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Mounding instability and incoherent surface kinetics

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Mounding instability in a conserved growth from vapor is analyzed within the framework of adatom kinetics on the growing surface. The analysis shows that depending on the local structure of the surface, kinetics of adatoms may vary, leading to disjoint regions in the sense of a continuum description. This is manifested particularly under the conditions of instability. Mounds grow on these disjoint regions and their lateral growth is governed by the flux of adatoms hopping across the steps in the downward direction. Asymptotically $\ln t$ dependence is expected in $(1+1)$ dimensions. Simulation results confirm the prediction. Growth in $(2+1)$ dimensions is also discussed.

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Mounding instability was experimentally observed and proposed by Johnson *et al.* [1] during the growth of GaAs on (001) GaAs substrate. Initially, activation difference [Schwoebel-Ehrlich (SE) barrier] [2] between adatoms hopping on the plane and the one crossing the step edge was considered responsible [3]. Later it was shown that edge diffusion can also lead to similar effects [4]. The necessary condition for this instability is to have current j_t, uphill on the tilted substrate [5]. One of the issues related to the growth of mounds has been the temporal dependence of the mound growth. Based on various forms of continuum equations the lateral growth is expected to have a time dependence $\sim t^s$, where s takes values from 0.0 to 1/4 [6]. Similarly, the width of the interface is predicted to follow the power law t^β with β varying from 1/3 onwards [6]. All the simulations that do not allow step dissociation invariably find that, for uphill j_t, steepening of mound sides occurs after some time, depending upon the strength of the j_t. The steepening sides finally form sharp ridges in the interface. The time when it just begins is referred to as the onset of instability. Most of the theoretical treatments and simulations do not extend to the region of instability. However, such instability is the asymptotic behavior under uphill j_t. In the growth equation approach, presence of these ridges is considered as singularities and it was proposed that a nonlocal equation may reconcile the presence of ridges with continuum description [7]. The predictions of growth equations [6] are based on the assumption that the underlying conserved growth equation, describing nonequilibrium growth, is valid over the entire substrate. In the following we show that for growth with uphill current, discontinuities appear on the substrate as a consequence of different kinetics in different regions, leading to different growth equations there. It is shown that the power law dependence is possible only for downhill or zero tilt dependent current. Thus above predictions [6] considered at zero tilt dependent current, but not for an uphill current. We argue this by establishing a correspondence between different kinetic processes on the interface and the terms in a growth equation. We consider a stepped region, as in Fig. 1, base region, as the adjoining bottom of two stepped regions and the top region as the highest joint region of two step regions. The kinetics of adatoms in each of these regions show that these regions offer different growth equations over time scales much smaller than τ_{ML}, the time for 1 ML deposition.

We consider growth on a one-dimensional substrate. Figure 1 shows the stepped region under consideration. Growth proceeds through randomly falling adatoms on the surface, which relax by diffusing on the stepped terraces. Adatoms with zero nearest neighbors (nn) are mobile while those with more than zero nn will have negligible mobility. Further, desorption and dissociation from the steps is also negligible at low temperature. Under the conserved growth conditions it is possible to write formally the growth equation in the form $\partial_t h(\mathbf{x},t) = \boldsymbol{\nabla}\cdot\mathbf{j}(\mathbf{x},t)+F$, where F is incident flux, $h(\mathbf{x},t)$ is height function and $\mathbf{j}(\mathbf{x},t)$ is particle current. An uphill current j_t indicates instability while the downhill current indicates the stable Edward-Wilkinson (EW) [8] type growth [5]. Let l_c be the average length traveled by an adatom before getting attached to another adatom or step. The density of steps can be expressed as $|m|/(1+|m|)$, where m is the local slope. Let P_A and P_B be the relative probabilities for hopping across the sites A and B in Fig. 1. By considering current due to the downward hops and that due to the in-plane hops separately, one can show that the resultant nonequilibrium current is given by [9]

$$\mathbf{j}_s = \frac{\hat{\boldsymbol{n}}|m|F(P_B-P_A)}{2(1+|m|)(l_c^{-1}+|m|a^{-1})}, \tag{1}$$

where $\hat{\boldsymbol{n}}$ denotes positive x direction. The presence of l_c^{-1} in the denominator accounts for the nucleation effect on larger terraces. In this expression, the local terrace width is $(l_c^{-1}+a^{-1}|m|)^{-1}$. However, due to the relative velocity between two adjacent terraces, the local terrace width changes. The

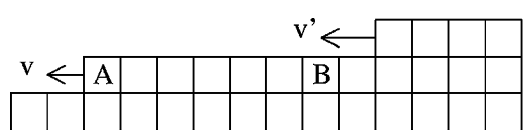

FIG. 1. A typical step structure formed during growth along positive slope. v and v' are velocities of the steps.

