Metal-Insulator Transition in Au Atomic Chains on Si with Two Proximal Bands

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One-dimensional atomic chains on Au/Si(557) feature two proximal 1D bands near the Fermi level, which were controversially attributed as a spinon-holon pair of a Luttinger liquid. Angle-resolved photoemission shows that only one band is metallic with the neighboring one gapped at room temperature. Furthermore, even the metallic branch is found to undergo a metal-insulator transition upon cooling, which follows a mean-field-type behavior. Scanning tunneling microscopy observes two apparently unequivocal chains on the surface, one of which exhibits periodicity doubling accompanying the metal-insulator transition. The surface 1D structure is thus concluded to be insulating at low temperature with a Peierls-type instability.

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One-dimensional (1D) *metallic* electron systems have attracted much attention due partly to their inherent instability, which leads to various broken symmetry ground states such as charge or spin density wave (CDW or SDW) and singlet or triplet superconductivity [1,2]. While all such ground states are the results of electron-phonon interaction, finite electron-electron interaction brings about an even more exotic state—the breakdown of the Fermi-liquid state with quasiparticles, which should then be described by the Luttinger liquid (LL) framework [3,4]. In contrast to the conventional bulk quasi-1D systems, 3D organic or inorganic crystals with strongly anisotropic atomic and electronic-band structures [5,6], recent studies showed that various atomic-scale 1D systems can be fabricated on solid surfaces [7–14]. A few such atomic chain systems on surfaces were shown to be metallic, especially for those on semiconductor surfaces such as Si, and, indeed, 1D metallic properties such as CDW [7] and LL [8] behaviors were discussed very recently.

Using a vicinal-cut surface with a regular step array provides a greater degree of freedom in fabricating 1D chain structures, where one may be able to control the interchain distance. Some recent efforts with vicinal Si surfaces were indeed successful; the 1D chain structures formed by Au on Si(557) [9], Si(553) [10], and Si(5 5 12) [11] were reported to feature 1D metallic bands with varying electron filling. Among them, the Au/Si(557) system was subject to special attention, where the two closely lying bands were observed with vanishing intensity at Fermi level (E_F) by angle-resolved photoemission (ARP) and were interpreted as the sign of spin-charge separation in LL (i.e., as the so-called spinon and holon bands). However, in contradiction with the fact that the spinon and holon bands of LL merge into one at E_F with a unique Fermi velocity, the later ARP study showed that the E_F crossings of the two bands are separated [9].

However, the origin of the proximal 1D metallic bands is still unclear, and they could not be reproduced by density functional calculations based on the available structural models [15–17]. Another question on this system concerns the ill-defined Fermi edges of the 1D bands in ARP spectra, which was originally attributed to the power-law behavior of the density of states (DOS) in LL [8] decorated with some effect due to the defects [18]. Most of all, one important and fundamental question is whether the metallic ground state of this half-filled 1D electronic system is really stable against Peierls instability as the previous ARP studies done at a low-temperature of 12 [8,18] or 100 K [9] assumed.

In the present Letter, in order to answer the above questions, we carefully reinvestigated the Au/Si(557) system with ARP and scanning tunneling microscopy (STM), focusing especially on the temperature dependence of the atomic and electronic-band structures. We first confirm the latest ARP result on the splitting of the two proximal bands at E_F [9]. While one of the two 1D bands exhibit clear Fermi-Dirac–type spectral shape on E_F at RT, ruling out the possibility of the power-law behavior [8] or the defect-induced Coulomb shift [18], the other band exhibits only marginal spectral weight. Furthermore, the one with normal metallic behavior at RT is found to show a metal-insulator transition, which saturates at 120 K with a band gap of 80 meV. STM imaging corroborates the phase transition indicating a periodicity doubling on part of the 1D chains at low temperature. It is thus concluded that the Au/Si(557) system is basically a normal 1D metallic system with an insulating ground state formed through a Peierls transition.

The photoemission measurements were performed using a high-resolution electron analyzer (SES-100, Gamma Data) and the He I radiation ($h\nu = 21.2$ eV) at a base pressure of 4×10^{-11} Torr. The nominal energy

and angular resolution is better than 13 meV and 0.2°, respectively. A Si sample 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 low-temperature STM apparatus (Omicron, Germany), where similar sample cooling and base pressure were available. A Si(557) surface with a regular 1D chain array was generated by annealing above 650 °C after the RT Au deposition onto the vicinal Si wafers [9], as confirmed by the LEED patterns with clear noninteger spots and consistent STM images.

