Excluded-volume interactions in tethered membranes

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The Monte Carlo method is applied to investigate the asymptotic behavior of self-avoiding polymerized membranes. We use a model described by a *local* Hamiltonian: repulsive interactions (hard spheres of diameter σ) act only between atoms whose degree of neighborhood (measured along the membrane) does not exceed a certain value l. For a particular value $\sigma=\sigma_c(l)$ the membrane undergoes a crumpling transition between asymptotically flat and crumpled states. We find that when l increases, $\sigma_c(l)$ decays to zero, and conclude that self-avoiding surfaces with purely repulsive interactions are asymptotically flat even for very small values of σ.

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I. INTRODUCTION

The tethered membrane, or surface, model was first introduced [1] as a generalization of linear polymers [2] and was intended to represent the properties of polymerized two-dimensional membranes [3]. In the past there were relatively few experimental realizations of the model, such as the spectrin network (membrane skeleton) [4] or exfoliated sheets of graphite oxide [5]. Recently, very large two-dimensional molecules have been produced by Stupp *et al.* [6]. From the theoretical point of view such membranes display richness that is absent from linear polymers: In the absence of excluded-volume interactions (i.e., when the surface is allowed to self-intersect) the soft model membrane is crumpled, and its root-mean-squared size R_g increases as $\sqrt{\ln L}$, where L is its linear size. However, as the bending rigidity of the membrane increases it undergoes a second-order transition [7,8] into a flat state. Such a (crumpling) transition has no analog in linear polymers, where the bending rigidity can only modify the persistence length. Properties of the flat state have been under intensive study during recent years. In Sec. II, a brief overview of the current theoretical status of the problem will be presented.

Introduction of excluded-volume interactions significantly modifies the behavior of the membranes. The first study of the problem [1,9] indicated that for self-avoiding membranes $R_g \sim L^{\nu}$ with $\nu \approx 0.8$. This result coincided with the prediction of a simple generalization [9] of Flory theory to polymerized membranes. Subsequent numerical studies, however, failed to recover such behavior [10–13] and concluded that self-avoiding membranes are always flat. While originally such a failure was attributed to technical problems related to details of the models, gradually a growing body of evidence appeared indicating that flatness is an inherent property of self-avoiding surfaces. Nevertheless, few studies observed the crumpled state [5,14,15]. In Sec. III we discuss in some detail the current state of the theory and experiments.

Due to growing experimental importance of the subject it is imperative to resolve the problem posed by the self-avoiding surfaces. Monte Carlo and molecular-dynamics studies of the problem exhibit very slow crossover effects. Such effects coupled with the difficulties to equilibrate large two-dimensional surfaces prevent the direct study of certain extreme situations. In particular, it is impossible to assess directly the asymptotic behavior of very flexible surfaces with very small excluded-volume interactions thus leaving the possibility to claim that for sufficiently small excluded-volume interactions the surfaces will remain crumpled. In this work we address the situation when such interactions are weak. Unlike the previous studies, we can both change the strength of the excluded volume parameter v, and the *range* l over which the excluded volume interactions are active, by restricting the repulsive interaction to a finite number of neighbors of each atom in the network. Since our Hamiltonian has interaction acting over a finite range (which can be gradually modified) we can trace the effect of the introduction of excluded-volume interactions step by step. In Sec. IV we describe our model and the numerical procedures. The results of the simulations are presented and analyzed in Sec. V. We show that for very small v the surfaces are crumpled. However, when vl^4 becomes of order of unity the surface becomes flat. Thus, in the limit $l \to \infty$, i.e., for true excluded-volume interactions, even infinitesimal excluded-volume interactions necessarily create *flat* membranes. In Sec. VI we consider the implications of the results and suggest directions of further theoretical study of the problem.

II. MEMBRANES—MODELS AND PROPERTIES

A polymerized membrane, or tethered surface, can be thought of as a two-dimensional network of monomers ("atoms") embedded in a three-dimensional space. Each atom i of such a network can be labeled by a two-dimensional index (or vector $\mathbf{x}$) indicating its position in

the network, and the spatial conformation of the membrane is described by a collection of three-dimensional vectors $\vec{r}(\mathbf{x})$ each of which describes the position of some atom $\mathbf{x}$ of the network. For the purposes of theoretical study it is convenient to generalize the model to a D-dimensional manifold embedded in a d-dimensional space. The most important property of such networks is their *fixed connectivity,* i.e., the unbreakable bonds between the nearest neighbors of the network.

A rough measure of the spatial extent of a membrane, or some other polymeric object, is provided by its shape tensor

$$T_{\alpha\beta}=\frac{1}{N}\sum_{i=1}^{N} r_{i\alpha}r_{i\beta}-\frac{1}{N^2}\sum_{i=1}^{N} r_{i\alpha}\sum_{i=1}^{N} r_{i\beta}\ , \tag{1}$$

where $r_{i\alpha}$ is the Cartesian component of the position vector of the ith atom. The trace of this tensor is denoted as the squared radius of gyrations R_g^2, and is equal to the mean-squared distance of the atoms from the center of mass of the membrane. In statistical-mechanical treatment of the manifolds one considers objects of (internal) linear extent L, i.e., containing $N\sim L^D$ atoms. Usually, one expects a power-law relation between L and the thermally averaged R_g^2, or the ordered eigenvalues of the shape tensor denoted $\lambda_1>\lambda_2>\cdots>\lambda_d$. The critical exponent ν can be defined from the relation

$$R_g^2\sim\lambda_1\sim L^{2\nu}\ . \tag{2}$$

Other eigenvalues of $T_{\alpha\beta}$ scale with exponents which do not exceed ν. In a particular case of $\nu=1$, when $\lambda_1\sim\lambda_2\sim\cdots\sim\lambda_D\sim L^2$, the configuration will be denoted as stretched or flat. In such a case one can introduce additional exponents describing the power-law scaling of the remaining eigenvalues $\lambda_{D+1},\ldots,\lambda_d$. All the cases at which $\nu<1$ will be denoted as "crumpled configurations." Obviously, the above notation does not exhaust all the possibilities, but it will suffice for the purposes of this work.