velocity difference will be proportional to $\partial j(x,t)/\partial x$. Including this dynamical effect, the expression for the current becomes

$$\mathbf{j}(x)=\frac{\hat{\boldsymbol{n}}|m|F(P_B-P_A)}{2(1+|m|)(l_c^{-1}+|m|a^{-1})}-\frac{\hat{\boldsymbol{n}}F}{4}\partial_x\left(\frac{|m|}{(1+|m|)(l_c^{-1}+|m|a^{-1})}\right)^2. \tag{2}$$

Next, we argue that every downward hop introduces height-height correlation; hence it will give rise to all the stabilizing terms in a growth equation. Under the tilt independent current conditions, the lowest of such terms is $\partial^3 h/\partial x^3$. Thus the current on the stepped surface will be

$$\mathbf{j}(x)=\frac{\hat{\boldsymbol{n}}|m|F(P_B-P_A)}{2(1+|m|)(l_c^{-1}+|m|a^{-1})}+\hat{\boldsymbol{n}}k\frac{\partial^3 h}{\partial x^3}-\frac{\hat{\boldsymbol{n}}|m|F}{4}\partial_x\left(\frac{|m|}{(1+|m|)(l_c^{-1}+|m|a^{-1})}\right)^2. \tag{3}$$

For small slopes, the above current generates a growth equation in the moving frame with average growth rate

$$\frac{\partial h(x,t)}{\partial t}=-\frac{F(P_B-P_A)l_c}{2}\frac{\partial^2 h}{\partial x^2}+\frac{Fl_c^2}{4}\frac{\partial^2}{\partial x^2}\left(\frac{\partial h}{\partial x}\right)^2-k\frac{\partial^4 h}{\partial x^4}+\eta(x,t), \tag{4}$$

where $\eta(x,t)$ is the Gaussian noise in the deposition with the property $\langle \eta(x',t')\eta(x,t)\rangle=\delta(x'-x)\delta(t'-t)$. For $P_A=P_B$, current is tilt free, and the corresponding equation has the Lai–Das Sarma–Villain [3,10] form.

Now we consider the kinetics of adatoms on the top and base regions over time scales much smaller than τ_{ML}. For the base and top regions, by definition the tilt dependent current is zero. On the top region, downward hops from two edges is the only process, leading to the growth term $k\partial^3 h/\partial x^3$. The asymmetric term is absent. On the base region only in-plane hops are allowed. Thus, only the Poisson growth is expected. These arguments indicate that for small time scales, three distinct regions exist on the interface.

In order that this distinction is relevant to growth, it is necessary that the regions remain well defined over time scales of the order of τ_{ML} or more. τ_{ML} is the minimum time dt in the growth equation (4), as it denotes the average time between the height fluctuations at a given site. Accordingly, a base region of width b will be irrelevant in growth dynamics if it shifts on an average by an amount of b or more in time τ_{ML}. Under this condition, the growth equation for stepped region is valid over the entire substrate. On the other hand it is relevant and appears as a discontinuity if the shift is smaller than b. The discontinuity manifests as a singular region in the growth, leading to sharp ridges separating adjacent mounds [7]. The power law dependence of growth fails under this condition and is replaced by $\ln t$ dependence. This dependence has not been previously mentioned in any of the simulations of growth although similar models have been studied in the past [7,11]. We have verified shifting of the base region in time for various SE barriers by inspecting consecutive time profiles of interfaces created in computer simulations of a $(1+1)$-dimensional model described below. From the model simulations, we find that the base region is relevant for uphill current, while it is irrelevant for zero or downhill current j_t.

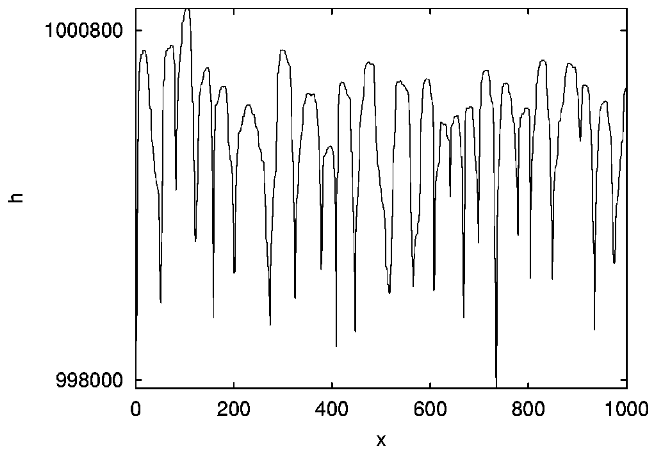

FIG. 2. Morphology of the surface in $(1+1)$ dimensions for an unstable growth after 10^6 layers. Parameter p is 0.6.

