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Discrete solid-on-solid model of interface with bending rigidity: Roughening versus crumpling

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We propose a discrete solid-on-solid model of interface having a specific type of bending rigidity; this type of bending rigidity was introduced originally by Widom [J. Chem. Phys. **84**, 6943 (1986)] to account for the energy cost associated with the bending of the oil-water interface in a lattice model of ternary microemulsions. We calculate the width of the interface as well as various height-height and normal-normal correlation functions through Monte Carlo simulation. We show that when this specific ''bending energy'' alone controls the out-of-plane thermal fluctuations of our discrete interface, the membrane is crumpled at all nonvanishing temperatures, but the nature of this crumpling is very different from that observed in the standard continuum models. We also show the effects of a pinning potential and present results on some nonequilibrium properties of the model. [S1063-651X(96)08508-X]

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I. INTRODUCTION

Amphiphilic molecules consist of a ''hydrophilic head'' and a ''hydrophobic tail''; the polar head prefers to be in contact with water, whereas the hydrocarbon chain constituting the tail tends to minimize contact with water [1]. Therefore, in a ternary microemulsion consisting of water, oil, and amphiphiles, a monolayer of amphiphiles is formed at the oil-water interface in such a way that the heads of the amphiphiles remain in contact with water while the tails remain dipped in oil [2]. Similarly, amphiphiles form bilayers in an aqueous medium; the amphiphiles in the two constituent monolayers of the bilayer are oppositely oriented so that the hydrophobic tails of all the molecules remain inside, thereby minimizing contact with water. The plasma membrane of red blood cells is a prototype example of such bilayers [3]. The monolayers and bilayers of amphiphilic molecules are often referred to as amphiphilic membranes.

The nature of the out-of-plane thermal fluctuations of interfaces and amphiphilic membranes as well as their effects on the equilibrium conformations have been investigated extensively over the last ten years [4–10] In the case of fluid interfaces, these fluctuations are controlled by the interfacial tension. On the other hand, the effective interfacial tension of amphiphilic membranes can be vanishingly small and, therefore, their out-of-plane thermal fluctuations are controlled by the bending rigidity [11]. The interfaces can exhibit a transition from a ''smooth'' phase to a ''rough'' phase at a temperature known as the roughening temperature [12]. Similarly, depending on the nature of the in-plane ordering, an amphiphilic membrane can exhibit a *crumpled* phase at sufficiently high temperatures [4–10]

The roughening and crumpling of isolated planar membranes have been studied using models in the solid-on-solid (SOS) approximation; there are neither ''overhangs'' nor ''droplets'' of the minority phase inside the majority phase in a SOS model. The results obtained from the discrete and continuum models of interfaces have been found to be qualitatively consistent with each other [13]. In this paper we propose a discrete SOS model for amphiphilic membranes, where the bending energy of the membrane is incorporated following a prescription suggested earlier by Widom [14] in order to account for the bending rigidity of the membrane at the oil-water interface in a lattice model of ternary microemulsions. (We use the terms interface and membrane interchangeably). We study the out-of-plane thermal fluctuations of this discrete SOS model using Monte Carlo (MC) simulations. We also investigate some dynamical aspects of this model through MC simulation and compare the results with the corresponding results derived analytically using a master equation approach.

II. MODEL

For simplicity, we formulate the model as a one-dimensional membrane (a curve) embedded in a two-dimensional space. Suppose the sites on a one-dimensional lattice parallel to the X axis (assumed horizontal) are labeled by the integers i $(i=1,2,3,\ldots,L)$ and the height of the membrane, measured from the reference horizontal line, is given by a single valued height function; h_i, the height at the ith site, can take only integer values. If h_i is constant and independent of i, then the membrane is flat everywhere. On the other hand, unequal heights at neighboring sites gives rise to bending of the interface; these bends are identical to the bendings of the amphiphilic monolayer at the oil-water interface in the Widom model.

The Hamiltonian for the model membrane proposed here is given by

$$H=j\sum_i |h_i-h_{i+1}|+k\sum_i (1-\delta_{h_i,h_{i+1}}), \tag{1}$$

where $\delta_{h_i,h_{i+1}}$ is the Kronecker δ function and the constant k is a measure of the bending stiffness of the membrane. However, note that an energy cost k is associated here with every bending of the interface over a single lattice spacing, which is also assumed to be identical to the linear size of all the molecules in the system [15]. In the limit $k=0$, this model reduces to the standard SOS model of an interface. This SOS model is quite different from the two SOS approximations to the Widom model introduced earlier by

Kahng *et al.* [16]. A comparison of the model of Kahng *et al.* and the model (1) proposed here will be made later in this paper. Our aim is to elucidate the consequences of the difference in describing the bending in the ''continuum scenario'' and ''discrete scenario.''

