# Self-avoiding tethered membranes with quenched random internal disorders

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Spatial conformations of a self-avoiding tethered membrane with a quenched in-plane disorder have been studied using Monte Carlo methods. The simulations have been performed for systems with various strengths of disorder $v$ and self-avoidance $b$. (1) The membrane with ''strong'' self-avoidance and weak disorder $(b,v)=(1.7,0.3)$ is in the usual flat phase and its shrinkage is small. Even if the temperature decreases the membrane does not show any clear evidence of a large buckling. (2) As the strength of disorder increases the membrane with ''weak'' self-avoidance ($b=3.0$) shows a phase transition from a usual floppy flat phase ($v=0.0$) to a crumpled phase ($v=0.75, \nu=0.85-0.92$), passing through a new flat phase with a small roughness exponent ($v=0.52, \nu_{\perp}=0.1\pm0.1$). We discuss the implication of the result, in particular its relevance to the understanding of the wrinkling transition in partially polymerized vesicles. [S1063-651X(96)10807-2]

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## I. INTRODUCTION

The statistical properties of polymerized membranes, or tethered surfaces, have been widely discussed in the past few years [1–3]. The polymerized membrane is a two-dimensional generalization of a linear polymer. At low-temperature a membrane with bending rigidity is asymptotically flat, and its radius of gyration $R_G$ increases as the linear internal dimension $L$ of the surface [3,4]. As a function of temperature, the membrane without self-avoiding interaction (phantom membrane) shows a crumpling transition from the low-temperature flat phase to a high-temperature crumpled phase ($R_G \sim \sqrt{\ln L}$) [3]. The properties of the flat phase have been studied extensively [4–7]. It is characterized by an anomalous elasticity with shear and compression moduli ($\mu$ and $\lambda$) that vanish and by a bending rigidity ($\kappa$) that diverges with decreasing wave number.

One of the surprising characters of the membrane is that the self-avoiding tethered membrane is flat when embedded in three-dimensional space [8–10]. Abraham and Nelson [11] discussed the origin of the phenomena: Entropic bending rigidity induced by the (next-nearest-neighbor) self-avoidance causes the crumpling transition [1] and the membrane becomes flat. This means that the flat phase of the self-avoiding tethered membrane is described by the Aronowitz-Lubensky (AL) fixed point associated with the flat phase of the phantom membrane [6]. Their discussion also means that the hard-sphere model has an inevitably large bending rigidity originated from the next-nearest-neighbor interactions and that these simulation did not purely investigate the effect of self-avoidance. That is, the hard-sphere model is inevitably rigid when the self-intersection is completely prohibited. In order to study the ''genuine'' self-avoidance effect, tethered membrane with hard spheres of smaller diameter (''weak'' self-avoidance) [9,10,12], and ''plaquette'' membrane model [13,14] have been employed [15]. Even in these cases, the membrane becomes flat and it was concluded that the self-avoiding tethered membrane is flat. Theoretically, using the Gaussian variational method, Guitter and Palmeri [16], Le Doussal [17], and Goulian [18] discussed the existence region of the crumpled phase and showed that the self-avoiding tethered membrane is flat in three-dimensional space. Higher-dimensional cases were also well described in the same framework [19]. However, as these theories do not describe the buildup of bending forces by the self-avoiding interaction and do not answer why the Gaussian approximation works instead of the Flory approximation, we have a wide gap between the theories and numerical simulations [12].

Recently, studies on the effects of quenched in-plane disorders have been performed. One of the most important effects of the disorder is the buckling transition [20]. Although the stable phase of a defect-free polymerized membrane is flat, the strains induced by a defect, such as a dislocation, can be accommodated by displacements in the normal direction, resulting in the buckling of the membrane. This process, which depends on a balance between in-plane stretching energy and curvature energy, occurs when

$$K_0 l^2/\kappa > \gamma. \tag{1.1}$$

Here, $K_0$ is the Young's modulus, $\kappa$ the bending rigidity, $l$ a length scale, and $\gamma$ a dimensionless constant of order $10^2$ [20,21]. The length scale $l$ depends on the nature of the defect. For example, in membranes of size $R$, $l=R$ for disclinations and $l=\sqrt{Rb}$ for a dislocation with the Burger's vector $\vec{b}$. Thus these defects always buckle in sufficiently large membranes, irrespective of the value of the elastic constants. On the other hand, for finite energy defects such as vacancies, interstitials, or tightly bound dislocation pairs, $l$ is an order of a lattice constant and the stability of the flat phase depends on the actual values of the elastic constants. This leads to the following interesting possibility of a buckling transition in an infinite system as a function of the temperature [21]. As the temperature $T$ decreases, $\kappa$ decreases and $K_0$ increases. At some temperature $T_B$, the condition (1.1) is satisfied and the membrane shows a buckling transition. We can assume that this process is also possible even if the membrane has the self-avoiding interaction. However,

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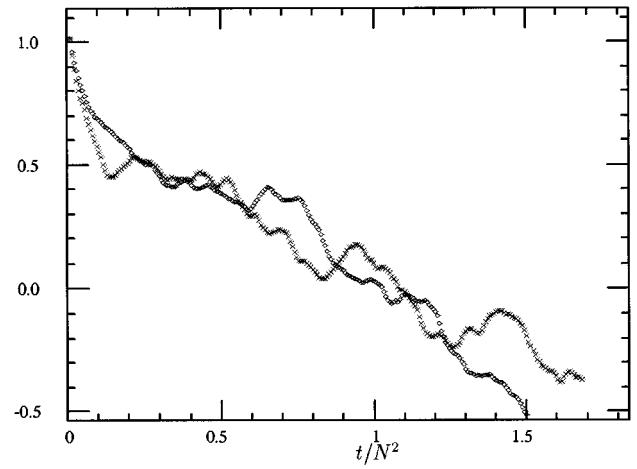


FIG. 1. Time dependence of autocorrelation $A_{Rg}$ for the radius of gyration with $N=547$. The curve ($\times$) depicts $A_{Rg}$ for $(b,v)=(3.0,0.75)$ (weak self-avoidance and strong disorder) and the curve ($\diamond$) represents $A_{Rg}$ for $(b,v)=(3.0,0.0)$ (regular hexagonal membrane).

the self-avoiding interaction does contribute to the bending rigidity as a ''remnant'' term [22]. If the interaction is too strong, the buckling condition is not satisfied and the membrane remains flat even if we quench the system to very low temperature.

Experimentally, Mutz, Bensimon, and Brienne discovered the ''wrinkling transition'' in partially polymerized lipid vesicles [23,24]. The membrane undergoes a reversible phase transition from a high-temperature phase, where the membrane is smooth and very fluctuating, to a low-temperature phase characterized by a rigid and highly wrinkled surface. Nelson and Radzihovsky [25,26] and Morse, Lubensky, and Grest [27,28] analyzed the stability of the flat phase of the (phantom) tethered membrane with randomnesses of the locally preferred metric and spontaneous curvature by the field theoretical method. It was concluded that at $T>0$ the weak short-range disorder is irrelevant and that the rigid membrane is still described by the pure flat phase AL fixed point. In order to explain the wrinkling transition, two possibilities are proposed. One is the strong disorder [29,30] and the other is the long-range correlation of the disorder [26,31], which is induced by unscreened disclinations [32]. On the other hand, the fact that the flat phase of the rigid phantom polymerized membrane is stable with respect to the weak disorders at $T>0$ implies that the flat phase of the self-avoiding tethered membrane is also stable with respect to the disorders. The situation, however, is not so simple. Mori and Wadati [33,34] discussed the existence region of the crumpled phase of the self-avoiding tethered membrane with disorders and proposed a possibility that the membrane is crumpled with $\nu=6/7$ at $d=3$.

