

Domain growth in the interacting adsorbate: Nonsymmetric particle jump model

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We show that an exponent of a powerlike time domain growth is determined not only by the conservation or nonconservation of the order parameter, but also by the asymmetry of single-particle jumps. Domains that have an anisotropic pattern, such as (2×1) , have a tendency to grow faster in a certain direction than they do in others. The rate of expansion in different directions depends on the barriers for single-particle jumps. As a result, dynamical behavior of systems which start in the same configurations and eventually reach the same equilibrium states is completely different. We show how differences in microscopic dynamics in a one-dimensional Potts model lead to different rates of domain growth. We observe a similar effect for a two-dimensional (2×1) ordering by changing the way in which a barrier for a jump depends on the number of neighboring particles. We show examples of the domain power growth, which are characterized by different exponents.

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I. INTRODUCTION

An assembly of interacting adsorbed adatoms on a crystalline solid surface undergoes a variety of phase transformations, the nature of which depends on the type of adatom-adatom interactions and the symmetry of the lattice of adsorption sites.¹⁻³ Many of these transformations result in complex adatom density patterns associated with different regions of the phase diagram for the system.¹ When a gas of adatoms is rapidly quenched below the line of the first-order phase transition, we observe an initial formation of small domains—droplets of the emerging new phase—already exhibiting the corresponding geometrical pattern. The growth of these domains is one of the most intensively studied problems in surface physics because it exhibits universal character that is characterized by only a few system parameters. Among them a crucial role is played by the relevant order parameter for the underlying phase transformation. The domain growth exhibits $t^{1/2}$ dependence for a nonconserved order parameter,⁴⁻⁸ and a $t^{1/3}$ dependence, called the Lifshitz-Slosov growth,⁷⁻¹¹ for a conserved one. Experimentally, it is found that slightly different systems either follow a $t^{1/2}$ growth law¹² or an almost diffusion-driven $t^{1/3}$ growth.^{9,13-16} On the other hand, for an open system in which particles can freely desorb and adsorb, the density is clearly nonconserved and the exponent is always $1/2$.⁷

It has been argued that the degeneracy of the order parameter, in a sense attributed to the Potts model,¹⁷ has no influence on the growth time dependence.⁵ For this reason, analy-

sis of the domain growth is usually made using a proper version of the Ising model with Glauber dynamics for a nonconserved order parameter or Kawasaki dynamics for a conserved one.^{8,9} For various complex experimental situations, like O/W(110),^{18,19} when domains exhibit (2×1) ordering structures, the usual “single site” density is not an appropriate order parameter and it is not clear if the inherent order parameter is conserved or not. In spite of an obvious importance of the above-mentioned issues for applications ranging from surface physics to metallurgy, theoretical attempts to provide an answer to the dynamical exponent puzzle are not numerous. Attempts to resolve this issue by the computer Monte Carlo simulation turned out to be inconclusive, the reason of that was attributed to finite sample size effects, to long lifetime of transients in ordered system, or to other computational difficulties. An important step toward solving the problem was made by Weinberg and co-workers,²⁰⁻²² who used a new version of the kinetic lattice gas model to describe migration of adatoms on a two-dimensional crystalline lattice. In their model, the migration proceeded by thermally activated adatom jumps via a series of states, which were also thermally activated. That model resulted in a different rate of transition from one adatom equilibrium location to another from the conventional value.^{20,21} Thermally activated states have been identified with the physisorbed adatom states of relatively higher energies. The existence of the physisorbed states was essential because it allowed for long adatom jumps resulting in a complete change of the system kinetics. Particle dynamics via such states results in

different than usual rates of the domain growth. In most cases, however, if a physisorbed state truly exists, then a transition to it leads to desorption rather than to surface migration. One obtains a similar domain growth effect with an unphysical choice of locally scaled jump rate, which results in a change of time scale for each particle jump individually.²² No explanation was provided as to why we observe differences in domain growth rates in cases where thermally activated particles jump between neighboring sites, with jump rates depending on the energy barriers only.

