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Structure and stability of the Si(1 0 5) surface

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Abstract

Recent experimental studies have shown that well-annealed, unstrained Si(1 0 5) surfaces appear disordered and atomically rough when imaged using scanning tunnelling microscopy. We construct new models for the Si(1 0 5) surface that are based on single- and double-height steps separated by Si(0 0 1) terraces, and propose that the observed surface disorder of Si(1 0 5) originates from the presence of several structural models with different atomic-scale features but similar energies. This degeneracy can be removed by applying compressive strains, a result that is consistent with recent observations of the structure of the Ge/Si(1 0 5) surface.
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The self-organized growth of Ge/Si quantum dots has been investigated extensively for more than a decade, driven by their potential applications as optoelectronic devices and nanoscale memories. In the early stages of growth, the quantum dots that have pyramidal shapes bounded by {1 0 5} facets evolve from stepped mounds without encountering any energetic barriers for their nucleation [1,2]. The absence of nucleation barriers has been explained by a competition between the strain-dependent, negative step formation energy and repulsive step–step interactions that have a weak dependence on strain [3,4]. The atomic con-

figuration of the (1 0 5) facets has also been elucidated; a rebonded step model for the Ge(1 0 5) surface under mismatch strain was found to play a crucial role for the stability of this surface [3,5–7].

While the structure of Ge/Si(1 0 5) surface has been recently elucidated, the Si(1 0 5) surface shows intriguing features that are not well understood. Experimental work by Tomitori et al. [8], Fujikawa et al. [5] and Zhao et al. [9] reveal that Si(1 0 5) is atomically rough even after careful annealing, and its scanning tunnelling microscopy (STM) image does not display 2D-periodicity. The analysis of Zhao et al. [9] suggests the presence of a structure for the Si(1 0 5) surface with large (0 0 1) facets and double-height steps. While a model for this surface with double-height steps was presented in [9], a study of its stability has not been attempted.

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Motivated by the recent STM investigations [5,9], we search for reconstructions of Si(1 0 5) based on (0 0 1) terraces separated by single- and double-height steps oriented along ⟨100⟩ directions. In addition to the currently accepted models of Si(1 0 5) (shown, for example, in Ref. [5]), we have found a few other possible structures based on double-height steps. We investigate the stability of these novel reconstructions, and propose that the roughness of the Si(1 0 5) surface is due to the coexistence of different structures with surface energies that are very close to each other. Furthermore, we find that compressive strain particularly favors a certain rebonded structure over all the others, which explains the atomically smooth and periodic structure of the Ge/Si(1 0 5) surface [5,8].

