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Direct evidence of the step-edge buckling at the Au/Si(557)-1×2 surface

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We have investigated atomic structure and electrical properties of the Au/Si(557)-1×2 surface by using scanning tunneling microscopy. We observe the doubled periodicity (×2) for the step-edge atoms even far away from defects at room temperature (RT), indicating no Peierls-type transition reported earlier. We further identify the Au atoms well resolved from Si atoms in the Au-Si-Au chain at RT, in good accord with the prevailing structural model. Our scanning tunneling spectroscopy data taken along the step-edge atoms unambiguously reveal that these step-edge Si atoms are metallic, and are buckled apparently with a charge transferred from down to up Si atoms. We find no significant thermal fluctuation of the buckled step edges at RT.

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One-dimensional (1D) nanowire in its ideal form can be a good playground in studying various exotic properties due essentially to the enhanced electron-electron and electron-phonon interactions because of its reduced dimensionality.[1] The Au/Si(557)-1×2 surface may be one of the quasi-1D systems self-assembled on semiconducting surfaces, showing two neighboring proximal bands near Fermi level.[2–13] The origin of these two proximal bands has been a subject of continued studies. At first, they have been attributed to the spin-charge separation of Luttinger liquid,[2] which has been ruled out by a subsequent angle-resolved photoemission spectroscopy (ARPES) study with an improved resolution.[3] Later, the system has been reported to undergo a charge-density wave (CDW) phase transition based on ARPES and scanning tunneling microscopy (STM) measurements.[4] This work reported that only one of the two bands is metallic while the other has a gap at room temperature (RT). One of the two chains in their STM images attributed to the step-edge atoms exhibits a periodicity doubling upon cooling undergoing a Peierls-type metal-insulator transition. Meanwhile theoretical calculations suggest that these bands are due to the spin-orbit splitting (Rashba effect) of a 1D band associated with the gold chains[5,6] and such changes in STM and ARPES upon varying temperature are likely to come from the thermal fluctuation of the step-edge atoms at RT, which may be frozen at low temperature (LT).[7]

X-ray diffraction (XRD) measurements[8] and first-principles calculations[9] suggested a structural model, which explains most of characteristic features of this system such as adatoms and step-edge atoms observed in STM. The atomic structure in the step edge, however, has been controversial, one study reports a ×1 periodicity at RT with a sparse ×2 periodicity only near defects while other shows the overall ×2 at RT suggesting no Peierls-type transition[11] or a different atomic structure.[12] Furthermore, our recent ARPES data do not indicate any evidence for the presence of a metal-insulator transition.[13] Therefore the origin and characteristic features of the two proximal bands of the Au/Si(557)-1×2 surface still remain under dispute. We here present evidence suggesting the metallic nature of the step-edge atoms arranged with the ×2 periodicity already at RT, not critically affected by the defects. We further show the presence of the step-edge buckling apparently with a charge transfer from down to up Si atoms. We compare the atomic arrangement of the buckled step edge with a theoretical model and discuss its electrical as well as thermal properties. Interestingly our STM images exhibits atomically well-resolved images of the Au-Si chains supporting the structural model proposed earlier.[8,9]

We have obtained our STM and STS data with a commercial STM (from SPECS) machine under a base pressure of 2.0×10^{-10} Torr. We used *n*-doped Si(557) wafer of resistivity 1–10 Ohm cm. The clean Si(557) surface was obtained by repeatedly flashing the sample up to 1250 °C followed by annealing at 850 °C, which appeared to be clean as confirmed by our STM image (not shown). We then deposited Au atoms for a coverage of 0.2 ML with the sample maintained at 650 °C. The sample was then annealed at 850 °C for about 30 s. The atomic and electronic structures of the Au/Si(557)-1×2 surface thus prepared have been studied by measuring STM images in a constant-current mode and STS data using an argon sputtered- and thus oxide removed-tungsten tip. The tip was moved along the direction parallel to the atomic chains to acquire the highest possible spatial resolution.

