

Role of Spin-Orbit Splitting and Dynamical Fluctuations in the Si(557)-Au Surface

Daniel Sánchez-Portal,^{1,*} Sampsa Riikonen,¹ and Richard M. Martin²

¹*Unidad de Física de Materiales CSIC-UPV/EHU, Donostia International Physics Center (DIPC), and Departamento de Física de Materiales, Universidad del País Vasco, Apdo. 1072, 20080 Donostia, Spain*

²*Department of Physics and Materials Research Laboratory, University of Illinois, Urbana, Illinois 61801, USA*

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Our *ab initio* calculations show that spin-orbit coupling is crucial to understand the electronic structure of the Si(557)-Au surface. The spin-orbit splitting produces the two one-dimensional bands observed in photoemission, which were previously attributed to spin-charge separation in a Luttinger liquid. This spin splitting might have relevance for future device applications. We also show that the apparent Peierls-like transition observed in this surface by scanning tunneling microscopy is a result of the dynamical fluctuations of the step-edge structure which are quenched as the temperature is decreased.

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Atomic-scale “wires” created on semiconductor surfaces have the potential to be the basis for nanoscale devices and to demonstrate the novel physics of one dimension. A specially attractive route to create such structures is provided by the use of stepped vicinal surfaces, which can be used as templates to deposit material creating very regular arrays of parallel metallic chains [1]. The Si(557)-Au reconstruction is one of these systems. The terraces of this surface are perpendicular to the (111) direction and have a width of ~ 19 Å. Each terrace contains a monatomic chain of gold atoms running parallel to the step edge. For this reason, the Si(557)-Au surface has been proposed as an experimental realization of a one-dimensional metal and has attracted much attention in recent years [2–8]. However, in spite of these efforts, the electronic structure of this system is not yet completely understood.

The first angle-resolved photoemission (ARP) study by Segovia *et al.* [2] found a spectrum dominated by a one-dimensional metallic band. This band was shown to split in two peaks near the Fermi level (E_F), and this was interpreted as a signature of separated charge and spin low-energy excitations as predicted by Tomonaga-Luttinger theory of the one-dimensional electron gas [9]. However, later photoemission data seemed to discard this interpretation. According to Losio *et al.* [3], the observed splitting would correspond to two distinct proximal bands that cross E_F at neighboring, although different, positions of the surface Brillouin zone. However, the origin of these bands was unclear. Finally, Ahn *et al.* [4] have recently suggested that only one of the bands is truly metallic and suffers a metal-insulator transition upon cooling. This observation was correlated with the temperature dependence of the scanning tunneling microscopy (STM) images: the step edge undergoes a periodicity doubling consistent with a Peierls-like instability. Thus, these authors concluded that at least one of the two proximal bands should be associated with the atoms forming the step edge.

In contrast with the electronic structure, the geometry of the Si(557)-Au reconstruction seems to be quite well established. A detailed model was recently proposed by Robinson *et al.* [6] on the basis of x-ray diffraction data, which has been corroborated by first-principles density function theory (DFT) calculations [8]. Unfortunately, the calculated band structure only presents *one* one-dimensional metallic band exhibiting a considerable dispersion and a width consistent with the experimental observations. Thus, the observed two-band photoemission spectrum remains unexplained from a theoretical point of view.

In this Letter, we present the results of DFT calculations that satisfactorily explain the observed ARP spectra and the temperature dependence of the STM images of the Si(557)-Au surface. The two proximal bands appear as a consequence of the spin-orbit (SO) splitting of the most dispersive surface state. The large effect of SO splitting on gold-derived surface states has been demonstrated previously by ARP experiments and calculations on Au(111) [10,11]. The origin and magnitude of the SO splitting is similar in the present case. The inversion symmetry is always broken at the surface, thus the spin splitting of bands with no spatial degeneracy becomes possible. This suggests that atomic wires formed by heavy atoms deposited on semiconducting surfaces could be used in the fabrication of spin transistors [12] and spin-filter devices [13]. Interesting phenomena can also appear associated with the competition between electron-electron interactions and SO coupling in one dimension [14,15].

Our results also show that the temperature dependence of the STM images stems from the buckling of the step edge, whose atoms alternate between up and down positions. A given “up-down” configuration and the reverse one are separated by a small energy barrier. At room temperature, the step edge fluctuates between both structures, and the STM only reflects the average surface electronic and atomic structure. Upon cooling, the step-

edge buckling can be revealed using STM, producing the apparent doubling of the periodicity.

