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An atomic-scale study of hydrogenated silicon cluster deposition on a crystalline silicon surface

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ABSTRACT

Controlled deposition of clusters on solid surfaces has attracted lots of attention in recent years, because of its potential application to tailoring the desired electronic properties of the resulting surfaces. We have carried out an atomic-scale study to understand the deposition mechanism. The molecular dynamics approach based on a modified Tersoff potential is used to simulate the deposition mechanism of hydrogenated silicon clusters on a crystalline silicon surface in detail. The important factors governing the deposition process such as impact energy and substrate temperature, are investigated for the hydrogenated silicon cluster $Si_{29}H_{24}$ on a H-terminated Si(100)-(2x1) surface.

1. Introduction

The controlled deposition of clusters on solid surfaces has attracted a lot of attention in recent years due to its potential application in tailoring the electronic properties of the resulting thin film. In particular, two types of deposition mechanisms are attractive: the destructive deposition of clusters and the soft-landing of cluster. Destructive deposition, e.g., ionized cluster deposition, holds promise in synthesizing high quality thin films at low temperatures. In this mechanism, the impact energy of the cluster is transferred to the fragmentary cluster atoms, thus becoming migration energy, and leading to high atomic mobility on the surface even at low temperatures [1]. In soft-landing deposition, the cluster is not destroyed upon landing on the substrate. This type of deposition is equally promising, as clusters exhibit interesting quantum properties [2,3] that are sensitive to their size and bonding states, and which can therefore be controlled. In both cases, the deposition of clusters instead of individual atoms or molecules attracts industrial applications due to its inherent possibility of high-speed thin film deposition.

In this work, we used model potential molecular dynamics simulations to study the controlled deposition of clusters on solid surfaces in order to understand the growth mechanisms and conditions. We begin our research with the simulation of the deposition of hydrogenated silicon clusters under low temperature plasma-enhanced chemical vapor deposition conditions.

2. Computational methods

To simulate the nanoparticle-surface interaction, the Ohira–Tersoff (OT) potential was employed as it has been shown to have a good performance in describing the structure and properties of amorphous Si (*a*-Si) bulk phases [4] and c-Si surfaces [5], and because it has been successfully used to study the radical–surface interactions [6–8], *a*-Si thin film growth mechanisms [9–11], and the H-induced crystallization of *a*-Si thin films [12–14]. The potential function form is described in [15]. The performance of the OT potential in describing the structure and properties of c-Si bulk is given in Table 1, which shows that the results of the OT potential are in good agreement with *ab initio* calculation results.

In this study, the filled fullerene stable configuration $Si_{29}H_{24}$ shown in Fig. 1 has been chosen to be the impinging cluster, as the structural and optical properties of this 1 nm nanoparticle have been studied and compared to theoretical and experimental findings [17,18]. The H-terminated Si(100)-(2x1) surface is used as the substrate, which consists of 288 silicon atoms (see Fig. 2). The dimension of this substrate is 21.72 Å × 21.72 Å × 10.86 Å with 2 dimensional periodic boundary conditions (PBC) applied in the surface plane. The top silicon layer is a reconstructed layer consisting of silicon dimers. The bottom two layers are kept rigid in their equilibrium positions to avoid translation of the substrate in space.

The substrate temperature is controlled by using a Berendsen thermostat [19] in the three layers above the rigid layers. These three layers also act as a heat reservoir which absorbs the heat during the collision. We used two substrate temperatures which represent the two extreme values employed in low temperature PECVD: 373 K and 573 K. The surface was heated up to the target temperature. Then, the surface temperature was maintained until the surface structure reached its equilibrium state, which took about 10 ps. Afterwards, the deposition of the $Si_{29}H_{24}$ cluster on the substrate was initiated. The

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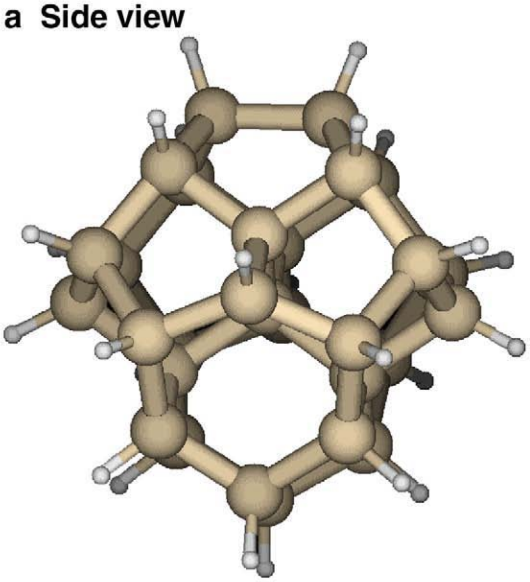
Table 1
Comparison of the energies and structures of c-Si and H-terminated Si(100)-(2x1) predicted by the OT potential with *ab initio* results.

c-Si	OT	*ab initio* [16]
Bulk cohesive energy (eV/atom)	4.6296	4.63
Lattice parameter (Å)	5.4320	5.43
H-terminated Si(100)-(2x1)	OT	*ab initio* [6]
Dimer bond length (Å)	2.44	2.4
H–Si–Si bond angle (°)	110.2	114.7

impact energies were selected from our results using the hard surface collision model [20]. We only employed the hard surface collision model to simulate the elastic scattering of the cluster from a hard surface, in order to obtain quantitative information about the cluster deformation energy.

