Coexistence of Two Different Peierls Distortions within an Atomic Scale Wire: Si(553)-Au

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(Received 13 July 2005; published 31 October 2005)

The ground state property of a Au-induced atomic wire array on a stepped Si(553) surface with interesting 1D metallic bands was investigated. Electron diffraction and scanning tunneling microscopy reveal an intriguing coexistence of triple- and double-period lattice distortions at low temperature. Angle-resolved photoemission observes both the nearly 1/3- and 1/2-filled bands to gradually open energy gaps upon cooling. We explain these unusual findings as due to the occurrence of Peierls distortions of triple and double periods on the two different atomic-scale chain elements, respectively, within a single unit wire. The two Peierls distortions are suggested to have different transition temperatures and little lateral correlation between each other.

DOI: 10.1103/PhysRevLett.95.196402 PACS numbers: 71.30.+h, 73.20.−r, 71.20.Dg

A strong updated motivation to study the electronic properties of one-dimensional (1D) metallic systems lies in the quest for the breakdown of the Fermi-liquid framework [1], which has long been one of the key ideas in condensed matter physics [2–6]. In very recent days, this field of research has been enriched by finding unprecedented forms of 1D material systems with metallic electrons such as carbon nanotubes [3] and atomic wire systems on solid surfaces [2]. Indeed, for metallic carbon nanotubes, the non-Fermi-liquid behavior was disclosed by few successful experiments [3–5].

For the case of surface atomic wires [2,7–13], most notably the regular arrays of atomic chains formed on stepped Si surfaces by the adsorption of metals, the observation of the spin-charge separation, an exotic manifestation of the non-Fermi-liquid property, was claimed recently [2]. However, on that specific system of Si(557)-Au, the band structure with two nearly half-filled bands was found to apparently deviate from the non-Fermi-liquid theory [9,11]. Furthermore, this system was found to undergo at low temperature (LT) a Peierls-type metal-insulator (MI) transition, where the charge modulation at the Fermi energy couples with a periodic lattice distortion (PLD) through the electron-phonon interaction [11,12,14]. While a different explanation for this transition was suggested very recently [15], the Peierls instability is intrinsic to 1D metallic systems, especially for the half-filled-band systems. Indeed, the Peierls-type MI transitions were observed also for other recent atomic wire systems with half-filled metallic bands [8,11,12].

Apart from its own importance and interests, the Peierls instability or the significant electron-phonon interaction may obscure the non-Fermi-liquid behavior at LT. This issue is not restricted to 1D metallic systems but poses an important current question in many different exotic electron systems including high-temperature superconductors [16,17]. In this respect, it is important enough to note the recent finding of a metallic atomic wire system with both nearly 1/2- and nearly 1/4-filled bands—Si(553)-Au [10,13]. This 1D system with especially a fractional band filling deviating from 1/2 was strongly suggested to have a metallic and, presumably, non-Fermi-liquid ground state at LT, which may overcome both the Peierls instability and the Mott insulating state [10].

In the present Letter, we detail the temperature dependence of the lattice and electronic structures of Si(553)-Au. By low-energy-electron diffraction (LEED), scanning tunneling microscopy (STM), and angle-resolved photoemission (ARP) measurements, we found (i) that the system exhibits *the apparent lattice distortions of* ×3 *and* ×2

