

Anomalous Roughness in Dimer-Type Surface Growth

Jae Dong Noh,^{1,2} Hyunggyu Park,^{1,3} and Marcel den Nijs¹

¹*Department of Physics, University of Washington, Seattle, Washington 98195-1560*

²*Center for Theoretical Physics, Seoul National University, Seoul 151-742, Korea*

³*Department of Physics, Inha University, Incheon 402-751, Korea*

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We point out how geometric features affect the scaling properties of nonequilibrium dynamic processes, by a model for surface growth where particles can deposit and evaporate only in dimer form, but dissociate on the surface. Pinning valleys (hilltops) develop spontaneously and the surface facets for all growth (evaporation) biases. More intriguingly, the scaling properties of the rough one dimensional equilibrium surface are anomalous. Its width, $W \sim L^\alpha$, diverges with system size L as $\alpha = \frac{1}{3}$ instead of the conventional universal value $\alpha = \frac{1}{2}$. This originates from a topological nonlocal evenness constraint on the surface configurations.

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The theory of nonequilibrium dynamic statistical processes has developed rapidly in recent years. Driven systems display intriguing scaling properties and can undergo various types of dynamic phase transitions [1]. Kardar-Parisi-Zhang-type surface growth is an example [2]. There, the properties of the depositing (evaporating) particles are not specified, but are implicitly presumed to be geometric featureless monomers. In surface-catalysis-type processes the geometric shape of the molecules matters. The onset of the catalytic process is associated with a so-called absorbing state dynamic phase transition [3–5]. Monomers give rise to directed percolation and dimers to directed Ising-type transitions. Subtle geometric features are known to be important in equilibrium crystal surface phase transitions as well. The competition between surface roughening and surface reconstruction depends on topological details of the crystal symmetry. Those determine whether a reconstructed rough phase can exist or not [6]. Geometric features are also important in diffusing particle systems. The shape of diffusing particles introduces conservations and leads to anomalous decay of particle density autocorrelations [7]. Therefore, the natural question arises, whether and how the shape of the deposited particles influences the growth and equilibrium properties of crystal surfaces.

Consider a crystal built from atoms of type X . Assume that deposition always takes place in dimer form, X_2 , aligned with the surface. The dimer attaches to two horizontal nearest neighbor surface sites. It loses its dimer character after becoming part of the crystal. Evaporation can take place only in X_2 molecular form, but a different partner is allowed. In this Letter we study the one dimensional (1D) version of this process. This can apply to step shapes during step-flow-type growth on vicinal surfaces. The adsorbed particles do not diffuse in this version of our model. However, topological features that drive our results are preserved, even when monomer diffusion is allowed but limited to terraces. Jumps across steps are unlikely due to

Schwoebel barriers [8]. So our main results do not alter in systems with diffusion.

We describe the 1D surface configurations in terms of integer height variables $h_i = 0, \pm 1, \pm 2, \dots$. They are subject to the so-called restricted solid-on-solid (RSOS) constraint, $h_i - h_{i+1} = 0, \pm 1$, and periodic boundary conditions, $h_{L+i} = h_i$. The dynamic rule is as follows. First, select at random a bond $(i, i + 1)$. If the two sites are not at the same height, no evaporation nor deposition takes place. If the two sites are at the same height, deposition of a dimer covering both sites is attempted with probability p , or evaporation of a dimer with probability $q = 1 - p$. Processes are rejected if they would result in a violation of the RSOS constraint.

The first surprise is that the surface always facets during growth and evaporation, although the surface is rough in equilibrium. The second surprise is that the equilibrium surface width $W \sim L^\alpha$ scales with an anomalous small exponent $\alpha = 0.29 \pm 0.04$. The data could be consistent with an even smaller value due to the strong finite size scaling corrections in Fig. 1(a).

1D surfaces, irrespective of being in equilibrium or in a stationary growing (evaporating) state, display, with only a few very specific exceptions, the universal roughness exponent $\alpha = \frac{1}{2}$. The up-down aspect of the steps becomes uncorrelated beyond a definite correlation length, and therefore the surface roughness obeys random walk statistics at large length scales, which implies $\alpha = \frac{1}{2}$. Our dissociating dimer deposition process circumvents this universal argument by means of a novel type of nonlocal topological constraint. The dimer aspect implies that all surface height levels must be occupied by an even number of particles. However, due to the dissociative nature of the dimers that information is not preserved locally. The “evenness” constraint is nonlocal. At local length scales the surface looks the same as in monomer deposition processes, but the global surface is much less rough. We checked this numerically. Define a window of length b .