To show how to obtain structural models for Si(1 0 5) in a systematic manner, we start from the bulk-truncated structure and attempt to reconstruct the surface in such a way that each atom has at most one unsatisfied (dangling) bond after reconstruction. We now present in detail the structures of Si(1 0 5) with single- and double-height steps. The bulk-terminated Si(1 0 5), given in Fig. 1(a), consists of Si(0 0 1) terraces of width $5a/4$ separated by steps of monatomic height $a/4$, where a is the lattice constant of Si ($a = 5.43$ Å). Terraces can be reconstructed by forming short rows of dimers; because the dimer rows are oriented at 45° angles with respect to the direction of the steps, every other atom on the step edges must be eliminated in order to lower the number of dangling bonds (db). Depending on the relative position of
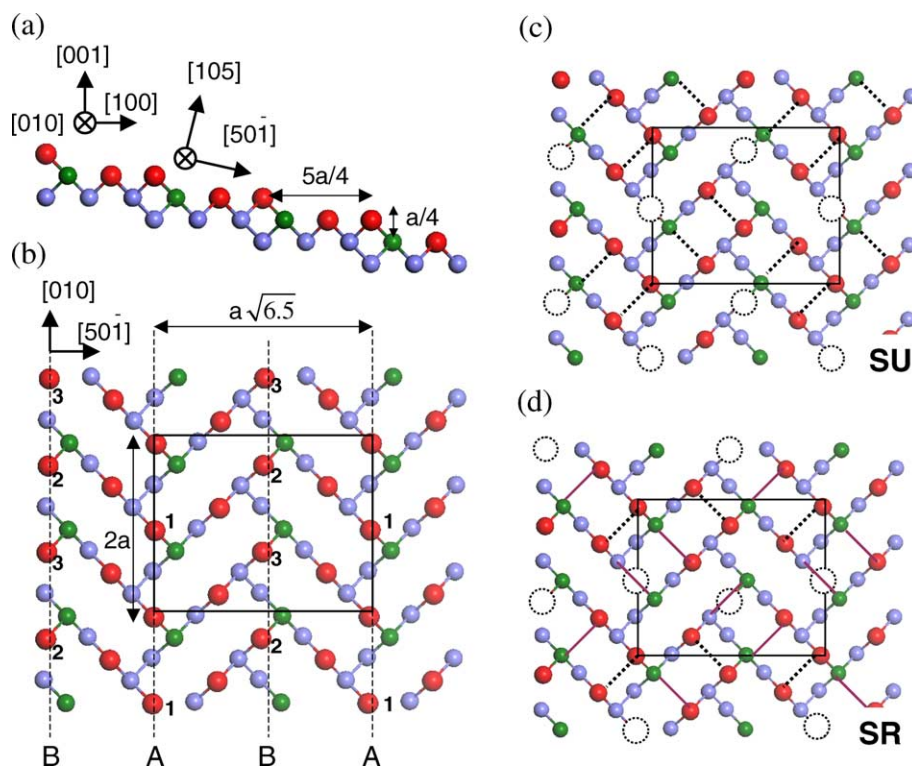

Fig. 1. Reconstructions of Si(1 0 5) with single-height steps and (0 0 1) terraces. (a) Side view and (b) top view of the bulk-truncated (1 0 5) surface. Atoms are colored according to their number of dangling bonds (db) before reconstruction: red = 2db, green = 1db, and blue = 0db. The (1 0 5) unit cells are marked by rectangles in (b)–(d). The single-height unrebonded (SU) and single-height rebonded (SR) models are shown in (c) and (d); these structures are obtained by eliminating atoms "1" and "2", and atoms "1" and "3", respectively (refer to (b)). The atoms that are removed are shown as open circles in (c) and (d). The remaining atoms are bonded as indicated by black dotted lines (dimer bonds) and purple solid lines (bridging bonds).

the atoms on the step edges that are eliminated, there are two distinct ways to achieve surface reconstruction. With the notation adopted in Fig. 1(b), one possibility is to eliminate atoms "1" on edges A and atoms "2" on edges B, and to then form dimers on the surface, as indicated by the dotted lines in Fig. 1(c). This is the model originally proposed by Mo et al. [10], and later named paired dimers [5]. Because the atoms on the terraces do not rebond at the step edges, we call this model single-height unrebonded (SU) [3].

Another way to achieve single-height reconstruction is by eliminating atoms "1" on edges A and atoms "3" on edges B (Fig. 1(b)) and then by creating bonds between the remaining surface atoms as indicated by the solid and dotted line segments in Fig. 1(d). This model, which we call single-height rebonded (SR) [3], was proposed by Khor and Das Sarma [11] and has recently been shown to appear on the side facets of the Ge quantum dots [5–7]. We note that there are two different types of bonds on this surface: the usual dimer bonds (dotted lines), and the bridging bonds (solid lines). The bridging bonds join a two-coordinated atom and a three-coordinated one, leaving the former with only one dangling bond and fully saturating the latter. Because the bridging bonds of the SR model are stretched, this reconstruction is strongly stabilized by the compressive strains present in Ge films deposited on Si surfaces [3,5,6]. Such rebonded models can also be found for structures that have double-height steps, as discussed below.

