Square lattice with attractive interactions

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We analyze the square lattice, which is allowed to fold on itself along its bonds in a two-dimensional embedding space, with bending energy (u) and attractive ($\omega<0$) or repulsive ($\omega>0$) interactions. We discuss two types of interaction, the first one is proportional to the contact area and the second one is proportional to the number of pairs of elementary squares which occupy the same place in the plane. We obtain the phase diagrams in the (u,ω) plane for both interactions. We also perform the numerical analysis for the first type of interaction and discuss its relation to the sequential folding transition of the polymerized membrane with the attractive interaction [F. F. Abraham and M. Kardar, Science **252**, 419 (1991)].

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I. INTRODUCTION

Recently there has been considerable interest in the phase transition of polymerized (tethered) membranes [1] with attractive interactions [2–6]. In a pioneering work [2,3], Abraham and Nelson found by molecular dynamics simulations that the introduction of attractive interactions between monomers leads to a collapsed membrane with fractal dimension 3 at sufficiently low temperature. Subsequently, Abraham and Kardar [4] showed that for open membranes with attractive interactions, as temperature decreases, there exists a well-defined sequence of folding transitions and then the membrane ends up in the collapsed phase. They also presented a Landau theory of the transition and in addition, they discussed that the folding transition is related to the unbinding transition of bimembranes. Liu and Plischke [5] carried out Monte Carlo simulations for a similar model and found that the membrane undergoes a phase transition from the high-temperature flat phase to the low-temperature collapsed phase passing through an intermediate crumpled phase. The intermediate crumpled phase exists over a certain range of temperature and its fractal dimension is estimated as $d_f=2.5$. Following this work, Grest and Petsche [6] extensively carried out molecular dynamics simulations of closed membranes. They considered flexible membranes; the nodes of the membrane are connected by a linear chain of n monomers. For short monomer chains, $n=4$, there occurs a first-order transition from the high-temperatures flat phase to the low-temperature collapsed phase, but no intermediate crumpled phase. For longer chains, $n=8$, the transition is either continuous or weakly first order. With the assumption of the continuous transition, the fractal dimension of the membrane at the transition is estimated as $d_f=2.4$. Mori and Wadati [7] discussed the phase transition of a D-dimensional (phantom) polymerized membrane with long-range attractive interaction ($r^{-\gamma}$) and showed that there are several types of phase transitions from a flat phase to a compact phase depending on the type of the interaction (γ) and the dimension of the membrane (D). Especially, when the interaction is short ranged, the transition is continuous. Based on this result, they discussed the possibility of the complete cancellation between the "entropic" rigidity from self-avoidance [2] and the negative rigidity from attractive interaction, which causes the crumpling transition of the membrane. This cancellation only occurs when the membrane is very flexible. Otherwise, the flat phase becomes unstable before the cancellation becomes complete and the membrane shows the sequential folding transition.

In order to describe the sequential folding transition of the membrane, the folding degrees of freedom of the membrane are important. The square lattice model was at first introduced by David and Guitter [8] as a simple model for polymerized membrane. The model is a discrete rigid-bond square lattice, which is allowed to fold on itself along its bonds in a two-dimensional embedding space. By changing the bending rigidity u, it shows a first-order transition from a completely flat phase ($u>0$) to a completely folded state ($u<0$) [8,10]. The triangular lattice case was also studied [9–11]. In this paper, we would like to study a square lattice model with attractive ($\omega<0$) or repulsive interactions ($\omega>0$) as a simple model that describes the sequential folding transition of the tethered membrane. As an interaction between different parts of the lattice, we study two types of interactions: the first one is a potential that is proportional to the contact area of the membrane and the second one is proportional to the number of pairs of the elementary squares that occupy the same place in the plane. We discuss possible phases and phase diagrams in the (u,ω) plane for each interaction. For the first type of interaction, we have also performed numerical studies.

The balance of our paper is as follows. In Sec. II, we define the square lattice model with bending rigidity (u) and interactions (ω). We consider two types of interactions; the first one is a potential that is proportional to the contact area of the lattice (CA type) and the second one is proportional to the number of pairs of elementary squares that occupy the same place in the plane (CP type). We treat these models at mean-field level and

study the stability of the flat phase ($u>0$) by attractive interactions and the one of completely folded state ($u<0$) by repulsive interactions. We obtain the phase diagrams in the (u,ω) plane for each potential. In particular, for the CA-type interaction, we find a partially folded state in the region ($u>0,\omega<0$). We perform numerical studies for the model with the CA-type interaction in Sec. III. We investigate the above theoretical results. In addition, we find a sequential folding transition of the lattice. We conclude in Sec. IV with a discussion about the difference between the potentials used here and the relation between the sequential folding transitions of polymerized membrane and the one of the square lattice.

II. MODEL SYSTEMS

We consider a model of foldings of a two-dimensional square lattice with $L\times L$ size. We consider all possible foldings of the lattice and each folding maintains the correct distances between the neighboring sites. Two configurations are identical if the positions of all corresponding sites (or vertex) coincide and this definition of the identical configuration does not distinguish between the different manners of folding that lead to the same final state. Figure 1(a) depicts such a lattice before the folding, while Fig. 1(b) shows the section of it in a folded state along its x axis. The membrane has of course two such sections. The membrane is constrained to a plane and the section is represented by a line of zero thickness that folds in $N=1,2,\ldots,L$ segments of successive length $x_1,x_2,\ldots x_N$ [14,15]. Correspondingly, the other section, which folds in M segments, is represented by $u_1,y_2,\ldots,y_M$. These segments' lengths x_i and y_j are multiples of lattice constant $a=1$. For convenience, in the theoretical calculation, we treat them as continuous quantities with uv cutoff (lattice constant $a=1$). The total lengths are fixed:

$$\sum_{i=1}^{N} x_i=L,\quad \sum_{j=1}^{N} y_j=L\ . \tag{2.1}$$

A configuration of the system is thus determined by a set of natural numbers (N,M) and a set of positive numbers $x_1,x_2,\ldots,x_N;y_1,y_2,\ldots,y_M$. Let the potential energy for a configuration of the square lattice be denoted by $U_{N,M}(x_1,x_2,\ldots,x_N;y_1,y_2,\ldots,y_M)$.

