

LATTICE GAS CELLULAR AUTOMATON MODELING OF SURFACE ROUGHENING IN HOMOEPITAXIAL GROWTH IN NANOWIRES

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Abstract

Our research addresses the problem of bridging large time and length scale gaps in simulating atomistic processes during thin film deposition. We introduce a new simulation approach based on a discrete description of atoms so that the unit length scale coincides with the atomic diameter. The interaction between atoms is defined using a coarse-grained approach to boost the computation speed. This approach does not heavily sacrifice the atomistic details in order to study structural evolution of a growing thin film on time scales in the order of seconds and even minutes. Our approach is inspired by lattice gas cellular automata models for chemically reacting systems, where individual particles interact with surrounding through assumed local driving forces. For homoepitaxial thin film deposition, the local driving force is the propensity of an atom to establish as many chemical bonds as possible to the underlying substrate atoms when it executes surface diffusion. Simulation results of Si layers deposited on a flat Si(001) substrate are presented.

Keywords: Computational materials science; surface growth; thin film deposition; lattice gas cellular automata.

1. INTRODUCTION

Bridging large time and length scale gaps in simulations of nanometer scale phenomena is an urgent issue in nanoscience and nanotechnology. While atomistic processes typically occur in picoseconds and have a characteristic length scale in nanometer, the typical sizes of final products, i.e., materials and devices, are

in micrometer or millimeter scale and their fabrication requires seconds, if not minutes, of various processings. This is best illustrated by deposition processes of semiconductor thin films. The main goal of these simulations is to understand atomistic processes leading to the formation of mesoscopic (10-1000 nm) structures—either bulk or surface—that serve as materials for various applications, ranging from lasers to single electron transistors.

The formation of mesoscopic structures is built upon various chemical bond breaking and formation events. Simulations based on quantum mechanical models produce a very accurate picture of these events. Despite this success, however, quantum mechanical (*ab initio*) simulations are unable to simulate systems with linear sizes around hundreds of nanometer (around 10^6 atoms in volume) due to increased computational complexity. Such large computational domains are necessary for realistically simulating thin film deposition and revealing interesting physics of structural evolution found in nanoclustering. Quantum mechanical simulations of thin film deposition also suffer from computational problems related to the very large time scale gap separating the atomistic processes in picoseconds or nanoseconds from the evolution of thin film morphologies in seconds or sometimes minutes. Although these problems are less severe in molecular dynamics simulations, in which interatomic potentials are assumed instead of calculated, the time and length scale gaps still persist since the molecular dynamics needs to evaluate repeatedly potential energy landscapes and their derivatives [1].

In this paper we introduce a new simulation approach based on a discrete description of atoms so that

the unit length scale coincides with the atomic diameter, but the interaction between atoms is defined using a coarse-grained description of surface forces. The goal of our simulation approach is to boost the computational speed and domain without heavily sacrificing the atomistic details in order to predict structural evolution of thin films on time scales of seconds and even minutes. This is done by placing the unit time scale in the nearest neighbour atomic jump time, which is around several microseconds for typical deposition conditions. Atomistic processes taking place faster than this unit time are therefore ignored.

Our approach is inspired by lattice gas cellular automata (LGCA) models for chemically reacting systems [2]. In the constructed LGCA for thin film deposition, the local driving force is given by the propensity of an atom to establish as many chemical bonds as possible to the underlying substrate atoms when it terminates its surface diffusion.

Our simulation approach is different from the kinetic Monte Carlo method in that (i) the unit time step in our simulation is explicitly determined by the nearest neighbour atomic jump, thus without assuming that atomistic processes having probabilities following a Poisson process [3], (ii) more than one adatom is allowed to diffuse on the surface at any given time, and (iii) surface atoms, be they adatoms or already incorporated into the film, always have finite probabilities to jump from their current sites. Despite these important differences, our approach shares with the kinetic Monte Carlo method the general idea that the local properties (such as chemical coordination, diffusion barrier, and presence of other nearby atoms) determine the direction the adatom will diffuse.