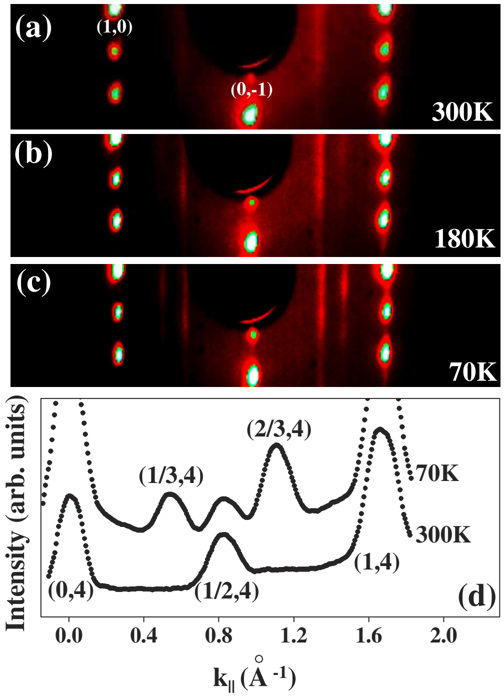

FIG. 1 (color online). LEED patterns of Si(553)-Au at (a) 300, (b) 180, and (c) 70 K with an electron beam energy of 88 eV. (d) LEED intensity profiles along the wire direction ($k_{\parallel}$) at 300 and 70 K.

periods along the different atomic chains within a unit wire and (ii) that the transition accompanies energy-gap openings for both nearly 1/2- and (1/3–1/4)-filled bands. The ground state of this system is, therefore, not metallic but an insulating broken-symmetry state against the original suggestion [10]. We relate the fractional band fillings of ~1/3 and ~1/2 with the ×3 and ×2 commensurate Peierls PLD's, respectively. The Si(553)-Au system is thus very unique with the coexisting commensurate Peierls distortions of double and triple periods within a single unit cell.

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) [11]. The nominal energy and angular resolutions were better than 15 meV and 0.15°, respectively. A commercial variable-temperature STM (Omicron, Germany) was used. A Si(553) substrate (12.5° offcut from the [111] orientation) was thermally cleaned. The temperature of a sample could be accurately controlled down to 50 (16) K by a He cryostat and a feedback-controlled heater for ARP (STM) measurements.

A Si(553) surface with a regular atomic wire array was generated by depositing Au at 920 K with postannealing [10]. The width of a single wire unit cell is 1.48 nm [Fig. 1(a) for LEED and Fig. 2(a) for STM] as composed of different atomic chains of the step-edge Si atoms, the Si atoms on terraces, and the Au atoms. The bright and dark chains in the STM image are tentatively assigned as the step-edge and terrace atomic chains, respectively, whose structural details are not clear yet [18]. At RT, while the step-edge chains exhibit only ×1 period protrusions, a few of the terrace chains show ×2 modulation as shown in Fig. 2(a), which is consistent with the previous STM studies [10,19]. This extra modulation will be discussed further below. In comparison, the Si(557)-Au [or Si(5 5 12)-Au] system, studied more extensively, has an opposite offcut direction and, thus, a different step-edge structure and a wider terrace to accommodate an extra atomic chain, the Si adatoms arranged into a ×2 period *a priori* [14,20–22].

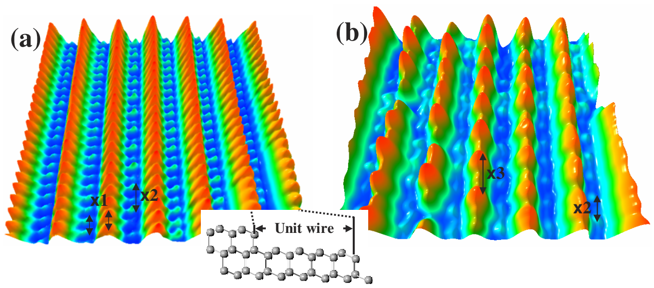

FIG. 2 (color online). STM topographs of Si(553)-Au at (a) 300 and (b) 45 K for an area of 10×10 nm^2 at 0.2 and 0.5 V biases (empty states), respectively. The ball and stick model in (a) shows the cross-sectional view of the unreconstructed Si(553) surface with steps and a narrow terrace.

