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A simple model of epitaxial growth

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Abstract

A discrete solid–on–solid model of epitaxial growth is introduced which, in a simple manner, takes into account the effect of an Ehrlich–Schwoebel barrier at step edges as well as the local relaxation of incoming particles. Furthermore a fast step edge diffusion is included in $2 + 1$ dimensions. The model exhibits the formation of pyramid–like structures with a well–defined constant inclination angle. Two regimes can be distinguished clearly: in an initial phase (I) a definite slope is selected while the number of pyramids remains unchanged. Then a coarsening process (II) is observed which decreases the number of islands according to a power law in time. Simulations support self–affine scaling of the growing surface in both regimes. The roughness exponent is $\alpha = 1$ in all cases. For growth in $1 + 1$ dimensions we obtain dynamic exponents $z = 2$ (I) and $z = 3$ (II). Simulations for $d = 2$ seem to be consistent with $z = 2$ (I) and $z = 2.3$ (II) respectively.

Among the different morphologies of growing surfaces in molecular beam epitaxy, the frequently observed formation and coarsening of pyramid–like structures with a well–defined slope is of particular interest [1, 2, 3, 4, 5]. This kind of behavior has been seen in experiments of molecular beam epitaxy in such diverse systems as Fe(001) [6, 7], Cu(001) [8], GaAs(001) [9, 10], and HgTe(001) [11]. The selection of a constant inclination angle can be associated with the compensation of slope–dependent uphill and downhill currents of deposited particles. This has been studied by numerical integration of continuum equations [1, 2, 4].

In the framework of our discrete model, particles are deposited on a d–dimensional simple cubic lattice with L^d sites. The prescription obeys a solid–on–solid restriction, i.e. neither overhangs nor holes will occur and the growing structure is completely characterized by integer height variables assigned to each of the sites. The time t associated with the growth process is defined as $t = N/L^d$ where N is the total number of deposited particles. We will consider a relaxation mechanism for the incoming particles as well as an additional diffusion process with high energy barriers at step edges. Only after the particle has reached its final position, a new one is deposited on the surface.

To begin with, the model is explained in terms of a one–dimensional substrate ($d = 1$) which is initially flat ($h_j(t = 0) = 0$ for $j = 1, 2, \ldots, L$) and satisfies periodic boundary conditions. A new particle is placed with equal probability anywhere on the surface, say at site i. If any of the neighbor sites j (with $|j - i| \leq R_{inc}$) offer a lower height $h_j < h_i$, the freshly deposited particle is moved to the lowest of these sites (a random one in case of a tie). This models the effect of particles reaching the substrate with a residual momentum and kinetic energy [12]. The parameter R_{inc} is called the *incorporation radius* of the model.

Without additional diffusion this prescription would coincide with the well–known *Random Deposition with Surface Relaxation* (RDSR) [2]. RDSR belongs to the Edwards–Wilkinson universality class the scaling behavior of which is known analytically [2, 13, 14]. In particular, the surface becomes flat in $2 + 1$ dimensions, i.e. its width increases only logarithmically with time and system size.

However, if the incoming particle is not yet bound to in–plane neighbors after relaxation, it is considered to diffuse along the surface. Now, an energy barrier at step edges can effectively surpress downward moves since the above mentioned residual momentum has been lost. In the following, this Ehrlich–Schwoebel barrier is taken to be infinite. Consequently, the diffusion is restricted to connected sites of equal height h_i. The main result of this Letter is that the interplay of this diffusion and the incorporation mechanism yields a stable slope which is controlled by R_{inc} only.

The second parameter of the model is the diffusion length l_D which marks the maximum distance that the particle can explore before it comes to rest. A simplistic prescription is used to replace the actual diffusion in $1 + 1$ dimensions: if a potential in–plane neighbor is available within a distance l_D the particle is placed there, otherwise it sticks to the deposition site. This mimics the nucleation of diffusing particles with a mean free path of order l_D [15], a quantity which depends on the ratio of the diffusion constant and the incoming flux (see e.g. [16]).