A model membrane in which the fixed connectivity has been enforced by appropriate nearest-neighbor interactions of the atoms in the network, but without any additional interactions, will be denoted as a *flexible phantom* membrane. (Here, and in the rest of the paper, the term "neighborhood" refers only to the *internal distances along the network.* It is *not* related to the positions of the atoms in the embedding space.) In the theoretical treatment of membranes it is convenient to treat the internal D-dimensional position vector $\mathbf{x}$ as a continuous variable. It is believed that the long length-scale properties should not depend on the fact that the microscopic Hamiltonian describes a system of discrete atoms. The continuous-$\mathbf{x}$ description of the problem therefore eliminates the explicit introduction of the microscopic scale. Such a description can be justified *a posteriori,* although there are no exact arguments which would allow such a description.

It has been demonstrated both numerically [9,16] and by an approximate analytical treatment [9] that the long length-scale properties of a flexible phantom membrane can be represented by a continuum Hamiltonian

$$\frac{\mathcal{H}_0}{k_BT}=\tfrac{1}{2}K\int(\nabla\vec{r})^2d^D\mathbf{x}\ , \tag{3}$$

where $(\nabla\vec{r})^2\equiv\sum_{\alpha=1}^{D}(\partial\vec{r}/\partial x_\alpha)^2$, while the "force constant" K has entropic origin and is approximately equal to the inverse of the mean squared distance (in the embedding space) between the (internally) neighboring atoms of the network. The Hamiltonian (3) is exactly solvable: It can be shown that for such a membrane $R_g^2\sim(1/K)\ln L$, and therefore $\nu=0$. Such a model membrane infinitely overfills the embedding space, and is obviously very remote from any realistic description of membranes.

Physical membranes cannot self-intersect. This property of self-avoidance, or excluded volume, is introduced into model systems via a short-range repulsive potential acting between *any* pair of atoms, independently on their internal distance in the network. The model repulsive potentials are usually either hard-sphere potentials, or somewhat "softer" strong inverse power-law potentials. In the theoretical treatment, the excluded-volume interactions are introduced by adding to (3) a term

$$\frac{\mathcal{H}_v}{k_BT}=\frac{v}{2}\int\int\delta^d(\vec{r}(\mathbf{x})-\vec{r}(\mathbf{x}'))d^D\mathbf{x}\,d^D\mathbf{x}'\ . \tag{4}$$

This is an obvious generalization of Edwards' Hamiltonian [17], which has been extensively used in the description of linear ($D=1$) polymers. It should be noted that the choice of the range of the repulsive potential can either completely prevent the self-crossing of the membrane, or leave it as an energetically costly possibility. A complete prevention of self-crossing of linear polymers may somewhat modify their dynamical behavior but has no influence on the thermodynamic equilibrium. For ring polymers such differences are more important, but, again, do not modify the important critical properties. By analogy with linear polymers, it is believed that the presence or absence of occasional self-intersections should not influence the long length-scale properties, although no systematic study of this point has been performed. In particular, the form (4) completely disregards this issue since it only preserves the correct second virial coefficient of interactions, and disregards other details of the microscopic repulsion. The virial coefficient and the parameter v are proportional $B\equiv\int(1-e^{-\mathcal{V}/k_BT})d^d\mathbf{r}$, where $\mathcal{V}$ is the interaction potential between monomers of the network. In self-avoiding polymers B (and v) are positive. Negative v in (4) of a system leads to a collapsed state. In this case, additional terms (such a repulsive many-point interactions) must be introduced into the continuum Hamiltonian to produce a realistic description of the membrane.

Introduction of bending rigidity into the membrane models may modify their long-range behavior: Rigid membranes are asymptotically flat [7]. Unusual properties of the flat phase (length-scale dependence of the in-plane elastic constants, and transverse fluctuations) have been extensively investigated during recent years [18]. As the rigidity of a membrane decreases (or the temperature increases), a phantom membrane undergoes a second-order phase transition [8,19] from an asymptotically flat

to a crumpled state. This transition has been investigated using renormalization-group methods [20]. Unfortunately, these methods rely on $1/d$ expansions, which are unable to provide reliable estimates for the $D=2$, $d=3$ case. Paczuski, Kardar, and Nelson [21] analyzed the crumpling transition in the presence of both bending rigidity and excluded-volume interactions within the framework of Landau-Ginzburg Hamiltonian, and concluded that the excluded-volume interactions are irrelevant at the flat phase until the transitions point. A numerical check of the relevance of the excluded-volume interactions at the transition point of a phantom membrane indicates [19] that such interactions should be relevant. There are no theoretical indications regarding the nature of the change which excluded-volume interactions can introduce into the crumpling transition

III. EXCLUDED-VOLUME EFFECTS

The first investigation [1,9] of tethered membranes with excluded-volume interactions was performed for a small system ($N=121$) of atoms. The repulsive interaction was a simple hard-sphere repulsion. That study found $\nu \approx 0.83$. Since for regular ($D=2$) membranes the excluded-volume interactions are relevant at any space dimension d, an ϵ expansion was developed in which both D and d are used as continuous variables, and the expansion is performed in the D-d plane [22]. While these expansions demonstrated the feasibility of the process, they did not provide an estimate of ν, since they have been performed only to the lowest order in ϵ. Some formal aspects of such treatment (such as renormalizability to the lowest order, and renormalizability of simpler models to all orders) have been checked [23]. None of these treatments was able to indicate anything pathological happening to two-dimensional membranes in $d=3$.

