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Band structure of Au monoatomic chains on Si(335) and Si(557) surfaces

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The electronic band structure of the Si(557) and Si(335) surfaces covered with monoatomic Au chains produced in UHV conditions, is investigated in detail by angle-resolved photoelectron spectroscopy (ARPES), especially for the surface state bands near the Fermi energy. The ARPES spectra in the plane parallel to step edges for Si(557)-Au vicinal surface show strongly dispersive electron energy bands, characteristic of one-dimensional structure. The band dispersion is also calculated within tight-binding model, with two adjustable coupling parameters t_1 and t_2, for the first and second neighbors along the chains, respectively, and compared with that determined from the photoemission experiment. The scanning tunneling microscopy (STM) imaging and reflection high energy electron diffraction (RHEED) studies enabled us to determine atomic chain separation and its internal structure. The study shows that the structural anisotropy of these surfaces induces highly anisotropic electronic structure.

Keywords: nanowires, vicinal surface, Si(335), Si(557), photoemission.

1. Introduction

In the last years photoemission has been well-established as a suitable tool for investigating occupied electronics states in solids [1]. Angle resolved ultraviolet photoemission spectroscopy (ARUPS), as a tool especially sensible to the surface, becomes an appropriate technique to study low-dimensional electronic states at the surface. The properties of electrons become more exotic as one progresses from the higher dimensional world into 1D [2]. The behavior of the electrons propagating in one-dimensional chain of atoms can be demonstrated for the 1D Tomonaga–Luttinger model [3, 4]. In such system the only excitation of the system consists of independently dispersing spin and charge density waves, called spinons and holons, respectively. These collective excitations have different group velocities, and run away from each other under the Fermi level. For these quasiparticles energy *vs*. momentum curves intersect, when approaching the Fermi energy.

Vicinal silicon substrates like Si(557) and Si(335) are adequate to perform and investigate 1D monatomic Au chains, since they have straight and regular step arrays. Moreover, the existence of the semiconducting band gap of the silicon substrate allows us to treat such metallic chains as unperturbed by the substrate.

LOSIO *et al*. [5], observed two narrowly spaced bands at the Si(557)-Au surface near the Fermi level with well defined splitting at the Fermi level. This is not expected from the Luttinger liquid theory.

In atomic chains a phase transition may occur. According to the Peierls theorem [6] a half filled metallic band can lower its total energy by developing a gap at the Fermi wave vector, which opens by pairing the atoms along the chain. One way out of the Peierls transition is an atomic chain anchored to the substrate, such as the Au chains on Si discussed here.

In the present work we demonstrate the capability of probing 1D electronic states of gold atom chains on stepped silicon Si(335), Si(557) surfaces, using photons with energy 21.22 eV. Basing on recorded spectra, we calculated surface band structure of the chains near the Fermi energy. In order to determine coupling parameters for first and second neighbors along the chains, we apply simple tight-binding model. Using polarized excitation radiation, we show that the electronic structures along and perpendicular to the chain are highly anisotropic. Scanning tunneling microscopy (STM) measurements confirm anisotropic character of the system.

2. Experimental

All experiments were performed in UHV conditions. The UHV chamber was equipped with a reflection high energy electron diffraction (RHEED) apparatus, a helium lamp, an electron energy analyzer VGX900IC, and nitrogen cooled manipulator. The vacuum system was also furnished with a quartz film-thickness monitor. The *p*-type, B-doped silicon with the specific resistivity of 1–2 Ωcm at room temperature, was used in all experiments. The substrates were cut from a commercial 2" Si crystal oriented by means of X-ray diffractometry technique with an accuracy of ±0.05 deg. Then, they were polished using diamond paste and Syton OP-S (Struers). The samples were etched in 1:10 HF/water solution for 10 seconds before they were placed inside the vacuum chamber. They were cleaned in UHV by flashing for one second to about 1500 K by direct current heating. In order to produce well ordered Si(335) surface, 0.28 ML of Au was deposited onto it and annealed for a minute at about 950 K, and next the temperature was gradually lowered to room temperature within five minutes. During Au deposition, the substrate temperature was kept at 650 K. In the case of Si(557) 0.18 ML of Au was deposited. Annealed procedure was the same. These processes were controlled *in situ* by RHEED. In both cases the base pressure was below 3×10^{-10} mbar. Detailed analysis of the RHEED pattern originating from these vicinal samples was described elsewhere [7, 8].