The potential energy will be taken to consist of the sum of two terms. First, as a bending energy, we assign energy κ per unit length of a fold. We denote an interaction between the elementary squares of the lattice as U_I and the potential energy for the system is written as

$$U_{N,M}(x_1,x_2,\ldots,x_N;y_1,y_2,\ldots,y_M)$$
$$=(N-1)\kappa L+(M-1)\kappa L+U_I \tag{2.2}$$
$$=U'_{N,M}+U_I\ . \tag{2.3}$$

As an interaction between different parts of the square lattice, we consider two types of interactions. The first one is a potential that is proportional to the contact area of the lattice. The contact area is determined by the area difference between the initial stretched form and the configuration considered. The potential is written as

$$U_{I,\mathrm{CA}}(x_1,\ldots,x_N;y_1,\ldots,y_M)=w(L^2-L_xL_y)\ . \tag{2.4}$$

Here, L_x and L_y are the widths in each direction of the lattice [Fig. 1(b)] and are represented as

$$\begin{aligned} L_x&=\max(x_1,x_1-x_2+x_3,\ldots)\\ &\quad-\min(0,x_1-x_2,\ldots)\\ L_y&=\max(y_1,y_1-y_2+y_3,\ldots)\\ &\quad-\min(0,y_1-y_2,\ldots)\ . \end{aligned} \tag{2.5}$$

We also consider a potential that is proportional to the number of pairs of elementary squares that share the same place in the plane (Contact Pair).

$$U_{I,\mathrm{CP}}(x_1,\ldots,x_N;y_1,\ldots,y_M)$$
$$=w\times(\text{number of pairs of contact elementary squares}). \tag{2.6}$$

In the usual discussion of polymer with attractive interaction, the second type is usually employed [12]. However, the attractive interaction of a polymer in poor solvent comes from that solute-solute and solvent-solvent contacts are preferred to solute-solvent contacts. From this point of view, we think the contact area potential $U_{I,\mathrm{CA}}$ is more natural than the contact pairs potential $U_{I,\mathrm{CP}}$ and we will discuss the difference between these potentials in the square lattice model.

For convenience we will use the reduced bending rigidity and potential strength,

$$u=\kappa/k_BT,\quad \omega=w/k_BT\ . \tag{2.7}$$

The partition function is given as

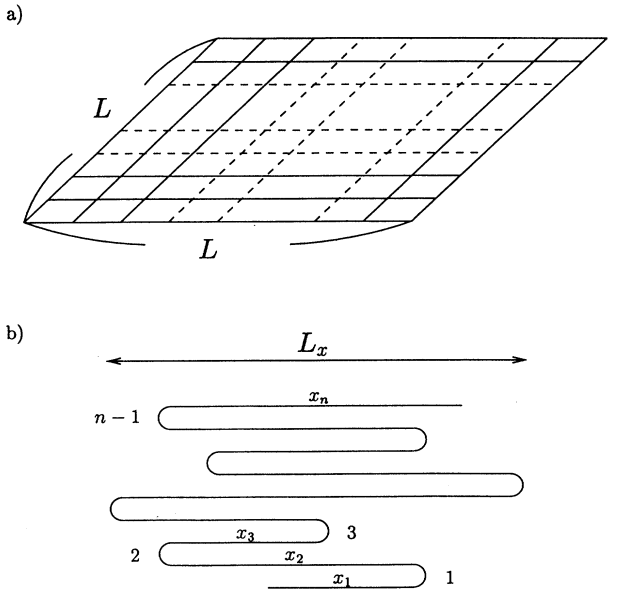

FIG. 1. (a) Stretched form of a square lattice of size $L\times L$. (b) Typical configuration of a section of the lattice. The folds have a vanishing length. The lattice is not completely folded to clearly indicate the configuration.

$$Z(u,\omega,L)=\sum_{N,M=1}^{\infty}\int_a^\infty dx_1\int_a^\infty dx_2\cdots\int_a^\infty dx_N\int_a^\infty dy_1\int_a^\infty dy_2\cdots\int_a^\infty dx_M \times\delta\left[\sum_{i=1}^{N}x_i-L\right]\delta\left[\sum_{j=1}^{M}y_j-L\right]\exp(-U_{N,M}/k_BT)\ . \tag{2.8}$$

The evaluation of the configuration sum is very difficult, we would like to treat the interaction term U_I at the mean-field level. At first we evaluate the partition function with the potential $U'_{N,M}$. By using Laplace transform [15], the partition function can be calculated easily and the expression is

$$Z(u,\omega=0,L)=\exp(2\eta_* L)\ . \tag{2.9}$$

Here η_* is a solution of the following equation,

$$\exp(a\eta_*)\eta_*=\exp(-uL)\ . \tag{2.10}$$

Then the free energy of the system is

$$\mathcal{F}_0\equiv-\ln Z(u,\omega=0,L)^2 =-2\eta_* L\ . \tag{2.11}$$

The condition (2.10) is rewritten as

$$\ln\eta_*+a\eta_*=-uL\ . \tag{2.12}$$

In the thermodynamic limit $L\to\infty$, the solution η_* is estimated as ($a=1$),

