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Fast kinetic Monte Carlo simulation and statistics of quantum dot arrays

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ABSTRACT

The paper introduces a new three-dimensional heteroepitaxial kinetic Monte Carlo (KMC) model for fast simulation of self-assembled quantum dot (QD) arrays. It represents a computationally efficient simplification of the ball-and-spring model and captures the most important features of heteroepitaxial growth. This conclusion is supported by our results obtained from KMC simulations of InAs QDs grown on GaAs(001) substrate at the following technologically relevant conditions: temperature in the range $T = 700–800$ K, deposition rate $F = 0.1–1.6$ ML/s, and a set of energy barriers derived from the literature. The main characteristics of QDs such as uniformity in size, aspect ratio, spatial ordering and others, are studied as functions of the most important parameters describing the growth process — substrate temperature, deposition rate and surface coverage.

1. Introduction

Due to the unique physical properties, semiconductor quantum dots (QDs) are important for applications in various advanced optoelectronic and single-electron devices — lasers, detectors, and many others (see [1] and references therein). Usually such practical applications require fabrication of QD arrays, consisting of millions of densely packed, equisized and uniformly distributed QDs [2]. Along with the techniques based on prepatterned substrates [3] one of the most promising methods for fabrication of QD arrays is the self-assembly (or self-organization) [4–7] which often gives even better results as regards the size and spatial distribution of the dots. By this method, the desired three-dimensional (3D) structures grow as a result of deposition of a certain semiconducting material on a substrate made of another material with the same crystalline structure but with lattice constant(s) differing significantly from the lattice constant(s) of the deposited compound. For most applications of QDs it is crucial to be achieved as equally shaped as possible dots, with high density, uniform spatial ordering and narrow size distribution. The main parameters which control these quantities are the substrate temperature T, the deposition rate F and the surface coverage θ.

Kinetic Monte Carlo (KMC) models (see e.g. [8–11]), molecular dynamics [12], and continuum models [13,14] have been used to understand the complicated process of QD self-assembly. Growth of Ge_xSi_{1-x}/Si(001) and In_xGa_{1-x}/Ga(001) nanostructures are the most widely studied prototypes. A 2D multiscale lattice model incorporating effective surface reconstructions is used in [8] for investigation of the pyramid-to-dome transition in the heteroepitaxial growth of Ge_xSi_{1-x} on Si. With this model, both shallow and steep facets are simulated and the results are explained by a theory based on simple forms of facet and elastic energies. Applying advanced algorithm for the solution of the elastic problem, the 3D KMC simulations presented in [9] study the deposition of InAs on patterned GaAs(001) surfaces. In a recent work [10], the same model is used to describe important features of heteroepitaxial growth including intermixing, wetting layer, critical thickness, and apparent critical thickness. Other aspects of heteroepitaxial KMC modelling concern the influence of different growth parameters such us temperature, deposition rate, interruption time (the time given to the system to equilibrate after the deposition is terminated) [15], and substrate anisotropy [16] on QD characteristics.

The KMC model for the simulation of strain-induced QD formation at the same computational cost as for homoepitaxial growth. It is utilized by using solely two parameters: elastic correction ΔW_1 and the wetting barrier ΔE_γ which are described below. Nevertheless, as shown by investigations of self-assembly characteristics, the model can simulate to sufficient extent the self-assembly of QDs. This means that our investigation is not only devoted to the effect of strain on the 2D island morphology in the submonolayer growth but also to the entire 3D process of self-assembly even in long running processes. Including other parameters corresponding to more complex systems most probably will successfully model more diverse heteroepitaxial objects. Although the histograms of island sizes have been commonly accepted in previous similar studies we use different diagrams for almost all quantities of the system during the time evolution and the population of QDs at given surface coverage accompanied with appropriate relative standard deviations. Our model is described in detail in Section 2. In Section 3, it is supported by the results from our simulations of the system InAs/GaAs(001). We do not include interruption time in the simulations and concentrate mainly on the influence of the growth parameters on the surface patterns obtained from abruptly terminated growth. The first reason for our assumptions is that these patterns serve as prepatterns for further system equilibration in the period of growth

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interruption. On the other hand, reaching an equilibrium state in experimental conditions may take a long time, which is often far from the capabilities of the KMC method [17]. Conditions of abruptly terminated growth are met, for instance, if capping or cooling down of the system is applied so that the nonequilibrium state is frozen [18]. The growth conditions (temperature, deposition rate and coverage) used in our study correspond mainly to the kinetically driven regime of growth at small overcritical coverages which is of crucial importance for the total self-assembly process.

