

Roughening of chemically reacting interfaces

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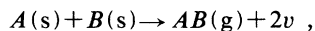
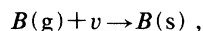
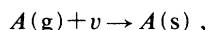
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There has been much interest recently in simple lattice-gas models of catalytic reactions. Using Monte Carlo simulations, we investigate interfacial roughening in a model of a Langmuir-Hinshelwood reaction which includes an effective repulsive interaction between the two reacting species. We measure the width of the reactive zone between the two reactants and the roughness of this interface. It is found that, when the interspecies repulsive interaction is nonzero, the interface roughness obeys the scaling law $w \sim t^\beta$, with $\beta = \frac{1}{4}$. When there is no interspecies interaction, the interface roughness scales as $t^{1/2}$. The reactive-zone width also scales as $t^{1/2}$ when there is no interspecies repulsion. With interspecies repulsion, however, we find that the reactive-zone width tends to a finite value at long times. We argue that in this case the interface should be described by the Kardar-Parisi-Zhang equation [Phys. Rev. Lett. **56**, 889 (1986)] with $\nu \neq 0$ and $\lambda = 0$.

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I. INTRODUCTION

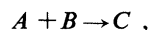
There is much interest in the structure of interfaces evolving under nonequilibrium conditions. The phenomenon of wetting [1–4] the surface structure of diffusion-limited aggregation (DLA) and Eden clusters [5], the structure of domain walls in lattice spin models [6,7], and thin-film growth and epitaxy [8–10] are some common examples. For lattice-gas models of Langmuir-Hinshelwood reactions, an interface exists between the two reactant species. Here, we investigate the roughening of this interface for a bimolecular model of a Langmuir-Hinshelwood reaction. The model that we study is described by the following elementary steps [11]:



where (g) and (s) denote gas-phase and adsorbed particles, respectively, and v denotes a vacant site on the lattice used to simulate the catalytic surface. The structure of the clusters formed in this model has previously been studied by Ziff and Fichthorn [12], who simulated the Langmuir-Hinshelwood reaction on an initially empty square lattice. As a result of reaction and subsequent desorption of nearest-neighbor AB pairs, clusters consisting entirely of A or entirely of B are formed. They suggested that these clusters are fractal, with a dimension of approximately 1.8 [12]. Meakin and Scalapino [13] investigated the time dependence of the vacancy density, and, on a square lattice, it was found to decay very slowly with time, approximately obeying the power law $N_v \sim t^{-0.056}$

[13]. In this paper we attempt to provide a better fundamental understanding of the time evolution of such reaction models by considering the evolution of the structure of an initially flat interface between domains of A and B . In particular, we show that the evolution of such an interface may be described by the ideas used in studying film growth.

It should be noted that interesting work [14–16] has been carried out on the reactive lattice-gas system



where the reactants A and B diffuse on the lattice. Beginning with a lattice on which the concentrations of A and B are $c_A = c_{A0}$ and $c_B = 0$ for $x < 0$, and $c_A = 0$ and $c_B = c_{B0}$ for $x > 0$, for the A and B are allowed to diffuse and react. The width of the reactive region is predicted to behave asymptotically as $t^{1/6}$ by a scaling theory [14], and this behavior is confirmed by a simulation study [15]. Here, we also study the width of the reactive region in our system, but, in addition, we consider the structure of the interface between the reactive region and each of the reactant domains. This interface is not defined in the $A + B \rightarrow C$ system.

II. SIMULATION AND DEFINITIONS

The bimolecular region system is simulated on a square lattice. Initially, the left half of the lattice is completely filled with particles of species A and the right half is completely filled with particles of species B . Two columns of sites between the A and B domains are left vacant in order to allow adsorption. Starting with this initial configuration, Monte Carlo simulations of the system are

performed, as described below. Periodic boundary conditions are applied in the vertical direction. The simulations are performed on sufficiently wide lattices that the reactive zone does not reach either the leftmost or the rightmost column of sites.