$$\eta_*=\begin{cases}\exp(-uL), & u>0\\ -uL, & u<0\ .\end{cases} \tag{2.13}$$

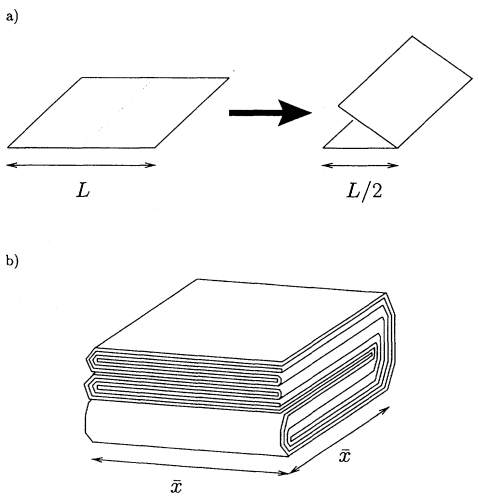

FIG. 2. (a) Folding transition of a flat membrane. Attractive interaction is considered to break the flat phase like in the figure. (b) Uniformly folded state of a square lattice. Each segment has equal length $\bar{x}=L/N$.

Free energy $\mathcal{F}_0$ is

$$\mathcal{F}_0=\begin{cases}-2L\exp(-uL), & u>0\\ -2uL^2, & u<0\ .\end{cases} \tag{2.14}$$

The total number of folds N_{total} is

$$N_{\text{total}}\equiv\langle N+M-2\rangle\equiv\frac{\partial}{L\partial u}\mathcal{F}_0 =\begin{cases}2L\exp(-uL)\to 0(L\to\infty), & u>0\\ 2L, & u<0\ .\end{cases} \tag{2.15}$$

This behavior completely coincides with the previous results [8,10,13].

Next we would like to study the effect of interactions on these results. From the above analysis, the lattice is in a completely flat phase ($u>0$) or completely folded state ($u<0$). When the lattice is in the flat state, the attractive interaction is considered to break the flat state as in Fig. 2(a). We assume the broken flat state is an uniformly folded state with $N=1+\frac{1}{2}N_{\text{total}}$ segments in each direction [Fig. 2(b)], that is each segment has equal length $\bar{x}\equiv L/N$. We estimate the effect of attractive interactions as

$$\langle U_{\text{CA}}\rangle=\omega(L^2-\bar{x}^2)\ , \tag{2.16}$$

$$\langle U_{\text{CP}}\rangle=\tfrac{1}{2}\omega N^2(N^2-1)\bar{x}^2\ . \tag{2.17}$$

On the other hand, when the repulsive force breaks completely the folded state, it is not clear how the folded state becomes unstable. We assume the above estimation holds even in this case. And in what follows, we also take the lattice constant $a=0$ for convenience. Then $\eta_*=\exp(-uL)$ and $N_{\text{total}}=2L\exp(-uL)$. However, when one interprets the results physically, the uv cutoff has to be taken into account. The total free energy is evaluated as

$$\mathcal{F}(u,\omega,L)=\begin{cases}-2\eta_* L+\omega(L^2-\bar{x}^2), & \text{CA case}\\ -2\eta_* L+\frac{1}{2}\omega N^2(N^2-1)\bar{x}^2, & \text{CP case}\ .\end{cases} \tag{2.18}$$

From the relation $N_{\text{total}}=2L\exp(-uL)$, we can express u in terms of N_{total},

$$uL=-\ln(N_{\text{total}}/2L)\ . \tag{2.19}$$

In the above estimation of interactions (2.17), we use the fold number $N=1+\frac{1}{2}N_{\text{total}}$, which is a function of u. In order to rewrite the free energy $\mathcal{F}_0(u,\omega,L)$ in terms of $(N_{\text{total}},\omega,L)$, we perform the Legendre transform with respect to u. Then the free energy $\mathcal{G}(N_{\text{total}},\omega,L)$ is

$$\mathcal{G}(N_{\text{total}},\omega,L)=uLN_{\text{total}}-\mathcal{F}(u,\omega,L)=\begin{cases}-N_{\text{total}}\ln N_{\text{total}}/2L+N_{\text{total}}-\omega L^2(1-N^{-2}), & \text{CA case}\\ -N_{\text{total}}\ln N_{\text{total}}/2+N_{\text{total}}-\frac{1}{2}\omega L^2(N^2-1), & \text{CP case}\ .\end{cases} \tag{2.20}$$

This free energy $\mathcal{G}$ has the following relation:

$$\frac{\partial\mathcal{G}}{\partial N_{\text{total}}}=uL\ . \tag{2.21}$$

In what follows, we discuss the effect of interactions on the completely flat phase and on the completely folded state. At first, we discuss the effect of attractive interactions on the completely flat phase. In this case, N_{total} is very small (~ 0) and the free energy $\mathcal{G}$ is roughly estimated as

$$\mathcal{G}(N_{\text{total}},\omega,L)=uL\langle N_{\text{total}}\rangle-\mathcal{F}(u,\omega,L)$$
$$=\begin{cases}-N_{\text{total}}\ln(N_{\text{total}}/2L)+N_{\text{total}}-\omega L^2N_{\text{total}} & \text{CA case}\\ -N_{\text{total}}\ln(N_{\text{total}}/2L)+N_{\text{total}}-\frac{1}{2}\omega L^2N_{\text{total}} & \text{CP case}\ .\end{cases} \tag{2.22}$$

Here we have discarded terms that are higher order in N_{total} or that do not depend on N_{total}. From the relation (2.21), N_{total} is given by

$$N_{\text{total}}=\begin{cases}2L\exp[-(uL+\omega L^2)], & \text{CA case}\\ 2L\exp[-(uL+\frac{1}{2}\omega L^2)], & \text{CP case}\ .\end{cases} \tag{2.23}$$

