PHYSICAL REVIEW B **94**, 241109(R) (2016)

Hidden Rashba spin-split states in a quasi-one-dimensional Au atomic chain on ferromagnetic Ni(110)

Takuya Warashina,[1] Munisa Nurmamat,[1] Koji Miyamoto,[2] Tatsuya Shishidou,[3] Masaki Taniguchi,[2] Akio Kimura,[1] and Taichi Okuda[2,*]

[1]*Graduate School of Science, Hiroshima University, 1-3-1 Kagamiyama, Higashi-Hiroshima 739-8526, Japan*
[2]*Hiroshima Synchrotron Radiation Center (HSRC), Hiroshima University, 2-313 Kagamiyama, Higashi-Hiroshima 739-0046, Japan*
[3]*Graduate School of Advanced Sciences of Matter, Hiroshima University, 1-3-1 Kagamiyama, Higashi-Hiroshima 739-8530, Japan*

(Received 23 September 2016; revised manuscript received 16 November 2016; published 15 December 2016)

An electronic structure of a Au atomic chain on Ni(110), which was investigated decades ago, has been reinvestigated by high-resolution spin- and angle-resolved photoemission spectroscopy. Clear evidence of a Rashba spin split, i.e., spin-polarization reversal between positive and negative wave vectors with respect to the symmetry point, has been observed in the electronic bands possessing quasi-one-dimensional character. The observed spin-split state caused by spin-orbit interaction (SOI) was overlooked in the previous spin-resolved photoemission study probably due to the dominant effect of exchange splitting of the Ni substrate than the Rashba effect and poorer resolution of the previous measurement. The system containing both the Rashba-type SOI and the exchange interaction with one-dimensional character will provide a platform to investigate the effect of exchange interaction on the spin-split states caused by SOI with low dimensionality.

DOI: 10.1103/PhysRevB.94.241109

Spin-polarized electronic states at materials' surfaces caused by strong spin-orbit interaction (SOI) and breaking of space inversion symmetry have attracted much attention because of the potential application for the next-generation spin-related electronics. Such spin-split surface bands that appear in Rashba systems [1,2] or in topological insulators [3,4] are shifted in k and protected by time-reversal symmetry resulting in the spin degeneracy at the time-reversal invariant momenta (TRIM). Because of the spin reversal with respect to the TRIMs, the suppression of backward scattering of the electron and the dissipationless electron transport is expected. In the case of a one-dimensional system since the direction of electron scattering is also restricted geometrically, more effective suppression of backscattering can be expected.

The effect of exchange interaction on the spin-split bands by SOI, however, is not established. For example, it has been reported that the spin degeneracy at the TRIM can be lifted by introducing some Mn doping in Bi_2Se_3, the archetypal three-dimensional topological insulator, accompanying the energy gap opening at the Dirac point and emergence of peculiar hedgehog spin structure [5]. However, a recent report suggested that the gap opening is not due to the ferromagnetic order or local magnetic moment of Mn atoms but due to the strong resonant scattering by the impurities [6]. In addition, it is reported that the magnetic impurities by evaporation on the topological insulators do not induce any significant change in the electronic states of topological surface states (TSSs) [7,8].

From the similarity of the origin of the spin-split surface states by the Rashba effect and TSSs, a similar effect on the spin-split surface states by a magnetic moment in the Rashba system is expected. Therefore, the exploration of systems where Rashba SOI and exchange interaction coexist is probably helpful to address the issue. As such systems Gd(0001) ($Z = 64$) and its surface monoxide as well as Tb(0001) ($Z = 65$) surfaces have been reported so far [9,10]. In these systems, a small k shift by switching the magnetization direction is observed by angle-resolved photoemission spectroscopy (ARPES) in the energetically split majority- and minority-spin bands by the large exchange interaction [9,10]. A similar asymmetric k shift of exchange split bands by switching magnetization is also recently reported on a Co(0001) film grown on a W(110) surface [11]. In this case, the interface of the film and high-Z W crystal ($Z = 74$) plays a role to induce the Rashba effect in the Co film. The effect observed in these examples is, however, the effect of Rashba SOI which shifts slightly the majority- and minority-spin bands, and the systems are not suitable to investigate the effect of exchange interaction on the spin-split bands caused by Rashba SOI.

