Simulation of islands and vacancy structures for Si/Ge-covered Si(001) using a hybrid MC-MD algorithm

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Abstract. A classical hybrid MC-MD algorithm is applied to study physically interesting phenomena, such as island stability and formation of dimer-vacancy structures, on Si/Ge-covered Si(001). The method introduces collective moves into the standard MC algorithm to allow the system to escape from metastable states. Simulation results are found to be in good qualitative agreement with experiments.

1 Introduction

Understanding the properties of thin layers on semiconductor surfaces is crucial for the development of new fabrication methods for a wide variety of nano-structure and optoelectronics applications. The Ge/Si(001) system is an ideal model for studies of heteroepitaxial growth where structural modifications, such as step-edge evolution or formation of defect structures, play a central role in determining the large-scale morphology of the surface.

Structural changes often involve propagation of long-range elastic effects. Studying such phenomena requires not only sufficiently large system sizes but also efficient simulation methods which can produce large configurational changes in the system. Semiconductor surfaces are often characterized by extremely complicated energy landscapes. In simulations of such systems, conventional algorithms become inefficient in dealing with long time-scale processes associated with escape from metastable states.

In order to overcome these problems, we have developed a new hybrid Monte Carlo - Molecular Dynamics algorithm which uses collective moves of several atoms to produce large-scale changes in the system. The version presented here is intended for the study of Si(001) systems, but the idea itself is quite general and could potentially be applied to other semiconductor surfaces. In the current version, the method is based on introducing long-range dimer displacements into the standard MC algorithm which significantly speed up the evolution of the system by allowing the system to overcome the large energy barriers associated with dimer diffusion. In this paper, we apply this method to study two physically interesting phenomena, namely the relaxation of islands and the formation of vacancy structures.

2 Simulation method

In this work, off-lattice constant-pressure Monte Carlo simulations [1] are used to model temperature-dependent structural properties of the Si- or Ge-covered Si(001) surface. The Si-Si, Si-Ge and Ge-Ge interactions are modeled using the classical Stillinger-Weber (SW) [2,3] potential which we have recently tested for use in finite-temperature simulations involving the Si(001) surface [4]. The system is modeled using slab geometry such that the unit cell is constructed along the [110], [1$\bar{1}$0] and [001] directions and periodic boundaries are applied in the x and y directions. The simulation slab consists of 20 atomic layers with the surface size varying from 20×20 to 40×40 atoms. The method is not limited to these system sizes but can be used to treat much larger systems (over 10^5 atoms).

In the standard constant-pressure Metropolis MC algorithm, two kinds of trial moves are attempted: small random displacements of individual particles (*single-atom moves*) and random variations of the sides of the simulation cell (*volume variations*) in order to keep the pressure constant at $P = 0$. In both cases, the acceptance probability is given by the Metropolis form [5]. More details can be found from Refs. [3,4].

The Si(001) surface consists of rows of dimerized atoms (2×1 reconstruction) which induces significant displacements of the atoms in the underlying layers. The potential barrier for a jump of a single dimer from one binding site to a neighboring site is extremely high, and therefore such events never occur if only small displacements of individual atoms are attempted in the simulation. Moreover, breaking a dimer bond would require a large amount of energy, which means that the two atoms must move simultaneously without breaking the bond. For this reason, traditional algorithms become inefficient in relaxing the system.

In order to overcome these problems we have developed a *hybrid MC-MD algorithm* [6,7] which introduces large dimer displacements to circumvent the high potential barrier between two binding sites. In this so-called *dimer-jump algorithm*, two atoms comprising a dimer undergo the same displacement (translation and a small rotation) which typically allows the particles to move a distance comparable to the Si lattice constant ($a_{0,Si} = 5.43$ Å).

Let us now consider the arrangement of atoms in the underlying layers. Fig. 1 shows a snapshot of a typical step edge on Si(001). We see that the four nearest atoms under a dimer have relaxed to near-tetrahedral positions, whereas those atoms which are not directly under the step are themselves dimerized. If we now displace one of the upper-layer dimers by an amount corresponding to the average distance between binding sites, the dimer lands in a position where its nearest neighbors are either too close or too distant. Moreover, the configuration at the initial site has become unfavorable to those atoms which were previously under the dimer. Consequently, the dimer jump will result in a very high-energy configuration which will almost certainly be rejected in the Metropolis trial.