This means that the completely flat phase becomes unstable with respect to both attractive interactions and the critical values ω_{critical} are

$$\omega_{\text{critical}}=\begin{cases}-u/L, & \text{CA case}\\ -2u/L, & \text{CP case}\ .\end{cases} \tag{2.24}$$

These values go to zero in the thermodynamic limit $L\to\infty$, which is very natural. Because the free-energy difference between the completely flat state and the one-fold state with a crease at its center is $\delta F=\frac{1}{2}\omega L^2+uL-\ln 2$ and it becomes negative for $\omega<-2u/L$ [2]. However, after some sequential foldings, the behaviors of the membrane are different for each interaction. In this case N_{total} is not small and Eq. (2.21) becomes

$$uL\sim\begin{cases}-\omega L^2/N^3, & \text{CA case}\\ -\omega L^2N, & \text{CP case}\ .\end{cases} \tag{2.25}$$

In the CP case, the effect of interaction becomes more and more important as N becomes large. And in the thermodynamic limit ($L\to\infty$), the bending rigidity becomes irrelevant and the membrane is in the completely folded state.

We also note that Eq. (2.21) has the following form:

$$uL=-\ln(N_{\text{total}}/2L)-\tfrac{1}{4}\omega L^2N_{\text{total}}\ . \tag{2.26}$$

When ω is negative, the right-hand side has a minimum. If u is small enough, the equation has no solution. The reason is that the free energy has no minimum and the model predicts a complete collapse of the membrane ($N\to\infty$). In order to avoid this catastrophe [12], we need to introduce three-body repulsive interactions such as

$$U_{I,\text{CT}}(x_1,\ldots,x_N;y_1,\ldots,y_M)=\omega'\times(\text{number of sets of three contact elementary squares})\ . \tag{2.27}$$

Then the free energy has the following additional term,

$$\langle U_{I,\text{CT}}\rangle=\tfrac{1}{6}\omega'L^2(N^2-1)(N^2-2)\ . \tag{2.28}$$

Then Eq. (2.21) is roughly,

$$-\omega=\frac{4u}{N_{\text{total}}L}+\frac{4}{L^2N_{\text{total}}}\ln(N_{\text{total}}/2L)+\tfrac{1}{6}\omega'N_{\text{total}}^2\ . \tag{2.29}$$

This equation has solutions N_{total} for any value of ω when $\omega'>0$. Especially, depending on the value of ω', there is the possibility that the equation has two solutions for some value of ω. Following the discussion about the coil-globule transition of a polymer, it corresponds to a discontinuous transition (change in N becomes discontinuous) [12].

On the other hand, in the CA case, the interaction becomes small as N becomes large and balancing the effect of bending rigidity and that of attractive interaction [Eq. (2.25)], N behaves as $L^{1/3}$. The behavior of the mean area is estimated to be $\langle L_x\times L_y\rangle\sim\bar{x}^2\sim L^{4/3}$. That is, the number of folds is small and the membrane is not in the completely folded state. We call this phase the partially folded state and analyze its behavior with a numerical method later.

Secondly, we study the stability of the completely folded state with respect to repulsive interactions. In this case, N_{total} is large ($\sim 2L$). In the CA case, the interaction is irrelevant and the completely flat phase is stable

with respect to the weak repulsive interaction. However, the situation is not so simple. In the completely folded state, the free energy per elementary square is $f_{\text{fold}}=2u+\omega$ and in the flat phase $f_{\text{flat}}=0$. From the discussion given in [10], the free energy of the system is given by

$$f_{\text{square}}=\min(f_{\text{fold}},f_{\text{flat}}) . \tag{2.30}$$

Consequently, there occurs a first-order phase transition between a completely flat phase ($2u+\omega>0$) and a completely folded state ($2u+\omega<0$). The intermediate folded state [$0<N_{\text{total}}<2(L-1)$] does not appear [16]. In the CP case, the repulsive interaction becomes very large and the completely folded phase becomes unstable. Even if N and ω are small, the repulsive interaction wins over negative bending energy in the thermodynamic limit ($L\to\infty$) and we think that the membrane is flat. However, in this case, the above estimation of the interaction (2.17) is not good. Because, when the membrane is almost flat, negative bending energy causes a crease at the edge of the membrane, not at its center. Even if the system size is large a fold can occur at its edge and the membrane is not in the completely flat phase.

Phase diagrams for both interactions are summarized in Fig. 3. Figures 3(a) and 3(b) correspond to the phase diagrams of the system with CA interaction and that of the system with CP interaction. In the domain ($u>0,\omega>0$), the membrane is in the completely flat phase ($N_{\text{total}}=0$) and in the domain ($u<0,\omega<0$), the membrane is completely folded ($N_{\text{total}}=2L$) for both interactions. The completely flat phase ($u>0$) becomes unstable by both attractive interactions ($\omega<0$), however, the resulting phases are different. In the CA case the membrane is partially folded and $N_{\text{total}}\sim L^{1/3}$ and the mean area behaves $\langle L_xL_y\rangle\sim L^{4/3}$. In the CP case, the membrane is completely folded. When the system size is finite ($L<\infty$), both systems show the sequential folding transition. In the domain ($u<0,\omega>0$), the folded state also becomes unstable in both systems. In the CA case, there occurs a first-order phase transition from the completely folded phase to the completely flat phase at the line ($2u+\omega=0$). In the CP case, the completely folded state becomes unstable by any amount of repulsive interaction and the membrane becomes flat.