In this context, a recently reported Fe film on W(110) is one of the examples that tries to show the effect of exchange interaction on the SOI-induced spin-split bands [12]. In the report the authors claimed that the Dirac-cone-like surface states [13] show the energy gap of 340 meV at the Dirac point when the thickness of the Fe film is 1.7 ML (monolayer) in which the magnetic moment of the Fe film is considered to be out of plane. The observation reminds us of the aforementioned energy gap opening of Bi_2Se_3 by Mn doping [5]. In the thicker and thinner Fe films in which in-plane magnetic moment is energetically more stable in the Fe/W(110) system, however, the expected shifts of the spin-split bands in k by the in-plane ferromagnetic order of the Fe film have not been observed, and the effect of exchange interaction on the SOI-induced spin-split bands is still not evident. Therefore, in order to further understand the effect of exchange interaction on the SOI-induced spin-polarized surface states the exploration and investigation of new systems with both strong SOI and exchange interaction, i.e., the systems consisting of heterostructures of heavy materials and ferromagnetic materials, must be important.

The Au/Ni(110) system is such a system being made up of heavy element Au and a ferromagnetic Ni substrate. The previous scanning tunneling microscopy (STM) observation revealed that the Au evaporation on Ni(110) gives rise to

*okudat@hiroshima-u.ac.jp

the formation of a quasi-one-dimensional Au atomic chain structure [14]. The band structure of the system has been observed by ARPES and shows clear one-dimensional character being consistent with the STM observation [15]. In the study, the minority-spin character in the one-dimensional state has also been observed by spin-resolved photoemission, and it is suggested that electron confinement is spin dependent at the interface of the Au atomic chain and Ni substrate. However, the possibility of the Rashba spin split was not discussed because the Rashba effect at materials' surfaces had not become aware at that moment.

Here, in this Rapid Communication, we report experimental evidence that the Au chain in the Au/Ni(110) system possesses Rashba spin-split states with one-dimensional character, which is proximal to the ferromagnetic substrate by high-resolution spin-ARPES measurement. The long spin coherent length expected in the one-dimensional Rashba system of the Au chain and the possible modification of the state by the exchange interaction by the Ni substrate is attractive as a potential application of the system for the spintronic devices.

The Ni(110) substrate was cleaned by standard Ar^+ sputtering and annealing at about 970 K. The Au atomic chain structure is self-assembled by Au evaporation on the Ni(110) substrate at room temperature using the tungsten filament whose evaporation rate was calibrated by quarts microbalance in advance. After the evaporation the sample was postannealed at 700 K for 15 min to get a better ordered chain structure. The sample characterization was performed by low-energy electron diffraction (LEED), Auger electron spectroscopy, and STM observation. As reported in the previous studies a density of the atomic chain and the distance between chains change as a function of Au coverage. Evaporation of $\theta_{\mathrm{Au}} =$ 0.7–0.9-ML Au results in well-ordered quasi-one-dimensional atomic chains as shown in Fig. 1(c) which was taken at 77 K using LT-STM (where LT represents low temperature) (Omicron GmbH) [16]. The observed chain structure with some undulation is in good agreement with the previous STM

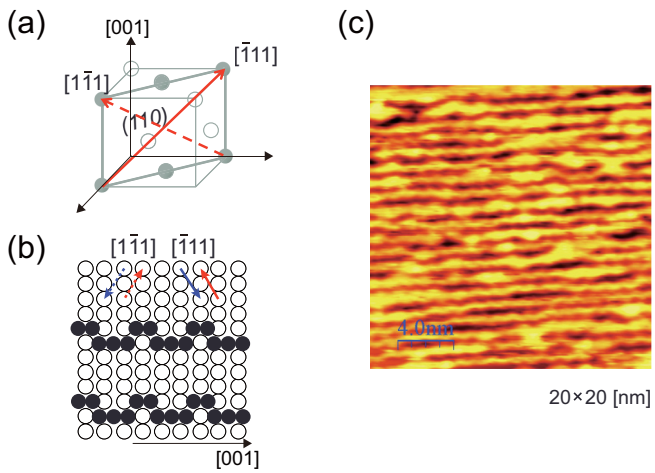

FIG. 1. (a) Schematic crystal structure of bulk Ni fcc and the magnetic easy axes. (b) Model of a Au chain (black circles) on a Ni(110) substrate (white circles) proposed from the STM observation by Nielsen *et al.* [14]. (c) Real-space image of a self-organized Au atomic chain on a Ni(110) substrate observed by scanning tunneling microscopy at $T = 77$ K with Au coverage of $\theta_{\mathrm{Au}} = 0.7$ ML.

study [14] in which a zigzag chain structure on the Au wetting layer is proposed.

