The role of step edge diffusion in epitaxial crystal growth

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The role of step edge diffusion (SED) in epitaxial growth is investigated. To this end we revisit and extend a recently introduced simple cubic solid-on-solid model, which exhibits the formation and coarsening of pyramid or mound like structures. By comparing the limiting cases of absent, very fast (significant), and slow SED we demonstrate how the details of this process control both the shape of the emerging structures as well as the scaling behavior. We find a sharp transition from significant SED to intermediate values of SED, and a continuous one for vanishing SED. We argue that one should be able to control these features of the surface in experiments by variation of the flux and substrate temperature.

Key words: Computer simulations; Models of surface kinetics; Growth; Surface Diffusion; Surface structure, morphology, roughness, and topography;
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1 Introduction

The investigation of non-equilibrium growth processes has attracted considerable attention in the past decades. Despite the lack of a general theory as in thermal equilibrium systems, significant progress has been achieved using the concept of self-affine growing surfaces (see [1] for an introduction and overview).

Molecular beam epitaxy (MBE) is a standard technique to obtain high quality crystals needed for e.g. semiconductor devices. In MBE particles are evaporated from an oven, cross a vacuum chamber and are deposited on a substrate. The particles diffuse on the growing surface and a relatively low incoming flux allows for the formation of nearly perfect crystals. Much effort has been

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devoted to a theoretical understanding of MBE growth and the various morphologies and scaling behaviors observed in experiments, see e.g. [2] for an overview of experimental investigations and [3] for a recent review of theoretical approaches. Several simple models have been proposed to describe specific aspects of MBE [4,5], but no unified scaling picture has yet emerged. Many of these models fail to explain the frequently observed slope selection, i.e. the formation of pyramids or mounds on the growing surface which show a constant inclination (independent of time) [6–12]. In terms of a continuum description slope selection has been suggested to be due to the balance of upward and downward currents on the surface [13,14]. Only recently, processes similar to step edge diffusion have been considered in the study of continuum equations [15,16]. So-called full diffusion models of MBE frequently take into account step edge diffusion [17–21]. There, microscopic energies are used to parameterize activated jump rates of the particles. However, simulations of these models are very time consuming which prevents a precise determination of the scaling laws. In the (commonly used) simplest models SED is not included explicitly [1] [17]. Whereas the effect of SED on submonolayer growth has been adressed by several authors [18–20], its influence on mound formation and coarsening has not been investigated [17,21].

Recently, we have introduced a simple cubic solid-on-solid model of epitaxial growth [22] in which slope selection emerges from the competition of two mechanisms:

(A) When a particle arrives at the surface its residual momentum perpendicular to the substrate enables it to move to a site of lower height, if such a site is available within a neighborhood of radius R_i.
(B) Otherwise, the particle diffuses on a terrace of the surface until it binds to an upward step or meets another diffusing particle. This latter *nucleation* is taken into account in a simple manner by means of a maximum diffusion length ℓ_D. Furthermore, a large Ehrlich-Schwoebel barrier rules out downward moves to lower terraces. Hence, such downward moves are only possible upon arrival through process (A).

The incorporation (A) of particles constitutes a downward current (downward funneling or transient diffusion [23,24]) whereas the Schwoebel effect (B) yields an effective upward current [25,26]. Both currents are slope dependent and their cancellation gives rise to the formation of mounds with a constant inclination as shown in [22]. At later stages, mounds grow and merge until a single one remains which occupies the entire substrate.

In this article we demonstrate that the features of the growing surface depend crucially on another mechanism, the diffusion of particles along step edges

[1] The diffusion along a step edge or the detachment away from a step have the same energy barrier.

(SED):

(C) A particle which is incorporated (A) or reaches an edge by diffusion (B) can hereupon move randomly along the edge until it finds an additional in-plane neighbor at a kink site. In analogy to process (B), SED is also limited by a maximum diffusion length ℓ_{SED}.

