Kinetic Monte Carlo simulation of quantum-dot nucleation and growth in PbSe/PbEuTe multilayers

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Abstract. We study the influence of different growth conditions on the structural properties of self-organized quantum-dot multilayers PbSe/PbEuTe(111). The kinetic Monte Carlo method is employed and the simulations show that mainly the quality of the dot ordering is affected when the growth conditions are slightly changed.

1. Introduction
Self-organization during a strain-driven heteroepitaxy of semiconductor nanostructures is a promising technique for fabrication of devices based on specific electrical and optical properties of quantum dots (QDs). Mechanisms of spontaneous formation of three-dimensional (3D) quantum dots in the Stranski-Krastanow growth mode as well as the self-assembling principles at their growth have been a field of intensive research in the last few years (for reviews see, e.g., Refs. [1, 2, 3]. For a prospective technological use the achievement of a narrow size distribution and a regular arrangement of the QDs is important. To this purpose the strain-mediated self-organization is advantageously applied at preparation of 3D superlattices of QDs, where the already grown islands influence positions and sizes of the islands in the subsequent layers [3, 4, 5].

Whereas the vertical correlations of dot positions are usually sufficiently ensured by the self-organization effect, the lateral ordering is considerably weaker. A unique example of a semiconductor system (grown on planar substrates) exhibiting good quality of vertical and lateral dot correlations are PbSe/PbEuTe(111) multilayers [6]. Depending on the thickness of the PbEuTe spacer layer, three different arrangements of a PbSe dot superlattice are observed (vertical and trigonal arrangements of the dots and a non-correlated mode), when also a pronounced hexagonal lateral ordering is formed in the both ordered cases [7, 8].

In the paper [9] we introduced a semi-atomistic kinetic Monte Carlo (KMC) model that we have developed in order to simulate the strain-mediated epitaxial growth of self-assembled multilayers. For the first time, this model enabled us to perform simulations of the dot nucleation and growth in multilayers consisting of tens of periods with hundreds of 3D quantum dots. We reproduced all three experimentally observed dot arrangements in the PbSe/PbEuTe(111) multilayers, moreover, with a good quantitative agreement. In this work we want to demonstrate a stability of the results in case the growth conditions are slightly altered. This is important since the experimental samples are also investigated within a range of growth conditions and, further, the conditions are adjusted (measured) only with a limited precision.
2. KMC model description

We shortly summarize the main features of our model (for details see Ref. [9]). The KMC method models a growth simulation as a sequence of random events taking place on a surface [10, 11, 12]. The basic included events are the deposition of adatoms onto the surface and their subsequent surface diffusion. Since the adatom diffusion is a thermally activated process, the hopping rate \( k \) of an adatom onto a nearest-neighbor site is given by the Arrhenius-like formula

\[
k_{i \rightarrow f} = k_0 \exp(-E_{i \rightarrow f}/k_B T),
\]

where \( k_0 = 2k_B T/h \sim 10^{13} \text{s}^{-1} \) is a characteristic vibrational frequency and \( T \) is the substrate temperature. \( E_{i \rightarrow f} \) denotes the energy barrier which an adatom must overcome at a hop from the initial site \( i \) to the final site \( f \). In our approach based on incorporation of the elastic strain to the adatom diffusion we model \( E_{i \rightarrow f} \) as a sum of two terms: a substrate term \( E_S \) and a strain-field contribution induced by buried dots,

\[
E_{i \rightarrow f} = E_S + K(w_f - w_i) V_0.
\]

\( w_i, w_f \) are the elastic energy densities at the sites \( i \) and \( f \) calculated from the strain tensor \( \epsilon_{ij} \) and the elastic constants \( C_{ijkl} \) as \( w = 1/2 C_{ijkl} \epsilon_{ij} \epsilon_{kl} \). The strain tensor on the surface of wetting layer, calculated in the approach of elastic continuum, comprises the pseudomorphic deformation of the wetting layer and the individual strain contributions from all QDs buried under the just growing layer. \( V_0 \) is the volume of one adatom (one PbSe molecule) and the coefficient \( K > 0 \) is a correction factor which absorbs all simplifications and neglects made in the model; above all, the omission of strain field caused by growing dots in Eq. (2).

The key simplification of our model is that the shape of growing dots is not atomistically simulated but it is taken \textit{ad hoc} from the experiment. Thus the dots are modeled as trilateral pyramids with \{100\} side facets [6] and only their size changes during the simulation. The main advantage of this approximation is that the simulations are substantially faster, so a larger area and more multilayer periods can be simulated, which is more important for the self-organization effect study. Since the dots are not treated atomistically and thus the described model is not bond-counting, a critical-nucleus concept is introduced in a special way. We consider a nonzero effect study. Since the dots are not treated atomistically and thus the described model is not

3. Simulation results

As we adumbrated in the introduction we performed simulations under slightly different conditions (with respect to those used in [9]) to test the stability of our results. A fundamental influence on the epitaxial growth is possessed by the substrate temperature \( T \) and the deposition flux \( F \). From this reason we altered just them, whereas the other model parameters remained constant (namely: \( E_S = 0.93 \text{eV}, N_c = 100 \) PbSe molecules, \( K = 7 \), total PbSe coverage of \( 5 \text{ML} \) and its part used for the formation of dots of \( 1.2 \text{ML} \), and simulation grid size of \( 2048 \times 2048 \) cells). For lucidity, let us label as \( A \) the growth conditions with \( T = 370 \circC \) and \( F = 0.12 \text{ML/s} \) used in [9], and as \( B \) the changed ones with \( T = 360 \circC \) and \( F = 0.08 \text{ML/s} \).