The electronic structure of Au/Ni(110) has been investigated by spin-ARPES at the ESPRESSO end station [17] where three-dimensional spin vector analysis can be carried out using two orthogonally set highly efficient very-low-energy-electron-diffraction (VLEED) spin polarimeters combined with a Scienta R4000 hemispherical analyzer [18]. The end station is located at the beamline BL-9B of the Hiroshima Synchrotron Radiation Center which equips the quasi periodic APPLE II undulator [19]. The incidence angle of synchrotron radiation light ($h\nu = 22.0$ eV) and light from a He discharge lamp (He I α; $h\nu = 21.22$ eV) is fixed 50° from the analyzer lens. The overall experimental resolutions were set to $\Delta E \sim$ 20 meV and $\Delta\theta \sim 0.3°$ and $\Delta E \sim 25$ meV and $\Delta\theta \sim \pm 0.7°$ in spin-integrated and spin-resolved ARPES measurements. All results presented here were taken at a sample temperature of 15 K and at a base pressure of 6×10^{-9} Pa [20].

Figure 2 shows (a) the band structure of Au/Ni(110) ($\theta_{\mathrm{Au}} = 0.7$ ML) taken along the atomic chain direction with an unpolarized He discharge lamp as well as (b) the constant energy contours (CECs) at the binding energies (E_B) of 1.2 and 2.3 eV. In addition to the Ni 3*d* bands at around $E_B =$ 0–1.5 eV, other bands labeled S1–S3 have been observed at the higher E_Bs [21]. The S1 state shows paraboliclike band dispersion along the Au chain direction which is in good agreement with the previous study [15]. One dimensionality of the band is clearly seen in the CECs at $E_B = 1.2$ and 2.3 eV showing an almost straight line along $\boldsymbol{k}$ perpendicular to the chain. On the other hand S2 and S3 states are newly observed in this Rapid Communication. Whereas the S3 state shows strong one-dimensional character as seen in the CEC plots, the S2 state that has upward convex parabolas with the maximum at $k_{\parallel} \sim \pm 0.59$ Å^{-1} shows some undulation in the CEC at $E_B = 2.3$ eV implying the non-negligible interaction of the states between adjacent one-dimensional chains. Since the Au 5*d* band can extend to around $E_B \sim 2$ eV the observed surface bands probably consist of Au 6*s*, 6*p*, and 5*d* states. The large dispersion of the surface states, however, suggests that the bands are derived mainly from Au 6*s* and 6*p* states.

Using *p*- and *s*-polarized light from the APPLE-II undulator one can investigate the character of the wave function of the states. In the case of *p*- (*s*-) polarized light the electric-field vector is in (perpendicular to) the electron-scattering plane which is spanned by light incidence and a photoelectron propagation vector as in the inset of Fig. 2(c) and one can excite the states having even (odd) symmetry s, p_x, and p_z (p_y) with respect to the scattering plane. As in Fig. 2(c) the S1 state is observed with *p*-polarized light but not with *s*-polarized light. On the contrary the S2 state is only observable with *s*-polarized light. Considering the aforementioned selection rule of dipole transition the S2 (S1) state has odd (even) symmetry with respect to the scattering plane. Since the scattering plane is parallel to the Au atomic chain in our measurement the S2 state has a wave function spatially spreading perpendicular to the atomic chain and can interact with the adjacent chain. In addition, the observed undulation of the band in the CEC of the S2 state suggests the validity of this simple interpretation. Note that the reason why the S2 state was not observed in the previous photoemission study can also be explained by the