ARUPS measurements were performed at low temperatures (130 K). In photoemission experiments HeI line with energy 21.22 eV was used. The radiation was *p*-polarized. Angular and energy resolution of the analyzer were fixed to be 1.5 deg and 50 meV, respectively.

3. Results and discussion

The STM images shown in Fig. 1 clearly demonstrate the 1D chainlike reconstructions of these surfaces. The width of the terraces was established to be equal $5\frac{2}{3}$ and $3\frac{2}{3}$ lattice constant towards $[\bar{1}\bar{1}2]$ direction for Si(557) and Si(335) surfaces, respectively. The STM experiments clearly exhibit the existence of one monoatomic chain along $[\bar{1}10]$ for Si(335)-Au surface and two chains for Si(557). The chains for Si(335)-Au are less compact. The lattice period along the chain is doubled for both of them. KRAWIEC *et al*. [9] in their calculations suggest, in the case of Si(557)-Au, the existence of one chain originating from Si and the second from Au.

Figure 2 shows that the structural anisotropy, visible in the STM images, appears also in the ARUPS spectra. Spectra were taken for two orientations of the sample: with atomic chains perpendicular (1) and parallel (2) to the radiation incident plane. It was realized by rotation of the sample by 90 deg around normal to the surface. The off-normal

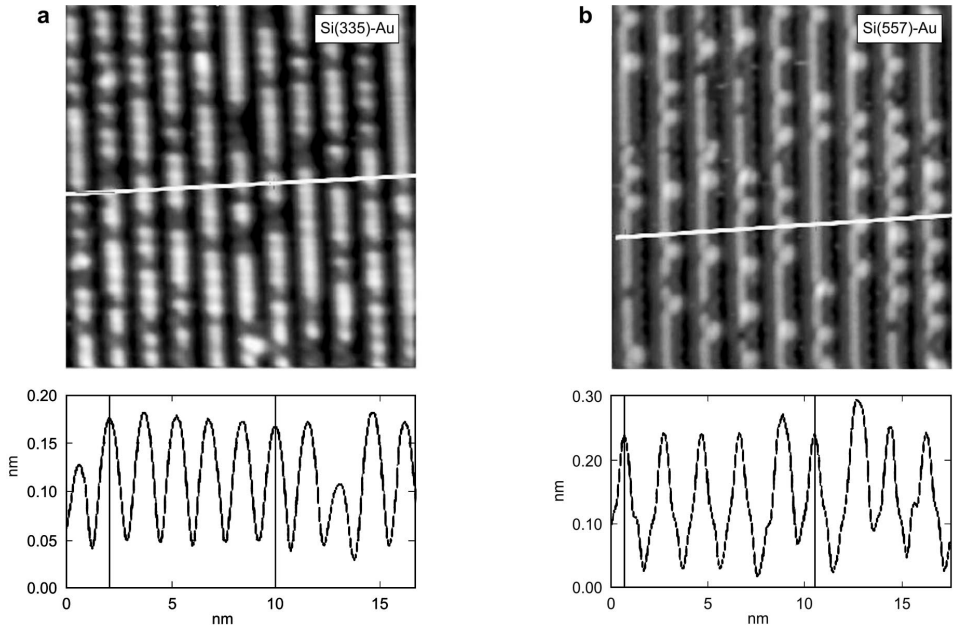

Fig. 1. STM images of monoatomic chains of Au onto Si vicinal surfaces: 0.28 ML of Au deposited on Si(335) (**a**), 0.18 ML of Au on Si(557) (**b**). The chains for case Si(335) are less compact as in the Si(557).