In this paper we shall focus on the deposition of Si atoms on a flat Si(001) substrate. Since both thin film and substrate are of identical chemical nature, there is no lattice constant mismatch between them, so that the thin film is assumed to store zero elastic energy. The homoepitaxial deposition of Si on Si(001) is commonly classified under the Frank-van der Merwe (FM) growth mode, where it is expected that the deposition will mainly proceed by a layer-by-layer fashion if the arriving Si atoms have enough chance to equilibrate by finding the minimum energy surface sites.

2. LGCA MODEL OF SURFACE ROUGHENING

In simulating the Si atoms deposited on a flat Si(001) substrate, we will use a one-dimensional lattice of length L with periodic boundary conditions. The lattice sites are denoted by $r = 0, 1, \dots, L - 1$; and the simulation time is discrete and indexed by $k = 0, 1, \dots$

The state of an individual site r at time k is described by the integer-valued variable $h(r, k)$, which represents the site's film height (thickness). We initialise the LGCA with the values $h(r, 0)$ for all r at time $k = 0$. To proceed with the deposition simulation, we require a sequence of uniformly distributed random integers, which determines where the Si atoms will land [4].

The state $h(r, k) \geq 0$ represents the height, i.e. number of one atomic-height layers, of the growth structure on the lattice at site r and time k . If $h(r, k) > 0$ then the particle at the topmost layer at site r is called the adatom at site r . These adatoms are thus precisely given by those at $h(r, k)$. All deposited atoms at site r below the adatom are called bulk atoms. Once an adatom becomes a bulk atom by virtue of the presence of a freshly landing atom above the adatom, then this bulk atom is not allowed to move anymore. We thus assume that only adatoms are mobile and can move to neighbouring lattice sites. The rules that describe their motion will be outlined below.

Since the topmost layer atoms are always mobile with different probabilities, we shall define the layer thickness function $H(r, k)$ that defines the volume of the bulk atoms:

$$H(r, k) = \max\{0, h(r, k) - 1\}. \quad (1)$$

The time evolution of the LGCA from time k to time $k + 1$ proceeds in three sub-steps: (i) particle landing, (ii) adatom velocity computation, and (iii) adatom motion. Each sub-step is performed at each lattice site r independently from the other lattice sites and the next sub-step is started only after the previous sub-step has been completed on all sites of the lattice. We introduce the intermediate time steps k' and k'' , which are reached when the first and the second sub-steps are completed, respectively.

In order to connect the LGCA to realistic surface growth processes, we must relate the simulation time k to the real deposition time t . We assume that each simulation step from k to $k + 1$ corresponds to an effective time step denoted by Δt_{eff} , leading to $t = k\Delta t_{eff}$. The effective time step Δt_{eff} corresponds to the average time taken by adatoms to jump to the nearest atomic site (including the waiting time before the actual hop). To achieve computational efficiency, we fix Δt_{eff} to a constant value given by

$$\Delta t_{eff}^{-1} = \frac{k_B T}{2\pi\hbar} \exp\left(-\frac{E_c}{k_B T}\right), \quad (2)$$

where k_B is the Boltzmann constant, $\hbar$ is the Planck constant divided by 2π , E_c is the surface corrugation (potential) energy E_c for a particular material and T is the growth temperature.

2.1 Adatom Landing

The deposition of adatoms on the surface is modeled by the adatom landing sub-step. We assume that there is a constant and spatially homogeneous flux of atoms, from which adatoms are randomly deposited on the surface. The probability p_l of adatom landing per unit simulation time and lattice site is directly related to the experimental growth rate through the simple fact that the number of atoms in the simulation must match the number of atoms in experiments. During the course of the simulations, we found that it is computationally inefficient to perform a random trial for each lattice site; such an algorithm would require L random trials per time step. Another difficulty is the very small value of p_l , which can go as low as 10^{-9} . To overcome both computational hurdles, we boost the probability of landing by assuming that at most one adatom can land on the entire 1D surface in each execution of the adatom landing sub-step. This landing probability is now equal to Lp_l , and we define a uniform random variable that determines where the adatom, if created, will land. Thus, we require only two random number generations. Once the adatom lands on a particular lattice site r , we increase the height at that site by unity. The surface height after this sub-step is denoted by $h(r, k')$.

