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Published in:
Quantum Dots: Fundamentals, Applications, and Frontiers

DOI:
10.1007/1-4020-3315-X_6

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Document Version
Publisher's PDF, also known as Version of record

Publication date:
2005

Link to publication in University of Groningen/UMCG research database

Citation for published version (APA):

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OFF-LATTICE KMC SIMULATIONS OF STRANSKI-KRASTANOV-LIKE GROWTH

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Abstract We investigate strained heteroepitaxial crystal growth in the framework of a simplifying (1 + 1)-dimensional model by use of off-lattice kinetic Monte Carlo simulations. Our modified Lennard-Jones system displays the so-called Stranski-Krastanov growth mode: initial pseudomorphic growth ends by the sudden appearance of strain induced multilayer islands upon a persisting wetting layer.

1. Introduction

In addition to its technological relevance, epitaxial crystal growth is highly attractive from a theoretical point of view. It offers many challenging open questions and provides a workshop in which to develop novel methods for the modeling and simulation of non-equilibrium systems, in general. In particular, strained heteroepitaxial crystal growth attracts significant interest as a promising technique for the production of, for instance, high quality semiconductor films. Recent overviews of experimental and theoretical investigations can be found in, e.g. [1, 2, 3]. A particularly attractive aspect is the possibility to exploit self-organizing phenomena for the fabrication of nanostructured surfaces by means of molecular-beam epitaxy (MBE) or similar techniques.

In many cases, adsorbate and substrate materials crystallize in the same lattice but with different bulk lattice constants. Frequently, one observes that the adsorbate grows layer by layer, initially, with the lateral spacing of atoms adapted to the substrate. The misfit induces compressive or tensile strain in this pseudomorphic film and, eventually, misfit dislocations will appear. These relax the strain and the adsorbate grows with its natural lattice constant far from the substrate, eventually.

Dislocations will clearly dominate strain relaxation in sufficiently thick films and for large misfits. In material combinations with relatively small misfit an alternative effect governs the initial growth of thin films: Instead of growing layer by layer, the adsorbate aggregates in three-dimensional (3D) structures. The term 3D islands is commonly used to indicate that these structures are spatially separated. The situation is clearly different from the emergence of mounds due to the Ehrlich–Schwoebel (ES) instability [1, 2], for instance.

At least two different growth scenarios display 3D island formation: In Volmer–Weber growth, such structures appear immediately upon the substrate when depositing the mismatched material. The situation resembles the formation of non-wetting droplets of liquid on a surface. This growth mode is frequently observed in systems where adsorbate and substrate are fundamentally different, an example being Pb on a graphite substrate [1].

In the following we concentrate on the so-called Stranski–Krastanov (SK) growth mode, where 3D islands are found upon a pseudomorphic wetting layer (WL) of adsorbate material [1, 2, 3]. Two prominent prototype SK systems are Ge/Si and InAs/GaAs where, as in almost all cases discussed in the literature, the adsorbate is under compression in the WL. In order to avoid conflicts with more detailed definitions and interpretations of the SK growth mode in the literature we will resort to the term SK-like growth in the following. It summarizes the following sequence of phenomena during the deposition of a few monolayers (ML) of material:

1. The layer by layer growth of a pseudomorphic adsorbate WL up to a kinetic thickness $h^*_{WL}$.
2. The sudden appearance of 3D islands, marking the so-called 2D-3D or SK transition
3. Further growth of the 3D islands, fed by additional deposition and by incorporation of surrounding WL atoms
4. The observation of separated 3D islands of similar shapes and sizes, on top of a WL with reduced stationary thickness $h_{WL}$.

Admittedly, the above operative definition already reflects our interpretation of SK growth to a certain extent. Besides these basic processes a variety of phenomena can play important roles in the SK scenario, including the interdiffusion of materials and the segregation of compound adsorbates. These effects are certainly highly relevant in many cases, see [4, 5] and other contributions to this volume. However, SK-like growth is
observed in a variety of material systems which may or may not display these specific features. For instance, intermixing or segregation should be irrelevant in the somewhat exotic case of large organic molecules like PTCDA deposited on a metal substrate, e.g. Ag(111). Nevertheless, this system displays SK-like growth in excellent accordance with the above operative definition [6].