In order to resolve the domain growth puzzle, we suggest in the present paper a different approach to the domain growth analysis based on the modification and generalization of our studies on surface diffusion.^{23–26} In these publications, we have shown that in the case of a closed system (conserved density), the diffusion coefficient has a general dependence on microscopic details of the adatoms dynamics, for example, on modifications to the single-particle jump barriers due to interactions between adatoms. It is precisely that which makes our kinetic lattice gas model essentially different from that introduced in Refs. 20 and 21. In our model there is no place for thermally activated states, instead the intersite energy barrier heights are cooperatively modified by the adatoms' mutual interactions.^{23,24} Our approach was motivated by *ab initio* calculations for the O/W(110) cluster.^{24,25} It has also been shown that the presence of In adlayer lowers the energy barrier by 0.7–0.12 eV for diffusion of nitrogen on GaN (0001) surface.²⁷ This result indicates that the interaction between adatoms may change the potential-energy landscape drastically, changing the behavior of the adsorbed species qualitatively from virtually immobile to effectively diffusing ones.

The role of interparticle interactions for a particle at the top of a barrier should be decided experimentally. Their presence was crucial in understanding diffusion in the CO/Ru(001) system²⁸ and for rapid rearrangements in the devil staircase Pb/Si structure.²⁹ In what follows, we will show that basically the same idea of the barrier modification due to adatom interactions is sufficient to account for the rich variety of scenarios for the domain growth dynamics without the necessity of any further assumptions.

Our next goal is to explain the (2×1) domain growth as seen, for example, in the O/W(110) system using the dynamic Potts model. A simple lattice gas can be mapped onto the Ising model in a natural way by identifying the presence of a particle with one Ising state and its absence with the other. On the other hand, for ordered states of lattice gases, such as the (2×1) structure, we have four different patterns of particles which can be mapped onto four different Potts states. Here, one Potts spin represents a basic (4×4) cell of the ordered system. This basic cell includes 16 lattice sites, half of them occupied in the ideal ordering. If we assume that each cell is ordered, we can map them onto the four-state Potts¹⁷ model on a two-dimensional lattice. It has been shown⁵ that the number of Potts states has no bearing on the rate of domain growth. In Sec. II we present a four-state Potts model in which the mean domain size increases as a power function of time with different exponents. Such dynamics can be realized by a Potts model with four and more

states, when some domain walls are immobile. Section III presents different scenarios of (2×1) domain growth.

II. POTTS MODEL

First, consider a one-dimensional Potts model with $p=4$ states labeled $A, B, C,$ and D . We shall study the dynamics and growth of domains envisaged as a row of spins found in the same state, i.e., $AAAA\dots$, or $BBB\dots$. Let us start with the case when domain walls between all Potts states have the same dynamics and the order parameter (number of particles in given state, i.e., A particles) is not conserved. Each particle can change its state at any time, but the probability of a given change depends on the state of its nearest neighbors. Let us define them as follows:

$$AABB \rightarrow ABBB \quad \text{or} \quad AABB \rightarrow AAAB, \quad (1)$$

with the transition probability $q=1/2$ for each. The same rule applies to other permutations of the four Potts states. Jumps of type (1) describe the motion of domain walls between domains containing at least two atoms. Single-atom domains disappear, however, following the rules:

$$ABA \rightarrow AAA \quad \text{with probability } v = 1,$$

$$ACB \rightarrow AAB \quad \text{with probability } q = 0.5,$$

$$\text{or } ACB \rightarrow ABB \quad \text{with probability } q = 0.5, \quad (2)$$

and the same for similar domains formed by other Potts states. All other jumps are assigned zero probability. According to rules (1) and (2), domain walls diffuse freely until they reach another wall. The order parameter, understood as the total number of cells in a given Potts state, is not conserved in such a process because with each jump one state changes into another one. Diffusive motion of the domain walls determines the character of the domain growth. In a one-dimensional system, the number of the domain walls is equal to the number of domains. For a four-state Potts model, there are two possible ways of changing the number of domains and/or domain walls. A single-cell domain disappears when (i) two domain walls are replaced by one—this is the case when two domains next to the one which disappears are of a different type [cf. second and third lines in Eq. (2)]—or (ii) no new wall is created and the two domains neighboring the one which disappears are of the same type [cf. first line of Eq. (2)]. The latter possibility is the only one possible for a two-state Ising model. The rate of both of these allowed processes is related to the wall diffusion coefficient. The rate with which domain walls disappear is equal to the domain wall diffusion constant D divided by the mean-square distance between the walls $\langle d^2 \rangle \sim (L/N)^2$. Here L is the size of the system and N is the number of domains. The total rate of decrease of the number of domains is proportional also to their number N , thus

$$\frac{dN}{dt} \sim ND \left(\frac{N}{L} \right)^2. \quad (3)$$

As a result $N \sim t^{-1/2}$, which means that the average domain size grows as $\langle d \rangle \sim t^{1/2}$. Growth of the domain is thus univer-

sal and similar to that in other growth processes where the domain-wall diffusion is the main growth mechanism. The internal structure of the p Potts states plays no role in this type of domain growth.