2.2 Adatom Velocity Computation

Adatom decides the direction of motion during its surface diffusion based on the surface morphology. We also allow for the adatom to stay at its current site. To be consistent, we therefore allow for all atoms at the topmost layer to have finite probabilities to move. To affect the motion of an adatom initially at r , we define a velocity function:

$$\nu(r, k'') = \begin{cases} -1, & \text{the adatom moves to site } (r-1) \\ 0, & \text{the adatom stays} \\ 1, & \text{the adatom moves to site } (r+1) \end{cases} \quad (3)$$

The velocity $\nu(r, k'')$ is computed as a function of $h(r, k')$ and the heights of the neighbouring sites through the probabilities of executing these three motion options. To do this we define a set of independent random variables $\{\xi_\nu(r, k') | r = 0, 1, \dots, L-1\}$ with a range $\{-1, 0, 1\}$ such that

$$\begin{aligned} P[\xi_\nu(r, k') = -1] &= p^-(r, k'), \\ P[\xi_\nu(r, k') = 1] &= p^+(r, k'), \\ P[\xi_\nu(r, k') = 0] &= p^0 = 1 - p^-(r, k') - p^+(r, k'), \end{aligned} \quad (4)$$

where $P[\cdot]$ denotes the probability of its argument, and the diffusion probabilities: $p^+(r, k')$, $p^-(r, k')$, p^0 , depend on the surrounding surface morphology of the site r . The physical content of our simulation lies in the determination of these diffusion probabilities. These

probabilities depend exponentially on the difference between the number of bonds an adatom can make at the current site and the corresponding number when it moves to the left ($r-1$) or to the right ($r+1$). The determination of the number of bonds includes the presence of nearby adatoms. This consideration is an important distinction with the standard kinetic Monte Carlo simulation that considers only diffusion of one adatom at any given time.

2.3 Adatom Motion

Once the decision of the adatom motion direction is established, the height at the site r is subtracted by unity and the height of the site where the adatom is moving to is increased by unity. We do not restrict that possibly two adatoms move to a lattice site; in such a case, these two adatoms will be on top of each other.

3. Si/Si(001) DEPOSITION

We performed simulations of Si layers deposited on a flat Si(001) substrate. The parameters needed to run the simulations are as follows: (i) surface corrugation energy for Si(001) surfaces is 0.7 eV ($\approx 1.12 \times 10^{-19}$ Joule) [5]; (ii) unit broken (dangling) chemical bond energy is 2.3 eV [6]; (iii) lattice size is 500; and (iv) $h(r, 0) = 0$ for an initial condition of a flat substrate.

There are 4 simulation results reported here, which correspond to two growth rate values: $R_g \in \{0.1, 10\}$ monolayer (ML) per second, and two deposition temperatures: $T \in \{700, 800\}$ K. The final simulation time is chosen such that on average 5 monolayers were grown, i.e., $t_{end} = 50, 0.5$ seconds for $R_g = 0.1, 10$ ML per second, respectively. If we want to grow on average x monolayers with a growth rate R_g , then we must run the simulation to $k \approx x R_g^{-1} \Delta t_{eff}^{-1}$.

In all our simulations we observed that the average height $\bar{h}(k) := (1/L) \sum_{r=0}^{L-1} h(r, k)$ as a function of time k almost perfectly agrees with the expected growth $R_g k \Delta t_{eff}$ of the structure.

We analysed the surface roughening evolution by evaluating the interface width:

$$w(L, k) = \sqrt{\frac{1}{L} \sum_{r=0}^{L-1} [h(r, k)^2 - \bar{h}(k)^2]}. \quad (5)$$

$w(L, k)$ measures the roughness of the evolving thin film surface. For early times $t < t_1$, $w(L, k)$ behaves independently of temperature T for a constant R_g . Fig. 1, shows the graphs of the interface widths for the four cases. By comparing directly these graphs, we observe that the interface width decreases with increased growth rate R_g at a constant temperature T .