2. Description of the model

Our model is based on the solid-on-solid KMC model — individual atoms adsorb at a certain deposition rate F and occupy fixed sites in a simple cubic lattice of lateral size N lattice units (l.u.) with no overhangs. They are allowed to move discretely from the current site to one of its nearest neighbours. The rate F is defined as the number of atoms per time unit and per site, which arrive at their adsorption sites. Periodic boundary conditions are applied with respect to neighbours for energy barrier calculations and neighbours for moving. The important events included in the model are deposition and diffusion, while desorption is considered as negligible at the growth conditions used in the simulations [19]. The hopping rate of a surface atom is given by the Arrhenius law

$$R = \nu exp(-\Delta E/(k_B T)), \tag{1}$$

(ν being the attempt frequency, ΔE — hopping barrier, T — substrate temperature and k_B — Boltzmann's constant) [20]. The hopping barrier ΔE has several ingredients:

$$\Delta E = j_n E_n + j_{nn} E_{nn} + \Delta E_\gamma - \Delta W, \tag{2}$$

where the short-ranged interactions are based on the bond counting approach, that is, E_n and E_{nn} are the contributions from (in-plane and out-of-plane) nearest, resp. next-nearest neighbours and j_n, j_{nn} are the corresponding numbers of neighbours. The long-ranged elastic interactions due to strain are incorporated by using a simple modification of the ball-and-spring model which is explained below and is represented in the formula for the total energy barrier by *the elastic correction* ΔW. This elastic correction is applied when the atom is hopping up a step of height 1 monolayer (ML). According to the model assumptions, it depends only on the distance k (in l.u.) of the atom to the substrate and thus, will be further denoted by ΔW_k. *The wetting barrier* $\Delta E\gamma$ is an additional barrier for hopping down which originates from the wetting effects and is determined by the difference between the surface free energies of the deposited material and the substrate. In the present study, we assume that $\Delta E\gamma$ does not depend on the layer height, that is, the wetting layer is not taken into account (see Section 3.3).

The probability of a given configuration of the system at equilibrium is proportional to the time spent from the system in the configuration. This time is inversely proportional to the total rate R_{tot} for living this configuration:

$$R_{\text{tot}} = R_{\text{ads}} + R^{\uparrow} + R^{\downarrow} + R^{\rightleftarrows}, \tag{3}$$

where $R_{\text{ads}} = N^2 F$ is the total adsorption rate and

$$R^{\uparrow} = \nu \sum_{k=1}^{\infty} N_k^{\uparrow} exp\left(\frac{-\bar{j}_n^{\uparrow} E_n - \bar{j}_{nn}^{\uparrow} E_{nn} + \Delta W_k}{k_B T}\right), \tag{4}$$

$$R^{\downarrow} = \nu N^{\downarrow} exp\left(\frac{-\bar{j}_n^{\downarrow} E_n - \bar{j}_{nn}^{\downarrow} E_{nn} - \Delta E_\gamma}{k_B T}\right), \tag{5}$$

$$R^{\rightleftarrows} = \nu N^{\rightleftarrows} exp\left(\frac{-\bar{j}_n^{\rightleftarrows} E_n - \bar{j}_{nn}^{\rightleftarrows} E_{nn}}{k_B T}\right) \tag{6}$$

are the total rates for moving up, down and within the same layer, respectively. Here, $N_k^{\uparrow}$ is the number of possible hops of an atom from layer of height k to layer of height $k+1$ in the current state of the system; $N^{\uparrow} = \sum_k N_k^{\uparrow}$, $N^{\downarrow}$ and $N^{\rightleftarrows}$ are the total numbers of possible hops up, down and within the same layer, respectively; $\bar{j}_n^{\uparrow}$ ($\bar{j}_{nn}^{\uparrow}$), $\bar{j}_n^{\downarrow}$ ($\bar{j}_{nn}^{\downarrow}$) and $\bar{j}_n^{\rightleftarrows}$ ($\bar{j}_{nn}^{\rightleftarrows}$) are the average numbers of nearest (next-nearest) neighbours per a possible hop (average coordination numbers) in the corresponding directions.