A site is picked at random, and an adsorption event is attempted. If the site is already occupied, the attempt fails, and the procedure is repeated from the beginning. If the site is vacant, a particle of species A or B is chosen for adsorption with equal probability. If A is chosen, the number n of nearest-neighbor sites occupied by particles of species B is counted. The adsorption attempt is then successful with a probability equal to s^n , where $0 \leq s \leq 1$. Similarly, the probability of success for adsorbing a B particle is given by s^n where n is the number of nearest-neighbor sites occupied by A particles. This ansatz simulates an effective repulsive interaction between the A and the B species [17]. If adsorption fails, the procedure is repeated from the beginning. If adsorption of an A particle is successful, a nearest-neighbor site occupied by a B particle is chosen at random and reaction occurs; the newly adsorbed A particle and the chosen nearest-neighbor B particle desorb immediately. If there are no B particles in the nearest-neighbor sites, no reaction occurs, and the procedure is repeated. The successful adsorption of a B particle is similarly treated. The parameter s , which occurs in computing the probability of adsorption of a particle into a vacant site, is varied between zero and unity in the simulations.

For each iteration, regardless of the success of the attempted adsorption event, time is incremented by one Monte Carlo step. In our results we report the time in units of Monte Carlo steps per site (MCS) for lattices of size $(10\,000 \times 200)$. Typical configurations of the lattice are shown in Figs. 1(a) and 1(b) for $s=1.0$ and 0.5 , respectively. These configurations are obtained after times of approximately 6×10^3 and 6×10^4 MCS, respectively. Each column of lattice sites is labeled by x , with x tending to $-\infty$ far to the left and x tending to $+\infty$ far to the right. We label the rows by y . It can be seen in these two figures that there are two kinds of A particles: those which are connected by nearest-neighbor A particles to $x = -\infty$, and those which are not similarly connected. Similarly, the B particles can be divided into two kinds according to their connectivity to $x = +\infty$. It is, therefore, possible to define an A domain consisting of all the A particles connected by other A particles to $x = -\infty$, and a B domain consisting of all the B particles connected by other B particles to $x = +\infty$. The external perimeter of each of these domains consists of particles which border a "reactive" zone between the two domains. This reactive zone consists of "islands" of A and B embedded in a "sea" of vacant sites. We take the external (reactive) perimeter of the A domain to consist of those A particles in the A domain which have nearest-neighbor vacant sites belonging to this sea of vacant sites. Within each of the A and B islands and within the A and B domains, there can also be "lakes" with islands embedded, and so on. Hence there are clusters of A , of B , and of vacant sites in the reactive zone.

We can associate a spin $\sigma = -1$ with each particle of

species A , a spin of $\sigma = 1$ with each particle of species B , and a spin $\sigma = 0$ with each vacant site. Then the average spin $\langle \sigma(x) \rangle = m(x)$ for each column of lattice sites can be defined. Using this quantity, we can measure the width of the reactive zone by $w_m = (\langle x^2 \rangle_m - \langle x \rangle_m^2)^{1/2}$, where the averages are defined by

