Particle currents and the distribution of terrace sizes in unstable epitaxial growth

M. Biehl\textsuperscript{1,2}, M. Ahr\textsuperscript{3}, M. Kinne\textsuperscript{3}, W. Kinzel\textsuperscript{1}, and S. Schinzler\textsuperscript{1,2}

\textsuperscript{1}Institut für Theoretische Physik und Astrophysik, \textsuperscript{2}Sonderforschungsbereich 410
Julius-Maximilians-Universität Würzburg
Am Hubland, 97074 Würzburg, Germany
\textsuperscript{3}Lehrstuhl für Physikalische Chemie II
Friedrich-Alexander-Universität Erlangen-Nürnberg
Egerlandstr. 3, 91058 Erlangen, Germany

A solid–on–solid model of epitaxial growth in 1 + 1 dimensions is investigated in which slope dependent upward and downward particle currents compete on the surface. The microscopic mechanisms which give rise to these currents are the smoothening incorporation of particles upon deposition and an Ehrlich–Schwoebel barrier which hinders inter–layer transport at step edges. We calculate the distribution of terrace sizes and the resulting currents on a stepped surface with a given inclination angle. The cancellation of the competing effects leads to the selection of a stable magic slope. Simulation results are in very good agreement with the theoretical findings.

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Epitaxial growth has become a standard method for the production of high–quality crystals and films, needed for e.g. semiconductor devices. An overview of experimental techniques can be found in [1], for instance. Significant effort has been devoted to a theoretical understanding of the many morphologies and scaling behaviors that can be observed in epitaxial growth, see e.g. [2] for a review of theoretical approaches. Here we address the frequently observed phenomenon of mounds in unstable growth which has attracted considerable interest, see e.g. [3–8]. Specifically, we consider situations in which competing smoothening and steepening effects control the surface morphology and lead to the selection of a stable slope in the system.

We discuss potential microscopic mechanisms which result in the emergence of mounds and slope selection in the frame of a discrete (1 + 1)–dimensional model. The net particle currents as well as the distribution of terrace sizes on a surface of a given slope can be worked out and this allows then to evaluate the magic slope as well as the complete statistical properties of the emerging surface. The analysis complements previous theoretical investigations which address the mean terrace size only or neglect fluctuations explicitly in the spirit of Burton Cabrera Frank (BCF) theory [9]–[10]. We demonstrate that the full distribution of terrace sizes carries relevant information that should be taken into account. Our results suggest, for example, that it should be possible to identify relevant microscopic mechanisms from experimental data.

Our (1 + 1)–dimensional model obeys the solid–on–solid (SOS) restriction, i.e. the surface can be described by an integer array of height variables $h_k$. Single particles are deposited at randomly chosen sites $k \in \{1, 2, \ldots, L\}$. Upon arrival, an incorporation process moves a particle to the lowest available site within a neighborhood of $\pm R$ lattice constants, i.e. the site $j$ with $h_j = \min \{h_{k-R}, \ldots, h_k, \ldots, h_{k+R}\}$. In case of a tie, the site closest to the deposition is chosen. Only if this is still ambiguous, an additional random selection is performed. Such a smoothening mechanism, in absence of further effects, is commonly associated with the Edwards–Wilkinson universality class of growth [2].

The parameter $R = O(1)$ (in lattice constants) is termed the incorporation radius and sets the typical length scale of the process. Various interpretations of incorporation have been considered, including downward funneling on non–trivial lattices and knock–out–processes due to the momentum of incoming particles, see e.g. [11,12].

A particle which is, after deposition and possibly incorporation, not yet bound to a lateral neighbor diffuses on the surface by performing a random walk (RW) until it reaches an additional binding partner and becomes immobile or until it collides with another moving adatom and forms an island nucleus. In a density of diffusing particles, this nucleation process would result in a typical collision free path $l_D$. In the case of irreversible agglomeration on a flat substrate, and if islands of two or more adatoms are considered immobile, it has been shown that $l_D \propto (d/f)^{1/4}$ in (1 + 1) dimensions [13,14]. Here, $d$ is the diffusion constant and $f$ the incoming flux, with all lengths in dimensionless lattice constants.

