Terrace Sizes and Particle Currents in Epitaxial Growth

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We investigate a solid–on–solid model of epitaxial crystal growth in $1+1$ dimensions. The emerging distribution of terrace sizes and the resulting currents on surfaces with a given inclination angle are calculated analytically. Furthermore, we evaluate the stable *magic slope* which is selected due to the compensation of the competing effects. Results of Monte Carlo Simulations are in very good agreement with the theoretical predictions.

Key Words: crystal growth, molecular beam epitaxy, non–equilibrium, unstable growth, slope selection

1. Introduction

In recent years, epitaxial growth has become a standard method for the production of high–quality crystals and films. A review of experimental methods can be found in (1), for instance. A recent overview of theoretical approaches towards an understanding of the many observed morphologies and scaling behaviors is given in (2). Here we address the frequently encountered phenomenon of mound formation in unstable growth which has attracted considerable interest (3–10). More 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, see for instance (3–5,9,10).

Potential microscopic mechanisms are discussed which result in the formation of mounds and slope selection in the framework of a discrete $(1+1)$–dimensional model. Particle currents on the surface of a given slope as well as the emerging distribution of terrace sizes are worked out and this allows then to evaluate the magic slope as well as the statistical details of the emerging surface. Our analysis complements previous theoretical investigations which are limited to the mean terrace size only or neglect fluctuations explicitly (11) in the spirit of Burton Cabrera Frank (BCF) theory (2). A similar discrete model which includes an additional step–adatom attraction has been considered in (9). There, however, the analysis is also restricted to non–fluctuating terrace sizes.

We will show that the full distribution of terrace sizes determines the actual particle currents and carries relevant information about the growth mechanisms. According to our results, for example, it should be possible to identify the most relevant microscopic mechanisms from experimental data.

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2. The Model

Our $(1+1)$–dimensional model obeys the solid–on–solid (SOS) restriction, i.e. the surface is described by an integer array of height variables h_k, defects or overhangs are excluded. 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 $R = \mathcal{O}(1)$ (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. (9,12,13).

After deposition and possibly incorporation, a particle might not yet be bound to a lateral neighbor. It then 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. Such nucleation processes would result in a typical collision free path l_{D} in a density of diffusing particles. In the case of irreversible aggregation on a flat substrate, and if islands of two or more adatoms are immobile, it has been shown that $l_{\rm D} \propto (d/f)^{1/4}$ in $(1+1)$ dimensions [2,14]. Here, d is the diffusion constant and f the incoming flux, with all lengths measured in dimensionless lattice constants.

In [10], we represented the influence of nucleation events in an effective single particle picture and $l_{\rm D}$ fixes merely the typical distance of island nuclei in the first layers on a flat substrate. Throughout the following we will assume that, after the formation of mounds, the typical terrace sizes l on the stepped surface are much smaller than the diffusion length. Hence we can completely neglect nucleation on the stepped surface which is justified for small incoming flux or fast diffusion, respectively.

Here, the random walk comes to an end whenever the particle sticks irreversibly to an in–plane neighbor, i.e. when it reaches a terrace step. Attachment can occur from below and above, in principle, but the symmetry is broken due to the so–called Ehrlich–Schwoebel effect [2] (ES): An additional energy barrier E_{es} at step edges hinders downward moves of diffusing adatoms as this would involve weakly bound intermediate positions. Our model takes the ES effect into account by assigning a probability $p_{es} \propto \exp\,[\,-E_{es}/(kT)\,]$ to downward moves.

The Ehrlich–Schwoebel effect results in an uphill current of diffusing atoms since particles will stick to upper terraces preferably for any $p_{es} < 1$. However, the assumed incorporation process clearly causes a competing downhill current. Both effects are slope dependent and their potential cancellation gives rise to the formation of mounds with a well–defined inclination angle. This effect has been termed slope selection in the literature [3–5,9,10].

3. Analysis and Numerical Investigation

3.1 Distribution of terrace sizes

In a first step we will work out the distribution of terrace sizes which emerges in our model on surfaces with a given, fixed inclination. Next, we will calculate the mean displacement of a particle's final position from its deposition site. The latter corresponds to the net particle current on the growing surface and its zero as a function of the inclination angle determines the stable slope.