We now focus on the unreconstructed (1 0 5) surface with double-height steps shown in Fig. 2(a) and (b). Since the step height has doubled, the width of the terraces must also be doubled, in order to preserve the overall surface orientation. We found several models of double-height structures of Si(1 0 5), with different terrace structures (e.g. $(\sqrt{2} \times 1)$ or (2×1) reconstruction) and atomic bonding at the steps (e.g. rebonded or unrebonded), as explained below.

The simplest model based on $(\sqrt{2} \times 1)$ terraces is illustrated in Fig. 2(c), where no atoms are eliminated and dimers are formed as indicated by the dotted lines. Since no rebonding is present, we call this the double-height unrebonded (DU) structure. If we allow for the rebonding of atoms

on the lower terrace, then all the atoms on the step edges (denoted by C in Fig. 2(b)) must be removed, so that any surface atom would have fewer than two dangling bonds after reconstruction. The remaining atoms are then bonded as indicated by the solid and dotted lines in Fig. 2(d). Like the SR model, there are two types of surface bonds, the dimers and the bridges; we name this the double-height rebonded (DR) structure.

In the case of (2×1) reconstructed terraces, the dimer rows are oriented at 45° angles with the step edge. In analogy to the single-height case, we can eliminate every other atom on the step edges (rows C in Fig. 3(a)). The elimination of atoms on consecutive terraces can be done in-phase or out-of-phase, which leads to structures with different periodic lengths in the $[5\,0\,\bar{1}]$ direction, as shown in Fig. 3(b) and (c). Since both of these models involve rebonding at the step edges, we call them the DR1 and DR2 structures. From the DR1 structure we can obtain the unrebonded model of Zhao et al. [9] by removing another atom from each unit cell, as illustrated in Fig. 3(d); we label this the DU1 structure, to distinguish it from the DU model in Fig. 2.