The system evolves in time attempting to keep the value of R_{tot} as small as possible. If the elastic corrections $\{\Delta W_k\}$ are raised, it will try to reduce $N^{\uparrow}$, more hoppings up will be executed, and therefore, $N^{\downarrow}$ will be increased. The system will tend to grow in a 3D manner. The same effect is observed, if ΔE_γ is raised to a high enough value. And vice versa, if ΔE_γ is low enough, the system will not be able to compensate the excess of free energy by increasing $N^{\downarrow}$, more hoppings down will be performed, $N^{\uparrow}$ will be increased, and the value of R_{tot} will be lowered on account of the higher coordination numbers $\bar{j}_n^{\uparrow}$ and $\bar{j}_{nn}^{\uparrow}$. In particular, the conditions of $\Delta E_\gamma = 0$ (the case of full wetting) and small enough $\{\Delta W_k\}$ will favour the layer-by-layer growth mode.

The elastic corrections $\{\Delta W_k\}$ are calculated in the following way. Let α_s and α_f denote the bulk lattice parameters of the substrate and the deposited material, respectively. First, we obtain the vertical lattice spacing α_z of the film due to strain transformation of the lattice. It is supposed that every atom is connected with imaginary Hookean springs with its (up to 4) nearest lateral neighbours in the (x,y)-plane, (up to 2) nearest neighbours in the z-direction and (up to 12) next-nearest neighbours. Let k_1 and k_2 be the spring constants for lateral bonds and for diagonal bonds, correspondingly (we assume that the spring constants are the same for substrate and for film atoms). Now, consider the elastic energy stored in the springs connecting a given atom with its neighbours. The contribution of every lateral, resp. diagonal, neighbour in (x,y)-plane is equal to $1/2k_1(a_f - a_s)^2$, resp. $1/2k_2\left(\sqrt{2}a_f - \sqrt{2}a_s\right)^2$, for nearest neighbours in the z-direction $1/2k_1(a_f - a_z)^2$, and for out-of-plane next-nearest neighbours $1/2k_2\left(\sqrt{2}a_f - \sqrt{a_s^2 + a_z^2}\right)^2$. Further, we require having a mechanical equilibrium in a system consisting of a substrate and complete monolayers of deposited atoms. It is easily seen that (neglecting the local atom displacements) the total elastic energy of such a system is a sum of a term which does not depend on α_z and a multiple of the quantity

$$E_z = \frac{1}{2}k_1\left(a_f - a_z\right)^2 + 2k_2\left(\sqrt{2}a_f - \sqrt{a_s^2 + a_z^2}\right)^2. \tag{7}$$

Thus, the equilibrium state corresponds to the minimum of E_z with respect to the parameter α_z. Note that $a_f = (1+\varepsilon)a_s$, where ε is the *lattice mismatch* between the film and the substrate. Clearly, the minimum point of E_z depends only on the ratio k_1/k_2 but not on the absolute values of k_1 and k_2. Let us choose $k_1 = 2k_2$ which is in agreement with the elastic properties of one the most studied heteroepitaxial systems InAs/GaAs [9]. Then the value of $a_z = \delta a_s$ which minimizes E_z is determined by the unique positive solution δ of the equation

$$(3\delta - 1 - \varepsilon)\sqrt{1+\delta^2} = 2\sqrt{2}(1+\varepsilon)\delta. \tag{8}$$

Taking for example the system InAs/GaAs(001) with $\varepsilon = 0.07$, we get $\delta \approx 1.1045$, that is, $a_z = 1.1045 a_s$ in this case and the lattice is stretched in z-direction with further 3%.

Elastic correction ΔW_k contributes to the total energy barrier ΔE whenever the atom is attached to a step at height k and is attempting a hop to the upper terrace. Such a simplification of the heteroepitaxial KMC model is justified by the observation that the material supply to a growing island is governed by a ring-shaped area of compressive strain (see e.g. [21]). The calculation of ΔW_k is based on the assumption