We have applied the periodic boundary condition, i.e., $h_{L+i}=h_i$. In our MC simulation the system is allowed to evolve, starting from a specific initial condition, following the standard Metropolis algorithm [17]. We work with the parameters $J=j/(k_BT)$ and $K=k/(k_BT)$, where T is the temperature and k_B, the Boltzmann constant, is chosen to be unity in our units. Note that if the magnitudes of J and K are increased (decreased) simultaneously keeping the ratio J/K constant, it effectively mimics a decrease (increase) of the temperature for fixed j and k.

III. DEFINITIONS OF THE CHARACTERISTIC QUANTITIES OF INTEREST

The width W of the interface is defined through the relation

$$W^2(L,t)=\{\langle[h_i(t)-\langle h\rangle]^2\rangle\}_{\rm av}, \tag{2}$$

where $h_i(t)$ $(i=1,2,\ldots,L)$ is the height at the ith site at time t, the symbol $\langle X\rangle$ implies the average of X over all the lattice sites, i.e., $\langle X\rangle=(1/L)\Sigma_{i=1}^{L}X_i$, whereas the symbol $\{Y\}_{\rm av}$ implies average of Y over a large number of MC runs. The quantity $W(L,t)$ is monitored, for fixed L, as a function of time t; $W(L,\infty)$ is the width of the system in equilibrium.

We have also computed three different correlation functions. The *height-height* correlation function $C(x)$ is defined as

$$C(x)=[(h_0-h_x)^2]_{\rm av}. \tag{3}$$

The correlation function $G(x)$ is defined as

$$G(x)=[(h_0-\langle h\rangle)(h_x-\langle h\rangle)]_{\rm av}. \tag{4}$$

In the continuum formulation, de Gennes and Taupin [18] studied the correlation between the normals to the membrane at two spatially separated points. We have introduced a discrete counterpart of this normal-normal correlation appropriate for our discrete model. Only three directions are possible for the normal to our discrete interface—it can be directed either towards the left $(-\hat{x})$ or the right $(\hat{x})$, or it can be vertical. Therefore, we can define the ''normal'' n_i as follows:

$$n_i=+1 \quad \text{if} \quad h_i>h_{i+1},$$

$$n_i=0 \quad \text{if} \quad h_i=h_{i+1},$$

$$n_i=-1 \quad \text{if} \quad h_i<h_{i+1}.$$

In terms of these ''normals'' the *normal-normal* correlation function $N(x)$ for our discrete model is defined as

$$N(x)=[n_0n_x]_{\rm av}. \tag{5}$$

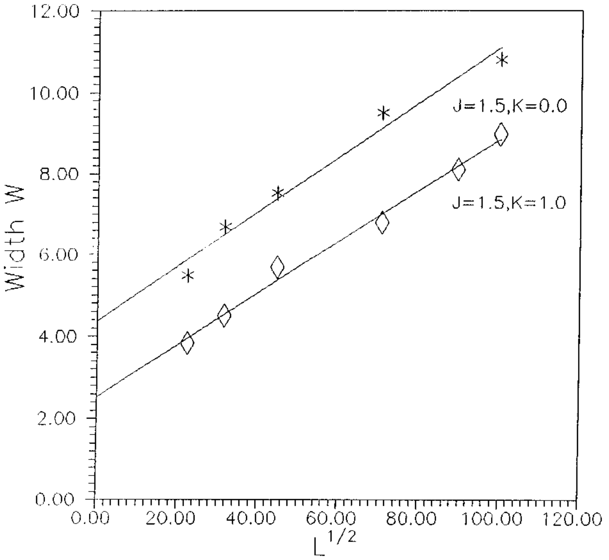

FIG. 1. The equilibrium width W of the interface plotted against $L^{1/2}$ for two different sets of values of the parameters J and K, namely, $J=1.5$, $K=0.0$ and $J=1.5$, $K=1.0$. The lines through the data points are the corresponding best straight-line fits.

IV. RESULTS AND DISCUSSION

In order to test our algorithm and computer program we have calculated $W(L,\infty)$ as a function of L first in the special case $K=0$, $J=1.5$, i.e., for the standard SOS model. From Fig. 1 we find that in this special case of $J\neq 0$, $W(L,\infty)\propto\sqrt{L}$, in agreement with the known result for the standard SOS model.