We have computed the surface energy of the structural models shown in Figs. 1–3 using two empirical models for atomic interactions, namely the Stillinger–Weber [12] and Tersoff [13] potentials. While the empirical potentials provide a reasonable description of stepped Si(0 0 1) surfaces (refer, for example, to [14]), they are not able to capture the tilting (buckling) of the dimers at the surface, which constitutes an important way of surface relaxation for Si(1 0 5). Further, it is precisely the tilting of the dimers that determines the major features of the STM images and helps in the identification of the atomic structure of the surface [5–7]. In order to capture the dimer tilting, we have used the charge self-consistent tight-binding method of Wang et al. [15], which accurately predicts the energy ordering of several dimer-tilted Si(0 0 1) structures [16]. With this method, for each of the Si(1 0 5) structural models described above, the total energy of the atoms in the simulation cell exhibits many local minima, and we search for the lowest energy structures by using a combination of molecular dynamics simulations and annealing.
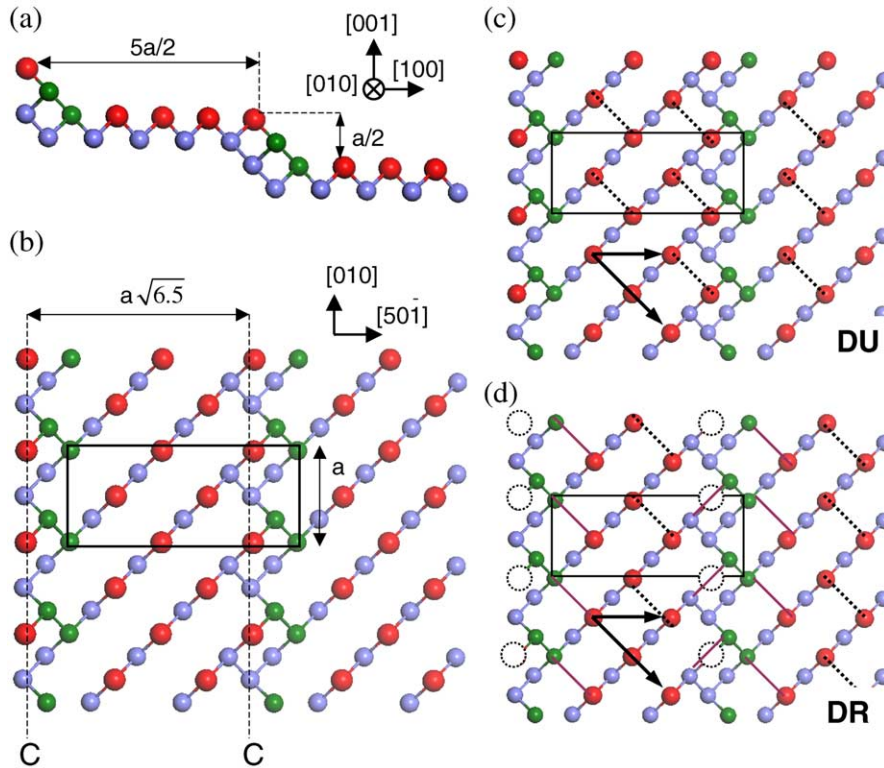
Fig. 2. Reconstructions of Si(1 0 5) with double-height steps and ($\sqrt{2} \times 1$)-Si(0 0 1) terraces. (a) Side view and (b) top view of the bulk truncated (1 0 5) surface with double-height steps. Atoms are colored according to the number of dangling bonds before reconstruction as explained in Fig. 1. The (1 0 5) unit cells are marked by rectangles in (b)–(d). The double-height unrebonded (DU) and the rebonded (DR) structures are shown in (c) and (d), respectively. The DR structure is obtained after elimination of all the atoms at the step edges (rows C in (b), open circles in (d)), followed by dimerization (black dotted lines) and rebonding (purple solid lines). The thick arrows in (c) and (d) represent the unit vectors of the ($\sqrt{2} \times 1$)-reconstructed terraces.