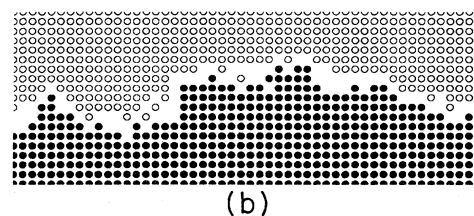
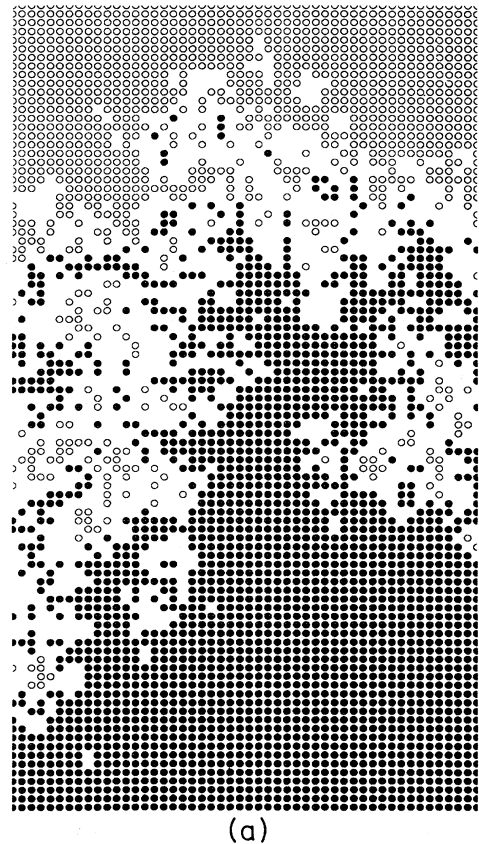


FIG. 1. (a) A typical configuration after approximately 6×10^3 MSC for $s=1.0$. Note the predominance of large clusters and overhangs in the interfacial region. (b) A typical configuration after approximately 6×10^4 MCS for $s=0.5$. The vacant sites are restricted to a relatively narrow zone between the A and B domains and, in contrast to (a), there is an absence of large clusters within the zone of vacant sites.

$$\langle x^n \rangle_m = \frac{\sum_{x=-\infty}^{x=+\infty} x^n [m(x+1) - m(x)]}{\sum_{x=-\infty}^{x=+\infty} [m(x+1) - m(x)]}. \quad (1)$$

Thus w_m is the root-mean-square width of the reactive zone, i.e., the region of the lattice where reaction events may possibly occur. For the case in which s is equal to unity, w_m is a suitable measure for the reactive-zone width. However, when s is not equal to unity, the width of the reactive zone, consisting of the domain of vacant sites between the A and B domains, reaches a constant value rather early in the simulations. As will be seen below, the quantity w_m in that case measures the width of meandering of the reactive zone.

Two measures which describe the roughness of the interfacial region between the two reacting species can be defined. The first measure of roughness is obtained by considering the distribution of sites forming the external perimeters of each of the A and B domains. Note that each of these domains has an external perimeter so that there are two interfaces which are statistically equivalent by symmetry. Using the distribution of external perimeter sites, we can obtain the average

$$\langle x^n \rangle_h = \sum_h x^n / N_h, \quad (2)$$

where N_h is the number of sites which form the external perimeter and the sum is over these sites. The roughness measure w_h is then defined as $w_h = (\langle x^2 \rangle_h - \langle x \rangle_h^2)^{1/2}$. This measure of roughness is similar to one that has been previously used to characterize the diffusion frontier or interface [18].

The second measure of roughness that we use is the same as the definition usually used in studies of film growth in models where there are holes and overhangs in the epitaxial layer [9,10]. Consider the external perimeter of the A domain. For each row y of lattice sites, there can be more than one lattice site belonging to the external perimeter. We define the interface position x_A for each row of sites to be given by the minimum value of x among all the external perimeter sites in that row. Then the roughness w_A of the interface between the reactive zone and the A domain is given by

$$w_A = (\langle x_A^2 \rangle - \langle x_A \rangle^2)^{1/2}, \quad (3)$$

where the averages are taken over all the rows of the lattice. A similar definition of w_B holds for the interface of the B domain, except that for the B domain, the interface position x_B for each row of sites is given by the maximum, rather than the minimum, value of x for the external perimeter sites in that row, i.e., the quantity $(x_B - x_A)$ for each row of sites is a measure of the width of the reactive zone for that row. In analogy with film growth models, the parameter s introduced earlier can be used to modify the roughness of the interface: the smaller the value of s , the smoother the interface. This is clearly illustrated in Figs. 1(a) and 1(b). Since the domains of A and B are equivalent we take the average of

w_A and w_B as a measure of the roughness w_r of the surface of these domains.