Despite the extensive investigation of SK growth, a complete detailed theoretical picture is still lacking, apparently. This concerns in particular the nature of the 2D-3D transition. One problem clearly lies in the richness of the phenomenon. On the other hand, the very diversity of SK-like systems gives rise to the hope that this growth scenario might be governed by a few basic universal mechanisms. Accordingly, it should be possible to capture and identify these essential features in relatively simple prototype systems.

This hope motivates the investigation of simplifying models without aiming at the reproduction of material specific details. Some of the key questions in this context are: under which conditions do a WL emerge and persist? How does its thickness before and after the SK transition depend on the growth conditions? Which microscopic processes trigger and control the sudden formation of 3D islands? How do the island size and their spatial arrangement depend on the parameters of the system?

Following earlier investigations of related phenomena, e.g. [8–11], we choose a classical pair potential ansatz to represent the interactions between atoms in our model. Here, we restrict our studies to the fairly simple case of a modified Lennard–Jones (LJ) system in 1 + 1 spatial dimensions, i.e. growth on a one-dimensional substrate surface. As we interpret our model as a cross-section of the physical (2 + 1)-dimensional case, we still use the common term 2D-3D transition for the formation of multilayer from monolayer islands.

We investigate our model by means of kinetic Monte Carlo (KMC) simulation, see e.g. [13] for an introduction and overview. This concept has proven useful in the study of non-equilibrium dynamical systems in general and, in particular, in the context of epitaxial growth, see e.g. [1, 2]. Most frequently, pre-defined lattices are used for the representation of the crystal. So-called solid-on-solid (SOS) models which neglect lattice defects, bulk vacancies, or dislocations have been very successful in the investigation of various relevant phenomena, including scenarios of kinetic roughening or mound formation due to instabilities [1, 2]. There is, however, no obvious way of including mismatch and strain effects in a lattice gas model. A potential route is to introduce additional elastic interactions between neighboring atoms in an effective fashion. In fact, such models of heterosystems have been studied in some
of the earliest KMC simulations of epitaxial growth [14, 15], see [16] for a recent example of a so-called ball and spring model. In alternative approaches the strain field of a given configuration is evaluated using elasticity theory as, for instance, in [17].

In order to account for strain effects more faithfully, including potential deviations from a perfect lattice structure, it is essential to allow for continuous particle positions. Given at least an approximation for the interatomic potentials, a molecular dynamics (MD) type of simulation [18] would be clearly most realistic and desirable, see [19] for one example in the context of heteroepitaxial growth. However, this method suffers generally from the restriction to short physical times on the order of $10^{-6}$ s or less. MBE relevant time scales of seconds or minutes do not seem feasible currently even when applying sophisticated acceleration techniques [20].

Here, we put forward an off-lattice KMC method which has been introduced in [7, 8, 11] and apply it in the context of the SK scenario. Some of the results have been published previously in less detail [12]. The paper is organized as follows. In the next section we outline the model and simulation method. Before analysing the actual SK-like growth in Sec. 4 we present some basic results concerning various diffusion scenarios in Sec. 3. In the last section we summarize and discuss open questions and potential extensions of our work.

2. Model and Method

In our off-lattice model we consider pairwise interactions given by LJ potentials of the form [18]

$$U_{ij}(U_0, \sigma) = 4U_0\left[\left(\frac{\sigma}{r_{ij}}\right)^{12} - \left(\frac{\sigma}{r_{ij}}\right)^6\right],$$

where the relative distance $r_{ij}$ of particles $i$ and $j$ can vary continuously. As a widely used approximation we cut off interactions for $r_{ij} > 3\sigma$.

The choice of the parameters $\{U_0, \sigma\}$ in Eq. (1) characterizes the different material properties in our model: interactions between two substrate (adsorbate) particles are specified by the sets $\{U_s, \sigma_s \equiv 1\}$ and $\{U_a, \sigma_a\}$, respectively. Instead of a third independent set we set $U_{sa} = \sqrt{U_sU_a}\sigma_s\sigma_a = \frac{1}{2}(\sigma_s + \sigma_a)$ for the inter-species interaction.