Vicinal Si(111) surfaces stabilized by Au atoms are hardly formed without defects because the surfaces contain both unstable steps and stable terrace structures. We find typically three types of defects, the excess Si atoms appearing as bright protrusions in STM images and two different vacancies appearing as dark vacancies along the step-edge and the adatom chains.[10] In order to maximize the domain size with reduced defects, the current direction has been chosen along the direction parallel to the length of the wire. Au has been slowly deposited with a constant flux while the sample was cooled with a rate slower than 1 °C/s.

In Fig. 1 we show the empty state STM images of a wide terrace (1500×1124 Å^2) (a) and of an enlarged image for a small area (300×141 Å^2) (b) with low defect density on the well ordered Au/Si(557)-1×2 surface. The number of the bright protrusions in (b) is approximately 18, less than half of the defects in previous STM work (45 protrusions over about 220×220 Å^2 region).[10] The line profile of a step edge marked by two white triangles in (b) is presented in (c). The

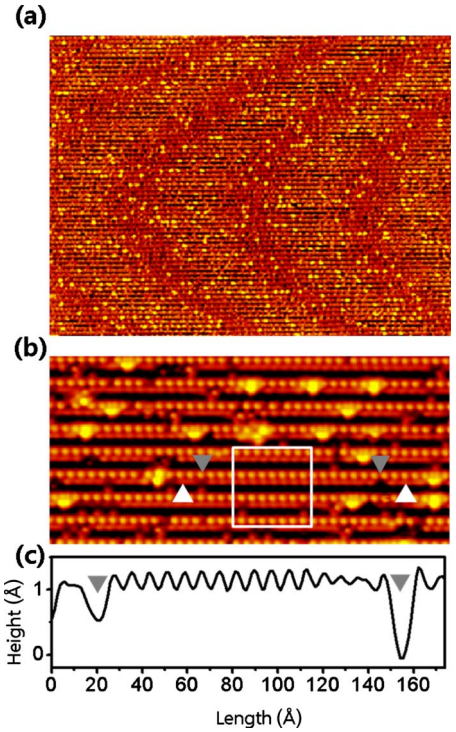

FIG. 1. (Color online) (a) A large scale empty state STM image (I_t=0.75 nA, V_s=1250 mV, 1500×1124 Å^2) taken at RT (b) Enlarged empty state image (I_t=1.0 nA, V_s=968 mV, 300×141 Å^2) taken from an area with reduced defects. (c) Line profile along the step edge marked by two white triangles in (b) showing a well defined doubled (×2) periodicity. The distance between two defects (reversed gray triangles) is about 13 nm.

wire between two defects, indicated by the reversed gray triangles, is about 13 nm long. Here, one clearly finds the ×2 periodicity in the line profile almost not affected by the defects. As mentioned earlier previous STM study shows a ×1 periodicity at RT, which becomes a ×2 periodicity through a Peierls-type metal-insulator transition below a transition temperature T_c=120 K.[10] They claimed that the ×2 periodicity was observed locally at RT only around defects, whereas other STM works reported the dominating ×2 periodicity over a large area at RT.[11,12]

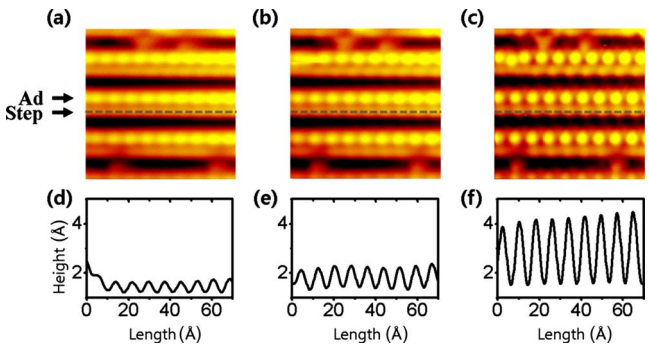

FIG. 2. (Color online) Empty state images (V_s=968 mV, 70×70 Å^2) and corresponding line profiles along the step edge [marked by the dotted lines in (a) to (c)] taken with three different tunneling currents ((a) 0.08 nA, (b) 0.14 nA, and (c) 1.00 nA).