In [15] nucleation is represented in an effective single particle picture and $l_D$ fixes the typical distance of island nuclei in the first layers on a flat substrate. After the formation of mounds, terrace sizes are much smaller than $l_D$, typically. Hence we will completely neglect nucleation on the stepped surface which is justified for small incoming flux or fast diffusion, respectively.

Here, the RW ends whenever the particle sticks irreversibly to a lateral neighbor, i.e. when it reaches a terrace step. Attachment can occur from below and above, in principle, but this symmetry is broken due to the so–called Ehrlich–Schwoebel (ES) effect [2]. An additional energy barrier $E_{es}$ at step edges hinders downward moves of diffusing adatoms as this would involve loosely bound intermediate positions. Our model takes
we have to distinguish the following cases:

To site  

a particle deposited on a site  

RW initiated at site  

obviously satisfies  

diffusion:  

for any  

because particles will stick to upper terraces preferably. Structures have built up the dent and their cancellation gives rise to the formation of mounds with a well-defined inclination angle. Once the structures have built up the magic slope, a coarsening process begins which decreases the number of mounds, see Fig. 3 for details.

We will first work out the distribution of terrace sizes which emerges in our model on surfaces with a given, fixed inclination. Further, we will calculate the mean displacement of a particle’s final position from its deposition site. The latter corresponds to the net particle displacement of a particle’s final position from its deposition site. The former is the central terrace: incorporation from exactly  

central terrace: incorporation from exactly  

with  

the incorporation process places the particle at its final position at site  

attaching to the lower terrace end.

(b)  

the particle performs a random walk until it reaches one of the trap sites  

moves to one of the neighboring positions  

with equal probability  

The asymmetry of the RW (b) is due to the ES-barrier present for jumps from site  

Denote with  

the probability for a downward move to site  

With probability  

the move is rejected and the particle remains at  

for the next time step, whereas with probability  

it jumps to  

A straightforward exercise yields the probability for a RW initiated at site  

by downward diffusion:  

which obviously satisfies  

For similar problems of this type see for instance Fig. 3.

Hence, the total probability  

deposition event to occur on terrace  

with subsequent downward diffusion is  

if  

else. The second case accounts for the fact that any particle directly deposited onto a terrace of width  

will be incorporated without performing diffusion.

The quantity  

which can be interpreted as the effective number of deposition sites which contribute to downward diffusion from terrace  

The prefactor  

is simply the constant probability for deposition on any of the sites in the system.

Now we can work out the probability  

for the central terrace  

to be shortened by the next deposition event. For  

one finds

The first contribution,  

represents deposition on any of the  

sites left of terrace  

Note that the outcome of the subsequent incorporation process is completely independent of the surface configuration, in particular of the left neighbor terrace width  

The second term [ . . . ] accounts for deposition on  

with final attachment to the upper terrace. Finally  

is the probability for the shortening of  

c through diffusion from terrace  

If  

only two processes can shorten the central terrace: incorporation from exactly  

sites left of  

and downward diffusion from terrace  

One obtains

Since  

can only increase at the cost of shortening  

terms at the same time, one obtains immediately the result

We proceed by assuming that in a population of terraces the distribution of their sizes factorizes, i.e. that a single, identical  

is sufficient to describe their statistics. For the limiting case of an infinite ES barrier  

the evolution of a terrace is independent of the entire configuration left of it, and the above property can be shown to hold true. In general, neighbor terraces clearly interact. Nevertheless, our simulations show that the assumption of identically distributed, independent terrace sizes yields excellent approximations, at the very least. Figure 3 shows, for instance, that the correlation coefficient of neighboring terrace sizes in a system with  

and  

vanishes within error bars.