As the lattice constant of a monatomic LJ crystal is proportional to $\sigma$, the relative misfit is given by $\epsilon = (\sigma_a - \sigma_s)/\sigma_s$. Here we consider only cases with $\sigma_a > \sigma_s$, i.e. positive misfits $\epsilon$ of the order of a few percent. For the simulation of SK-like growth we set $U_s > U_{sa} > U_a$. In such systems, the formation of a WL and potential layer-by-layer growth should be
favorable, in principle. If not otherwise specified, we have set the misfit
to $\epsilon = 4\%$, a typical value for SK systems, and used the LJ prefactors
$U_s = 1.0$ eV, $U_\alpha = 0.74$ eV, $U_{\alpha s} \approx 0.86$ eV.

Growth takes place on a substrate represented by six atomic layers,
with the bottom layer fixed and periodic horizontal boundary conditions.
In the following we mostly refer to systems with $L = 800$ particles per
substrate layer, additional simulations with $L = 400$ or $600$ revealed no
significant $L$-dependence of the results presented here. The deposition
of single adsorbate particles is performed with a rate $R_d = LF$, where $F$
is the deposition flux. As we interpret the substrate lattice constant as
our unit of length, flux and deposition rate (measured in ML/s) assume
the same numerical values. The rates of all other significant changes of
the configuration are given by Arrhenius laws. We consider only hopping
diffusion events at the surface and neglect bulk diffusion, exchange
processes or other concerted moves. Furthermore, diffusion is restricted
to adsorbate particles at the surface whereas jumps of substrate particles
onto the surface are not considered. As we will demonstrate in a forthcoming
publication, these simplifications are justified for small misfits $\epsilon$
in the LJ system because the corresponding rates are extremely low, see
also [7, 8].

The rate $R_i$ for a particular event $i$ is taken to be of the form

$$R_i = \nu_0 \exp \left(-\frac{E_i}{k_B T}\right),$$

where $T$ is the simulation temperature and $k_B$ the Boltzmann constant.
For simplicity, we assume that the attempt frequency $\nu_0$ is the same
for all diffusion events. In order to relate to physical units we use $\nu_0 = 10^{12}$ s$^{-1}$ wherever numerical results are given. The activation barriers
$E_i$ are calculated on-line given the actual configuration of the system.
This can be done by a minimal energy path saddle point calculation
[9, 18]. Here, we use a frozen crystal approximation, which speeds up
the calculation of barriers significantly (see [21] for an application in the
context of strained surfaces). Note that the calculations are particularly
simple in 1+1 dimensions: the path between neighboring local minima
of the potential energy is uniquely determined and the transition state
corresponds to the separating local maximum.

An important modification concerns interlayer diffusion. LJ systems
in 1+1 dimensions display a strong additional barrier, which hinders
such moves at terrace edges [1, 2]. This so-called Ehrlich–Schwoebel
(ES) effect is by far less pronounced in (2+1)-dimensional systems,
because interlayer moves follow a path through an energy saddle point
rather than the pronounced maximum at the island edge. In our investi-
gation of the SK-like scenario we remove the ES barrier for all interlayer diffusion events by hand. One motivation is the above mentioned overestimation in one dimension. More importantly, we wish to investigate strain-induced island formation without interference of the ES instability. Note that the latter leads to the formation of mounds even in homoepitaxy [1, 2].

The rates for deposition and diffusion are used in a rejection-free KMC simulation [13]. Using a binary search tree technique, one of the possible events \( j \) is drawn with probability \( R_j / R \), where \( R = (R_d + \sum_i R_i) \) is the total rate of all potential changes. Time is advanced by a random interval \( \tau \) according to the Poisson distribution \( P(\tau) = R e^{-R\tau} \) [13]. In order to avoid the artificial accumulation of strain due to inaccuracies of the method, the potential energy should be taken to the nearest local minimum by variation of all particle positions in the system. This relaxation process affects both, adsorbate and substrate atoms. In order to reduce the computational effort, we restrict the variation to particles within a radius \( 3\sigma_s \) around the location of the latest event, in general. The global minimization procedure is performed only after a distinct number of steps. It is important to note neither procedure leads to a substantial rearrangement. Significant changes of the activation energies due to relaxation signal the necessity to perform the global procedure more frequently.