Figure 1 shows the photoelectron intensities near E_F around $k_{\|} = 0.4$ Å^{-1}, which corresponds to the E_F crossings of the two 1D dispersing bands [9]. The two bands with *parallel dispersion up to* E_F are clear with an energy splitting of 0.3–0.4 eV and there is no sign for them to merge at E_F. As also shown in the momentum distribution curves of the photoelectron intensities of Fig. 1(b), the two bands have well separated E_F-crossing points of 0.35 and 0.41 Å^{-1}, which were determined to be the $k_{\|}$ points for the leading edges of the energy distribution curves (EDCs) approaching E_F closest. The crossing point of S2 coincides with the center of the Brillouin zone (the $\times 1$ zone) along the 1D chain direction, indicating half filling for S2 and slightly over half filling for S1. This result is fully consistent with that of Losio *et al.* [9] and confirms that the spin-charge separation expected in LL cannot explain the split bands.

Another experimental observation related to the LL behavior was the decaying spectral weight toward E_F without a well-defined Fermi cutoff [8,18]; in LL the spectral function follows a power-law increase from E_F as a function of binding energy. Thus, we checked the spectral shape of each band near E_F carefully. Figure 2 shows the EDC of each band at the expected Fermi crossings. At RT (solid circles), the EDC of S1 obviously exhibits huge intensity at E_F, which is essentially the same as that of normal metal Ta (rectangles) and is fitted excellently by a Fermi-Dirac function convoluted by a Gaussian (instrumental) broadening. Note that E_F is located exactly at the middle of the leading edge of the EDC for S1. This clearly denies the power-law behavior of LL proposed previously [8].

However, what is still puzzling is that the S2 band does not exhibit any clear Fermi cutoff (the intensity cutoff at E_F) for any $k_{\|}$ points. At the expected k_F, as shown in Fig. 2(a), the leading edge of its EDC is shifted from E_F by 50 meV. This can be interpreted as due to a band gap or to other effects such as the power-law behavior of LL or the defect-induced Coulomb shift [18] as discussed previously. We tentatively attribute this as due to a band gap opened already at RT, as discussed in detail below.

Although we confirmed the normal metallic behavior of at least one 1D band at RT, it is still crucial to check whether the ground state at a sufficiently low-temperature is metallic or not in order to understand the difference from the previous ARP study which observed no E_F

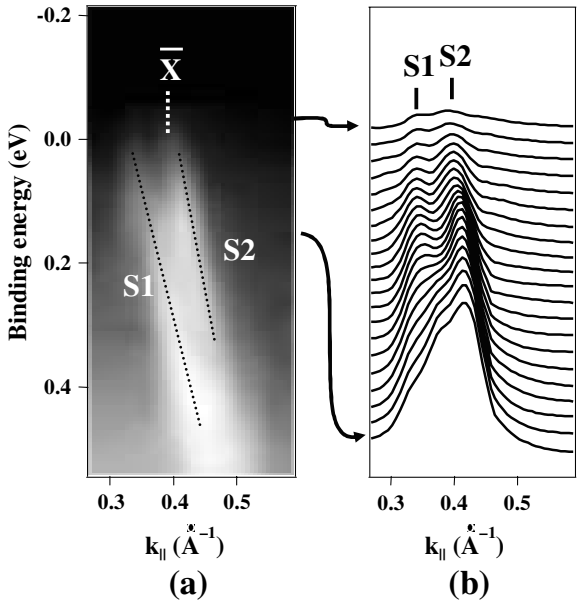

FIG. 1. Two highly dispersive S1 and S2 bands of Au/Si(557) at 300 K as shown (a) in the photoelectron intensity map in grey scale as a function of binding energy (as referenced to E_F) and momentum along the 1D chains $k_{\|}$ and (b) in the momentum distribution curves of the photoelectron intensity.

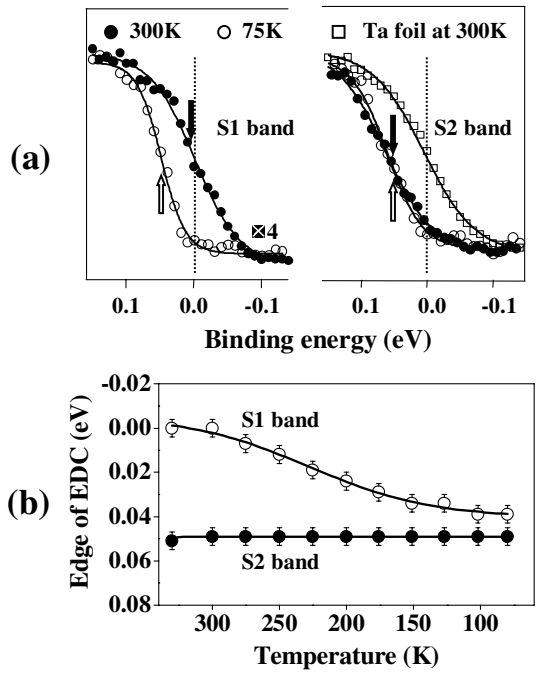

FIG. 2. (a) Energy distribution curves (EDCs) of the photoemission intensity for the S1 and S2 bands at the two k_F positions of 0.35 and 0.41 Å^{-1}, respectively, and (b) temperature dependence of the center-of-the-leading-edge positions of EDCs [see the arrows in (a)] for S1 and S2 at the corresponding k_F positions. 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 4.