For practically identical physical systems, however, the growth law becomes different when the rules for the microscopic atomic jumps are different. To see this, consider the same Potts model but now with the Kawasaki dynamics⁵—appropriate for a conserved order parameter. In that case of dynamics, the atomic jump results in the exchange of two neighboring spins

$$AAB \rightarrow ABA \quad \text{with probability } v = 0.1,$$

$$ACB \rightarrow ABC \quad \text{with probability } q = 0.5,$$

$$\text{or } ACB \rightarrow CAB \quad \text{with probability } q = 0.5. \quad (4)$$

The rule now is that neighboring spins exchange their states, but the total number of spins in the same state is preserved. The probability of this exchange is smaller when the other neighbor of the changing spin is in the same state. The rate of decrease of N , different from Eq. (3), is

$$\frac{dN}{dt} \sim -NDa \left(\frac{N}{L} \right)^3. \quad (5)$$

This rate is again proportional to the number of domains N , to the rate at which particles travel a distance from one domain to the other $\sim DN^2/L^2$, and to the inverse of a mean number of particles in one domain $L/(aN)$, where a is the distance between underlying lattice sites. This last factor reflects the fact that in order to annihilate a domain wall it is not sufficient that just one atom crosses from one domain to the other. Instead, for two domains to merge in this type of dynamics, it is necessary that all atoms which constitute one of the domains move to the other domain of the same type. For this reason, the rate of wall number decay is much lower than for the Glauber dynamics, so it has to be divided by the mean number of particles in the domain. Such a simple picture is sufficient to account for the difference in the domain growth laws for nonconserved (3) and conserved (5) order parameters. For the latter, we get $N \sim t^{-1/3}$.

When domains are built up between locally ordered particle assemblies, like (2×1) , which is the situation characteristic in systems where repulsive interactions are prevalent, the first type of domain dynamics with nonconserved order parameter seems to be more appropriate. At the same time, we observe in this case a wide variety of different exponents characterizing the domain growth. We will now show how that variety of exponents different from $1/2$ follows from our model when applied to the systems with a nonconserved order parameter and more than two.

Let us assume that $p=4$, and let us define the microscopic dynamics in such a way that all following walls $A|B$, $B|C$, $C|D$, and $D|A$ move by diffusion jumps with a probability $q_1=1/2$, as before, but that the walls $A|C$ and $B|D$ do not move at all, i.e., $q_2=0$. Two walls of the first type, say, $A|B$ and $B|C$, create one immobile wall $A|C$ when they meet each other. This wall, once created, will not be removed until

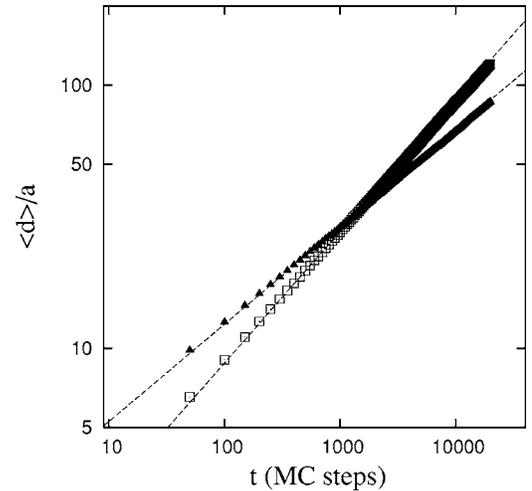


FIG. 1. Time dependence of the mean domain width, in units of lattice constant a , for one-dimensional systems of Potts spins that evolve according to the classical schema (squares)—all walls move, or the modified one (triangles)—with two types of walls blocked. Slopes of the plotted lines are 0.5 for the classical dynamics and 0.37 for the modified dynamics. Simulations were made for 3000 spins and 20 000 MC steps. Periodic boundary conditions were used. Results were averaged over 100 samples.

another mobile wall, $D|A$ or $C|D$, diffuses to its position. Therefore, the global domain dynamics consists of diffusion of mobile walls, leading to the creation of immobile walls which wait for another mobile wall to arrive to convert it into a mobile wall again. As a result, according to the Monte Carlo (MC) simulations, the domain size grows like $t^{0.37}$ (see Fig. 1). The dynamics slower than $t^{1/2}$ is, in this case, caused by correlations between mobile and immobile walls. An interesting fact is that even if the domain size grows with a coefficient smaller than $1/2$, the number of *mobile* walls still decays like $t^{-1/2}$. It is only the *total* number of the domain walls that decays like $t^{-0.37}$. A new immobile wall is created in a place where all other walls have already been removed, so the immobile and mobile walls are strongly anticorrelated.