We want to mention, that a similar model has been studied by Bartelt and Evans [27]. However, they achieved smooth step edges not by means of SED. Rather, they artificially enforced square based shaped islands by explicitly rearranging them after the attachment of particles without treating the intersection of islands in a realistic way.

In this paper we investigate the following limiting cases for ℓ_{SED} (note that all length scales will be given in lattice constants):

- *significant* SED (*i.e.* ℓ_{SED} of the order of the system size L) in Sec. 3.1,
- *vanishing* SED ($\ell_{\mathrm{SED}} = 1$, Sec. 3.2) which is shown to be comparable to growth with an additional Ehrlich–Schwoebel barrier for SED (Sec. 3.3) which we will refer to as *restricted* SED,
- and no SED ($\ell_{\mathrm{SED}} = 0$) in Sec. 3.4.

In addition, the crossover for intermediate values of ℓ_{SED} is investigated in Sec. 4. Concluding remarks are given in Sec. 5.

2 Model description

We consider the deposition of particles on a simple square lattice and assume that a solid-on-solid restriction is fulfilled, i.e. the surface can be described by an integer data array of heights $h(x, y)$. The discussion is restricted to models with an incorporation radius $R_i = 1$ in process (A). Greater values should only change the average terrace width of the resulting mounds. A simple argument for this was already given in [22]: the frequency of downward moves (A) on a given terrace of width ℓ_T is proportional to R_i. If $\ell_D > \ell_T$ the upward current is proportional to $\ell_T - R_i$. Only for $\ell_T = 2R_i$ these currents cancel and slope selection is achieved. The diffusion on terraces (B) is simulated explicitly as a random walk which is restricted to sites of equal height. The assumption of an infinite Ehrlich–Schwoebel barrier is not unrealistic. Values of 0.2 eV have been reported for metal homoepitaxy [28,29]. At room temperature this implies that diffusion is by a factor 2000 faster than jumps over step edges. Clearly, smaller Ehrlich–Schwoebel barriers increase the downward current and lead to larger terrace widths [30].

Starting from a flat substrate in MBE, the initial number of mounds can be controlled by the flux of the deposited particles. In our model, this effect (deriving from the interplay between many particles) is described in a simplistic manner: If a particle has not found an upward step edge after n_D moves it is treated as if it had met another particle and formed an immobile nucleus. The diffusion length ℓ_D is thus related to the distance of islands in the sub-monolayer regime and corresponds to the mean free path in a density of diffusing particles. On compact non-fractal terraces a maximum number of diffusion steps n_D corresponds to $\ell_D = \sqrt{n_D}$. Theories [31] and simulations [32] predict in this case that $\ell_D \propto (D/F)^{1/6}$ with the diffusion constant D and the flux F of arriving particles. In our model neither the temperature nor the flux can be varied explicitly but enters indirectly through ℓ_D. In all simulations presented here we have set $\ell_D = 15$, which merely fixes the initial number of mounds on the substrate [22]. Since SED is an effectively one-dimensional process we can replace it by a deterministic search for the nearest kink site within a distance $\pm\ell_{\rm SED}$ along the step edge. To begin with, we assume that no further restriction applies to the SED; the presence of a barrier at step edge corners will be discussed later. Again, the characteristic length $\ell_{\rm SED}$ can be related to the reduced flux f (per unit length) and the corresponding diffusion constant δ: $\ell_{\rm SED} \propto (\delta/f)^{1/4}$.

Only after the particle has reached its final position a new particle is placed randomly on the surface. This very efficient algorithm allows for the simulation of large systems over several decades in time which is well beyond the capabilities of conventional full diffusion models. As our model does not include the possibility of desorption, time can be expressed in terms of *monolayers*, i.e. the total number of deposited particles divided by the system size L^2.

3 Limiting cases

In all models considered here, an initial fast formation of mounds is observed. In the presence of SED (3.1,3.2,3.3) the slope remains unchanged in the subsequent coarsening process. Hence the ratio of the lateral mound size and their height remains constant. Only in the absence of SED (3.4) we observe time dependent slopes.