Figure 1 shows simulation results of three multilayers consisting from 20 periods grown under the conditions \( B \) for three different spacer thicknesses \( d_{SP} \) of 26, 45, and 60 nm. The first layer in all these multilayers is identical and contains completely disordered array of dots. The dot density in the layer \( n = 512 \text{µm}^{-2} \) distinctly differs from that obtained at the growth conditions \( A \) where it amounted to \( 442 \text{µm}^{-2} \). Regardless this fact, the final arrangement of the whole multilayers is \textit{qualitatively} the same for both growth conditions \( A \) and \( B \) (to compare see [9]). For \( d_{SP} = 26 \text{nm} \) we obtained again a vertically aligned multilayer [panels (a), (b) in Fig. 1], for
Figure 1. Panels (a), (c), and (e) display the top views on positions and sizes of the dots in the 20th periods of three multilayers simulated with different spacer thicknesses of 26, 45, and 60 nm, respectively. The two dashed lines always mark out the location and orientation of a vertical cross-section through the related multilayer. The cross-sections are displayed in panels (b), (d), and (f), respectively. The gray level of the QD-representing triangles indicates their complementary lateral coordinate in the given cross-section band. The left-bottom-corner insets in the upper panels show the 2D FFT power spectra of the given surface morphology, and the right-bottom-corner insets show the cross-correlation functions of the 19th and 20th dot-layer morphologies.

For $d_{SP} = 45$ nm an ABCABC stacking of dot layers is produced [panels (c), (d) in Fig. 1], and for $d_{SP} = 60$ nm an uncorrelated multilayer grew [panels (e), (f) in Fig. 1].

A quantitative comparison between the multilayers grown at the conditions A and B is given in Table 1. The initial dot densities $n_i$ in first layers and the dot densities $n$, mean lateral distances $L$ and correlation lengths $L_{corr}$ in the 20th layers (determined from the FFT power spectra) are compared. The values of $L$ remained constant (within the measurement error) both for $d_{SP} = 26$ nm and $d_{SP} = 45$ nm. It is due to the fact, that the lateral dot distance is primarily determined by the strain field distribution, i.e. by the spacer layer thickness and
Table 1. Comparison of dot-arrangement parameters (dot density $n$, lateral dot distance $L$, and lateral correlation length $L_{corr}$) at the topmost (20$^{th}$) period of multilayers simulated under two different growth conditions A and B (see the text) for three different spacer thicknesses $d_{SP}$. The quantity $n_i$ denotes the initial dot density in the first period.

| $d_{SP}$ (nm) | growth cond. | $n_i$ ($\mu$m$^{-2}$) | $n$ ($\mu$m$^{-2}$) | $L$ (nm) | $L_{corr}$ (nm) |
|---------------|--------------|------------------------|---------------------|---------|----------------|
| 26 A          | 442 ± 10     | 176 ± 10               | 87 ± 8              | 224 ± 30 |
| 26 B          | 512 ± 10     | 197 ± 10               | 89 ± 8              | 219 ± 30 |
| 45 A          | 442 ± 10     | 295 ± 10               | 65 ± 3              | 243 ± 30 |
| 45 B          | 512 ± 10     | 337 ± 10               | 65 ± 3              | 223 ± 30 |
| 60 A          | 442 ± 10     | 440 ± 10               | disordered          |         |
| 60 B          | 512 ± 10     | 507 ± 10               | disordered          |         |

the elastic constants which stayed identical. The dot densities $n_i$ and $n$ for $d_{SP} = 60$ nm are different, however, they do not change (out of the error) during the multilayer growth which confirms the uncorrelated mode. Thus the only one significant difference is in the lateral ordering quality (described by $n$ and $L_{corr}$) for $d_{SP} = 45$ nm. The more gradual ordering is caused by smaller modulation of the strain field in comparison with the thin spacer of 26 nm.

4. Conclusion
The presented results verified that the self-organization effect is primarily caused by the strain field distribution, namely by the strain field induced by buried dots, since no other mechanism is included in our model. Moreover, they further confirmed that the mean lateral dot distance in ordered multilayers does not depend on the growth conditions if they are varied in a small range, since the distance is also predetermined by the strain field. On the other hand, different growth conditions can influence mainly the quality of a QD superlattice ordering.

Acknowledgments
This work is a part of the research program MSM 0021620834 that is financed by the Ministry of Education of the Czech Republic; the work has been also supported by the EC SANDiE European Network of Excellence and by the project KAN400100652 of the Grant Agency of the Academy of Sciences of the Czech Republic.

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