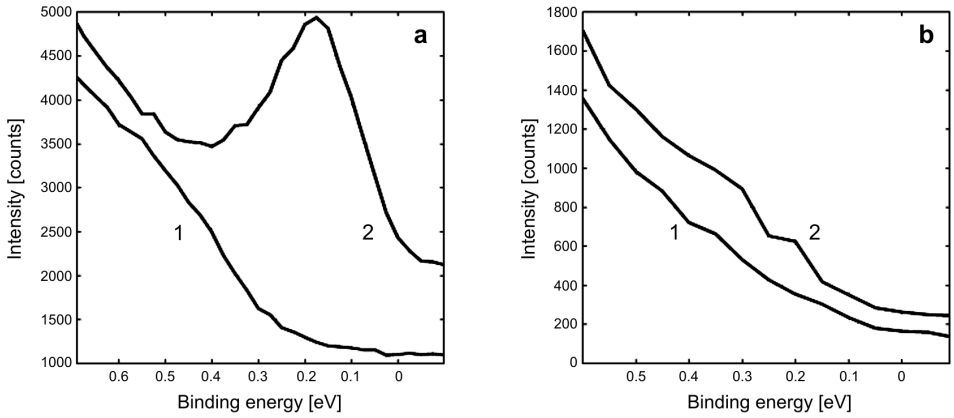

Fig. 2. ARPES spectra taken in two perpendicular direction across (1) and along (2) of the monoatomic chains on Si(557)-Au (**a**) and Si(335)-Au (**b**). The spectra at particularly figures were recorded at the same off-axis angle and polarization (see text). The off-axis was 34.5 deg (**a**), and 30.5 deg (**b**). For direction of incoming radiation along $[\bar{1}10]$ (**a**, spectrum 2) occupied states below Fermi level are clearly visible.

angles (angle between surface normal and analyzer) was 34 deg for Si(557)-Au (Fig. 2**a**) and 30 deg for Si(335)-Au (Fig. 2**b**). The observed anisotropy in the spectra, clearly exhibit 1D character of the system. The occupied electronic states of Si(557)-Au are highly dispersive as it is shown in Fig. 3. Such significant dispersion excludes their identification as Shockley surface states.

The Brillouin zones (BZ) of the Si(335)-Au and the Si(557)-Au are quite similar. The distance in ΓK direction is equal to 0.82 $Å^{-1}$ in reciprocal space for both surfaces. ΓK direction corresponds to the $[\bar{1}10]$ in real space. Spacing between terraces is 12.6 Å and 19.2 Å for Si(335) and Si(557), respectively, which gives 0.25 and 0.16 $Å^{-1}$ along $\Gamma M'$ direction for these surfaces. Basing on the simple tight-binding model, the band structure schematically showed in Fig. 4 is expected [5, 10, 11]. According to Fig. 4, the band crosses the Fermi level at the half length of the surface BZ. The marked area points to the region which was measured by photoemission. The set of spectra for Si(557)-Au is shown in Fig. 3. The binding energy is defined with respect to the Fermi level. The metallic character of the surface indicates that the Peierls transition to the semiconducting state is avoided. The spectrum for Si(335)-Au is different. For Si(557)-Au highly dispersive peaks are observed, whereas for the Si(335)-Au are not. The dispersion is very weak because of the presence of highly localized electronic state. It can be due to the small length of the chains caused by defects and the smaller width of the terraces entailed weakness of the 1D effects.