This analysis does not contradict the above analyses of the flat phase, because it does not forbid the existence of any flat phase. That is, when the membrane is very rigid, the membrane may become flat and a crumpled phase occurs only when the effective bending rigidity of the membrane is small. In fact, Morse, *et al.* [35] carried out molecular-dynamics simulation for self-avoiding polymerized fluid membrane. The model contains disclinations and dislocations all over the membrane. They have used the hard-sphere model with a diameter that completely prohibits the self-intersections of the membrane. They concluded that the membrane is asymptotically flat and those sites with more

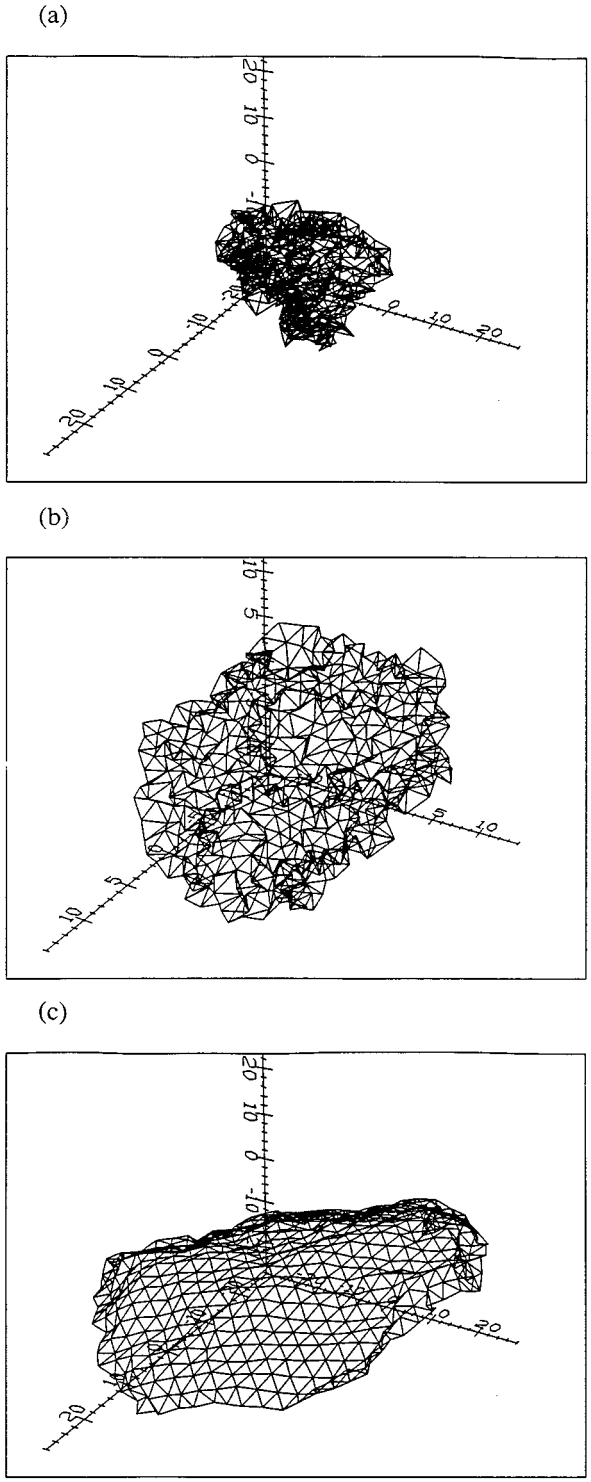


FIG. 2. (a) Typical configuration of a tethered membrane with weak self-avoidance and strong disorder $(b,v)=(3.0,0.75)$ and 547 monomers in three-dimensional space (after $20N^2$ Monte Carlo steps). (b) Typical configuration of a tethered membrane with strong self-avoidance and weak disorder $(b,v)=(1.7,0.3)$ and 547 monomers in three-dimensional space (after $6N^2$ Monte Carlo steps). (c) Typical configuration of a tethered membrane with no disorder $(b,v)=(3.0,0.0)$ and 547 monomers.

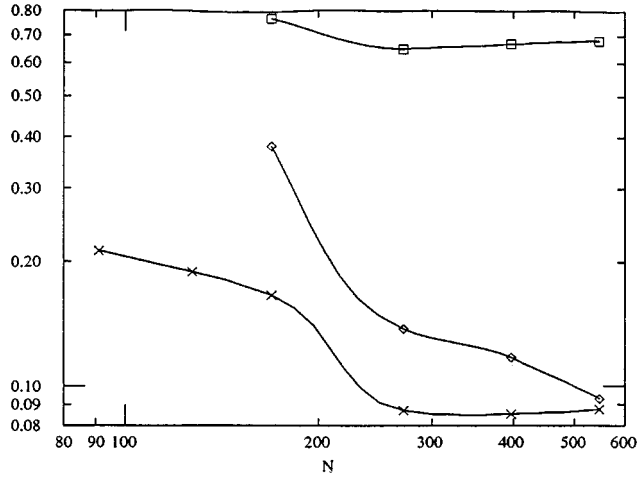


FIG. 3. The shrinkage parameter $P_{\text{shrink}}$ as a function of monomer number $N$. The curve ($\times$) depicts $P_{\text{shrink}}$ for $b=3.0$, $v=0.75$; the curve ($\diamond$) depicts $P_{\text{shrink}}$ for $b=3.0$, $v=0.52$; and the curve ($\square$) represents $P_{\text{shrink}}$ for $b=1.7$, $v=0.3$.

than six neighbors buckle only locally. In order to conclude whether the ''genuine'' self-avoiding tethered membrane with disorders can be crumpled or not, it is necessary to investigate very soft (small bending rigidity) membranes. Another motivation for studying the weak self-avoiding case is that we can expect a large conformational transformation. From the condition (1.1) the buckling transition occurs more easily when the effective bending rigidity is small. Following this point of view, we have performed Monte Carlo studies of a similar model proposed by Morse *et al.* [36]. As a tethering potential between nearest-neighbor monomers, we use the following potential

$$U_{\text{NN}}(r)=k(r^2-b^2)^2, \tag{1.2}$$

where $k$ is the force constant and the constant $b$ denotes the equilibrium bond length. We studied the model for several choices of $b$ [37,38] and a fixed value of the diameter of the hard sphere at $\sigma=1.0$. By changing the bond length $b$, we can simulate the membrane with several values of effective bending rigidity and various flexibility. The result is summarized as follows. When the reference bond length is small ($b=1.7$), the membrane is asymptotically flat and it does not show large conformational transformation, which is consistent with the result by Morse *et al.* When we take $b=3.0$ (weak self-avoidance case), the membrane shows a large shrinkage (density becomes about seven times) and it is considered to be in a crumpled phase with $\nu=0.87\pm0.02$ [38].

