nonlinear couplings between the out-of-plane displacements \(z\) and the in-plane phonon degrees of freedom \(u\), the renormalized long-wavelength bending rigidity and elastic constants differ considerably from their microscopic values [4]. These renormalized elastic constants enter an effective long-wavelength free energy [10, 19] for the transformed variable \(z_{\lambda}\),

\[
\beta \mathcal{H}_{\text{eff}} = \frac{1}{2} \sum_{\lambda} \kappa_{R}(\lambda) \lambda^{4} |z_{\lambda}|^{2}, \tag{7}
\]
with \( \kappa_R(\lambda) \sim \lambda^{-\eta} \) and \( \eta = 2(1 - \zeta) \).

In the flat state, the expectation value of the smallest eigenvalue of the moment of inertia tensor for a subdomain containing \( N_s \) vertices is

\[
< \lambda_1(N_s) > = \frac{1}{2N_s^2} \left( \sum_{i,j} (z_i - z_j)^2 \right),
\]  

(8)

where the sums run over all monomers in the subdomain. In the continuum limit, for a disc of area \( A_s \), (8) becomes

\[
< \lambda_1(A_s) > = \frac{1}{A_s} \left[ A_s \int_{A_s} d^2r < z^2(x) > - \int_{A_s} d^2r \int_{A_s} d^2r' < z(x)z(x') > \right].
\]  

(9)

Using the mean-field eigenfunctions (6), we can transform expression (9) and evaluate the resulting expectation values using (7). We thereby neglect any renormalization effects on the eigenfunctions; the resulting expression for \( < \lambda_1(N_s) > \) is therefore evaluated in the mean-field approximation, employing, however, the correct value of \( \eta \). In performing the resulting sums, we have used \( \eta = 0.6 \) \((\zeta = 0.7)\), as well as both \( \sigma = 1/3 \), the value of the Poisson ratio for a two-dimensional harmonic triangular lattice [20, 21], and \( \sigma = 0 \). We have also cut off the sums in \( I \) and \( n \) when \( \lambda_{nl} \approx 2 \). The result for \( \sigma = 1/3 \) is given by the solid line, and that for \( \sigma = 0 \) by the dashed line in figure 2; \( N_s = 3R(R+1)+1 \), with \( R \) the radius of the subdomain of the disc of radius 12. The agreement is quite satisfactory for \( \sigma = 1/3 \). For smaller values of \( \sigma \) the scaling function develops a more pronounced dip for large \( N_s \), and for large values of \( \sigma \), the increase in \( < \lambda_1(N_s) > \) near the edge of the membrane becomes more pronounced. The value for \( \zeta \) we obtain in this way is somewhat larger than what we found from an analysis of the smallest eigenvalue of the moment of inertia tensor.

![Graph](https://via.placeholder.com/150)

Fig. 2. — \( < \lambda_1(N_s) > \) as a function of the number of vertices \( N_s \) in the subdomain. The solid line is the scaling function obtained using \( \sigma = 1/3 \) and \( \eta = 0.6 \) \((\zeta = 0.7)\). The dashed line was obtained using \( \sigma = 0 \).

Further information concerning the roughness of the membrane can be obtained by studying the behavior of the unit normal vectors of the elementary triangles of the surface. Let \( M_j(\alpha) \) be the projection of the unit normal of triangle \( \alpha \) along the eigenvector \( \hat{e}_j \) of the moment.
Fig. 3. — Typical configuration of a tethered network consisting of 469 vertices (after $10^6$ MCS). (a) Projection on $xz$ plane. (b) Projection on $yz$ plane.
of inertia tensor. Following reference [12], we have evaluated $< M_j^2 > = [\frac{1}{N_{\Delta}} \sum_{\alpha} M_j(\alpha)]^2$ (where $N_{\Delta}$ is the number of elementary triangles) for a network containing $N = 469$ vertices. We find $< M_1^2 >= 0.056$, $< M_2^2 >= 1.3 \times 10^{-3}$, and $< M_3^2 >= 5.4 \times 10^{-4}$. As expected for an oriented, flat membrane, both $< M_1^2 >$ and $< M_2^2 >$ are essentially zero. The rather small value for $< M_1^2 >$ implies that the surface is very rough. A typical configuration of a tethered network with $N = 469$ vertices is shown in figure 3.