3.1 Significant SED

In our earlier investigation of the limit of significant SED ($\ell_{\rm SED} = \mathcal{O}(L)$) we observed two distinct regimes: First, pyramids (their number is of the order of L^2/ℓ_D^2) with a well defined slope form and their square shaped bases reflect

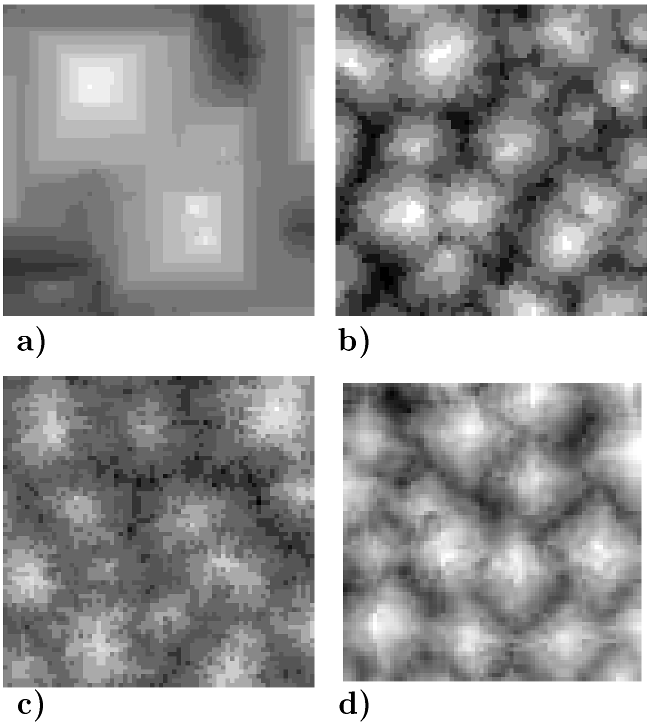

Fig. 1. Snapshots of the surfaces after growth of 64 (panel a) or 128 monolayers (all others). Windows of size 70×70 sites are shown; simulations were performed on a square lattice with $L = 140$, $\ell_D = 15$. Pictures correspond to: $\ell_{\rm SED} = 2L$ (panel a), $\ell_{\rm SED} = 1$ (b), no SED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

the lattice structure, cf. Fig. 1(a). Then, a coarsening process starts which is characterized by two scaling exponents. The surface width

$$w = \sqrt{\langle (h(x,y) - \langle h \rangle)^2 \rangle}$$

increases according to a power law $w \propto t^{\beta}$ with a growth exponent $\beta \approx 0.45$. We observe that w saturates after a time $t_{\times} \propto L^z$ at a value $w_{sat} \propto L^{\alpha}$ where the dynamic exponent z satisfies $z = \alpha/\beta$. This corresponds to the scaling picture of Family and Vicsek for self-affine surfaces [33]. In the saturation state only one pyramid of the size of the system remains in our model. As a direct consequence of slope selection the saturation width increases linearly with L. Hence the roughness exponent is $\alpha = 1$ (and $z = 1/\beta \approx 2.20$).

It is intuitively clear that fast SED should result in a very rapid coarsening of the structures. Whenever two pyramids merge, as for instance shown in Fig. 1 (a), material is moved efficiently along the straight edges towards the region where the base terraces touch and a large density of kink sites is provided. Hence fast SED fills the inner corners at the regions of contact.