We note that in the above analysis we have observed a large shrinkage and a crumpled phase only in the ''weak'' self-avoiding case. There remains the possibility that such findings may be an artifact of the model, because the self-intersection is not necessarily prohibited. We cannot exclude such possibility in the hard-sphere model when we analyze the self-avoiding membrane with small effective bending rigidity. However, it is important that the membrane becomes crumpled even in the weak self-avoiding case. Up to now, almost all theoretical works dealt with the stability of the flat phase of the (phantom) rigid polymerized membrane (or stability of the AL fixed point). Even if self-intersection is not

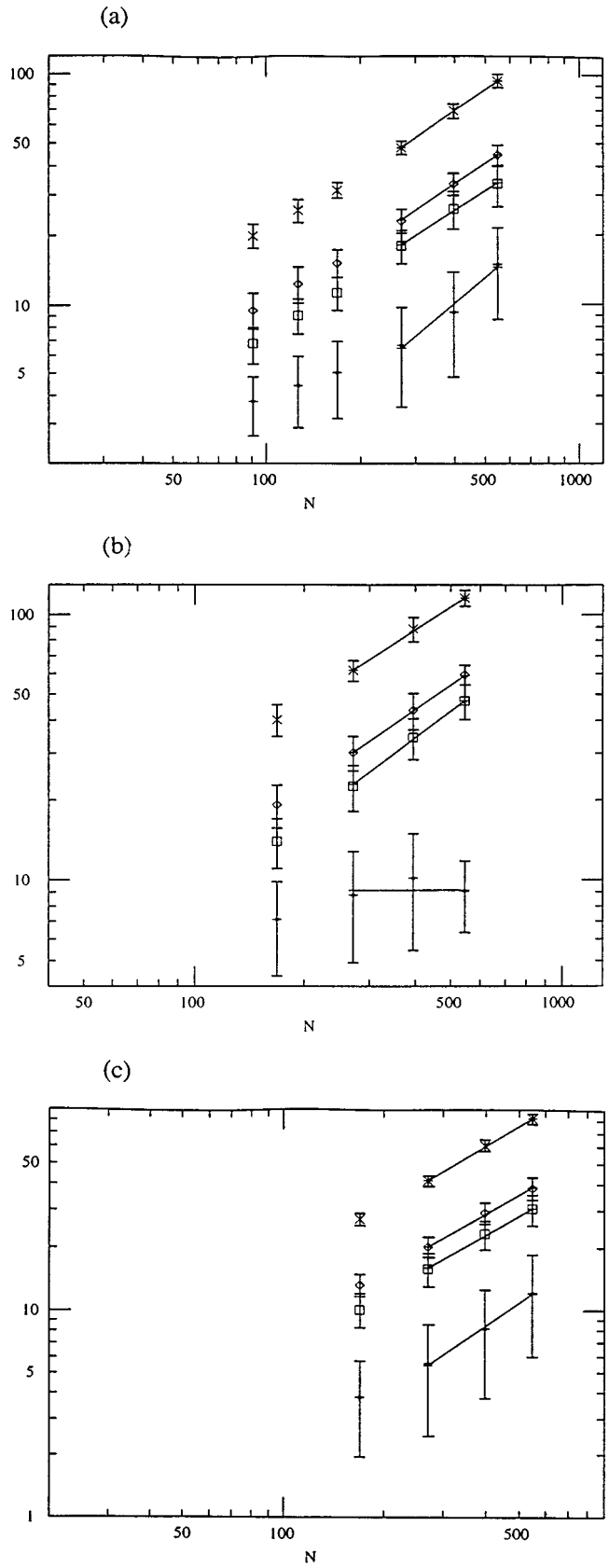


FIG. 4. Scaling plot of the mean square radius of gyration $\langle R_G^2\rangle(\times)$ and expectation values $\langle\lambda_1\rangle(\diamond)$, $\langle\lambda_2\rangle(\square)$, $\langle\lambda_3\rangle(+)$ of the eigenvalues of the moment of inertia tensor for membranes with $(b,v)=(3.0,0.75)$ (a), $(b,v)=(3.0,\sim0.52)$ (b), and $(b,v)=(1.7,0.3)$ (c). (a) The solid lines have slopes 0.94,0.92,0.88,1.17 from top to bottom; (b) 0.89,0.96,1.04,0.00; (c) 0.95,0.93,0.93,1.13.

TABLE I. Scaling exponents of $R_G$ and $\lambda_i$. These exponents are determined from the data for $L=19$, 23, and 27.

| $(b,v)$ | $\nu$ | $\nu_1$ | $\nu_2$ | $\nu_3$ |
|---|---|---|---|---|
| (1.7,0.0) | 1.00 ± 0.01 | 0.97 ± 0.03 | 1.14 ± 0.06 | 0.74±0.37 |
| (1.7,0.3) | 0.95 ± 0.01 | 0.93 ± 0.02 | 0.93 ± 0.05 | 1.13±0.36 |
| (3.0,0.0) | 1.12 ± 0.01 | 1.13 ± 0.02 | 1.32 ± 0.09 | 0.68±0.48 |
| (3.0,0.52) | 0.89 ± 0.01 | 0.96 ± 0.03 | 1.04 ± 0.05 | 0.00±0.2 |
| (3.0,0.75) | 0.94 ± 0.01 | 0.92 ± 0.02 | 0.88 ± 0.05 | 1.17±0.27 |

completely forbidden, when the usual flat phase becomes unstable in numerical studies, the AL fixed point becomes unstable by disorder. We can say that the disorders are relevant to the behavior of the membrane.

In this work, we shall report Monte Carlo studies of another model for self-avoiding tethered membrane with a quenched in-plane disorder. Introducing two kinds of monomers [big ($p_i=1$) and small ($p_i=-1$)], which are chosen at random on the network, the reference length between the $i$th and $j$th monomer $b_{ij}$ varies among monomer pairs by the rule $b_{ij}=b+(p_i+p_j)v$. With the model we can change the strength of disorder at our convenience. This disorder has a short-range correlation and it is the same with the disorder in the polymerized fluid membrane [39]. Phantom case of the model with bending rigidity was at first studied by Kantor [40] and at low temperature the shape of the membrane deviated from a flat configuration and it settled into a partially folded ground state. We shall study the effect of self-avoidance on this model without bending rigidity apart from that induced by the self-avoidance. Especially, when the bending rigidity of the membrane is small or the diameter of the hard sphere is small (''weak'' self-avoidance), we like to see whether the membrane is crumpled or not and whether the flat phase becomes unstable or not. We also like to see whether the membrane shows the buckling transition as temperature decreases when the membrane is flat at high temperature [20,21]. The outline is as follows. At first, we study the model at a fixed temperature with (1) ''strong'' self-avoidance and weak disorder $(b,v)=(1.7,0.3)$ and (2) ''weak'' self-avoidance and several strengths of disorder $(b=3.0)$. We note that, in these models we do not prohibit completely the self-intersection even in the case (1) and that the name ''strong'' is not truly strong. Then, in the case (1), where the membrane becomes asymptotically flat at some temperature, we study the quenching process and see whether there occurs a large conformational transformation.