3. Diffusion Processes

Before analysing the SK-like scenario, we compare the barriers for hopping diffusion in various settings on the surface. The investigation of systems like Ge/Si(001) reveal a very complicated scenario due to anisotropies and the influence of surface reconstructions [22, 23]. For Ge on Ge(111) the barrier for hopping diffusion is higher on the surface of a compressed crystal, whereas diffusion is faster on relaxed Ge [23].

Lennard–Jones or similar models do not, in general, reproduce this feature. Schroeder and Wolf [21] consider \((2+1)\)-dimensional single species LJ systems and evaluate the diffusion barrier for a single adatom on surfaces in various lattice types. Among other results they find that mechanical compression of the crystal lowers the barrier for surface diffusion. However, in the mismatched two species system, it is more important to compare diffusion on (a) the substrate, (b) the WL and (c) the surface of partially relaxed islands.

The strong adsorbate-substrate interaction \( (U_{as} > U_a) \) favors the formation of a WL, but it also yields deep energy minima and a relatively high diffusion barrier for adsorbate particles on the substrate. This ef-
fect is much weaker for particles on a complete wetting monolayer of adsorbate and, hence, the corresponding diffusion barrier is significantly lower. The faster diffusion further stabilizes the WL, as deposited particles will reach and fill in gaps easily. In principle, the trend extends to the following layers. However, due to the short range nature of the LJ potential the influence of the substrate essentially vanishes on WL of three or more monolayers. With the example choice $\epsilon = 4\%$ and $U_s = 1.0$ eV, $U_a = 0.74$ eV, $U_{ad} \approx 0.86$ eV, we find an activation barrier of $E_{a}^{0} \approx 0.57$ eV for adsorbate diffusion on the substrate and $E_{a}^{1} \approx E_{a}^{2} \approx 0.47$ eV for diffusion on the first and second adsorbate layer, respectively.

For the SK scenario the diffusion on islands of finite extension is particularly relevant. Figure 1 shows the barriers for diffusion hops on islands of various heights located upon a wetting monolayer. We wish to point at two important features: (i) Diffusion on top of islands is, in general, slower than on the WL and the difference increases with the island height. In our model, this is an effect of the partial relaxation or over-relaxation in the island top layer. (ii) Depending on the lateral

![Figure 1. Diffusion barriers as obtained in our model for a single adatom on a flat symmetric multilayer island with 24 base particles and height 1, 3, 5 layers (bottom to top curves). Symbols represent the activation energies for hops from the particle position $x$ to the left neighbor site. The island is placed on top of a single WL, interaction parameters are given in the text of section 3. The leftmost barriers correspond to downward jumps at the island edge with suppression of the ES effect. Horizontal lines mark the barrier for adatom diffusion on the WL (lower line) and on perfectly relaxed adsorbate material (upper line).](image)
island size and its height, there is a more or less pronounced diffusion bias towards the island center, reflecting the spatially inhomogeneous relaxation. A similar effect has been observed in (2 + 1)-dimensional LJ systems [21]. Note that (ii) has to be distinguished from the diffusion bias imposed by the ES effect, which would be present even in homoepitaxy and with particle positions restricted to a perfect undisturbed lattice. Clearly, (i) and (ii) favor the formation of islands upon islands and hence play an important role in SK-like growth. They concern adatoms which are deposited directly onto the islands as well as particles that hop upward at edges, potentially. As we will argue in the following section, upward diffusion moves play the more important role in the 2D-3D transition of our model.

4. SK-Like Growth Scenario

In our investigation we follow a scenario which is frequently studied in experiments [24, 25]. In each simulation run a total of 4 ML adsorbate material is deposited at rates in the range $0.5 \text{ ML/s} \leq R_d \leq 9.0 \text{ ML/s}$. After deposition is complete, a relaxation period with $R_d = 0$ of about $10^7$ diffusion steps follows, corresponding to a physical time on the order of 0.3 s. As we have demonstrated in [11], strain relaxation through dislocations is not expected for, say, $\epsilon = 4\%$ within the first few adsorbate layers. Indeed misfit dislocations were observed in none of the simulations presented here. Results have been obtained on average over at least 15 independent simulation runs for each data point.