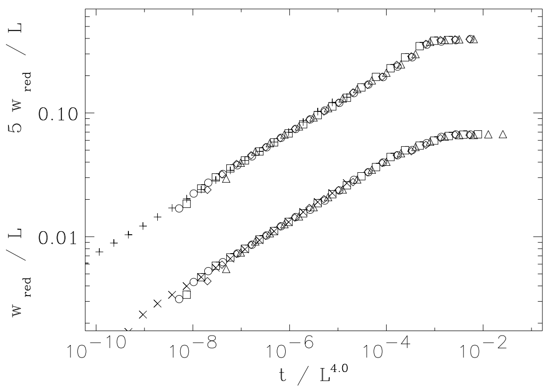

Fig. 2. The evolution of the reduced surface width for the model with restricted SED with $\ell_{\rm SED} = 2$ (upper curve, w_{red} multiplied by 5) and without a barrier at step edge corners with $\ell_{\rm SED} = 1$ (lower curve). Here, the values $w_i = 0.62$ (upper) and $w_i = 0.67$ (lower curve) have been used for all system sizes, $L = 80(\triangle), 100(\diamond), 128(\square), 140(\bigcirc), 256(\times), 512(+)$. Averages are over 10 independent simulations, standard error bars would be smaller than the size of the symbols.

3.2 Vanishing SED

Systems in which the particles are nearly immobile at step edges still exhibit slope selection with $\alpha = 1$, but otherwise a significantly different scaling behavior is found. In this section we will investigate the limiting case $\ell_{\rm SED} = 1$. The extension to larger $\ell_{\rm SED}$ of order one will be postponed until Sec. 4. Fig. 2 (lower curve) shows the evolution of the surface width with time for $\ell_{\rm SED} = 1$. Here, an intrinsic width w_i has been taken into account. Following [1] we have plotted $w_{red} = \sqrt{(w^2 - w_i^2)^2}$ where w_i is an L-independent contribution to the total width. In our model, one source of the intrinsic width is clearly the initial slope selection process. The value of $w_i = 0.67$ is close to the estimate $\ell_D/(2R_i\sqrt{72}) \approx 0.88$ for perfect square pyramids of size ℓ_D and slope $1/2R_i$ which are formed within the first t_0 monolayers of growth. As w_i is less than one monolayer, it can be neglected in the long time limit. Curves for different system sizes collapse under the scaling assumption for the reduced width w_{red} with $\alpha = 1$ and $z = 4.0$ respectively $\beta = 0.25$.

The lack of a rapid transportation of material along edges has two consequences for the surface morphology. First, as expected, the shape of the growing mounds is rounded, as can be seen in Fig. 1(b). No long range smoothening mechanism favors the orientation of edges along the lattice vectors. Furthermore, the coarsening is driven mainly by fluctuations of the particle deposition and hence is much slower than the SED assisted process.

The observed exponent $z = 4.0$ coincides with the results of [13,14] obtained in continuum models without explicit SED mechanism. It is also interesting to note the agreement with a recent hypothesis of Tang, Šmilauer, and Vvedensky [34] that purely noise assisted coarsening should be characterized by $z = 2+d$ for growth on d-dimensional substrates in the presence of slope selection.

A particular difficulty should be noted with respect to the observation of the saturation behavior. In the late stages of growth only a few mounds are present. The merging of these last structures seems to depend crucially on the actual configuration, i.e. their relative position. This was already observed for the case of fast SED and can lead to a cascade like increase of w close to saturation with *metastable* configurations persisting over relatively large times.

3.3 Ehrlich–Schwoebel effect for SED

Next we discuss the influence of an energy barrier for SED around corner sites. In analogy to the Schwoebel-effect one can argue that moves around the corners of terrace edges should be suppressed. Indeed, there is experimental and theoretical evidence for such an effect, see for instance [18,35] and references therein.

As a clear limiting case we consider the presence of an infinite barrier at terrace corners, referred to as restricted SED in the following. Hence, the movement of particles through SED (process C) is always limited to a straight portion of the edge. If no additional binding partner can be reached within a distance $\pm\ell_{\mathrm{SED}}$, the particle remains immobile, hereby increasing the density of kinks. We find that the favored shape of terraces corresponds to squares which are tilted with respect to the lattice vectors by 45^o (*diamonds*) and offer the maximum possible number of kink sites. In Fig. 1(d) a snapshot of an accordingly growing surface is shown. Pyramids with diamond shaped bases and a stable slope have emerged in contrast to the rounded *cones* of Fig. 1(b). This observation becomes obvious if one considers an initially square shaped pyramid base. A particle diffusing at a straight step edge will be reflected by the corners which leads to an effective flux towards the center of the step edge. Therefore, extended terraces aligned with the lattice axis are unstable.