In order to achieve a much better acceptance rate, we have introduced a scheme in which the the local environments around the initial and the landing site are relaxed prior to the acceptance test. The dimer itself is also included in this group of atoms. The relaxation is performed using standard Molecular Dynamics with Velocity Verlet for integrating the equations of motion. Initial velocities are assigned from the Maxwell-Boltzmann distribution at the simulation temperature and constant-temperature conditions are maintained using velocity rescaling. In most cases, a relaxation of 7-10 MD steps using a time step of 10^{-15} s is enough for achieving a good acceptance rate.

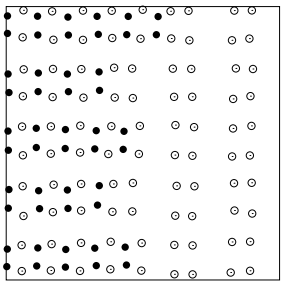

Fig. 1. Snapshot of a typical S_{B} step edge on the Si(001) surface after relaxation at 700 K. The open and solid circles are lower and upper terrace atoms, respectively.

3 Relaxation of islands and step edges

As the first application example of the method described above, we investigate the relaxation of islands and step edges on Si(001). We begin the simulations with a square Si island on the Si(001) surface, which enables us to study both the S_{A} and S_{B} steps at the same time (see Fig. 2 for an illustration of the different step edges). The system is allowed to evolve using the dimer-jump algorithm until equilibrium is reached, and no further qualitative changes in the configuration are then observed.

Figure 2 shows typical snapshots of an island at two different temperatures. Looking first at the island on the left, we notice that the shape of the island has changed to a slightly anisotropic form. The S_{A} and S_{B} steps have evolved in a very different manner: the S_{A} steps are longer and have a smooth appearance, whereas the S_{B} steps are clearly more rough and irregular. This behavior is consistent with STM images of Si islands and step edges which all show very similar features as observed in our simulations (see e.g. [8,9]).

Comparison of the two figures at 700 K and 930 K shows that island stability is clearly dependent on temperature. At 700 K, the island changes its shape during equilibration but stays compact and well defined for very long

simulation times. In contrast, when $T = 930$ K, the island dissociates very quickly. If we reduce the initial island size, we observe that the dissociation takes place at much lower temperatures (see e.g. Refs.[6,7]). These observations indicate that there exists a temperature-dependent critical size beyond which islands are stable against dissociation and smaller islands will decay. It is not the aim of this work to address the subject in detail, but from these results it is evident that the algorithm is suitable for such an investigation.

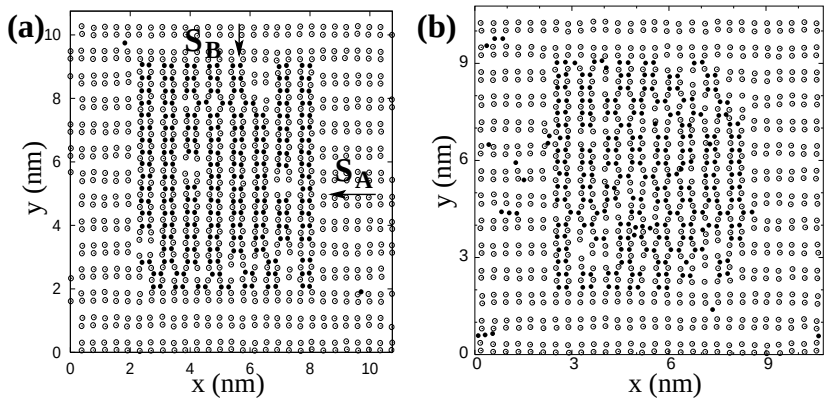

Fig. 2. Snapshots of typical Si islands on Si(001). (a) $T = 700$ K, after 7×10^4 MCS. (b) $T = 930$ K, after 4×10^3 MCS. The solid circles are adatoms and the open circles are atoms in the first surface layer.