Next, we computed the width of the interface as a function of L for the more general case $K\neq 0$. $J\neq 0$ simultaneously. In order to test the effect of nonvanishing K on the roughness of the SOS interface we chose the same value of J, namely, $J=1.5$, as before, while K was chosen to be 1.0. It is clear from Fig. 1 that the nonvanishing bending rigidity tends to suppress the out-of-plane thermal fluctuations thereby reducing the width of the interface, but the roughening exponent remains the same as in the special case $K=0$. In other words, in the general cases, when both J and K have nonvanishing values, the interface is rough but not crumpled.

The advantage of the SOS model (1) is that one can ''switch off'' J and thus choosing nonvanishing K can have a discrete model of interface with purely bending rigidity only. Therefore, we next computed $W(L,t)$ in the other special case $J=0$, $K\neq 0$. We found that $W(L,t)$ kept increasing forever with increasing time t. In Fig. 2 the solid curves represent $W(L,t)$, plotted against $L^{1/2}$ for three different values of t, when $K=3.0$; the corresponding data for $K=1.0$ are shown by the dashed curves in the same figure. Note that smaller K corresponds to an effectively higher temperature T. From Fig. 2 it is clear that (a) for a fixed L the interface is wider at a higher temperature than at a lower temperature after the elapse of the same time interval beginning from the flat conformation at $t=0$, and (b) at any given t, W is independent of L but $W(L,t)\to\infty$ as $t\to\infty$ for all L.

Thus the actual area of the membrane in equilibrium is infinitely large, if $J=0$ but $K\neq 0$, at all $T\neq 0$, *even for all finite values of the basal area (L in d=1).* Does it necessarily

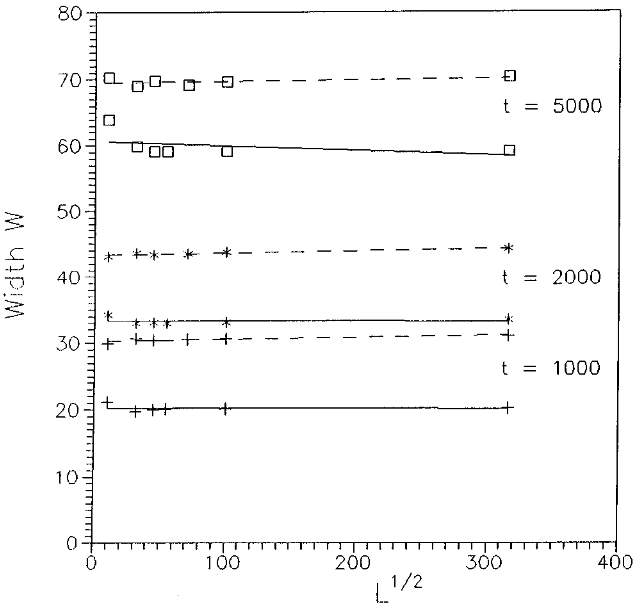

FIG. 2. The width W of the interface plotted against $L^{1/2}$ for the parameter values $J=0.0$, $K=3.0$ (full curves) and $J=0.0$, $K=1.0$ (dashed curves) at three different times, namely, $t=1000$, $t=2000$, and $t=5000$ Monte Carlo steps.

imply that the membrane is crumpled at all nonvanishing temperatures?

In order to answer this central question we shall now summarize the three different definitions of crumpling. All these definitions yield identical results when applied to standard continuum models of membranes and, therefore, are usually regarded as equivalent; however, our discrete lattice model, with $J=0$ and $K\neq 0$, satisfies the criterion of crumpling set by one of these definitions, but appears to be smooth when examined against the two other definitions. (i) If the ratio of the excess area δA created by thermal fluctuations and the original basal area A of the membrane diverges in the thermodynamic limit, the membrane is crumpled. This criterion of crumpling is certainly satisfied by our lattice model (1) at all $T\neq 0$ if $J=0$ but $K\neq 0$. (ii) If the interfacial width W scales as $W\sim L^{\zeta}$ with $\zeta>0$, the membrane is rough. Moreover, if $\zeta>1$ for $L\ll\xi_p$, where ξ_p is the persistence length [18], then the membrane is crumpled on length scales much longer than ξ_p; thus all crumped membranes are rough but the converse is not necessarily true [7]. From Fig. 2 we conclude that, after very early times (when no clear trends emerge due to the presence of transients), one can extract an effective roughening exponent ζ_{eff} for all longer times, and one finds that $\zeta_{\mathrm{eff}}=0$. This, however, would imply that at all times, except the very early stages, during its growth the membrane remains effectively smooth. (iii) If the correlation between the normals to the membrane at two spatially separated points follows $\langle\vec{n}(\vec{r})\vec{n}(0)\rangle\sim e^{r/\xi_p}$, then the membrane is defined as crumpled on length scales much larger than ξ_p [18]. But we neither find any exponential decay in the "normal-normal correlation function" $N(x)$ nor observe any qualitative difference in $N(x)$ computed for $K=1.0$ and $K=5.0$ [see Figs. 3(a) and 3(b)], which correspond to an interface of fixed bending rigidity k at two different effective temperatures.

