Molecular Dynamics of Tethered Membranes

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(Received 11 January 1989)

By molecular-dynamics simulation, we investigate the possible existence of a crumpling transition for a model of tethered membranes, where the particles are tethered by a continuous potential. For distant-neighbor interactions, the potential is repulsive and contains a variable hard-core diameter parameter. By varying this parameter, we are able to study in detail the effect of self-avoidance. Our results suggest the interpretation that self-avoiding two-dimensional tethered membranes are asymptotically flat, even without an explicit bending rigidity, and that there is no crumpling transition except for "phantom" membranes.

PACS numbers: 64.60.−i, 82.65.−i

In recent years there has been considerable interest in the large-scale properties of random surfaces in the context of both field theory[1] and condensed-matter theory.[2] Condensed-matter systems in which surfaces and interfaces play a significant role include microemulsions, lipid bilayers, vesicles, and suspensions of monolayers of exfoliated layered crystals. While much of the statistical mechanics of two-dimensional surfaces embedded in a higher-dimensional space remains to be understood, it is known that there is no single universality class which encompasses all such systems.[3]

Recently, Kantor, Kardar, and Nelson[4] introduced an interesting model of random surfaces, namely, the tethered membrane. In this model, particles (hard spheres of diameter σ) are joined in a fixed pattern by flexible tethers of length l. For $l \leq (3)^{1/2}\sigma$, the membrane is self-avoiding in that it cannot intersect itself without the overlap of hard spheres. Without further parameters, the properties of this membrane are temperature independent and determined by the entropy. Kantor, Kardar, and Nelson[4] argued, on the basis of a Flory mean-field theory, that such membranes are crumpled; i.e., the radius of gyration scales as $R_g \sim L^{\nu}$, where L is a characteristic size of the membrane, and the exponent ν takes on the value 0.8 for two-dimensional membranes free to move in three-dimensional space. Monte Carlo simulations and renormalization-group arguments[4,5] seemed to support this conclusion.

Recently, Plischke and Boal[6] have questioned this result on the basis of larger and more detailed simulations. By examining the shape of tethered self-avoiding membranes they reached the conclusion that these systems are asymptotically flat at large length scales. In particular, they found that the eigenvalues of the inertia tensor have the scaling behavior $\lambda_1 \sim L^{2\nu_1}$ and $\lambda_3 \sim L^{2\nu_3}$, where λ_1 and λ_3 are the smallest and largest eigenvalues, respectively. The exponents were consistent with the values $\nu_1 \approx 0.7$ and $\nu_3 \approx 1.0$, corresponding in the limit of large size to a rough but flat object.

These results are surprising when taken together with the fact that "phantom" membranes are crumpled[7] and undergo a finite-temperature transition to a flat phase if the membrane has explicit bending rigidity. In a phantom membrane the particles interact only with nearest neighbors on the network, and there is a great deal more phase space available. The ideal phantom membrane consists of particles with zero diameter, and one finds[4,7] in the absence of bending rigidity that $R_g \approx [\ln(L)]^{1/2}$ for large L. The two results may be reconcilable if there is a critical value of the particle diameter (for fixed tethering length) at which there is a transition from a crumpled phase to a flat phase. We note that the ideally crumpled (Gaussian) phase of the phantom membrane cannot exist for any nonzero diameter of the particles since $[\ln(L)]^{d/2}/L^2 \rightarrow 0$ as $L \rightarrow \infty$ for a two-dimensional membrane in a d-dimensional space.

In this Letter we report the results of large-scale molecular-dynamics simulations of a model for tethered membranes. Our model is slightly different from that of Refs. 4 and 6 in that tethering is enforced by a continuous potential. The particles on the network are arranged in a triangular array and interact with their nearest neighbors through the potential

$$V_{\mathrm{NN}}(r) = \begin{cases} 4\epsilon[(1/r)^{12} - (1/r)^6 + 1/4], & 0 < r < 2^{1/6}, \\ 0, & 2^{1/6} \leq r < 2^{1/6} + l, \\ 4\epsilon[(1/r')^{12} - (1/r')^6 + 1/4], & 2^{1/6} + l \leq r, \end{cases} \quad (1)$$

where $r' = 2(2^{1/6}) + l - r$. The region $2^{1/6} < r < 2^{1/6} + l$ is thus force free and equivalent to the flexible string of other models[4,6] of tethered membranes. In our calculations we have taken $l = 0.5$. Self-avoidance is generated by the interaction between particles which are not

nearest neighbors on the network. We take this interaction to be

$$V_d(r) = \begin{cases} 4\epsilon[(\sigma/r)^{12} - (\sigma/r)^6 + 1/4], & 0 < r < 2^{1/6}\sigma, \\ 0, & r \geq 2^{1/6}\sigma, \end{cases} \quad (2)$$