Soon after the first numerical studies of the crumpling transition in phantom membranes [8,19], an attempt was made to investigate that transition in the presence of excluded-volume interactions [10]. Surprisingly, it was found that the transition had completely disappeared, and the surfaces were flat for any value of the bending constant. This study found that λ_1 increases as L^{ν}, with $\nu \approx 0.96$, while λ_3 (the smallest eigenvalue of the shape tensor) increases as L^{ζ}, with $\zeta \approx 0.65$. The natural conclusion was that the surface remains asymptotically flat ($\nu=1$) with strong transverse fluctuations. The same result has been obtained in a large-scale molecular-dynamics simulation [11] of a system containing $N=4219$ atoms. The discrepancy between these results and Ref. [1] was explained by the fact that the original study used very small systems, where the crossover effects were strong, and, in addition, the measurement of $R_g^2=\lambda_1+\lambda_2+\lambda_3$, instead of separate evaluation of λ_i's, enhanced the crossover effects, since R_g^2 is a sum of three terms, the smallest of which increases slowly with L. These studies made it clear that the effects of the excluded volume interactions are more subtle than was thought before. Since it is impossible to determine numerically whether the exponent ν is *exactly* unity or somewhat smaller, the subsequent studies concentrated on checking the consistency of the concept of flat surface. Boal *et al.* [12] have shown that, starting from a folded initial configuration, the surfaces tend to unfold during the equilibration. An indirect support of the possibility to have "stiffness" induced by excluded-volume interactions is also provided by simulations [24] of certain networks with fractal [25] connectivity.

An interesting argument has been advanced [15] to explain the failure of numerical simulations to observe the crumpled phase of membranes: an excluded-volume interaction between nearest-neighbor atoms locally restricts the bending of the membrane and therefore acts as a bending force constant. This effective rigidity is proportional to the temperature, and therefore the change of the temperature does not modify the strength of the effect. Thus bending rigidity which had been accidentally introduced into the model systems prevented creation of the crumpled phase. Indeed, the "diameter" of atoms is usually not much smaller than the distance between the neighboring atoms, and therefore the induced effective rigidity is large enough to bring the entire system into the "usual" flat phase. Such an explanation attributes the presence of the flat phase to the bending forces, and *not* to the excluded-volume interactions, and therefore the appearance of the flat phase *is an artifact* of the computer model rather than an essential property of excluded-volume interactions. Bending forces can be reduced by, e.g., diluting the two-dimensional network. Simulations of diluted networks [26] failed to recover the crumpled state.

A different computational method of enforcing self-avoidance in model systems is using a triangulated surface at which no atom (vertex) can cross a plane of a triangle created by any three neighboring atoms [27]. This can be represented as a potential which depends on the positions of *four* atoms. While this is very different from the two-atom repulsive potentials, the asymptotic behavior is expected to be unmodified, since, in the renormalization-group language, the two-particle potential will be generated under the rescaling. However, for such models one may expect severe crossover effects. In particular, a modification of boundary conditions can produce very different results [27]. A detailed analysis of such a model [28] demonstrates that the membranes are flat, although the crossovers are extremely slow.

Due to the difficulties in reaching a definite conclusion in $d=3$ it is important to consider higher-dimensional cases. Recently, Grest [13] found that $\nu=1$ in $d=3$, $\nu=0.95\pm0.05$ in $d=4$, and $\nu=0.85\pm0.05$ in $d=5$. The picture arising from these results is as follows: the exponent ν increases with decreasing d and reaches the value of $\nu=1$ at (or near) $d=d_l\approx 4$. Below this "lower critical dimension" the two-dimensional membranes are flat. Not surprisingly, for $d<d_l$ the behavior of the membrane cannot be explained in such a simple manner as for $d>d_l$. (This resembles the problems of *linear* polymers with power-law interaction below their lower critical dimension [29].) Although this explanation does not contradict the "effective rigidity" argument, it states that the flat phase is *not* an artifact of a computer model, but will always be generated by excluded-volume interac-

tions. Heuristic arguments regarding the existence of d_l have been advanced by several authors ($d_l=3$ in Ref. [30], $d_l=4$ in Ref. [31]). Unfortunately, those conjectures are not based on exact analysis, and cannot deduce the value of d_l with any degree of certainty.

While most of the numerical studies reached the conclusion that self-avoiding surfaces are flat, there are several important studies with opposite indications: In an experimental study of graphite oxide sheets in an aqueous solution [5] Hwa and co-workers succeeded by changing the contents of acetone in the solution to observe compact ($\nu=\frac{2}{3}$) and crumpled ($\nu=0.79\pm0.03$) phases. Two recent numerical studies [14,15] considered membranes, which besides the excluded volume interactions contained an interparticle (short-range) attraction term. In these models flat, crumpled and compact phases were seen for various values of temperature. However, the temperatures at which crumpled and/or compact phases were observed corresponded to negative B, i.e., *negative* v in (4). Therefore these results are examples of a more complicated case, which cannot be represented by the simple Edwards-type Hamiltonian.