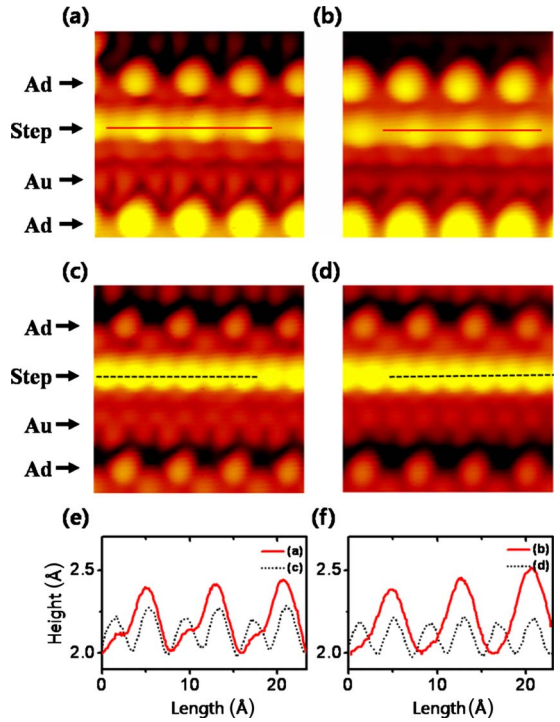

FIG. 3. (Color online) Empty state images ((a) I_t=1.03 nA, V_s=423 mV, (b) I_t=1.07 nA, V_s=734 mV) and filled state images ((c) I_t=−1.03 nA, V_s=−423 mV, (d) I_t=−1.07 nA, V_s=−734 mV) taken from a defect free region marked by the white box in Fig. 1(b). We find several features not observed before, such as the extra Si and Au rows at the bottom of the step and the buckling of the step-edge. Line profiles along the step edge ((e) and (f)) marked by the red (gray) solid lines and black dotted lines.

Figure 2 shows the empty state images taken with three different tunneling currents (a–c) and the corresponding line profiles (d–f) along the step edge (marked by the dotted lines in (a– c)) in a defect free region [white square in Fig. 1(b)]. As the tip gets closer to the surface in STM, one obtains more accurate geometrical information with an increased tunneling current. As shown in Figs. 2(d)–2(f), the modulation becomes more prominent as tunneling current increases. We thus find the presence of the ×2 modulation along the step edge with a much increased tunneling current than used in previous STM measurements. This strongly indicates that the step-edge atoms have a geometrical modulation different from that of the ×1 structure already at RT. We further emphasize that our STM images have been taken from a defect free region so that there is apparently no defect-induced periodicity doubling as claimed earlier.[4] One may also argue that the tip may influence the atoms during closer approach, acting like a defect that triggers the ×2 modulation. If the ×2 modulation is generated by the tip, one would expect to see both possible phases of the ×2 modulation, not exclusively one, as shown Fig. 1(b).

Previous STM studies[4,10–12] have identified mostly the adatoms and step-edge atoms in good agreement with XRD measurement as well as theoretical results. In Fig. 3, we show empty state images [(a) and (b)] and filled state images [(c) and (d)] taken with four different bias voltages at RT. The scanning area is 30×30 Å^2. The empty state images

