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Electronic transition of Au-induced atomic chains on Si(5 5 12): A strong similarity with the Au/Si(5 5 7) system

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We investigated in detail the electronic structures of the Si(5 5 12) surface with Au adsorbates, which features one-dimensional (1D) atomic chains and a 1D metallic band. From angle-resolved UV photoelectron spectroscopy, we resolved that the nearly half-filled 1D band is actually composed of two parallel branches; one is metallic, but the other is insulating with an energy gap of 50 meV below E_F at room temperature. An energy gap evolves gradually from 270 K on the metallic branch, saturating at 75 K with a size of 40 meV below E_F, while the insulating branch is intact. These behaviors are essentially identical with those recently reported on the Au/Si(5 5 7) system [Phys. Rev. Lett. **91**, 196403 (2003)]. The scanning tunneling microscopy imaging indicates that these surfaces possess common 1D atomic chain structures, while the interchain distance is different. This explains the very similar electronic structure and transition of the two systems.

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Recently, various atomic-scale one-dimensional (1D) structures have been realized on solid surfaces, mostly in the form of atomic wires or chain structures[1–12] formed through self-organization processes. Some of these systems, especially on Si surfaces, were shown to indeed possess 1D (or strongly anisotropic) metallic (or quasimetallic) electronic bands.[1–6] Moreover, a few interesting and exotic 1D physical properties have been suggested to show up on the atomic wire (chain) systems, such as the 1D charge-density-wave formation through Peierls instability for In chains on Si(1 1 1) (Ref. 1) and Au-induced chains on Si(5 5 7),[6] and the realization of atomic-scale memory on Au-induced chains on Si(1 1 1).[11]

In particular, vicinal Si surfaces with regular and uniform step arrays have been used to prepare 1D chain structures with Au adsorbates.[2,3,5–7,9,10] Various substrates were used, such as Si(5 5 7), Si(7 7 5), Si(5 5 3), Si(3 3 5), and Si(5 5 12), which have different interchain distances and different atomic structures; the detailed atomic structures of the above systems are not clear yet. Electronic band structures of the Au-induced chain structures are also different over the different substrates, while one common aspect may be the presence of nearly half-filled 1D parabolic bands approaching the Fermi level.[2,3,5–7] For the Si(5 5 7) (Refs. 3 and 6) and Si(5 5 3) (Ref. 5) surfaces, two very close and parallel bands were clearly resolved, which were originally suggested as the sign of the spin-charge separation of Luttinger liquid.[2] However, two more recent angle-resolved photoemission (ARP) studies showed that the band dispersions of the two proximal bands do not follow the theoretical prediction for the spin-charge separation case; the two bands have different Fermi velocities instead of a unique one, that is, a merged band at Fermi level, expected for Luttinger liquid.[3,6] It was further shown that the two proximal 1D bands of Au/Si(5 5 7) have characteristically different behaviors; one band is metallic and the other is gapped at room temperature.[6] More importantly, the metallic branch was found to undergo a Peierls-type metal-insulator transition.[6] The phase transition was clearly related to a specific element of the 1D chain structure through scanning tunneling microscopy (STM) and spectroscopy (STS) measurement.[6,13] It is thus of interest as to how the metallicity and the ground states vary over the different chain systems with different band structures in relation with the inherent instability and non-Fermi liquid nature of a 1D metallic system.

In this paper, we focus on the 1D chain system induced by Au on Si(5 5 12). It was reported that Au adsorption on Si(5 5 12) at high temperature induces a rearrangement of the step structure into a regular array of (3 3 7)×2 facets,[4,10,12] which accompanies the change of Au adsorption structure and the metallization of the surface.[12] The resulting system, called Au/Si(3 3 7)×2 hereafter, was reported to feature a "single" 1D metallic band.[4] If this report is correct, then this system could be the one with the simplest band structure among the 1D chain systems mentioned above, and thus can be a system of choice to investigate the noble 1D electronic behaviors. However, in the present high-resolution ARP measurements, we observed that the 1D metallic band is actually composed of two parallel branches, which are very similar to those of Au/Si(5 5 7) (Refs. 3 and 6) and Au/Si(5 5 3).[5] As in the case of Au/Si(5 5 7) reported recently, only one branch is metallic, the other being gapped at room temperature, and the metallic branch undergoes a gradual metal-insulator transition upon cooling. The origin of this similarity is explained from the identical 1D building block structure of these systems, presumably the Si step edge, as suggested by the STM imaging.