IV. MODEL SYSTEM AND SIMULATION PROCEDURE

While the results in high d indicate that two-dimensional membranes in $d=3$ should be flat, it is difficult to ascertain this claim *directly* in $d=3$ due to various computational artifacts of the models. The natural thing would be usage of a model which is described as closely as possible by the Hamiltonian $\mathcal{H}_0+\mathcal{H}_v$ given by (3) and (4) without any additional terms. It is known [9] that any simple nearest-neighbor interaction produces (3) already at length scales of few lattice distances. Therefore we use a simple potential

$$\mathcal{V}_{\rm NN}(\vec{r})=\begin{cases}0 & \text{for } r<b\\ \infty & \text{for } r>b\ ,\end{cases} \tag{5}$$

to describe the nearest-neighbor bond. Such a choice corresponds to (3) with $K\approx 1/b^2$. The excluded-volume interaction can now be introduced by a hard-sphere repulsion potential

$$\mathcal{V}_{\rm rep}(\vec{r})=\begin{cases}\infty & \text{for } r<\sigma\\ 0 & \text{for } r>\sigma\ .\end{cases} \tag{6}$$

As long as the parameter σ/b is small, the description of the repulsive part should be close to (4). In what follows, we measure the spatial distances in units of maximal nearest neighbor distance, thus setting $b=1$, while σ becomes dimensionless parameter. When $\sigma\ll 1$ no additional effective interactions are generated on the length-scales of several lattice distances.

So far the model does not differ from most of the models used to describe self-avoiding surfaces. Unfortunately, when $\sigma\ll 1$ it is more difficult to establish the long length-scale behavior. In particular, in Ref. [12] the authors encountered some difficulties in determining whether the crumpled phase and the crumpling transition exist for very small σ. To overcome those difficulties we considered a variant of the problem, where the excluded volume interactions have been enforced only up to a certain neighborhood level: We considered a two-dimensional triangular array of atoms embedded in $d=3$. In an infinite array every atom has six nearest neighbors [one (internal) lattice unit away from that atom] six next-nearest neighbors ($\sqrt{3}$ units away), etc. Thus all neighbors can be grouped into levels which are a particular distance away from a given atom. If we restrict the repulsive interaction (6) to act only between atoms whose *internal* distance along the lattice does not exceed a certain number, we will have a model with *local* interactions. The (internal) range l of the interactions will be defined as the square root of the number of atoms in the interacting neighborhood, e.g., if nearest and next-nearest neighbors interact then $l^2=1+6+6=13$. Obviously, these are not true excluded-volume interactions, and on length scales larger than l the system could be represented by a continuous *local* Hamiltonian

For any finite l, and $\sigma=0$, we have the usual crumpled flexible phantom membrane. As σ increases, the effective rigidity of the system also increases and at some particular value $\sigma_c(l)$ the surface will undergo a transition into the flat state. Since the interactions are *local,* this will be the *regular* [(un)crumpling] transition. Transitions of this type can be relatively simply detected [19]. The main aim of this work is to determine the dependence of σ_c on l as the range of the interaction grows.

The simulation has been performed on a surface which was a hexagon excised from a triangular lattice embedded in three-dimensional space. We used hexagons of two sizes: 14 and 20 lattice spacings between the opposite corners, i.e., they were $L=15$ and 21 atoms across, and contained the total of $N=169$ and 331 atoms, respectively. These are systems of moderate size, which both provide the ability to see the asymptotic behavior and can be well equilibrated. Since all the potentials of the model are either 0 or ∞, the Monte Carlo (MC) procedure is independent of temperature: Every attempted move is either permitted, and therefore accepted, or forbidden by the infinite potential, and therefore rejected.

Usually, an elementary move in a MC procedure consists of random selection of an atom and an attempt to move it in a random direction by certain amount s. The equilibration time is determined by the slowest modes which correspond to large length-scale motions. We made an attempt to accelerate the equilibration procedure by making the following "collective" moves: The procedure begins by a random choice of an atom of the network. Then a random decision is made regarding how many shells of neighbors will be moved together with that atom. All the atoms are moved in the same randomly chosen direction. The atom in the center of the moving block is moved by some amount s_0, while rest of the atoms are moved by smaller amounts s determined by their (internal) distance h from the center of the block: $s=s_0(1-h/h_0)$, where h_0 is the (internal) distance of the atom just outside the moving block. While this procedure reminds numerous cluster motion algorithms which have been recently developed to accelerate the

equilibration processes, it suffers from a serious drawback: As the size of the moving cluster increases, so does the probability that the move is forbidden because one of the atoms violates one of the potential restrictions. This problem was partially mended by making s_0 dependent on the number of atoms n inside the moving block: We found that a choice $s_0 = s_* / \sqrt{n}$ enabled us to keep the move acceptance rate almost independent of the size of the moving block. With $s_* = 0.2$ we were able to maintain the acceptance rate of at least $\frac{1}{2}$ for all our simulations.