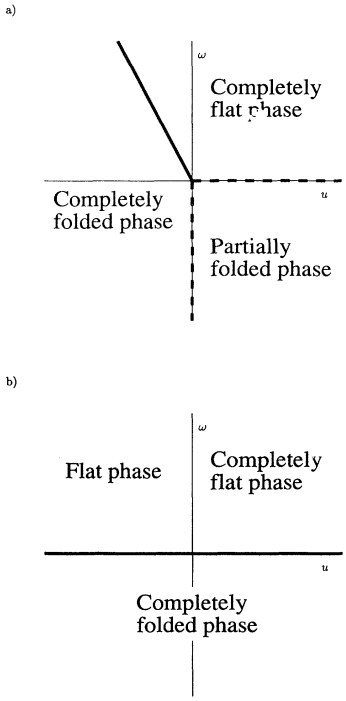

FIG. 3. (a) Phase diagram in the (u,ω) plane for the square lattice with CA-type interaction. Three first-order lines $\omega=-2u\,(u<0)$, $u=0(\omega<0)$, and $\omega=0(u>0)$ separate the three phases, completely flat phase, completely folded state, and partially folded state. (b) Phase diagram for the square lattice with CP-type interaction. One first-order line $\omega=0$ separates the completely folded state from the flat phase.

III. NUMERICAL STUDIES

In this section, we will study the square lattice with CA interaction numerically. In the previous section, we have treated the segment lengths as continuous quantities. Hereafter, we return to the original discrete system.

The potential we consider is

$$U_{N,M}=U'_{N,M}+U_{I,\text{CA}}(L_x,L_y) , \tag{3.1}$$

and we perform the configuration sum directly. From the definition of $U_{I,\text{CA}}$ [Eq. (2.4)], the above potential only depends on the folding number $(N-1,M-1)$ and the width of each section (L_x,L_y). We can interpret each section as a one-dimensional random walk with $L-1$ steps and it has 2^{L-1} states. The main part of numerical calculation is the number counting of states, which is specified by the final length l ($=L_x,L_y$ in the previous section) and the bending number b ($=N-1,M-1$). Table I shows such numbers that represent the degeneracies of corresponding (l,b) states for the $L=12$ case. Hereafter, we denote such a number as $D(l,b)$.

Then we execute the following "standard" calculation to evaluate the partition function and other thermodynamic quantities for each set of parameters (u,ω). The partition function is evaluated as

$$Z(u,\omega,L)\equiv\sum_{L_x,L_y=1}^{L}\sum_{N,M=0}^{L-1}D(L_x,N)D(L_y,M)\times\exp[-U_{N,M}(N,M;L_x,L_y)] \tag{3.2}$$

where

$$U_{N,M}(N,M;L_x,L_y)=uL(M+N-2)-\omega L_xL_y . \tag{3.3}$$

Note that we have moved the energy origin by a constant $-\omega L^2$ from the definition of the previous section. The thermal average $\langle\ \rangle$ of some physical quantity $O(N,M,L_x,L_y)$ is defined as

TABLE I. The number $D(l,b)$ of the states that have the length l and the b times folds for an 11-step random walk, obtained by exact enumeration on the computer.

l \ b	0	1	2	3	4	5	6	7	8	9	10	11
1	0	0	0	0	0	0	0	0	0	0	0	1
2	0	0	0	0	0	1	7	20	30	25	11	0
3	0	0	0	1	10	56	102	140	80	30	0	0
4	0	0	1	16	74	139	192	100	55	0	0	0
5	0	0	6	38	84	140	84	70	0	0	0	0
6	0	1	13	38	86	70	77	0	0	0	0	0
7	0	2	8	34	32	56	0	0	0	0	0	0
8	0	2	12	20	44	0	0	0	0	0	0	0
9	0	2	4	18	0	0	0	0	0	0	0	0
10	0	2	11	0	0	0	0	0	0	0	0	0
11	0	2	0	0	0	0	0	0	0	0	0	0
12	1	0	0	0	0	0	0	0	0	0	0	0

$$\langle O(N,M,L_x,L_y)\rangle \equiv \sum_{L_x,L_y=1}^{L} \sum_{N,M=0}^{L-1} D(L_x,N)D(L_y,M)O(N,M,L_x,L_y)\exp[-U_{N,M}(N,M;L_x,L_y)]/Z[u,\omega,L] \ . \tag{3.4}$$

The specific heat, mean area, and mean folding number are calculated as

$$(\text{specific heat}) \equiv \langle U_{N,M}^2 \rangle - \langle U_{N,M} \rangle^2 \ ,$$

$$(\text{mean area}) \equiv \langle L_x \times L_y \rangle \ ,$$

$$(\text{mean bending number}) \equiv \langle N_{\text{total}} \rangle \equiv \langle N+M-2 \rangle \ .$$

In what follows, we present the results of numerical studies. Figure 4 depicts the mean bending number, the mean area, and the specific heat for membranes of size $L=30\times30$ for parameters u and ω in the range $-0.3<u,\omega<0.3$. These seem to imply that there are (at least) three different phases. In the region $\omega>0$ and $\omega>-2u$, the membrane is in the completely flat phase. The mean bending number is zero and the mean area is maximam L^2. In the region $u<0$ and $\omega<-2u$, the membrane is in the completely folded state. The mean bending number is $2(L-1)$ and the mean area is 1. In the remaining region ($u>0,\omega<0$), the membrane is in

