Adsorbate-induced reconstruction of an array of atomic wires: Indium on the Si(553)-Au surface

J. R. Ahn,[1,*] P. G. Kang,[2] J. H. Byun,[2] and H. W. Yeom[2,†]
[1]*BK21 Physics Research Division and Sungkyunkwan University Advanced Institute of Nano Technology (SAINT), Sungkyunkwan University, Suwon 440-746, Republic of Korea*
[2]*Center for Atomic Wires and Layers and Institute of Physics and Applied Physics, Yonsei University, Seoul 120-746, Korea*
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The In-induced surface reconstruction of the Si(553)-Au surface has been studied using the combined experiment of low-energy-electron diffraction, scanning tunneling microscopy, and angle-resolved photoemission spectroscopy. Low-energy-electron diffraction revealed that In adsorbates interact actively with the surface above 150 °C, widening the terraces uniformly and forming a new atomic wire array. This wire structure has a $\times 2$ period along the wires, where the phase coherence across the wires was much better than that of the pristine Si(553)-Au surface. The In-induced uniform terrace widening was confirmed by scanning tunneling microscopy. More interestingly, the In adsorbates alter the metallic atomic wires of the Si(553)-Au surface with highly dispersive one-dimensional bands into insulating ones with still large dispersion.

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One-dimensional (1D) electronic systems have attracted considerable interest on account of exotic phenomena such as Peierls instability and Tomonaga-Luttinger liquid.[1–4] In order to investigate such 1D phenomena in a real system, electrons need to be delocalized along only a specific direction. This is observed as highly dispersive 1D energy bands in angle-resolved photoemission spectroscopy. There have been few 1D structures with highly dispersive 1D bands because it is not guaranteed that all 1D structures have sufficient electron delocalization.

Recently, few 1D structures with highly dispersive bands were observed on Si surfaces: the In/Si(111)-4×1 surface[2,5–8] and the Au/Si(111)-5×2 surface.[9,10] The three 1D metallic bands of the In/Si(111)-4×1 surface are related to interesting phenomena such as 1D charge density wave (CDW) formation,[2] interband interactions within multiple metallic bands,[5] and the coexistence of metallic and CDW phases.[6–8] The Au/Si(111)-5×2 surface with 1D bands was reported to show a continuous transition in dimensionality[9] as well as a local electronic structure of a disordered 1D metallic system.[10] More recently, stepped Si surfaces have been used to provide various 1D structures. The Au adsorption on stepped Si surfaces, Si(557)-Au and Si(553)-Au, were found to produce multiple 1D metallic bands, where the number and band fillings of the 1D bands depend on the substrates.[3,4,11,12] These systems have exhibited interesting phenomena such as the Peierls instability on the Si(557)-Au surface,[3] the coexistence of $\times2$ and $\times3$ Peierls distortions within a single unit wire on the Si(553)-Au surface,[4] and spin-orbit-split 1D metallic bands.[13]

In this work, we have attempted to modify the electronic and atomic structures of atomic wires by adsorbing metal atoms in order to provide another degree of freedom in fabricating atomic wires. Indium atoms were deposited on the metallic atomic wires on the Si(553)-Au surface. The terrace width of the Si(553)-Au surface was widened uniformly by In adsorbates. The In-adsorbed Si(553)-Au surface was observed to have $\times2$ order along the wires with a much longer coherent length across the wires. Note that the room temperature (RT) $\times2$ order of the Si(553)-Au surface is due to the local $\times2$ charge density modulation caused mainly by defects. The In-induced reconstruction produces insulating atomic wires with similar electronic and atomic structures to those of the pristine Si(553)-Au surface.

The photoemission measurements were performed using a high-resolution electron analyzer (SES-100, Gamma Data) and a high-flux monochromatized He I radiation ($h\nu$ =21.2 eV).[4] The nominal energy and angular resolutions were better than 15 meV and 0.15°, respectively. A commercial variable-temperature scanning tunneling microscope (STM) (Omicron, Germany) was used.[4] A Si(553) substrate (12.5° offcut from the [111] orientation) was cleaned thermally. A Si(553) surface with a regular atomic wire array was generated by depositing Au at 650 °C with a postannealing.[4] Indium atoms were deposited using a heated graphite crucible.