The paper consists of the following. In Sec. II, we describe the model and numerical procedures. The results of the simulations are presented and analyzed in Sec. III. The membrane with weak self-avoidance $(b=3.0)$ shows large shrinkage even if the strength of disorder is small $(v\sim 0.2)$. The membrane with weak disorder $(v\sim 0.52)$ is asymptotically flat and roughness exponent $\nu_\perp$ is extraordinarily small $(\nu_\perp \simeq 0.1\pm 0.1)$. The membrane with strong disorder $(v=0.75)$ seems to be crumpled and the exponent for the radius of gyration $R_G$ is 0.85–0.92. The membrane with strong self-avoidance and weak disorder $(b,v)=(1.7,0.3)$ is asymptotically flat and its relative shrinkage as compared with the membrane without disorder is small. Even if the temperature decreases, the shape of the membrane does not show large folding, which is observed in the phantom case. On the contrary, the membrane becomes more and more flat. In Sec. IV, we discuss the implications of the results and suggest directions of further studies.

## II. MODEL SYSTEM AND SIMULATION PROCEDURE

The model that we study consists of hard spheres with diameter $\sigma$ connected in a two-dimensional triangular array embedded in a three-dimensional space. The position of the $i$th atom in such a network is denoted by $\vec{r}_i$. In the simulation a hexagonal sheet ($L\leqslant 27$ monomers across) with $N=(3L^2+1)/4$ monomers excised from the triangular lattice has been used. The connectivity of the system is fixed by keeping nearest-neighbor atoms on the lattice connected by a tethering potential [41]

$$U_{\rm NN}(r_{ij})=k({r_{ij}}^2-{b_{ij}}^2)^2. \tag{2.1}$$

Here $r_{ij}$ denotes the distance between $i$th and $j$th monomers and $b_{ij}$ denotes the equilibrium distance (bond length), which varies among atom pairs but is kept frozen during the simulation. The force constant $k$ of the springs connecting the nearest neighbors is the same everywhere and is represented as $\kappa=\epsilon_0/\sigma^2$ in terms of an arbitrary energy unit $\epsilon_0$. The bond lengths have been chosen by the following procedure [40]: every atom $i$ is independently assigned a random number $p_i=\pm 1$, representing big or small atoms, respectively. The bond length between atom $i$ and neighboring atom $j$ is set as $b_{ij}=b+(p_i+p_j)v$. Thus the bond length between two big atoms is $b+2v$, the bond length between two small atoms is $b-2v$, while the distance between a big and a small atom is $b$. Such a choice maintains an average bond length $b$. In order to take into account the effect of self-avoidance, we add the following potential between all pairs of monomers:

$$U(r)_{\rm SA}=\begin{cases} 0 & {\rm r}>\sigma \\ \infty & \text{otherwise.} \end{cases} \tag{2.2}$$

Hereafter, we fix the diameter of the hard sphere at $\sigma=1.0$. Note that the tethering potential does not restrict the length between nearest-neighbor monomers. Even if we take $b<\sqrt{3}$ and $v=0$, we cannot completely forbid the self-intersection at finite temperature. In the simulation we have studied two cases: The first case is $b=1.7$ [(1):''strong'' self-avoidance] and the second case is $b=3.0$ [(2):''weak'' self-avoidance] [42]. Case (1) corresponds to a rigid membrane and case (2) corresponds to a membrane with small bending rigidity. About the strength of disorder, in the case

(1), we have fixed $v=0.3$, which is close to the maximal contrast between the long and the short bonds as allowed by the algorithm. In case (2), we have at first studied extensively two cases $v=0.3\times3.0/1.7\sim0.52$ (weak disorder) and $v=0.75$ (strong disorder). The reason we take $v\sim0.52$ is that such a choice corresponds to the same strength of disorder with case (1), because the ratios $(v/b)$ are equal. Then we have varied the strength of disorder in the range $0.0<v<0.95$. For comparison, we have also performed Monte Carlo study of the model with no disorder, $(b,v)=(3.0,0.0)$ and $(b,v)=(1.7,0.0)$. About the force constant $k$, we have studied the behavior of the membrane at a temperature $k/k_BT=1.0$ and used the unit $T=1.0(k/K_B=1.0)$. At this temperature, the fluctuation of the distances between monomers is small. When we decrease the temperature in the case (1), the simulation begins at $T=10.0$, which was decreased from time to time by a factor 3. This corresponds to rapid cooling and there remains the possibility that the membrane is trapped in some local minimum [43]. However, we think that even with such rapid cooling we can see whether there occurs buckling instability, since it results in a large conformational transformation of the membrane.

An elementary move in Monte Carlo simulation consists of randomly choosing an atom and moving it by a distance $s$ in a randomly chosen direction. In all the simulations we have taken the displacement $s\leqslant0.2$. A trial move is accepted or rejected according to the conventional procedure of comparing $\exp(-\Delta E/k_BT)$, where $\Delta E$ is the energy difference between the configuration before and after the trial move, with a random number chosen from the interval $0-1$. We define the Monte Carlo time unit as a time required to perform $N$ elementary moves.

In order to investigate the number of time steps (defined in terms of Monte Carlo steps) required for thermalization, we have estimated the relaxation time by calculating the autocorrelation function of the observables such as radius of gyration and eigenvalues of the inertia tensor. The autocorrelation function of a physical quantity $O$ is defined as

$$A_O(t)=(\langle[O(t'+t)-\langle O(t')\rangle][O(t')-\langle O(t')\rangle]\rangle)/\langle[O(t')-\langle O(t')\rangle]^2\rangle, \quad (2.3)$$

where the average $\langle\ \rangle$ is performed over the time $t'$. Figure 1 depicts an example of such measurement for an $L=27(N=547)$ surface at temperature $T=1.0$. The lines (crosses) and (diamonds) correspond to the autocorrelation function $A_{Rg}(t)$ of the radius of gyration, respectively, for $(b,v)=(3.0,0.75)$ and $(b,v)=(3.0,0.0)$. From the figure we see $N^2$ Monte Carlo time units is sufficient for equilibration and independence between samples at the system size $N=547$. In all the simulation, we have used $N^2$ Monte Carlo steps as our time unit and we have performed an equilibrium process over times ranging between $30N^2$ and $500N^2$ Monte Carlo time units. We have also performed the disorder average for $N\leqslant169$ with some samples. When we vary the temperature or the strength of disorder, we have used a triangular lattice with 271 monomers.

