Surface roughness evolution in a solid-on-solid model of epitaxial growth

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Abstract

The paper presents results from kinetic Monte Carlo simulations of kinetic surface roughening using an important and experimentally relevant model of epitaxial growth – the solid-on-solid model with Arrhenius dynamics. A restriction on diffusing adatoms is included allowing hopping down only at steps of height one monolayer in order to prevent possible unrealistic events of jumping from arbitrarily high steps. Simulation results and precise analytic expressions representing the time evolution of surface roughness do not depend on the substrate size and clearly put forward the conclusion that for any basic set of parameters the model approaches in asymptotic limit the usual random deposition process with growth exponent $\beta = 1/2$. At high temperatures, it is preceded by a long transient regime characterized by a smooth surface and described by a power law with $\beta = 3/4$.

Keywords:
Kinetic roughening, Growth exponent, Universality class, Solid-on-solid model, Kinetic Monte Carlo simulation

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1. Introduction

Surface morphology in epitaxial growth is a characteristics which often has a strong impact on important technological properties of the films. Two

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of the main analytic tools for quantitative description of its temporal evolution are surface width and height-height correlation function. The surface width $w$ can be defined in different ways depending on the measure used to estimate surface height fluctuations. The most common definition of $w$, the root-mean-square roughness (RMSR), is implemented in many experimental measurement techniques – Scanning Tunneling Microscopy, Atomic Force Microscopy, Stylus Profilometry, etc.

RMSR is defined by the expression

$$w(L, t) = \langle (h(t) - \bar{h}(t))^2 \rangle^{1/2},$$

where $h(t)$ is the surface height at a time $t$, the overbars denote spatial average, and the angular brackets stand for suitable ensemble average over simulation or experimental trials on sample domains of size $L \times L$. Instead of mean height $\bar{h}(t)$, one can equivalently use the term surface coverage $\theta$.

One of the most important concepts concerning the global surface width $w$ is the famous Family-Vicsek dynamic scaling principle [1] which states that

$$w(L, t) = L^\alpha f(t/L^{\alpha/\beta}),$$

where the scaling function $f(x)$ satisfies $f(x) \sim x^\beta$ for $x \ll 1$ and $f(x)$ is approaching a constant for $x \gg 1$. Eq. 2 means that $w(L, t) \sim t^\beta$ at any time $t$ smaller than a crossover time $t_c \sim L^{\alpha/\beta}$ and $w(L, t) \sim L^\alpha$ for $t \gg t_c$. The growth exponent $\beta$ describes $w$ in the initial period of surface roughening while the roughness exponent $\alpha$ characterizes the roughness of the saturated surface. Alternatively, the scaling of the correlation function $G(r, t) = \langle (h(x + r, t) - h(x, t))^2 \rangle$ can be investigated, where $h(x, t)$ is the surface height at a point $x$ and time $t$ and $x, r \in \mathbb{R}^d$ (in case of $d$-dimensional substrate).

The fundamental importance of the scaling principle lies in the fact that it implies similarity of surface morphologies obtained at different length scales and equal other parameters.

In recent decades, much theoretical and experimental research has been concentrated on scaling behavior of surface width and correlation function in non-equilibrium conditions, with the main objective to predict and classify the roughness evolution in epitaxial models. Growth systems of different kinds has been examined by kinetic Monte Carlo (KMC) simulations (see e.g. [2, 3, 4, 5, 6, 7, 8, 9, 10]) and the corresponding continuum field
approaches based on partial differential equations (the models of Mullins- 
Herring (MH) [11, 12], Edwards-Wilkinson (EW) [13], Kardar-Parisi-Zhang 
(KPZ) [14], Villain-Lai-Das Sarma (VLDS) [15, 16], etc.). As a result of 
this research we now know that important from experimental point of view 
growth systems can be grouped into different universality classes according to 
their scaling parameters in the regime of non-equilibrium surface roughening.

Despite the intensive work on the subject, the general behavior of surface 
roughening which depends on large number of growth parameters is not yet 
completely clear even for commonly used epitaxial processes. The simplest 
settings of the problem for which an exact solution is known correspond to 
the random deposition model [17, 18] with $\alpha = \infty$ and $\beta = 1/2$. In this solid-
on-solid (SOS) model with a simple cubic lattice the atoms are deposited 
randomly on top of previously adsorbed ones and remain immobile at their 
adsorption sites. To the same class belong models with nonzero intralayer 
diffusion but no interlayer transport (for example, due to large step-edge 
barriers).