Figure 3(b) shows the detailed energy bands near $\bar{K}$ as measured by ARP for Si(553)-Au at RT. The whole band dispersions are given schematically in Fig. 3(a) [10,13]. The three parabolic bands $S1$–$S3$ reach the Fermi energy (E_F) at $k_\parallel = 1.30$, 1.25, and 1.07 Å^{-1}, respectively, in the second Brillouin zone. The electron fillings of $S1$, $S2$, and $S3$ are measured to be 0.59, 0.51, and 0.30 ± 0.02, respectively, where 1 represents a filled band. These are qualitatively consistent with the previous report: 0.56, 0.51, and 0.27 for $S1$–$S3$, respectively [10,13]. The small discrepancy for $S1$ and $S3$ may be due to the finite $k_\perp$ dispersions [10,13]. In any way, one can say that the band structure is composed of two nearly 1/2-filled bands and the other band with 1/3–1/4 filling. The two nearly 1/2-filled bands of $S1$ and $S2$ are very similar to those of Si(557)-Au [9,11] and Si(5 5 12)-Au [12] but the fractionally filled band $S3$ is unique for Si(553)-Au [10,13].

For the case of Si(557)-Au and Si(5 5 12)-Au, $S2$ with an almost exact 1/2 filling exhibits an energy gap (about 50 meV below E_F) even at RT, while $S1$ with a slightly larger filling exhibits a metallic energy-distribution curve (EDC) [11]. The RT spectral characteristics of $S1$ and $S2$ are also the case for Si(553)-Au; $S1$ is metallic but $S2$ has an energy gap of 40 meV below E_F as shown in Fig. 4. Note

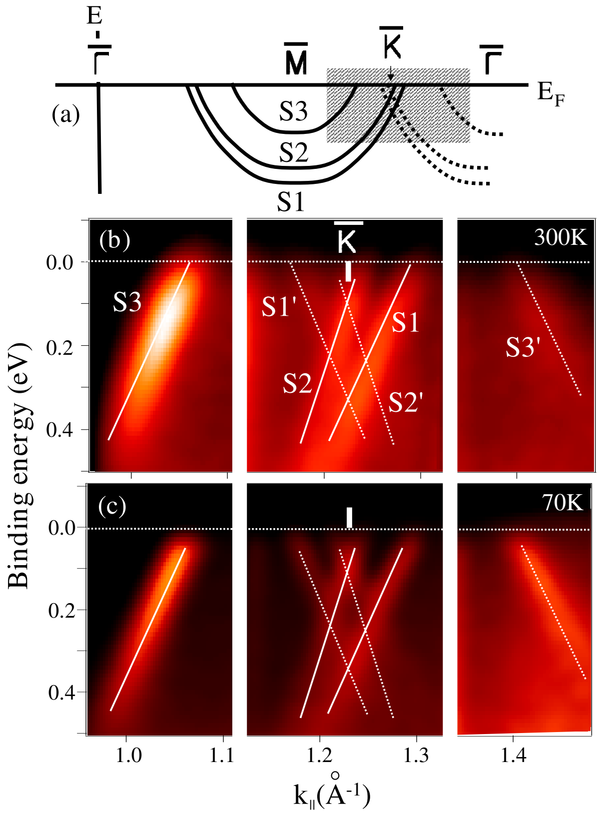

FIG. 3 (color online). (a) Schematics of the 1D metallic bands of Si(553)-Au along the wire direction ($k_\parallel$) [10,13]. Experimental energy bands near the Fermi momentums [the shaded region in (a)] as measured by ARP at (b) 300 and (c) 70 K.