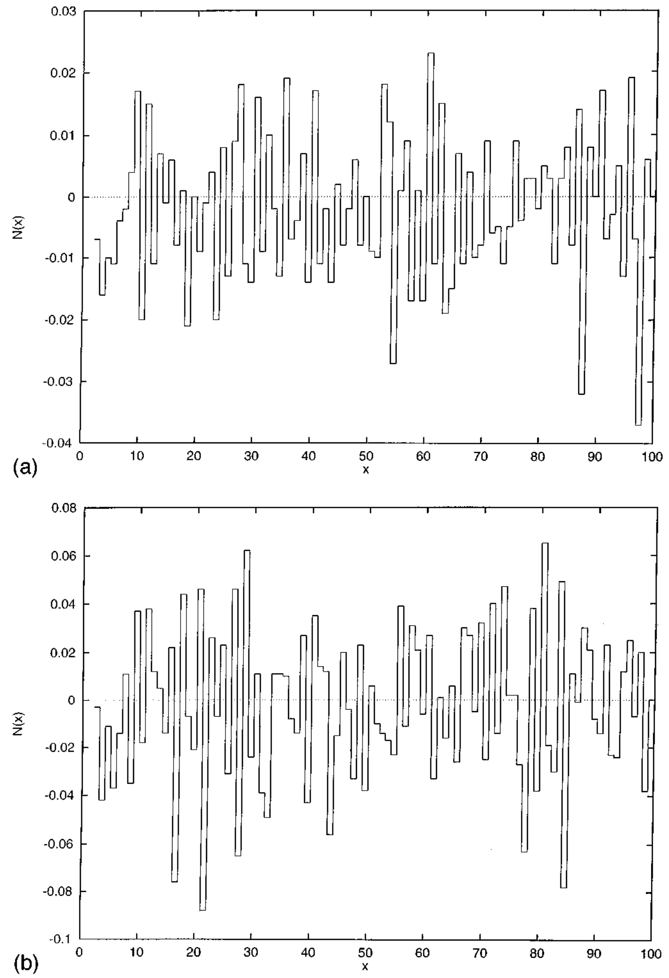

FIG. 3. The "normal-normal correlation function" $N(x)$ for (a) $K=5.0$, (b) $K=1.0$.

In Figs. 4(a) and 4(b) we present the height-height correlation function $C(x)$ for $K=5.0$ and $K=1.0$, respectively. The corresponding correlation functions $G(x)$ are plotted in Figs. 5(a) and 5(b), respectively. Noting, as before, that $K=1.0$ corresponds to an effectively higher temperature than $K=5.0$ for the same fixed bending stiffness k, we conclude that the only effect of a higher temperature in both these figures is the larger magnitude of the corresponding correlation function.

In order to understand these unusual features of the out-of-plane thermal fluctuations in model (1), let us compare schematic typical configurations for this model with those of standard continuum models. Since wiggles exist in the rough phase of the continuum model on all length scales (smaller wiggles are part of the bigger ones which, in turn, are parts of even bigger ones, and so on) the width increases with increasing L. The absence of wiggles of all sizes leads to $\zeta=0$ in our lattice SOS model. Moreover, it is obvious from these schematic representations of typical configurations why the normal-normal correlation exhibits a periodicity in our lattice SOS model. Note that the diverging area of the lattice membrane in equilibrium arises from the diverging area (length in $d=1$) of the vertical regions of the membrane.