In this model, on a one-dimensional substrate, adatoms are rained randomly. An atom with one or more nn is incorporated in the crystal. An adatom with zero nn is allowed to hop n number of times at the most. If it acquires a nn, then no further hops are allowed. If the number of hops are exhausted, it is incorporated at the final site after n hops. A parameter p is introduced, such that $p>0.5$ corresponds to the positive SE barrier. $p=1$ is the case of the infinite SE barrier. This model is similar to that used by Krug [11] in connection with the effect of a detailed balance on the asymmetric term in the growth equation. The model mimics the realistic growth in $(1+1)$ dimension at low temperature and without desorption. We have measured $\langle h_i h_j\rangle$ correlations for various values of p and used the first zero crossing as the measure of the size of the mound. Figure 2 shows the well developed mounds with $p=0.6$. Note the deep ridges formed due to high step heights of the steps forming the ridges. These ridges are the discontinuities of the base region. The mound growth in this case is then decided by the transport of adatoms across the ridges. We estimate the lateral mound growth rate by appealing to the diffusional kinetics of atoms. The growth proceeds by expansion of a larger mound at the cost of a smaller one [7]. Thus the ridge moves laterally in the direction of the expansion of the larger mound. A smaller mound generally makes a smaller angle with respect to the substrate. Thus, relatively longer terraces are present on this mound. The diffusional addition to the ridges is mainly from these terraces, resulting in the shift of the ridge in the direction shrinking the smaller mound. Thus, we assume that adatoms are added from the smaller mound, diffusionally. The diffusional rate of displacement is $dl=D_s^{1/2}t^{-1/2}dt$ on a plane

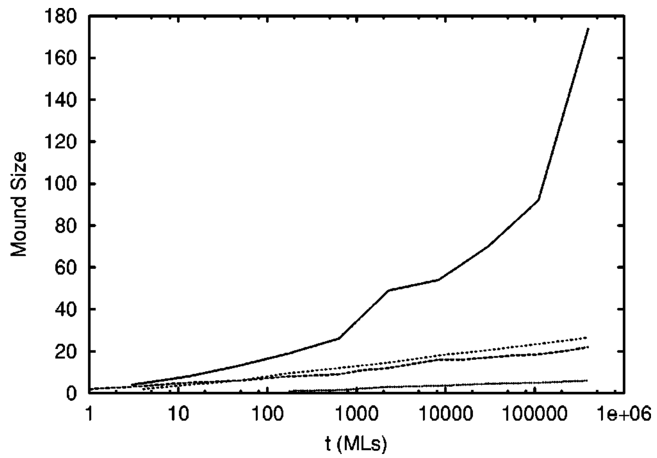

FIG. 3. Time evolution of lateral growth in $(1+1)$ dimensions. The values of parameter p are 0.5, 0.6, 0.7, and 0.8, respectively, for the curves from top to bottom in the figure. The substrate size is $L=10\,000$.

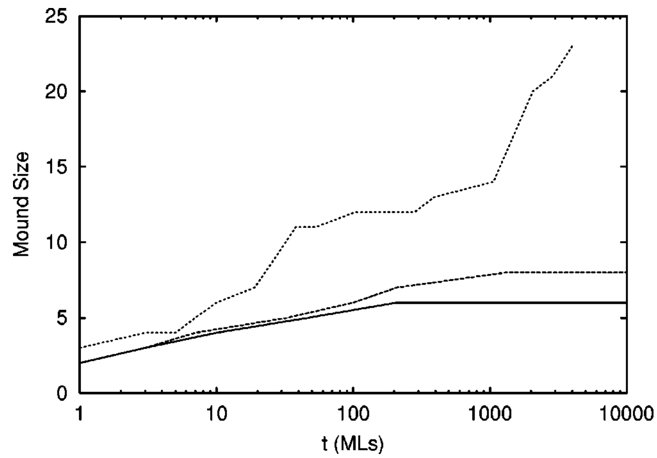

FIG. 4. Time evolution of lateral growth in $(2+1)$ dimensions. The values of parameter p are 0.35, 0.6, and 0.7, respectively, for the curves from top to bottom in the figure. The substrate size is 300×300 for the simulation.

surface in time dt. However, for a ridge to move laterally, it must be filled at least up to the first step height. For sharp ridges as in Fig. 2, the step height of a ridge may be taken to be $\sim w$, the rms height fluctuation (width). Hence, the displacement for a ridge will be $dl_r = pD_s^{1/2}t^{-1/2}aw^{-1}dt$, where, p is the relative fraction of adatoms crossing the step edge and a is the lattice constant. For $w\sim t^{1/2}$, the growth of mounds is proportional to $\ln t$.