In standard MC procedures a single atom is moved by an amount s_*, and therefore the center of mass of the object moves by amount s_*/N. In our procedure an elementary move consists of an attempt to move n atoms leading to a displacement of the center of mass by an amount $s_* \sqrt{n}/N$. We define the MC time unit as a time required to perform N elementary moves. In these units the diffusion constant of the center of mass of the object will be $s_*^2 n/N$. The extra factor n, which should be taken as an average n used in the procedure, shows that diffusion process is accelerated, and therefore the relaxation times (which can be estimated as times needed for the object to diffuse its own R_g) are decreased. Since the algorithm allows partial vectorization it is indeed advantageous in use on parallel computers. For scalar computations the advantage of such method is not clear, since the time consumed by an elementary move also increases.

For each choice of parameters σ and l we performed equilibration process over times ranging between $8N^2$ and $40N^2$ MC time units. Since R_g^2 of the systems in this simulation did not exceed 4, the equilibration time exceeded the time τ_0 required for the system to diffuse its own R_g by more than an order of magnitude. We expect this time to suffice for reaching equilibrium and for collection of reliable statistics (e.g., measurement of R_g^2 with at least 10% accuracy). The time τ_0 does not necessarily provide a good estimate of the equilibration time, e.g., near the crumpling transition we can expect a critical slowing down of the processes. Therefore we verified the equilibration by a direct measurement of several autocorrelation functions. Figure 1 depicts an example of such measurement for $L = 15$ surface with interaction range $l = \sqrt{13}$. The solid and dotted lines correspond to the autocorrelation function $A_\lambda(t)$ of the largest eigenvalue (λ_1) of the shape tensor for $\sigma = 0.35$ and $\sigma = 0.25$, respectively. The autocorrelation function is defined as $A_\lambda(t) \equiv (\langle \lambda_1(t'+t)\lambda_1(t') \rangle - \langle \lambda_1(t') \rangle^2) / \langle \lambda_1(t')^2 \rangle$, where the averaging $\langle\ \rangle$ is performed over the time t'. Both values of σ are close to σ_c, thus representing the worst situation when the system is in the intermediate shape between the crumpled and flat conformations. Even in this case we see equilibration within few times N^2 MC time units. The dashed and dot-dashed lines in Fig. 1 represent (for the same respective pair of σ's) the autocorrelation function $A_{\mathbf{v}}(t)$ of a vector $\mathbf{v}$ which connects to opposite corners of the hexagon. The function is defined as $A_{\mathbf{v}}(t) \equiv \langle \mathbf{v}(t'+t) \cdot \mathbf{v}(t') \rangle / \langle \mathbf{v}(t')^2 \rangle$ and measures the rotation of the entire surface. It decays to zero when the original orientation is "forgotten." Notice that,

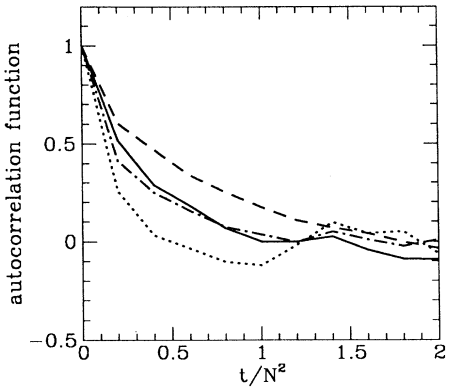

FIG. 1. Time dependence of normalized autocorrelation functions for $L = 15$ surface with interaction range $l^2 = 13$. Solid and dotted lines depict A_λ (see text) for sphere diameters 0.35 and 0.25, respectively. Dashed and dot-dashed lines represent A_v for the same values of diameters, respectively.

for larger σ and therefore flatter conformations, the rotation is slower. Similar behavior was observed in several additional autocorrelation functions, leading us to the conclusion that our results represent thermally equilibrated averages with reasonable accuracy.

V. RESULTS

For hexagon sizes $L = 15$ and 21 we performed MC equilibrations for four different ranges l of the repulsive interactions. We chose $l^2 = 13$, 19, 37, and 55. For fixed l we performed simulations for a large sequence of values of the diameter σ of the repulsive potential in order to locate the transition point from the crumpled to flat state. As the parameter σ increases, reasonably sharp transition can be seen. Figure 2 depicts the dependence of λ_i's on

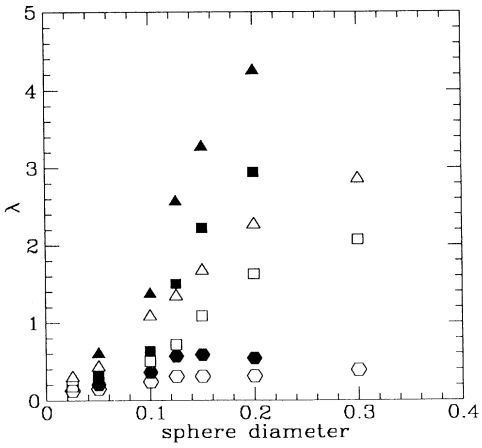

FIG. 2. Dependence of largest (triangles), intermediate (squares), and smallest (hexagons) eigenvalues of the shape tensor on the sphere diameter, for $L = 15$ (open symbols) and $L = 21$ (solid symbols) for interaction range $l^2 = 37$.

the sphere diameter σ for $l^2=37$. The open and solid symbols represent $L=15$ and 21 cases, respectively. We notice that the smallest eigenvalue of the shape tensor λ_3 shows very little dependence on σ, while two remaining eigenvalues increase by a significant factor in the range of parameters $0.1<\sigma<0.2$. Within this range, the values of λ_1 and λ_2 change from values typical of crumpled phantom membranes ($\sim \ln L$) to values typical of flat membranes ($\sim L^2$). The transition is, obviously, sharper for larger membranes. Similar behavior is observed for other values of l. However, the "sharpness" of the transition does depend on l: for $l=13$ the transition is very sharp and can therefore be found with large accuracy, while for larger l's the finite size effects become more severe, and it is more difficult to determine the position of σ_c.