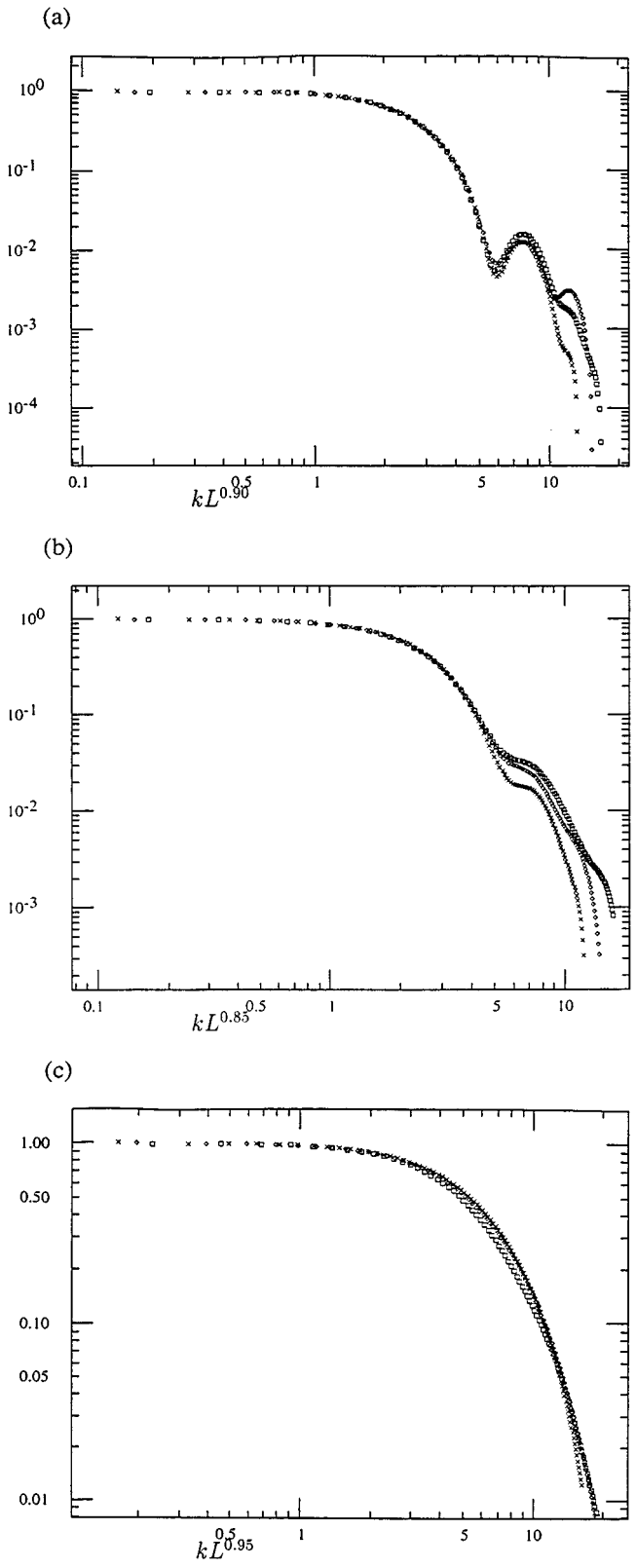


FIG. 5. (a) The structure factor $S_1(q)$ plotted as a function of the scaling variables $qL^{0.90}$ for $N=271$ ($\times$), 397 ($\diamond$), and 547 ($\square$). (b) The structure factor $S_2(q)$ plotted as a function of the scaling variables $qL^{0.85}$ for $N=271$ ($\times$), 397 ($\diamond$), and 547 ($\square$). (c) The ''perpendicular'' structure factor $S_3(q)$ plotted as a function of the scaling variables $qL^{0.95}$ for $N=271$ ($\times$), 397 ($\diamond$), and 547 ($\square$).

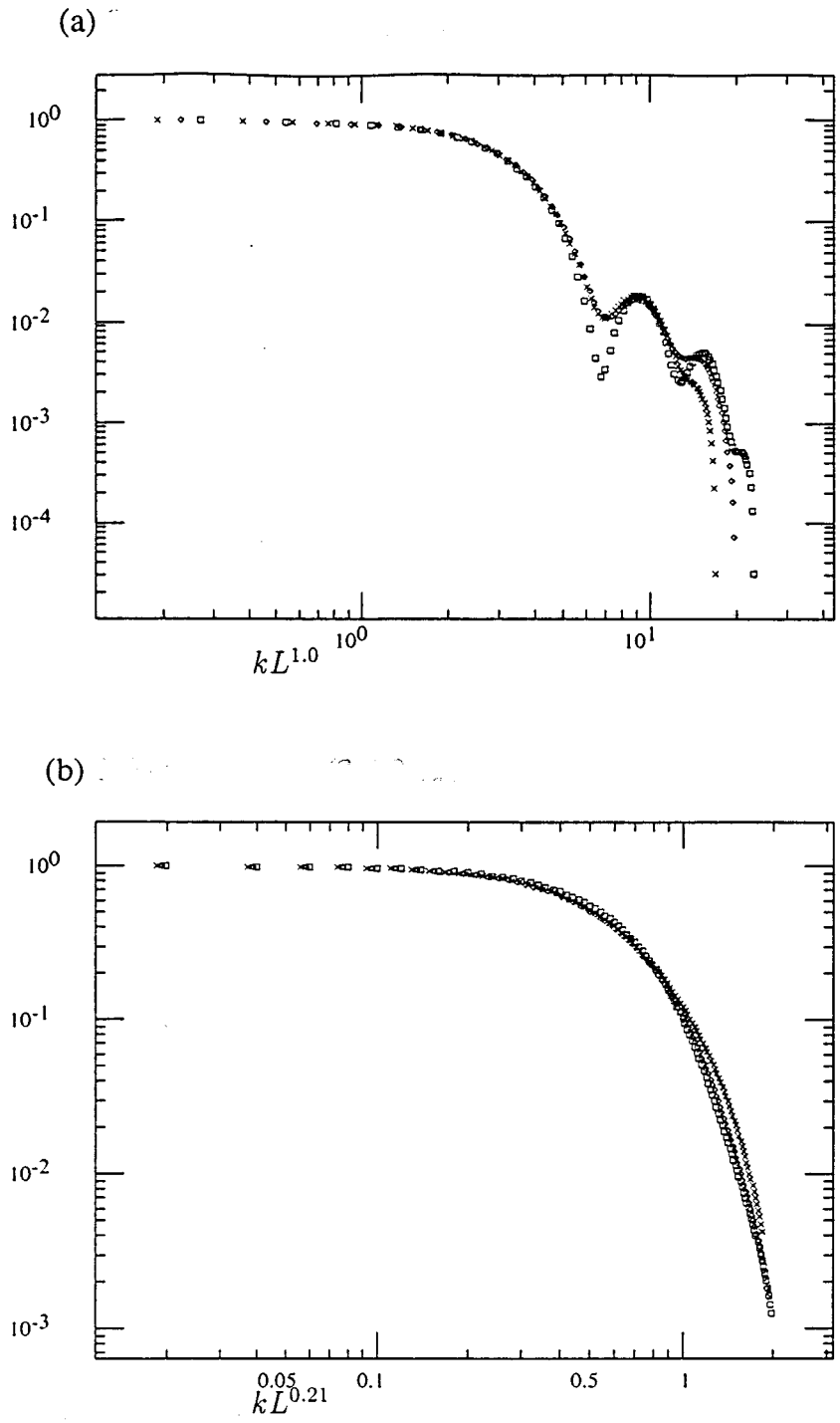


FIG. 6. (a) The structure factor $S_1(q)$ plotted as a function of the scaling variables $qL^{1.0}$ for $N=271$ ($\times$), 397 ($\diamond$), and 547 ($\square$). (b) The ''perpendicular'' structure factor $S_3(q)$ plotted as a function of the scaling variables $qL^{0.21}$ for $N=271$ ($\times$), 397 ($\diamond$), and 547 ($\square$).

### III. RESULTS

Typical spatial conformations of the 547-monomer membranes are presented in Fig. 2. The membrane with weak self-avoidance and strong disorder $(b,v)=(3.0,0.75)$ shows a large shrinkage. The membrane is highly crumpled as compared with the membrane with no disorder $(b,v)=(3.0,0.0)$ [Fig. 2(c)]. On the other hand, the spatial conformation of the membrane with ''strong'' self-avoidance and weak disorder $(b,v)=(1.7,0.3)$ exhibits clear anisotropy [Fig. 2(b)]. The membrane seems to be flat and, apart from some local buckling, the shrinkage is small.