The diffusion process alone (without relaxation) does not alter the frequencies at which values h_j occur in comparison with simple *Random Deposition* (RD). Thus, formally, $\langle h \rangle = t$ and $w(t) = (\langle h^2 \rangle - \langle h \rangle^2)^{1/2} \propto t^{\beta}$ with $\beta = 1/2$ where $\langle \ldots \rangle$ denotes the average over all lattice sites. The width w of the surface grows indefinitely and no saturation due to lateral correlations is observed. However, the spatial distribution of h–values is changed drastically. The infinite Ehrlich–Schwoebel barrier results in the formation of separate *islands* of linear size $\propto l_D$ on the flat substrate. Without local relaxation no coarsening is possible, the number of islands and their sizes remain constant after a short transient. However, their height and thus the slopes grow indefinitely resulting in the RD power law. The profile of a single island

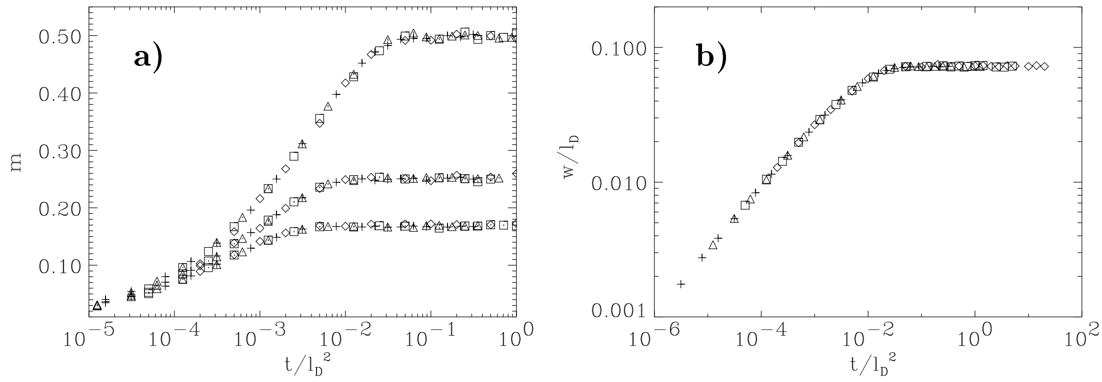

Figure 1: Growth in $1+1$ dimensions: evolution of a single island for various system sizes $L = l_D = 100(\diamond)$, $200(\Box)$, $400(\triangle)$, and $800(+)$. Results were averaged over 50 independent runs, error bars would be smaller than the symbols. a) shows the average local slope m vs. scaled time for three different values of the incorporation radius (top to bottom: $R_{inc} = 1, 2, 3$). In b) the evolution of the surface width w is displayed for the same system sizes and $R_{inc} = 1$.

is given by the inverse of a cumulative binomial distribution (i.e. an inverse error function in the continuum limit) as was observed independently by Krug [17].

The net effect of the infinite Ehrlich–Schwoebel barrier is an uphill current of deposited particles which can be compensated for by the relaxation process described above. The resulting slope selection is most clearly studied for $l_D = L$ when only one island forms. Figure 1(a) shows for this case the evolution of the average local height difference $m = \langle\, | h_{j+1} - h_j \,|\, \rangle$ for three different values of R_{inc}. (The profiles produced by our model are strictly monotonic along the slopes of the pyramids, otherwise m would systematically over–estimate their inclination.) The curves for different system sizes $L = l_D$ collapse when time is scaled with l_D^2, indicating that after a characteristic time $t_m \propto l_D^2$ a constant slope is reached.

Note that if a particle is deposited on a terrace of length n, it jumps downward with probablity R_{inc}/n whereas it is added to the upper terrace with probability $1 - R_{inc}/n$. Therefore, the selected slope for which uphill and downhill currents cancel is $m_{sat} = (2\,R_{inc})^{-1}$ in excellent agreement with the simulations. A more detailed presentation of this argument, also in terms of a continuous description of the step dynamics, will be given in a forthcoming publication [18].

