Large out-of-plane spin polarization in a spin-splitting one-dimensional metallic surface state on Si(557)-Au

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We report evidence of a spin-split one-dimensional metallic surface state for the system of Si(557)-Au obtained by means of spin- and angle-resolved photoelectron spectroscopy. Strikingly, considerably large out-of-plane spin polarization which is locked to one specific direction has been observed which is in good agreement with the one-dimensional Rashba spin-splitting surface states caused by the highly anisotropic wave function of the gold chains predicted by first-principles calculation.

Low-dimensional systems are well known as sources for unprecedented and intriguing physical phenomena. These effects arise when electron-confinement and symmetry-breaking effects become dominant. If strong spin-orbit interaction (SOI) is added to this list, then a momentum-dependent spin splitting of the electronic states is likely to happen. This phenomenon, initially derived from the relativistic Dirac equation, is commonly known as the Rashba and its Hamiltonian is written as

$$H_{\text{SOI}} = -\frac{\hbar^2}{4m^2c^2}(\nabla V \times \vec{p}) \cdot \vec{\sigma}, \qquad (1)$$

where V is the electronic potential, $\vec{p}$ the momentum operator, and $\vec{\sigma}$ the vector of Pauli matrices. Unlike the Zeeman splitting, the Rashba spin splitting does not result in a finite global spin orientation in two-dimensional systems as observed in recent experiments.[1] On the other hand, a global spin projection is expected for one-dimensional systems—where electron propagation is restricted to one spatial direction—as theoretically predicted for thin cross-sectional nanowires.[2]

An experimental demonstration of a one-dimensional Rashba effect is yet unavailable. Model systems for this would be Au chains grown on vicinal silicon surfaces such as Si(335), Si(553), Si(557), and Si(775).[3] Particularly, Si(557)-Au appears to be an excellent candidate since its structure has been well established both by x-ray diffraction[4] and by density-functional theory (DFT) calculations.[5] The resulting structure is illustrated in Fig. 1 and shows its main building blocks: Au atomic chains (γ), which grow at the center of the terraces, Si adatoms (α), having a double ($\times 2$) periodicity, and Si honeycomb chains (η). The valence-band electronic structure of this system shows an evident one-dimensional metallic character with parabolic energy dispersion along the chain direction and almost straight Fermi-surface lines perpendicular to them, as observed in angle-resolved photoelectron spectroscopy (ARPES).[3]

Special attention—and a strong controversy—has been generated by the experimental finding that the parabolic bands were doubled. At first, these double bands in Si(557)-Au were interpreted as a spin-charge separation in a Tomonaga Luttinger liquid.[6] Subsequent high-resolution ARPES measurements[7] ruled out this hypothesis after revealing that the two bands do not merge at the Fermi energy. A later first-principles calculation proposed this splitting to be caused by a large SOI, in analogy to the case of the two-dimensional spin-split surface state of Au.[8] Other groups interpreted the origin of these electronic states based on purely structural mechanisms after the experimental finding by temperature-controlled ARPES of differences in the electronic character of each band [one showing a metal-insulator transition at 120 K while the other being insulating (semiconducting) already at room temperature].[9] Furthermore, dimerization of the step-edge Si atoms was reported by low-
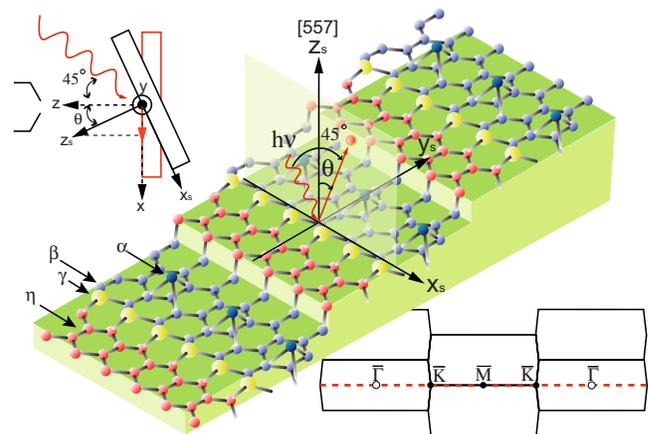

FIG. 1. (Color online) Schematic illustration of Si(557)-Au model and the SBZ as well as the experimental geometry. The atoms labeled α (dark blue), γ (yellow), and η (red) correspond to Si adatoms, Au atoms, and honeycomb-structured Si atoms, respectively. (S)ARPES spectra have been acquired along gold chain direction (x_s) which corresponds to the (red) dashed line in the SBZ.

temperature scanning tunneling microscopy at the transition temperature[10] which could naturally explain each surface state as originating from different surface structure.