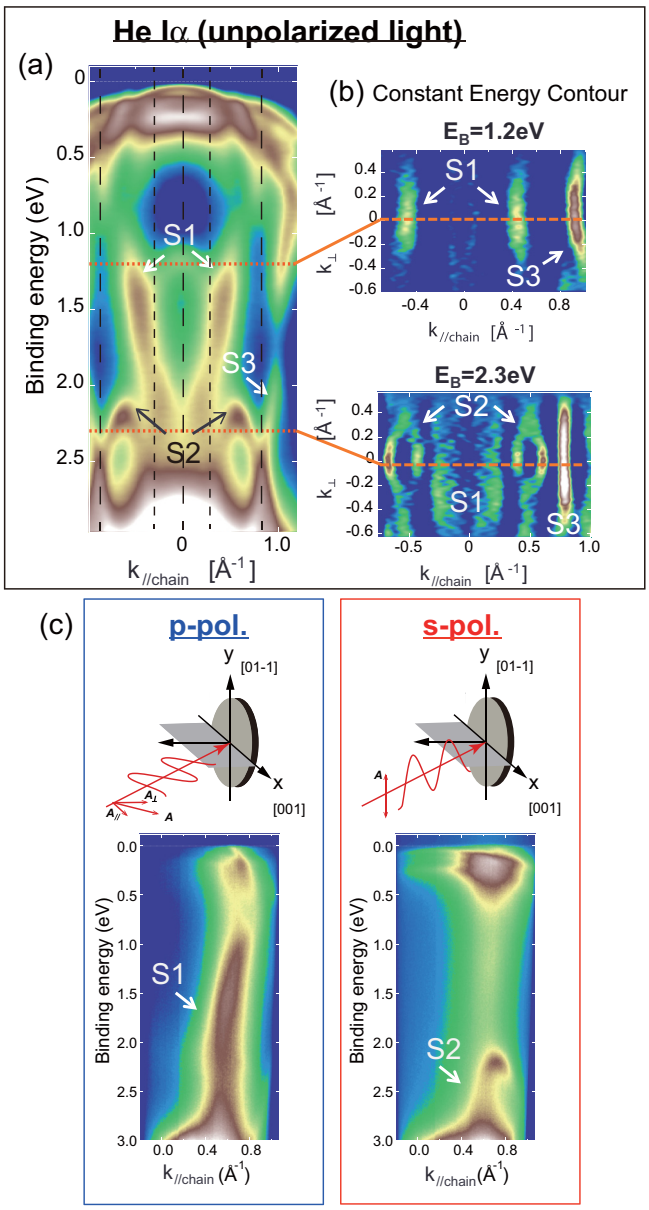

FIG. 2. (a) Band dispersion along a Au atomic chain observed by angle-resolved photoemission spectroscopy at $T = 15$ K with an unpolarized He discharge lamp ($h\nu = 21.22$ eV). Three surface states labeled S1–S3 are observed in addition to Ni $3d$ bands which is almost the same as the clean Ni(110) surface. (b) Constant energy contours of the bands at the binding energies (E_B) of 1.2 and 2.3 eV show one dimensionality of these bands derived from the Au atomic chain. Small undulation of the S2 state suggests the non-negligible interaction between chains in this state. (c) ARPES results taken with synchrotron radiation light ($h\nu = 22$ eV) using different photon polarizations (p polarized and s polarized). The S1(S2) state is selectively observable with p- (s-) polarized light.

polarization dependence of photoemission intensity since the previous photoemission measurement was performed using only p-polarized light [15].

Since the surface state bands are derived from the Au atomic chain there is a possibility that the state is spin polarized by the strong SOI of Au. Figure 3 shows the results of

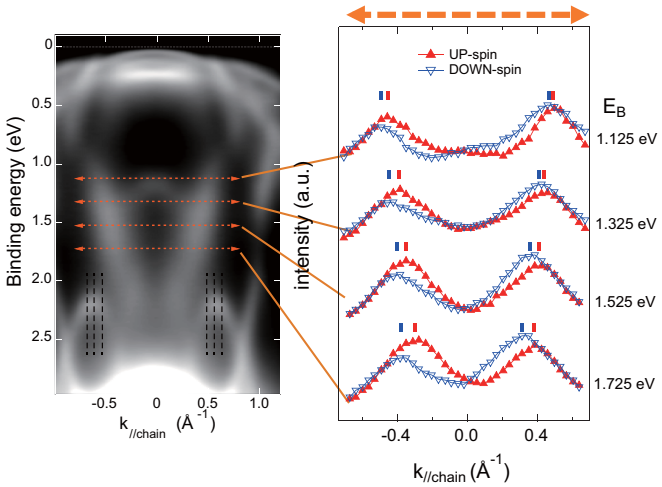

FIG. 3. (Right) Spin-resolved momentum distribution curves (spin-MDCs) taken at the binding energies indicated with dotted lines in the ARPES band map (left). The red (blue) triangles represent the intensity of the up- (down-) spin photoemission signal. The opposite shift of up- and down-spin states in $k_{\parallel \mathrm{chain}}$ strongly suggests the Rashba spin splitting of the S1 state.