As in previous works [8–10], the inertia tensor of the system

$$T_{\alpha\beta}=(1/N)\sum_i r_i^\alpha r_i^\beta-(1/N^2)\sum_{i,j} r_i^\alpha r_j^\beta \qquad (3.1)$$

is diagonalized for each configuration in the data set and the eigenvalues are numbered according to their magnitudes as $\lambda_1>\lambda_2>\lambda_3$. The directions of the principal axes are given by the eigenvectors $\vec{e}_j$ corresponding to $\lambda_j$. In Fig. 3, we

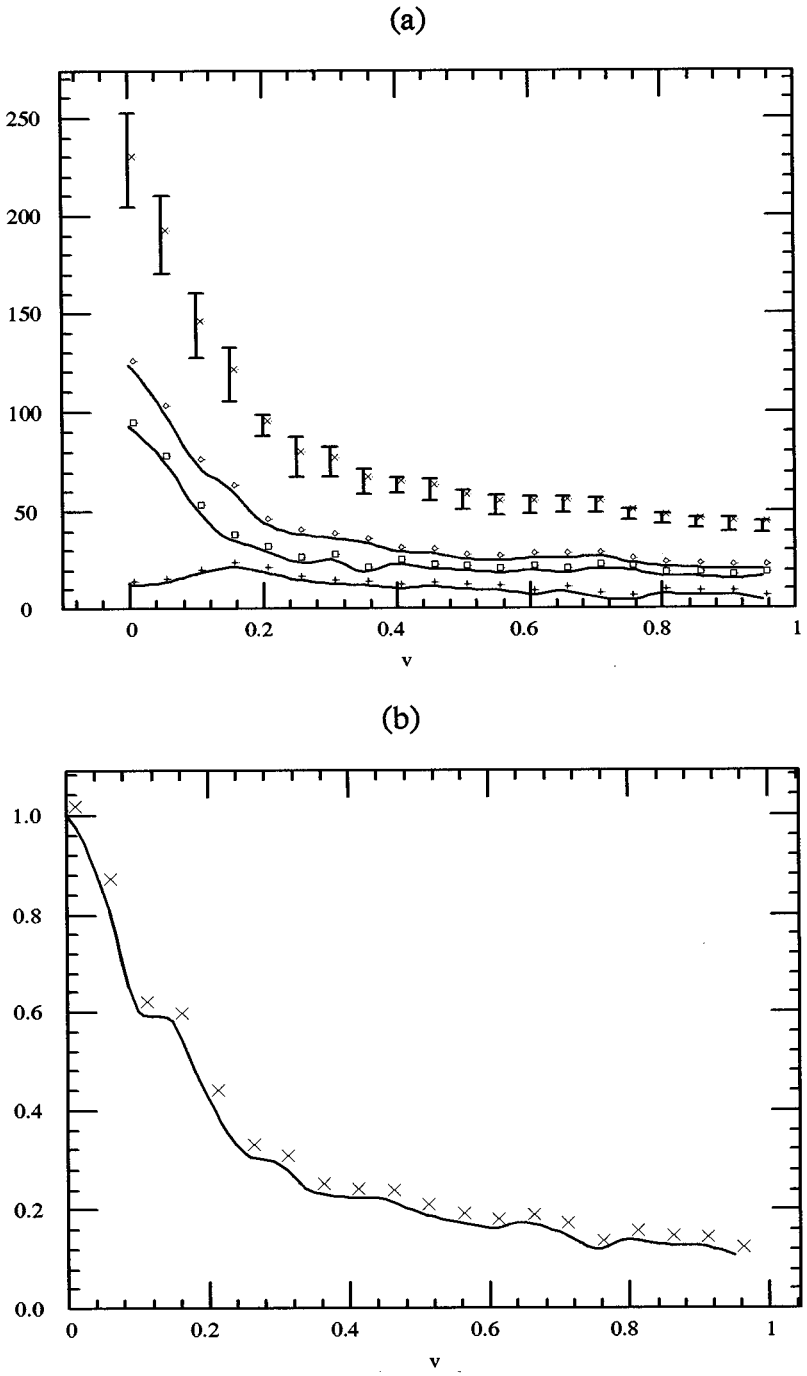


FIG. 7. (a) Plot of the mean square radius of gyration $\langle R_G^2 \rangle$ $\times(\times)$ and expectation values $\langle \lambda_1 \rangle(\diamond)$, $\langle \lambda_2 \rangle(\square)$, $\langle \lambda_3 \rangle(+)$ of the eigenvalues of the moment of inertia tensor for membranes with $(b,N)=(3.0,271)$. (b) The shrink parameter $P_{\text{shrink}}$ as a function of strength of disorder $v$. $N=271$ and $b=3.0$.

plot the eigenvalues and the square of the radius of gyration $R_G^2(=\lambda_1+\lambda_2+\lambda_3)$ versus $N$. The exponents $\nu$ and $\nu_i$ are defined as

$$R_G^2 \sim L^{2\nu}, \qquad \lambda_i \sim L^{2\nu_i}. \tag{3.2}$$

When the membrane is flat, the exponents $\nu$, $\nu_1$, and $\nu_2$ should coincide ($R_G^2 \sim \lambda_1 \sim \lambda_2 \sim L^{2\nu}$) and $\nu=1.0$. We call the exponent $\nu_3$ the roughness exponent $\nu_\perp$ ($\nu_\perp < 1.0$) in this case. When the membrane is crumpled, all the exponents coincide and $\nu < 1.0$.

Before presenting the results of these exponents, we shall quantify the shrinkage of the membrane. In order to characterize the shrinkage of the membrane, especially the difference between the membranes with and without disorder, we introduce the following shrinkage parameter [37]:

$$P_{\text{shrink}} = \frac{V_{\text{membrane with disorder}}}{V_{\text{membrane without disorder}}}, \tag{3.3}$$

where $V$ is defined as

$$V = \sqrt{\lambda_1 \lambda_2 \lambda_3}. \tag{3.4}$$

This shrinkage parameter means the inverse of the relative density of the membrane. In Fig. 3, we show the shrinkage