However, some recent works have brought back the Rashba-type spin-splitting scenario: First, a recent high-resolution ARPES study on the related system of Si(553)-Au showed the characteristic anticrossing of the surface states at ×2 zone boundary.[11] Second, the observation of a particular surface plasmon on Si(557)-Au points toward a surface state which is spin split.[12]

It is evident that this controversy of the double surface state can only be settled by providing direct evidence of the spin structure of the system. In this Rapid Communication, we present the first spin- and angle-resolved photoelectron spectroscopy (SARPES) measurements of the surface states of Si(557)-Au. As the results, we observe that the surface states show unambiguous spin splitting which is consistent with the first-principles calculation including SOI of Au atoms (Rashba effect). Strikingly, the observed spin polarization is locked one-specific direction that is not in-plane but contains significantly out-of-plane component which, from our first-principles calculations, we attribute to the aniso-tropic spatial distribution of the wave function of Au.

Si(557)-Au surface was prepared by deposition of 0.2 ML Au onto the atomically clean and well-ordered Si(557) surface[13] at a substrate temperature of 650 °C with a post-annealing at 850 °C.[3] The Au coverage was calibrated from a time extrapolation of the reconstructions of Si(111)$\sqrt{3}$ × $\sqrt{3}$-Au ($\theta = 0.67$ ML) and Si(111)5×2-Au ($\theta = 0.4$ ML). The quality and cleanliness of these samples were checked by reflected high-energy electron diffraction, low-energy electron diffraction, x-ray photoemission, and Auger electron spectroscopy. During measurements the surface was re-freshed by short flashes at 850 °C (Ref. 7) and checked af-terward by gauging the quality of the surface states.

ARPES and SARPES measurements were performed at beamlines BL-18A and BL-19A of the Photon Factory (PF), KEK, respectively. The spin-integrated ARPES measurement was done using a SCIENTA SES-100 analyzer, whereas for SARPES measurement we used a homemade high-efficient very-low-energy-electron-diffraction-type spin detector com-bined with a large hemispherical photoelectron energy ana-lyzer (SPECS GmbH. PHOIBOS-150).[14] In this work we set energy and angular resolutions of ~60 meV and ±0.2° for the ARPES and ~100 meV and ±1° for the SARPES mea-surements, respectively. The effective Sherman function of the spin detector was 0.33 as determined from Ni(110).[14] As illustrated in Fig. 1, the angle between the incident synchro-tron beam and the analyzer entrance lenses was fixed at 45°.

