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A simple model of epitaxial growth: the influence of step edge diffusion

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Abstract

By studying limiting cases of a simple cubic solid–on–solid model we demonstrate that step edge diffusion (SED) may play a crucial role in epitaxial growth. Our model exhibits the formation and coarsening of pyramids or mounds with a constant inclination angle (slope selection). A comparison of very fast and slow SED shows how the details of this process control both the shape of the emerging mounds as well as the coarsening dynamics. In the absence of SED, slope selection is suppressed and true self–affine scaling is observed.

Molecular beam epitaxy (MBE) has become a standard technique for the production of high quality crystals [1]. Atoms or molecules are evaporated from an oven and then deposited on a substrate. These particles or *adatoms* diffuse on the growing surface and a relatively low incoming flux allows for the growth of nearly perfect crystals. A variety of growth scenarios and surface morphologies has been found in MBE experiments, see e.g. [2] for an overview. For example, the formation of three–dimensional structures which display a constant inclination angle (slope selection) is frequently observed instead of *layer–by–layer* or *step–flow* growth.

Considerable progress towards a theoretical understanding of such non-equilibrium growth processes has been achieved by applying the concept of self–affine height profiles, see [3] for an introduction and overview. A recent review of theoretical approaches to MBE growth can be found in [4]. The investigation of continuous differential equations aims mainly at the identification of potential universality classes and their scaling behavior. Evidently, computer simulations of simplified discrete models play a key role in this context. Several such models have been proposed to describe certain aspects of MBE, see e.g. [5, 6].

In terms of the continuum description slope selection in three–dimensional growth can be explained as a consequence of compensating upward and downward currents on the surface (e.g. [7, 8]). So far, step edge diffusion has not been included explicitly in such continuous models. So–called full diffusion models of MBE frequently display slope selection and take into account step edge diffusion [7,9–13]. There, microscopic energies are used to parameterize the rates of various activated processes in a kinetic Monte Carlo simulation. However, realistic modeling is very time consuming which impedes the precise determination of scaling laws.

In the following we revisit a recently introduced simple cubic solid–on–solid model of epitaxial growth [14, 15]. Our aim is to single out a limited set of microscopic mechanisms and to investigate their relevance for the macroscopic properties of the growing surface.

Due to the SOS restriction, which allows no overhangs or holes, the surface is fully described by assigning integer height variables $h(x, y)$ to the L^2 sites of a square lattice (with periodic boundary conditions, in the following). As we do not include desorption processes in our model, the mean height is given by $\langle h(x, y)\rangle = N/L^2$ after N particles have been deposited on the initially flat substrate. This number of *monolayers* can be interpreted as a time scale ($t = \langle h\rangle$) of the growth dynamics. Following the standard analysis [3] we will investigate the scaling of the surface width $w = (\langle h^2\rangle - \langle h\rangle^2)^{1/2}$ with time and system size.

At each time step, one particle is deposited at a randomly chosen site of the lattice. The first important ingredient of our model is the *incorporation* of incoming particles:

(I) If a lower site is available within a neighborhood of radius R_i, an arriving particle is moved the lowest such site (a random one in case of a tie). In the following this search is restricted to nearest neighbors of the deposition site, i.e. $R_i = 1$.

This local relaxation reflects the fact that incoming particles are able to penetrate the crystal due to their residual momentum perpendicular to the surface. Within our model it is irrelevant whether the deposited particle itself performs a downward move or a perhaps more realistic *knock–out* process occurs [12, 13]. A model which includes only random deposition and process (I) is well–known and widely believed to belong to the Edwards–Wilkinson universality class [3]. Potential non–universal dependences on details of the relaxation are discussed by D.P. Landau in this volume.

A particle which has not been incorporated according to (I) diffuses on the surface, i.e. it performs a random walk on a terrace. Two restrictions apply to this diffusion process in our model:

(II a) Terrace diffusion ends when a particle meets an in–plane neighbor which could be provided at an upward step or, in principle, by another diffusing particle. This latter *nucleation of islands* is taken into account in a simplified manner by limiting the diffusion to a maximum number n_D of steps, after which the particle is considered immobile. On compact non–fractal terraces this corresponds to a characteristic length $\ell_D = \sqrt{n_D}$.

(II b) Downward moves of diffusing particles onto lower terraces are hindered by a so–called Ehrlich –Schwoebel barrier at step edges [3]. Here, we assume this barrier to be very large and simply rule out such moves. Hence, the only possible *inter layer transport* of adatoms occurs by means of process (I).

The diffusion length ℓ_D corresponds to the mean free path in a density of particles and controls the typical distance of *island seeds* in the sub-monolayer regime. (II a) represents the interplay between many mobile adatoms in an effective single particle picture. Hence it is possible to add a new particle only after the last one has become immobile at its final position.