The analysis simplifies significantly if terrace sizes are considered to be uncorrelated. The above expressions were obtained for a given triple of terraces  

By averaging  

over  

and  

respectively, one obtains the mean probabilities  

for  

if  

else: 

if  

else.

where the r.h.s. involve only the width of the considered terrace itself, the frequency  

vanishing ter-
race sizes and the mean values $\langle l \rangle = \sum_{j=0}^{\infty} j \rho(j)$ and $\langle \Delta \rangle = \sum_{j=0}^{\infty} \Delta(j) \rho(j)$, see Eq. (3).

The evolution of terraces according to (3) produces a stationary distribution $\rho(l)$, if $p(l+1) w(l+1 \to l) = p(l) w(l \to l+1)$, hence

$$p(l+1) = p(l) \frac{\langle l \rangle - \langle \Delta \rangle + [1 - p(0)] \Delta(l)}{l+1 - \Delta(l+1) + \langle \Delta \rangle} \tag{6}$$

for $l \geq 0$. This relation is implicit, since all $p(l)$ have to be known for the evaluation of the averages on the r.h.s. In the particular case of an infinite ES barrier, Eq. (3) reads $p(l+1) = p(l) \langle l \rangle / (l+1)$ which is satisfied by the Poissonian $\rho(l) = \lambda^l e^{-\lambda} / l!$ with mean $\langle l \rangle = \lambda$.

In order to obtain the stationary $p(l)$ for $p_{es} > 0$ on a surface with a given mean terrace size $\lambda$, we replace $\langle j \rangle$ with $\lambda$ and $\langle \Delta \rangle$ with an adjustable parameter $D$ in Eq. (3). The quantities $p(0)$ and $D$ are determined such that $\sum_{l=0}^{\infty} p(l) = 1$ is satisfied and $\langle l \rangle = \lambda$ is reproduced self-consistently. Note then, that, by construction, $p(l)$ guarantees $\langle \Delta \rangle = D$ as well. In the numerical treatment, sums are truncated at a value $l_{\text{max}}$, with the resulting $p(l_{\text{max}})$ small enough to justify the truncation a posteriori.

A particle deposited at, say, lattice site $i$ will become immobile at a final position $j \neq i$ after incorporation and diffusion, in general. The expected displacement $\delta = \langle j - i \rangle$ depends on the distribution of terrace sizes in the system. Taking into account all possible displacement processes and their corresponding probabilities one finds

$$\delta = \frac{1}{2} R(R+1) - \frac{1}{2} \sum_{l=0}^{R-1} p(l) [(R-l)(R-l+1)] + \sum_{l=R+1}^{\infty} p(l) \left( -l(l-R-\Delta(l)) + \frac{1}{2}l(l+1) - \frac{1}{2}R(R+1) \right), \tag{7}$$

where the $p(l)$ obtained from (3) have to be inserted for a given $\langle l \rangle = \lambda$. Here, the first line corresponds to the expected (positive, downward) effect of incorporation and the second represents the total (negative, upward) contribution of diffusion. In the limiting case $p_{es} = 0$, Eq. (3) reduces to $\delta = R \langle l \rangle - \langle l \rangle^2 / 2$, exploiting the fact that $\Delta(l) = 0$ in this case and $\langle l^2 \rangle - \langle l \rangle^2 = \langle l \rangle$ for the Poisson distribution.

Figure 3 shows the result of Monte Carlo simulations of the growth process in a system of size $L = 1000$ for the model with $R = 2$ and different values of $p_{es}$, where boundary conditions were used to fix $\langle l \rangle = \lambda$. We have displayed the particle current $\delta / \lambda$ on the surface as a function of $\lambda$. Note that $\delta$ in Eq. (3) was obtained for the normalization $\sum_{l=0}^{\infty} p(l) = 1$ which corresponds to a fixed number of terraces. In systems with a fixed number $L$ of lattice sites, an additional factor has to be introduced as the number of terraces grows like $1 / \lambda$.