Our slabs contain four silicon bilayers, the bottom surface saturated with hydrogen, with a total of 115 atoms. Because of this large supercell, we use the SIESTA code [16,17] to study the energy landscape and the structural properties of the Si(557)-Au surface. The details of the calculations are similar to those presented in Refs. [7,8] for the same system. Since the present version of SIESTA does not allow to include the SO interaction, we have used the VASP code [18,19] to study its effect on the electronic structure of our relaxed structures. The VASP calculations utilized projected-augmented-wave potentials with a plane-wave cutoff of 250 eV, which proved to be sufficient to obtain converged electronic band structures.

Figure 1(a) shows the relaxed structure of the Si(557)-Au reconstruction. This structure is almost identical to the experimentally proposed model [6,8]. The larger, filled circles stand for the gold atoms occupying silicon substitutional positions on the middle of the terraces. The corresponding band structure, along the direction parallel to the steps, is shown in Figs. 1(b) and 1(c). Panel (b) shows the results from a non spin-polarized VASP calculation using the local density approximation (LDA) for the exchange-correlation potential. This band structure is almost identical to that already reported in Ref. [8] using the SIESTA code. Several surface bands and resonances can be identified, all of them with negligible dispersion in the direction perpendicular to the steps. The different symbols reflect their main atomic character, as obtained from the projection of the charge onto nonoverlapping spheres

centered on each atom. The unoccupied band marked with open triangles comes from the adatoms [labeled *ad* in Fig. 1(a)], while the occupied one is related to the restatoms (labeled *rest*). In principle, every atom in the step edge has a dangling bond pointing perpendicularly to the step, which would give rise to a very flat half-filled band. This unstable situation leads to a buckling of the step edge that doubles the unit cell and forms two bands marked by open squares in Fig. 1. The band with larger weight in the “up” (*B*) atoms is fully occupied, while the band associated with the “down” (*A*) atoms has a small occupation [20]. Notice that the step-edge bands have a very small dispersion. Thus, they cannot explain the observed ARP spectra, as was recently suggested by Ahn *et al.* [4]. Contrary to the initial interpretation of the ARP data [2], none of the surface bands in the proximity of E_F has a clear Au 6s character. This is a direct consequence of the larger electron affinity of gold as compared to silicon: the 6s Au character appears several eV below E_F . The sole surface bands exhibiting an appreciable gold component in Fig. 1 are marked with circles. These bands mainly come from the 3p states of the silicon atoms neighboring to the Au chains (atoms *D* and *C*). However, they also show a strong contribution to the Si-Au bonds. We find a flat band (open circles) associated with the Au-Si_{*D*} bonds, and a dispersive (filled circles) corresponding to the Au-Si_{*C*} bonds. This last band is the *only one* that presents a dispersion and width consistent with the photoemission data.

Figure 1(c) depicts the same band structure once the effect of the SO interaction has been included in a non-collinear spin calculation. We still get a non spin-polarized ground state. The changes are negligible for most surface bands. This is expected since most of them are localized in regions far from the gold atoms or have a very small dispersion, i.e., small group velocities. However, the dispersive Au-Si_{*C*} band develops a considerable SO splitting. This brings the calculated band structure in reasonable agreement with the experimental spectra. The experiments [3,4] show a ~ 300 meV splitting near E_F . This splitting exhibits a linear dependence as a function of k_{\parallel} with a ~ 1.2 eV \AA slope. This can be compared with our calculated ~ 200 meV splitting at E_F and ~ 1.4 eV \AA slope.

Even though the two SO-split bands are a robust feature predicted for this surface, the states at the Fermi energy are strongly affected by the other bands shown in Figs. 1(b) and 1(c). In our calculations, both SO-split bands are metallic; however, there is a band gap just above E_F . This gap relates to the presence of a row of adatoms in the terrace which induces an alternation of the Si_{*C*}-Au-Si_{*C*} bond angle between 101.8° and 109.6° . The presence of this gap drives the surface very close to becoming semiconducting. The metallicity of the system is due to the very small partial occupation of the upper

