Quasi-One-Dimensional Nature of the Rashba States of Au Wires on Si(557) Surface

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Abstract

The electronic band structure of Si(557) surface adsorbed with Au wires is studied by means of first-principles density-functional-theory calculations. The spin-orbit split metallic bands are associated with the Au-Si zigzag chain and their asymmetric electron density around the Au atoms due to the on-site orbital hybridization explain large Rashba-type splitting and out-of-plane spin polarization. It is found that the Rashba metallic states have strong one-dimensional (1D) character along the Au wires but non-negligible two-dimensional distribution of the wave functions, implying not exactly genuine, only quasi 1D states.

Keywords: Rashba effect, Electronic band structure, Surface state, DFT calculation, Si(557) surface, spin-orbit coupling

1. Introduction

Rashba effect is a spin-splitting phenomenon of an electronic state without magnetic field [1, 2]. Its microscopic origin is the spin-orbit coupling (SOC) associated with broken inversion symmetry in analogous to Dresselhouse effect [3]. Since the measurement of spin-splitting surface states of Au(111) with angle-resolved photoelectron spectroscopy (ARPES), a variety of Rashba states at surface systems have been studied experimentally and theoretically. Among them, we have investigated some general aspects of the Rashba effect at surfaces on the basis of the band theoretical point of view. The key factors governing the magnitude of the Rashba effect are shown to be large SOC, namely large atomic-number Z , and the asymmetric electron density around the nucleus induced by broken inversion. [5] The importance of the factors originates in the dominant contribution of the gradient of Coulomb potential from nucleus in SOC [6] and may reasonably explain why Ag(111) surface states does not show any observable size of Rashba splitting though the magnitude of SOC in Ag is nearly one third of that in Au. The SOC band splitting and corresponding spin structure at several typical surfaces have been analyzed and classified by using the group theory with a $k \cdot p$ perturbation approach. [7] Interestingly, it is shown that the two-dimensional (2D) states with certain irreducible representations in the particular groups of k (Γ_6 in C_{3v} , and Γ_6 and Γ_7 in C_{4v} , for

example) always behave Rashba-type k -linear band splitting and vortical spin structure, regardless of the time reversal symmetry. This clearly explains the observed Rashba band splitting at K point that is *not* any of the time reversal invariant momenta (TRIM), in Bi/Si(111) surface. [8]

Recently, one-dimensional (1D) metallic states have attracted much attention because of fundamental interests in their quasiparticle nature and of possible applications to nano-devices. In this context, Au wires on Si(557) surface provides us a suitable example to be studied. Photoemission measurements of Au wires on Si(557) surface have shown two-peak structures indicative of the spin and charge separation in the Luttinger liquid theory. [9] Soon later, an *ab initio* density-functional-theory (DFT) calculation has claimed that such two-peak structures may be explained by the appearance of two metallic bands associated with Au-Si bonds. [10] However, further microscopic character of the two metallic bands has not been clarified because of uncertain surface structure adopted in the calculation, though the structure of vicinal Si surface adsorbed with Au was optimized to some extent. Some experimental efforts have been paid to determine the stable structure of the Au/Si(557) surface with three-dimensional x-ray diffraction (XRD) measurements by Robinson *et al.*, [11] and with scanning tunneling microscopy (STM). [12] Finally, a DFT study has confirmed Robinson's model theoretically stable for the Au/Si(557) surface, which are drastically relaxed from the ideal vicinal Si surface. [13] The electronic band structure of the optimized surface structure is quite consistent with the previous ARPES measurements. Besides Si(557), the other Au/Si surface systems with odd miller indices have been synthesized and studied by ARPES. [14]

Concerning the microscopic origin of the doublet metallic bands in Au/Si(557), Sánchez-Portal *et al.* have shown in their DFT calculation that SOC has a crucial role to realize two 1D bands measured in ARPES. [15] The band doublet was identified by ARPES as Rashba-type spin splitting in a 1D chain structure. [16] Sánchez-Portal *et al.* have further investigated from first principles the relation between the electronic and atomic structures in Au/Si(557). [17] An undoubted proof of the Rashba origin for the doublet structure in Au/Si(557) has been made by spin-resolved ARPES measurements by Okuda *et al.* [18] Clear spin polarization in the metallic bands has been found to be the Rashba-like and consistent with first-principles calculations. Furthermore, a large out-of-plane spin component has been observed and interpreted with highly anisotropic wave function of the Au wires induced by orbital hybridization with the neighboring Si surface atoms.