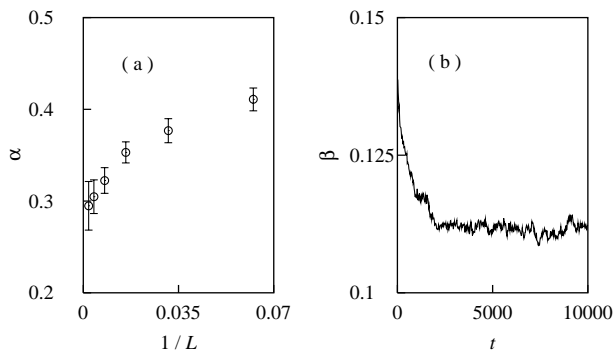


FIG. 1. Finite size scaling estimates of the critical exponents at the equilibrium point $p = q$ for (a) the stationary state surface width $W \sim L^\alpha$ using $L - 2L$ pairs for $L = 2^4, \dots, 2^9$. (b) The temporal surface width $W \sim t^\beta$, using $t/10 - t$ pairs at $L = 2^{13}$.

The surface roughness scales as $W \sim b^\alpha$, with $\alpha = \frac{1}{2}$ for $b \ll L$, but crosses over to the global finite size scaling exponent $\alpha \approx 0.29 \pm 0.04$ for $b \rightarrow L$.

We performed a detailed numerical study of the properties of even-visiting random walks [9] that are globally restricted to visit each site an even number of times. The results, together with analytical scaling arguments, yield the value $\alpha = \frac{1}{3}$. This value lies within the numerical error bars in Fig. 1(a) for the dimer deposition model.

The details of our random walk study are rather technical and will be presented elsewhere [10], but the essence can be captured by the following intuitive scaling argument. Consider the even-visiting random walks for time interval $0 < t < T (= L)$. We assign a defect variable to each site, to mark that it has been visited by the random walker an odd/even number of times up to time t . The even-visiting constraint is satisfied when all defects disappear at time T . Initially, the random walker does not feel the constraint and diffuses freely for $t < \tau_{\text{free}} \ll T$. The defects are uniformly spread over a region of size $\xi \sim \tau_{\text{free}}^{1/2}$. Then it stops spreading and starts to heal the defects. The healing time for a single defect in the region of size ξ is of the order of ξ^2 . By assuming that the healing process for each defect is independent, we estimate the total healing time $\tau_{\text{heal}} \sim \xi^{d+2}$ with spatial dimensionality d . As $\tau_{\text{heal}} \gg \tau_{\text{free}}$, we conclude that $\xi \sim T^{1/(2+d)}$, i.e., $\alpha = 1/3$ for $d = 1$. Existence of a time scale τ_{free} explains the crossover behavior of the surface roughness in the window length b . Similarly, we can argue that the surface width scales with $\alpha = \frac{1}{3}$ in generalized $n \geq 2$ dissociating n -mer-type deposition processes.

The dynamic critical exponent z at the equilibrium point follows from how the surface width diverges as a function of time, $W \sim t^\beta$. We find numerically that $\beta = 0.111 \pm 0.002$; see Fig. 1(b). This suggests the value $z = 3$, since $z = \alpha/\beta$.

The equilibrium surface roughness is unstable with respect to growth and evaporation. It facets immediately.

This phase transition is second order. The correlation lengths that characterize the faceted structure diverge. Before addressing this issue we need to describe and explain the faceted phase. The valleys in the growing surface are sharp and the hilltops rounded; see Fig. 2. This shape is inverted for $p < q$. The faceting is caused by the spontaneous formation of pinning valleys during growth (pinning hilltops during erosion, for $p < q$). Consider, for example, dimers on a flat surface for $p > q$. Odd segments between them act as the nuclei of pinning valleys. Such valleys cannot be filled by direct deposition. The only way to get rid of them is by lateral movement of the subhills.

In finite systems the surface grows in shocks. An initial rough or flat configuration grows fast at first, but pinning valleys appear randomly at all surface heights. The interface develops into a rough faceted structure with many subhills and growth almost stops. From here on the surface advances only when subhills anneal out by the lateral movement of the pinning valleys. The annealing time of a subhill scales exponentially with its size. This exponentially slowing down healing process leads ultimately to a faceted W shape with only two remaining pinning valleys. Their lifetime diverges exponentially with the lattice size. After their demise the surface experiences a growth spurt, and the cycle restarts all over.