Despite the obvious differences in surface morphology, the scaling behavior of the systems with and without restriction of SED is approximately the same for small ℓ_{SED}. In Fig. 2 we compare the evolution of the corresponding (reduced) width w_{red}; the data rescale under the assumption $z = 4.0$ and $\beta = 0.25$ in both cases. Again, the coarsening is driven mainly by deposition noise and hence it is relatively slow compared to the model with fast, unrestricted SED.

Note that the actual value of ℓ_{SED} should be largely irrelevant in restricted

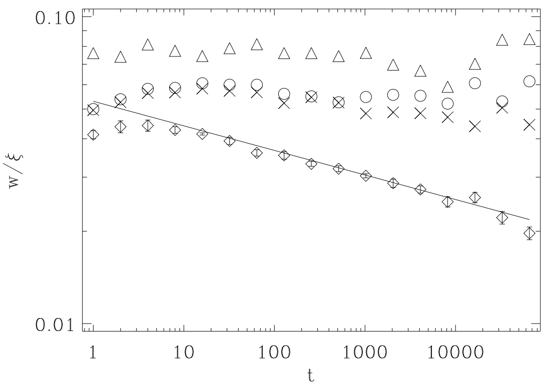

Fig. 3. Temporal evolution of the surface width w divided by the typical distance ξ on the surface. The symbol ($\triangle$) represents the model with restricted SED, here $\ell_{\rm SED} = 2$, system size $L = 512$, 11 independent simulation runs (the last 6 points are only from 3 runs). Unrestricted SED: ($\times$) $\ell_{\rm SED} = 1$, $L = 256$, 70 runs; ($\bigcirc$) $\ell_{\rm SED} = 2$, $L = 256$, 70 runs. In the absence of SED ($\diamond$, $L = 512$, 44 runs, the last two points are from 21 runs, standard error bars are shown) w/ξ follows a power law, the solid line corresponds to $(\alpha - 1)/z = -0.08$.

SED. After a ($\ell_{\rm SED}$-dependent) transient, diamond shaped terraces should always emerge, along which particles can diffuse only a few steps in any case. We will study the validity of this simplifying argument in a forthcoming project.

3.4 Growth without SED

Fig. 1(c) shows a snapshot of a surface grown in the absence of SED. As for unrestricted SED with small but non-zero $\ell_{\rm SED}$, the emerging structures are rounded. However, the total lack of smoothening SED leads to very rugged terrace edges. As we will see below, the system does not select a stable constant slope of the mounds. Apparently, the absence of well defined extended terraces makes the effective cancellation of downhill and uphill currents impossible. Clearly, our simple argument yielding a mean terrace width $2R_i$ is not applicable here.

The above mentioned difficulty in observing saturation is particularly pronounced for $\ell_{\rm SED} = 0$. *Metastable* surface configurations with very few remaining mounds persist already in fairly small systems and reduce the usefulness of scaling plots analogous to Fig. 2. Hence, we have determined first β from the simulation of large systems without attempting to achieve saturation. We find a value of $\beta \approx 0.20$, which clearly demonstrates the importance of step edge diffusion: the mere presence of SED with $\ell_{\rm SED} = 1$ already changes the growth exponent to $\beta = 0.25$ as shown above.