![Fig. 4. - The structure factors $S_3(q)$ (o), $S_1(q)$ (o), and $S(q)$ (x) plotted as a function of $q$ for $L = 25$ ($N = 469$).](image)

Finally, we have also determined the directionally averaged static structure factor

$$S(q) = \frac{1}{N^2} \sum_{i,j} \left\langle \frac{\sin(qr_{ij})}{qr_{ij}} \right\rangle,$$

where $r_{ij} = |r_i - r_j|$, as well as

$$S_j(q) = S(q) = \frac{1}{N^2} \sum_{i,j} \left\langle e^{iq(r_i - r_j)} \right\rangle,$$

for $q = q \hat{e}_j$, $j = 1 (z)$ and 3 ($x$), where the $\hat{e}_j$ are the eigenvectors of the moment of inertia tensor. The average in (11) is taken with respect to the frame of reference fixed to the principle axes of the membrane.

Our results for these three structure factors are shown in figure 4 for a membrane of size $L = 25$ ($N = 469$). As can be seen, the three structure factors behave very differently and converge only at $q$-values that correspond to the length of a single bond. The oscillations in $S_3(q)$ arise from scattering of a two-dimensional planar object with, in this case, a fairly diffuse interface. This has been discussed in detail by Abraham and Nelson [10]. For large $q$, one expects $S_1(q) \sim q^{-2/\ell}$. In the present case, however, fluctuations are so strong, that this regime is not yet reached, even for a network with $L = 25$. In contrast, this behavior is readily
observed in string-and-bead networks of the same size [10]. As a result of the rapid decrease in $S_1(q)$, the directionally averaged scattering function $S(q)$ appears to scale as $q^{-2.7}$ over a relatively large $q$-range. The small dip near $q = 1$ is a vestige of the structure in $S_3(q)$.

Finally, scaling plots of $S(q)$ vs $qL^{0.75}$ and $S_1(q)$ vs. $qL^{0.63}$ obtained using data for membranes of size $N = 271$ and $N = 469$ are shown in figures 5a and 5b, respectively. The excellent collapse of the data for $S(q)$ is consistent with an isotropic crumpled membrane with the fractal dimension $d_f \approx 2/0.75 \approx 2.7$. Indeed, not only does the data collapse for this value of $d_f$, but the slope in the regime $2\pi/a > q > 2\pi/R_g$ is also consistent with $S(q) \approx q^{-d_f}$. The solid line
in figure 5a is proportional to \((qL^{0.75})^{-2}\). In contrast with the behavior of \(S(q)\), however, \(S_1(q)\) scales with \(qL^\zeta\), with \(\zeta = 0.63\), as expected for a flat membrane. This value for \(\zeta\) agrees with that obtained from an analysis of the smallest eigenvalue of the moment of inertia tensor.

4. Discussion.

In this paper we have presented results for the conformation and scaling behavior of two-dimensional tethered networks obtained using a model for extremely flexible triangulated surfaces. The model is constructed so that the resulting network is very flexible and can fold in on itself without any cost in energy; it is therefore much rougher than the commonly studied string-and-bead models. Although the network is still asymptotically flat, the large out-of-plane fluctuations result in a directionally averaged structure factor which exhibits a nontrivial scaling behavior reminiscent of a crumpled object with a fractal dimension \(d_f \approx 2.7\). This result agrees with the conclusions of a recent study of “giant-hole membranes” [17] in which the local flexibility of the standard string-and-bead model of tethered surfaces is enhanced by replacing the interparticle bonds by tethered strings several particles long. It must be stressed, however, that this result is a finite size effect, and ultimately, for very large system sizes, the anisotropic behavior described in references [10 and 16] would be observed. An important open question is just how large the system needs to be before this occurs. Although our work is restricted to membranes of diameter \(L \leq 25\) bond units, the results indicate that the behavior we observe should persist to considerably larger systems sizes.

The model we have studied is very “rough” at short length scales, and the amplitude of both the out-of-plane and in-plane fluctuations are substantially larger than in the standard string-and-bead models for tethered surfaces. The analysis of reference [16] therefore predicts that there should be a large \(q\)-regime in which \(S(q)\) scales as \(q^{-4/\zeta+2}\). We find a substantially slower decrease in \(S(q)\) with \(q\); note, however, that the behavior we observe lies between the two regimes \(q^{-3+\zeta}\) and \(q^{-4/\zeta+2}\) predicted in reference [16]. It is therefore possible that our data lie in the crossover region between these two scaling regimes and that substantially larger system sizes will be required to see the predicted behavior.

Finally, the graphite oxide membranes studied in reference [5] are claimed to have an aspect ratio on the order of 1000. What is important, however, is the size of the membranes in terms of the elementary units which fold. A correct interpretation of the behavior observed in graphite oxide membranes will require a more detailed characterization of the microscopic structure and conformation of this material.

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[20] The Poisson ratio $\sigma$ in (5) is the same as the two-dimensional Poisson ratio for the corresponding shell. See reference [18].