In Fig. 3, a plot of mound size vs time on a semilog scale clearly shows that for $p>0.5$, i.e., for the uphill current j_t, the mounds growth is $\ln t$. Also shown is the case for $p=0.5$. $j_t=0$ for this case. We plot the length corresponding to the first maximum in height-height correlations for this case. The curve on the semilog plot is the exponential showing a power law dependence. Correlation length $\xi\sim t^{1/4}$ in this case. In fact it can be shown [9] that the corresponding equation describes Das Sarma–Tamborenea [12] model to which the tilt independent growth equation reduces for large slopes. Further, for $p<0.5$, asymptotically, the EW growth is recovered. Thus a single growth equation describes the growth over an entire substrate for zero and down hill j_t. If the SE barrier is small or uphill j_t is small, power law growth may be observed as a transient before $\ln t$ dependence sets in. In the present model dissociation from steps is not included so that the detailed balance is not followed. If this is included and the current is still uphill, then $\ln t$ dependence continues for growth in $(1+1)$ dimension. This also explains the behavior under an infinite SE barrier of growth [13] where mounds do not grow laterally beyond l_c. In this case, in the absence of downward hops, the base region remains fixed spatially at all times leading to Poisson growth.

Above arguments are true in any dimension. In $(2+1)$ dimensions, mound formation is observed experimentally as well as in simulations [1,11]. Various predictions are referred to in the Introduction regarding the time evolution of the mounds. The present analysis suggests that the $\ln t$ dependence in $(1+1)$ dimensions is the *upper limit* for lateral development of the mounds in $(2+1)$ dimensions for uphill j_t, while power law dependence is possible only for zero or down hill j_t. A slower rate for mound growth is expected in $(2+1)$ dimensions for the following reason. A given mound is surrounded by four or more mounds. The probability that such a mound happens to be the smallest among the surrounding ones, including itself, is very small. A given mound may be reduced in one direction, but it may increase in other direction owing to a smaller mound there. Thus, instead of consumption, shift of mounds is more likely on a two-dimensional substrate. In order to find the time dependence of the mound growth in $(2+1)$ dimensions, we have used the same model described above, except that the rules apply in two directions on a square lattice. In addition, we have included edge diffusion with *no edge barriers*. It is observed that edge diffusion suffices to induce uphill current so that even if the diffusion of single adatoms is unbiased, mound formation is observed. In the absence of edge diffusion, but with unbiased single adatom diffusion, an EW-type growth is obtained [14]. A noise reduction technique [15] is employed with a reduction factor of up to 5, wherever needed. The growth of the mound size is monitored in the same way as for the $(1+1)$ dimensions, using zero crossing for the correlations $\langle h_i h_j\rangle$. Figure 4 shows the plot of the mound size as a function of time on semilog plot. Clearly, after an initial growth like $\ln t$, the curve tends to saturation, confirming the slower growth rate. By varying parameter p, a condition close to the tilt independent current is obtained. The growth in that case follows $t^{1/4}$ power law. From the arguments leading to Eq. (4), in $(2+1)$ dimensions, we find that the asymmetric term will be ineffective if the step edge tension is lower so that the step morphology is wavy or fingered. This is so because the terrace size can be reduced by step movements in the orthogonal directions as well. Thus only the $\nabla^4 h$ term contributes, leading to $\beta=1/4$ and $z=4$ in $(2+1)$ dimensions. Clearly, this observation suggests that in experimental growth, if the SE barrier is very small (but nonzero), at low temperature, the growth rate of mounds can be $t^{1/4}$ in the transient region. If the edge tension is high so that steps are straight and less wavy, the asymmetric term can contribute with β and z, characteristics of a Lai–Das Sarma like equation [10] in the transition region.

In conclusion, we have shown that the growth from a vapor on a surface proceeds via, in principle, a heterogeneous dynamics. The stepped, base, and top regions on the surface allow different growth dynamics. As a result the spatial scalability breaks down over time scales much smaller than τ_{ML}. The effect is distinctly observable under uphill current on the tilted substrate leading to mounds separated by ridges. The kinetics across the mounds suggest a $\ln t$ dependence in $(1+1)$ dimensions, which is verifiable in a suitable model. A slower growth is predicted in $(2+1)$ dimensions, which is also observed in a model simulation. Power law dependence for mounds is only for zero or downhill current.

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