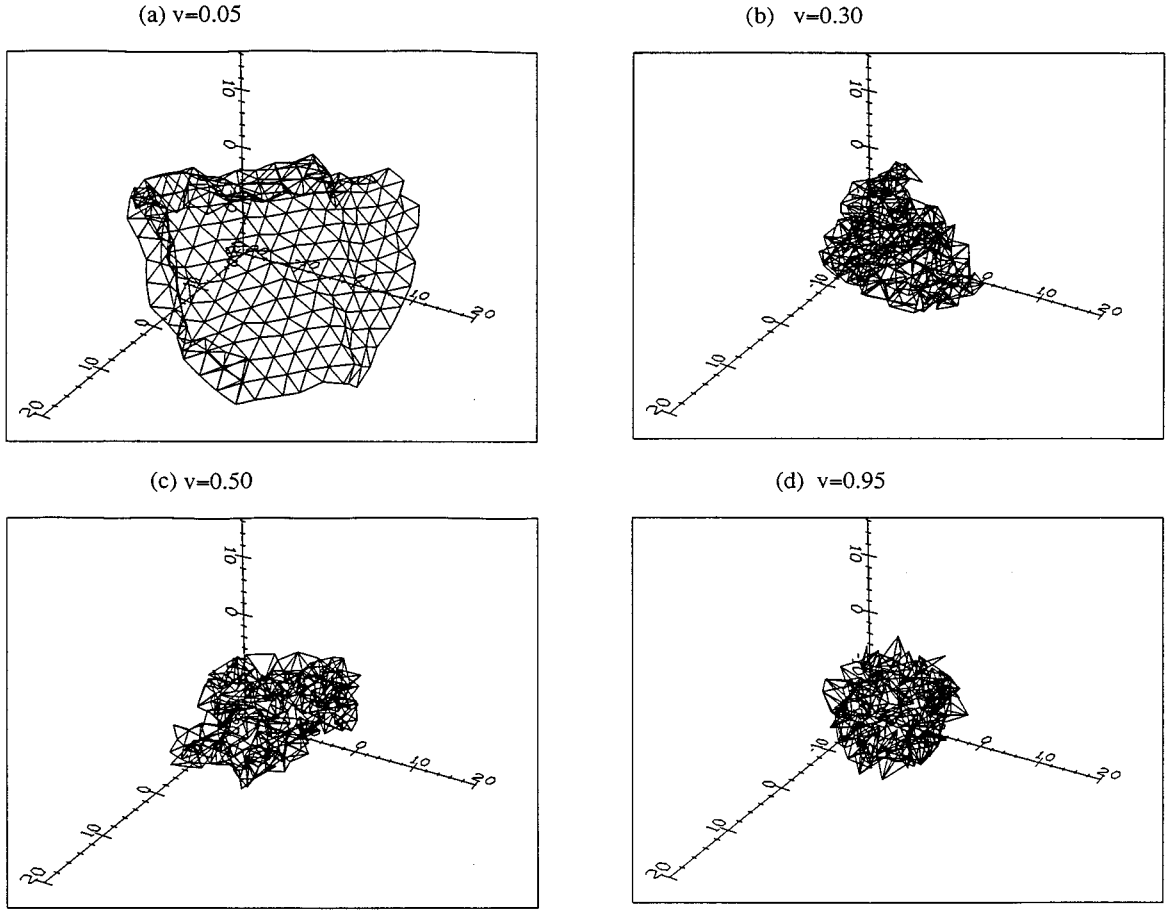


FIG. 8. Typical spatial conformation of the two-dimensional membrane with weak self-avoidance ($b=3.0$) and (a) $v=0.05$, (b) $v=0.3$, (c) $v=0.50$ in a flat membrane, and (d) $v=0.95$ in a crumpled membrane.

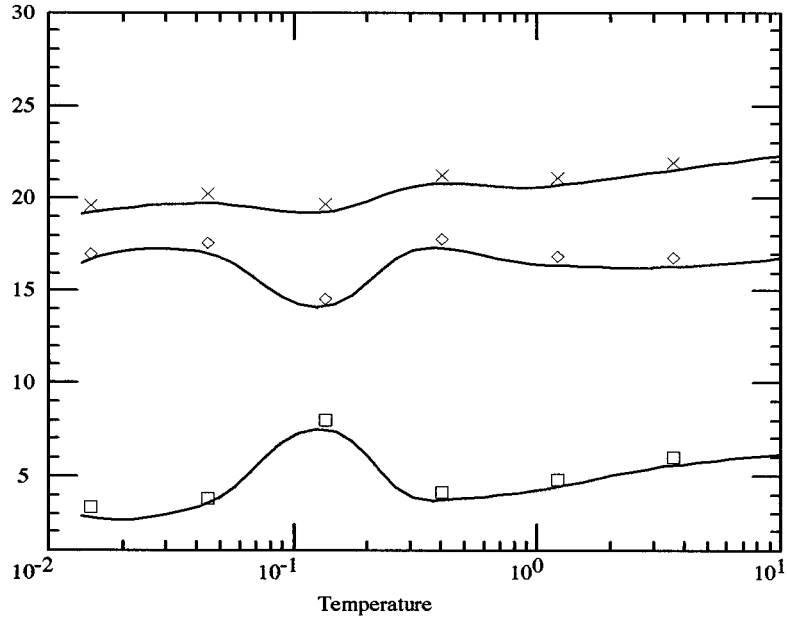


FIG. 9. Temperature dependence of the three (ordered) eigenvalues $\lambda_1>\lambda_2>\lambda_3$ of the inertia tensor of the membrane with $(b,v)=(1.7,0.3)$ and $N=271$.

parameter as a function of the monomer number $N$ for each case. When the self-avoidance is weak and the bending rigidity of the membrane is small ($b=3.0$), we can see a large shrinkage. With $N=547$ the relative density becomes about 11 times and it does not depend on the strength of disorder ($v=0.52$ and 0.75). On the other hand, the membrane with strong self-avoidance and weak disorder $(b,v)=(1.7,0.3)$ shows a small shrinkage. We think that in this case the disorder does not change the shape of the membrane drastically and only local buckling occurs, as we have seen in the spatial configuration. This result completely coincides with that of the simulation of polymerized fluid membrane [35,38].

In what follows, we shall study the asymptotic behavior of the membrane. At first, we study the scaling behavior of the eigenvalues of the inertia tensor. In Fig. 4 and Table I, we summarize the results. In pure cases $(b,v)=(3.0,0.0)$ and (1.7,0.0), the exponents $\nu$, $\nu_1$, and $\nu_2$ are apparently larger than $\nu_3$ and the membrane is asymptotically flat. However, the exponents $\nu$, $\nu_1$, and $\nu_2$ are larger than 1 and this means the membrane with (3.0,0.0) tends to stretch and there remains the effect of the boundary fluctuation or the force constant $k$ is not strong enough. Also in the case $(b,v)=(3.0,0.52)$, the exponents $\nu$, $\nu_1$, and $\nu_2$ are larger than $\nu_3$ and the membrane is anisotropic (flat). We see that the exponent $\nu_\perp$ is very small $\nu_\perp=0.0\pm0.2$. Such small undulations are caused by the large shrinkage of the membrane. As we increase the strength of disorder $v$, the usual flat phase becomes unstable by the disorder and the membrane is in a new flat phase. In the cases $(b,v)=(3.0,0.75)$ and (1.7,0.3) all the exponents $\nu$ and $\nu_i$ are large and we see no clear evidence of an anisotropy. However, the membrane with $(b,v)=(1.7,0.3)$ shows only a small shrinkage (Fig. 3) and its spatial conformation shows clear anisotropy [Fig. 2(b)]. We think that the membrane is flat in this case. On the other hand, the membrane with (3.0,0.75) exhibits a large shrinkage and its spatial conformation is also highly crumpled. The membrane with (3.0,0.52) is in a new flat phase and the membrane with (3.0,0.75) has stronger disorder. The exponent $\nu_3$ drastically changes from $\nu=0.0\pm0.2$ to $1.17\pm0.27$; it is natural to think that the membrane with (3.0,0.75) is not in the usual flat phase nor in the new flat phase. We assume it is in the crumpled phase.