Both the ARPES and SARPES spectra have been acquired along Au chain structure, x_s direction, which corresponds to the dashed line shown in the surface Brillouin zone (SBZ) (Ref. 15) (see Fig. 1). This was done by rotating the sample against the $y = y_s$ direction using a photon energy $h\nu = 34$ eV. Since our spin detector can measure transverse spin (P_x: polarization along the x direction of the spin detector) and longitudinal (P_y: y direction) spin components, P_x con-tains a variable contribution of P_{z_s} and P_{x_s} components when the sample is rotated around the $y = y_s$ axis as illustrated in Fig. 1.
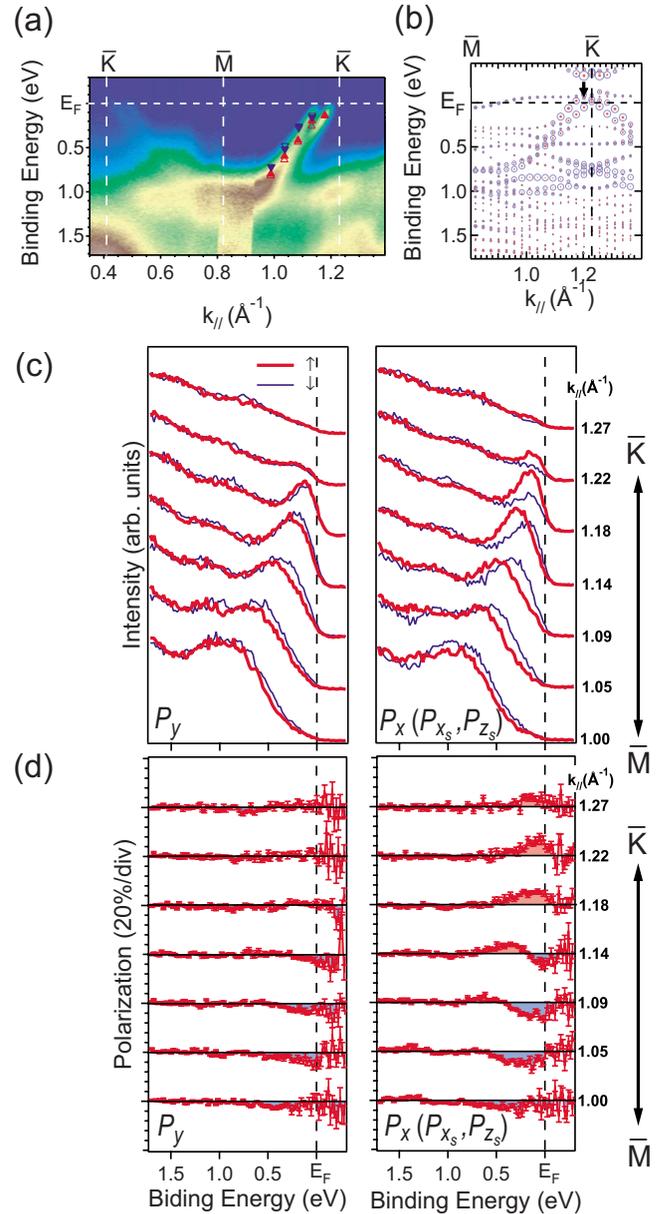

FIG. 2. (Color online) $E - k_\parallel$ maps obtained by (a) spin-unresolved ARPES measurements along the Au chains and (b) by the first-principles calculation including SOI. (c) SARPES spectra of the surface states with P_y (P_{y_s}) and P_x (P_{x_s} and P_{z_s}) polarization components in the spin-detector (sample) coordinate system and (d) its corresponding polarization spectra. Peak positions of (c) are plotted in (a) by triangles (see text).

Figure 2(a) shows a color scale map of the spin-unresolved photoemission intensity vs E and $k_\parallel$. These ARPES data were acquired at $T = 150$ K around the $\overline{\text{M}}$ point of the second SBZ. As has been previously reported, well-resolved double parabolas which disperse to the Fermi en-ergy (E_F) are observed. The photoemission intensity is angle dependent, such that stronger surface state intensities are re-corded at higher emission angles, as previously reported.[7,16] In our measurement we did not observe a clear indication of an energy gap in the S_2 state as reported in Ref. 9. The energy resolution of our ARPES measurement is, however,
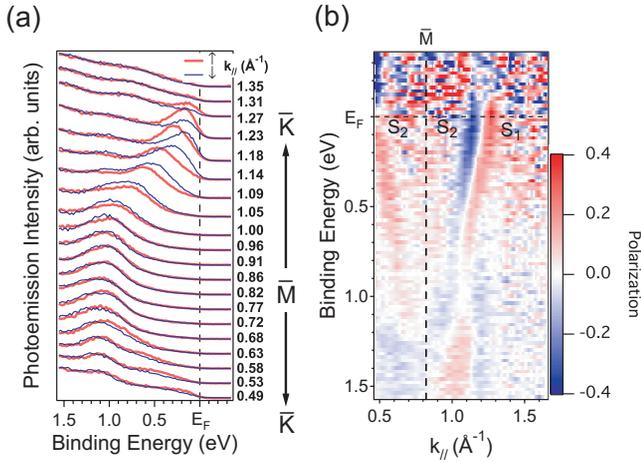
FIG. 3. (Color) (a) SARPES spectra of the P_x (P_{x_s} and P_{z_s}) component in a wider k range than Fig. 2(c). (b) $E-k_\parallel$ polarization map of P_x (P_{x_s} and P_{z_s}) for the same k range as in (a).