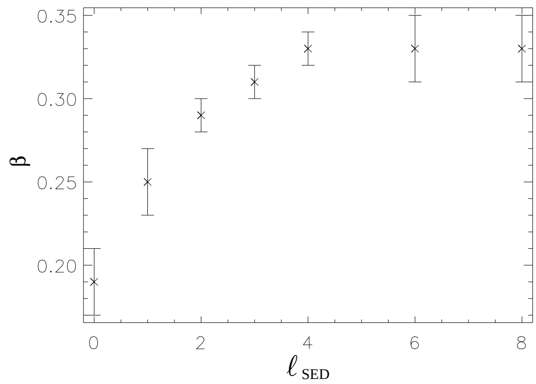

Fig. 4. Continuous rise of β from 0.25 to 0.33 for small values of ℓ_{SED}. $2^{15} = 32768$ ML were deposited on a system of size 256×256. The diffusion length was set to $\ell_D = 15$. The $w(t)$ curve was averaged over at least 7 independent simulation runs. Error bars are standard deviations of a nonlinear fit to the power law in the range 4 ML – 32768 ML.

In order to determine a second scaling exponent we have analysed the Fourier transform of the growing surfaces. For all simulations at all times a pronounced maximum in the Fourier spectrum is observed, which can be used to extract a typical distance ξ on the surface. The scaling behavior of ξ, which corresponds to the typical mound size, is given by the dynamic exponent: $\xi \propto t^{1/z}$. Numerically we obtain a value of $z < 4$ which together with $\beta \approx 0.20$ implies $\alpha < 1$ in the absence of SED. In Fig. 3 the quantity w/ξ is plotted which scales like $w/\xi \propto t^{(\alpha-1)/z}$ with time. The upper three curves correspond to the system with slow SED ($\ell_{\mathrm{SED}} = 1, 2$) and the model with restricted SED (here $\ell_{\mathrm{SED}} = 2$). In all these cases w/ξ is roughly constant for large times as expected from the observed slope selection with $\alpha = 1$. However, in the absence of SED the quantity follows a power with an exponent $(\alpha - 1)/z$ in the vicinity of -0.08, i.e. $\alpha \approx 0.7$, which clearly indicates the absence of slope selection. Note that these findings do support the hypothesis of [34] that for general noise assisted coarsening $2\beta + d/z = 1$, which is fullfilled in this case with $\beta = 0.2$, $\alpha = 0.7$, and $z = \alpha/\beta = 3.5$.

The consequences of $\alpha < 1$ for the persistence of mounds in the limit $t \to \infty$ remains unclear at this stage, even though we have realized $t \approx 10^5$. Our main point here, however, is to demonstrate the absence of slope selection.

4 Crossover behavior

We now turn to the investigation of intermediate values of ℓ_{SED}. One could speculate that for any finite $\ell_{\mathrm{SED}} \ll L$ the limiting case of vanishing SED (*i.e.* $\ell_{\mathrm{SED}} = 1$) should be reached as $t \to \infty$. However, fig. 4 supports a value of

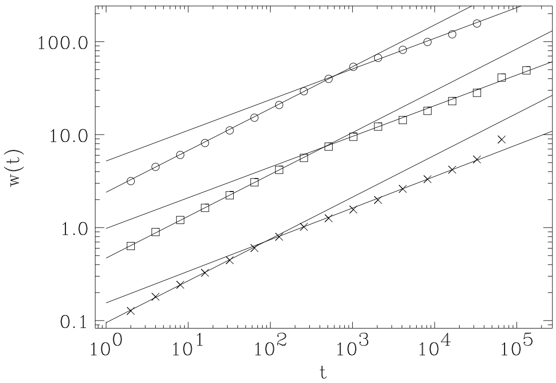

Fig. 5. Crossover from fast SED to vanishing SED for large, intermediate values of ℓ_{SED}. The straight lines correspond to $w \propto t^{0.33}$ and $w \propto t^{0.45}$. The simulations were carried out on a 1024×1024 lattice with $\ell_D = 15$. The different symbols correspond to $\ell_{\mathrm{SED}} = 50$ ($\times$), 80 ($\square$), and 100 ($\bigcirc$).