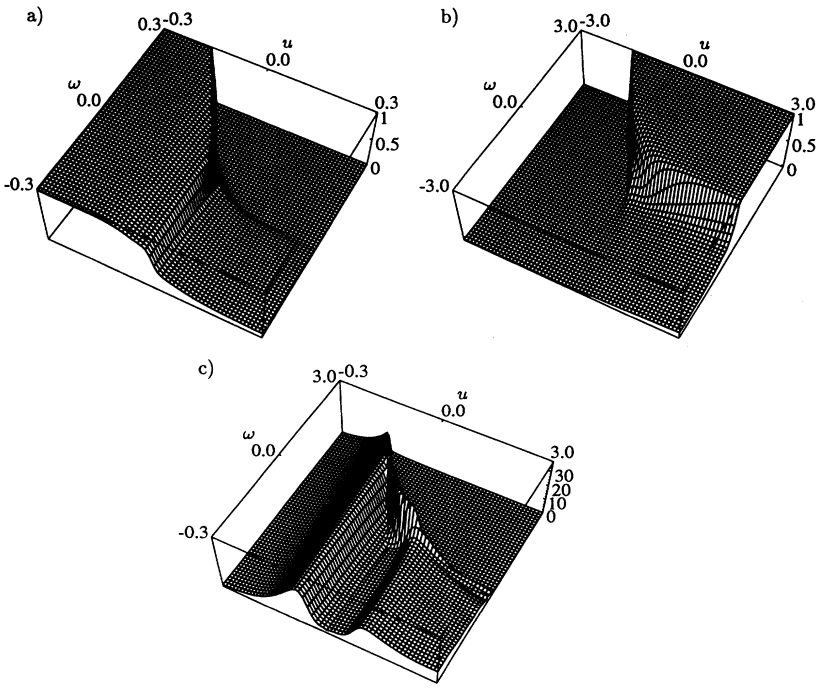

FIG. 4. Mean bending number (a), mean area (b), and specific heat (c) for squares lattice of size $L=30\times30$ for parameters u and ω in the range $-0.3<u,\omega<0.3$. The first two are normalized by their maximum values.

the partially folded state. Both the mean bending number and the mean area are small but nonzero. We also find a first-order transition from the completely flat to the completely folded state at the line $2u+\omega=0$. Among them all, the emergence of a "partially folded state" on the fourth quadrant of the (u,ω) plane is interesting. In this region, in order to minimize the potential of the system, both bending number and lengths L_x, L_y have to be small. From Table I, we can see that such a configuration does not exist and equilibrium configuration has to manage it. In the previous section, by balancing these energies, the mean bending number and the mean area are estimated as $N_{\text{total}} \sim L^{1/3}$ and $\langle L_x \times L_y \rangle \sim L^{4/3}$. We have estimated the exponent μ of the mean bending number $\langle N_{\text{total}} \rangle \sim L^{\mu}$ as $\mu \simeq 0.39 \pm 0.05$. We also evaluated the exponent ν of the mean area $\langle L_x \times L_y \rangle \sim L^{\nu}$ as $\nu \simeq 1.30 \pm 0.07$ (Fig. 5). The prediction of the mean-field theory is good. When there is no interaction $(u=0, \omega=0)$, the membrane is a direct product of two random walks in one direction [8]. In this free case, the mean bending number is of course $\frac{1}{2}L$ and the exponent $\mu=1.0$. From Fig. 5(c) the exponent for the mean area is $\nu=1.55$. This means that the swell of the partially folded state is smaller than that of the free case.

Next, we will discuss the phase transition between these phases. Previously we have discussed the phase transition between the completely flat phase and the completely folded phase at a line $2u+\omega=0$; now we will discuss the other two cases. At first, we study the transition from the completely flat phase to the partially folded phase. In the previous section, we discussed that the membrane with finite L shows a sequential folding transition. Figure 6 shows the behavior of the membrane. We have fixed the bending rigidity at $u=0.5$. Figure 6(a) shows the behavior of the mean bending number as we change the strength of attractive interaction ω, which is scaled with u/L, for system size $L=8,16,24$. In each system size, the curve runs up in a rather discrete manner like a staircase and its value at each plateau is almost integers. The length of each plateau becomes longer as ω becomes large. A more detailed graph around the origin for the membrane of size $L=30\times 30$ is shown in other figures [Fig. 6(b)], with the behavior of the mean area [Fig. 6(c)]. One notices that the first "jump" of the bend-