Such anticorrelation is possible when the Potts spins have at least four states and dynamics is defined in such a way that the immobile walls are created and destroyed all the time during growth process. Indeed, in the example above we made immobile two types of walls out of the total of six, and there is always a natural path to creating or destroying any of them. In a three-state Potts model, there are only three types of domain walls and if one of them is immobile, then it can be easily destroyed, but there is no natural way of creating a new one. This explains the need for at least four Potts states.

III. SURFACE 2×1 DOMAINS

The Potts model example above shows that when we have several different types of domains with the walls between them moving at different rates, the global domain size growth can eventually be characterized by exponents other than $1/2$ even if the order parameter is not conserved. Its value depends on the details of the microscopic dynamics.

Let us now discuss a two-dimensional system of particles which interact by attractive nearest-neighbor interactions, $J_1/(k_B T) = -1$, and repulsive next-nearest-neighbor interactions, $J_2/(k_B T) = 1$.²⁵ Such system orders forming the well-known (2×1) structure.^{18,19} There are four different domain types within this structure. Microscopic dynamics of particles depends, in general, not only on the energies of their initial equilibrium states but also on the height of potential barriers between the initial and final states. In the standard version of the dynamics, all barriers are chosen to be of the same height and the particles move much faster along than across the ordered lines of the (2×1) domain structure.²⁵ However, even in this case, the movement of walls between the ordered structures is more complicated than assumed in standard models, so that more detailed analysis is needed to understand the details of the wall motion.

We can change the relative speed of motion of different walls by modifying the rates of individual particle jumps. The individual particle jump rate from state i to state j has to fulfill a detailed balance condition so that the formula for the rate can be written as

$$W(i \rightarrow j) = W_0 e^{\beta E_i} e^{-\beta E_{ij}^B}, \quad (6)$$

where $\beta = 1/k_B T$ and W_0 , providing an intrinsic time scale, is the jump rate of an isolated particle. The factor $e^{\beta E_i}$ is standard, with

$$E_i = J_1 \sum_{nn} n_k + J_2 \sum_{nnn} n_k \quad (7)$$

equal to the energy of the particle at site i . Here nn and nnn denote, respectively, summations over the nearest and the next-nearest-neighbors k of site i , and n_k is the occupation number (0 or 1) of a neighboring site of this type. The factor $e^{-\beta E_{ij}^B}$ in Eq. (6) depends on the interaction on the barrier. Here, $E_{ij}^B = E_{ji}^B$ is the system energy modification due to interactions when the hopping particle is at a saddle point between the sites i and j . The interparticle interaction dependence of E_{ij}^B results in a possibility of different dynamics in systems with the same equilibrium properties,^{23,24,26} since the equilibrium properties of the system are determined by E_i 's only. For our system, we set

$$E_{ij}^B = J' \sum_{k \langle ij \rangle} n_k, \quad (8)$$

where $k_{\langle ij \rangle}$ represents the sum over all sites neighboring the saddle point between the sites i and j , and J' is the interaction of the hopping particle at the saddle point with its nearest neighbor.

As we have already mentioned, in the most often used models with $J' = 0$, any additional particle moves very fast along the rows, whereas its motion across the rows is much slower. A jump across the row consists of two consecutive jumps: a particle jumps out of an ordered row and creates a hole, which is subsequently filled by the added particle. The

ratio of particle jump rates along and across the row is, for $J' = 0$,

$$r = e^{3\beta J_2}, \quad (9)$$

and increases with decreasing temperature. In Eq. (9), $3J_2$ is the energy difference between the energy of a particle residing momentarily between the rows and that for the particle within the row. For $J' \neq 0$, the saddle point energy difference has to be added in the exponent, resulting in

$$r = e^{\beta(3J_2 - 2J')}. \quad (10)$$

It follows that the dynamics of domain walls depends on the details of the ‘‘single particle’’ microscopic dynamics, with the possibility of obtaining domain growth of different