Let us consider a triple of terraces on an inclined surface with a central terrace c of width l_c (lattice sites $j = 1, 2, \dots l_c$) and its neighbors ℓ to the left (r to the right) with size l_ℓ (l_r). We include formally the case $l_c = 0$, corresponding to a height difference of 2 lattice sites between terraces ℓ and r Without loss of generality, we will assume that the surface height decreases to the right. For a particle deposited on a site k of terrace c with $l_c > R$ we have to distinguish the following cases:

(a) $k > l_c - R$: the incorporation process places the particle at its final position at site $l_c + 1$, attaching to the lower terrace end.

(b) $1 \le k \le l_c - R$: the particle performs a random walk until it reaches one of the *trap sites* $j = 1$ or $j = l_c + 1$ where it comes to rest. This includes deposition at site $k = 1$ without subsequent diffusion.

A diffusing particle located at a site j with $2 \le j \le l_c - 1$ moves to one of the neighboring positions $j \pm 1$ with equal probability $1/2$. The asymmetry of the RW (b) is due to the ES–barrier present for jumps from site $j = l_c$. We denote with $p_{es}/2$ the probability for a downward move to site $l_c + 1$. With probability $(1 - p_{es})/2$ the move is rejected and the particle remains at $j = l_c$ for the next time step, whereas with probability $1/2$ it jumps to $l_c - 1$.

A straightforward exercise yields the probability for a RW initiated at site k to end in $j = l_c + 1$ by downward diffusion: $q(k, l_c) = \,(\,(k-1)\,p_{es}\,)/\,(\,1+(l_c-1)p_{es}\,)$ which obviously satisfies $q(1, l_c) = 0$ for all p_{es}. For similar problems of this type see for instance [9,15].

Hence, the total probability $d(l_c)$ for a deposition event to occur on terrace c with subsequent downward diffusion is $d(l_c) = \Delta(l_c)/L$ with

$$\Delta(l_c) = \sum_{k=1}^{l_c - R} q(k, l_c) = \frac{(\,l_c - R\,)\,(\,l_c - R - 1\,)p_{es}}{2 + 2\,(\,l_c - 1\,)p_{es}} \qquad (1)$$

if $l_c > R$ and $\Delta(l_c) = 0$ else. The second case accounts for the fact that any particle directly deposited onto a terrace of width $l_c \le R$ will be incorporated without performing diffusion.

The quantity $\Delta(l_c)$ can be interpreted as the effective number of deposition sites which contribute to downward diffusion from terrace c. The prefactor $1/L$ of $d(l_c)$ is simply the constant probability for deposition on any of the sites in the system.

Now we can work out the probability $\widetilde{w}(l_c \to l_c - 1)$ for the central terrace c to be shortened by the next deposition event. For $l_c > R$ one finds

$$\begin{aligned} \widetilde{w}(l_c \to l_c - 1) &= \,(\,R + [l_c - R - \Delta(l_c)]\, + \Delta(l_\ell)\,)/L \\ &= \,(\,l_c - \Delta(l_c) + \Delta(l_\ell)\,)/L. \end{aligned} \qquad (2)$$

The first contribution, R, represents deposition on any of the R sites left of terrace c. Note that the outcome of the subsequent incorporation process is completely independent of the surface configuration, in particular of the left neighbor terrace width l_ℓ. The second term $[\dots]$ accounts for deposition on c with final attachment to the upper terrace. Finally $\Delta(l_\ell)/L$ is the probability for the shortening of c through diffusion from terrace ℓ.

If $0 < l_c < R$, only two processes can shorten the central terrace: incorporation from exactly l_c sites left of c and downward diffusion from terrace ℓ. One obtains $\widetilde{w}(l_c \to l_c - 1) = (l_c + \Delta(l_\ell))/L$ and we finally observe that Eq. (2) is valid for any $l_c \ge 1$ since $\Delta(l_c) = 0$ for all $l_c \le R$. Obviously, $\widetilde{w}(l_c \to l_c - 1) = 0$ if $l_c = 0$ already.

Since l_c can only increase at the cost of shortening l_r at the same time, one obtains immediately the result

$$\widetilde{w}(l_c \to l_c+1) = \begin{cases} \frac{1}{L}\,(\,l_r - \Delta(l_r) + \Delta(l_c)\,) & \text{if } l_r > 0 \\ 0 & \text{if } l_r = 0. \end{cases} \tag{3}$$

We proceed by assuming that in a *population* of terraces the distribution of their sizes factorizes, i.e. that a single, identical $p(l)$ is sufficient to describe their statistics. For the limiting case of an infinite ES barrier ($p_{es} = 0$) 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 1 shows, for instance, that the correlation coefficient of neighboring terrace sizes in a system with $L = 1000$ and $p_{es} = 0.2$ vanishes within error bars.

