

## Diffusion of Si and Ge dimers on Ge (001) surfaces

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We have studied the various diffusion pathways of Si and Ge dimers on the Ge (001) surface using scanning tunneling microscopy. The adsorbed dimers can be classified into two categories: Dimers adsorbed on top of the substrate rows and dimers adsorbed in the troughs between the substrate rows. There are three different diffusion pathways for the dimers: Along the substrate rows, across the substrate rows, and in the troughs between the substrate rows. The activation barriers for diffusion of these three pathways have been determined for both Ge and Si dimers on Ge (001). The barriers for dimer diffusion of the system Ge/Ge (001) are slightly lower than for the Si/Ge (001) system. As compared to Si on Si (001) the activation barriers for dimer diffusion on Ge (001) are significantly lower. © 2003 American Institute of Physics. [DOI: 10.1063/1.1533107]

### I. INTRODUCTION

The diffusion of atoms on a solid surface is a fundamental problem in surface science, one that has attracted attention over many years. Imaging techniques with atomic resolution, such as field ion microscopy and scanning tunneling microscopy (STM), have provided us a remarkable look at the variety of ways in which atoms and small molecules diffuse on surfaces. Thermally activated diffusion from substrate site to substrate site can occur either via a simple hop mechanism or via an exchange mechanism. Atomic exchange refers to a process in which an atom on top of a surface replaces an atom in the first layer of the surface, while the replaced atom moves to the top.

When the diffusing species is a dimer, i.e., a cluster of two atoms, rather than a single atom the variety of diffusion processes that can be observed is enhanced. For instance, dimers can diffuse as a two-atom unit or one atom at a time. In the latter process the two atoms may come back immediately after dissociating or it is also possible that they will move away from each other and continue to perform simultaneous random walks until they meet each other (or another atom) and form a dimer again.

A detailed knowledge of diffusion is a prerequisite for the understanding many surface processes, among them, for instance are crystal growth techniques such as molecular-beam epitaxy, and chemical-vapor deposition. These modern growth techniques have made possible the production of solid-state devices that have revolutionized the electronic and computer industries. Device applications have in turn fueled a continuing effort, whose aim is to understand and ultimately control the physics of growth processes, especially for the important case of semiconductor materials. During the last decade silicon-germanium growth has re-

ceived much attention. Silicon-germanium heterostructures allow the creation of a new class of materials based on modulated band-gap engineering. However, the lattice mismatch between silicon and germanium gives rise to strain-induced roughening of the semiconductor film, which is a major factor limiting the fabrication of coherently strained multilayers. On the other hand, the strain-induced roughening is of benefit for the self assembly of quantum dots where the interest is to create regularly spaced three-dimensional islands of uniform size.

In an attempt to understand the silicon-germanium epitaxial growth in more detail the homoepitaxial growth of Si (001) has often served as a model system. Interestingly, the epitaxial growth of Si and Ge on the closely related Ge (001) surface received much less attention. This is surprising because from the viewpoint of surface perfection the epitaxial growth of Ge (001) turns out to be a much better model system than Si (001).<sup>1</sup> It is common knowledge that at room temperature the semiconductor group IV(001) surfaces, i.e., silicon and germanium (001), exhibit a  $(2 \times 1)$  reconstruction: Surface atoms dimerize, eliminating one dangling bond per atom to lower the surface free energy. Along with the dimerization goes the development of an anisotropic surface stress tensor. The reconstructed  $(2 \times 1)$  surface is under a compressive stress perpendicular to the dimer bond and under a tensile stress along the dimer bond. This dimerization was proposed by Schlier and Fansworth<sup>2</sup> in the late 1950s and in 1985 experimentally confirmed for silicon (001) by scanning tunneling microscopy.<sup>3</sup> Two different dimers are observed: symmetric dimers and asymmetric dimers. The asymmetric dimer has one atom that buckles out of the surface plane and one atom that buckles inward. The symmetric appearing dimers buckle very rapidly between the two buckled configurations. At room temperature the Si (001) surface exhibits the  $(2 \times 1)$  reconstruction. The state with the lowest-surface free energy at room temperature of Ge (001), is not a

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simple ( $2 \times 1$ ) reconstruction but an ordered striped pattern consisting of ( $2 \times 1$ ) and  $c$  ( $4 \times 2$ ) domains.<sup>4</sup> In retrospect, the occurrence of this structure is not surprising at all, but can be explained in the framework of a well-established strain relaxation theory.<sup>5</sup> The reason why this structure has not been observed before so far is that it requires extremely clean and defect-free surfaces. For the Si (001) surface it is inherently difficult to prepare clean surfaces with defect densities low enough for this pattern to develop. For the Ge (001) surface, however, this requirement can be met by extremely careful preparation procedures.

