Mounding Instability and Incoherent Surface Kinetics

S.V. Ghaisas
Department of Electronic Science, University of Pune, Pune 411007, India

(March 22, 2002)

Mounding instability in a conserved growth from vapor is analysed within the framework of adatom kinetics on the growing surface. The analysis shows that depending on the local structure on 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.

Mounding instability was experimentally observed and proposed by Johnson et. al [1] during 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 effect [4]. One of the issues related to growth of mounds has been the temporal dependence of mound growth. Based on various forms of continuum equations the lateral growth is expected to have a time dependence \( t^\beta \) where \( s \) takes values from 0.0 to 1/4 [5]. Similarly, the width of the interface is predicted to follow the power law \( t^\beta \) with \( \beta \) varying from 1/3 onwards [3]. All these predictions are based on the assumption that the underlying conserved growth equation describing non equilibrium growth, is valid over the entire substrate. In the following we show that under the conditions of instability and low temperatures, this assumption fails. Consequently the growth of mounds is governed laterally by the adatom kinetics across the mound boundaries. We show this by developing growth equation over a stepped surface in 1+1 dimensions using kinetics of adatoms and steps. This helps establish the correspondence between kinetic processes and terms in the growth equation. We assume that only mechanism of relaxation is by diffusion of adatoms. This allows identification of process and corresponding term uniquely. These assumptions are expected to be valid at low temperature, where evaporation is negligible. Once the kinetic processes leading to various terms in growth equation are identified, presence or absence of such terms in various regions on the surface can be predicted. This allows us to classify different regions on the surface according to the growth equation followed there.

Consider growth on a one dimensional substrate. Fig.1 shows the stepped region under consideration. Growth proceeds through randomly falling adatoms on the surface that 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

\[
\partial_t h(x, t) = \nabla \cdot j(x, t) + F, \quad \text{where, } F \text{ is incident flux, } h(x, t) \text{ is height function and } j(x, t) \text{ is particle current. An uphill current on a tilted substrate indicates instability while downhill indicates stable Edward-Wilkinson (EW) } \| \text{ type growth.}\]

Let \( l_d \) be the average length travelled by an adatom before getting attached to another adatom or step. The density of steps can be expressed as \( \frac{\hat{n}|m|}{l_d + |m|a^{-1}} \). 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 seperately, one can show that the resultant nonequilibrium current is given by

\[
j_s = \frac{\hat{n}|m|F(P_B - P_A)}{2(1 + |m|)(l_d^{-1} + |m|a^{-1})} \quad (1)
\]

Where \( \hat{n} \) denotes +ve x direction. Presence of \( l_d^{-1} \) in the denominator accounts for the nucleation effect on larger terraces. In this expression, local terrace width is \( l_d^{-1} + a^{-1}|m| \). However due to the relative velocity between two adjacent terraces, the local terrace width changes. The velocity difference will be proportional to the \( \frac{\partial j(x, t)}{\partial x} \). Including this dynamical effect, the expression for the current becomes,

\[
j(x) = \frac{\hat{n}|m|F(P_B - P_A)}{2(1 + |m|)(l_d^{-1} + |m|a^{-1})} - \frac{\hat{n}F}{4} \frac{\partial j(x, t)}{\partial x} \left( \frac{|m|}{(1 + |m|)(l_d^{-1} + |m|a^{-1})} \right)^2 \quad (2)
\]

Next, we argue that every downward hop introduces height-height correlation, hence will give rise to all the stabilizing terms in a growth equation. Under the tilt independent current conditions, the lowest of such terms is \( K \frac{\partial^2 h}{\partial x^2} \). Thus the current on the stepped surface will be,

\[
j(x) = \frac{\hat{n}|m|F(P_B - P_A)}{2(1 + |m|)(l_d^{-1} + |m|a^{-1})}
\]