Figure 1 shows the temperature dependence of the In-adsorbed Si(553)-Au surface. The low-energy-electron diffraction (LEED) pattern of the Si(553)-Au surface at RT is 1×2 (hereafter 1×2-Au), as shown in Fig. 1(a).[4] The In adsorption of 1 ML (monolayer) on the 1×2-Au surface at RT changed the LEED pattern into a diffuse 1×1 phase. With increasing annealing temperature, the diffuse 1×1 phase changed gradually into a 1×2 phase which was optimized at 290 °C. A closer inspection revealed this In-induced 1×2 phase (hereafter 1×2-In) to have different long-range orders along and across the wires from the 1×2-Au phase. First, the period in k space across the wires was reduced from 0.42 to 0.35 Å^{-1}, as shown in Fig. 2(a). This suggests that the terrace width of the surface increases from 1.48 to 1.80 nm. The increased width agrees closely with a single unit cell of the lattice constant of the Si substrate across the wires, as shown in Figs. 3 and 4. Second, the $\times2$ LEED streaks of the 1×2-Au phase were sharpened into the spots in the 1×2-In phase. This suggests that the coherent length across the wires related to the $\times2$ period along the wires is enhanced. The change in long-range order indicates that the In adsorbates induce surface reconstruction. The LEED pattern and its line profiles across the wires at 550 °C became identical to those of the 1×2-Au surface, as

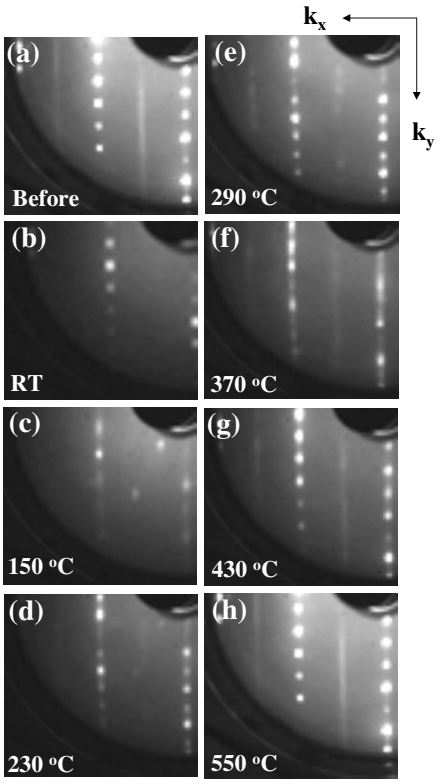

FIG. 1. The LEED pattern at a beam energy of 80 eV of the In-adsorbed Si(553)-Au surface at different annealing temperatures from RT to 550 °C. The 1×2 LEED pattern of the Si(553)-Au surface before In adsorption is shown in (a).

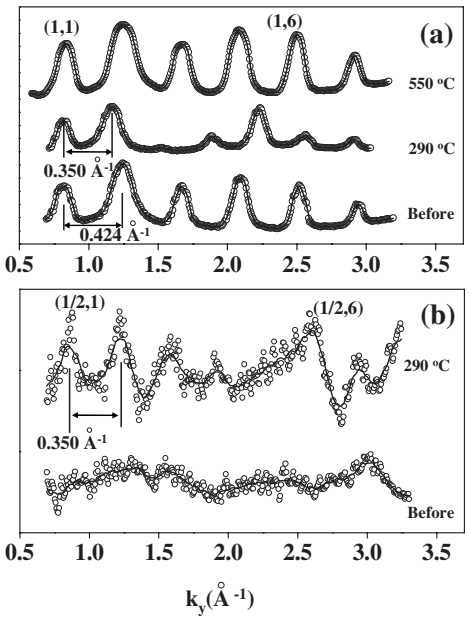

FIG. 2. Annealing-temperature dependence of the line profiles across the wires of the (a) integer ($\times1$) and (b) noninteger ($\times2$) spots of the LEED patterns of the In-adsorbed Si(553)-Au surface. Here, the solid lines are the smoothed lines of the raw dates denoted by the open circles.