It is the aim of the present article to study in real space the diffusion of Si and Ge dimers on the clean Ge (001) surface. A comparison with the well-known system Si/Si (001) will be made. We will show that for these systems three diffusion pathways are active: Diffusion of on-top dimers over the substrate rows, diffusion across the substrate rows, and diffusion in the troughs. The barriers for diffusion of Si and Ge dimers on Ge (001) are both significantly lower as compared to Si/Si(001).

## II. EXPERIMENT

The experiments were performed in an ultrahigh vacuum system equipped with a scanning tunneling microscope (STM). The base pressure of the system is  $\sim 5 \times 10^{-11}$  mbar. The nearly intrinsic and nominal flat Ge (001) samples were cleaned by various cycles of 800 eV Ar<sup>+</sup> sputtering and annealing at 1100 K. After equilibration to room temperature  $\sim 0.5$ –1% of a monolayer of Ge or Si was deposited from a thoroughly degassed, heated wafers at  $\approx 3 \times 10^{-4}$  monolayers per second (ML/s). Subsequently, the samples were transferred to the STM for imaging. The complete procedure of deposition and transferring into STM takes about 10 to 15 min. The STM experiments were performed in the temperature range from 300 K up to at maximum 400 K.

## III. RESULTS AND DISCUSSION

### A. Adsorption sites

In this work, a large number of STM images, taken after room temperature deposition of Si or Ge atoms on the Ge (001) surface, have been analyzed. The smallest observed entities turn out to be ad-dimers, i.e., bonded pairs of adatoms. The reason that no isolated Si or Ge monomers are found is because they are very mobile on the Ge (001) surface. The adsorbed dimers can be divided in two classes: Dimers residing on top of the substrate dimer rows and dimers residing in the trough between the substrate rows. In Fig. 1, a schematic diagram of the various plausible adsorption sites for an isolated dimer is shown. The on-top dimers have their dimer bonds aligned either along the substrate dimer bonds (A configuration) or perpendicular to the substrate dimer bonds (B configuration). The trough dimers can also appear in two configurations. The trough dimer bond can be aligned along the substrate dimer bonds (C configuration) or perpendicular to it (D configuration). The on-top dimers are easy to recognize because the dimers appear bright in filled-state and empty-state images. The C-type

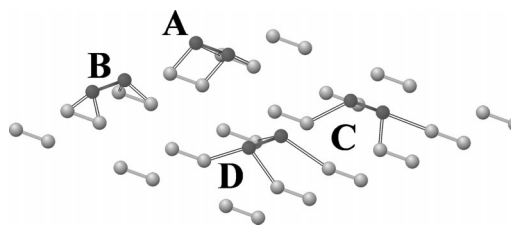


FIG. 1. Schematic diagram of ad-dimer adsorption sites on Ge (001). There are two stable adsorption sites on top of the substrate rows (A and B) and two adsorption sites in the troughs (C and D). The intermediate configuration labeled A/B is not shown.

trough dimer, however, appears only bright in empty state images. Experimentally most of these dimer structures prove to be accessible kinetically and they can be observed in STM (Table I). From a population analysis one can, in principle, establish their relative energies. Because of the kinetic barriers involved in the various diffusion processes, it can however be very difficult to achieve equilibrium on a surface. Increasing the temperature to overcome these barriers does not always help since the dimers then start to nucleate and form islands. The calculated energy differences (see Table II) of the Si/Ge dimers on Si or Ge (001) exhibits the same overall trend.<sup>6</sup> In all cases, the B-type structure on top of a substrate dimer row is lowest in energy. The “on-top” A and “trough” C structures are somewhat higher in energy. The “trough” D structure is substantially higher in energy for all systems. For the system Si/Si (001) isolated A, B, and C-type dimers are observed at room temperature.<sup>7–11</sup> For the system Ge on Ge (001) the on-top dimers are found in the type B configuration or occasionally in an intermediate state labeled A/B<sup>12,13</sup> (see Fig. 2). Whether or not these A/B dimers are really in an intermediate state<sup>14</sup> or rapidly rotating back and forth between the A and B configurations<sup>15</sup> is not yet fully clear. Recent calculations for the system Ge on Ge (001) have pointed out that besides the two already known stable on-top adsorption sites (A and B) a third local minimum exists.<sup>14</sup> The angle between the substrate dimer bonds and the adsorbed dimer in this local minimum is about 45°. Another interesting point is that the calculations are in favor of a dissociative rotational process rather than a nondissociative rotational process. Besides the on-top B and A/B dimers also type-C trough dimers are observed quite frequently (see Fig. 2). For the system Si on Ge (001) isolated on-top dimers are only found exclusively in the A/B configuration.<sup>16,17</sup> Because Si atoms might mix with Ge atoms of the substrate the nature of the ad-dimers can vary from pure Si–Si to mixed Si–Ge