The incorporation (I) constitutes a downward current of particles whereas the Schwoebel effect (II b) results in an effective upward current. Both currents are slope dependent and their cancelation gives rise to the formation of mounds with a stable inclination angle as discussed in [14]. There, a simple argument is given which shows that the system achieves the constant slope $1/(2R_i)$ after a short transient. The argument relies on the existence of extended terraces with a more or less smooth border. One necessary condition for the latter is that $\ell_D \gg R_i$, otherwise the frequent occurrence of nucleation disturbs the equilibrium between up– and downward currents.

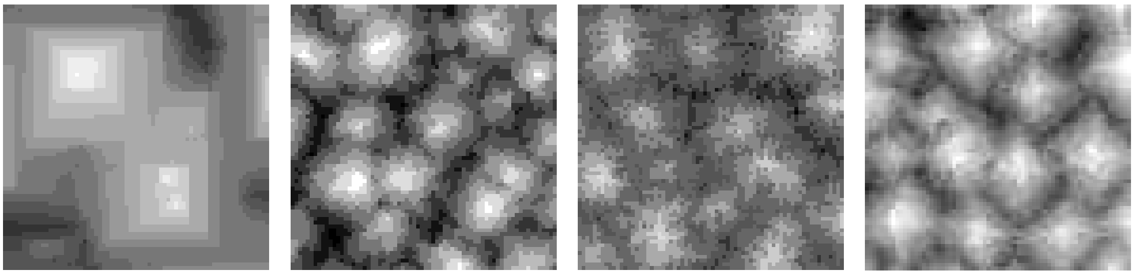
Here we will focus on the role of another mechanism which turns out to control the features of the growing surface to a large extent, the diffusion along step edges (SED):

(III) A particle which is incorporated (I) or sticks to an edge after diffusion (II) performs a random walk along the edge until it reaches an additional neighbor at a kink site. In analogy to (II a), SED is also restricted by a maximum diffusion length ℓ_{SED} along the edge.

In the following we summarize our results concerning particularly clear–cut limiting cases of the model.

The limit of arbitrarily fast step edge diffusion was already studied in [14]. It corresponds to setting $\ell_{SED} = \mathcal{O}(L)$ in the model, which guarantees that every particle is moved to the closest kink site along the edge. Consequently, terrace edges are smooth and aligned with the lattice vectors. Initially, on the order of L^2/ℓ_D^2 pyramids form with square shaped bases and a constant slope $1/2$. Then a coarsening process begins during which pyramids merge. Fig. 1 (a) shows a snapshot of a correspondingly growing surface. As a consequence of slope selection, the surface width is inversely proportional to the decreasing number of pyramids, we find $w \propto t^\beta$ with a growth exponent $\beta \approx 0.45$. A finite system reaches its saturation state with only one remaining pyramid after a characteristic time $t_{sat} \propto L^z$. Obviously, the saturation width satisfies $w_{sat} \propto L^\alpha$ with $\alpha = 1$ in the presence of slope selection and the dynamic exponent is given by $z = \alpha/\beta = 1/\beta \approx 2.20$. It is intuitively clear that fast SED should lead to a very rapid coarsening of structures. Whenever two pyramids merge as shown in Fig. 1 (a), particles are moved efficiently along the straight edges to the region of contact, where a large number of kink sites is provided.

The model still exhibits slope selection with $\alpha = 1$ when particles are nearly immobile at step edges. Here we concentrate on the case $\ell_{SED} = 1$, i.e. particles can perform only one step along the edge. As can be seen in Fig. 1 (b), the shape of the emerging mounds is rounded. Due to the lack of a long range smoothening



a) **b)** **c)** **d)**

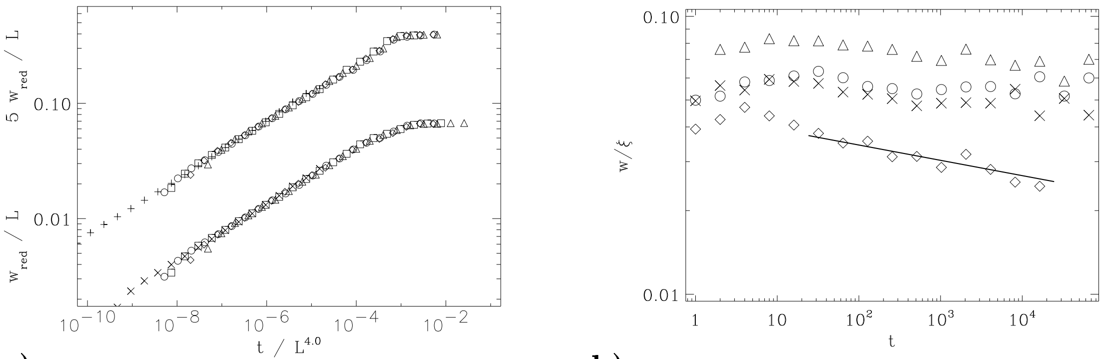
Figure 1: Snapshots of the surfaces after growth of 64 (a) or 128 monolayers (all others). Windows of size 70×70 sites are shown; simulations were performed on a square lattice with $L = 140$, $\ell_D = 15$. Pictures correspond to: $\ell_{SED} = 2L$ (panel a), $\ell_{SED} = 1$ (b), no SED ($\ell_{SED} = 0$) (c), and SED in the presence of an infinite barrier at corners (d) with $\ell_{SED} = 2$.