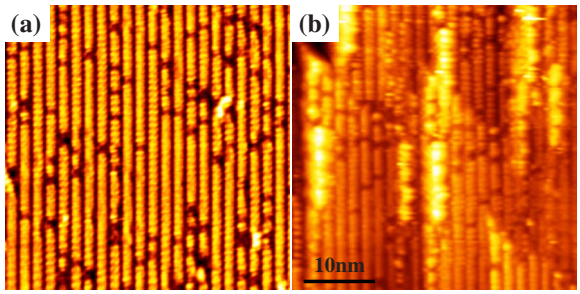

FIG. 3. (Color online) Empty-state STM images (V_s=1.0 V) of the (a) flat and (b) step-bunched regions of the 1×2-In surface.

shown in Figs. 1(h) and 2(a). This suggests that the In adsorbates are desorbed at 550 °C and the 1×2-In surface changes into the 1×2-Au surface, which is also supported by the STM images and band structures, as described below.

The In-induced changes in the real space were visualized by STM [Figs. 4(a) and 4(b) for 1×2-Au and Figs. 3, 4(c), and 4(d) for 1×2-In]. The width of the single wire unit cell of the 1×2-Au surface is 1.48 nm and the single wire unit cell is composed of different atomic chains of step edge Si atoms, Si stoms within the terraces, and Au atoms.[4,12,14] The Si atoms at the step edge have two broken bonds and the Au atoms substitute for Si surface atoms within the terrace.[12] As detailed recently, the bright and dark chains, which were observed in both the filled-state and empty-state STM images [see Figs. 4(a) and 4(b)], correspond to the step edge and terrace atomic chains, respectively.[15] Recent STM experiments showed that the terrace and step edge atomic chains of the 1×2-Au surface undergo charge density modulations from the RT $\times1$ to the low temperature (LT) $\times2$ period and from the RT $\times1$ to the LT $\times3$ period, respectively.[4,14] At RT, a part of the terrace chains shows a local $\times2$ charge density

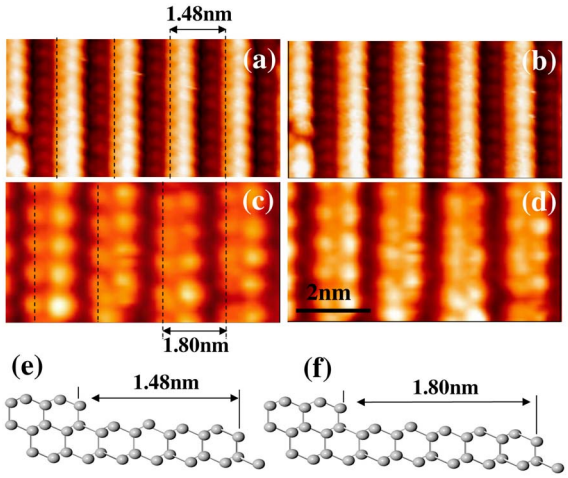

FIG. 4. (Color online) (a) Filled-state (V_s=−0.2 V) and (b) empty-state (V_s=0.2 V) STM images of the 1×2-Au surface and (c) filled-state (V_s=−0.2 V) and (d) empty-state (V_s=0.2 V) STM images of the 1×2-In surface. The atomic structures of the bulk-terminated 1×1 surfaces with the same terrace widths with the (e) 1×2-Au and (f) 1×2-In surfaces, respectively.