Much more multifarious is the behavior of the kinetic roughening in the 
presence of interlayer transport. It is usually described by power laws $w(\theta) \sim \theta^\beta$ where $\beta > 0$ or by logarithmic curves ($\beta = 0$) as in Family [19] and EW 
models for 2-dimensional (2D) substrates [20, 21, 22, 23]. Among the first 
studied systems are the homoepitaxial metal systems with body-centered 
and face-centered cubic crystal geometries. Particularly important for surface 
smoothing appears to be the process of downward funneling (DF) – the atoms 
deposited at step edges funnel down until reaching prescribed adsorption sites 
in lower layers. At $T = 0$ K, DF dynamics produces EW asymptotic behavior 
$w(\theta) \sim \log(\theta)$ [24].

In general, all groups of theoretically studied epitaxial models differ in 
their relaxation rules which in turn determine the corresponding universality 
class. While only several universality classes are commonly known due 
to their applicability in experimental work, the total number of different 
dynamic scaling scenarios seems to be infinite like the infinite number of 
possible (artificial) ways of choosing the smoothing rules in the model.

An important property of scaling theory is that the time axis can be 
divided into periods in which the surface roughness $w$ is represented by (pos-
sibly unstable) power laws $w \sim \theta^{\tilde{\beta}}$. The exponent $\tilde{\beta}$, taking its values in 
the interval $[0, 1]$, becomes usually smaller in every following period (the two 
extreme values 0 and 1 correspond to the asymptotic steady state and the
steepest roughening, respectively). Universality classes are well-known with
the set of exponents defining the final asymptotic steady state ($\theta \to \infty$) but
any pre-asymptotic transient period can be also very important for applica-
tions especially if it complies with experimental requirements. For example,
every growth process with a fast enough surface relaxation usually begins
with a 2D layer-by-layer mode that under certain conditions could be suffi-
cient for the concrete experiment.

In the early atomistic SOS models the surface is relaxed in such a way
that the global roughness increases monotonically until reaches an asym-
ptotic steady state determined by the substrate size. This is caused by the
fact that after adsorption, the adatoms perform a single hop and move ei-
ther directly to a site of local minimum height (EW-universality class) or
to some of its nearest neighbors of any height (VLDS-universality class).
Traditionally, the VLDS-universality class is considered as a correct enough
description of conserved epitaxial growth process, but there exist microscopic
situations in which it seems to be unsatisfactory. A natural complication of
early models is a KMC model based on Arrhenius activated diffusion, called
in the literature also realistic or collective diffusion model stressing on the
fact that detailed motion of the deposited atoms is included. As it is pointed
out in [25], the main difficulty with the analysis of RMSR in realistic models
is that all involved processes as diffusion, nucleation, atom incorporation at
steps, etc., take place over a wide range of time scales. Such schemes are
appropriate for simulations below the roughness transition allowing to avoid
equilibrium surface roughening and ignore evaporation. A crucial parameter
is the substrate size which not only determines the asymptotic state, but if
it is not large enough compared to the adatom diffusion length, can also be
a limiting factor for obtaining the correct scaling behavior.

Several SOS models with Arrhenius dynamics are used in the literature
for understanding the dynamic scaling of conserved epitaxial growth. One of
the first growth models with detailed description of surface diffusion in 1+1D
is published in [26] where the effective exponents ($\alpha_{\text{eff}}, \beta_{\text{eff}}$) are found to vary
from $(\infty, 1/2)$ (random deposition) to approximately $(0, 0)$ with increasing
the temperature, and at intermediate temperatures a possible growth expo-
nent $\beta$ of a new universality class is proposed. The fundamental question for
existence of a universality class which includes the SOS model with Arrhenius
dynamics is addressed in a number of works (see e.g. [15, 16, 27, 4, 28, 29]).
Most of the papers show temperature dependence of the effective growth exponents, crossovers from linear to nonlinear growth exponents and do not
exclude the possibility for unstable growth. The membership of SOS model in VLDS class is finally proved in [30, 31, 32] and only recently, an analytic formula for \( w \) is derived in [33] in the case of irreversible growth:

\[
w = \frac{L^\alpha}{R^{1/2}} f(Rt/L^{\alpha/\beta}),
\]

with \( \alpha = 2/3, \ \beta = 1/5 \) (2+1D case, see also [15, 16]).