The transition from the crumpled to flat state can be easily seen by visually inspecting the spatial conformations of the membranes. Figure 3 depicts such conformations of the membrane which have been quantitatively described in Fig. 2. Figure 3(a) shows the conformation in the crumpled state for $\sigma=0.1$. This picture is visually indistinguishable from pictures which we have for $\sigma<0.1$, i.e., it is strongly crumpled. Figure 3(b) depicts the $\sigma=0.2$ case, which is close to the estimated transition point. The configuration in Fig. 3(c) corresponds to $\sigma=0.3$, i.e., above the transition point, and well in the flat phase.

As can be seen from Fig. 2, the sharpest signature of the transition point is found in the behavior of the largest eigenvalue $\lambda_{\max}=\lambda_1$ of the shape tensor. Since the exact determination of the transition point is difficult both due to statistical errors and due to rather strong finite size effects, we attempted to find proper variable in terms of which the graphs for different l's collapse to a single line, at least in the neighborhood of the transition point. There is no theory describing the buildup of the effective bending constants from excluded-volume interactions under rescaling. However, we can get a clue by inspecting the importance of weak excluded-volume interaction on various length scales. It has been shown [9] that for a finite membrane of linear size L with small excluded-volume parameter v, the importance of the interaction grows with increasing L. In particular, the membrane can no longer be considered as a phantom membrane (in $d=3$) if $K^{3/2}vL^4$ exceeds unity.

Since the repulsive interactions of our model are limited to internal distance l, we will define (in our dimensionless units) a parameter $p\equiv vl^4$, where $v\equiv\sigma^3$, which serves as a natural indicator of the importance of the repulsion within the range in which it is active. Figure 4 depicts the dependence of $\lambda_{\max}$ on this parameter for all values of l and for both system sizes L. Plots of this type cannot collapse the data for two different values of L, since the small-p end of the graph has a different functional dependence on L from the large-p end. Thus the solid symbols ($L=21$) should be inspected separately from the open symbols ($L=15$). (The values of $\lambda_{\max}$ for $L=21$ have been multiplied by $\frac{3}{5}$ to bring them to comparable values with the results for $L=15$.) Although no good collapse is seen, we can make few observations: (a) the transition region indeed appears for p of order of unity; (b) the spread of the data points in the transition region (between $\log_{10}p=0$ and $\log_{10}p=1$) is smaller for larger L, i.e., the collapse somewhat improves with increasing L; (c) if, instead of using p we would use directly the excluded-volume parameter v, the transition points would spread out over more than two decades. Statistical errors do not permit an accurate determination of the "best scaling variable": The data collapse is almost as good (or, "as bad") for the combination vl^5. However, for the choices

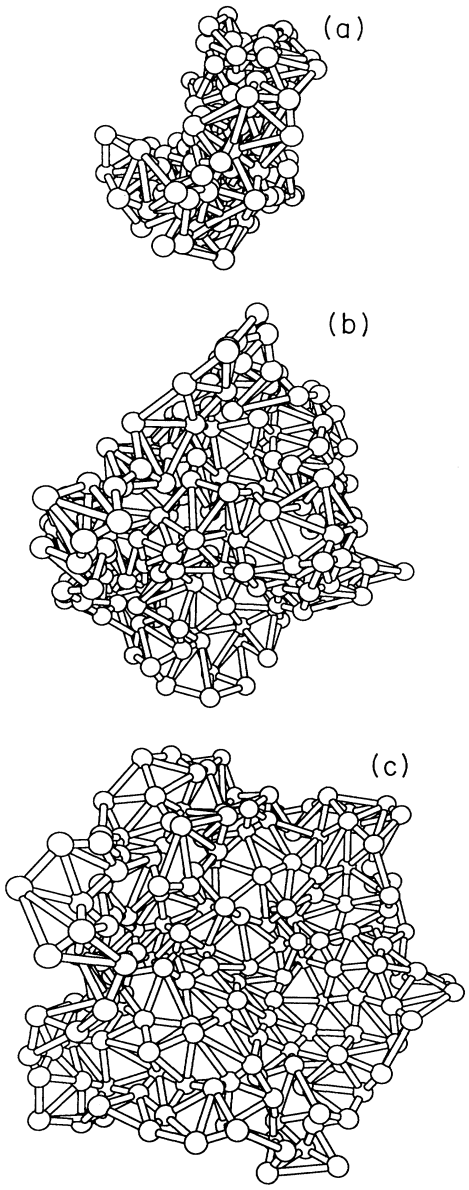

FIG. 3. Spatial conformations of membranes with $L=21$ and $l^2=37$ for sphere diameters (a) 0.10 (crumpled phase), (b) 0.20 (close to the transition point), and (c) 0.30 (flat phase). Spheres represent positions of the atoms; the depicted diameter of the spheres is not related to the range of the repulsive interactions. Cylinders represent the bonds between the nearest-neighbor atoms.