FIG. 4. (Color online) Calculated charge distribution of the Au-Si chain structure in the plane determined by Au (γ) and Si (β) atoms (dashed line in the upper inset) around $\bar{K}$ at E_F indicated by arrow in Fig. 2(b). A side view of the model used in the calculations is shown in the inset. A considerable large anisotropy for the charge distribution along the y_s direction [perpendicular to the chain (x_s)] is found at the Au atoms which is believed to cause the out-of-plane spin component (large red arrow in the model).

not enough to investigate precisely the metal-insulator transition and we will not discuss it further.

SARPES spectra and its polarization of the surface states for longitudinal (P_y) and transverse (P_x) spin-detector directions are presented in Figs. 2(c) and 2(d). The spin-up states (red thick lines) correspond to y and x directions in Fig. 1 for P_y and P_x, respectively, having opposite sense for the spin-down states (blue thin lines). Spin polarization is present in both longitudinal and transverse directions: energetically shifted spin-up and spin-down states are clearly observed at $k_\parallel = \sim 1.05$ Å^{-1}, ~ 1.09 Å^{-1}, and ~ 1.14 Å^{-1}. The spin-split states disperse toward the E_F and lose its total intensity at $k_\parallel = \sim 1.22$ Å^{-1} for the spin-down state and at ~ 1.27 Å^{-1} for the spin-up state implying the Fermi-level crossing of these states. Peak positions of spin-up and spin-down states for P_y and P_x in Fig. 2(c) are also plotted in Fig. 2(a) as filled and empty triangles pointing up and down. This observation indicates clearly that the pair of parabolic surface bands in Fig. 2(a) originates from spin-split surface states.

If the origin of this spin splitting is Rashba effect as predicted by recent first-principles calculation, a polarization reversal at time-reversal symmetry points, such as the $\bar{M}$ point ($k_\parallel \sim 0.82$ Å^{-1}) would be observed. In Fig. 3(a) transverse spin (P_x) SARPES spectra of the surface states are shown for an extended k range ($0.49 < k_\parallel < 1.35$ Å^{-1}). Above $k_\parallel = 1.05$ Å^{-1}, spin-split surface states dispersing toward E_F are clearly observed. For wave vectors below 1.00 Å^{-1} the spin splitting decreases and becomes practically zero as it approaches toward the bottom of the parabola, the point $\bar{M}$ of the second SBZ. Crossing the $\bar{M}$ point, although the peak positions of the spin-up and spin-down states are difficult to determine because of the low spectral intensity, the energy shift between spin-up and spin-down states appears again and the binding energy of spin-up state becomes now smaller than that of spin-down state. In other words, a polarization reversal at $\bar{M}$ point is observed. This polarization reversal is also seen in the angle-resolved polarization map of Fig. 3(b) where the polarization reversal of S_2 states against the time-reversal symmetry point, $\bar{M}$ (vertical dashed line) is clearly
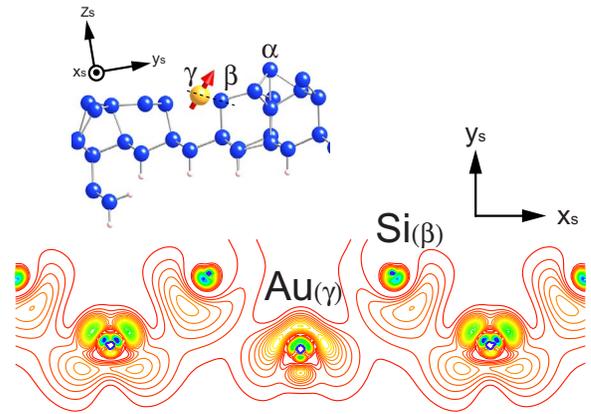
seen. These observations give evidence that the investigated one-dimensional, metallic double surface states become spin split by the Rashba effect rather than the photoemission final-state effect by the SOI.[17,18] This fact is also in agreement with the previous work obtained from first-principles calculations[8] and high-resolution ARPES measurement on Si(553)-Au.[11]