In our simulations we observe the complete scenario of SK-like growth as described in the introduction. Illustrating mpeg movies of the simulations are available upon request or directly at our web pages [26]. A section of a simulated crystal after deposition and island formation is shown in Figure 2. Ultimately, the formation of islands is driven by the relaxation of strain. As shown in the figure, material within the 3D island and at its surface can assume a lattice constant close to that of bulk adsorbate. On the contrary, particles in the WL are forced to adopt the substrate structure.

During deposition, monolayer islands located on the WL undergo a rapid transition to bilayer islands at a well-defined thickness $h^*_{\text{WL}}$. For the systematic determination of $h^*_{\text{WL}}$ we follow [24] and fit the density $\rho$ of 3D islands as $\rho = \rho_0 (h - h^*_{\text{WL}})^\alpha$, finding comparable values of the exponent $\alpha$. Figure 3 displays the results for two different substrate temperatures $T$ and various deposition rates $R_d$. The increase of $h^*_{\text{WL}}$ with decreasing $T$ agrees qualitatively with several experimental findings, see [25] as one example. Heyn [4] discusses the effect of adsorbate/substrate
interdiffusion on the $T$-dependence of $h_{WL}^*$ in the InGaAs system. Reassuringly, our result is consistent with his findings without intermixing.

A key observation is the significant increase of $h_{WL}^*$ with increasing deposition flux. Qualitatively the same flux dependence is reported for InP/GaAs heteroepitaxial growth in [25]. This behavior leads to the conclusion that the emergence of islands upon islands, i.e. the SK transition, is mainly due to particles performing upward hops onto existing monolayer islands. If, on the contrary, the formation of second or third layer nuclei by freshly deposited adatoms was the dominant process, one
would expect more frequent nucleation and an earlier 2D-3D transition at higher growth rates. Note again that we have suppressed the ES effect explicitly which would also lead to more frequent mound formation at higher deposition rates [1, 2].

In our model, upward diffusion is the limiting effect and sets the characteristic time for the SK transition. The frequency of these processes strongly depends on the temperature and it has to be compared with the deposition rate. A high incoming flux will fill layers before particles can perform upward hops, and hence it will delay the 2D-3D transition, cf. Fig. 3. These considerations, together with the arguments of [25], suggest a functional dependence of $h_{WL}^*$ on $F$ and $T$ of the form

$$h_{WL}^* = h_0 \left( \frac{F}{R_{up}} \right)^\gamma,$$

where $R_{up}$ is the Arrhenius rate for upward hops. In our simulations we find a typical value which corresponds to an activation barrier $E_{up} \approx 1.0$ eV close to the transition. Using this value we have performed a non-linear fit according to Eq. (3) based on the data for $T = 500$ K. Inserting the obtained parameters $h_0 \approx 3.0$ ML and $\gamma \approx 0.2$ in Eq. (3) yields good agreement for $T = 480$K, as well. Note that the value of $\gamma$ is expected to vary with the material system in the range $0 < \gamma < 0.5$ according to [25]. Our assumption that upward diffusion is more important than nucleation of deposited adatoms on top of islands is further supported by the observation that multilayer islands tend to form on a WL of, say, two monolayers thickness even without particle deposition.

After the 2D-3D transition, islands grow by incorporating newly deposited material, but also by consumption of the surrounding WL. Very large islands are observed to split by means of upward diffusion events onto their top layer [26]. The migration of WL particles towards and onto the islands can also extend into the relaxation period after deposition ends. Eventually, a stationary WL thickness $h_{WL} \approx 1$ is observed in our example scenario with $U_{as} \approx 0.86$ eV. By increasing the strength of the adsorbate/substrate interaction we can achieve, e.g. $h_{WL} \approx 2$ for $U_{as} \approx 2.7$ eV, but it is difficult to stabilize a greater stationary WL thickness due to the short range nature of the LJ potential.