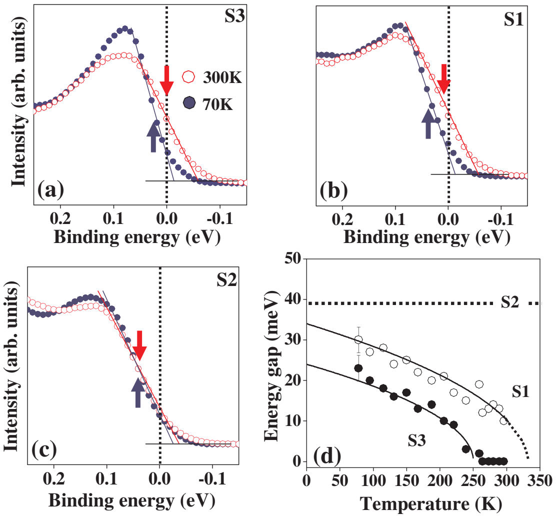

FIG. 4 (color online). Temperature dependence (300 and 70 K) of the photoelectron EDC's of (a) $S3$, (b) $S1$, and (c) $S2$, respectively, at the corresponding Fermi wave vectors. The arrows indicate the centers of the leading edges of the EDC's. (d) More detailed temperature dependence of the energy gaps below E_F of $S3$ (solid circle), $S1$ (open circle), and $S2$ (dotted line). For $S1$ and $S3$, the temperature dependences $\Delta(T)$ are reasonably fitted with the Bardeen-Cooper-Schrieffer function for a relatively weak electron-phonon coupling, $\Delta(T) = \Delta(T = 0)(1 - T/T_c)^{-1/2}$, with T_c the transition temperature [23].

the contrasting spectral intensities of $S1$ and $S2$ at E_F. However, a more quantitative comparison with the Fermi edge of a normal metal and a detailed curve fitting analysis suggests the existence of a very small gap of a 10 meV scale for $S1$ [Fig. 4(b)]. This will be discussed further below. The origin of the solid energy gap on the 1/2-filled $S2$ band has not been established yet, as well as its origin [11,14,15]. On the other hand, the nearly 1/3-filled band $S3$ on Si(553)-Au exhibits another very obvious metallic EDC as shown in Fig. 4(a). In conclusion, at RT, Si(553)-Au is given with an insulating band ($S2$) and two metallic (or quasimetallic) bands ($S1$ and $S3$). The contrasting spectral differences among $S1$–$S3$ at E_F were not noticed in the previous ARP study [10].

The 1D metallic band structure described above undergoes drastic changes at LT along with a characteristic structural transformation. As shown in Fig. 1, the RT $1 \times' 2'$ ($'$ for very weak 1/2 streaks) LEED pattern gradually transforms into a complex structure with both 1×3 and 1×2 orders at LT. The transition is much more obvious for the newly formed 1/3 superspots [Fig. 1(d)], but the enhancement of the 1/2 streaks can also be noticed. The detailed temperature dependence of the 1/3 spots further indicates that the phase transition is of a second order type with a transition temperature close to 250 K.

The STM imaging at 50 K gives a much clearer view of the structural transformation [Fig. 2(b)]. At LT, most of the step-edge chains show a very conspicuous $\times 3$ modulation, especially at empty states, with one large and one small protrusion in each $\times 3$ cell. Along with this new modulation at LT, the population of the terrace chains with the $\times 2$ structure increases significantly, and, in fact, almost no $\times 1$-period chains are noticed. That is, *the step-edge and terrace chains undergo the transitions from the* $\times 1$ *lattice into* $\times 3$ *and* $\times 2$ *PLD's, separately.* As mentioned above, the $\times 2$ PLD on the terrace chains can be found even at RT locally (see also the weak 1/2 streaks in LEED), which may suggest that the transition temperature for the $\times 2$ PLD is higher or is more sensitive to the perturbation imposed by random defects [14].