The mechanism for lateral movement of pinning valleys is the exchange of active bonds between ramps. Active bonds are locations along the ramp where a dimer can deposit or evaporate. Most steps on the ramps are only one or two atomic units wide and therefore dynamically inactive; see Fig. 2. Active bonds move up or down the ramps by deposition or evaporation of dimers. The growth bias $p > q$ gives them an upward drift. This must lead to an exponential distribution.

Figure 3(a) shows the logarithm of the active bond distribution, $\rho(x)$, versus the horizontal distance x from the center of a hilltop for various values of $p > q$. The straight lines for large x confirm the exponential distribution of active bonds along the ramps, $\rho(x) \sim \exp[-x/\xi_f]$. We determined this from odd lattice sizes, in particular, $L = 257$, where the surface contains an odd number of pinning valleys and therefore reaches a V-shaped stationary state in which it remains pinned at all times.

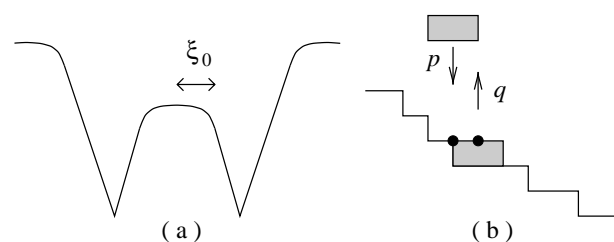


FIG. 2. Hill structures in the faceted growth phase. (a) Schematic structure with two pinning valleys. ξ_0 is the characteristic width of the hilltops. (b) Active bonds (the filled circles) along a local segment of a faceted ramp.

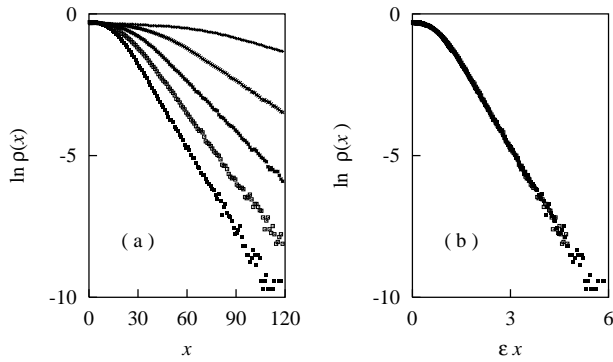


FIG. 3. Active bond distribution $\rho(x)$ at $L = 257$ for $p = 0.505, 0.51, 0.515,$ and 0.52 (from top to bottom). Beyond ξ_0 , $\rho(x)$ decays exponentially. (b) Collapse of the data from (a) according to Eq. (1), with $\beta_\rho = 0$ and $\nu = 1$, demonstrating the hills preserve their shape.

Every now and then an active bond moves in the opposite direction, against the flow, and reaches the valley bottom. That pinning valley moves by two lateral lattice constants when the active bond jumps across onto the other ramp. The probability for this is very small, and scales exponentially with the ramp length, but it is larger from the lower ramp, and therefore the valley bottom moves in the direction of the lower hill, and actually accelerates, because that lower hill keeps shrinking.

Near the rounded hilltops, the surface remains highly active and initially the active bond density does not decrease significantly with x . This defines a second characteristic length scale, ξ_0 , representing the flatness of the rounded hilltops; see Fig. 2. Surprisingly, both lengths, ξ_f and ξ_0 , diverge at $p = q$. Figure 3(b) shows that the curves in Fig. 3(a) collapse according to a single length scaling form

$$\rho(\epsilon, x) = \epsilon^{\beta_\rho} \mathcal{F}(\epsilon^\nu x) \quad (1)$$

with $\nu = 1.0(1)$ and $\beta_\rho = 0.0(1)$. This means that on approach of the $p = q$ critical point the hills maintain their shape in the sense that ξ_0 and ξ_f diverge simultaneously and with the same exponent $\xi_0 \sim \xi_f \sim (p - q)^{-1}$.

It is surprising that both length scales of the faceted phase diverge at the equilibrium point. The structure of the rough phase would be much more complex when one of them remained finite. This actually happens in the following generalization of the model.

Recently, Alon *et al.* [11] added to the conventional monomer-type RSOS model growth the constraint that evaporation from flat segments is forbidden. It remains unclear how this can be experimentally implemented, but the interesting aspect of their model is the presence of a roughening transition, belonging to the directed percolation universality class [3], and unconventional roughness properties at this transition. Our model becomes a directed Ising (DI) type [4,5] generalization of this when we dis-

able digging on flat surface segments. Modify the evaporation probability q to rq when both neighbors are at the same height as the update pair $(i, i + 1)$, i.e., $h_{i-1} = h_i = h_{i+1} = h_{i+2}$. At $r = 0$, the no-digging limit, it becomes impossible to dig into the crystal layers beneath the current lowest exposed level. That level itself becomes frozen as well when it fills up completely.