The photoemission measurements were performed using a high-resolution electron analyzer (SES-100, Gamma Data) and He I radiation ($h\nu = 21.2$ eV) at a base pressure of 4×10^{-11} Torr. The nominal energy and angular resolution are better than 13 meV and 0.2°, respectively. A Si(5 5 12) sub-

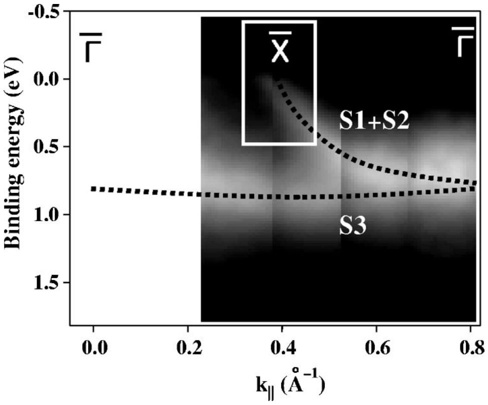

FIG. 1. Photoelectron intensity map of the surface state bands S1, S2, and S3 of Au/Si(3 3 7)$\times$2 measured at 300 K in gray scale as function of binding energy (as referenced by E_F) and momentum along the 1D chain $k_{\|}$. Dotted lines are drawn as guides for the surface state bands.

strate was cryogenically cooled down to 75 K and could be maintained at a specific temperature stably by a feedback-controlled heater. STM images were obtained by a commercial STM apparatus (Omicron, Germany). The Si(5 5 12) surface is miscut by 30.5° from the [0 0 1] to the [1 1 1] orientation and has a unit cell consisting of two (3 3 7) and one (2 2 5) facets. The Au/Si(3 3 7)$\times$2 surface with a regular 1D chain array was generated by depositing Au on a Si(5 5 12) surface held at 650 °C,[4,10] as confirmed by the characteristic low energy electron diffraction patterns with clear noninteger spots and consistent STM images. This surface is composed of a unit of two (3 3 7) facets in which the interchain distance is 3.14 nm. That is, the surface prepared with Au adsorbates and high-temperature annealing has a different step arrangement as well as a different Au adsorption structure, as revealed by core-level photoemission.[12]

The Au/Si(3 3 7)$\times$2 surface was reported to show a highly dispersive single metallic band by ARP.[4] This report is confirmed as shown in Fig. 1. A parabolically dispersing band seems to cross E_F near the zone center ($\overline{X}$) and another very flat band (denoted as S3) appears at a binding energy of 0.9 eV. Similar bands at 0.9 eV were observed for other 1D chain systems on stepped Si surfaces with Au.[4,5] However, a closer look with higher energy and angular resolution than before reveals more complex details of the band near E_F.

Figure 2(a) shows the detailed ARP spectra near the E_F-crossing of the dispersing band. These data clearly indicate that the dispersive metallic band in Fig. 1 and in the previous report is *split into two proximal and parallel bands*. The adjacent bands have an energy spliting of 0.23 eV and cross E_F at $k_{\|}=0.35$ (denoted as S1) and 0.40 (S2) Å^{-1}, respectively. These band dispersions are almost identical with those observed on Au/Si(5 5 7) as redrawn in Fig. 2(b) for comparison;[6] the adjacent bands are resolved better on Au/Si(3 3 7)$\times$2. The 1D chain system of Au/Si(5 5 3) was also reported to show a very similar two-band structure, but with an extra quarter-filled band.[5] As discussed previously for Au/Si(5 5 7), these band dispersions do not fit with the spin-charge separation behavior in Luttinger liquid.[6]

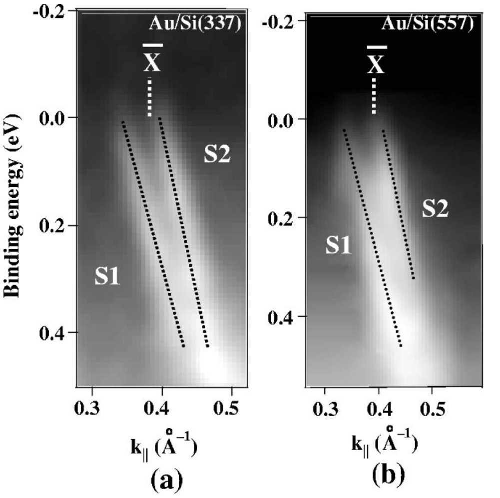

FIG. 2. Two highly dispersive S1 and S2 bands of (a) Au/Si(3 3 7)$\times$2 and (b) Au/Si(5 5 7) at 300 K in the photoelectron intensity map in gray scale along the 1D chain $k_{\|}$ near the zone center ($\overline{X}$).