the spin-ARPES observation in which the spin polarization perpendicular to the electron momentum [i.e., perpendicular to the atomic chain; y direction in Fig. 2(c)] is measured as a function of electron momentum (i.e., spin-resolved momentum distribution curve; spin-MDC). Such spin-MDCs are measured with a He discharge lamp at $E_B = 1.125$, 1.325, 1.525, and 1.725 eV, respectively, which are at the dotted lines in the ARPES band mapping (left-hand side of Fig. 3). All the spin-MDCs show clear shifts of the peak positions in k between up- and down-spin states. Namely, the peaks of the down-spin state shift to negative wave vectors whereas those of the up-spin state shift to positive wave vectors with respect to $\bar{\Gamma}$. The opposite shift with respect to the $\bar{\Gamma}$ point for the opposite spin state strongly suggests that the observed shift is caused by the Rashba effect. The value of shifts at the constant energy cut is expected to be constant in different E_B's in the ideal Rashba spin-split bands of a two-dimensional electron gas. The average value of the shift is $\Delta k_R = 0.025 \pm 0.08$ Å^{-1} [22]. The observed Δk_R is larger than that in Au(111) ($\Delta k_R = 0.024$ Å^{-1} [23,24]) and smaller than the one in Au/Si(557) ($\Delta k_R = 0.050$ Å^{-1} [25]).

In the previous study Pampuch and co-workers also have performed a spin-ARPES observation on the S1 band [15]. They have reported a minority-spin character of the S1 state that is antiparallel to the majority-spin polarization of substrate Ni. The minority character of the S1 state is explained by considering the spin-dependent electron scattering at the interface of the Au atomic chain and the substrate Ni in analogy with spin-polarized quantum well states in the Cu/Co(100) system [26,27]. Since the spin-ARPES measurement was performed with a magnetized Ni substrate in the previous paper the large spin-polarization change as the function of energy in their spin-resolved energy distribution curve (spin-EDC) may hinder the relatively small spin-polarization change by the Rashba effect. The lower-energy and angular resolutions ($\Delta E = 150$ meV and $\Delta\theta = \pm 2^\circ$) of the previous

measurement also hamper observing the smaller spin polarization caused by the Rashba effect. On the contrary, in our measurement the Ni substrate is not magnetized after the sample preparation where the Ni substrate is annealed higher than its Curie temperature (627 K). If we assume that the magnetic domain in the sample is substantially smaller than the beam spot size of our measurement ($\sim 1 \times 2$ mm) the observed spin polarization can solely be attributed to the Rashba SOI. Thus, the spin-ARPES results of the previous study and the present Rapid Communication do not conflict each other. In addition, in the previous study peak broadening of the S1 state as the function of electron momentum is observed. Although the peak broadening was attributed to the Bragg reflection gap expected by the band folding caused by the $\sim \times 3$ periodicity of the zigzag structure (three times of the Ni lattice constant derived from the zigzag chain model in Ref. [14]) in the previous study a clear indication of a gap opening is not observed at the zone boundary of $\times 3$ periodicity [dashed line at 0.297 Å^{-1} in Fig. 2(a)] in our measurement. The wider peak width at the larger emission angle observed in the previous study, however, can be caused by the combination of poorer momentum resolution and the steeper band dispersion and/or the band split by the Rashba effect.

As we discussed above the contribution of the exchange interaction in the observed spin polarization is probably canceled out. However the rather large peak width of the S1 state might come from the contribution of the exchange split bands from different domains. As shown by the arrows in Fig. 1(a) the easy magnetization axes of Ni are in the $\langle 111 \rangle$ direction and correspond to the $[1\bar{1}1]$ or the $[\bar{1}11]$ direction on the (110) plane. The contribution of exchange coupling along the chain direction can shift the Rashba spin-split bands energetically as indicated in Fig. 4 and discussed in Ref. [9]. As illustrated in the figure the direction of the energy shift should be opposite between opposite magnetization directions. On the nonmagnetic substrate (i.e., multidomain substrate) contributions from these different domains which are equally distributed can be observed together in the ARPES measurement and cause peak broadening of the S1 band. It should be noted that even with the contributions of many domains the shift by the Rashba effect should be observed correctly and the Δk_R value by the Rashba effect is not affected by the mixture of different magnetic domains as simulated in Fig. 4. This speculation for the peak broadening can be examined with the magnetized substrate using a picture-framed Ni(110) crystal [28] in the near future.