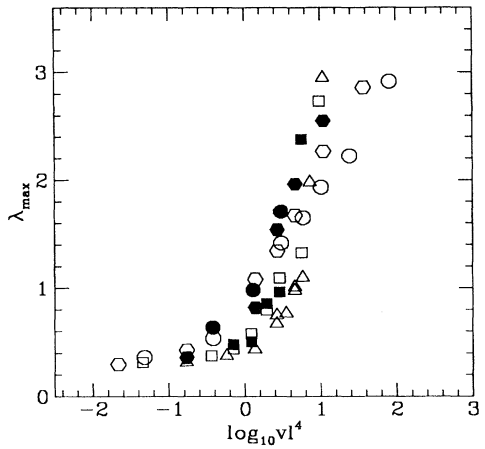

FIG. 4. Semilogarithmic plot of the dependence of the maximal eigenvalue of the shape tensor on the scaling parameter vl^4 (see text). Triangles, squares, hexagons, and circles correspond to $l^2=13$, 19, 37, and 55, respectively. Open symbols represent the results for $L=14$, while the solid symbols represent $L=21$. The values of λ_{max} for $L=21$ have been reduced by a factor of $\frac{5}{3}$ to bring them to a similar scale as the results for $L=15$.

vl^3 or vl^6 the quality of the graph deteriorates significantly. Despite this uncertainty in the value of power of l it is obvious that the critical value of v for which the transition from the crumpled to flat phase appears decreases with increasing l (as fourth or fifth power of l^{-1}).

Assuming that the transition indeed occurs when $vl^4=\sigma^3 l^4$ reaches value of order unity, we may attempt to plot our estimates of the critical sphere diameter σ_c as a function of $l^{-4/3}$. Such a plot suffers from all the uncertainties evident from Fig. 4. We arbitrarily defined σ_c as the value for which λ_{max} reaches $\frac{2}{3}$ of its maximal possible value. This value can be determined only with poor accuracy, due to statistical fluctuations in the data points and due to uncertainty regarding the extrapolation of the result to $L\to\infty$, since only two values of L are available. In our (subjective) estimate these errors are smaller than 10% for $l^2=13$, and may be as large as 20% for $l^2=55$. Figure 5 depicts the l dependence of such an estimate of σ_c. Despite the uncertainties in the determination of the points of this graph the general trait is obvious: $\sigma_c\to 0$ for $l\to\infty$.

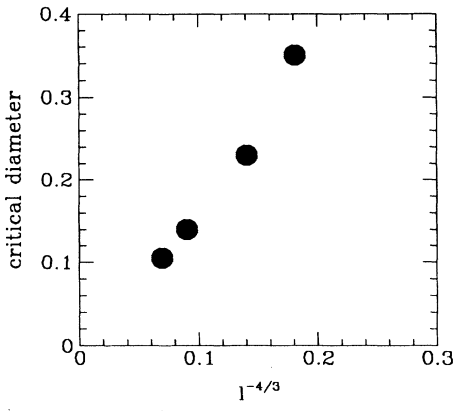

FIG. 5. Estimated critical sphere diameter as a function of the interaction range l.

VI. DISCUSSION

In this work we attempted to investigate self-avoiding surfaces with $K\approx 1$, and with very small excluded volume. In such a model, all other interactions are negligible on the length scales of few lattice spacings. By limiting the range l over which the repulsive interactions are active we converted the original problem into a sequence of problems with *local* interactions which gradually converges to the true excluded-volume interaction case. By investigating the sequence of crumpling transition points we have shown that self-avoiding surfaces are flat even for very small values of excluded volume v, when the system is correctly represented by the generalized Edwards Hamiltonian. Flatness of the surface therefore is *not* an artifact of computer models but an essential property of a self-avoiding surface.

An approximate picture of surface becoming flat emerges: On short length scales excluded-volume interactions are not felt, and the surface is crumpled like a regular flexible phantom membrane. At certain length scales excluded-volume begins to significantly modify the shape of the phantom object. At that scale excluded-volume interactions already produce effective rigidity which suffices to keep the entire surface flat. Beyond that length scale the surface remains flat because of its rigidity, and self-avoiding interactions no longer play a role. It is reasonable that the repulsive interaction will not play a significant role until $p=vl^4\approx 1$. However, it is far from being obvious why the crumpling transition should appear exactly at the same scale. Our results are not sensitive enough to exclude the possibility that the transition appears for v somewhat larger or smaller than predicted from the relation $p\approx 1$.

These results are specific to $d=3$, which is probably below the lower critical dimension of "polymerlike" behavior of the surfaces. We should keep in mind, however, that these results *do not* exclude existence of crumpled configurations of surfaces with attractive interactions. Such crumpled states are *not* analogous to the usual (crumpled) state of self-avoiding linear polymers, and cannot be simply investigated using an Edwards-type Hamiltonian. Since existence of nonflat states has been demonstrated both experimentally, and numerically it would be interesting to build a proper theoretical description of those phenomena. However, these phenomena probably cannot be described in the language familiar from linear polymer physics (θ point and similar concepts), i.e., *real* self-avoiding surfaces require a more complicated theoretical description.

Despite recent progress in the investigation of self-avoiding surfaces, a wide gap seems to have opened be-

tween the theory and numerical simulations. Several important problems have to be addressed by the theory: (a) We need to find a way to determine the lower critical dimension of the (polymerlike) self-avoiding membrane problem, and (b) below the critical dimension we need to find a proper way to describe the buildup of bending forces by the excluded volume interactions.