an alignment of edges with the lattice vectors is not favored here. Apart from this qualitative feature we find that also the scaling behavior is different from the case of fast SED. Figure 2 (a) (lower curve) shows that the (reduced) surface width increases with time according to a growth exponent $\beta \approx 0.25$, and accordingly $z \approx 4$. This corresponds to a coarsening of mounds which is much slower than for fast SED. Long range transportation of material along the edges is impossible and the coarsening is essentially driven by fluctuations in the deposition process. The observed exponents coincide with results obtained in continuum models without explicit SED mechanism [7, 8]. They also agree with a recent hypothesis of Tang *et al.* [16] that purely noise assisted coarsening should be characterized by $z = d + 2$ for $(d + 1)$–dimensional growth in the presence of slope selection.

Preliminary simulations for $\ell_{SED} = 2$ suggest that $\beta \approx 0.27$ in this case. Currently it is unclear whether the scaling behavior changes continuously with increasing ℓ_{SED} or is indeed universal for all finite values of $\ell_{SED} = \mathcal{O}(1)$.

Fig. 1 (c) shows a surface grown in the absence of SED. As for SED with small but non–zero ℓ_{SED}, the emerging structures are rounded. However, the total lack of smoothening SED leads to very rugged terrace edges. The system does not display slope selection but truly self–affine scaling with $\alpha < 1$ and and $\beta \approx 0.20$. Apparently, the absence of well defined extended terraces makes the effective cancelation of downhill and uphill currents impossible. By performing a Fourier analysis of the height profile we have obtained the typical mound size ξ of the system. Fig. 2 (b) displays the temporal evolution of the ratio w/ξ which evolves according to a power law with exponent $(\alpha - 1)/z \approx -0.05$, i.e. $\alpha \approx 0.8$ and $z \approx 4.0$. As expected w/ξ is found to be roughly constant for all cases which display slope selection ($\alpha = 1$). It is important to note that our findings contradict the hypothesis of [16] that $2\beta + d/z = 1$ for general noise assisted coarsening.

Finally, we address the influence of an additional energy barrier which hinders SED moves around corner sites in analogy to the Schwoebel effect. Experimental



a) **b)**

Figure 2: a) Evolution of the reduced width (see [3]) $w_{red} = (w^2 - w_i^2)^{1/2}$ for the model with restricted SED with $\ell_{SED} = 2$ (upper curve, $5 \times w_{red}$) and without a barrier at step edge corners with $\ell_{SED} = 1$ (lower curve). Here, the intrinsic widths $w_i = 0.62$ (upper) and $w_i = 0.67$ (lower) have been subtracted for all system sizes, $L = 80(\triangle), 100(\diamond), 128(\square), 140(\circ), 256(\times), 512(+)$. Averages are over 10 independent simulations, error bars would be smaller than the symbol size.
b) The width w divided by the characteristic length ξ parallel to the substrate. The latter is obtained from a Fourier analysis of the height profile. System sizes are $L = 512$ and 256 respectively ($\circ$, $\times$), the symbols represent: ($\triangle$) the model with restricted SED (here $\ell_{SED} = 2$) and without restriction for $\ell_{SED} = 1$ ($\times$) and $\ell_{SED} = 2$ ($\circ$) respectively. In the absence of SED ($\diamond$) w/ξ follows a power law, the solid line corresponds to $(\alpha - 1)/z = -0.05$.

and theoretical evidence for such an effect is presented in [10, 11], for instance. As a limiting case we consider an infinite barrier at corners, referred to as restricted SED in the following. The movement of particles through SED (III) is thus always limited to a straight portion of the edge. In Fig. 1 (d) a snapshot of an accordingly growing surface is shown. Well–defined pyramids with square shaped bases, tilted w.r.t. the lattice vectors by 45^o, and a stable slope have formed in contrast to the rounded *cones* of Fig. 1 (b). Despite the obvious differences in surface morphology, the scaling behavior with and without the restriction of SED is approximately the same for small ℓ_{SED}. In Fig. 2 (a) we compare the evolution of the corresponding (reduced) width w_{red}, the data is consistent with $z = 4$ and $\beta = 1/4$ in both cases. The value of ℓ_{SED} should be largely irrelevant in restricted SED. After a (ℓ_{SED}–dependent) transient, diamond shaped terraces should always emerge, along which particles can diffuse only a few steps in any case.

In summary, our findings demonstrate clearly that the qualitative and quantitative features of epitaxial crystal growth depend crucially on the presence and the detailed properties of step edge diffusion. This offers one possible explanation for the large variety of morphologies and scaling behaviors that can be observed in experiments and computer simulations. Forthcoming investigations should address the precise determination of scaling exponents in larger systems, in particular we will investigate the behavior for intermediate values of ℓ_D and ℓ_{SED}. The study of more realistic models, for instance with finite energy barriers at downward steps and corners should allow for a more quantitative comparison with experimental results.

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