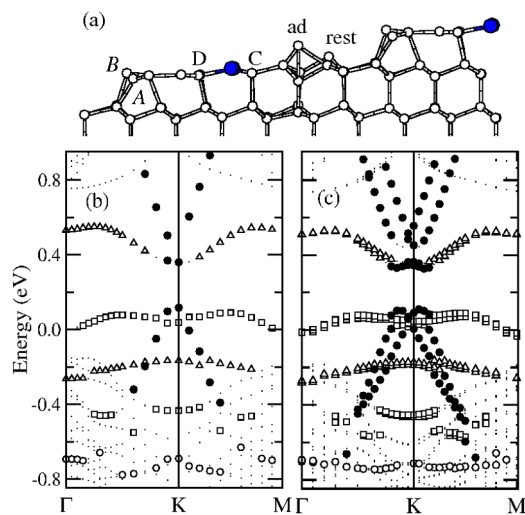


FIG. 1 (color online). (a) Calculated equilibrium structure of the Si(557)-Au reconstruction. The corresponding electronic band structure is shown for a calculation not including (b), and including (c), the spin-orbit interaction. Energies are referred to the Fermi level. Surface states have been marked with different symbols according to their main atomic character (see text).

step-edge band that pins the position of E_F . A description of exchange and correlation beyond LDA would quite likely cause a widening of the step-edge gap, leading to a semiconducting band structure at zero temperature.

The ground-state structure and the electronic bands shown in Fig. 1 perfectly explain the low-temperature STM images of the surface [4]. The step edge exhibits a buckling, with A atoms lying $\sim 0.4 \text{ \AA}$ below B atoms. Furthermore, a high contrast between these two types of silicon atoms is guaranteed. Empty-state STM images will preferentially show A atoms, while filled-state images will reflect the location of B atoms.

The origin of the temperature dependence of the STM images is clarified in Fig. 2, where the stability of the step-edge buckling respect to thermal vibrations is analyzed using SIESTA. Figure 2(a) shows the energy landscape as a function of the position of atom A , i.e., the lower atom of the step edge in the ground-state structure. We progressively increase its height, Z_A (see the filled symbols in Fig. 2). At each step, the whole structure is relaxed while keeping Z_A as a constraint. As expected, the energy increases quadratically for small displacements from the equilibrium position ($Z_A = 0$). The height of the B atom (Z_B) remains almost unchanged during this process as shown in Fig. 2(b) and, although the magni-

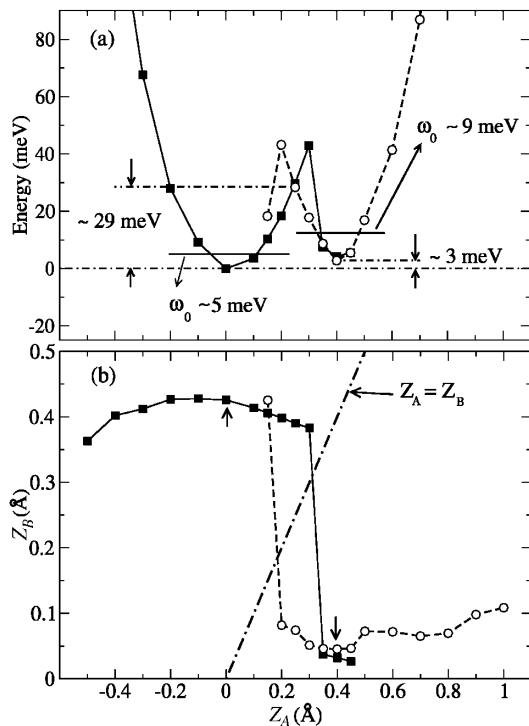


FIG. 2. Total energy (a) and position of the B atom (b) as a function of the height of atom A . Heights are referred to that of atom A in the ground-state configuration. The arrows in panel (b) mark the position of the two energy minima. The energy landscape shows two wells separated by a small energy barrier of ~ 29 meV, and estimated zero-point energies of $\omega_0 \sim 5$ and 9 meV.

tude of the gap decreases as Z_A approaches Z_B , we can still unambiguously identify the fully and partially occupied step-edge bands, respectively, with the atoms B and A . However, for larger displacements, the situation is very different. When $Z_A \geq Z_B$, the structure becomes unstable and collapses into a new configuration with an inverted step-edge buckling. Atom A is now situated $\sim 0.4 \text{ \AA}$ above atom B . This sudden change is accompanied by a charge transfer from atom B to atom A so that the filled step-edge band can be always assigned to the atom occupying the highest position.