The present work is devoted to an analytic description of surface roughness \( w \) obtained by KMC simulations with an alternative to the commonly used SOS model presumably improving some of its potential ”weaknesses” and describing well enough the conserved epitaxial growth with Arrhenius dynamics.

2. Motivation and a short description of the model

In most SOS models with Arrhenius dynamics any adatom is allowed to perform a hop down no matter how large the step height is and this artificial rule is the main reason for formation of asymptotic steady state. In order to satisfy the requirement to produce no overhangs, the adatom should slide in one move only along a column wall while eventually sits on top of the neighboring column. As it is noted in [28], this is unrealistic and causes anisotropy between horizontal and vertical surface diffusion. Such a compromise with jumps down of arbitrary height has been used in simulations since models including detailed vertical diffusion would be much more time consuming. There exist also models keeping the height difference between any two adjacent columns smaller than a prescribed global number [34, 35]. Such a strategy avoids the potential ”weakness” of the model described above but apparently introduces another one and thus, also fails to identify the real class corresponding to SOS schemes with Arrhenius dynamics.

Searching for a possibly better solution of the discussed problem we propose the following rule for surface diffusion. Let us assume an atom is sitting at a step of height larger than 1 and is allowed to perform only moves down of height 1. If after the first vertical move down along the column wall the atom possesses only lateral neighbors (at least two), this will lead with a high probability to an overhang which violates the condition for conservativeness. If it has only one lateral neighbor, the activation energy for such a hop will be higher than activation energy in the case of one lateral and one vertical
neighbor beneath because of the lower number of nearest neighbors at the final position of the move. If the difference between the two activation energies is high enough one can allow interlayer transport only downwards at steps of height 1. This is what is assumed in our model (diffusion upwards is not allowed and only irreversible growth is investigated in the current settings).

Our simulations with relatively low values of diffusion-to-deposition ratio $R$ produce a surface morphology characterized with formation of voids, cracks, pits, etc. which is known often to occur in real growth conditions at low temperatures. With increasing of $R$, however, in long time periods the concentration of such irregularities is limited to a low value, eventually vanishing at $R \to \infty$.

Generally, two main factors are responsible for time evolution of surface morphology – deposition competes with surface diffusion. Since these mechanisms are controlled by different parameters, for a precise comparison with experimental results, simulations and theoretical models need to take into account at least a basic set of growth parameters which include temperature $T$, deposition rate $F$, diffusion rate $D$ and coverage $\theta$. Following this strategy, we build a model based on a simple cubic geometry with the typical assumptions: the atoms adsorb on top of already deposited ones (thus, no overhangs are allowed) at a constant rate $F$; they diffuse on terraces randomly to one of its nearest neighbors at a rate $D$ (Eq. 4) and desorption has negligible values; for the sake of simplicity, in this study only irreversible aggregation is considered – the atoms become immobile after attaching to an island or another atom (corresponding to zero or negligible detachment rate); an important difference with the previous conserved SOS models is that the atoms can move to another layer only by hopping one monolayer (ML) down with the same rate (no ES barrier); diffusion rate $D$ satisfies an Arrhenius law:

$$D = \nu \exp(-\Delta E/k_B T),$$

(4)

where $\Delta E$ is the diffusion barrier for an adatom on a terrace. The attempt frequency is chosen to be $\nu = 2k_BT/h$ ($k_B$ is Boltzmann’s constant, $h$ – Planck’s constant).

Note that the models with a single dominating kinetic process differing in some of the parameters (temperature, activation energy, attempt frequency or deposition rate) can be unified by considering only the diffusion-to-deposition ratio (variable) $R = D/F$. Therefore, the surface evolution can be fully described by two independent variables - the coverage $\theta$ and $R$. 
The two extreme values of $R$ are $R \to 0$ ($\Delta E$, $F$ are sufficiently large or $T$ – sufficiently low) and $R \to \infty$ ($\Delta E$, $F$ are sufficiently small or $T$ – sufficiently high). The latter case corresponds to an ideal smoothly growing film with an oscillating $w$ while the former one is the random deposition model (the "benchmark" model as it is called in [23], see also [36, 17, 18]) in which the atoms are randomly deposited at a constant rate on top of the previously adsorbed ones, the columns grow independently and obey Poisson statistics:

The probability $P_k(t)$ – a given column to be of height $k$ at a time $t$ is

$$P_k(t) = \frac{(Ft)^k}{k!} \exp(-Ft), \quad (5)$$

the coverage $\theta = \bar{h}(t)$ is

$$\bar{h}(t) = \sum_{k=0}^{\infty} k \frac{(Ft)^k}{k!} \exp(-Ft) = Ft, \quad (6)$$

and the RMRS $w$ is given by

$$w(t) = \left( \sum_{k=0}^{\infty} \frac{(Ft)^k}{k!} \exp(-Ft)(k - Ft)^2 \right)^{1/2} = (Ft)^{1/2} = \theta^{1/2}. \quad (7)$$

Similar problems are considered in [18] for different adsorption-site geometries and different structures. For adsorption at four-fold hollow sites of a face-centered or body-centered cubic geometry, the columns do not grow independently, and the asymptotic kinetic roughening has the form $w \sim \theta^3$ where $\beta \approx 1/4$.

All simulations in our study are performed on a substrate of size $100 \times 100$, with deposition rate $F = 0.01$ ML/s, temperature $T = 800^\circ$C, $\Delta E$ mainly in the range $1.85 - 2.75$ eV and time $t$ up to $4.0 \times 10^6$ s. Translated to ($R, \theta$)"terminology", this means $R = 10^2 - 10^7$ and $\theta = 0 - 4.0 \times 10^4$. The conclusions from our simualtions are not hindered by the relatively moderate substrate size and coverage since the results presented below as well as single trials on larger substrates ($200 \times 200$ and $300 \times 300$) show no dependence of RMSR on the system size which in turn means that the problem does not exhibit finite size effects. We measure the roughness at coverages $\theta$ with uniformly distributed fractional part and therefore, in the case of ideal layer-by-layer growth ($R \to \infty$), it is expected to oscillate between 0 and 1/2 with a mean value

$$w_\infty = \int_0^1 \sqrt{x - x^2} dx \approx 3.9 \times 10^{-1}. \quad (8)$$
3. Results and discussion

In order to get a feeling of the effective growth exponent we first examine the curve of \( \log w \) (shown in Fig. 1 (left) for two particular energies corresponding to \( R = 3.6 \times 10^4 \) and \( R = 1.0 \times 10^5 \)). One can distinguish between three different growth modes. After the initial increase of the growth exponent \( \beta \) (not shown here), a stable period with \( \beta = 0.75 \) follows which length is an increasing function of \( R \) (see black dotted line). This is the first growth regime (it can be seen also in Fig. 1 (right) for higher values of \( R \)). The second straight line with a slope 1 (brown dotted line) shows the existence of a second (short unstable) growth mode, where \( \beta \) increases from 0.75 to values equal to 1 (for \( R = 3.6 \times 10^4 \)) or greater than 1 (for \( R = 1.0 \times 10^5 \)). The highest value of \( \beta \) clearly corresponds to the inflection point of the function \( \log w \) which changes from convex in the first period with \( \beta = 0.75 \) to concave in a third period where the curve approaches the line 0.5 log(\( \theta \)). This suggest \( \beta = 0.5 \) as a strong candidate for a growth exponent in the asymptotic state or equivalently, \( w \) tends to the random deposition power function \( \theta^{1/2} \) as \( \theta \to \infty \) and in asymptotic sense, the model coincides with random deposition model. Stronger evidences and precise expressions for \( w \) as a function of \( R \) and \( \theta \) follow below.

Least squares approximations provide the following analytic expressions of \( w \) in the transient regime with \( \beta = 0.75 \) and in the asymptotic regime with \( \beta = 0.5 \), correspondingly:

\[
\tilde{w}_1(R, \theta) = 9.64 R^{-1/2} \theta^{3/4} \tag{9}
\]
and
\[ \tilde{w}_2(R, \theta) = g(\theta; \lambda, \xi)\theta^{1/2}, \] (10)
where the function \( g \) is defined by
\[ g(\theta; \lambda, \xi) = \exp[\exp[-(\log \theta/\lambda)^\xi] - 1], \] (11)
the parameters \( \lambda \) and \( \xi \) depend only on \( R \) and some of their values are presented in Table 1. However, it should be emphasized that the function \( g \) used in the definition of \( \tilde{w}_2 \) is not unique, the values of \( \lambda \) and \( \xi \) are approximate and may undergo slight changes if results from longer simulations are used. In Fig. 2 (left), simultaneous approximation of \( w \) by \( \tilde{w}_1 \) and \( \tilde{w}_2 \) is shown in the cases \( R = 3.6 \times 10^4 \) and \( R = 1.0 \times 10^5 \) (the only ones in our simulations with \( L = 100 \) where both periods with \( \beta = 0.75 \) and \( \beta = 0.5 \) are clearly visible). In the right part of the same figure, only the functions \( \tilde{w}_1 \) are shown for three different values of \( R \). Fig. 3 also presents approximations of \( w \) by \( \tilde{w}_1 \) for higher deposition-to-diffusion ratio \( R \) (left) and by \( \tilde{w}_2 \) for lower \( R \) (right). These observations are clear evidences (but of course, not strict proofs) that the scaling behavior of the epitaxial model under consideration is described in asymptotic limit by
\[ w(R, \theta) \sim \theta^{1/2}, \] (12)
that is, for \( \theta \to \infty \) it coincides with the model of random deposition. In other words, the described growth process is a new representative of the oldest and simplest universality class random deposition with \( \alpha = \infty \) and

| \( R \)       | \( \lambda \) | \( \xi \)   |
|-------------|--------------|------------|
| \( 4.7 \times 10^2 \) | 3.60        | -3.54     |
| \( 1.4 \times 10^3 \) | 4.27        | -3.96     |
| \( 4.1 \times 10^3 \) | 5.61        | -6.12     |
| \( 1.2 \times 10^4 \) | 6.69        | -6.29     |
| \( 3.6 \times 10^4 \) | 7.99        | -9.12     |
| \( 1.0 \times 10^5 \) | 9.81        | -14.37    |

Table 1: The values of \( \lambda \) and \( \xi \) used in the definition of \( g(\theta; \lambda, \xi) \).
\( \beta = 0.5 \). Furthermore, it possesses a pronounced (pre-asymptotic) transient regime obeying the law

\[
\begin{align*}
  w(R, \theta) & \sim R^{-1/2} \theta^{3/4}. \\
\end{align*}
\]  

(13)

Not surprisingly, the prefactor \( R^{-1/2} \) in Eq. 13 is the same as in the analytic formula for \( w \) derived from traditional SOS model with Arrhenius dynamics (see [33, page 4]), which is an indirect evidence for the validity of our model. As we mentioned in the introduction, due to this prefactor, at low temperatures or for small \( R \) the surface is covered by a large number of unevennesses and vice versa – extremely low concentration of such irregularities is observed if \( R \) is large enough. For comparison, snapshots of the surface can be seen in Fig. 4 at coverages \( \theta = 17 \) ML for \( R = 9.1 \times 10^5 \) (left) and \( \theta = 14 \) ML for

Figure 2: Approximation of \( w \) simultaneously by \( \tilde{w}_1 \) and \( \tilde{w}_2 \) (a) and by \( \tilde{w}_1 \) (b) for different values of \( R \).

Figure 3: Approximation of \( w \) by \( \tilde{w}_1 \) for higher values of \( R \) (left) and by \( \tilde{w}_2 \) for smaller \( R \) (right).
$R = 4.1 \times 10^3$ (right).

Figure 4: Snapshots of the surface at coverage $\theta = 17$ ML for $R = 9.1 \times 10^5$ (left) and $\theta = 14$ ML for $R = 4.1 \times 10^3$ (right). The color changes from green to red with increasing the height.

The first results from simulations with included nearest-neighbor interaction energy in the definition of activation barrier $\Delta E$ exhibit the same characteristics as in the case of irreversible growth. As could be expected, allowing detachment of atoms from islands leads only to prolongation of the pre-asymptotic regime with growth exponent $\beta = 3/4$. Simulations allowing hopping down at steps of limited height greater than 1 perhaps would also be of interest.

4. Conclusions

We have proposed an alternative of the commonly used solid-on-solid model with Arrhenius dynamics. The results from first simulations with our model show that kinetic roughening of the surface for intermediate values of diffusion-to-deposition ratio $R$ is successfully described by two stable growth regimes – a transient pre-asymptotic regime with growth exponent $\beta = 3/4$ and an asymptotic state with $\beta = 1/2$ (coinciding with random deposition). Our considerations and precise analytic results derived from the simulations
confirm the validity and applicability of the presented KMC model. We have performed also some simulations with the extended definition of $\Delta E$ including nearest-neighbor interactions which seemingly do not alter the profile of temporal roughness evolution.

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