spectral weight at 12 K [8]. Naturally, at this point, one may suspect that there may be a phase transition below RT. We thus measured the temperature dependence of the whole 1D band dispersions and the detailed spectral shapes near E_F down to 70 K. We found that the band dispersion is rigid against the change of the temperature as also noted previously [8], but there is a strong overall shift due to the surface photovoltage (SPV) effect. The SPV shifts, which increase as the temperature decreases, were carefully compensated by comparing the angle-integrated valence-band spectra including very sharp Au $5d$ and Si bulk features with an accuracy better than 10 meV. After this correction, in contrast to the invariant band dispersions, the spectral shape near E_F was found to exhibit a systematic and distinctive temperature dependence.

Figure 2(b) shows the leading edge position of the EDCs of S2 (solid symbols) and S1 (open symbols) at the two k_F positions shown in Fig. 2(a). While the leading edge of S2 does not change except for a slight sharpening, that of S1 shows a gradual shift to higher binding energy, which saturates at about 120 K with a shift amounting to 40 meV. As noticed in Fig. 2(a), this leading edge shift is obviously a sign of a gap opening for the S1 band. It should be stressed again that this leading edge shift cannot be explained by any uncorrected SPV shift since the leading edge of S1 (also all other spectral features not shown here) shows no such systematic shift. Assuming a symmetric gap, we estimate the band gap being 80 meV for this temperature-dependent metal-insulator transition, which was found to be completely reversible. Now, it is clear that the previous ARP study at 12 K should have found no clear Fermi cutoff due to this gap-opening transition at 120–270 K for S1 and to the gap of S2 opened already at RT.

Since the S1 and S2 bands are almost half filled and 1D, one can naturally invoke Peierls instability to explain the metal-insulator transition. Since the Peierls transition involves periodic lattice distortion (PLD) and CDW formation, which can be probed by structural probes, we first investigated the LEED patterns in detail at different temperatures. However, this turned out to be difficult since the surface shows $\times 2$ streaks in LEED already from RT. This must be due to the apparent $\times 2$ protrusions along the chains on the whole surface as observed by the previous RT STM study [9]. Nevertheless, we observe a nontrivial intensity increase (and a decrease of the width) of the streaks during the phase transition indicating an increased $\times 2$ order, which is thought to reflect some structural changes on the surface. However, a much more clear evidence of the PLD or CDW could be found from low-temperature STM imaging.

Figure 3 shows the empty-state STM images at two different bias voltages (left and right) taken at RT [3(a) and 3(c)] and 78 K [3(d) and 3(e)]. The RT images show two prominent chains of protrusions (denoted as α and β); one with a strong $\times 2$ modulation along the chains and the other without. A comparison of this image with the structure model proposed recently [Fig. 3(b)] [19] leads straightforwardly to identify the β chains with the adatom chains and the α with the step edge. Then the $\times 2$ modulation is explained by the alternating occupation of the Si adatoms, which is a natural consequence of the threefold coordination of adatoms. Moreover, the lack of any obvious $\times 2$ modulation on α is explained by the stability of the $\times 1$ step-edge reconstruction [see Fig. 3(b)] [16,17]. It should be noted that the Au atom chains incorporated into the terrace Si layer are not imaged. In contrast to the present result, the previous RT STM study reported two prominent chains of protrusions with a common $\times 2$ modulation. We also observe such $\times 2$ modulation on the α chains (the step edges) but only around some defect sites. This difference is not fully understood yet but could be due to the different defect density and distribution [20,21].

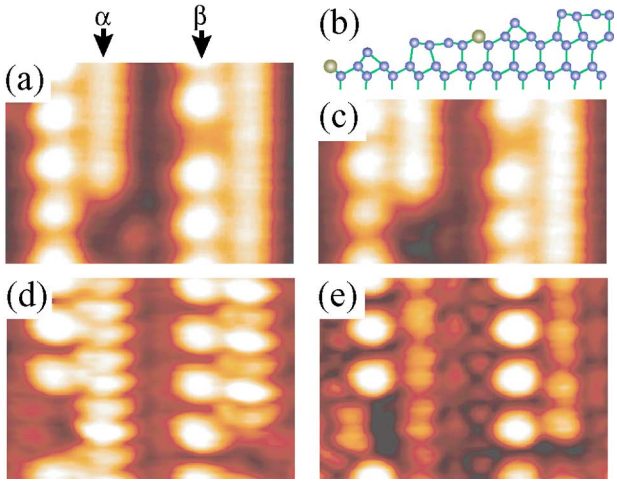

FIG. 3 (color online). Empty-state STM images at (a),(c) 300 and (d),(e) 78 K with a sample bias of (a),(d) $V_s = 1.0$ and (c),(e) 0.7 V. The recent structural model of Au/Si(557) [16,17,19] is shown in (b) schematically, where the large and small circles denote Au and Si atoms, respectively.