1
formed due to high step heights of the steps forming the ridges. The model used for this growth will be described later. It suffices to know that its a growth with finite diffusion of adatoms and finite SE barrier. We estimate the growth rate by appealing to the diffusional kinetics of atoms. The growth proceeds by expansion of a larger mound at the cost of smaller one [11]. Thus the ridge proceeds in one direction. A smaller mound generally poses 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 to the shift of ridge in the direction of smaller mound. Thus, we assume that adatoms are added from the smaller mound, diffusionaly. The diffusional rate of displacement is \( d = D_s^{1/2}t^{-1/2}dt \) on a plane surface in time \( dt \). However, for a ridge to move laterally, it must be filled at least up to first step height. For sharp ridges as in Fig.2, the step height of ridge may be taken to be \( \sim w \), the rms height fluctuation (width). Hence, the displacement for a ridge will be \( d_r = pd_{st}^{1/2}t^{-1/2}aw^{-1} \). Where, \( p \) is relative fraction of adatoms crossing the step edge and \( a \) is lattice constant. For \( w \sim t^{1/2} \), the growth of mounds is proportional to \( int \).

We verify the \( int \) dependence for a 1+1 dimensional model that mimics the growth at low temperature. In this model, on a one dimensional substrate, adatoms are rained randomly. An atom with one or more \( mn \) is incorporated in the crystal. An adatom with zero \( mn \) is allowed to hop \( n \) number of times at the most. If it acquires a \( mn \), then no further hops are allowed. If number of hops are exhausted, it is incorporated at the final site after \( n \) hops. A parameter \( p \) is introduced, such that for \( p > 0.5 \) hopping across a step in the downward direction is difficult. \( p = 1 \) is the case of infinite SE barrier. We have measured \( < h_i h_j > \) correlations for various values of \( p \) and used the first zero crossing as the measure of the size of the mound. In Fig. 3, plot of mound size Vs. time on a semi log scale clearly shows that for \( p > 0.5 \) i.e. for positive SE barrier, the mounds growth is \( int \). Also shown is the case for \( p = 0.5 \). We plot length corresponding to the first maximum in height-height correlations for this case. The curve on semi-log plot is exponential showing a power law dependence. Correlation length \( \xi \sim t^{1/4} \) in this case. In fact it can be shown [8] that corresponding equation describes Das Sarma - Tamborenea (DT) [3] model to which the tilt independent growth equation reduces for large slopes. We find that for \( p < 0.5 \), asymptotically, EW growth is recovered. Thus, the base and top regions move at least in phase with the \( \xi \) to provide rough surface. 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 \( int \) dependence continues for growth in 1+1-dimension.
dimensions, mound formation is observed experimentally as well as in simulations \[1,13\]. Various predictions are referred in the introduction above regarding the time evolution of the mounds. The \( \text{int} \) dependence in 1+1- dimensions is the upper limit for lateral development of the mounds in 2+1- dimensions. This is so because, a given mound is surrounded by four or more mounds. Probability that such a mound happens to be the smallest amongst 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 mound growth in 2+1- dimensions, we have used 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, EW type growth is obtained \[14\]. Noise reduction technique \[13\] is employed with reduction factor of 5. The growth of mound size is monitored in the same way as for the 1+1- dimensions, using zero crossing for the correlations \( \langle h_i h_j \rangle \). Fig.4 shows the plot of mound size as a function of time on semi-log plot. Clearly, after an initial growth like \( \int t \), the curve tends to saturation, confirming the slower growth rate. By varying parameter \( p \), a condition close to 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 asymmetric term will be ineffective if step edge tension is lower so that steps 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 \( \nabla^4 h \) term contributes, leading to \( \beta = 1/4 \) and \( z = 4 \) in 2+1- dimensions. Clearly, this observation suggests that in experimental growth, if SE barrier is very small (but nonzero), at low temperature 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, asymmetric term can contribute with \( \beta \) and \( z \), characteristics of a Lai-Das Sarma like equation \[9\] in the transition region.

In conclusion, we have shown that growth from vapor on 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. The effect is distinctly observable for unstable growth leading to mound formation. The kinetics across the mounds suggest a \( \text{int} \) 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.

\[\begin{array}{c}
 V \\
 A \\
 B \\
 V' \\
\end{array}\]

FIG. 1. A typical step structure formed during growth along positive slope. \( v \) and \( v' \) are velocities of the steps.
FIG. 2. Morphology of the surface in 1+1- dimensions for an unstable growth after $10^6$ layers. Parameter $p$ is 0.6.

FIG. 3. Shows 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 300 X 300 for the simulation.

FIG. 4. Shows 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 $L = 10000$. 