While the single island forms, the width w of the corresponding surface increases until it reaches its saturation value $w_{sat} = m_{sat}\, L\,/\sqrt{48}$. Thus, the roughness exponent is $\alpha = 1$ as expected for a constant slope. Simulations suggest a growth exponent $\beta = 1/2$ which corresponds to a dynamic exponent $z = 2$ in agreement with the time scale found for the slope selection process. Apparently, the relaxation process is basically irrelevant for the small time behavior and w increases as in Random Deposition. Figure 1(b) displays the evolution of w for various system sizes,

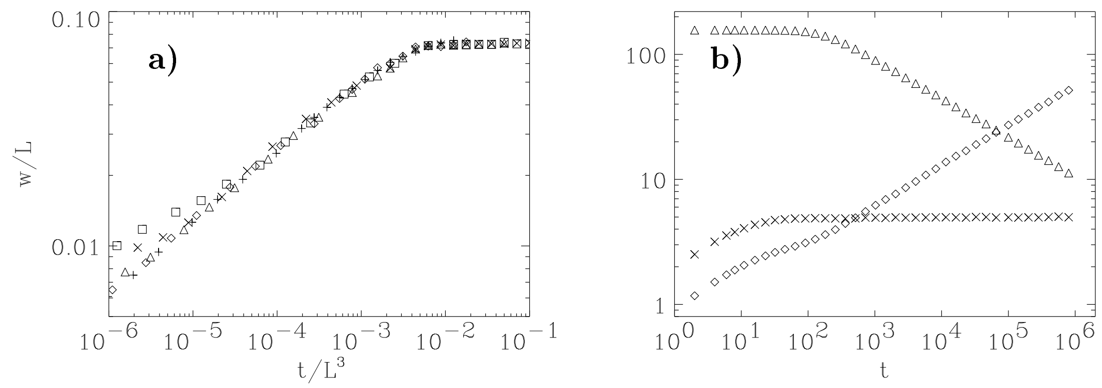

Figure 2: The dynamics of the model for $l_D = 20 \ll L$ in 1+1 dimensions: a) surface width vs. time, rescaled according to the assumption $z = 3$ and $\alpha = 1$ for system sizes $L = 200(\square)$, $283(\times)$, $400(\triangle)$, $566(\diamond)$, and $800(+)$. Averages were performed over 50 runs with standard errors smaller than the symbol size. In b) the evolution of $w(\diamond)$, $10 \cdot m(\times)$, and n_{isl} $(\triangle)$ is shown for a system with $L = 5000$ and $l_D = 20$ on average over 40 simulations. After the slope is selected, surface width and number of islands evolve according to $w \propto t^{1/3}$ and $n_{isl} \propto t^{-1/3}$.

rescaled according to the assumed behavior.

Next we investigate the dynamics of many islands. For $1 \ll l_D \ll L$ the number of initially formed pyramids is of the order of L/l_D. The small time behavior is dominated by the slope selection process which is completed after the characteristic time $t_m \propto l_D^2$. In the second phase fluctuations in the deposition of particles induce a coarsening process in which smaller islands are merged into larger ones [19]. The number of islands n_{isl} decreases according to a power law: $n_{isl} \propto t^{-1/3}$. This exponent coincides with theoretical predictions for noise driven coarsening in one–dimensional systems [3, 19] and can be related to domain growth in magnetic systems [20, 21, 22]. Accordingly, the surface width increases like $w \propto t^{1/3}$. After a time of order L^3 the system reaches its saturation state: a single remaining pyramid with the above given constant inclination.

Fig 2(a) shows results of simulations with $l_D = 20$. Under a rescaling according to $z = 3$ and $\alpha = 1$ the curves for different system sizes collapse for times greater than the L–independent value t_m. The above scaling law is very well confirmed by Monte Carlo results provided the number of islands is still fairly large. Fig 2(b) shows the results for $L = 5000$ and $l_D = 20$ with n_{isl} decreasing from about 150 to 10. A more detailed analysis indicates that, close to saturation, the pairwise merging of the last few islands occurs at characteristic times and yields a cascade–like increase of w.

It is important to note that the condition $l_D \gg 1$ has to be satisfied in order to produce the observed behavior. Otherwise, nucleation is likely to occur even on small terraces, the above mentioned monotonicity of the profile is lost, and n_{isl} can fluctuate strongly in time. The detailed behavior of systems with a small diffusion length $l_D = \mathcal{O}(1)$ will be the objective of forthcoming investigations.

In the following we discuss a generalization of the model to 2+1 dimensions. We assume $R_{inc} = 1$ and the diffusion process is simulated explicitely as a random walk under the constraint that the particle remains at the same height. The walk stops when the particle reaches an in–plane neighbor or has accomplished n_D jumps. This corresponds to a typical diffusion length $l_D \approx \sqrt{n_D}$ provided the set of accessible sites has a compact shape (unlike, for example, a fractal cluster).