III. RESULTS AND DISCUSSION

As the reaction proceeds, the interface becomes progressively rougher, and there is no time-independent characteristic length scale in the problem. Thus we expect the interface roughness w to scale as $w \sim t^\beta$ [8-10]. We performed simulations for values of s ranging from 0.5 to 1.0 using lattices of heights ranging between 100 and 10 000. The data shown in Figs. 2(a) and 2(b) are the results of simulations using lattices of height 10 000 lattice constants for $s=1.0$ and 0.5, respectively. For each value of s , 100 simulation runs was performed and averaged.

For $s=1.0$, the exponent β is equal to 1/2 for both w_h and w_r . Although both w_h and w_r scale in the same manner with time, w_h is always larger than w_r . The width of the reactive zone, as measured by w_m , also in-

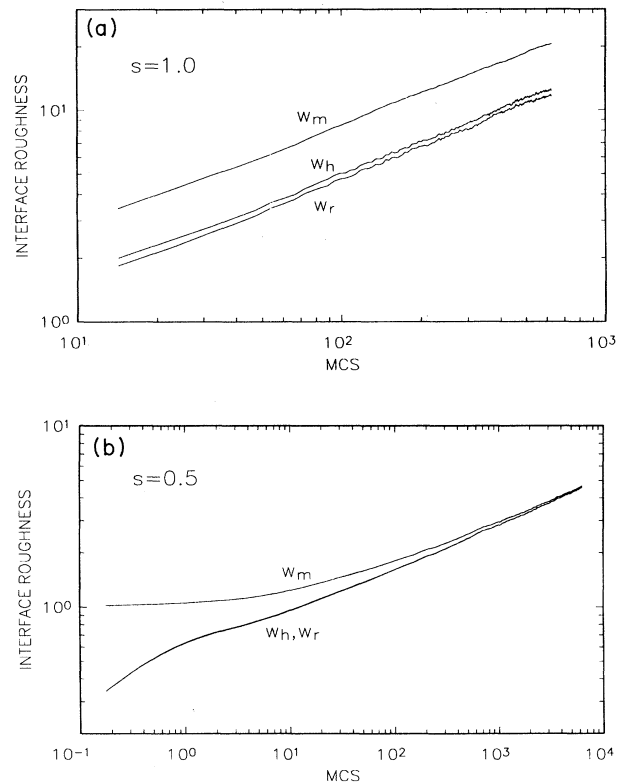


FIG. 2. (a) Time dependence of w_h , w_r , and w_m for the case in which $s=1.0$. (b) Time dependence of w_h , w_r , and w_m for the case in which $s=0.5$.

creases with time as $t^{1/2}$, and is always larger than both the roughness measures w_h and w_r . For $s=0.5$ the exponent β is found to be $1/4$ for both w_h and w_r . In contrast to the case in which $s=1.0$, w_h and w_r are equal to each other when $s=0.5$. Initially, the width w_m of the reactive zone is larger than w_h and w_r . However, for short times w_m apparently does not obey a power law and grows slower than w_h and w_r . Therefore w_h and w_r approach w_m with time. Consequently, in the long-time limit, w_m becomes equal to the roughness of the interface defined by the other two metrics. For our simulations with $s=0.5$, this occurs after approximately 10^4 MCS. Subsequently, both the roughness of the interface and the width of the reactive-zone scale with time as $t^{1/4}$. We have also performed simulations for values of s between 1.0 and 0.5. These simulations all show results similar to the case in which $s=0.5$.