modulation, as shown in Figs. 4(a) and 4(b) albeit not clearly. This is believed to produce the ×2 streaks in the 1 ×2-Au LEED pattern. The STM images of the 1×2-In surface show that the In adsorption increases the terrace width by 0.32 nm, as shown in Fig. 4. Two different types of atomic chains within the single wire unit cell were observed in both the filled-state and empty-state STM images of the 1×2-In surface, which is composed of rows of bright protrusions with the same ×2 period. The two atomic chains are tentatively assigned to be located within the terrace (brighter chain) and at the step edge (darker one). The combined LEED and STM experiments reveal the In adsorbates to widen the terrace of the 1×2-Au surface uniformly by a single lattice constant. In addition, the In adsorbates produce two different types of atomic wires located at the step edge and within the terrace, respectively, as observed on the 1 ×2-Au surface. Both the atomic chains have ×2 periods along the wires with longer coherence lengths across the wires than those of the 1×2-Au surface. The In-induced terrace widening results in step bunching at most of the surface, as shown in Fig. 3(b). The step bunching occurs at every fifth terrace on average, which is reasonable because the terrace width of the 1×2-In surface increases by a single unit cell compared with that of the 1×2-Au surface. The In coverage of the 1×2-In surface was estimated from the intensities of the In core-level spectra, which were measured with $h\nu$=105 eV. The relative intensity of the In core-level spectrum of the 1×2-In surface to that of the In/Si(111)-4 ×1 surface (data not shown) ranged from 0.3 to 0.6. The In coverage of the 1×2-In surface was estimated to range from 0.3 to 0.6 ML because the In coverage of the In/Si(111)-4 ×1 surface is 1 ML.[2,5–8] This suggests that four or six In atoms exist in the 1×2-In unit cell. After annealing at 550 °C, the STM image became identical to that of the 1 ×2-Au surface, as shown from the LEED pattern of the sample annealed at 550 °C.

The structural phase transition from the 1×2-Au to the 1×2-In surface was further studied by photoemission spectroscopy, as shown in Fig. 5. The band structure of the 1 ×2-Au surface at RT is composed of three fractional-filled 1D bands, two nearly half-filled bands (S_1 and S_2), and a single roughly third-filled band (S_3).[3,12] The three 1D bands have dual electronic properties: the S_1 and S_3 bands are metallic but the S_2 band is semiconducting with a small energy gap of 40 meV at RT. The temperature-dependent band structures shown in Figs. 5(b)–5(g) were measured at near k_F=1.07 Å^{-1} of the S_3 band. The In adsorption at RT results in the disappearance of the S_3 band, which is partly due to the disordering of the surface observed by LEED. With increasing annealing temperature, the new energy band S_3' evolves, which disperses approaching k=1.25 Å^{-1} at the Fermi level. The intensity of the S_3' band was optimized at 290 °C. At higher temperatures, the S_3' band fades away with the concomitant reappearance of the S_3 band. The temperature dependence of the S_3' band is consistent with that of the 1×2-In LEED pattern, which proves that the S_3' band originates from the 1×2-In surface.

Figure 6 shows the whole band structure along the wires of the 1×2-In surface prepared at the optimized temperature. Four energy bands were found to be located within the