For Si(557)-Au the splitting at the Fermi level is observed which is clearly visible in Fig. 5, where the second derivative of the measured intensity is shown. For Si(557)-Au surface two bands are observed. One band crosses the Fermi level inside

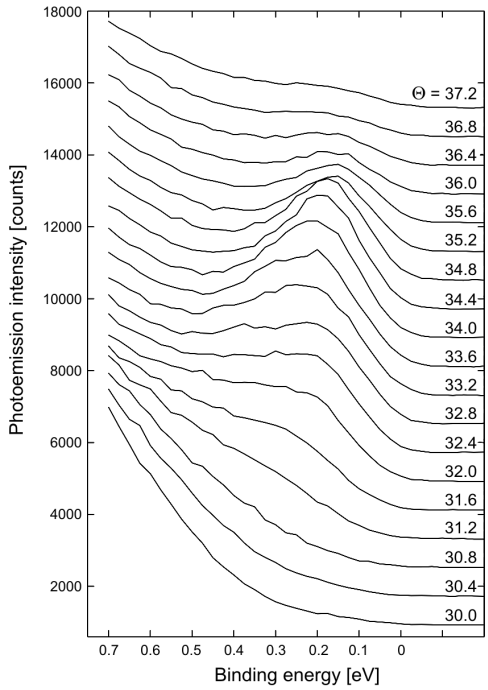

Fig. 3. Angle resolved photoemission set of spectra taken from Si(557)-Au along [$\bar{1}$10] direction (Θ is an angle between normal to the sample and analyzer).

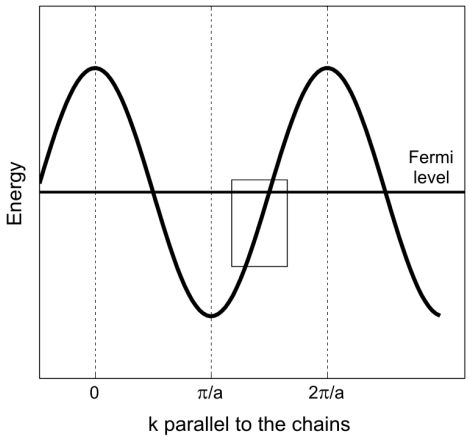

Fig. 4. Schematic drawing of tight-binding band topology for vicinal surfaces. The photoemission sampled region was marked as the rectangle and it is just in a half way of the second surface Brillouin zone.

the half length of the BZ which corresponds to half filling. Second band spreads slightly further and cuts the Fermi level outside the half length of the BZ indicating that the filling of the band is somewhat larger than 0.5. These reconstructed Au vicinal surfaces have an even electron count per unit cell which seems to defy conventional wisdom that an even counting produces a semiconductor and an odd a metal. The splitting

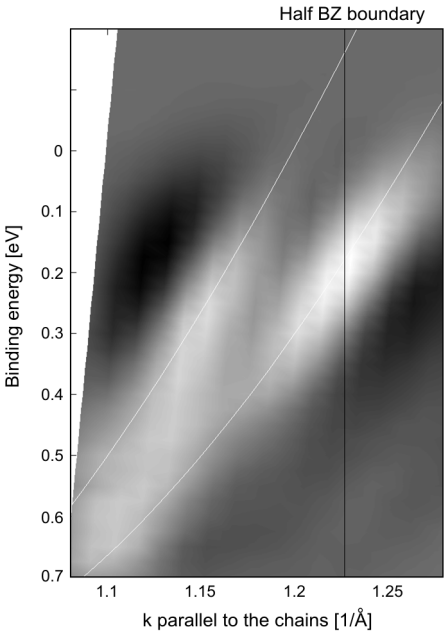

Fig. 5. Map of the intensity second derivative. Highly dispersive peak is observed for Si(557)-Au. Marked thin black line is placed in a half way of the second Brillouin zone, equal to 1.23 1/Å. The fitting tight-binding band structure is marked by thin white lines.