Hereupon the particle performs an additional diffusion (random walk at constant height) along the step edge until it is bound to (at least) two in–plane neighbors or until n_{D2} moves have been accomplished. We would like to emphasize that without step edge diffusion the properties of the model are changed drastically. For instance, we find a roughness exponent $\alpha \approx 0.8$ for $n_{D2} = 0$ whereas the expected slope selection with $\alpha = 1$ is confirmed for $n_{D2} \gg 1$. The crossover is in itself interesting but here we concentrate on the model with $l_{D2} \propto \sqrt{n_{D2}}$ large enough (of order L) to ensure that the particle reaches the nearest accessible kink site. In this regime the bases of the growing pyramids are virtually of square shape, cf. Fig. 3, whereas smaller values of l_{D2} yield rounded cross–sections.

The overall behavior is qualitatively the same as in 1+1 dimensions. First, a constant slope builds up which is observed most clearly in the formation of a single pyramid ($\sqrt{n_D} \approx L$). Figure 4(a) shows that here the surface width obeys a scaling relation $w = L^\alpha \cdot g(t/L^z)$ where $g(x \gg 1) = const$. Again, slope selection occurs ($\alpha = 1$). The systematic deviation from the prediction $m_{sat} = 1/2$ is of order $1/L$ in finite systems, cf. inset in Fig. 4 (b). This correction is taken into account in Fig. 4 by scaling w with $(m_{sat}L)$ rather than with L.

The observed dynamic exponent is in good agreement with $z = 2$. If $g(x) \propto t^\beta$ for $x \ll 1$, this would imply $\alpha/z = \beta = 1/2$ as in $1+1$ dimensions. Yet, the simple power law seems to hold only for very short times. This can be seen clearly in the inset of Fig. 4(a) where the effective exponent obtained from two consecutive data points is plotted. As already observed in 1+1 dimensions, the surface width increases initially like in RD with $\beta = 1/2$. The relevance of the incorporation mechanism is determined by the number of step edges which is independent of the pyramid size in 1+1 dimensions. However, for $d = 2$ it increases with the island radius already in the submonolayer regime. Thus, the deviation from RD–behavior becomes noticeable much earlier.

Preliminary results concerning the coarsening of many islands ($l_D \ll L$) are shown in Fig. 4(b). The data seem to be consistent with a dynamic exponent $z \approx 2.3$ ($\beta \approx 0.43$) in this regime. Simulations with small and intermediate values of n_{D2} indicate that β is significantly smaller when the step edge diffusion is slower. However, a more precise determination of this value is necessary and should be based on the simulation of larger systems. At the current stage a definite conclusion concerning the scaling behavior cannot be drawn. The above mentioned analogy to the dynamics of domains in magnetic systems would imply $1/\beta = z = 3$ but is unlikely to carry over to epitaxial growth in 2+1 dimensions [3]. Several authors

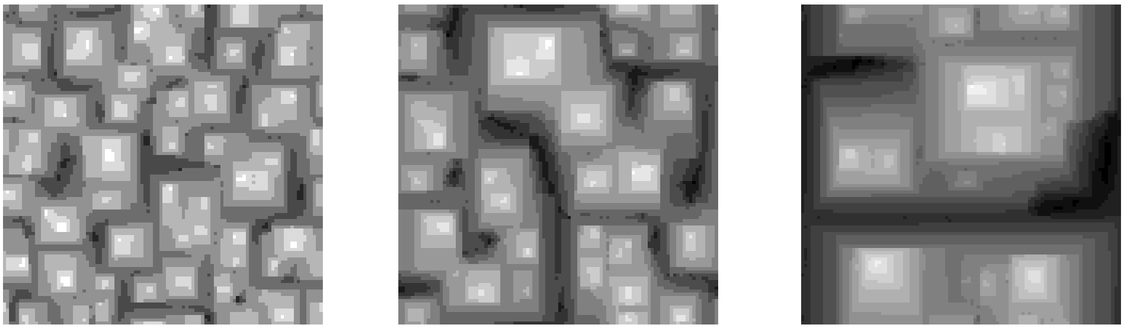

Figure 3: Snapshots (density plots) of the evolution of the surface morpholgy in $d = 2$ at times $t = 4, 16, 64$. The system size is $L = 100$, the model parameters are $R_{inc} = 1$, $l_D = 15$, and $l_{D2} = 200$.

obtain values $\beta \leq 0.25$ from the numerical studies of continuum equations (see [3] and refs. therein). A recent argument by Tang *et al.* [19] suggests that $z = 2 + d$ for noise–driven coarsening.