For very steep surfaces, $\lambda \to 0$, the displacement approaches the limiting value $R$, representing the fact that every deposited particle is shifted by $R$ lattice constant in the incorporation and then comes to rest. In the limit of vanishing slope, our model yields a diverging negative “upward” current. This is an artifact of completely neglecting nucleation, which inevitably becomes important as $\lambda \to \infty$ and imposes a maximal displacement on the order of $l_0$.

On mounded surfaces, bottom and top terraces limit the extension of inclined flanks. Any slope that results in a net uphill current according to Eq. (3) will steepen this portion of the surface and vice versa. Accordingly, a mean terrace width $\lambda_o$ will be stabilized which corresponds to the zero of $\delta(\lambda)$. In the presence of an infinite ES–barrier we find the exact relation $\lambda_o = 2R$. A naive and not quite correct argument was used in (3) to obtain the same result. It is instructive to check that the magic slope cannot be obtained from the condition that the mean displacement vanishes on a particular terrace of size $\lambda$. This would correspond to setting $p(l) = \delta_{l,\lambda}$ in Eq. (3) and gives results analogous to the BCF–like treatment in (3) which does not account for fluctuating terrace sizes. For $p_{es} = 0$ one obtains, e.g., $\lambda = 2R+1 = \lambda_o + 1$.

Fig. 4 shows the frequency of terrace sizes as observed in two different systems which both stabilize the mean $\lambda_o = 6$. In one we have set $p_{es} = 0$ and $R = 3$, the second example corresponds to $R = 2$ and $p_{es} = 0.258$. Computer simulations show excellent agreement. Systems with a very pronounced ES–effect produce a narrow distribution with a very low frequency $p(0)$ of step bunching, i.e. zero terrace sizes. As a limiting case one finds $p(0) = e^{-\lambda_o} = e^{-2R}$ for infinite ES–barrier ($p_{es} = 0$).
On the contrary, step bunching is observed with a much larger frequency in cases with a weaker ES–effect where the distribution \( p(l) \) is much broader. Fig. 3 displays \( \lambda_0 \) and the variance \( \sigma^2 \) of the terrace size distribution as functions of \( p_{es} \). Note that \( \sigma^2 \) grows drastically with increasing \( p_{es} \), indicating significant deviations from the Poissonian for infinite ES–effect.

The analysis of experimental data is frequently based on the simple assumption of random, non–interacting terrace sizes on vicinal surfaces. This leads to the geometric distribution \( p(l) = (1 - 1/\lambda)^{l-1}/\lambda \) with \( \langle l \rangle = \lambda \), see e.g. [1] for a discussion. Note that \( p(l) \) differs significantly from the type of statistics that we find in our model, cf. Fig. 3. In particular, step bunching is much more frequent in this simple picture: \( p_0(0) = (\lambda-1)^{-1} \).

In summary we have presented a microscopic model of unstable epitaxial growth in which it is possible to derive the net particle currents on surfaces of a given inclination. For the first time it is possible to work out the full distribution of terrace sizes in such a system. Further, we were able to calculate the stable mean terrace size and the corresponding statistical properties of the surface. We have restricted ourselves to the analysis of (1+1)–dimensional growth in this work. However, our results should carry over to a more realistic (2+1)–dimensional picture to a large extent, whenever terrace edges do not meander significantly.

Our findings allow for a qualitative interpretation of experimental results in systems which display slope selection: frequent step bunching and a broad distribution hint at a relatively weak ES–barrier. Narrow distributions with little or no step bunching indicate that a significant ES–effect is present but is compensated for by smoothening effects like downhill funneling.

Extensions of this work will concern desorption and its influence on the growth process. Preliminary results indicate that a significant desorption rate can trigger a transition from slope selection to rough growth and we expect non–trivial effects in the statistics of terrace sizes.

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