exhibit the well-resolved adatoms as well as the step-edge atoms with the ×2 periodicities. One also notices that the images further show two more lines which are identified as Si atoms just below the step-edge atoms and Au atoms when compared to the structural model.[8,9] This is the observation revealing all the detailed terrace structure of the Au/Si(557)-1×2 surface including the Au atoms at RT. The filled state images also show additional well-resolved surface structures. Since the atomic structure observed in our STM images agrees quite well with the structural model,[8,9] we thus safely discuss the features around step edge within the frame work of the structural model. (e) and (f) are the line profiles of the step edge in the empty and filled state images for bias voltages $V_s = \pm 423$ mV and ± 734 mV, respectively. The periodicity of the topographic maximum at the empty state images appears to be ×2. Alternating shoulders having the same periodicity are additionally observed at the bias voltage $V_s = +423$ mV. The intensity associated with the shoulders increases upon changing the bias voltage from positive to negative. Interestingly the step-edge line in the filled state image at $V_s = -734$ mV reveals almost ×1 periodicity probably with a very weak ×2 modulation. We could not completely reverse the intensities from the step-edge atoms by either changing the bias voltage[12] or the defects mentioned above in our STM experiment.

Tunneling current mainly depends both on the distance between sample and tip and the decay length.[14,15] If the distance or the decay length is not enough to allow the overlap between the wave functions of the sample and of the tip, there appears no feature in STM images. Since the parabolic bands of the Au/Si(557)-1×2 surface have a considerable momentum parallel to the surface, the decay length of the wave function should be short. In order to allow a tunneling process to occur between the wave function of the sample with a short decay length and that of the tip, the distance has to be reduced. Therefore, we have used a high tunneling current of 1.0 nA to obtain a well-resolved detailed feature in the middle of the terrace.

In order to identify the electrical properties of the step-edge atoms, we have taken the conductance dI/dV maps as shown in Fig. 4(a) taken at various bias voltages over the region covering the step edge as well as the adatom lines. The scanned area is 20×20 Å^2. The number of spatial points to obtain the dI/dV maps over the scanning region is 32 points ×32 points. The I/V spectra have been measured at all the spatial points for a chosen tunneling current of 0.03 nA and a bias voltage ranging from −0.129 V to +0.208 V. The dI/dV spectra, containing the information of density of states (DOS) mapped on a bias voltage plane, have been obtained by numerically differentiating the tunneling current versus voltage $I(V)$ curves. STM images with atomic resolution have also been taken during the STS measurements (not shown). We find a negligibly small drift and no significant change on the tip during the scans. Interestingly the dI/dV maps displayed in Fig. 4(a) shows the feature corresponding only to the step-edge region. The DOS associated with the atoms of the step edge shows the spatial difference in intensity with respect to the applied bias voltage. One notes that the intensity of the dI/dV map for the filled states has been shifted downward in contrast to the upward shift for