Our results are summarized in Table 1, where the surface energies of the Si(1 0 5) models computed using empirical [12,13] and tight-binding [15] potentials are given. The table also contains the number of dangling bonds per unit area for each model. We note that both the Stillinger–Weber (SW) and the Tersoff (T3) potentials yield large energy penalties for the dangling bonds on the surface, giving an energy ordering similar to that predicted by bond counting. On the other hand, the tight-binding (TB) description of atomic interactions allows the structures with large numbers of dbs/area (SU, DU and DU1) to relax via dimer buckling, leading to an entirely different ordering of the reconstructions. It can be seen from Table 1 that the surface energies of the (1 0 5)

models are spread over an interval of 8–12 meV/Å^2 when the empirical potentials are employed. In the case of tight-binding, this interval is only ≈ 4 meV/Å^2, due to a stronger relaxation of the unrebonded structures.

For all the potentials that have been used, we find that the SR model has the lowest surface energy among all the reconstructions considered; this is in agreement with previous work [3,5–7] where only the SU and SR models are compared. A closer look at the TB values in Table 1 shows that the SR, DU and DR2 models have energies that fall within ≈ 1 meV/Å^2 of the energy of SU, indicating a near-degeneracy of the lowest energy surface. Furthermore, our molecular dynamics simulations show that there are other local minima
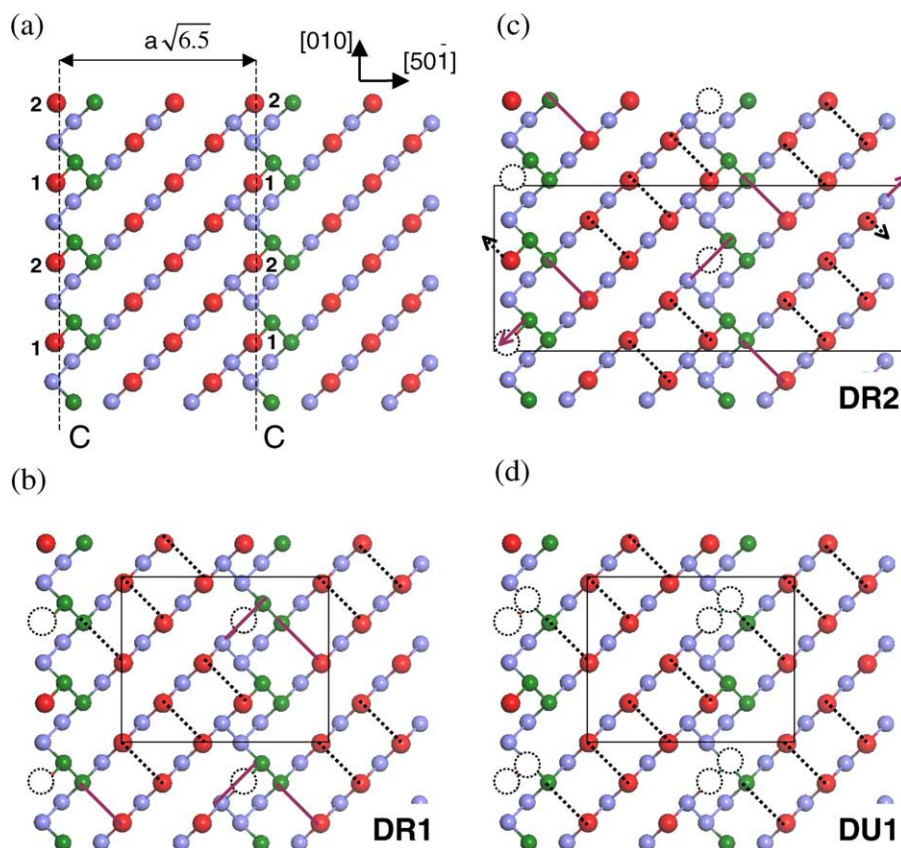
Fig. 3. Reconstructions of Si(1 0 5) with double-height steps and (2×1)-Si(0 0 1) terraces. Fig. 3(a) shows the top view of the bulk-truncated (1 0 5) surface with double-height steps. Atoms are colored according to the number of dangling bonds before reconstruction as explained in Fig. 1. Since the dimer rows on the terraces are oriented at 45° with respect to the step edges C, every other atom on the step edges is eliminated upon reconstruction. The elimination can proceed in-phase (atoms "1" on all step edges) or out-of-phase (atoms "1" on a given step edge and atoms "2" on the next edge). Since rebonding at the step edges is present in both models (purple solid lines), we label them DR1 (b) and DR2 (c). The atoms that have been removed to achieve the reconstruction are shown as open circles in (c) and (d). The (1 0 5) unit cells are marked by rectangles with the dimensions $2a \times a\sqrt{6.5}$ for DR1 and $2a \times 2a\sqrt{6.5}$ for DR2. The unrebonded model DU1 presented in Ref. [9] can be obtained by removing one more atom from the DR1 unit cell as shown in (d).

(with different bond bucklings) in the same energy interval. Due to this high degeneracy, we propose that several models can be present simultaneously on the Si(1 0 5) surface, which can explain the key features observed in experiments—disorder and roughness. Because energy differences are small, the entropy associated with the spatial distribution of different (1 0 5) unit cells would be important even at low temperatures, explaining the lack of 2D-periodicity of the STM images [5,9]; the atomic-scale roughness observed in the STM images can be generated by a random arrangement of

the single- and double-height structures that are close in energy. The proposal can be tested experimentally by imaging (zooming in) different areas or "patches" of the large-scale Si(1 0 5) samples. Preliminary work along these lines has been reported in [5,9].