Now, one may be curious to find out the effect of a pin-

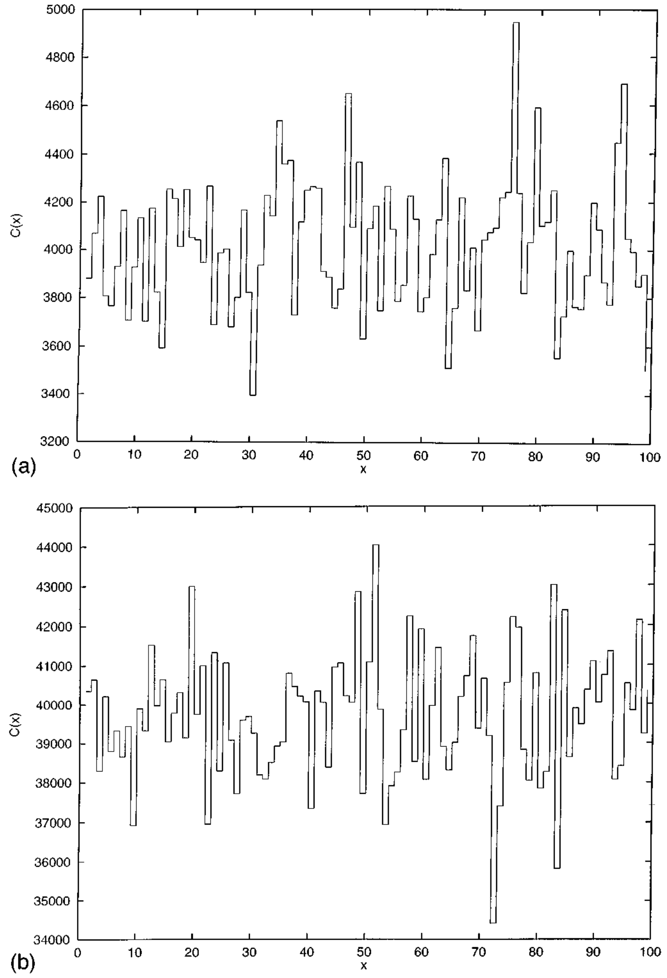

FIG. 4. The "height-height correlation function" $C(x)$ for (a) $K=5.0$, (b) $K=1.0$.

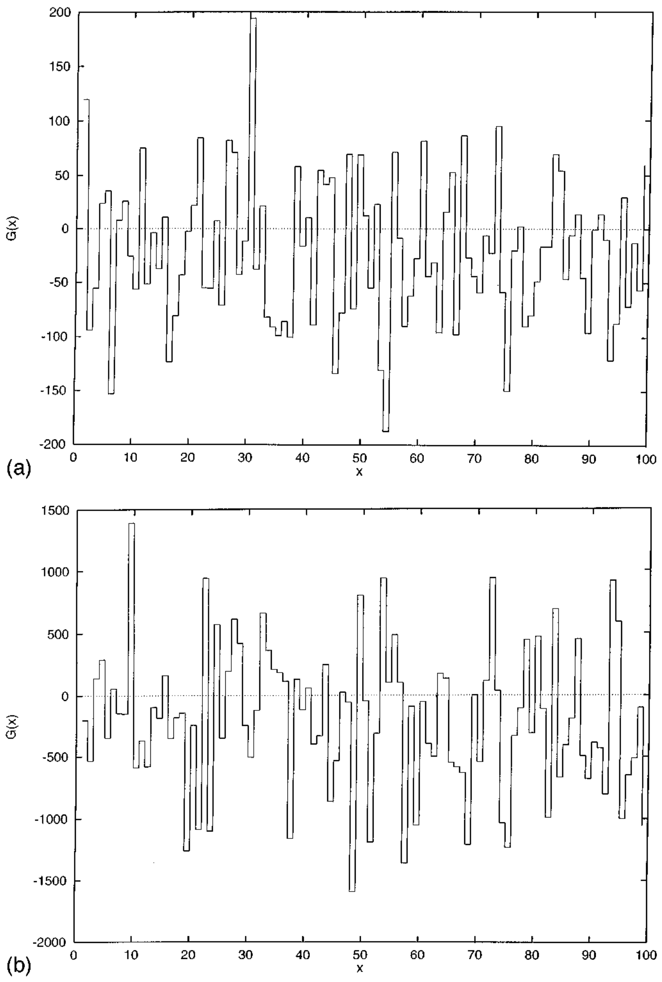

FIG. 5. The correlation function $G(x)$ for (a) $K=5.0$, (b) $K=1.0$.

ning potential, e.g., gravity, on the equilibrium structure of the interface. We choose the pinning potential to be $g\Sigma_{i=1}^{L}|h_i|$, where the constant g determines its strength. From now on, the symbol G will denote $G=g/(k_BT)$. In the standard SOS limit, i.e., when $K=0$ but $J\neq 0$, we observe that (Fig. 6) the interface becomes smooth, even for pinning potentials as weak as $G=0.015$. In the other limiting case, namely, $J=0$, but $K\neq 0$, we found that (Fig. 7) even pinning potentials as weak as $G=0.03$ lead to a finite value of the width of the interface in equilibrium. Moreover, since the pinning potential tends to suppress out-of-plane thermal fluctuations, the larger value of G leads to a smaller value of the equilibrium thickness of the interface.