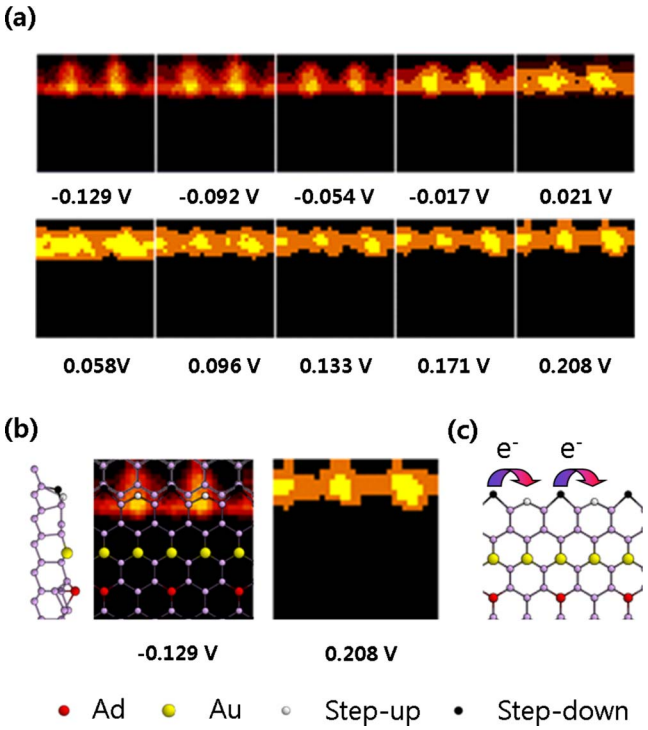

FIG. 4. (Color online) (a) dI/dV maps taken at various bias voltages. (b) two dI/dV maps measured at the filled state (−0.129 V) (left) and at the empty state (0.208 V) (right). Side and top views of the structural model of the Au/Si(557)-1×2 surface is attached to the dI/dV map of the filled state. One notes significant DOS near the Fermi level for all the bias voltages indicating the metallic nature of the step-edge chains. Red (big dark gray), yellow (big gray), white (light gray), and black circles represent adatoms, Au atoms, step-up, and step-down atoms, respectively. (c) Schematic drawing illustrating the charge-transfer process from the step-down to the step-up atoms.

empty states. It is important to note the significant DOS for all the bias voltages in Fig. 4(a) suggesting the metallic nature of the step-edge atoms around the Fermi level. In order to identify the locations of each atom in the dI/dV map, we have overlapped the dI/dV map of the filled state over the well-known structural model of the Au/Si(557)-1×2 surface with a bucking in the step-edge Si chains. One clearly notices that for the filled (empty) state, DOS is mostly localized at the "step-up" ("step-down") atoms among the step-edge Si chain. This may be understood in terms of a charge transfer from the "step-down" atoms to the "step-up" atoms accompanied by the buckling of the step-edge Si chains. Consequently, the "step-up" atoms are electron rich and the "step-down" atoms are electron poor. We note that this interpretation results in the atomic arrangement slightly different from that proposed by Riikonen & Sánchez-Portal.[7] Their up-atoms appear to be our down-atoms and vice versa for their down-atoms. Figure 4(c) illustrates the electron charge-transfer process between the two types of atoms of the buckled step edge. The good reproducibility of our STS data suggests no prominent thermal fluctuation for the step-edge atoms at least at RT. Unfortunately we have not been able to obtain any reliable STS data at LT because of the significant

thermal drift induced by the temperature difference between the tip and the sample.

Previous ARPES experiments for the Au/Si(557) surface showed two parabolic bands, crossing the Fermi level.[4,13] Similar proximal bands have also been observed for the Au/Si(553) surface.[16,17] Recent theoretical and experimental studies account for the presence of such proximal metallic bands of the systems mainly by the spin-orbit splitting of the one-dimensional Au-Si chains.[5,6,18] Our recent ARPES data also supported such an explanation, which also showed the presence of a satellite peak in the Au $4f$ core-level spectrum reflecting a plasmon excitation in the metallic Au-Si chains.[13] However, using STM we find no direct evidence for the metallic character of the Au chains by themselves. Since the decay length of wave function is inversely proportional to the momentum parallel to the surface, the wave function of the parabolic bands of the Au/Si(557)-1×2 surface, which cross the Fermi level at a higher momentum, would have a short decay length. This explains why the Au-Si chains showed no metallic character in the dI/dV map.

In summary, we have studied properties of the Au/Si(557)-1×2 surface focused on the nature of the step-edge atoms by carefully obtaining STM and STS data with an enhanced spatial resolution at RT. We find that the $\times2$ periodicity dominates both along the adatom chains and the step-edge chains not much affected by the presence of defects. This observation is in sharp contrast to the previous study reporting the prevailing $\times1$ periodicity along the step-edge chains with a sparse $\times2$ periodicity only near the defects at RT. We thus find no evidence for a possibility of a Peierls-type metal-insulator transition to induce the $\times2$ periodicity at temperatures below Tc=120 K as claimed earlier. Our STS data further show the presence of significant DOS near the Fermi level indicating the metallic nature of the surface and also reveal the significant buckling of the step-edge chains due to the charge transfer between up- and down-atoms. With the enhanced spatial resolution in our STM images, we further identify the presence of Au atoms in the Au-Si-Au chains well separated from their neighboring Si atoms, which has not been identified in earlier STM studies.

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