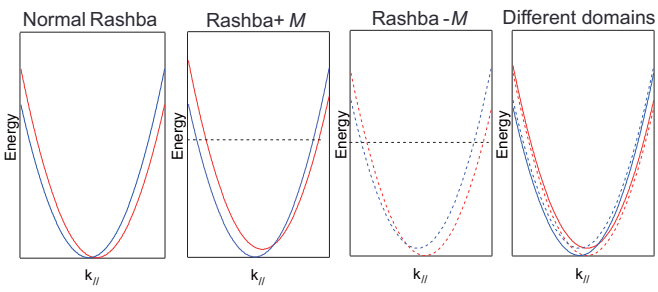

FIG. 4. Schematic of the expected Rashba spin-split band modification by exchange interaction induced by the ferromagnetic substrate. In the case of the nonmagnetic (=multidomain) substrate, contributions from both positively and negatively magnetized domains are mixed up.

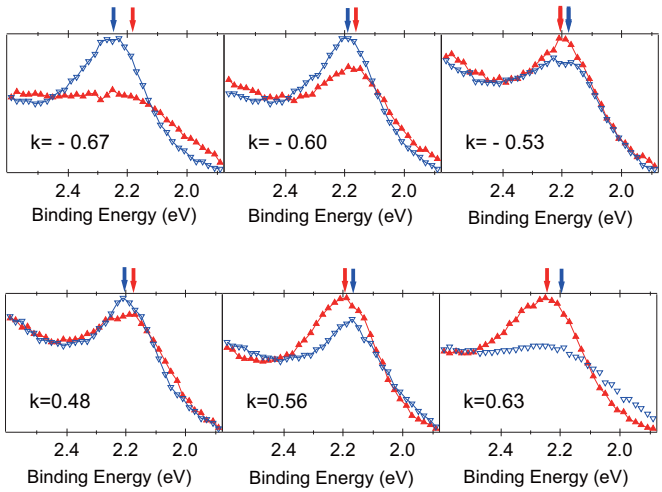

FIG. 5. Spin-resolved energy distribution curves of the S2 state taken at the k points indicated by the vertical dashed lines in the ARPES band map in Fig. 3. The red (blue) upward (downward) triangles represent the intensity of the up- (down-) spin photoemission signal. Clear spin-polarization reversal with respect to the $\bar{\Gamma}$ point ($k = 0$ Å^{-1}) is observed. The difference in the peak position between up- and down-spin states is bigger at larger $|k|$ values.

Finally we have also measured the spin polarization of the S2 state. Spin-EDC at several k points that are indicated in Fig. 3 by vertical dashed lines are presented in Fig. 5. The peak positions of up- and down-spin states are indicated with blue and red arrows in the figure. It looks that the energy difference of the up- and down-spin bands reduces as the wave vector approaches $\bar{\Gamma}$ both in negative and positive k values. Clear spin polarization and its inversion with respect to the $\bar{\Gamma}$ point suggest that the S2 state is also a Rashba spin-split state.

In conclusion, we have revisited the system of a Au atomic chain on a Ni(110) surface and investigated the spin texture of the sample by high-resolution spin- and angle-resolved photoemission spectroscopy. Au-induced three electronic states having one-dimensional character have been observed in the self-assembled Au atomic chain by 0.7-ML Au on the Ni(110) surface. Clear experimental evidence of the Rashba spin split in the quasi-one-dimensional electronic states is observed in the system. The system with one-dimensional Rashba spin-split states on a magnetic substrate will provide a platform to study the effect of exchange coupling on the SOI-induced spin-split states.

This work was partly supported by KAKENHI, Grant-in-Aid for Scientific Research Kiban(B), Kiban(A), and Wakate(B) of the Japan Society for the Promotion of Science (Grants No. 19340078, No. 23244066, and No. 25800179). The experiments were performed with the approval of the Proposal Assessing Committee of the Hiroshima Synchrotron Radiation Center (Proposal No. 12A-25).