In the absence of any pinning potential we did not observe any qualitative difference in the correlation functions at different temperatures, except the difference in their magnitudes. But, in the presence of a nonvanishing pinning potential, we observed interesting qualitative differences in the nature of the correlation functions evaluated at different temperatures. The correlation functions $N(x)$, $C(x)$, and $G(x)$ are plotted in Figs. 8–10, respectively, in each of which part (a) corresponds to $K=4.0$, $G=0.4$, whereas part (b) corresponds to $K=1.0$, $G=0.1$. Note that the effective temperature of the system corresponding to parts (b) is four times that of parts (a) in each of Figs. 8–10. Clearly, there are

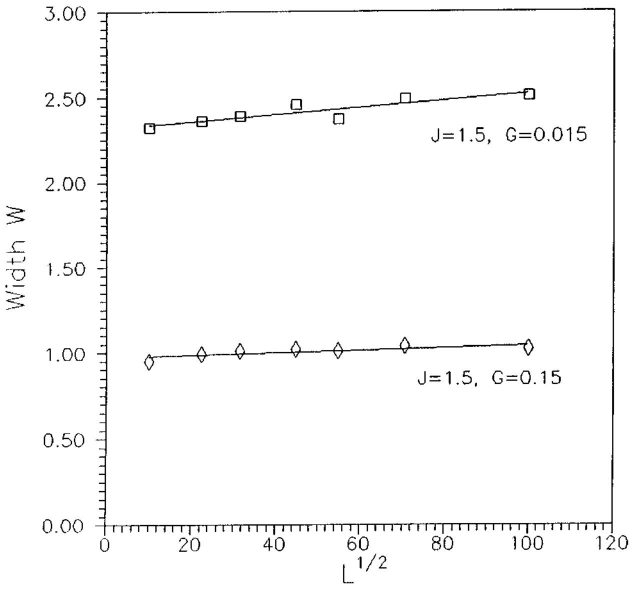

FIG. 6. The equilibrium width W of the interface plotted against $L^{1/2}$ for two different sets of the parameters J, K, and G, namely, $J=1.5$, $G=0.015$, and $K=0.0$ and $J=1.5$, $G=0.15$, and $K=0.0$.

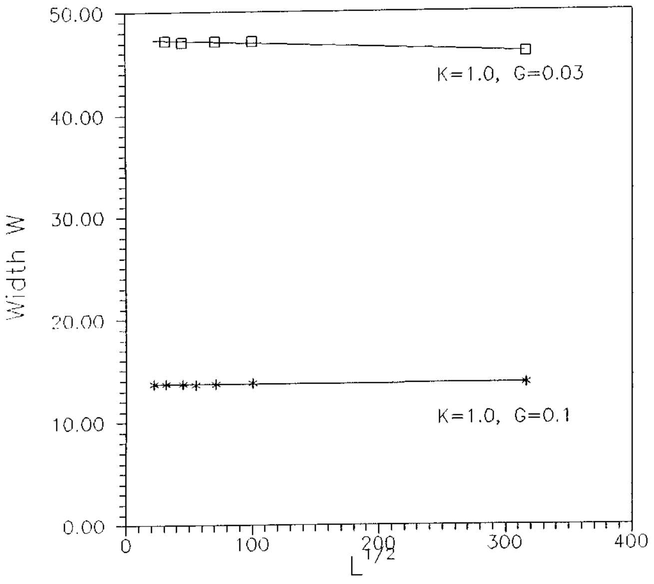

FIG. 7. The equilibrium width W of the interface plotted against $L^{1/2}$ for two different sets of the parameters J, K, and G, namely, $K=1.0$, $G=0.03$, and $J=0.0$ and $K=1.0$, $G=0.15$, and $J=0.0$.

