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Absence of a crumpling transition in strongly self-avoiding tethered membranes

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We report extensive simulations of a model for tethered self-avoiding membranes with bending rigidity. These simulations have been performed at finite and infinite temperature for membranes whose stretched configurations are hexagonal with linear dimensions L in the range 5–19. We have analyzed the shape of the clusters by calculating the eigenvalues λ_i, $i=1-3$ of the inertia tensor and the structure factor for wave vectors in the direction of the eigenvectors of this matrix. We find that the smallest eigenvalue scales, for all temperatures investigated, as $\lambda_1 \sim L^{\nu_1}$ with $\nu_1 \approx 0.65$; the two larger eigenvalues scale as L. Thus, in the thermodynamic limit $L \to \infty$, the tethered self-avoiding membrane is flat but rough, in contrast to previous conclusions [Y. Kantor and D. R. Nelson, Phys. Rev. Lett. **58**, 2774 (1987); Phys. Rev. A **36**, 4020 (1987)], and in contrast to the behavior of membranes without self-avoidance which display a thermodynamic phase transition between a crumpled high-temperature phase and a low-temperature flat phase [Y. Kantor, M. Kardar, and D. R. Nelson, Phys. Rev. Lett. **57**, 791 (1986); Phys. Rev. A **35**, 3056 (1987)].

The study of random surfaces is of great interest from both a conceptual and practical point of view. On the practical side, the static and dynamic properties of systems such as microemulsions, biological membranes, and vesicles seem to be determined by the characteristics of two-dimensional interfaces embedded in three-dimensional space. On the conceptual side, there are indications that simple models of random surfaces may display novel phase transitions which are at this point poorly understood. One such model—the tethered membrane model—was introduced recently by Kantor, Kardar, and Nelson.[1] In its simplest form the system consists of hard spheres connected in a fixed geometry by flexible strings (see Fig. 1). To prevent self-intersection of the membrane, the maximum length of the strings between the centers of the spheres, for a triangular network of interconnections, must be less than or equal to $\sqrt{3}d$ where d is the hard-sphere diameter. In this simple model, there are no energy parameters and the free energy of the membrane is simply its entropy. Entropy generates an effective elasticity—the free energy can be taken to be of the form $F=-KR^2$ where R is the radius of the volume occupied by the membrane—and this tends to favor a collapsed configuration. The hard-core exclusion competes with this effect and it is this competition which creates the possibility of an interesting phase even in the absence of a finite-energy parameter. We note that if the strings are longer than $\sqrt{3}d$, the membrane is self-avoiding in the sense that particles have an excluded volume but not "properly" self-avoiding in that the surface can intersect itself. We believe that self-intersections are crucial for the existence of a crumpled phase in equilibrium.

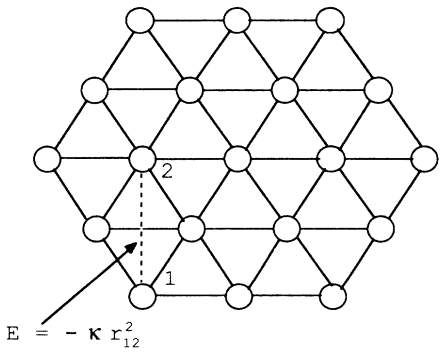

FIG. 1. Planar configuration of a tethered membrane of linear dimension $L=5$. The solid lines represent flexible strings, the dashed line indicates the second-neighbor interaction responsible for the bending rigidity.

Previous numerical work on tethered membranes can be divided into two categories. First, because of the long relaxation times of simulations, Kantor, Kardar, and Nelson[1] studied a version of the model in which the hard-core interaction between particles is ignored except for nearest neighbors in the network. This means that the membrane can intersect itself and hard spheres can overlap. Their work on this "phantom" network clearly indicated a high-temperature crumpled phase with the property $R_g \sim (\ln L)^{1/2}$ (L^ν, with $\nu=0$), where L is the linear dimension of the network. This form of the radius of gyration can be derived analytically for a free surface with fixed connectivity. A Migdal renormalization-group analysis also indicated that the form of the nearest-neighbor interaction is irrelevant.[1]

For properly self-avoiding surfaces only infinite temperature simulations have been carried out prior to the present work. Kantor *et al.*[1] studied membranes with $N \leq 225$ particles with a stretched configuration in the shape of a parallelogram. They calculated, among other properties of the membrane, the spherically averaged structure factor

$$S(\mathbf{k}) = S(k) = \frac{1}{N^2}\left\langle \sum_{\mathbf{x},\mathbf{x}'} e^{i\mathbf{k}\cdot[\mathbf{r}(\mathbf{x})-\mathbf{r}(\mathbf{x}')]} \right\rangle . \quad (1)$$

The behavior of this function provides the most persuasive

numerical evidence for the existence of a crumpled phase. When plotted as a function of the scaled variable kL^{ν} with $\nu=0.83$ for different L, the data for $S(k)$ collapses rather well to a single curve. The behavior of the radius of gyration is also consistent with the functional form $R_g \sim L^{0.8}$, the exponent predicted for self-avoiding surfaces by a Flory argument.[1]