3. Results and discussion

Guided by the results of our hard surface collision simulations, we selected three different impact energies: 0.3 eV/atom, 2 eV/atom and 5 eV/atom.



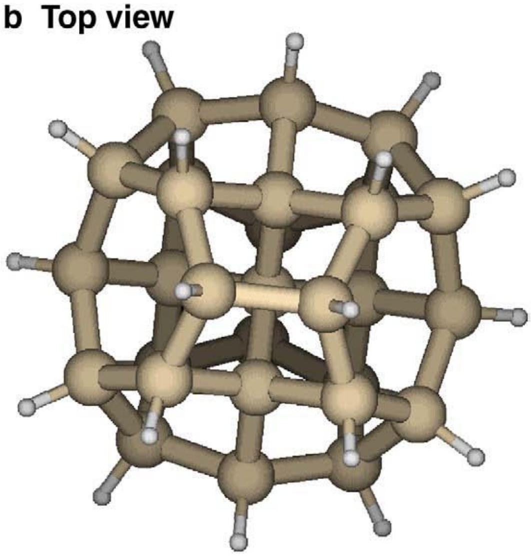


Fig. 1. The initial configuration of the hydrogenated silicon cluster $Si_{29}H_{24}$. Big balls represent silicon atoms while small ones represent hydrogen atoms.

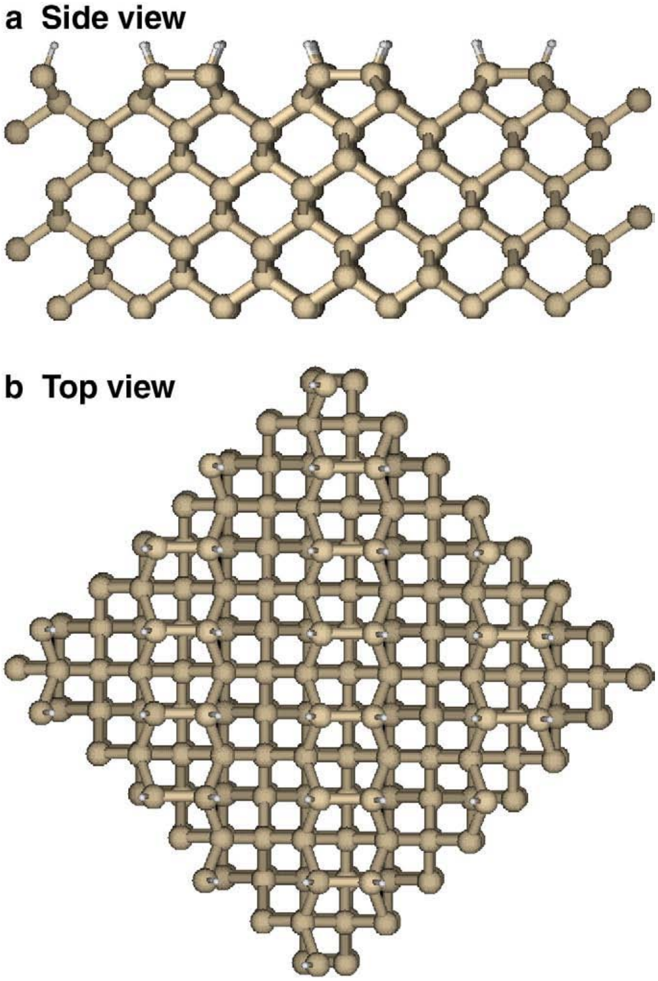


Fig. 2. The initial configuration of the H-terminated Si(100)-(2x1) surface. The big balls represent silicon atoms while the small ones represent hydrogen atoms.

Figs. 3 and 4 demonstrate the results of collisions for different impact energies at substrate temperatures of 373 K and 573 K, respectively. The deposition mechanisms are quite similar for those two substrate temperatures. When the impact energy is low (0.3 eV/atom), the cluster remains intact on the surface; with a medium impact energy (2 eV/atom), the cluster partially dissociates and spreads out on the substrate; while at a high impact energy (5 eV/atom), not only does the cluster dissociate, but it also penetrates into the surface. At this high impact energy, the deposition mechanisms are slightly different for the two substrate temperatures: at the higher substrate temperature, the cluster penetrates deeper into the substrate.

Biswas et al. [21] suggested that epitaxial growth requires that each impinging cluster dissociates and spreads in a uniform layer on the substrate. Therefore, we chose the collision at a medium impact energy where the cluster deforms and spreads out on the surface for more detailed study. As there is no significant difference at the medium impact energy for the two investigated temperatures, the low temperature case was chosen for illustration. Figs. 5 and 6 show the results obtained from the cluster-surface collision at an impact energy of 2 eV/atom and a surface temperature of 373 K. Fig. 5 demonstrates the structural evolution of the cluster during the collision. Fig. 6(a) displays the time evolution of the instantaneous temperature of the first silicon layer and Fig. 6(b) shows the temporal spread-out of the kinetic energy for this layer. Combining the results of Figs. 5 and 6(a), we see that the temperature of the first silicon layer increases