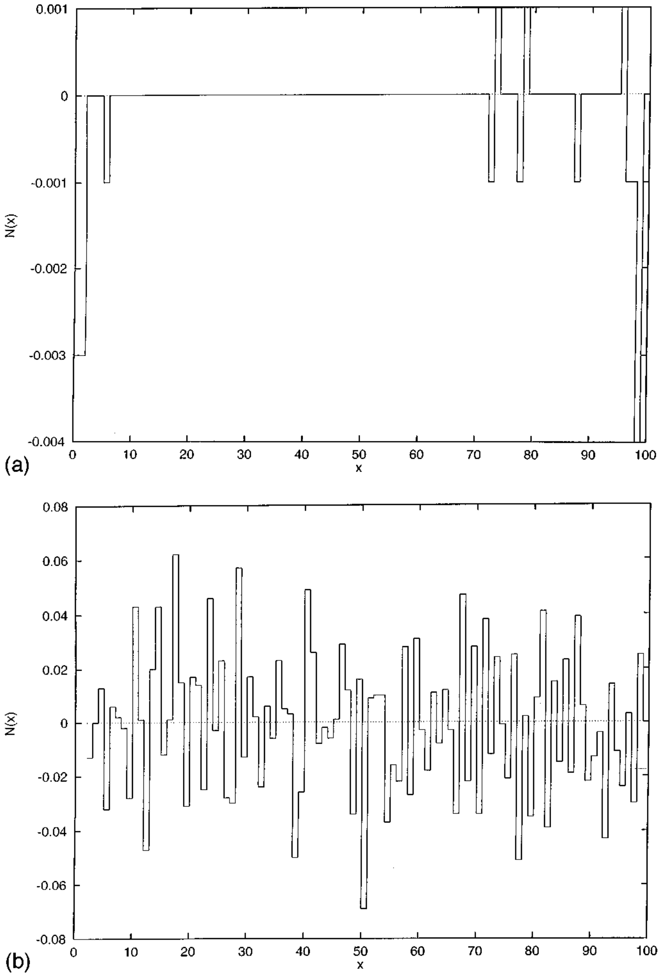

FIG. 8. The "normal-normal correlation function" $N(x)$ for (a) $K=4.0$, $G=0.4$, (b) $K=1.0$, $G=0.1$.

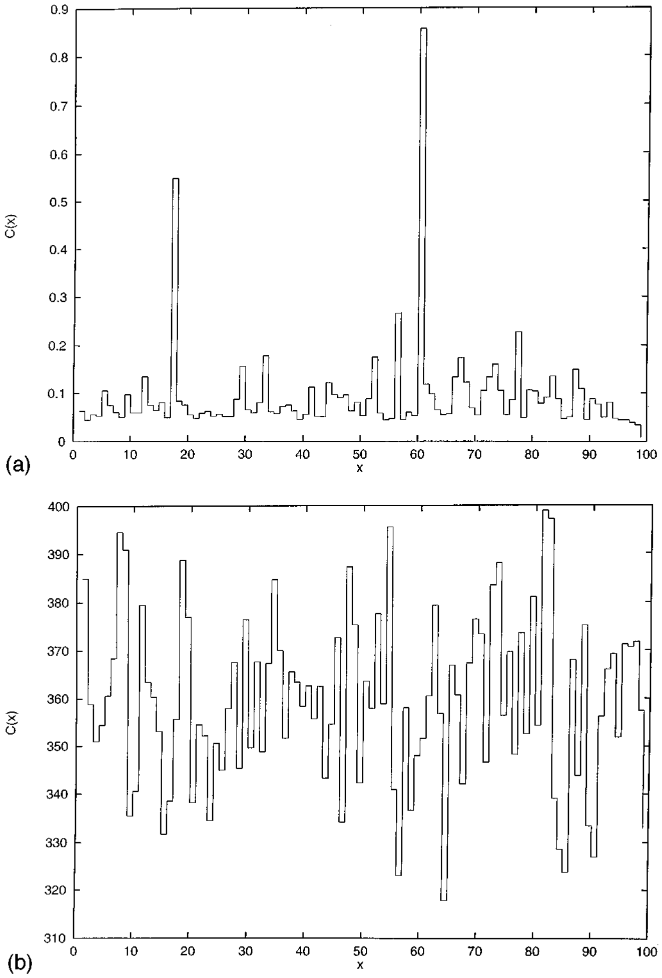

FIG. 9. The "height-height correlation function" $C(x)$ for (a) $K=4.0$, $G=0.4$, (b) $K=1.0$, $G=0.1$.

wider flat portions of the interface at lower temperatures in equilibrium.

Finally, we studied the dynamics of the model interface analytically [19] in the special limit $J=0$, $G=0$, but $K\neq 0$, by using a standard master equation approach [20]. The width was found to grow with time following the "growth law" $W^2\propto t$. Our MC data in Fig. 11 are consistent with this growth law.

We can now compare our model with the model of Kahng *et al.* [16]. The special SOS models considered by Kahng *et al.* do *not* include any bending energy of the type included in our model. Thus, interactions in their SOS models are similar to the first term on the right-hand side of our Eq. (1). But, in addition to the interaction between the nearest-neighbor columns, they also allow interactions between farther-neighbor pairs of columns; the latter was motivated by the similar farther-neighbor interactions between the spin pairs in the Widom model [14]. The roughening of the interface in the Widom model has been investigated in two dimensions recently by Maiti and Chowdhury [21].