A generalization of this model is obtained if one incorporates rigidity into the Hamiltonian. Curvature energies are known to be important in determining the properties of microemulsions and membranes[2] and, here, they serve to set a temperature scale. If the infinite-temperature phase (zero rigidity) is indeed crumpled, we have the possibility of a thermodynamic phase transition between this phase and a low-temperature flat phase. In our calculations we have introduced rigidity by means of *repulsive* springs between second-neighbor atoms on the triangular network. Thus our model has the Hamiltonian

$$\beta H = -\frac{\kappa}{2}\sum_{\mathrm{NNN}} [\mathbf{r}(\mathbf{x})-\mathbf{r}(\mathbf{x}')]^2 , \tag{2}$$

where κ is the rigidity parameter, $\mathbf{x}$ is a two-dimensional vector which specifies the position of a particle on the underlying network, $\mathbf{r}(\mathbf{x})$ is the position in the three-dimensional space, and the sum extends over second neighbors on the network. The hard-core exclusion and tethering constraints have not been explicitly stated in (2). In a separate study of their phantom surface, Kantor and Nelson[3] found a phase transition between the ideally crumpled phase $[R_g \sim (\ln L)^{1/2}]$ and a low-temperature flat phase $(R_g \sim L)$ as a function of κ [these authors used a different form of the bending energy than (2) but for small curvatures the two energy parameters are equivalent].

In our simulations we have studied self-avoiding surfaces in a hexagonal stretched configuration (see Fig. 1). For diameter L the membrane consists of $N=(3L^2+1)/4$ particles and we have studied systems up to $L=19$ $(N=271)$. Conventional Monte Carlo simulations with a variable stepsize $\leq 0.2\sqrt{3}d$ were carried out and the length of the flexible tethers was taken to be small enough that it was impossible for the membrane to pass through itself in a single Monte Carlo step without overlap of hard spheres. A measure of the relaxation time between two statistically independent configurations of phantom membranes is given by the Rouse relaxation time[1] $\tau_R \approx N/s^2$, where s is the stepsize. The infinite temperature simulations were run for at least 1000 Rouse relaxation times and, for the largest membrane, for 2000 relaxation times. For finite temperature, the simulations (for $L \leq 11$) were run for 500 relaxation times. In all cases we were satisfied that the expectation values showed no tendency to drift after an initial transient which lasted, for $L=19$, for roughly $200\tau_R$.

We first attempted to find a critical point by determining the temperature dependence of the specific heat, the radius of gyration, and the shape parameter $A=\lambda_1/\lambda_3$, where λ_1 is the smallest and λ_3 the largest eigenvalue of the inertia tensor (matrix elements $I_{jm}=\langle r_j r_m\rangle - \langle r_j\rangle\langle r_m\rangle$ where angular brackets indicate averaging over particle positions in a given configuration). In contrast to the phantom surfaces,[3] the specific heat is a smoothly increasing function of κ with no hint of an emerging singularity as the size of the network is increased. Similarly, we found no evidence of a transition in either A or R_g. Thus, we decided to reexamine the infinite-temperature case. For our more symmetric hexagonal networks, the two largest eigenvalues of the inertia tensor are nearly equal and considerably larger than the smallest eigenvalue. As function of L, the parameter A *decreases:* $A(L=5)=0.17$, $A(L=11)=0.12$, and $A(L=19)=0.095$.

In view of this strong anisotropy, we separately determined the structure factor for wave vectors projected along the eigenvectors of the inertia tensor. Thus, if $\mathbf{e}_1$, $\mathbf{e}_2$, and $\mathbf{e}_3$ are the eigenvectors corresponding to λ_1, λ_2, and λ_3 in a given configuration, we define $\mathbf{k}_j = k\mathbf{e}_j$ and calculate

$$S_j(k) = \frac{1}{N^2}\left\langle \sum_{\mathbf{x},\mathbf{x}'} e^{i\mathbf{k}_j\cdot[\mathbf{r}(\mathbf{x})-\mathbf{r}(\mathbf{x}')]} \right\rangle , \tag{3}$$

where the angular brackets indicate averaging over the configurations of the Monte Carlo run. The results of these calculations of $S_1(k)$ and $S_3(k)$ are displayed in Figs. 2 and 3 plotted on a log-log scale as functions of the scaled variables $q_1 = kL^{\nu_1}$ with $\nu_1=0.65$ in the case of S_1 and $q_3 = kL^{\nu_3}$ with $\nu_3=0.975$ in the case of S_3. The numbers quoted above for the exponents ν_j provide the best (visual inspection) collapse of the data for a large range of q and S. We have also carried out a more formal analysis of the structure factor by choosing a value of S_1 or S_3 (in the range $0.03 \leq S \leq 0.3$) and then determining the value of k, corresponding to this value of S, as function of L. The resulting (k,L) points were fitted to the functional form $k=aL^{\nu}$. This procedure yields $\nu_1=0.65\pm0.05$ and $\nu_3=0.96\pm0.05$. Since $\nu_j \leq 1.0$, we conjecture that