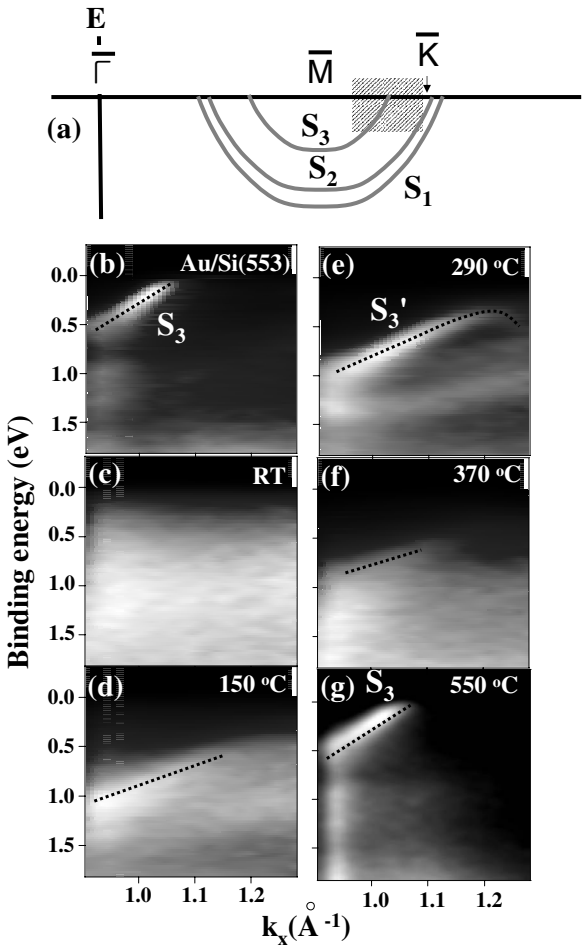

FIG. 5. (a) Schematic diagram of the band structure of the 1 ×2-Au surface and [(b)–(g)] the temperature dependence of the band structure of the In-adsorbed 1×2-Au surface near the shaded region in (a) from RT to 550 °C, which were measured with $h\nu$ =21.2 eV. Here, the dashed lines are drawn as a guide.

bulk band gap. Therefore, they are safely assigned as surface bands. Two of them (S_1' and S_3') are dispersive with bandwidths larger than 1.5 eV, while others (S_4 and S_5) are very flat. The S_1' and S_3' bands with a band minimum at k =0.8 Å^{-1} approach k=1.40 and 1.25 Å^{-1}, respectively, as they disperse to a low energy and are insulating with energy gaps of about 0.60 and 0.35 eV, respectively. The backfoldings of the S_1' and S_3' bands at the Brillioun zone boundary (k=1.23 Å^{-1}) of the ×2 period along the wires are not obvious but are expected to occur in order to satisfy the clear ×2 period observed in LEED.

The band structure of the 1×2-In surface can be understood by two possible scenarios. The first is that the energy bands have completely different origins from those of the 1 ×2-Au surface due to the In-induced significant reconstruction. The second is that the In adsorption modifies the band structure of the 1×2-Au surface without dramatically changing its characteristics. We further consider how the In adsorbates can modify the electronic structure of the 1 ×2-Au surface. The energy dispersions of the S_1' and S_3' bands are quite similar to those of the S_1/S_2 and S_3 bands of

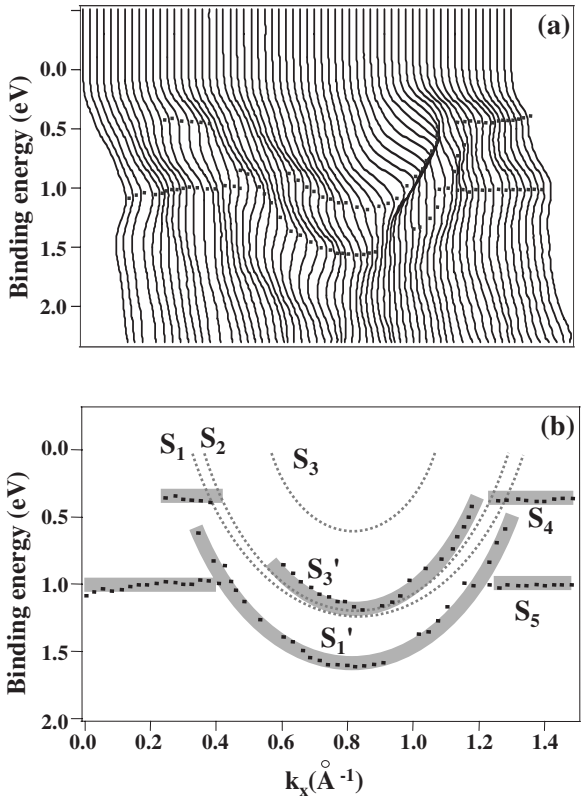

FIG. 6. (a) Energy bands of the 1×2-In surface measured with $h\nu=21.2$ eV, which are redrawn in comparison with those of the 1×2-Au surface in (b). The peaks of the energy distribution curve in (a) are indicated by the solid rectangles. In (b), the energy bands of the 1×2-Au surface are drawn by the dotted lines and the energy bands of the 1×2-In surface are guided by the thick solid lines.