Finally, we discuss the properties of 3D islands emerging in SK growth. As an example, their base length $b$ is measured as the number of particles in the island bottom layer. The results discussed in the following were obtained at the end of the relaxation period with $R_d = 0$. Whereas mean values do not change significantly, fluctuations are observed to decrease with time in this phase. We observe that, for fixed temperature, the island size decreases with increasing deposition flux, as observed in several
experimental studies [25]. However, \( \langle b \rangle \) becomes constant and independent of \( T \) for large enough deposition rate, cf. Fig. 4. Corresponding behavior is found for the island density and their lateral distance, hinting at a considerable degree of spatial ordering [12]. The saturation behavior further demonstrates the importance of upward hops versus aggregation of deposited particles on islands. The latter process would yield a continuous increase of the island density with \( R_d \). We find a narrower distribution of island sizes with increasing \( R_d \) [12]. The island size distribution in the saturation regime will be studied in greater detail in forthcoming investigations.

We conclude our discussion by noting that, in the saturation regime of high growth rates, the typical island size follows a simple power law: \( \langle b \rangle \propto 1/\epsilon \), cf. Fig. 4. Very far from equilibrium, the only relevant length scale in the system is the relative periodicity \( 1/\epsilon \) of the adsorbate and substrate lattices. This characteristic length was already found to dominate in the formation of misfit dislocations for larger values of \( \epsilon \) [11].

5. Conclusion and Outlook

Despite its conceptual simplicity and the small number of free parameters, our model reproduces various phenomena of heteroepitaxial
growth. We believe that, with a proper choice of interaction parameters, our model should be capable of reproducing all three prototype growth modes: layer-by-layer growth (for very small misfits), Volmer–Weber (for $U_{\text{ad}} < U_a$) and, as demonstrated here, the Stranski–Krastanov mode. Our work provides a fairly detailed and plausible picture of the latter. The key features are:

(i) The strong adsorbate/substrate interaction favors the WL formation and results in a relatively slow diffusion of adatoms on the substrate. Diffusion on the WL is significantly faster and the corresponding barrier decreases with the WL thickness.

(ii) Strain relaxation leads to a pronounced bias towards the island center on top of finite mono- and multilayer islands located on the WL. In addition, diffusion is slower on top of the partially relaxed islands than on the WL, in our model.

Whereas (a) favors the formation and persistence of the WL, (b) clearly destabilizes layer by layer growth. We find that the microscopic process which triggers the transition is upward diffusion of adatoms from the WL and at island edges. The corresponding barriers decrease with the WL thickness analogous to (a). As a result of the competing effects, the 2D-3D transition occurs at a critical thickness which depends upon $T$ and $R_d$ as suggested in Eq. (3).

We hypothesize that strain effects induce spatial modulations with a characteristic length scale $\epsilon^{-1}$ and thus control the island size far from equilibrium. We find a corresponding saturation regime for large enough deposition fluxes. The precise mechanism of the size selection will be the subject of a forthcoming project. A related open question concerns the crossover from kinetically controlled island sizes to the equilibrium behavior which should be achieved in the limit of very small $R_d$. Several arguments [2, 17] suggest that the typical island size close to equilibrium should be of order $\epsilon^{-2}$.

Further investigations will concern the effects of intermixing and segregation which have been excluded from our model, so far. To this end we will consider the co-deposition of both species and allow for exchange diffusion at the substrate/adsorbate interface. In order to test the potential universality of our results, we will introduce different types of interaction potentials in our model. Ultimately, we plan to extend our model to the relevant case of 2+1 dimensions and to more realistic empirical potentials for semiconductor materials, e.g. [21].
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Acknowledgments

F. Much has been supported by the Deutsche Forschungsgemeinschaft. M. Biehl thanks the organizers and all participants of the NATO ARW on Quantum Dots: Fundamentals, Applications, and Frontiers for pleasant and useful discussions. We also thank B. Voigtländer for communicating the results of [23] prior to publication.

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[26] mp3 movies and other illustrations are available from the web pages http://www.physik.uni-wuerzburg.de/~biehl/~much.