In order to see further the shape of the membrane with weak self-avoidance ($b=3.0$), we investigate the structure factors defined by

$$S(\vec{k})=\frac{1}{N^2}\left\langle \sum_{i,j} \exp i\vec{k}\cdot[\vec{r}_i-\vec{r}_j]\right\rangle. \tag{3.5}$$

The angular brackets indicate the average over equilibrated configurations. In Fig. 5, we show the structure factors $S(k\vec{e}_1)=S_1(k)$, $S(k\vec{e}_2)=S_2(k)$, and $S(k\vec{e}_3)=S_3(k)$, plotted as a function of the variable $kL^\nu$ for $271\leqslant N\leqslant 547$ and $v=0.75$. Although the scaling for the ''perpendicular'' structure factor $S_3$ is not good enough to estimate the exponent $\nu_3$, we estimate $\nu=0.90-0.85$ from other structure factors. Figure 6 depicts the structure factors $S_1$ and $S_3$ for the membrane with $(b,v)=(3.0,0.52)$. The scaling behavior for $S_1$, which is a function of $kL$, indicates that the membrane is flat. From the scaling of the perpendicular structure factor $S_3$, we estimate the roughness exponent $\nu_\perp=0.21$. This value is within the error bar of the previous estimate from the scaling of $\lambda_3$ and is a reliable one [44].

From these analysis, we think that the membrane with (1.7,0.3) is flat and the membrane with (3.0,0.52) is in a new flat phase with a small roughness exponent $\nu_\perp=0.1\pm0.1$. The membrane with (3.0,0.75) is in a crumpled phase with $\nu=0.85-0.92$, which is near the theoretically predicted value $\nu=6/7$ [34].

In order to see the behavior of the conformational transformation and the shrinkage of the membrane with weak self-avoidance caused by the random stress, we have investigated the behaviors of the membrane with various strength of disorder. The strength of disorder is in the range $0.0<v<0.95$. Figure 7(a) depicts the behavior of $R_G$ and other eigenvalues of the inertia tensor as a function of $v$. Figure 7(b) shows the corresponding shrinkage parameter, respectively. Even if the strength of disorder is small, $v\sim0.2$, the shrinkage is large and the density becomes 2.5 times. That is, the membrane's shape changes significantly. After that, the shrinkage parameter and other quantities do not change much. In Fig. 8, we depict the conformational transformation of the membrane as we change the strength of disorder $v$. When the disorder is weak $v=0.05$, the membrane is smooth and we can see its anisotropic nature. As the disorder becomes large, $v=0.30-0.50$, the membrane's conformation changes dramatically and we see large shrinkages. The membrane with strong disorder $v=0.95$ seems to be crumpled and takes an isotropic conformation.

Secondly, we have studied the cooling of the membrane with strong self-avoidance and weak disorder $(b,v)=(1.7,0.3)$. If the bending rigidity and the elastic constants change as we decrease the temperature $T$, the buckling condition (1.1) may be satisfied. At the temperature $T=1$, the membrane seems to be flat from the above analysis. The temperature dependence of the spatial conformations of the membranes is depicted in Fig. 9. As the temperature decreases, at $T\sim0.3$, a peak occurs in $\lambda_3$, however, overall the membrane becomes flatter and flatter. We see no apparent evidence of foldings or large buckling, which was seen in the phantom case near $T=0$ [40]. We have also obtained other thermodynamic functions such as internal energy and specific heat. We do not find any singularity in the data. This means that the remnant bending rigidity prevents the membrane from drastic buckling within the range of $T$ studied here.

## IV. CONCLUSIONS

We summarize the results obtained in the previous section. First we have studied asymptotic behavior of the membrane with the following sets of parameters $(b,v)=(1.7,0.0),(1.7,0.3)$ (''strong'' self-avoiding case) and (3.0,0.0),(3.0,0.52),(3.0,0.75) (''weak'' self-avoiding case).

(1) For membranes with weak self-avoidance ($b=3.0$), there occurs large shrinkages by the disorder. The relative density with the $v=0.0$ case becomes 8–9 times even if the strength of disorder is small $v\sim0.52$ (Fig. 6). As the strength of disorder increases, the membrane exhibits a phase transition from a highly fluctuating flat phase ($v=0.0$) to a

crumpled phase ($\upsilon = 0.75, \nu = 0.85$–$0.92$) passing through a new flat phase ($\upsilon = 0.52, \nu_\perp = 0.1 \pm 0.1$) (Fig. 8). This result agrees well with the theoretical prediction $\nu = 6/7$, but a larger system size and an average over randomness are necessary in order to estimate the exponent precisely. The model does not completely forbid the self-intersection. These findings may arise from this. That is, if the self-intersection is completely prohibited, such behaviors may not occur. However, even if the self-intersection is completely forbidden, highly crumpled and floppy conformation is possible in a self-avoiding plaquette membrane model [13,14]. We therefore think that large shrinkages can occur in the membrane with complete self-avoidance when the bending rigidity is small.

(2) A membrane with strong self-avoidance and weak disorder $(b,\upsilon) = (1.7,0.3)$ is asymptotically flat and its shrinkage is small. On the other hand, the membrane with the same strength of disorder and weak self-avoidance $(b,\upsilon) = (3.0,0.52)$ shows a large shrinkage. One possible reason for the difference is that the former one has a larger effective bending rigidity than the latter one. The condition (1.1) is not satisfied and the buckling transition does not occur in the strong self-avoidance case. In order to see whether there occurs some large conformational transformation by thermal buckling transitions, we have studied the cooling process of the membrane with (1.7,0.3). The membrane becomes flatter and flatter as the temperature decreases and we find no folding or large buckling between the temperature about $0.01 < T < 10.0$. This indicates that the bending rigidity cannot be so small even if the temperature is very low. As the ''entropic'' rigidity contribution to the total bending rigidity of the flat membrane does not depend much on the temperature, the buckling condition is not satisfied even if the temperature becomes small.

From these results, we discuss the possibility of thermal conformational transformation of the ''weak'' self-avoiding tethered membrane with quenched disorder. Introducing the ''bare'' bending energy $\kappa$ into the model ($b = 3.0$), we may be able to make the membrane smooth and flat again. Then we may see a large buckling of the membrane by decreasing the temperature according to the picture explained in the Introduction [21]. Such transformation is possible only for the weak self-avoidance case, otherwise remnant entropic rigidity prevents the condition (1.1) from being satisfied in the strong self-avoidance case. This possibility may be relevant to the understanding of the wrinkling transition in partially polymerized vesicles [23].

Our results also show that the following mechanism explains the wrinkling transition [26]. In the experiment, the partial polymerization at high temperature presumably results in a sparse but percolating network of covalent bonds and the flat shape is stable. As the temperature decreases, a crystalline order sets in within these lipid areas and quenched random stress will appear. This process corresponds to the increase in $\upsilon$ in our study and large transformation occurs and a wrinkled structure appears. That is, our results indicate that the wrinkling transition can be a mechanical buckling transition, which we found in this work.

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ture decrease [4], however the former one does not depend on $T$ so much. This explanation seems to contradict with the picture that the induced ''entropic'' rigidity is proportional to temperature [11], it is not so. When the membrane becomes flat, the configuration of the membrane does not sweep the entire phase space. The change in temperature $T$ only change the strength of connectivity and it does not contribute to the former one apart from the latter thermally induced one. So the former one, ''entropic'' bending rigidity in the flat phase, contributes as a remnant one to the total bending rigidity.

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