The consequences of the apparent PLD's at LT on the electronic structure are obvious. Figure 3(c) shows the LT band dispersions, which overall look similar to those at RT except for the reduced thermal broadenings of the spectral features and the enhanced intensities of the umklapp features $S2'$ and $S3'$. The umklapp features are due to the $\times 2$ potential [10,13], and its enhancement at LT is consistent with the structural changes observed, i.e., the enhanced $\times 2$ order at LT. There are, however, subtle but more important changes at E_F, especially for the two metallic bands of $S1$ and $S3$, as detailed in Fig. 4. Both *$S1$ and $S3$ open energy gaps below RT* as indicated by the leading edge shifts of the corresponding EDC's and the decreased spectral weights at E_F. In clear contrast, the insulating $S2$ band maintains its solid gap [Fig. 4(c)]. The gap opening is most clear for $S3$, the ~1/3-filled band, where the transfer of the spectral weight from E_F into the quasiparticlelike feature centered at ~0.08 eV is apparent [Fig. 4(a)]. Here any possible extrinsic spectral shifts, for example, that due to the surface photovoltage effect, were corrected accurately by the internal references of the Si bulk band, Au 5d peaks, and, furthermore, the persistent EDC leading edge of $S2$. The sizes of the energy gaps of $S1$ and $S3$, measured quantitatively as shown in Fig. 4(d), increase gradually upon cooling to reach 30 and 23 meV, respectively, at 70 K. This gradual gap opening seems not saturated even at 70 K and is fully consistent with the intensity change of the 1/3 superspots in LEED.

One can straightforwardly relate the LT $\times 3$ ($\times 2$) PLD to the gap opening on $S3$ ($S1$) with an electron filling close to 1/3 (1/2). That is, we interpret that the gap openings of $S1$ and $S3$ with fractional fillings are accompanied by PLD's of nearby commensurate periods instead of incommensurate PLD's (or charge density waves, CDW's) imposed by the exact nesting vectors. Such a "commensurability effect" is understood from the fact that the energy gain due to a CDW formation in a realistic lattice system should compete with the energy cost due to the strain produced by a corresponding, especially incommensurate, PLD [23]. Generally speaking, the structural and electronic transitions observed can consistently be understood from the Peierls instability due to the electron-phonon interaction. This is further corroborated by (i) the size of the energy gaps, which is compatible with the phonon energy scale, and (ii) the mean-field-theory-type temperature evolution of the energy gaps shown in Fig. 4(d). However, the present case is much more complicated than usual with two distinct PLD's and the corresponding energy gaps. The different sizes of the energy gaps would lead to different transition temperatures (T_c) as suggested in Fig. 4(d). That is, the transition on the terrace chains with the ×2 PLD seems to have a higher T_c than the other, which is evidenced by the presence of the partial ×2 order at RT in LEED and STM and also by the small gap on $S1$ at RT mentioned above. The present system is, thus, analogous to the unique double Peierls transitions, $T_c = 59$ and 149 K, observed for a quasi-1D metallic compound of $NbSe_3$ [23,24].

The uniqueness of the present system is that the two different commensurate PLD's separately exist on neighboring atomic chains within an atomic-scale unit wire of ~1.5 nm. What is surprising is that the two adjacent PLD's of ×3 and ×2 periods seem not correlated spatially, i.e., do not form a 1×6 lattice. This is unusual if we consider only the lattice distortion, since the chain elements are closely entangled in any way through the strong covalent bond network. Such transverse lateral correlations between the PLD's within a unit wire as well as between the different wires are to be investigated further in addition to the structural details of the complex PLD's at LT.

The phase transition is obviously against the previous suggestion that the atomic wires in Si(553)-Au may exhibit a metallic and, moreover, non-Fermi-liquid ground state due to a fractionally filled band overcoming the Peierls instability destined to a 1D metal [10]. Along with other examples of In/Si(111), Si(557)-Au, and Si(5 5 12)-Au, this may indicate that the surface atomic wire systems have generally soft and susceptible lattices upon Peierls instability as composed of only a single (supported) atomic layer. However, further studies on the quantitative measurement of the electron-phonon interaction strength and the possible coexistence of the Peierls gap and the spin-charge separation [16,17] are in order, which require careful experiments at a much lower temperature.

The detailed comparison of the present phase transition with those of Si(557)-Au and Si(5 5 12)-Au is obviously important considering the very similar 1/2-filled bands. In particular, a recent theoretical calculation on Si(557)-Au argued that the apparent ×2 PLD on the step-edge chain is due to the quenching of thermal fluctuation between two degenerated (buckled) structures along the chains [15]. However, as discussed extensively in our recent work [14], such an order-disorder-type transition cannot straightforwardly provide a gap-opening mechanism for the obvious metal-insulator transition. This is clear in the well-established case of the Si(001) surface with an order-disorder phase transition due to the dynamical fluctuation of buckled dimers [25]. Note also that the step-edge structure is expected to be largely different between Si(557)-Au and Si(553)-Au. A further discussion on this comparison, however, should wait for reliable structural models, especially for Si(553)-Au.