One common feature of our model and the Widom model [14] is that there are bending energies associated with bendings over one lattice spacing, which is assumed to be identical to the size of each molecule. Another lattice model, which associates bending energies over bendings of the in-

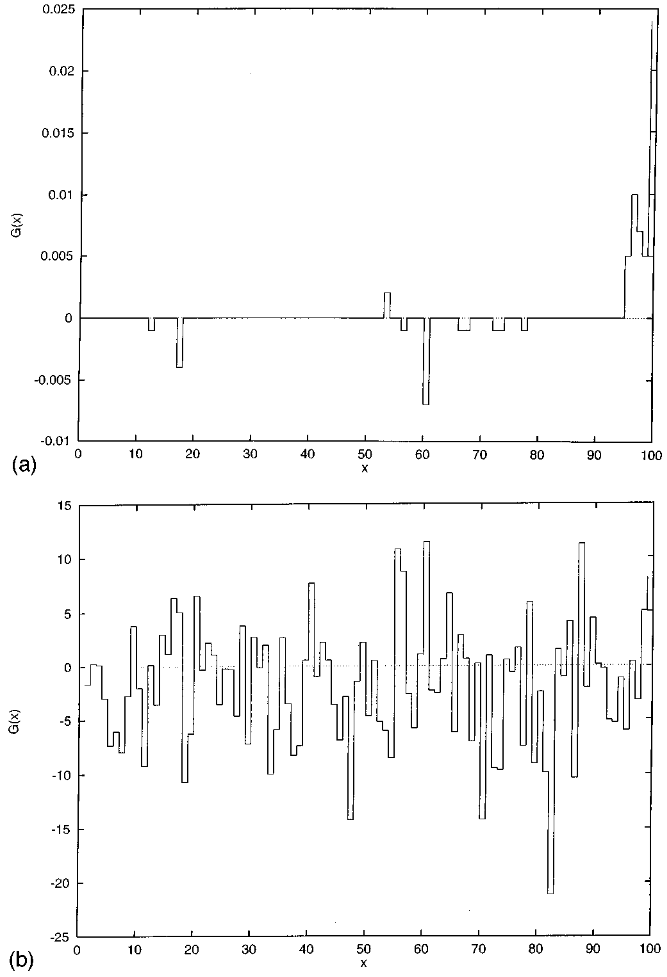

FIG. 10. The correlation function $G(x)$ for (a) $K=4.0$, $G=0.4$, (b) $K=1.0$, $G=0.1$.

terface over single lattice spacings, has been found to mimic reliably the energies associated with bendings of interfaces over truly molecular dimensions [22]. However, the reliability of such bending energies in mimicking the energies associated with bendings over longer length scales remains to be explored in detail.

V. CONCLUSION

In this paper we have proposed a discrete model of an interface with a specific form of the bending energy: this bending energy is associated with bending of the interface over as short a length scale as the size of a molecule. When the out-of-plane thermal fluctuations of this interface are controlled only by this bending stiffness, it is crumpled, but the nature of the crumpling is very different from that observed in the continuum models. We have shown that although the conformation of the interface in this case satisfies one particular definition of a crumpled manifold, it does not satisfy two other widely used definitions of crumpling although all these three dimensions are known to be mutually consistent when applied to the continuum models.

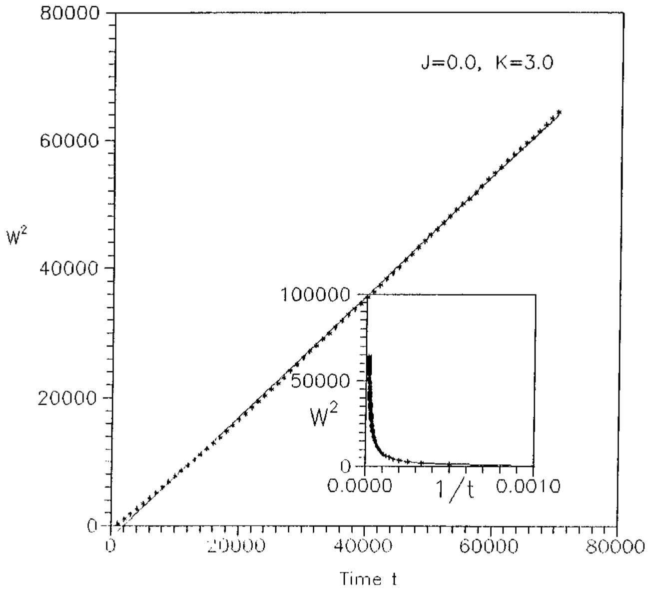

FIG. 11. The square of the interfacial width plotted against time t for $J=0.0$, $K=3.0$, and $G=0.0$. The inset shows the same data plotted against $1/t$ to emphasize the fact that, when J and G vanish but K has a nonvanishing value, the interfacial width diverges in the limit $t \rightarrow \infty$.

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