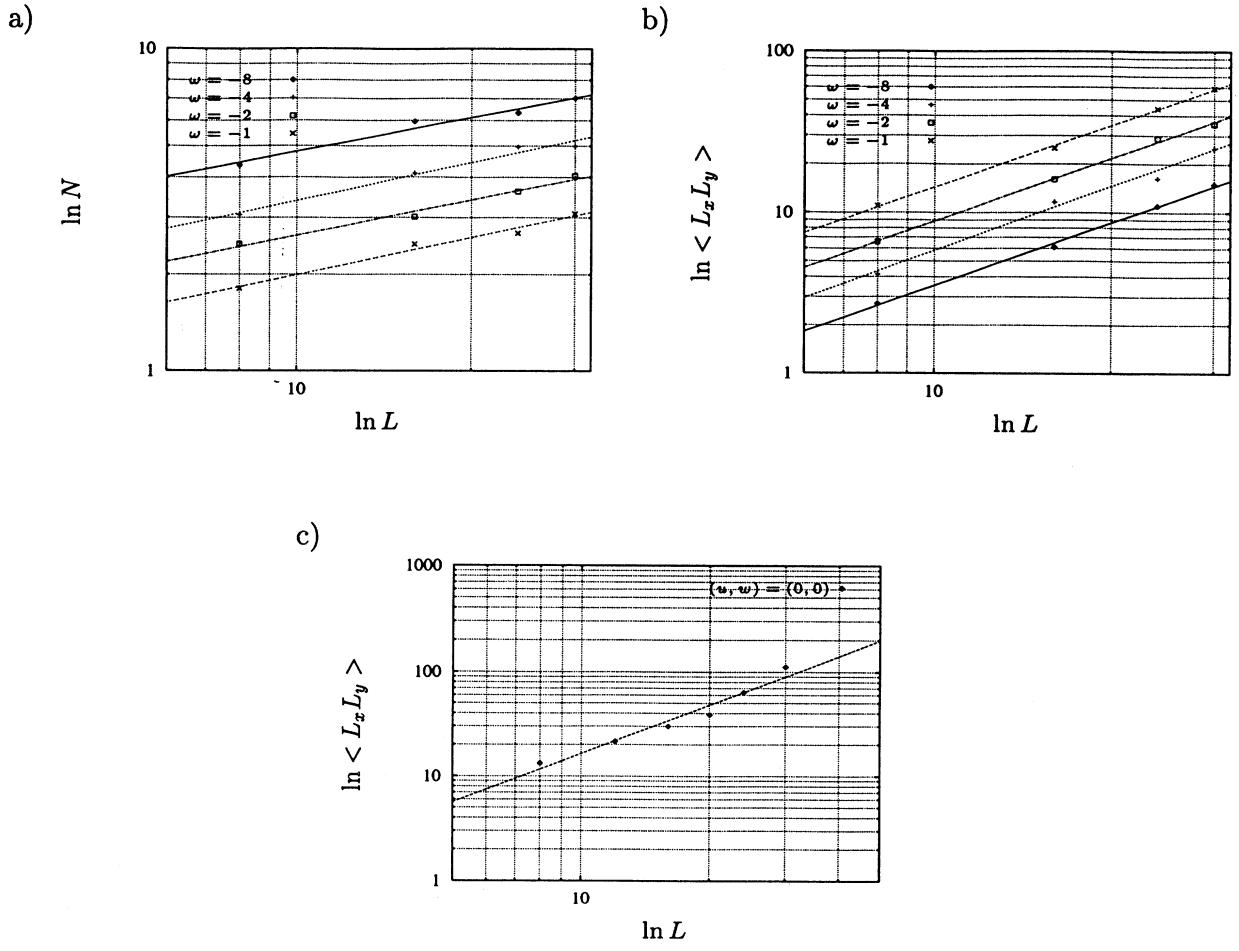

FIG. 5. System size dependence of mean bending number (a) and of mean area (b). Bending rigidity u is fixed at $u=0.5$. The lines indicate a fit of the data yielding exponents μ and ν (mean bending number $\sim L^{\mu}$ and mean area $\sim L^{\nu}$). The exponents are $\mu=0.39\pm0.05$ and $\nu=1.30\pm0.07$. (c) System size dependence of mean area for free system $(u,\omega)=(0,0)$ $(\nu=1.55)$.

ing number from zero to one occurs at $w=-2u/L$ while the area becomes half of L^2, which implies that the membrane folds on itself and a crease neatly divides it in half. This can be easily understood from the viewpoint of the energetic competition between flat and singly folded state in the previous section. As the bending number becomes large, the energy gain by a fold becomes small and the length of plateau becomes longer. We conclude that this behavior of the membrane is a sequential folding transition of the membrane.

The phase transition between the partially folded state and the completely folded state is discontinuous. Because the mean bending number $\langle N_{\text{total}} \rangle$ changes its behavior from $\langle N_{\text{total}} \rangle \sim L^{1/3}$ to $\langle N_{\text{total}} \rangle = 2(L-1)$ and this change is not continuous. In addition, we can easily describe the double peak in the specific heat for the case $\omega=0$ [13] (see Fig. 7). These peaks approach each other as the system size L becomes large and in the thermodynamic limit ($L \to \infty$), it becomes δ-function-like and corresponds to a first-order transition. These two peaks clearly remain in the domain $\omega<0$ and we think that the transition from the partially folded ($u>0$) to a completely folded state ($u<0$) is a first-order transition.

IV. DISCUSSIONS AND CONCLUSIONS

In this paper, we have studied the square lattice with bending rigidity u and interaction ω. As an interaction between different elementary squares of the lattice, we have discussed two types. The first one is a potential that is proportional to contact area (CA), and the second one is a potential that is proportional to the number of con-