Figure 4 shows the phase diagram. The rough equilibrium point broadens into a rough phase (the shaded area). Along the *DI-E* phase boundary, the surface growth is zero. Inside the rough phase the surface grows, but slowly. Its scaling properties are complex and obscured numerically by huge corrections to scaling. The surface width W seems to grow logarithmically in time for $L > 2^{10}$, and maybe the stationary state width scales logarithmically as well, but extracting the true scaling properties is a rather hopeless endeavor.

The origin of this complexity is easy to pin point. The rough surface grows slowly, although the bare coupling constants $p < q$ favors evaporation. It performs a delicate balancing act. The surface erosion picture from $r = 1$ still holds along faceted ramp segments. There the surface evaporates due to the downward drift velocity of active bonds along the slope, but this is frustrated by the emergence of pinning hilltops. The surface grows at flat surface segments due to an upward pressure created by the reduced digging probability factor r , but the formation of pinning valleys limits this. Moreover, the nonlocal evenness constraint is at work as well. Growth and evaporation are dynamically balanced only along the faceting transition line *DI-E* (Fig. 4). Everywhere else the rough surface grows slowly.

The properties of the two faceted phases confirm the above intuitive picture. The erosion faceted phase signals the local stability of eroding ramps inside the rough phase, while the growth faceted phase indicates that flat segments persist. We have numerical evidence showing that the

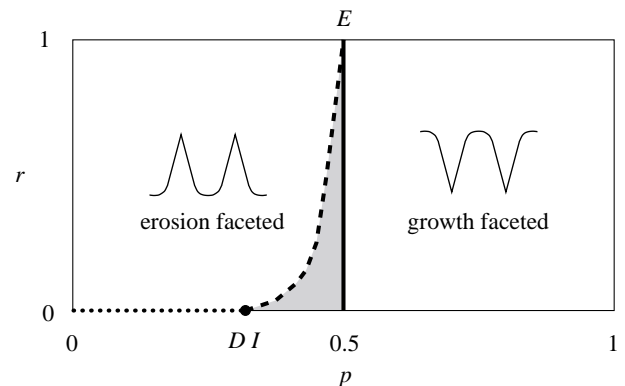


FIG. 4. Phase diagram of the generalized model: The rough equilibrium point E at $r = 1$ broadens into a slowly growing rough phase (shaded) when the digging probability r is reduced. The dotted line to the left of the *DI* critical point at $r = 0$ represents the smooth phase.

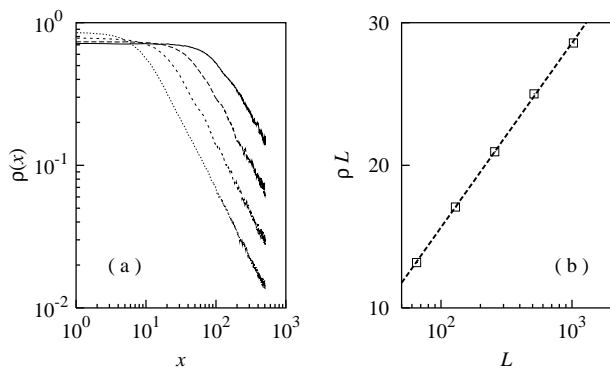


FIG. 5. Scaling at the $p = q$ transition line: (a) Active bond distribution $\rho(x)$, along $p = q$ for system size $L = 1025$ at $r = 0.9, 0.8, 0.6$, and 0.2 (ordered from the top down). $\rho(x) \sim x^{-1}$ decays algebraically beyond a nondiverging hilltop flatness length scale ξ_0 . (b) Scaling of the total active bond density $\bar{\rho} \sim \ln L/L$ at $p = q$ and $r = 0$.

active bond characteristic length ξ_f of the erosion faceted phase does not diverge along the roughening transition line $DI-E$ for $r < 1$. This confirms that eroding ramps remain locally stable. On the other side of the phase diagram, the flatness length scale ξ_0 of the growth faceted phase does not diverge along the $p = q$ roughening line. This confirms the persistence of locally stable flat segments inside the rough phase.