Since these bands are 1D and nearly half-filled, one may expect Peierls instability at a sufficiently low temperature, which was already observed on Au/Si(5 5 7).[6] Figure 3(a) shows the *k*-resolved energy-distribution curves (EDC) of the photoemission spectra at the Fermi-level crossings (i.e., at k_F) for the S1 and S2 bands. It is shown that only the S1 band is metallic at room temperature with a Fermi-Dirac dis-

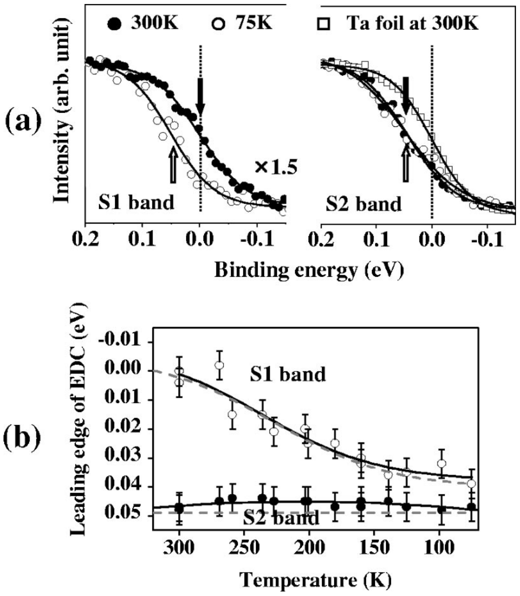

FIG. 3. (a) EDC's of the photoemission intensities for the S1 and S2 bands at the corresponding k_F's of 0.35 and 0.40 Å^{-1} on Au/Si(3 3 7)$\times$2, respectively, and (b) temperature dependence of the center-of-the-leading-edge positions of EDC's [see the arrows in (a)] for S1 and S2 on Au/Si(3 3 7)$\times$2 at the corresponding k_F positions. Similar results for the Au/Si(5 5 7) system are drawn as dotted lines (Ref. 6). The EDC of a typical normal metal (Ta) is given for comparison in (a) and, due to the low intensity of S1 at E_F, its EDC in (a) is enlarged by a factor of 1.5.

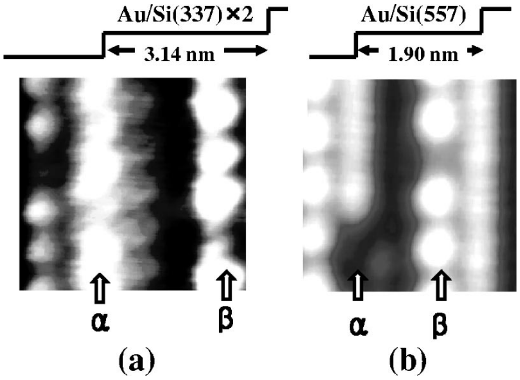

FIG. 4. Empty-state STM images of (a) Au/Si(3 3 7)×2 and (b) Au/Si(5 5 7) (taken from Ref. 6) with a sample bias of V_s =1.0 V at 300 K, where the terrace sizes of the stepped surfaces are drawn at the top of each image for comparison.

tribution of the spectral weight while the S2 band has a significantly reduced intensity at E_F; compare the EDC's of S1 and S2 with that of a normal metal Ta (open squres). The leading edge of the S2's EDC is shifted from E_F by 50 meV, as indicated by the solid arrow in the figure. It is thus likely that the S2 band is insulating with a bandgap of 100 meV assuming a symmetric gap centered at E_F.