For the case in which $s=1.0$, the fluctuations in particle density resulting from the formation of clusters in the reactive zone play a significant role in interfacial roughening. This situation is illustrated in Fig. 1(a). For $s=0.5$, a typical interfacial configuration of which is seen in Fig. 1(b), such fluctuations are insignificant. We have mentioned that w_m is a measure of the width of the reactive zone and, hence, the region over which clusters can be expected. Since w_m increases indefinitely with time, one might suspect that fluctuations resulting from cluster formation are more important than we have implied for $s=0.5$. For the case in which $s=1.0$, w_m is a good measure of the width of the reactive zone. For $s=0.5$, however, w_m is not a good measure of this width. This is because, as may be seen in Fig. 1(b), the region over which reaction can occur consists of only a narrow zone of vacant sites between the A and B domains. Fluctuations in particle density within each of the A and B domains are insignificant, in contrast to the case for which s is equal to 1.0. Thus, for $s=0.5$, w_m is really a measure of the width of meandering of the reactive zone.

The width of the reactive zone for $s=0.5$ is better measured by the quantity $\langle x_B - x_A \rangle$, with x_B and x_A defined in Sec. II. From the simulations we find that this quantity diverges with time for $s=1.0$ (with an exponent of approximately 0.48), but for s less than unity, it converges to an s -dependent value. A plot of some simulation results is shown in Fig. 3. Hence we conclude that for s less than unity the length scale of the clusters in the reactive zone becomes infinitesimally small compared to the length scale of the interfacial roughness.

We now consider the results for $s=1.0$. It can be seen that the width w_m of the reactive zone behaves diffusively. Since this width is obtained from the average spin $m(x) = \langle \sigma(x) \rangle$ of the sites in each column x , it is, thus, interesting to compare the interfacial roughness in our reaction system with that in the diffusion system $\partial \sigma / \partial t = \nabla^2 \sigma$, where the boundary conditions are $\sigma(-\infty, y) = -1$ and $\sigma(\infty, y) = 1$, and the initial condition is a step function at $x=0$. For this diffusion system, the width of the average spin $m(x) = \langle \sigma(x) \rangle$ profile also satisfies $w_m \sim t^{1/2}$. The scaling properties of the diffusion hull have been studied previously by Sapoval, Rosso, and

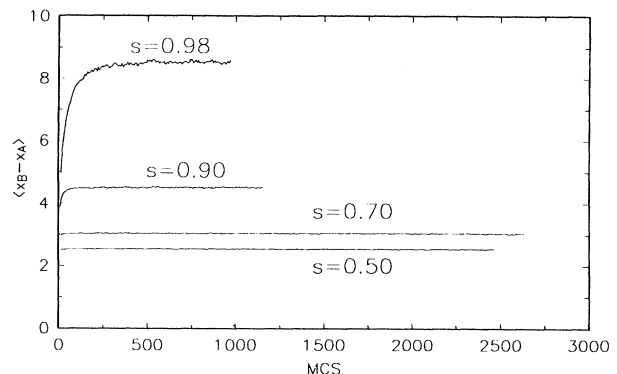


FIG. 3. Time dependence of the reactive-zone width measured by $\langle x_B - x_A \rangle$ for s less than unity.

Gouyet [18]. It can be deduced from their results that the roughness measure w_h obey the scaling $w_h \sim t^\beta$, where β is approximately 0.286 implying that, in the long-time limit, the diffusion hull is smooth on the length scale of w_m which scales as $t^{1/2}$. In the case of the reaction system studied here, however, we find that w_h also scales as $t^{1/2}$. Therefore the roughness of the external perimeter in this model grows as fast as the width of the reactive zone, i.e., the “fingers” of the A and B domains have the same length scale as the width of the reactive zone. This difference between the diffusion system and the reaction system arises from particle-particle correlations that exist in the latter as a result of the removal of nearest-neighbor AB pairs. In the diffusion system, the distribution of particles is nonuniform but random. In the reaction system it is nonuniform and nonrandom, and one is more likely to find an AA or a BB nearest-neighbor pair in the reaction system than in the diffusion system.