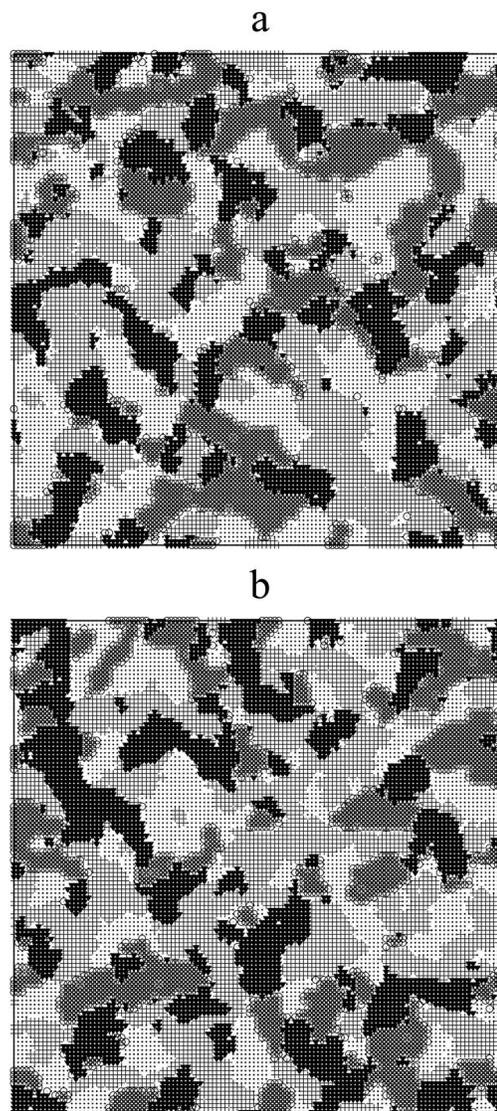


FIG. 2. Examples of the domain pattern resulting from (2×1) ordering. The upper panel shows a system with dynamics I, after 2 000 000 MC steps, and the lower panel is for dynamics II after 1 600 000 MC steps. The mean size of domains in both cases is the same $\langle S \rangle \approx 92$ units. Each unit has 16 lattice sites. The size of the system is 480×480 lattice sites.

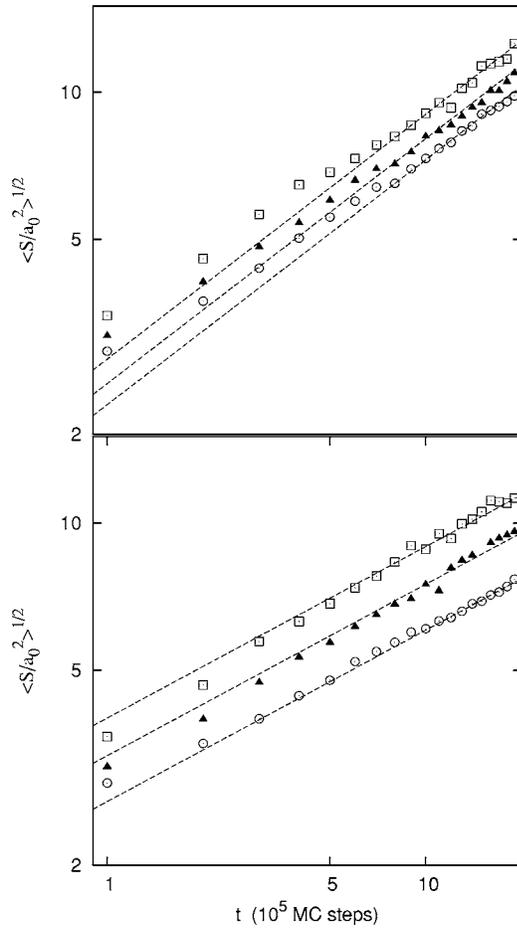


FIG. 3. Time dependence of the mean (2×1) domain width at different temperatures. $\langle S/a_0^2 \rangle^{1/2}$ is the mean domain area expressed as a number of 4×4 basic unit cells. Upper panel shows results for the system that evolves according to dynamics I; lower panel is for dynamics II. Results for $k_B T = 0.45 J_1$ are denoted by squares, for $k_B T = 0.42 J_1$ by triangles, and for $k_B T = 0.4 J_1$ by circles. At each temperature, 20 points are shown, separated by 10 000 MC steps. The slope of lines in the upper panel is 0.5, and in the lower panel it is 0.35.

characters. We have used our standard MC simulation setup to analyze a system with coverage $1/2$. At this coverage, the system orders at $k_B T_c = 0.48 J_1$.²³ We study the domain ordering at several temperatures below T_c for two system sizes: 480×480 and 800×800 . The evolution time was 2 000 000 MC steps. The initial state was a random configuration of particles corresponding to the state of the system at a very high temperature. The system evolves according to MC rules defined by the transition rates (6). We identify domains by comparing 4×4 fragments of the lattice with ordered patterns of four possible domains. Therefore, 16 lattice sites form one basic domain unit. Its length we denote by a_0 . Domain sizes are then measured in such units. We assume the error level 3, meaning that we identify part of the system as a particular domain only when it disagrees with a sample pattern at no more than three points. In Fig. 2 we show how the domains look like when such analysis has been done after some time, for two different versions of dynamics: $J'/(k_B T) = 1$ (dynamics I) and $J'/(k_B T) = -1$ (dynamics II).