At a lower temperature, the spectral weight of S1 at E_F decreases gradually with the shift of the EDC leading edge, indicating clearly a metal-insulator transition. The leading edge shifts were quantitatively measured as function of temperature, as shown in Fig. 3(b). The bandgap gradually develops from about 270 K and saturates at about 130 K with a size of 40 meV below E_F. This bandgap is rather clear in the EDC, as shown in Fig. 3(a). During this measurement, the surface photovoltage shift was corrected carefully by comparing an angle-integrated spectra with Au 5*d* and Si bulk features. In clear contrast, the EDC of S2 does not exhibit any noticeable change keeping insulating with the same energy gap. As also shown in Fig. 3(b) in dashed lines, this temperature-dependent behavior mimics that of Au/Si(5 5 7) and is well fitted with the mean-field theory expectation for S1. That is, the energy gap is fitted well with the BCS function, $\Delta(T)/\Delta(T=0)=(1-T/T_c)^{1/2}$, providing a consistent value of a transition temperature of $T_c=270$ K, where $\Delta(T)$ is an energy gap at T. It is thus obviously concluded that the 1D chains of Au/Si(3 3 7)×2 has the same electronic band structure and transition with those of Au/Si(5 5 7). One difference noticed is that the EDC of the insulating band has some appreciable intensity at E_F for Au/Si(3 3 7)×2 both at room and low temperature. The origin of the spectral intensity, which is independent of temperature, is not understood yet, but we move into the discussion of the origin of the similarity between Au/Si(3 3 7)×2 and Au/Si(5 5 7).

The proximity in the band structures of Au/Si(5 5 7) and Au/Si(3 3 7)×2 can be understood from their atomic structures, as revealed by STM measurements shown below. Figure 4(a) shows empty-state STM images of the Au/Si(3 3 7)×2 at room temperature. The Au-induced facets exhibit a period of 3.14 nm, that is, the terrace size of two (3 3 7) facets along $[66\bar{5}]$. The image further reveals two nonidentical chains (α and β) on a single (3 3 7)×2 unit. Besides, the two chains have obviously different periodicity; the α and β chains have ×1 and ×2 periodicity along $[\bar{1}10]$, respectively, which was also observed on Au/Si(5 5 7), as shown in Fig. 4(b). This indicates that Au/Si(3 3 7)×2 contains the same 1D structural building blocks with Au/Si(5 5 7).[6,15]

On Au/Si(5 5 7), first-principles density functional calculations showed the α and the β chains correspond to a Si step edge and a Si-adatom chain, respectively.[14] Although no structural model has been available for Au/Si(3 3 7)×2, we straightforwardly extend this interpretation to the case of Au/Si(3 3 7)×2 from the similarity of the STM images.[6] Since the extensive STS study on the Au/Si(5 5 7) surface indicated that the metallic band (S1) is related to the α chain (presumably the step edge Si atoms),[13] the similar band structure, at least the common S1 band, is natural. The metal-insulator transition is expected subsequently since the α chain was observed to undergo periodicity doubling transition upon cooling on Au/Si(5 5 7). The band gap opening in the density of states localized on the α chain was also confirmed through STS measurements.[13] That is, the very similar electronic transition of these two 1D systems is understood from the common metallic chain structure (the α chain) within them. Note further that the metal-insulator transition does not seem to be affected by the difference of the interchain distances between each 1D unit; 3.14 and 1.90 nm on Si(3 3 7)×2 and Si(5 5 7), respectively. In more detail, the separation between the α and β chains is also different; 1.05 and 0.63 nm on Si(3 3 7)×2 and Si(5 5 7), respectively. These separations are equally one-third of the terrace size of each surface although the reason for this and other structural details are not at all clear as yet. Further studies on the atomic structures of these surfaces are highly required.

In summary, in contrast to the previous ARP reports, the dispersive metallic band of Au/Si(3 3 7)×2 is resolved into two bands by high resolution ARP. Unexpectedly, the proximal bands are composed of a metallic and an insulating band at room temperature as observed on Au/Si(5 5 7).[6] The similarity can be understood from the existence of the same Au-induced 1D chain structures, especially the step edge Si chains on both surfaces, as revealed by the STM measurements. Upon lowering the temperature, the metallic band becomes insulating with an energy gap of 40 meV below E_F at 75 K. This behavior is also consistent with the metal-insulator transition on Au/Si(5 5 7) due to the periodicity doubling on the step edge Si chains. It is interesting enough that similar 1D bands have been observed on various 1D Au-decorated Si stepped surfaces, Si(3 3 7), Si(5 5 7), and Si(5 5 3). However, the origin of the dispersing bands, especially the insulating band S2, is unknown as yet, and the pseudogaplike EDC of S2 on Au/Si(3 3 7)×2 is to be investigated further.

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