with σ a variable parameter. The parameter σ is a measure of an "effective" hard-core radius of the particles. The case $\sigma = 0$ corresponds to the phantom membrane; $\sigma = 1$ to the strongly self-avoiding membrane in which self-intersections are impossible. We have considered finite systems that are hexagonal in shape[6] and characterized by a linear dimension L. A hexagonal sheet of size L contains $(3L^2+1)/4$ particles. We have simulated membranes up to size $L = 75$ (4219 particles) and for 10^6 to 8×10^6 total time steps, the longer times corresponding to the larger clusters. The largest previously reported simulations[6] were for membranes with $L = 19$ (271 particles). The procedure is a straightforward molecular-dynamics calculation. The membrane is initially in a flat configuration and the particles are given random displacements and zero velocities. All clusters equilibrate to a mean reduced temperature k_BT/ϵ between 0.6 and 0.7, and have zero total linear and angular momentum. The classical equations of motion are integrated forward in time, and the appropriate microcanonical ensemble averages are calculated.

Both the phantom and self-avoiding membranes are characterized by long relaxation times and large fluctuations in equilibrium. Figure 1 shows snapshots of molecular-dynamics configurations which display folding of the 4219-atom particle membrane, an effect which contributes to slow relaxation and large fluctuations. The figure shows four snapshot configurations at (0.65, 0.80, 0.95, and $1.1)\times10^6$ time steps, each configuration being viewed along the z axis (left-hand picture), the y axis (upper right-hand picture), and the x axis (lower right-hand picture), respectively. The membrane is initialized about the x-y plane. These snapshots show the "folding" motion, which typically has a period of $\approx 5\times10^5$ time steps for this large size cluster. Between these folding configurations, the membrane returns to a nearly "flat" hexagonal form. In order to determine if our results are characteristic of equilibrium, we have calculated the autocorrelation functions of the macroscopic variables of interest and have estimated the relaxation times. In all cases, the molecular-dynamics simulation was carried out for at least 100 such relaxation times and, except for $L = 75$, for a much longer period.

The essential results of our calculations are displayed in Figs. 2 and 3. In Fig. 2 we plot $\ln(R_g)$ as a function of $\ln(L)$ for a range of σ. The straight lines joining the

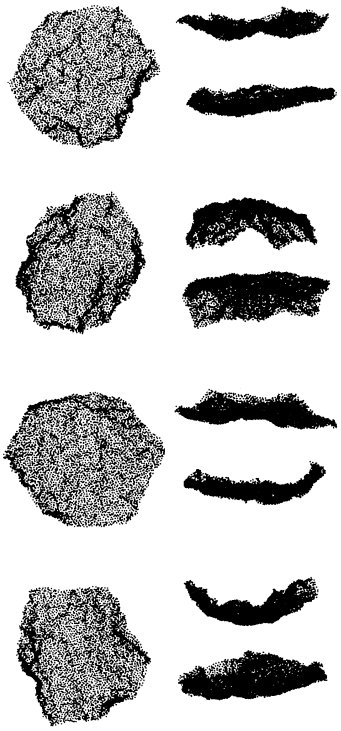

FIG. 1. Configurations of a self-avoiding tethered membrane of 4219 particles ($L = 75$) and $\sigma = 1$. The individual particles are denoted by dots, and tethered bonds are not drawn. From top to bottom, the different views for the four configurations are at times (0.65, 0.80, 0.95, and $1.10)\times10^6$ time steps, respectively.

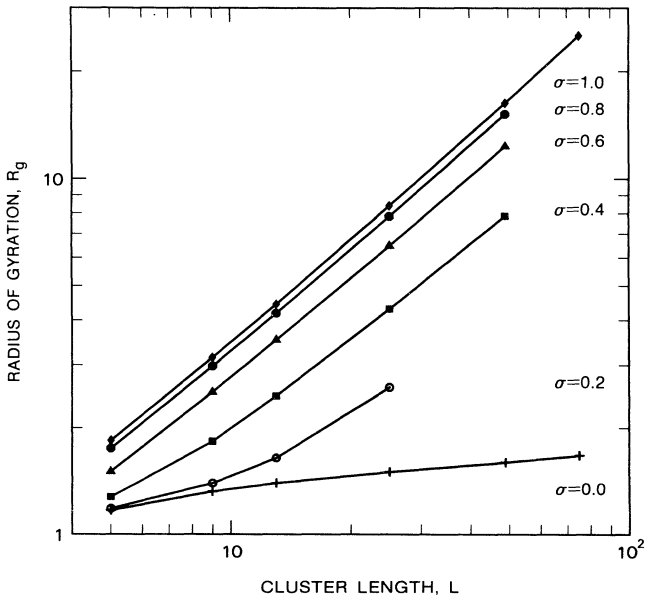

FIG. 2. Log-log plot of the radius of gyration R_g of tethered membranes for various values of the parameter σ. Straight-line segments are drawn through the data points and have slope equal to the effective exponent $\nu(L)$ which, for all $\sigma \neq 0$, increases with L and is consistent to the asymptotic value $\nu(\infty) = 1$.