the Si(553)-Au surface, respectively, as shown in Fig. 6(b).[4,12] For this reason, the S_1' and S_3' bands can be believed to form through rigid downward shifts of the S_1/S_2 and S_3 bands of 0.35 and 0.50 eV, respectively. This suggests that the valence electrons of In adsorbates saturate the partially filled dangling-bond-like states of the 1×2-Au surface through adsorbate-substrate bond formation. Another explanation is that the S_3' band is equivalent to the S_1/S_2 bands and the S_1' band is created at the expense of the S_3 band. This suggests that the 1×2-Au surface is reconstructed significantly by the In adsorbates.

The S_3' and S_1' bands of the 1×2-In surface are located between binding energies of 1.5 and 0.35 eV. In addition, STM images with a sample bias (V_s) ranging from −0.2 to −1.0 V show only the terrace and step edge atomic chains [see Fig. 4(c) for $V_s=-0.2$ V]. This leads naturally to the conclusion that the S_3' and S_1' bands originate from the two chains. Similarly, the origins of the parabolic bands of the 1×2-Au surface are also ascribed to the terrace and step edge atomic chains [see Fig. 4(a)].[4] The terrace and step edge atomic chains of both the 1×2-Au and 1×2-In surfaces were observed in both the filled-state and empty-state STM images, as shown in Fig. 4. Furthermore, the 1×2-In surface has nearly the same distance between the two atomic chains with that of the 1×2-Au surface. From the resemblances between the 1×2-In and 1×2-Au surfaces, it is more reasonable to interpret that the S_1' and S_3' bands have similar origins to the S_1/S_2 and S_3 bands, respectively. The remaining question is how the band structures before and after the In-induced uniform terrace widening can be similar. The electronic and atomic structures of the Au-adsorbed stepped Si surfaces produce some indications.[16] The number and band fillings of the dispersive 1D energy bands on the Au-adsorbed stepped Si surfaces depend on their step directions, where their terrace widths only change their band fillings slightly.[16] Both the Si(335)-Au and Si(557)-Au surfaces have two half-filled bands and both the Si(553)-Au and Si(775)-Au surfaces have two half-filled bands and a single third-filled band. The uniform widening of the 1×2-Au terrace by a single lattice constant makes the Si(553) surface transit into the Si(332) surface, where terrace widening by two lattice constants produces the Si(775) surface.[16] This can support that the 1×2-In surface has a similar band structure to the 1×2-Au surface and its surface bands originate from Si structures rather than from Au or In structures, as found on the 1×2-Au surface. On the other hand, the band fillings can also be changed by electron transfer from the In adsorbates passivating the 1×2-In surface. In any case, the atomic structure of the 1×2-In surface needs to be determined before a decisive assignment of the origins of the S_1' and S_3' bands can be made.

In summary, the In-induced atomic and electronic phase transitions of the Si(553)-Au surface were studied using the combined LEED, STM, and potential energy surface experiments. The In adsorption results in the uniform terrace widening of the Si(553)-Au surface across the wires by 0.32 nm, which agrees closely with a single unit cell of the Si substrate. Furthermore, the In adsorbates drive $\times2$ order along the wires similar to the 1×2-Au surface but its coherent length across the wires is much longer than that of the 1×2-Au surface. The single wire unit cell of the 1×2-In surface with a width of 1.80 nm is composed of two different types of atomic wires located at the step edge and within the terrace, respectively, which is in close resemblance to the 1×2-Au surface. The step edge and terrace atomic chains produce two highly dispersive 1D bands with an energy gap of 0.35 and 0.6 eV, respectively. Therefore, In adsorbates drive the two metallic atomic chains on the 1×2-Au surface to change into the two insulating atomic chains with the same degree of anisotropy in the band structure.

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*jrahn@skku.edu
†yeom@yonsei.ac.kr

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