In this paper, some detailed band-theoretical analysis of the electronic structure in Au/Si(557) will be presented to gain a deep insight of the Rashba effect in 1D metallic states. Particular attention will be paid to the proper interpretation of the Rashba states relevant to the time reversal symmetry, and to the 2D character of their wave functions.

Figure 1: (Color online) Surface structure model of Au/Si(557): (a) bird's eye view and (b) top view. Large (yellow), medium (blue), and small (white) balls represent Au, Si, and H atoms, respectively. Three arrows denote the primitive translation vectors ($\mathbf{a}$, $\mathbf{b}$, and $\mathbf{c}$) assumed in the periodic slab model and the parallelepiped indicates the corresponding unit cell. The vectors $\mathbf{a}$ and $\mathbf{b}$ represent the two-dimensional periodicity of the (557) surface plane while the vector $\mathbf{c}$ is taken as the normal direction to the plane for the construction of supercell.

2. Surface Structure Model and Methods

The general description of the structures of the vicinal Si surfaces adsorbed with Au wires are given in the literature. [14] The surface structure determined by the previous DFT calculation [13, 19] is shown in Fig. 1 and used in the present calculation. This surface structure is more complicated than that originally considered as mentioned in the preceding section. The surface model consists of 47 Si, 2 Au, and 14 H atoms in the unit cell. The additional 14 H atoms are used to terminate the dangling bonds of Si atoms at the bulk side. It should be noted that the two-dimensional (2D) lattice vectors ($\mathbf{a}$, $\mathbf{b}$) in Fig. 1 are of the true translational symmetry of Au/Si(557) while the lattice vector ($\mathbf{c}$) is actually arbitrary and only for the setup of the periodic slab model assumed. The present choice of $\mathbf{c}$ is perpendicular to the (557) surface plane. The size of the vacuum region in the slab model that is the length of $\mathbf{c}$ was confirmed to be sufficiently thick by checking the band structure for different thickness values.

Figure 2 shows the reciprocal lattice points and the surface Brillouin zone of Au/Si(557) constructed from the 2D lattice vectors ($\mathbf{a}$, $\mathbf{b}$) of Fig. 1. The direction of the Au wires that is taken along the k_x axis is slightly deviated from one of the reciprocal lattice vectors ($\Gamma - A - \Gamma$). This is because the lattice vector $\mathbf{b}$ cannot be chosen perpendicular to the Au wires when the other lattice vector $\mathbf{a}$ is parallel to the wires.

Figure 2: Surface Brillouin zone of Au/Si(557). Open circles denote the two-dimensional reciprocal lattice points. Thick solid lines ($\Gamma - A - \Gamma$, $\Gamma - B - \Gamma$, and $\Gamma - Y - \Gamma$) indicate the lines connecting the neighboring reciprocal points. The direction of the Au wires is along the k_x axis. Note that A , B , and Y points are the time reversal invariant momenta in addition to Γ . A broken line denotes the zone boundary line of which the center is A .

First-principles DFT approach with the slab model shown above is adopted to calculate the electronic structure of Au/Si(557). Scalar relativistic equations are solved self-consistently with the second-variation step for the inclusion of SOC during iterations. Local-density approximation is assumed for exchange and correlation in the calculation. All the DFT calculations for the slab model are performed by using all-electron full-potential linearized augmented plane wave (FLAPW) method implemented as the HiLAPW package. Some numerical details follow our previous surface DFT calculations. [5, 8, 18]

3. Results and discussion

In the previous DFT calculation, [15] the electronic band structure of the Au/Si(557) surface was drawn along the line ($\Gamma - K - M$ in Ref. [15]) that is parallel to the Si steps, equivalently the Au wires, showing the doublet feature of the metallic bands originating from SOC. At that time, the term of the Rashba effect was not explicitly stated though certain similarity to the SOC splitting on Au(111) was pointed out. The SOC-split surface bands in Fig. 1(c) of Ref. [15] indeed look like the Rashba origin. It was, however, not clear to catch the exact k position at which the split bands are degenerate in the Brillouin zone and its relation to the time reversal symmetry. As illustrated in Fig. 2, the k line on the k_x axis parallel to the Au wires that was intended to study is *not* a special symmetry line in the 2D Brillouin zone. To clarify the time reversal symmetry on the surface bands, the electronic band structure is plotted in Fig. 3 on $\Gamma - A - \Gamma$, where A is one of the TRIM and the spin flip behavior is expected around it, as well as on the k line along the k_x axis parallel to the Au wires as the same as the previous DFT result. [15] Clear time reversal symmetry can be seen in the largely dispersive metallic bands around A that is one of the TRIM in the Brillouin zone. The band structures on the both lines really resemble each other because they are very close in the Brillouin zone (Fig. 2)