We have also examined the strain-dependence of the different surface reconstructions. By calculating the energies of all the model structures for three values of an applied equibiaxial strain (−1%, 0%, +1%), we find that the near-degeneracy of the Si(1 0 5) surface can be removed when the surface

Table 1
Surface energy of Si(1 0 5) reconstructions calculated using the Stillinger–Weber potential (SW) [12], the Tersoff potential (T3) [13], and the self-consistent tight-binding method of Wang et al. (TB) [15]. The last column indicates the number of dangling bonds (db) per surface area expressed in units of $a^2\sqrt{6.5}$, where a is the bulk lattice constant of Si

	SW (meV/Å^2)	T3 (meV/Å^2)	TB (meV/Å^2)	Bond counting $(db/a^2\sqrt{6.5})$
SU	99.63	99.40	83.54	6
SR	90.39	90.79	82.78	4
DU	102.24	99.36	84.84	6
DU1	99.35	99.00	87.03	6
DR	96.24	95.09	87.03	4
DR1	96.27	96.64	85.22	5
DR2	95.99	96.26	83.48	5

is subjected to a compressive state of strain. As illustrated in Fig. 4, even a compressive strain which is as small as −1% further stabilizes the SR model over the other models. This result is supported by the recent observations of Fujikawa et al., who showed that the initially rough, unstrained, Si(1 0 5) surface becomes smooth after being subjected to a compressive mismatch strain through the deposition of three monolayers of Ge [5].

The origin of the strain-dependence of the surface energy lies in the arrangement and type of the atomic bonds around the step edges: If a structure contains significant rebonding (i.e., has bridging bonds that are stretched compared to the bulk bonds), compressive stresses tend to lower its surface energy. Indeed, this trend emerges from the

data presented in Fig. 4: all rebonded structures show a decrease of their energy (relative to SU) in compression. In contrast, the energy gap between the unrebonded structures (DU and SU, DU1 and SU) remains almost constant for the range of strains investigated here. This finding is fully consistent with our previous work on the formation energies of ⟨1 0 0⟩ steps [3], as well as with the results of Refs. [5,6,8] on Ge/Si(1 0 5).

In summary, we have constructed a set of structural models for Si(1 0 5) and analyzed their stability using empirical [12,13] and tight-binding [15] potentials. Our study shows that the presence of single- and double-height reconstructions on Si(1 0 5) can explain the experimentally observed [5,9] atomic roughness and disorder of this surface. Three double-stepped reconstructions (DU, DR1,
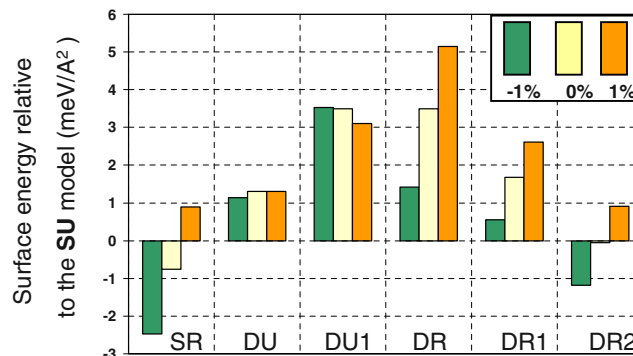

Fig. 4. Surface energies of different Si(1 0 5) structures measured with respect to the surface energy of the SU model, computed with the tight-binding method [15] for three values of applied equibiaxial strain: +1%(orange), 0%(yellow) and −1%(green). While there are three models (SR, DU, DR2) that have surface energies within ∼1 meV/Å^2 of the surface energy of SU at zero strain, a small amount of compressive strain (−1%) removes this near-degeneracy and strongly stabilizes the SR model over all the other models.

DR2) were found to have lower surface energies than the double-step model proposed in [9]. The atomic bonding at the step edge determines the strain-dependence of the surface reconstructions and leads to the strain-induced stabilization of the SR structure. Future experiments on strained Si(1 0 5) surfaces (produced, for example, by bending) would be invaluable in gaining further insight into the evolution of surface roughness as a function of strain. The models presented here may also serve as building blocks for other structures, for example, the quenched (1×4)-Si(1 0 5) observed in [9].

Acknowledgements

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