4 Formation of vacancy structures

Experimental studies have produced consistent evidence that dimer vacancies (DV) are intrinsic defects of the Si(001) surface [10]. They are known to cluster into distinctive complexes (e.g. 2, 2+1, 3 DVs etc.) [11], and under certain conditions, elongated vacancy islands are also observed to form. In the case of Ge growth on Si(001), an ordered vacancy-line pattern forms spontaneously on the surface and the periodicity of the pattern depends on the thickness of the Ge layer [12]. This so-called $2 \times n$ reconstruction has a profound influence on the large-scale morphology of the surface, leading to e.g. a reversal of the step-edge roughness and surface stress anisotropy.

We began to investigate the vacancy-line formation by comparing the relative energy of different $2 \times n$ reconstructions. The details of the calculations are given in Ref. [13]. The results show that the $2 \times n$ pattern is stabilized by the Ge layer and increasing Ge coverage leads to a decrease in the optimal spacing between the vacancy lines. Our results are in excellent agreement with experimental observations, which confirms that our classical

model seems to capture the essential features of dimer vacancies in Si(001) surfaces. As the next step, it is interesting to ask whether the ordering process can be simulated using the dimer-jump algorithm.

Figure 3 shows two snapshots from simulations using the dimer-jump algorithm for a pure Si(001) surface and for a surface covered with 2 ML of Ge. In both cases, we used the same initial configuration which consists of a random distribution of dimer vacancies. All dimers are allowed to move on the surface during the simulation, which in effect means that the vacancies migrate. We notice that on the clean Si(001) surface, the vacancies show a tendency to cluster together but the overall arrangement is random. On the Ge-covered surface, on the other hand, we observe that clear segments of vacancy lines are forming (one line passes through the whole system and two shorter segments have also developed). Once formed, the vacancy lines are very stable and do not dissociate.

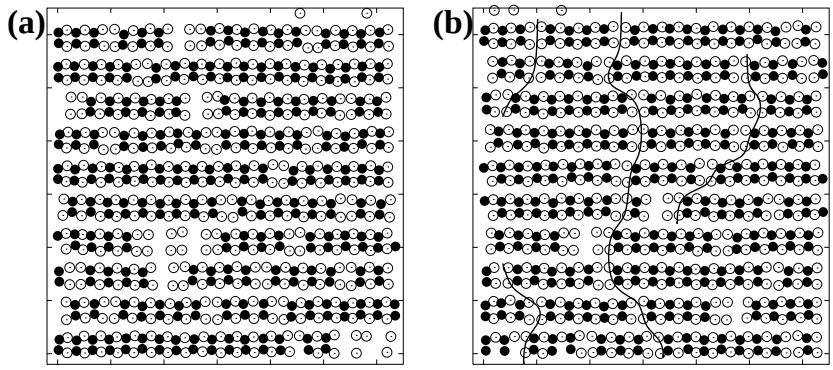

Fig. 3. Formation of dimer vacancy structures at $T = 930$ K. (a) Clean Si(001). (b) 2 ML of Ge on Si(001). In both cases, the initial configuration was a random distribution of vacancies.

The drawback in these particular simulations is that we were forced to use a rather high temperature (930 K) in order to have sufficiently many dimer jumps accepted within a reasonable simulation time. However, the equilibrium shape of the vacancy lines is expected to be quite kinked and disordered at this temperature. Therefore, an approach such as parallel tempering (see e.g. [14]) could be useful in speeding up the equilibration at lower temperatures. Nevertheless, even these first results are promising and suggest that the dimer-jump algorithm gives the possibility to perform atomic-scale simulations of large systems such that experimentally accessible length scales are within reach.

5 Conclusion

We have used a newly developed hybrid MC-MD algorithm to study the relaxation of 2D Si islands on Si(001) and the formation of vacancy structures on clean and Ge-covered Si(001). The method is designed to overcome problems related to long time and length scales in physical phenomena occurring on semiconductor surfaces. This approach has the advantages of being suitable to off-lattice simulations and utilizing bulk-fitted potentials without the introduction of any additional parameters. The simulation results are in good qualitative agreement with experiments, and the investigation suggests that the algorithm is also suitable for quantitative studies.

Acknowledgments

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