TABLE I. Experimentally observed dimer adsorption sites after room-temperature deposition and imaging.

Adsorption site	Ge/Ge (001)	Si/Ge (001)	Si/Si (001)
A	No	No	Yes
B	Yes	No	Yes
A/B	Yes	Yes	No
C	Yes	? <sup>a</sup>	Yes
D	No	? <sup>a</sup>	No

<sup>a</sup>It is not settled yet if the features measured in the trough are C dimers, D dimers, or even triatomic clusters.

TABLE II. Relative energies for different adsorption configurations of Si or Ge ad-dimers on Ge (001) and Si (001).<sup>17</sup> Energies in eV/dimer.

	Ge/Ge (001) (eV)	Si/Ge (001) (eV)	Si/Si (001) (eV)
A	0.18	0.24	0.01
B	0	0	0
C	0.23	0.26	0.28
D	0.80	0.63	0.95

or even pure Ge–Ge.<sup>18–20</sup> Currently, there is no real consensus about the nature of the trough features of the system Si/Ge (001). Besides a C (Ref. 21) or a D dimer<sup>16</sup> also a triatomic cluster<sup>22</sup> has been proposed recently. In Table I, an overview of the experimentally observed dimer adsorption configurations is given.

## B. Diffusion

After having discussed the various stable adsorption sites of dimers we now move to their dynamic behavior at temperatures near or above room temperature. The first real-space diffusion studies of dimers on Si (001) were performed by Dijkkamp, van Loenen and Elswijk.<sup>23</sup> With a scanning tunneling microscope held at a stable temperature in the range from room temperature to 400 K, these authors were

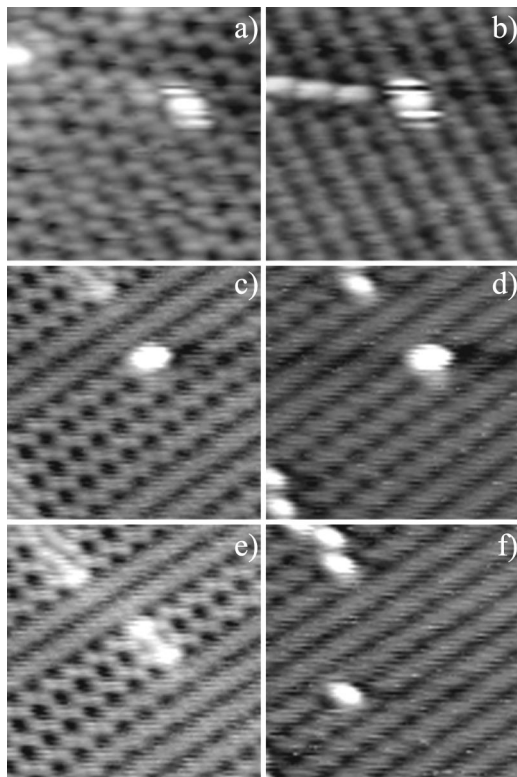


FIG. 2. Scanning tunneling microscopy images of isolated ad-dimers on Ge (001). Tunneling current (a)–(b) 1 nA and (c)–(f) 0.7 nA. Sample bias (a), (c), and (e)  $-1.6$  V, (b), (d), and (f)  $+1.6$  V. Image size  $7 \text{ nm} \times 7 \text{ nm}$ . (a) Filled state image of an A/B-type Si ad-dimer on Ge (001). (b) Empty state image of an A/B-type Si ad-dimer on Ge (001). (c) Filled state image of a B-type Ge ad-dimer on Ge (001). (d) Empty state image of a B-type Ge ad-dimer on Ge (001). (e) Filled state image of a C-type Ge ad-dimer on Ge (001). (f) Empty state image of a C-type Ge ad-dimer on Ge (001).