The analysis simplifies significantly if terrace sizes are considered to be uncorrelated. The above expressions (2,3) were obtained for a given triple of terraces $\{\,\ell, c, r\,\}$. By averaging $\widetilde{w}(l_c \to l_c \pm 1)$ over $p(l_\ell)$ and $p(l_r)$, respectively, one obtains the mean probabilities $w(l_c \to l_c \pm 1)$:

$$w(l_c \to l_c-1) = \begin{cases} \frac{1}{L}\,(\,l_c - \Delta(l_c) + \langle\Delta\rangle\,) & \text{for } \; l_c > 0 \\ 0 & \text{for } \; l_c = 0 \end{cases} \tag{4}$$

$$w(l_c \to l_c+1) = \,(\,\langle l\rangle - \langle\Delta\rangle + [1-p(0)]\Delta(l_c)\,)/L \tag{5}$$

where the r.h.s. involve only the width of the considered terrace itself, the frequency $p(0)$ of vanishing terrace sizes and the mean values $\langle l\rangle = \sum_{j=0}^{\infty} j p(j)$ and $\langle\Delta\rangle = \sum_{j=0}^{\infty} \Delta(j) p(j)$, see Eq. (1).

The evolution of terraces according to (4,5) produces a stationary distribution $p(l)$, 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. (6) reads $p(l+1) = p(l)\langle l\rangle/(l+1)$ which is satisfied by the Poissonian $p(l) = \lambda^l\, e^{-\lambda}/l!$ 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 λ, we replace $\langle l\rangle$ with λ and $\langle\Delta\rangle$ with an adjustable parameter D in Eq. (6). The quantities $p(0)$ and D are determined such that $\sum_l p(l) = 1$ is satisfied and $\langle l\rangle = \lambda$ is reproduced self–consistently. Note that then, by construction, (6) guarantees $\langle\Delta\rangle = D$ as well. In the numerical treatment, sums are truncated at a value l_{max}, with the resulting $p(l_{\max})$ small enough to justify the truncation.

3.2 Particle Currents

Assume a particle was deposited at, for instance, lattice site i. It will finally become immobile at some final position $j \neq i$ after incorporation and diffusion, in general. The expectation value of the displacement $\delta = \langle j - i\rangle$ clearly depends on the distribution of terrace sizes in the system. Taking into account all possible displacement processes in our model and their corresponding probabilities one finds

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

where the $p(l)$ obtained from (6) 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. (7) 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 1 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 δ/λ on the surface as a function of λ. Note that δ in Eq. (7) was obtained for the normalization $\sum_l p(l) = 1$ 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 $1/\lambda$.

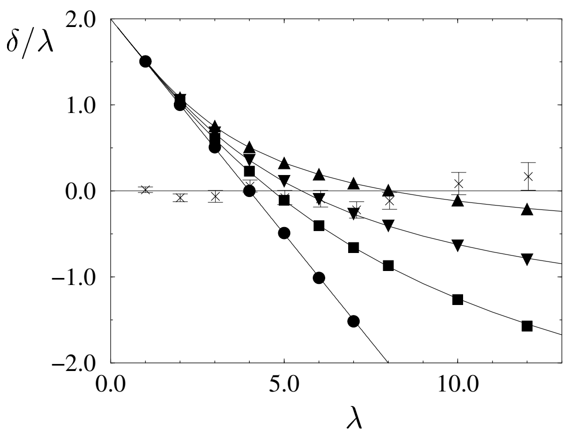

FIG. 1. Mean particle displacement δ/λ vs. $\lambda = \langle l\rangle$, cf. Eq. (7), displayed for $R = 2$ and different ES barriers. The symbols represent simulation results obtained with $L = 1000$ for $p_{es} = 0(\bullet), 0.1(\blacksquare), 0.2(\blacktriangledown), 0.4(\blacktriangle)$. All averages are over 100 independent runs and standard error bars would be smaller than the symbols, where not shown. In addition, the correlation coefficient $\rho = (\langle l_a l_b\rangle - \langle l\rangle^2)/(\langle l^2\rangle - \langle l\rangle^2)$ for neighboring terraces a and b is displayed for the case $p_{es} = 0.2$; crosses and error bars correspond to $10 \cdot \rho$.