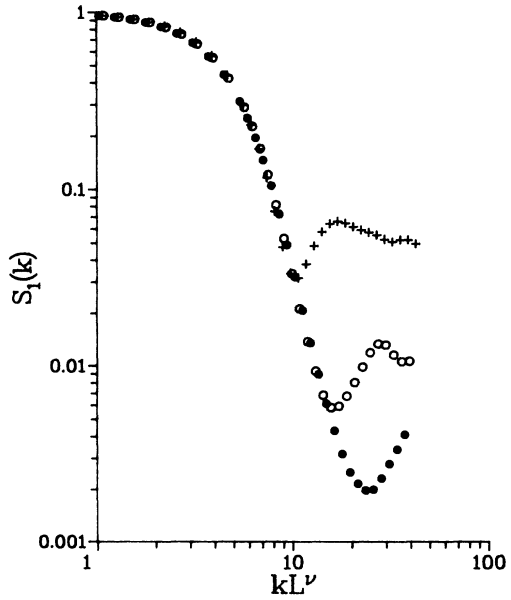

FIG. 2. The "perpendicular" structure factor $S_1(k)$ for wave vectors in the direction of the eigenvector corresponding to the smallest eigenvalue of the inertia tensor plotted as function of the scaled variable kL^{ν} with $\nu=0.65$. Crosses, $L=5$ $(N=19)$; open circles, $L=11$ $(N=91)$; filled circles, $L=19$ $(N=271)$.

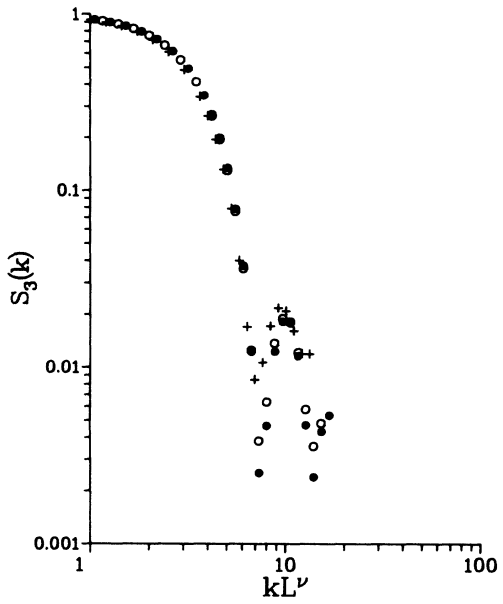

FIG. 3. The in-plane structure factor $S_3(k)$ ($\mathbf{k}$ in the direction of the eigenvector corresponding to the largest eigenvalue of the inertia tensor) plotted as function of the scaled variable kL^ν with $\nu=0.975$. Symbols correspond to the same size membranes as in Fig. 2.

$\nu_3=1.0$, indicating that even in the absence of bending rigidity the self-avoiding tethered membrane is flat at large length scales.

We note that our numerical results are not inconsistent with those of Ref. 1. If one plots the spherically averaged structure factor (1) as function of the appropriately scaled wave vector $q=kL^\nu$, a reasonable collapse of the data is obtained for $\nu\approx 0.8$. However, over most of the range of q, the largest contribution to the spherically averaged structure factor comes from S_1 which becomes small near $k\approx\pi/\sqrt{\lambda_1}$. The contributions from wave vectors lying in the "plane" of the membrane become negligible at smaller wave vectors because of the strong anisotropy.

We have also determined the properties of the self-avoiding membranes for bending rigidity $\kappa\leq 1.0$. In the phantom membrane, the phase transition to the low-temperature flat phase takes place at $\kappa\approx 0.15$ and this obviously provides an upper bound for the critical rigidity in the self-avoiding case. To within our numerical uncertainty, the characteristic exponents ν_1 and ν_3 are independent of the rigidity parameter, indicating that even at low-temperatures tethered membranes are rough (in the sense that λ_1 diverges in the thermodynamic limit), with a nontrivial dependence of the width on the linear dimension of the network.

Finally, we comment briefly on the renormalization-group analysis of the crumpling transition.[4] This work has been based on a generalization of the Edwards model[5] of polymers in which the hard-core exclusion of the particles is modeled by a δ-function potential. Thus, the cost in energy of a self-intersection of the membrane is finite rather than infinite as in the system that we have simulated. We have some evidence, based on a few simulations of tethered spheres of small diameter, that the possibility of self-intersections may be crucial for the existence of a crumpled phase. If confirmed, this result is in marked contrast to the case of bead and string models of polymers[6] where the hard-sphere diameter is an irrelevant variable and whose universal properties are correctly described by the Edwards model. Further properties of these interesting systems as well as simulations of considerably larger systems are in progress and will be reported elsewhere.

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