To illustrate this, we present some of the details of the latter. Recall that along $r = 1$, the active bond distribution $\rho(x)$ for different p collapses onto a single curve [Fig. 3(b)]. For $r < 1$ this fails. At the transition point $p = q$, $\rho(x)$ scales algebraically, as $\rho(x) \sim x^{-1}$, see Fig. 5(a), but only beyond the central flat part of the hills. The flatness length scale, ξ_0 remains finite. Its value varies as $\xi_0 \sim |1 - r|^{-\nu}$, with $\nu = 1.0(1)$ along the line $p = q$. In Fig. 5(a), ξ_0 can be as large as $\xi_0 \approx 40$. This explains the poor finite size convergence of the surface roughness inside the rough phase.

The scaling properties of the $p = q$ faceting transition follow from the behavior of the active bond density. Numerically, the surface width scales at $p = q$ as $W \sim L$, as in the faceted phase. From the perspective of the rough phase the faceting transition takes place when the total active bond density, $\bar{\rho} = \frac{1}{L} \int_0^L \rho(x) dx$, vanishes, because in the faceted phase the ξ_0 segments of the rounded hilltops are of measure zero compared to the ramp segments. We find $\bar{\rho} \sim (q - p)^{\beta_\rho}$ with β_ρ very close to 1. At $p = q$ itself, the power law $\rho(x) \sim x^{-1}$ predicts that ρ scales with system size as $\bar{\rho} \sim \ln L/L$. The numerical data in Fig. 5(b) confirm this. Finally, $\bar{\rho}$ decays at $p = q$ algebraically in time with exponent $0.32(1)$. This suggests a dynamic exponent $z = 3.1(2)$.

Erosion below the currently lowest exposed level becomes strictly forbidden at $r = 0$. The evaporation faceted

phase becomes flat, and a DI-type roughening transition takes place at $p = p_{DI} = 0.317(1)$. Hinrichsen and Ódor [12] already documented this. They independently introduced the $r = 0$ limit of our model. They also report that the surface width scales at p_{DI} as $\sqrt{\log(t)}$, and inside the rough phase as $\log(t)$.

In summary, the presence of a nonlocal topological constraint on equilibrium surface configurations in dissociating dimer-type surface growth leads to anomalous reduced surface roughness, with exponent $\alpha = \frac{1}{3}$ instead of the conventional value $\alpha = \frac{1}{2}$. Moreover, the growing (evaporating) surface is always faceted, due to the spontaneous creation of pinning valleys (hilltops). Under other circumstances, in particular, when the digging probability on flat surface segments is being suppressed, an intermediate slowly growing rough phase appears with complex scaling properties and strong corrections to scaling, due to the presence of large internal length scales.

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- [1] A.L. Barabási and H.E. Stanley, *Fractal Concepts in Surface Growth* (Cambridge University Press, Cambridge, 1995); V. Privman, *Nonequilibrium Phase Transitions in Lattice Models* (Cambridge University Press, Cambridge, 1996).
 - [2] M. Kardar, G. Parisi, and Y.C. Zhang, *Phys. Rev. Lett.* **56**, 889 (1986); T. Halpin-Healy and Y.C. Zhang, *Phys. Rep.* **254**, 215 (1995).
 - [3] J. Marro and R. Dickman, *Nonequilibrium Phase Transitions in Lattice Models* (Ref. [1]).
 - [4] W. Hwang, S. Kwon, H. Park, and H. Park, *Phys. Rev. E* **57**, 6438 (1998); M.H. Kim and H. Park, *Phys. Rev. Lett.* **73**, 2579 (1994).
 - [5] P. Grassberger, F. Krause, and T. von der Twer, *J. Phys. A* **17**, L105 (1984); H. Takayasu and A. Yu. Tretyakov, *Phys. Rev. Lett.* **68**, 3060 (1992); N. Inui and A. Yu. Tretyakov, *Phys. Rev. Lett.* **80**, 5148 (1998).
 - [6] M. den Nijs, in *The Chemical Physics of Solid Surfaces and Heterogeneous Catalysis*, edited by D. King (Elsevier, Amsterdam, 1994), Vol. 7, Chap. 4.
 - [7] G.I. Menon, M. Barma, and D. Dhar, *J. Stat. Phys.* **86**, 1237 (1997).
 - [8] P. Politi, G. Grenet, A. Marty, A. Ponchet, and J. Villain, cond-mat/9906289.
 - [9] G.M. Cicuta and M. Contedini, math/9903063.
 - [10] J.D. Noh, H. Park, D. Kim, and M. den Nijs (to be published).
 - [11] U. Alon, M.R. Evans, H. Hinrichsen, and D. Mukamel, *Phys. Rev. Lett.* **76**, 2746 (1996).
 - [12] H. Hinrichsen and G. Ódor, *Phys. Rev. Lett.* **82**, 1205 (1999).