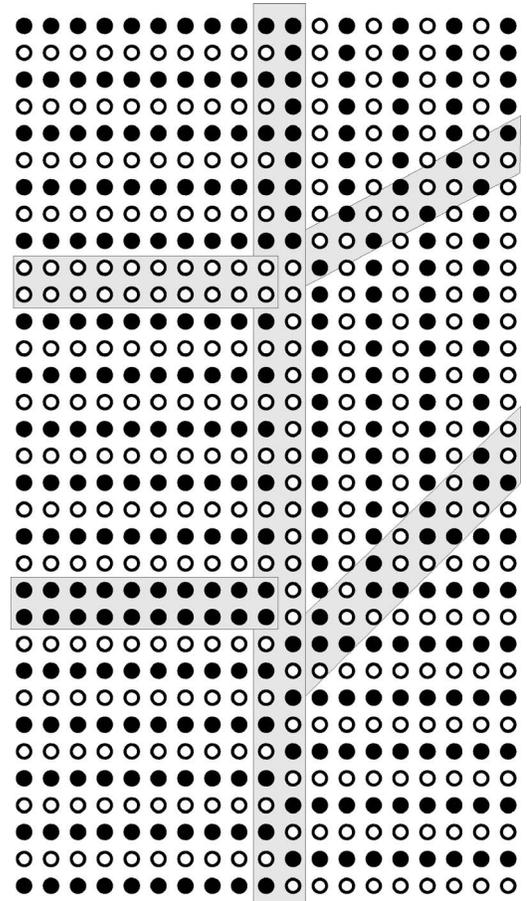


FIG. 4. Examples of domain walls between different (2×1) domains.

According to Eq. (10), these correspond to low and high anisotropy of particle motion, respectively. The two pictures in Fig. 2 have been chosen in such a way that the mean domain size, measured as its mean area, is the same in both cases. No qualitative differences can be seen between the two pictures.

The mean domain size, however, behaves differently as a function of time in both cases. We can see in Fig. 3 that for $J' = 1$ (low anisotropy) we clearly have the $t^{0.5}$ growth law and for $J' = -1$ (high anisotropy) the growth is according to $t^{0.35}$. The domain growth dynamics is different due to different interactions of the particle while being at the saddle point between its initial and final states. The equilibrium properties of both systems are the same. In the system under study, there are four basic types of domains and there are more than six possible domain wall types. Examples of such domain walls are presented in Fig. 4. Some of them move very slowly. In fact, they can be considered as almost immobile, resulting in the slowing down of the net domain growth. The mechanism that leads to different dynamics in cases I and II appears then to be identical to that discussed earlier for the one-dimensional Potts model. The essential feature of this dynamics is the immobilization of certain types of domain walls.

It appears that the domain growth dynamics in systems with more than two states for a nonconserved order param-

eter depends on the details of the microscopic dynamics of particle motion. Such dynamics should result in different relative speeds of domain-wall motion. We have proposed a simple model which can be easily analyzed following the results obtained for a one-dimensional four-state Potts system. This system with a nonconserved order parameter results in the usual $t^{1/2}$ diffusional domain growth for classical microscopic dynamics. However, when the symmetry of states is broken by making two domain walls immobile, we end up with a completely different net growth dynamics governed by the $t^{0.37}$ time dependence. We have shown that similar growth laws are obtained for more realistic (2×1) domains in a two-dimensional system with attractive and repulsive interactions. We have shown that differences in particle interaction, with the rest of the system, when that

particle occupies the top of the barrier, lead to different domain growth dynamics. We hypothesize, therefore, that different power-law time dependencies of the domain growth are observed in many systems^{12–16,18,19} due to similar reasons. The character of the saddle-point interaction was determined for some systems by comparison of theoretical models with experimental data for the diffusion coefficient.^{28,29} The rate of net domain growth is another experimental probe that can help to determine the actual microscopic jump dynamics in a given system.

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