The reversed buckling configuration is also stable against small displacements (see the open symbols in Fig. 2). Its energy is only ~ 3 meV (~ 5 meV) larger than that of the ground-state structure within the LDA (generalized gradient approximation, GGA). For large displacements we find a transition back to the ground-state structure (open symbols in Fig. 2). This means that the roles of atom A and B can be interchanged without any appreciable change of the global structure of the surface and the total energy. This is not surprising if we consider that, for the structure shown in Fig. 1(a), the two step edge sites only become inequivalent when we consider their registry with the adatom row in the same terrace, which is located at $\sim 12 \text{ \AA}$ from the step edge. Thus, the energy landscape as a function of the height of the atoms in the step edge is formed by two similar wells. The energy barrier separating both structures turns out to be quite small, ~ 29 meV (~ 25 meV) within LDA (GGA).

From the total energy curves and the corresponding displacements of all the atoms in the supercell, the zero-point energies are estimated to be $\omega_0 \sim 5$ meV for the ground-state structure and ~ 9 meV for the reversed buckling one. This, together with the magnitude of the calculated barrier, shows that at low-temperatures, both structures are indeed stable for long times. The simulated images using Tersoff-Hamann theory [21] can be found in Fig. 3(a), the upper part of the panel corresponding to the ground-state configuration and the lower part to the reversed step-edge buckling. The images are very similar. They are dominated by two chains showing a double periodicity along the step-edge direction in good agreement with low-temperature images reported for this system [3,4]. One of the chains is the row of adatoms. The other corresponds to the step edge, where only every other atom is visualized.

The situation changes at higher temperatures. The time necessary to flip between the different step-edge configurations gets shorter as the temperature increases. Given the size of the calculated energy barrier, at room temperature, the STM images will only show a time average of the electronic and atomic structure of the surface. This has been modeled in Fig. 3(b) by averaging the STM images of the two structures. As a consequence, the double periodicity is lost and all the atoms in the step edge appear with similar intensities. This explains the

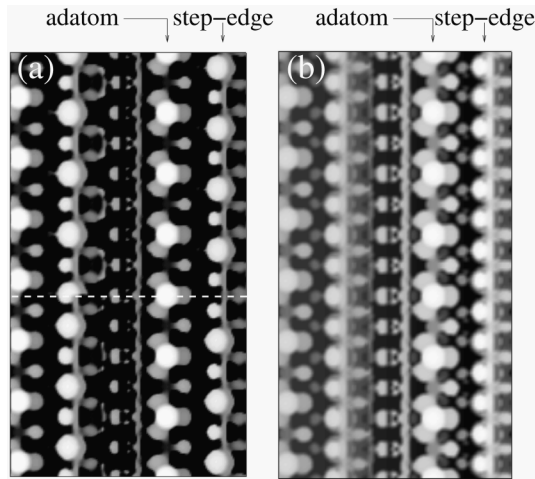


FIG. 3. Simulated STM images at low (a) and high temperature (b) for a +0.7 V bias voltage (empty states). The upper part of panel (a) corresponds to the ground-state structure, while the lower part is obtained with the reversed step-edge configuration. Panel (b) combines both images.

apparent periodicity doubling observed by Ahn *et al.* [4] upon cooling. Therefore, the mechanism behind the change of appearance of the STM images of the Si(557)-Au surface is similar to that proposed to explain, for example, the $3 \times 3 \rightarrow \sqrt{3} \times \sqrt{3}$ transition in Sn/Ge(111) [22].

Our results also provide a plausible scenario to explain the observation of a metal-insulator transition in this surface [4]. The Fermi energy—and thus the occupation and metallicity of the SO-split bands—depends strongly upon the position of the upper step-edge band. Small changes may drive the system from metallic to insulator and conversely. Furthermore, as the temperature increases, the step-edge structure is expected to fluctuate at an increasing rate. Associated with this atomic movement, there is an electron dynamics that can be interpreted as a successive closing and opening of the step-edge gap or, alternatively, as an increasing width of the step-edge energy levels. Therefore, we can expect changes in the spectral weight at E_F as the temperature is increased.

In summary, we report on first-principles DFT calculations unveiling the origin of several phenomena observed in the Si(557)-Au surface. The experimental electronic band structure is successfully reproduced, pointing out the important role played by the spin-orbit coupling in this system. From a general perspective, this raises the question of whether systems composed by atomic-scale wires of heavy atoms on semiconducting substrates can be used to create or transport spin-polarized currents, and thus be useful for future electronic devices. The temperature dependence of the STM images can be easily explained by the presence of two structurally inequivalent silicon atoms in the step edge

that fluctuate at room temperature between two positions. The presence of an electronic gap slightly above E_F together with the step-edge fluctuation can also explain large changes in the bands near the Fermi energy as a function of temperature.

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*Electronic address: sqbsapod@sc.ehu.es

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