We can also gauge the importance of cluster formation at the interface by comparing w_h with w_r . If overhangs in the interface are important, then w_h will be different from w_r . This, of course, is the situation for $s=1.0$. For $s=0.5$, however, w_h and w_r are the same [cf. Fig. 2(b)]. Overhangs are thus not significant for $s=0.5$. Since the formation of clusters is related to the presence of overhangs (more overhangs implying easier cluster formation), we conclude that for $s=0.5$ fluctuations due to cluster formation are not important.

We have defined an A domain and a B domain in our reaction system. Between these domains is a domain of vacant sites which shares an interface with each of the A and B domains. Thinking in terms of film growth, we can consider the evolution of the reaction system to be the growth of this “film” of vacant sites. There are “overhangs” and “holes” in the domain of vacant sites just as in, for instance, ballistic deposition models [9,10] of film growth. However, there are two differences between the growth of the domain of vacant sites studied here and the growth of various deposition models.

First, in the reaction model, growth can occur at any point along the internal or external perimeter of the domain, whereas, in film-growth models, deposition of

particles, and hence film growth, occurs only at exposed points of the external perimeter. Second, in the reaction models, as mentioned earlier, there are clusters of vacant sites embedded in the A and B domains, and, conversely, there are also clusters of A and B particles embedded in the domain of vacant sites. Upon adsorption of a single A or B particle, possibly followed by subsequent reaction and desorption of an AB pair, the connectivity of these clusters to each of the domains may be changed. For example, a cluster of vacant sites initially completely embedded in the A domain and separated from the domain of vacant sites by only a single A particle will become connected to this latter domain if that A particle reacts and is removed from the lattice. Thus, in contrast to film growth models where deposition and thermal desorption involves only one particle at a time, in the reaction model, the film of vacant sites can "adsorb" or "desorb" more than one vacant site at a time. This type of fluctuation in the interface positions has been observed in diffusion fronts where it has been referred to as intercalation noise [18,19].

Despite these differences it is, nevertheless, interesting to discuss the roughening of the film of vacant sites in our reaction system by considering the general stochastic differential equation [8–10,20–22] which describes film growth,

$$\frac{\partial h}{\partial t} = R(\nabla h, \nabla^2 h, \dots) - \nabla j_{\text{eq}} + \eta. \quad (4)$$

At least in the case of $s=0.5$, the simulation results can be rationalized using this equation. Here, R is the contribution associated with the dynamics of the addition or removal of particles from the film, j_{eq} is the flux due to thermal diffusion, and η is the δ -function correlated noise. The Edwards-Wilkinson [22] and the Kardar-Parisi-Zhang (KPZ) [8] equations are two special cases of this general form. For the reaction system that we study here, no thermal diffusion occurs and j_{eq} is zero.

We argue that this equation should be sufficiently general to describe the reaction interface when the interface structure does not include overhangs and holes. The rates of adsorption on and desorption from the zone of vacant sites at any point can then be described by the term R , which depends on the local nature of the reaction interface. This is adequate as long as the adsorption and desorption are local events and do not involve, for instance, the desorption of a cluster of A particles as a result of the reaction of a single A particle which connects this cluster to the A domain. Such an event cannot depend on just the local nature of the interface because the connectivity of the particles in the cluster must be considered. When s is not equal to unity, overhangs and holes are not significant since $w_r = w_h$. This implies that the contribution of cluster (nonlocal) desorption or adsorption is not significant. Thus the time evolution of the zone of vacant sites occurs through the addition or removal of single sites, and, therefore, it is plausible that Eq. (4) with R depending only on the local nature of the inter-

face and with η δ -function correlated in space and time can describe the interfacial evolution for s not equal to unity.

We now consider the form of the term $R(\nabla h, \nabla^2 h, \dots)$. In our model we have included a repulsive interaction between an A (B) particle attempting to adsorb in a vacant site and the nearest-neighbor adsorbed B (A) particles. A consequence of this interaction is a higher rate of removal of vacant sites from the "hills" than from the "valleys" in the film of vacant sites. As a result of this, there is a term of the form $\nu \nabla^2 h$ in R , where ν may be interpreted as a surface tension [8–10,20–22].