At a low temperature of 78 K, as shown in Figs. 3(c) and 3(d), the STM images exhibit a drastic difference; the α chains show a clear $\times 2$ modulation along the chains, most likely caused by dimerization within a $\times 2$ unit cell. The dimerization is symmetric in some chains and asymmetric in others, which is not fully understood yet but is suspected to be influenced by defects. Irrespective of the details of the new $\times 2$ modulation on the step edges, it is clear that there are structural changes on the surface, which is consistent with the metal-insulator transition observed in ARP spectra.

As supported by the above STM imaging of the extra $\times 2$ modulation along the chains at low temperature, we suggest that the origin of the metal-insulator transition is Peierls instability of the metallic part of the 1D chains. This implies that the metallic band at RT (S1) comes from the electronic states localized on the step edges. Our

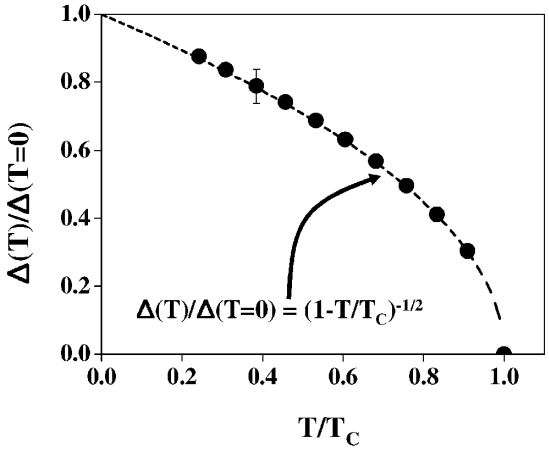

FIG. 4. Temperature dependence of the energy gap of the S1 band is fitted with the mean field theory (dashed line), $\Delta(T)/\Delta(T=0) = (1 - T/T_c)^{-1/2}$, where T_c and $\Delta(T)$ are a critical temperature and an energy gap, respectively.

site-specific scanning tunneling spectroscopy (STS) also shows the opening of an energy gap in the α chain at 78 K from a nearly gapless state at RT. The details of the STM and STS results will be reported elsewhere [20]. One can note further that the size of the band gap of S1 is qualitatively consistent with the mean field theory expectation of 3.52 times the transition temperature (T_c) with $T_c =$ 270 K. Furthermore, as shown in Fig. 4, the temperature dependency of the gap size is reasonably well fitted by the mean field theory of $\Delta(T)/\Delta(T=0)$ being proportional to $(1 - T/T_c)^{1/2}$ providing a consistent value of $T_c = 260$ K. Thus, we conclude that the phase transition is a Peierls transition with a moderate electron-phonon interaction.

Even with the present work, the origin of the S2 band is not clear and our assignment of the S1 band to an electronic state of the step edge needs to be confirmed by *ab initio* calculations. At present, the theory is not successful in reproducing the two parallel 1D bands even with the most elaborated structure model discussed above [16,17]. We speculate that the two bands would have different structural origin considering the complex and multiple chain structures on this surface. Furthermore, although the spectral behavior of the S1 band is rather well understood here, the band gap or simply the vanishing spectral weight of S2 band at E_F is unclear. We tentatively suggest that S2 band is also gapped even at RT since (i) the size of the band gap of S2 is slightly larger than that of S1 and (ii) the E_F crossing of S2 band is closer to the zone center indicating a possibly stronger electron-phonon interaction, which can easily lead to a higher T_c above RT. In this case, the present system is an analogy of $NbSe_3$, where two CDW transitions occur at different temperatures due to slightly different chains [1]. The experimental or theoretical verification of these postulates is certainly beyond the scope of the present work and is left for future studies.

In summary, the atomic and electronic structures of the Au-induced 1D chain structure on Si(557) were reinvestigated at 75–300 K. We observed that the two proximal 1D bands near E_F exhibit contrasting behaviors, one with a 50 meV band gap even at RT featuring no temperature dependency but the other with a normal metallic Fermi edge at RT exhibiting a temperature-induced gradual gap opening at 120–270 K. The STM study indicates an extra $\times 2$ periodicity modulation at 78 K along the step-edge 1D chain. This surface is, thus, concluded to be insulating below 120 K through a Peierls-type metal-insulator transition denying the previous Luttinger liquid interpretation.

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