The relevance of our simple model for molecular beam epitaxy is readily recognized if one identifies the parameter l_D with the typical island distance in the submonolayer regime. Several authors have calculated this length for the simple case in which two adatoms already form a stable nucleus ([16] and refs. therein). For $d = 2$ one obtains $l_D \propto (D/F)^6$ ($(D/F)^4$ in $d = 1$ [15]) where D is the diffusion constant and F the flux of arriving particles. A helpful simplification of our model is due to the infinite Ehrlich–Schwoebel barrier. However, even with a finite (but non–vanishing) barrier the qualitative scenario will be the same. Extensions of the BCF theory [23, 24] which include an incorporation mechanism also yield slope selection [25]. There, however, the tops of pyramids are rounded. Furthermore the terrace width depends on the actual energy barriers and the temperature of the system [18].

In summary, we have presented a simple model which reproduces essential features of epitaxial growth. Local relaxation leads together with surface diffusion in the presence of a high Ehrlich–Schwoebel barrier to the selection of a stable slope of growing islands. We identify two different scaling regimes (both with roughness exponent $\alpha = 1$): the initial slope selection (I) and the coarsening of pyramids (II). For growth on a one–dimensional substrate we find $z = 2$ in regime (I) and $z = 3$ in (II). Simulations in $d = 2$ suggest the same dynamic exponent for (I) and a value of $z \approx 2.3$ in regime (II). Apart from a more accurate measurement of these exponents in larger systems, forthcoming investigations will address the crossover behavior for $l_D = \mathcal{O}(1)$, the precise influence of the step edge diffusion length l_{D2}, the effect of a finite Schwoebel barrier, and a continuous description of the step dynamics [18].

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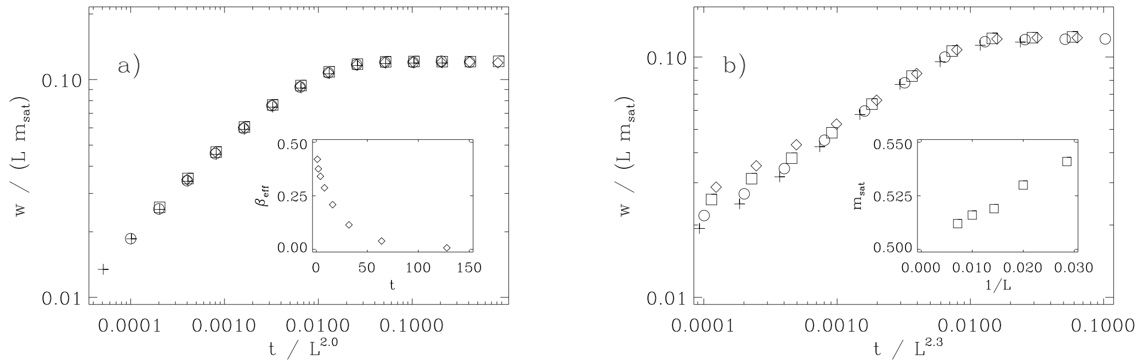

Figure 4: Scaling in 2+1 dimensions for system sizes $L = 140(+)$, $100(\circ)$, $70(\Box)$, and $50(\Diamond)$ with $l_{D2} = 2L$. Results were averaged over 30 independent runs with error bars smaller than the symbols. Fig. a) shows the surface width vs. time for one island ($\sqrt{n_D} \approx L$), rescaled according to $z = 2.0$ and $\alpha = 1$. The inset displays the effective exponent calculated from two consecutive data points with $L = 50$. In b) the surface width is plotted vs. time for the model with coarsening ($n_D = 200$), rescaled according to $z = 2.3$ and $\alpha = 1$. The inset shows the saturation slope vs. $1/L$.

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