Figure 3: (Color online) Electronic band structures of Au/Si(557) with spin-orbit coupling on (a) $\Gamma - A - \Gamma$ and (b) the k_x axis parallel to the Au wire. The Fermi energy is taken at the origin of energy. The size of open circle marks is proportional to the d component with up (red) and down (blue) spin of the Au atom. The spin quantization axis is assumed perpendicular to the (557) surface plane.

and the corresponding wave functions must be quite analogous. One more set of doublet metallic bands with negligibly small Rashba splitting can be seen in the electronic structure around the Fermi energy. This may be attributed to the edge state (or dangling bond state) relatively localized at the every two Si atoms along the step edge. [15]

As explained in the previous report, [18] the Rashba-split metallic bands consist of Au d and p orbitals strongly hybridized with the neighboring Si p orbitals. Due to the important contribution from the neighboring Si p orbitals, the metallic bands should be called the Au-Si zigzag chain states. Asymmetric electron density around the Au atoms induced by the on-site hybridization of the different-parity orbitals leads to such large Rashba splitting and dominant out-of-plane spin polarization (see Fig. 4 of Ref. [18]). Almost no s component of the Au atoms is involved in the wave functions due to the deep position of the s states (mass-velocity effect). This is a similar feature to the surface states in the Au(111) surface [5] and it is interesting to see how the situation is changed in the case of Ag/Si(557) as a future study. **In this connection, the s level shift by the mass-velocity effect in Au and Ag can be estimated to be -1.67eV and -0.45eV , respectively, from the difference in the conduction band bottom (Γ_1) between non-relativistic and scalar relativistic bulk band structures.**

In order to analyze the 1D nature of the metallic bands, the electronic band structures on some other lines pointing different directions in the Brillouin zone

Figure 4: (Color online) Electronic band structures of Au/Si(557) with spin-orbit coupling (SOC) on (a) $\Gamma - B - \Gamma$, (b) $\Gamma - Y - \Gamma$, and (c) the zone-boundary line centering at A (broken line in Fig. 2). The Fermi energy is taken at the origin of energy. The size of open circle marks is proportional to the d component with up (red) and down (blue) spin of the Au atom. The spin quantization axis is assumed perpendicular to the (557) surface plane. Panel (d) is drawn on the same line as (c) but without SOC, that means that both spin states are degenerate.

are depicted in Fig. 4. Not surprisingly, the band structure on $\Gamma - B - \Gamma$ is almost equivalent to that on $\Gamma - A - \Gamma$, implying another proof of the 1D character not only of the metallic bands but also of other states around the Fermi energy. To see the 1D nature in a more quantitative manner, two lines perpendicular to the Si steps and Au wires, $\Gamma - Y - \Gamma$ and a zone-boundary line centering at A are chosen in the Brillouin zone (see Fig. 2). Interestingly the metallic states possessing large Au d component also show clear Rashba-type spin splitting around A on the line almost perpendicular to the Au wires. Disappointingly no Au- d dominant states appear near the Fermi energy on $\Gamma - Y - \Gamma$. Since the Rashba splitting and the band width due to hopping integrals perpendicular to the Au wires are expected to be of the similar order, the electronic band structure is depicted without SOC in Fig. 4 (d). The obtained band width is indeed small, approximately 0.02eV for the metallic bands around the Fermi energy. This band width is unexpectedly not too small because the distance between the Au wires, equivalently between the (557) step edges exceeds 19Å. This means that the metallic states may have small, but finite 2D character of the wave functions to certain extent on the surface. Furthermore the metallic states show to possess the dominant out-of-plane component of spin.

Figure 5 shows the electron density (wave function squared) of the Rashba-split state near the Fermi energy around A on $\Gamma - A - \Gamma$ plotted on two cross

Figure 5: (Color online) Electronic wave function squared of the Rashba-split metallic state near the Fermi energy around A on $\Gamma-A-\Gamma$ (Fig. 3) in Au/Si(557). Density contour plots are on the ac and bc planes. The parallelepiped denotes the unit cell used in the calculation, the same as in Fig. 1. Large amplitude is seen along the Au (large yellow balls) and neighboring Si zigzag structure but certain two-dimensional extension crossing the terrace of the surface. In the area highlighted with red, the density exceeds $0.001e/\text{Bohr}^3$.

sections of which one is parallel to the Au wire (ac plane) and the other is crossing the terrace of Si(557) surface (bc plane). Large amplitude of the density can be indeed found in the region of the Au and neighboring Si zigzag structure while its wave function extends laterally crossing the terrace of the (557) surface. This 2D nature of the metallic states may account for the finite dispersion of the states perpendicular to the Au wires as shown in Fig. 4. It is also quite natural to see in Fig. 5 that the metallic state remarkably diminishes the wave function toward the bulk direction.