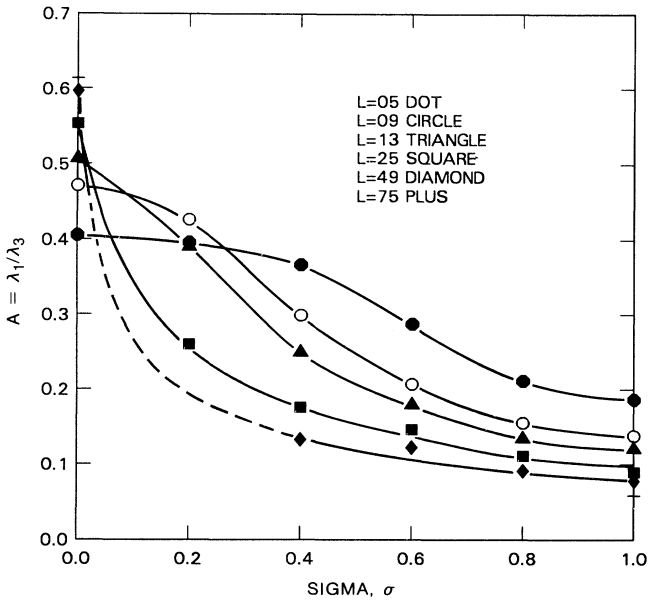

FIG. 3. The shape parameter $A=\lambda_1/\lambda_3$ plotted as a function of σ for various L. As discussed in the text, we believe that this shows that tethered membranes with $\sigma \geq 0.2$ are asymptotically flat and suggests that the same conclusions hold for all $\sigma > 0$.

data points have a slope equal to the effective exponent $\nu(L)$ in the scaling relation $R_g \sim L^{\nu}$. We see a typical crossover effect toward larger values of ν as L becomes larger for all values of σ, except $\sigma=0$. For $\sigma=1$, the exponent ν is unity. The data for $0.4 \leq \sigma \leq 1$ strongly suggest $\nu(\infty)=1$ for these σ's. For $\sigma=0.2$, we did not simulate large membranes because of the large computational burden, and the finite-size effects dominate. But we conjecture that this system also has $\nu(\infty)=1$. By contrast, the decreasing effective exponent of the phantom membrane is consistent with the expected logarithmic functional form $R_g \sim (\ln L)^{1/2}$ and was verified on a semilog plot.

We have also determined the eigenvalues of the inertia tensor I, the matrix elements of which are given by $I_{jm}=\langle (r_j-\langle r_j\rangle)(r_m-\langle r_m\rangle)\rangle$, where the angular brackets indicate an average over a molecular-dynamics configuration. The sum of these eigenvalues is equal to R_g^2 for the particular configuration. We denote the smallest eigenvalue by λ_1, the largest by λ_3, and in Fig. 3 display the "shape" function $A=\lambda_1/\lambda_3$ as function of σ for various L. The curves are a guide to the eye.

For all $\sigma \geq 0.2$ the shape function decreases with increasing L, indicating that the membranes are becoming more flat as they become larger. Conversely, the phantom membrane becomes more symmetric as the size is increased. We have analyzed the eigenvalues in terms of the assumed functional forms $\lambda_j \propto L^{2\nu_j}$ and find that the data strongly support $\nu_2=\nu_3=1.0$ for $\sigma \geq 0.4$ and are consistent with $\nu_1 \approx 0.8$, a somewhat larger value than that reported in Ref. 6. For $\sigma=0.2$, the finite-size effects that are also apparent in Fig. 2 prevent an accurate determination of the exponents.

The data presented in Fig. 3 also suggest that *any* degree of self-avoidance *may* lead to a flat configuration at large L, although more data for low σ are clearly desirable. The values of σ at which the curves for L_1 and L_2 intersect $[A(L_1)=A(L_2)]$ decrease steadily as the values of L_1 and L_2 increase. We can interpret these intersection points as a sequence of approximations to the critical value σ_c at which an infinite membrane becomes flat, and we see that this value is clearly less than 0.2 and may well be zero. If this conjecture is correct, the tethered membrane has the property, also found in polymers, that there is a single fixed point which controls the large-scale behavior of the system no matter how small the hard-core diameter of the particles. The addition of bending rigidity to the Hamiltonian can only make the system more flat, and we therefore conjecture that there is no finite-temperature crumpling transition in self-avoiding tethered membranes.

The flat phase found, at least at large σ, also seems to be intrinsically interesting. The fact that the smallest eigenvalue of the inertia tensor diverges as $L^{2\nu_1}$ with $\nu_1 \approx 0.8$ indicates that the membrane is rough. At the moment we have no analytic theory which predicts this behavior although the works of Nelson and Peliti[8] and of Aronowitz and Lubensky on solid membranes[9] do predict a nontrivial exponent for the transverse correlation function.

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