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[1] Y. Kantor, M. Kardar, and D. R. Nelson, Phys. Rev. Lett. **57**, 791 (1986).
[2] P. G. de Gennes, *Scaling Concepts in Polymer Physics* (Cornell University Press, Ithaca, 1979).
[3] *Statistical Mechanics of Membranes and Surfaces,* Proceedings of the Fifth Jerusalem Winter School for Theoretical Physics, edited by D. R. Nelson, T. Piran, and S. Weinberg (World Scientific, Singapore, 1989).
[4] A. Elgsaeter, B. T. Stokke, A. Mikkelsen, and D. Branton, Science **234**, 1217 (1986); C. F. Schmidt, K. Svoboda, N. Lei, I. B. Petsche, L. E. Berman, C. R. Safinya, and G. S. Grest, *ibid.* **259**, 952 (1993).
[5] T. Hwa, E. Kokufuta, and T. Tanaka, Phys. Rev. A **44**, 2231 (1991); X. Wen, C. W. Garland, T. Hwa, M. Kardar, E. Kokufuta, Y. Li, M. Orkisz, and T. Tanaka, Nature **355**, 426 (1990).
[6] S. I. Stupp, S. Son, H. C. Lin, and L. S. Li, Science **259**, 59 (1993). See also E. L. Thomas, *ibid.* **259**, 43 (1993).
[7] D. R. Nelson and L. Peliti, J. Phys. (Paris) **48**, 1085 (1987).
[8] Y. Kantor and D. R. Nelson, Phys. Rev. Lett. **58**, 2774 (1987).
[9] Y. Kantor, M. Kardar, and D. R. Nelson, Phys. Rev. A **35**, 3056 (1987).
[10] M. Plischke and D. H. Boal, Phys. Rev. A **38**, 4943 (1989).
[11] F. F. Abraham, W. E. Rudge, and M. Plischke, Phys. Rev. Lett. **62**, 1757 (1989).
[12] D. H. Boal, E. Levinson, D. Liu, and M. Plischke, Phys. Rev. A **40**, 3292 (1989).
[13] G. S. Grest, J. Phys. (France) I **1**, 1695 (1991).
[14] D. Liu and M. Plischke, Phys. Rev. A **45**, 7139 (1992).
[15] F. F. Abraham and D. R. Nelson, J. Phys. (Paris) **51**, 2653 (1990); Science **249**, 393 (1990).
[16] Y. Kantor and M. V. Jarić, Europhys. Lett. **11**, 157 (1990).
[17] S. F. Edwards, Proc. R. Soc. London Ser. A **85**, 613 (1965).
[18] J. Aronowitz and T. C. Lubensky, Phys. Rev. Lett. **60**, 2634 (1988); E. Guitter, F. David, S. Leibler, and L. Peliti, J. Phys. (Paris) **50**, 1789 (1989); R. Lipowsky and M. Girardet, Phys. Rev. Lett. **65**, 2893 (1990); S. Leibler and A. C. Maggs, *ibid.* **63**, 406 (1989); P. Le Doussal and L. Radzihovsky, *ibid.* **69**, 1209 (1992).
[19] Y. Kantor and D. R. Nelson, Phys. Rev. A **38**, 4020 (1987).
[20] F. David and E. Guitter, Europhys. Lett. **5**, 709 (1988); M. Paczuski and M. Kardar, Phys. Rev. A **39**, 6086 (1989).
[21] M. Paczuski, M. Kardar, and D. R. Nelson, Phys. Rev. Lett. **64**, 2638 (1988).
[22] M. Kardar and D. R. Nelson, Phys. Rev. Lett. **58**, 1289 (1987); **58**, 2280(E) (1987); Phys. Rev. A **38**, 966 (1988); B. Duplantier, Phys. Rev. Lett. **58**, 2733 (1987); J. A. Aronowitz and T. C. Lubensky, Europhys. Lett. **4**, 395 (1987).
[23] B. Duplantier, Phys. Rev. Lett. **62**, 2337 (1989); B. Duplantier, T. Hwa, and M. Kardar, *ibid.* **64**, 2022 (1990); F. David, B. Duplantier, and E. Guitter, *ibid.* **70**, 2205 (1993); T. Hwa, Phys. Rev. A **41**, 1751 (1989).
[24] E. Duering and Y. Kantor, Phys. Rev. B **40**, 7443 (1989); E. Levinson, Phys. Rev. A **43**, 5233 (1991).
[25] B. B. Mandelbrot, *The Fractal Geometry of Nature* (Freeman, San Francisco, 1982).
[26] G. S. Grest and M. Murat, J. Phys. (Paris) **51**, 1415 (1990); M. Plischke and B. Fourcade, Phys. Rev. A **43**, 2056 (1991).
[27] J.-S. Ho and A. Baumgärtner, Phys. Rev. Lett. **63**, 1324 (1989); A. Baumgärtner, J. Phys. (France) I **1**, 1549 (1991).
[28] D. M. Kroll and G. Gomper (unpublished).
[29] See, e.g., Y. Kantor and M. Kardar, Europhys. Lett. **9**, 53 (1989).
[30] E. Guitter and J. Palmeri, Phys. Rev. A **45**, 734 (1992).
[31] M. Goulian, J. Phys. (France) II **1**, 1327 (1991); P. Le Doussal, J. Phys. A **25**, L469 (1992).