4. Concluding remarks

The electronic character of the SOC-split metallic bands in Au/Si(557) has been analyzed by means of the first-principles DFT calculations. The calculated bands clearly show Rashba-type time reversal symmetry at the boundary center points in the directions nearly parallel and perpendicular to the Au wires. The states are found to be not only predominantly of 1D character along the Au wires but also finite 2D extension of the wave functions on the surface. **It is quite interesting to further investigate the electronic states of 1D structures grown on the other vicinal surfaces.** Quite recently, properly genuine 1D electronic nature of the edge state has been discovered for the Bi single bilayer system that is believed to be a 2D topological insulator. [20, 21, 22] In this context, searching for the edge states of 2D topological insulators might be a further opportunity for investigating the 1D electronic states.

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References

- [1] E. I. Rashba, Sov. Phys. Solid State **2**, 1109 (1960).
- [2] Yu A. Bychkov and R. I. Rashba, J. Phys. C **17**, 6039 (1984).
- [3] G. Dresselhaus, Phys. Rev. **100**, 580 (1955).
- [4] S. LaShell, B. A. McDougall, and E. Jensen, Phys. Rev. Lett. **77**, 3419 (1996).
- [5] M. Nagano, A. Kodama, T. Shishidou, and T. Oguchi, J. Phys.: Condens. Matter **21**, 064239 (2009).
- [6] G. Bihlmayer, Yu M. Koroteev, P.M. Echenique, E. V. Chulkov, and S. Blügel, Surf. Sci. **600**, 3888 (2006).
- [7] T. Oguchi and T. Shishidou, J. Phys.: Condens. Matter **21**, 092001 (2009).
- [8] K. Sakamoto, H. Kakuta, K. Sugawara, K. Miyamoto, A. Kimura, T. Kuzumaki, N. Ueno, E. Annese, J. Fujii, A. Kodama, T. Shishidou, H. Namatame, M. Taniguchi, T. Sato, T. Takahashi, and T. Oguchi, Phys. Rev. Lett. **103**, 156801 (2009).
- [9] P. Segovia, D. Purdle, M. Hengsberger and Y. Baer, Nature **402**, 504 (1999).
- [10] D. Sánchez-Portal, J. D. Gale, A. Garcia, and R. M. Martin, Phys. Rev. B **65**, 081401(R) (2002).
- [11] I. K. Robinson, P. A. Bennett, and F. J. Himpsel, Phys. Rev. Lett. **88**, 096104 (2002).
- [12] J. N. Crain, A. Kirakosian, K. N. Altmann, C. Bromberger, S. C. Erwin, J. L. McChesney, J.-L. Lin, and F. J. Himpsel, Phys. Rev. Lett. **90**, 176805 (2003).
- [13] D. Sánchez-Portal and R. M. Martin, Surf. Sci. **532-535**, 655 (2003).
- [14] J. N. Crain, J. L. McChesney, F. Zheng, M. C. Gallagher, P. C. Snijders, M. Bissen, C. Gundelach, S. C. Erwin, and F. J. Himpsel, Phys. Rev. B **69**, 125401 (2004).
- [15] D. Sánchez-Portal, S. Riikonen, and R. M. Martin, Phys. Rev. Lett. **93**, 146803 (2004).
- [16] I. Barke, F. Zheng, T. K. Rügheimer, and F. J. Himpsel, Phys. Rev. Lett. **97**, 226405 (2006).
- [17] S. Riikonen and D. Sánchez-Portal, Phys. Rev. B **76**, 035410 (2007).

- [18] T. Okuda, K. Miyamaoto, Y. Takeichi, H. Miyahara, M. Ogawa, A. Hara-sawa, A. Kimura, I. Matsuda, A. Kakizaki, T. Shishidou, and T. Oguchi, Phys. Rev. B **82**, 161410(R) (2010).
- [19] S. J. Riikonen, PhD Thesis, Universidad del País Vasco (2007).
- [20] S. H. Kim, K.-H. Jin, J. Park, J. S. Kim, S.-H. Jhi, T.-H. Kim, H. W. Yeom, Phys. Rev. B **89**, 155436 (2014).
- [21] I. K. Drozdov, A. Alexandradinata, S. Jeon, S. Nadj-Perge, H. Ji, R. J. Cava, B. A. Bernevig, and A. Yazdani, arXiv:1404.2598v1 (2014).
- [22] A. Takayama, T. Sato, S. Souma, T. Oguchi, and T. Takahashi, unpublished (2014).