able to observe “in real time” the preferential diffusion of small silicon clusters along the substrate rows. However, at that time it was impossible to determine unambiguously the size of the diffusing species. They showed by measuring the mean-square displacement of the clusters that they performed a one-dimensional random walk. Several years later, Swartzentruber<sup>24</sup> measured directly in an elegant way the preferential diffusion of on-top dimers along the substrate rows with a modified scanning tunneling microscope denoted as atom-tracker scanning tunneling microscope. In this technique, a conventional STM tip is locked onto a selected dimer using two-dimensional lateral feedback. Once locked, the feedback electronics maintain the tip over the dimer tracking its coordinates as it diffuses over the surface. When a diffusion event occurs the atom tracker quickly relocates to the dimer’s new position (the atom tracker requires several ms (5–25 ms) to relock onto an dimer’s new coordinates after a diffusion event). The average jump frequency  $\nu$  of a dimer is expected to follow the Arrhenius relation for a thermally activated process:  $\nu = \nu_0 e^{-E/kT}$ , where  $E$  and  $\nu_0$  are the activation energy and attempt frequency,  $T$  the temperature, and  $k$  is Boltzmann’s constant. By plotting  $\ln \nu$  versus  $1/kT$  one can determine the activation energy and attempt frequency from the slope and the intercept, respectively. Before giving an overview of the diffusion rates of Si and Ge dimers on Ge (001), we first briefly discuss the Si on Si (001) system. For diffusion of an on-top Si dimer along the substrate rows of Si (001) the activation barrier and attempt frequency were found to be  $0.94 \pm 0.09 \text{ eV}$  and  $10^{12.8 \pm 1.3} \text{ s}^{-1}$ ,<sup>19</sup> respectively (in Ref. 10 an activation barrier of  $1.09 \pm 0.05 \text{ eV}$  and attempt frequency  $10^{13.2 \pm 0.6} \text{ s}^{-1}$  were found). For Si on Si (001) this pathway is the only diffusion pathway that is active in the range below about 400–450 K.

However, in 1997, Borovsky, Krueger and Ganz<sup>10</sup> showed experimentally that by increasing the substrate temperature to 450 K or above, the Si dimers are able to diffuse across the substrate dimer rows of Si (001). At these temperatures, dimers diffuse along the tops of substrate rows very rapidly, but occasionally the dimer jumps to a trough site. As is clear this diffusion process consists of two separate events: A jump from an on-top site to a trough site and a jump from a trough site to an on-top site (activation barrier  $1.36 \pm 0.06 \text{ eV}$ , assuming that the attempt frequency is the same as for the on-top diffusion pathway, i.e.,  $10^{13.2 \pm 0.6} \text{ s}^{-1}$  Ref. 10). The anisotropy between a long row and across dimer diffusion is about a factor of one thousand at 450 K. Besides the two already-discussed diffusion pathways, also a third diffusion pathway (diffusion of a trough dimer along the trough) turns out to be active [activation barrier  $1.21 \pm 0.09 \text{ eV}$ , assuming again an attempt frequency of  $10^{13.2 \pm 0.6} \text{ s}^{-1}$  (Ref. 10)].

In Table III, the diffusion barriers for the diffusion of Si and Ge dimers on the Ge (001) surface are presented. The diffusion of an on-top dimer along the substrate rows of Ge (001) is the fastest diffusion pathway with an activation barrier of  $0.82 \text{ eV}$  assuming an attempt frequency of  $10^{13} \text{ Hz}$  for a Si dimer versus an activation barrier of  $0.83 \text{ eV}$ , assuming an attempt frequency of  $10^{13} \text{ Hz}$ . In Fig. 3, the various pathways of Ge dimers on Ge (001) are shown. The diffusion rate



TABLE III. Diffusion barriers for the diffusion of a Si (Ge) dimer on Ge (001) compared to the diffusion of a Si dimer on Si (001). In order to compare the results we have set the attempt frequency to the attempt frequency we measured for the on-top diffusion of Si and Ge dimers on Ge (001), i.e.,  $10^{13}$  Hz.

Diffusion pathway	Ge/Ge (001)	Si/Ge (001)	Si/Si (001)
Along (on top)	0.82 eV ( $\nu_0=10^{13}$ Hz)	0.83 eV ( $\nu_0=10^{13}$ Hz)	0.94–1.09 eV <sup>a</sup>
Along (trough)	0.92 eV	1.0 eV	1.21 eV <sup>b</sup>
Across	0.95 eV	1.0 eV	1.36 eV <sup>b</sup>

<sup>a</sup>References 10 and 22.