In our reaction model, it is also clear that the growth velocity of the film is dependent on the local slope of the film surface. The density of sites where growth can occur is higher if the film surface is along one of the principal axes of the lattice than if it is along a diagonal. Consequently, there is lateral coupling in the interface evolution, and we expect nonlinear terms to be present in R . To lowest order, we can, therefore, expect a term of the form $\lambda |\nabla h|^2$ to occur in R , where λ gives the strength of this lateral coupling [8]. In order to test this idea, simulations were performed with a diagonally oriented interface between the A and B domains. The average width $\langle x_A - x_B \rangle$ of the zone of vacant sites for $s=0.5$ that was obtained is the same as that obtained in the simulations discussed above in which the interface between A and B is oriented along one of the axes of the square lattice. In both cases $\langle x_A - x_B \rangle$ is approximately 2.55 ± 0.05 lattice constants. Since $\langle x_A - x_B \rangle$ is determined by a balance between reactive adsorption and nonreactive adsorption of A and B particles onto the square lattice and, hence, depends on the structure of the interface, we conclude that the structure of the interface is not dependent upon its average orientation. Therefore $R(\nabla h, \nabla^2 h, \dots)$ must not contain any term dependent on the slope of the interface, and the equation for the time evolution of the interface is of the KPZ form $\partial h / \partial t = \lambda |\nabla h|^2 + \nu \nabla^2 h + \eta$, with $\nu \neq 0$ and $\lambda = 0$ (Edwards-Wilkinson equation). For this case the exponent β is equal to $\frac{1}{4}$. This is supported by the simulation results for w_h and w_r for $s=0.5$ (and for other values of s between 1.0 and 0.5).

When $s=1.0$, we have seen that the roughness increases as $t^{1/2}$, and not as $t^{1/4}$. Intercalation noise becomes important in this case, and the equation describing the interfacial evolution must be different from that for the case in which s is not unity. It was pointed out above that in the reaction model there are clusters embedded in each of the A , B , and vacancy domains, and these clusters can participate in the growth of the vacancy domain. For the case in which $s=1.0$, the contribution of this process to the growth of the vacancy domain dominates the contribution from the addition or removal of a single vacant site. Thus it is reasonable that the interfacial evolution equation is reduced to $\partial h / \partial t = \eta$, and we then expect an exponent β equal to $\frac{1}{2}$. However, we should point out that the noise in the reaction system may not be δ -function correlated in space and time, in which case the equation $\partial h / \partial t = \eta$ would not necessarily yield an exponent of $\beta = \frac{1}{2}$.

IV. CONCLUSIONS

We have studied the interface between adjacent domains of reactants in a bimolecular Langmuir-Hinshelwood reaction. It was found that the scaling exponent of the interface roughness depends upon the interaction between the reacting species. For the case $s < 1$, which implies a repulsive interaction between the two species, the interfacial roughness scales as $t^{1/4}$. We argue that for s less than unity the KPZ equation with $\nu \neq 0$ and $\lambda = 0$ is a valid description of the interfacial roughening, and thus a scaling of $t^{1/4}$ is to be expected. Our argument depends upon the width of the reactive zone becoming infinitesimally small compared to the roughness of the reaction interface in the long-time limit. The simulation results support this for the case when s is less than unity.

For $s = 1$, the reactive-zone width increases indefinitely with time. The reactive zone itself becomes occupied by clusters which grow indefinitely. Therefore, for the case in which s is equal to unity, the idea of a reactive zone restricted to a narrow region between two adjacent domains is not valid. Since the clusters participate in the roughening of the reactive interface, we conclude that the "noise" term in the KPZ equation dominates the terms in the interface evolution equation which describes the addition or removal of single sites to the zone of vacant sites.