$\beta = 0.33$ for all $\ell_{\mathrm{SED}} \geq 4$. In these simulations we deposited $2^{15} = 32768$ ML but no indication for a crossover to $\beta = 0.25$ was found. We believe that the mean terrace width (here: $2R_i = 2$) determines the value of ℓ_{SED} at which $\beta = 0.33$ is reached. Clearly, the lateral fluctuations of the step edges are related to SED. Higher values of ℓ_{SED} reduce the lateral fluctuations. On the other hand, if the fluctuations become so large that they sense the terrace border, the growth behavior should change. We will investigate the validity of this speculative argument in a forthcoming project. We should be able to confirm our hypothesis by increasing R_i or reducing the Ehrlich–Schwoebel barrier, both yielding an increased mean terrace width.

Both exponents, $\beta = 0.33$ and $\beta = 0.45$, are relevant values of the growth exponent as can be most easily identified in fig. 5. There, we investigate the crossover for models with relatively high values of ℓ_{SED} compared to the terrace width. Therefore, the simulation of large systems was necessary and we chose $L = 1024$. The time needed for the simulations was several weeks on a HP9000/715. As a consequence, the data are calculated from a single simulation run. Nevertheless, our results support the following picture: Initially, the structures are small compared to ℓ_{SED}. Hence, we obtain the scaling behavior of significant SED with $\beta = 0.45$. When the typical size ξ of the structures becomes of the order of ℓ_{SED} the SED is no longer significant and the growth exponent drops down to $\beta = 0.33$. The time $t_\times$ when the crossover occurs therefore depends on ℓ_{SED} like $t_\times \propto \ell_{\mathrm{SED}}^z$ (since $\ell_{\mathrm{SED}} \approx \xi \propto t_\times^{1/z}$) which is in accordance with the data presented in fig. 5. It should be possible to observe the scaling with $\beta = 0.45$ in experiments if SED is sufficiently fast in the investigated material. One might speculate that this is the case in [8] where a value $\beta \approx 0.5$ was observed in the growth of Cu(001) at $T = 200$K.

5 Conclusions

In summary, our findings show qualitatively and quantitatively that the features of epitaxial growth processes depend crucially on the presence of step edge diffusion and its detailed properties. This offers a possible explanation for the large variety of surface morphologies and scaling properties observed in experiment and simulation. In particular, since the SED–constant δ is controlled by temperature, low (high) values of $\ell_{\rm SED}$ should be realized at low (high) T. Hence, the observation of $\beta \approx 0.25$ at low T and $\beta \approx 0.5$ at high T in [8] could be due to the influence of SED. In this case, the change of the scaling exponent should be reflected by a change of the morphology.

We find slope selection (hence $\alpha = 1$) for all simulations with non–zero step edge diffusion. With $\ell_{\rm SED} = 1$ we find $\beta = 0.25$ resp. $z = 4.0$. With increasing step edge diffusion the growth exponent rises continuously to $\beta = 0.33$. For intermediate values of $2R_i \ll \ell_{\rm SED} \ll L$ we observe initially the growth exponent of SED–assisted coarsening $\beta = 0.45$. When the size of the structures becomes comparable to the step edge diffusion length, β changes to $\beta = 0.33$. Preliminary results for finite values of the Ehrlich–Schwoebel barrier with our model (not discussed here) as well as kinetic Monte–Carlo simulations [36] indicate that the coarsening behavior is not affected by the actual value of the Ehrlich–Schwoebel barrier. Clearly, small Ehrlich–Schwoebel barriers increase the time for mound formation and the asymptotic scaling exponents will be more difficult to obtain via computer simulations.

Assuming mass currents driven by surface energetics, the authors of [34] obtain also $z = 4$ in $d = 2$ dimensions. Even though SED might be interpreted as such a mass current, we obtain a significantly different result for $\ell_{\rm SED} \geq 4$. The reason for this discrepancy remains unclear at present.

Forthcoming investigations will address more realistic models, for instance with finite energy barriers at downward steps for which we expect more insight into the increase of the growth exponent from $\beta = 0.25$ to $\beta = 0.33$. For additional barriers in step edge diffusion at corners we expect a non-trivial crossover behavior, as well.

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