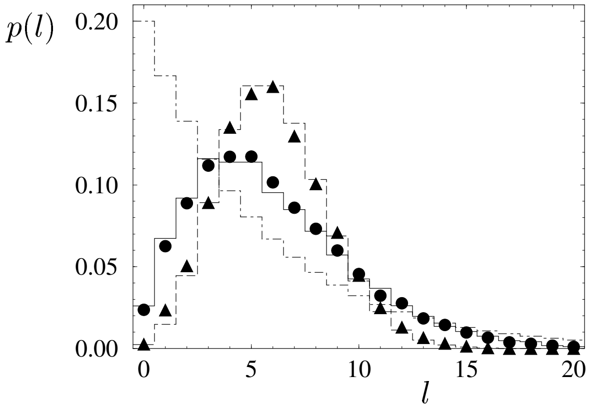

FIG. 2. Frequency $p(l)$ of terrace sizes in two cases with $\langle l\rangle = \lambda_o = 6$. The dashed line shows the theoretical prediction for $R = 3, p_{es} = 0$, triangles represent simulations (as in Fig. 1); the solid line and bullets correspond to $R = 2, p_{es} = 0.258$. The dot–dashed curve displays a geometric distribution with $\langle l\rangle = 6$ for comparison.

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 $l_{\rm D}$.

3.2 Slope selection

The extension of inclined flanks is obviously limited on mounded surfaces by bottom and top terraces. Any slope that results in a net uphill current according to Eq. (7) will steepen this portion of the surface and vice versa. Accordingly, a mean terrace width λ_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 [10] to obtain the same result. It is important to note that the correct magic slope cannot be obtained from the condition that δ vanishes on terraces of one particular size $\hat\lambda$. This would correspond to setting $p(l) = \delta_{l,\hat\lambda}$ in Eq. (7) and gives results analogous to those of [9,11] which do not account for fluctuating terrace sizes. For $p_{es} = 0$ one obtains, e.g., $\hat\lambda = 2R{+}1 = \lambda_o{+}1$.

The frequency of terrace sizes as observed in two different systems which both stabilize the mean $\lambda_o = 6$ is shown in Fig. 2. In one system we set $p_{es} = 0$ and $R = 3$, the second example corresponds to $R = 2$ and $p_{es} = 0.258$. Our computer simulations show excellent agreement in both cases. 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 λ_o and the variance σ^2 of the terrace size distribution as functions of p_{es}. Note that σ^2 grows drastically with increasing p_{es}, indicating significant deviations from the Poissonian for infinite ES–effect.

In the analysis of experimental data one frequently employs the simplifying assumption of random, non–interacting terrace sizes on vicinal surfaces. This leads to the geometric distribution $p_g(l) = (1-1/\lambda)^{l-1}/\lambda$ with $\langle l\rangle = \lambda$, see e.g. [16] for a discussion. Note that $p_g(l)$ differs significantly from the type of statistics that we find in our model, cf. Fig. 2. In particular, step bunching is much more frequent in this simple picture: $p_g(0) = (\lambda-1)^{-1}$.

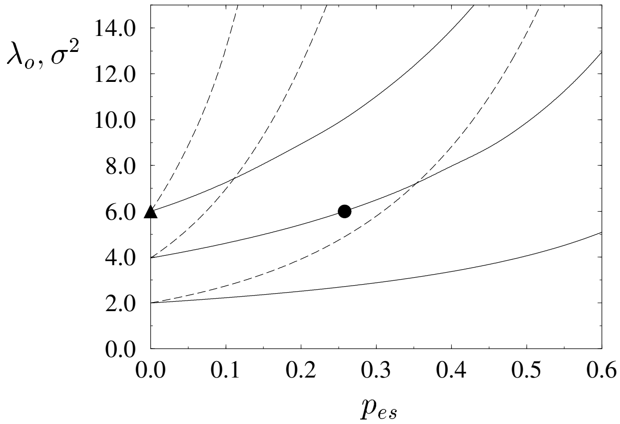

FIG. 3. The selected mean λ_o (solid lines) and $\sigma^2 = \langle l^2\rangle - \lambda_o^2$ (dashed) vs. p_{es}. Pairs of curves correspond to (from below) $R = 1, 2, 3$. Note that for $p_{es} = 0$ we find $\lambda_o = \sigma = 2R$. The symbols represent two choices of (p_{es}, R) which result in $\lambda_o = 6$, cf. Fig. 2.

4. Summary

We have presented a microscopic model of unstable epitaxial growth in which it is possible to calculate the net particle currents on surfaces with a given inclination. For the first time it is possible to work out the full distribution of terrace sizes in such a system. Furthermore, 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 $(2+1)$–dimensional growth, 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.

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