In summary, we observed a new metal-insulator transition on the Au-induced atomic wires on Si(553) with triple 1D bands. The metal-insulator transition involves the gap openings of two 1D bands with nearly 1/2 and ~1/3 fillings, which are related to the ×2 and ×3 commensurate Peierls distortions within a single unit wire at LT, respectively. The system is thus not metallic against a recent suggestion to observe a non-Fermi-liquid behavior but falls into a complex and interesting broken-symmetry ground state.

This work was supported by MOST through Center for Atomic Wires and Layers of the CRi program.

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[1] J. M. Luttinger, J. Math. Phys. (N.Y.) **4**, 1154 (1963).

[2] P. Segovia *et al.*, Nature (London) **402**, 504 (1999).

[3] H. Ishii *et al.*, Nature (London) **426**, 540 (2003).

[4] M. Bockrath *et al.*, Nature (London) **397**, 598 (1999).

[5] Z. Yao *et al.*, Nature (London) **402**, 273 (1999).

[6] R. Joynt *et al.*, Science **284**, 777 (1999).

[7] J. Nogami, B. Z. Liu, M. V. Katkov, C. Ohbuchi, and N. O. Birge, Phys. Rev. B **63**, 233305 (2001).

[8] H. W. Yeom *et al.*, Phys. Rev. Lett. **82**, 4898 (1999); J. R. Ahn *et al.*, *ibid.* **93**, 106401 (2004).

[9] R. Losio *et al.*, Phys. Rev. Lett. **86**, 4632 (2001).

[10] J. N. Crain *et al.*, Phys. Rev. Lett. **90**, 176805 (2003).

[11] J. R. Ahn, H. W. Yeom, H. S. Yoon, and I.-W. Lyo, Phys. Rev. Lett. **91**, 196403 (2003).

[12] J. R. Ahn, H. W. Yeom, E. S. Cho, and C. Y. Park, Phys. Rev. B **69**, 233311 (2004).

[13] J. N. Crain *et al.*, Phys. Rev. B **69**, 125401 (2004).

[14] H. W. Yeom *et al.*, Phys. Rev. B **72**, 035323 (2005).

[15] D. Sánchez-Portal, S. Riikonen, and R. M. Martin, Phys. Rev. Lett. **93**, 146803 (2004).

[16] F. H. L. Essler and A. M. Tsvelik, Phys. Rev. Lett. **88**, 096403 (2002).

[17] L. Bartosch, Phys. Rev. Lett. **90**, 076404 (2003).

[18] K. D. Ryang, P. G. Kang, J. R. Ahn, S. Jeong, and H. W. Yeom (unpublished).

[19] J. N. Crain and D. T. Pierce, Science **307**, 703 (2005).

[20] I. K. Robinson, P. A. Bennett, and F. J. Himpsel, Phys. Rev. Lett. **88**, 096104 (2002).

[21] D. Sánchez-Portal, J. D. Gale, A. García, and R. M. Martin, Phys. Rev. B **65**, 081401(R) (2002).

[22] D. Sánchez-Portal *et al.*, Surf. Sci. **532**, 655 (2003).

[23] George Grüner, *Density Waves in Solids* (Addison-Wesley, Reading, MA, 1994).

[24] J. Schäfer *et al.*, Phys. Rev. Lett. **87**, 196403 (2001).

[25] Y. Enta, S. Suzuki, and S. Kono, Phys. Rev. Lett. **65**, 2704 (1990); K. Hata, Y. Sainoo, and H. Shigekawa, *ibid.* **86**, 3084 (2001).