explains why the surfaces becomes metallic despite an even electron count per unit cell. Each of half filled nearly degenerate bands contributes one electron. The existence of these bands suggests two nearly identical orbitals within the unit cell that generate bonding/antibonding combinations [2]. The simple tight binding model was introduced to reproduce band topology observed in Fig. 5. This was realized employing the Hamiltonian that contains only intra-chain couplings t_1 and t_2 for first and second neighbors in the chain, respectively. The interactions between the chains and between a chain and a substrate are neglected. The tight binding bands with the two couplings parameters are given by:

$$\begin{aligned} E(k_x) &= E_0 + t_1\left[\exp(ik_x a) + \exp(-ik_x a)\right] + t_2\left[\exp(ik_x 2a) + \exp(-ik_x 2a)\right] \\ &= E_0 + 2\left[t_1\cos(k_x a) + t_2\cos(2k_x a)\right] \end{aligned}$$

where E_0 is a reference energy, a is a lattice constant along the Au chain. Three parameters E_0, t_1, t_2 were used to fit the equation to the measured band structure for

T a b l e. Band parameters for Au chains on Si(557).

	First band	Second band
E_0 [eV]	0.02±0.02	0.3±0.02
t_1 [eV]	0.61±0.03	0.77±0.03
t_2 [eV]	0.1±0.03	0.07±0.03

vicinal surface Si(557)-Au. The coupling parameters discussed here are presented in the Table.

4. Conclusions

We combine STM real space studies and reciprocal space photoemission studies. It is clearly shown that structural anisotropy demonstrated by STM induces also electronic band structure anisotropy. ARUPS enables us to determine the unusual surface states induced by the Au atoms at vicinal Si(111) surfaces. Observed bands splitting disagrees with Luttinger liquid theorem since spinon and holon bands cross at Fermi level separately. Such measurements and the existence of metallic bands are encouraged to prepare vicinal surfaces with different Miller indexes, and to investigate them in the hope that they turn up interesting electronic properties.

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References

[1] HÜFNER S., *Photoelectron Spectroscopy*, Springer Series in Solid-State Sciences, Vol. 82, Springer, Berlin 1995.

[2] HIMPSEL F.J., ALTMANN K.N., BENNEWITZ R., CRAIN J.N., KIRAKOSIAN A., LIN J.-L., MCCHESNEY J.L., *One-dimensional electronic states at surfaces*, Journal of Physics: Condensed Matter **13**(49), 2001, pp. 11097–113.

[3] TOMONAGA S., *Remarks on Bloch's method of sound waves applied to many-fermion problems*, Progress of Theoretical Physics **5**(4), 1950, pp. 544–69.

[4] LUTTINGER J.M., *An exactly soluble model of a many-fermion system*, Journal of Mathematical Physics **4**(9), 1963, pp. 1154–62.

[5] LOSIO R., ALTMANN K.N., KIRAKOSIAN A., LIN J.-L., PETROVYKH D.Y., HIMPSEL F.J., *Band splitting for Si(557)-Au: is it spin-charge separation?*, Physical Review Letters **86**(20), 2001, pp. 4632–5.

[6] PEIERLS R.F., [In] *Quantum Theory of Solids*, Clarendon, Oxford 1955, p. 108.

[7] JALOCHOWSKI M., STROZAK M., ZDYB R., *Gold-induced ordering on vicinal Si(111)*, Surface Science **375**(2-3), 1997, pp. 203–9.

[8] ZDYB R., STROZAK M., JALOCHOWSKI M., *Gold-induced faceting on Si(533) surface studied by RHEED*, Vacuum **63**(1-2), 2001, pp. 107–12.

[9] KRAWIEC M., *et al.*, (unpublished).

[10] CRAIN J.N., MCCHESNEY J.L., FAN ZHENG, GALLAGHER M.C., SNIJDERS P.C., BISSEN M., GUNDELACH C., ERWIN S.C., HIMPSEL F.J., *Chains of gold atoms with tailored electronic states*, Physical Review B: Condensed Matter and Materials Physics **69**(12), 2004, pp. 125401/1–10.

[11] HIMPSEL F.J., ALTMANN K.N., CRAIN J.N., KIRAKOSIAN A., LIN J.L.; LIEBSCH A., ZHUKOV V.P., *Photoelectron spectroscopy of atomic wires*, Journal of Electron Spectroscopy and Related Phenomena **126**(1-3), 2002, pp. 89–99.

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