The resulting spin direction of the polarized surface states merits further discussion. In an ideal two-dimensional Rashba case, as extracted from Eq. (1), the spin direction must be orthogonal to both the potential gradient, ∇V (surface-normal direction) and the electron momentum, $\vec{p}$, so that the spins are contained within the surface plane and point tangential to the Fermi surface. Reducing the dimensionality to a one-dimensional case should result in a defined direction along x_s for $\vec{p}$ so that the spin direction would have to be perpendicular to the latter, i.e., have a y_s (longitudinal) direction. However, what we observe in Figs. 2(c) and 2(d), is that the spin polarization is larger along the transverse direction $[P_x (=P_{x_s}, P_{z_s})]$ compared to the longitudinal one $[P_y (=P_{y_s})]$. According to Eq. (1), it is most unlikely that the spin should point along the x_s direction, that is, parallel to $\vec{p}$. Having in mind that P_x is a mixture of P_{x_s} and P_{z_s}, it is straightforward to conclude that this large transverse spin component must be due to an out-of-plane spin component (P_{z_s}) projected onto the x direction as illustrated in Fig. 1. The polar angle for the surface states observation in our SARPES measurements is around 25° ($\sim k_\parallel = 1.1$ Å^{-1}), so that a P_{z_s} component projected onto the x direction would yield about 50% of the total signal, which is therefore observable by our spin P_x channels.

To understand the origin of this P_{z_s} component we have performed first-principles calculations on a Si(557)-Au repeated slab model[8] depicted in the inset of Fig. 4. Our numerical results stem from DFT calculations within the local-density approximation with the all-electron full-potential

linearized augmented plane-wave method. The spin splitting and spin structure have been investigated by switching on and off the spin-orbit coupling terms.

From these calculations the surface double bands which are in good agreement with our observed spin-polarized surface states and with previously reported DFT calculation[8] are reproduced as in Fig. 2(b) supporting that the spin splitting is caused by Rashba effect. The wave-function character of these surface states corresponds to hybridized Au $5d$ and Au $6p$ orbitals [filled and open circles in Fig. 2(b), respectively], which is consistent with electronic states originating from the Au chains and not from the Si step-edge structure. But this does not yet explain the existence of the observed out-of-plane component. Instead, we have calculated the charge density of the parabolic bands, the wave-function squared, $|\psi_{k,n}|^2$ around the Au one-dimensional chain. The charge distribution of the plane formed by the Au (γ in Fig. 1) and the Si (β) atoms (dashed line in the inset of Fig. 4) close to the $\bar{K}$ point at near E_F indicated by arrow in Fig. 2(b) is illustrated in Fig. 4. It is remarkable to observe that adjacent Au atoms are inequivalent, in resemblance to the $\times 2$ periodicity along chain direction found for Si adatoms (α), as reported from diffraction experiments and which upholds a band folding scenario close to the $\bar{K}$ point. Furthermore, the charge distribution surrounding the Au atoms is quite anisotropic along y_s direction, most likely because of the zigzaglike structure of the Au (γ) and Si (β) chains. In this way, the potential gradient should have considerable y_s component so that previously unexpected P_{z_s} spin polarization can arise and even become dominant. Indeed, this is illustrated by the red arrow in the inset of Fig. 4: the calculated spin direction is almost perpendicular to the Au(γ)-Si(β) plane, which is consistent with our SARPES observations.

In conclusion, we have investigated the spin structure of the nearly degenerated double surface states of Si(557)-Au by SARPES. Clear Rashba-type spin-split surface states are experimentally observed, which are corroborated by our first-principles calculations including relativistic effects. These findings allow us to shed some light over the controversial nature of these surface states, finally settling this matter as originating from a Rashba effect. A dominating out-of-plane spin component has been observed and qualitatively explained by our calculation which we attribute to the one-dimensional structure of the Au chains. Further experimental and theoretical investigations are, however, expected to confirm this out-of-plane spin polarization is due to the intrinsic initial-state effect rather than the photoemission final-state effect.[18] The demonstration of locked, dominant out-of-plane spin polarization in one-dimensional metallic surface states will very likely stimulate revitalized research in metallic one-dimensional systems within the spintronics and theory communities.

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