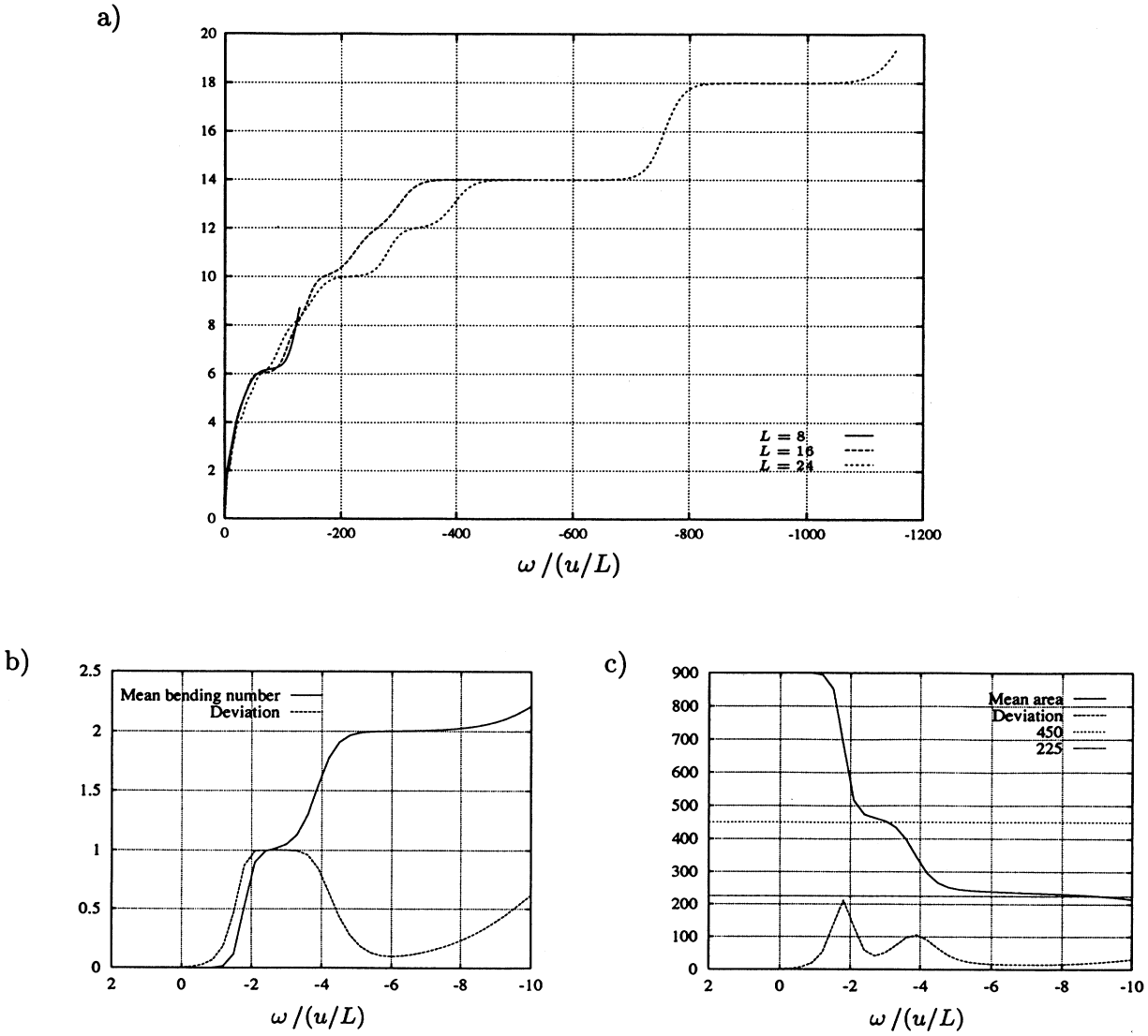

FIG. 6. (a) Mean bending number $\langle N_{\text{total}} \rangle$ vs strength of attractive interaction ω, which is scale with u/L for $L=8,16,24$ at $u=0.5$. Mean bending numbers are quantized and plateaus emerge. (b),(c) More detailed graphs for mean bending number and mean area around the origin for $L=30$. When a first folding transition occurs, the mean area becomes half. This means that a crease neatly divides the membrane in half.

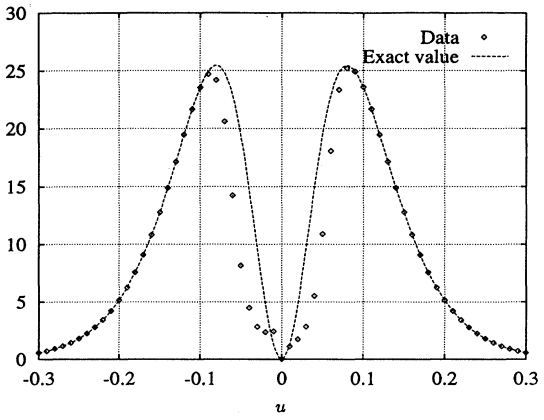

FIG. 7. Specific heat for $L=30$ with $\omega=0$. The solid line is exact results (see Ref. [13]) and the dots are from numerical data.

tact pairs of elementary squares (CP). Especially, we have analyzed the stability of the completely flat phase ($u>0$) with respect to attractive interactions ($\omega<0$) and that of the completely folded state ($u<0$) with respect to repulsive interactions ($\omega>0$). Both attractive interactions destroy the flat phase and these systems show a sequential folding transition when the system size is finite. However, the resulting states are different. In the CA case the membrane is partially folded and in the CP case the membrane is completely folded. Repulsive interactions also destroy the completely folded state. In the CA case, there occurs a first-order phase transition between a completely folded state to a completely flat state at the line $2u+\omega=0$. In the CP case, any amount of repulsive interaction makes the membrane flat. These results are summarized in Fig. 3.

We have also performed numerical studies for the CA case and confirmed several theoretical results. Especially, there occurs a sequential folding transition. This means that we can use the square lattice model with attractive interactions as a simple model for the sequential folding transition of a tethered membrane. Thermodynamical behavior of the partially folded state is also studied and its total folding number and its area behave as $N_{\text{total}}\sim L^{0.39}$, $\langle L_x L_y\rangle\sim L^{1.30}$. This phase is more "compact" than the phase of the free membrane $(u,\omega)=(0.0,0.0)$. On the other hand, the behavior of the membrane with CP interaction in the region $(u>0,\omega<0)$ is not clear when the three-body force exists. We cannot compare the behavior of the membrane with these interactions. Experiments on the polymerized membrane in poor solvent showed that the membrane is in a compact phase after several foldings [17,18]. However, we cannot decide which interaction is better than the other interaction in our analysis. From this point of view, the square lattice model with bending rigidity and CP interaction and three-body interaction deserves for future study.

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[13] In the free case $\omega=0$, we can calculate the partition function and other thermodynamic quantities for the discrete system as

$$Z[u,\omega=0,L]=\{1+\exp(-uL)\}^{2(L-1)},$$

$$N_{\text{total}}=2(L-1)/\{1+\exp(uL)\}$$

$$\rightarrow\begin{cases}0, & \text{as } u\rightarrow\infty\\ L-1, & u\rightarrow 0\\ 2(L-1), & u\rightarrow-\infty\end{cases},$$

$$C(\text{specific heat})=u^2\frac{\partial^2}{\partial u^2}\ln Z(u,\omega=0,L)$$

$$=2(L-1)\frac{(uL/2)^2}{\cosh^2 uL/2}.$$

These results fit very well with numerical studies on the u axis (Fig. 7).
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[16] The free energy difference with system size $L\times L$ between the completely flat phase and the one folded state with a crease at its edge is $\delta F=(u+\omega)L$ and it is positive in the region $2u+\omega>0$ and $\omega>0$. Before the one folded state appears, the completely folded state appears at the line $2u+\omega=0$ and the intermediate folded state does not

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