<sup>b</sup>Reference 10.

of Ge and Si dimer to diffuse across the substrate rows at room temperature is  $10^{-3}$  and  $10^{-4}$  Hz, respectively. If we assume an attempt frequency of  $10^{13}$  Hz, these rates translate into activation barriers of 0.95 and 1.0 eV, respectively. Finally, the diffusion of a trough dimer in the trough at room temperature is also significantly faster for a Ge dimer than a Si dimer (0.92 versus 1.0 eV). In summary, the barriers for diffusion on Ge (001) are lower, as is the anisotropy of the diffusion as compared to Si (001). The difference between the systems Ge/Ge (001) and Si/Ge (001) is only small.

Another issue concerning the diffusion of dimers on (001) surfaces that remains to be addressed is the detailed atomic pathway of a diffusing dimer. In particular, does the

dimer break up into two atoms during a diffusion step or does it simply remain bound? For diffusion of on-top Si dimers over the substrate dimer rows of Si (001) atom-tracking experiments<sup>24</sup> reveal that the jumps are always to nearest-neighbor sites. This is strong evidence in favor of a simple model where the dimer remains bound during diffusion. However, recent atom-tracking studies<sup>25</sup> of a silicon dimer moving in the trough of Si (001) reveal that besides single jumps also double jumps and even triple jumps do occur quite frequently. The presence of these long jumps provides evidence in favor of a model in which the dimer bond breaks up completely and its two atoms move nearly independently along the trough until they meet again and reform the dimer. For the diffusion of Si and Ge dimers on top and across the substrate rows of Ge (001) our measurements also indicate that the dimer stays intact during the diffusion process. For the diffusion of a Si or Ge dimer in trough of Ge (001) we observe double jumps quite frequently. We suggest that the preference for these double jumps is related to the buckling registry in the direct proximity of the diffusing trough dimer. As has been pointed out in Ref. 12, an isolated C dimer pins the buckling registry into two local  $c$  ( $4 \times 2$ ) arrangements which are out-of-phase with each other. Exactly at the location of the C dimer the two regions with out-of-phase  $c$  ( $4 \times 2$ ) buckling registry meet. A jump of a C dimer to one of the two neighboring trough sites must therefore be accompanied with a phase shift of  $90^\circ$  of both local  $c$  ( $4 \times 2$ ) regions, i.e., a concerted motion of many surface dimers in the direct proximity of the diffusing trough dimer. Note that for a double jump along the trough *only* four surface dimers have to change their buckling registry.

Finally, we briefly studied the plausible diffusion pathways of an on-top Ge ad-dimer along the substrate dimer row directions using *ab-initio* calculations. The calculations [GAMESS (Ref. 26)] were performed by using an unrestricted shell Hartree-Fock method with a SBK (Stevens-Basch-Krauss-Cundari) effective core potential and SBK basis set for Ge and H atoms.<sup>27</sup>  $\text{Ge}_{23}\text{H}_{20}$  cluster was used to simulate diffusion of the on-top ad-dimer along the substrate rows [see Fig. 4(a)]. Unphysical dangling bonds on cluster boundary were terminated by H atoms. Potential energy surface (PES), was mapped out by calculating the adsorption energy depending on the displacement position,  $\Delta_1$  and  $\Delta_2$ , of both adatoms [see Fig. 4(a)]. The potential energy surface was generated with an equidistant grid with the spacing between the grid points of 0.32 Å. In the regions around the transition states displacements were 0.1 Å. The coordinates of the atoms in the two top surface levels and heights of the two adatoms were optimized. PES for the diffusion of the on-top Ge ad-dimer along the substrate row is shown in Fig. 4(b). The two most favorable paths for the on-top diffusion are shown in Fig. 4(b). Path I is the correlated motion of the ad-dimer and goes along with a stretching of the ad-dimer, whereas for pathway II, the ad-dimer stay fully intact. These pathways are similar to the ones obtained for Ge and Si ad-dimers on the Si (001) surface by Lu, Wang, and Ho.<sup>28</sup> Obtained energies for the barriers are 0.91 and 0.89 eV for the pathways I and II, respectively. The effect of cluster size