Then, it is plausible that $\partial h / \partial t = \eta$, which implies the exponent $\beta = \frac{1}{2}$, describes the interface evolution.

It should be noted that our results are consistent with simulation results for the roughening of the interface between the spin-up and spin-down domains of an Ising model [7]. There, the interface roughness is simply taken to be the quantity w_m that we have defined here. For temperatures below the critical temperature, w_m increases as $t^{1/4}$. At the critical temperature, it was found that w_m increases as $t^{1/2}$ [7]. The arguments that we have used to justify describing the reaction interface with the KPZ equation can similarly be used to understand the Ising model simulations [7]: below the critical temperature the length scale of spin clusters is not so large as the roughness of the interface, and the time evolution of the latter may be described by the KPZ equation with $\nu \neq 0$ and $\lambda = 0$. At the critical temperature fluctuations become important, and the growth of the interfacial region between the spin-up and the spin-down domains becomes dominated by the addition or removal of clusters rather than by the addition or removal of single spins.

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- [1] R. Pandit, M. Schick, and M. Wortis, *Phys. Rev. B* **26**, 5112 (1982).
 - [2] R. Lipowsky, *J. Phys. A* **18**, L585 (1985).
 - [3] M. Grant, K. Kaski, and K. Kankaala, *J. Phys. A* **20**, L571 (1987).
 - [4] K. Binder, in *Kinetics of Ordering and Growth at Surfaces*, edited by M. G. Lagally (Plenum, New York, 1990), p. 31.
 - [5] *Kinetics of Aggregation and Gelation*, edited by F. Family and D. P. Landau (North-Holland, Amsterdam, 1984).
 - [6] K. K. Mon, K. Binder, and D. P. Landau, *Phys. Rev. B* **35**, 3683 (1987).
 - [7] D. Stauffer and D. P. Landau, *Phys. Rev. B* **39**, 9650 (1989).
 - [8] M. Kardar, G. Parisi, and Y. C. Zhang, *Phys. Rev. Lett.* **56**, 889 (1986).
 - [9] F. Family, *Physica A* **168**, 561 (1990).
 - [10] J. Krug and H. Spohn, in *Solids Far From Equilibrium: Growth, Morphology, and Defects*, edited by C. Godreche (Cambridge University Press, Cambridge, 1991).
 - [11] E. Wicke, P. Kumman, W. Keil, and J. Scheifler, *Ber. Bunsenges. Phys. Chem.* **4**, 315 (1980).
 - [12] R. M. Ziff and K. A. Fichtorn, *Phys. Rev. B* **34**, 2038 (1986).
 - [13] P. Meakin and D. J. Scalapino, *J. Chem. Phys.* **87**, 731 (1987).
 - [14] L. Galfi and Z. Racz, *Phys. Rev. A* **38**, 3151 (1988).
 - [15] Z. Jiang and C. Ebner, *Phys. Rev. A* **42**, 7483 (1990).
 - [16] H. Taitelbaum, S. Havlin, J. E. Kiefer, B. Trus, and G. H. Weiss, *J. Stat. Phys.* **65**, 873 (1991).
 - [17] For $s = 1$, the model is similar to the model investigated in Ref. [12]. In their model the adsorption rate is infinitely high and thus no vacancies exist on the lattice.
 - [18] B. Sapoval, M. Rosso, and J. F. Gouyet, *J. Phys. (Paris) Lett.* **46**, L149 (1985).
 - [19] B. Sapoval, *Philos. Mag.* **B 59**, 75 (1989).
 - [20] D. E. Wolf and J. Villain, *Europhys. Lett.* **13**, 389 (1990).
 - [21] S. Das Sarma and P. Tamborenea, *Phys. Rev. Lett.* **66**, 325 (1991).
 - [22] S. F. Edwards and D. R. Wilkinson, *Proc. R. Soc. London Ser. A* **381**, 17 (1982).