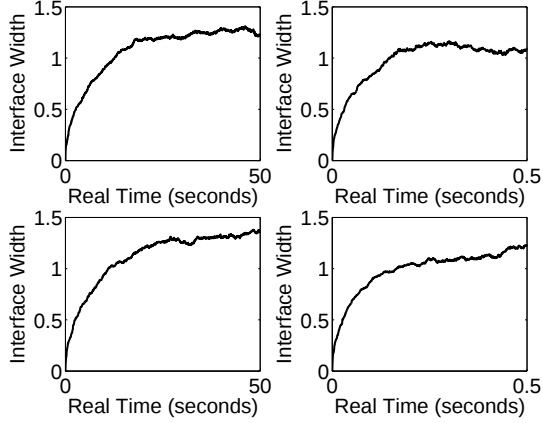


Fig. 1. Growth temperature is 700 K (top row) and 800 K (bottom row); growth rate is 0.1 ML/s (left column) and 10 ML/s (right column).

This “kinetic stabilisation” behaviour is commonly observed in homoepitaxial deposition, in which a higher growth rate can suppress surface roughening. Physically, this can be explained by the dominance of the uniformly advancing growth front due to the deposition flux in comparison to the processes that control the adatom diffusion [7].

We also computed the step density as defined by

$$\rho_{step}(k) = \frac{1}{2L} \sum_{r=0}^{L-1} \sum_{r' \in \{r-1, r+1\}} [1 - \delta_{h(r,k), h(r',k)}], \quad (6)$$

where $\delta_{j,k}$ is the Kronecker δ -function, which is equal to unity if $i = j$ and is equal to zero otherwise. The step density ρ_{step} above precisely measures the number of surface height discontinuities, i.e., steps. The step density of a flat surface is zero, whereas the step density of a surface that has no two neighbouring sites of the same height is equal to unity. Fig. 2, shows the evolution of the step density for the four cases. At $T = 700K$, the growth rate of 10 ML/s induces a faster step density increase, although at the same time reaches a stable value faster than the 0.1 ML/s growth rate does. This behaviour does not occur at 800 K; instead, both growth rates produce almost identical step density evolutions. Increased temperature increases the rate of adatom diffusion jump and allows a greater mobility for the adatoms. The equilibrium adatom density on the surface at 800 K is thus dominated by the diffusing surface atoms, rather than by the arriving atoms from the deposition flux. This behaviour was observed for GaAs homoepitaxy [8].

4. SUMMARY

In summary, we have introduced a new method to simulate thin film deposition based on LGCA. The

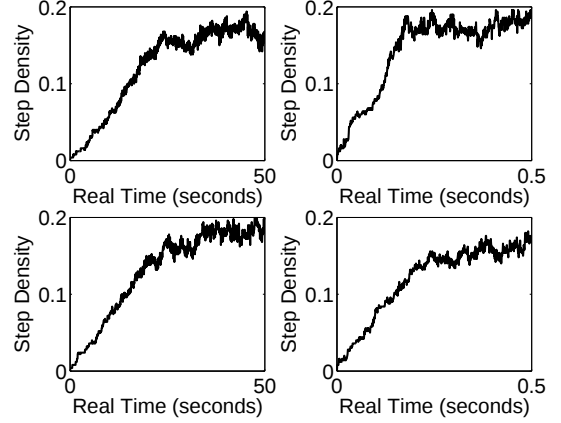


Fig. 2. Growth temperature and rate are as in Fig. 1.

Si/Si(001) simulations we performed have produced consistent results with what have been observed experimentally. The number of parameters required to run the simulations is minimal; thus our method is quite appropriate for independently evaluating experimental results and, importantly, for predicting the structural evolution of various semiconductor homoepitaxial thin film systems.

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