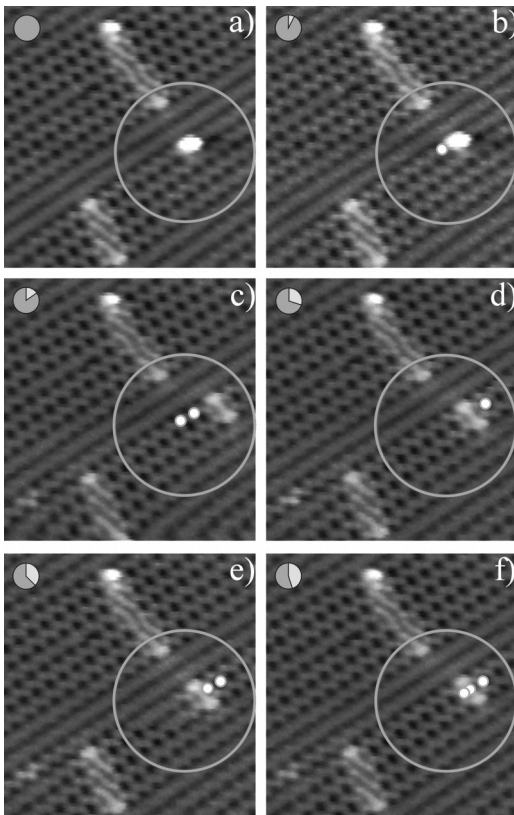


FIG. 3. Various diffusion pathways for a Ge ad-dimer on Ge (001) at room temperature. Tunneling current 0.7 nA, sample bias  $-1.6$  V, image size  $11 \text{ nm} \times 11 \text{ nm}$  and time lapse between subsequent images is 276 s. (a)–(b) Diffusion of an on-top ad-dimer along the substrate row (b)–(c) diffusion across the substrate row (c)–(f) diffusion of a trough ad-dimer in the trough.

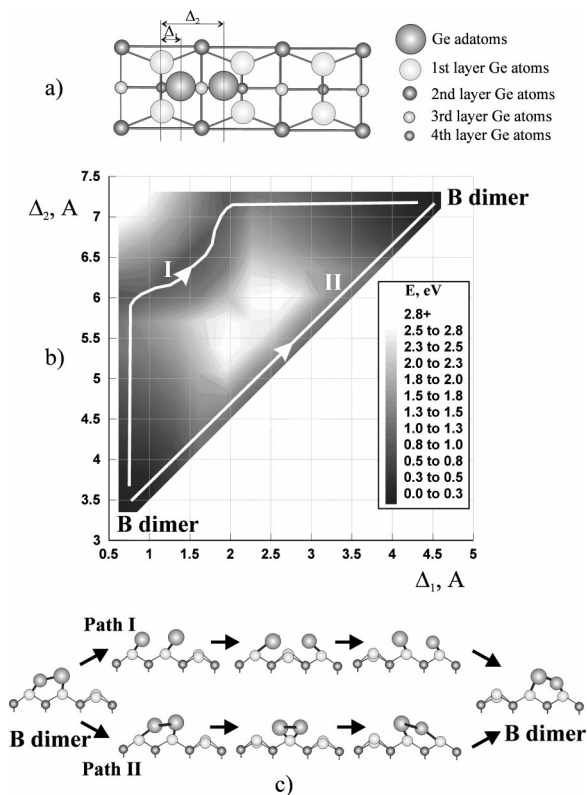


FIG. 4. (a)  $\text{Ge}_{23}\text{H}_{20}$  cluster model used to simulate diffusion of an on-top Ge ad-dimer along the substrate dimer row of Ge (001). Hydrogen atoms are not shown. (b) Potential energy surface (PES) depending on the displacements  $\Delta_1$  and  $\Delta_2$  of the adatoms. The two most plausible diffusion pathways are outlined. (c) Atomic configurations of the diffusion pathways I and II for an on-top Ge ad-dimer diffusing along a substrate row of Ge (001).

was checked by calculating the energies of the transition states on a larger cluster model of  $\text{Ge}_{43}\text{H}_{36}$ . The energies of the transition states obtained by large cluster are 0.96 and 0.92 eV for the pathways I and II, respectively, which are very close to the values 0.92 and 0.89 eV of the smaller cluster. The energies of the transition states of the two most plausible pathways are practically the same. The activation barrier for diffusion of an on-top Ge dimer along the substrate rows of Ge (001) is found to be  $\sim 0.9$  eV and is in agreement with the experimental value of 0.82 eV.

In summary, we have determined the diffusion rates for the diffusion of Ge and Si dimers over the substrate rows, across the substrate rows, and in the troughs of the Ge (001) surface. The barriers for Si and Ge dimer diffusion on Ge

(001) are lower, as is the anisotropy of the diffusion as compared to Si/Si (001).

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