[1] E. I. Rashba, Sov. Phys. Solid State **2**, 1109 (1960).
[2] Y. A. Bychkov and E. I. Rashba, JETP Lett. **39**, 78 (1984).
[3] C. L. Kane and E. J. Mele, Phys. Rev. Lett. **95**, 146802 (2005).
[4] B. A. Bernevig and S. C. Zhang, Phys. Rev. Lett. **96**, 106802 (2006).
[5] S.-Y. Xu, M. Neupane, C. Liu, D. Zhang, A. Richardella, L. A. Wray, N. Alidoust, M. Leandersson, T. Balasubramanian, J. Sánchez-Barriga, O. Rader, G. Landolt, B. Slomski, J. H. Dil, J. Osterwalder, T.-R. Chang, H.-T. Jeng, H. Lin, A. Bansil, N. Samarth, and M. Z. Hasan, Nat. Phys. **8**, 616 (2012).
[6] J. Sánchez-Barriga, A. Varykhalov, G. Springholz, H. Steiner, R. Kirchschlager, G. Bauer, O. Caha, E. Schierle, E. Weschke, A. A. Ünal, S. Valencia, M. Dunst, J. Braun, H. Ebert, J. Minár, E. Golias, L. V. Yashina, A. Ney, V. Holý, and O. Rader, Nat. Commun. **7**, 10559 (2016).
[7] M. R. Scholz, J. Sánchez-Barriga, D. Marchenko, A. Varykhalov, A. Volykhov, L. V. Yashina, and O. Rader, Phys. Rev. Lett. **108**, 256810 (2012).
[8] M. Ye, S. V. Eremeev, K. Kuroda, E. E. Krasovskii, E. V. Chulkov, Y. Takeda, Y. Saitoh, K. Okamoto, S. Y. Zhu, K. Miyamoto, M. Arita, M. Nakatake, T. Okuda, Y. Ueda, K. Shimada, H. Namatame, M. Taniguchi, and A. Kimura, Phys. Rev. B **85**, 205317 (2012).
[9] O. Krupin, G. Bihlmayer, K. Starke, S. Gorovikov, J. E. Prieto, K. Döbrich, S. Blügel, and G. Kaindl, Phys. Rev. B **71**, 201403(R) (2005).
[10] O. Krupin, G. Bihlmayer, K. M. Döbrich, J. E. Prieto, K. Starke, S. Gorovikov, S. Blügel, S. Kevan, and G. Kaindl, New J. Phys. **11**, 013035 (2009).
[11] P. Moras, G. Bihlmayer, P. M. Sheverdyaeva, S. K. Mahatha, M. Papagno, J. Sánchez-Barriga, O. Rader, L. Novinec, S. Gardonio, and C. Carbone, Phys. Rev. B **91**, 195410 (2015).
[12] K. Honma, T. Sato, S. Souma, K. Sugawara, Y. Tanaka, and T. Takahashi, Phys. Rev. Lett. **115**, 266401 (2015).
[13] K. Miyamoto, A. Kimura, K. Kuroda, T. Okuda, K. Shimada, H. Namatame, M. Taniguchi, and M. Donath, Phys. Rev. Lett. **108**, 066808 (2012).
[14] L. P. Nielsen, F. Besenbacher, I. Stensgaard, E. Laegsgaard, C. Engdahl, P. Stoltze, and J. K. Norskov, Phys. Rev. Lett. **74**, 1159 (1995).
[15] C. Pampuch, O. Rader, T. Kachel, W. Gudat, C. Carbone, R. Klasges, G. Bihlmayer, S. Blugel, and W. Eberhardt, Phys. Rev. Lett. **85**, 2561 (2000).
[16] For STM analysis, the free software WSXM was used, http://www.wsxmsolutions.com/.
[17] T. Okuda, K. Miyamoto, H. Miyahara, K. Kuroda, A. Kimura, H. Namatame, and M. Taniguchi, Rev. Sci. Instrum. **82**, 103302 (2011).
[18] T. Okuda, K. Miyamoto, A. Kimura, H. Namatame, and M. Taniguchi, J. Electron Spectrosc. Relat. Phenom. **201**, 23 (2015).
[19] S. Sasaki, A. Miyamoto, K. Goto, M. Arita, T. Okuda, T. Mitsuyasu, K. Fujioka, H. Namatame, and M. Taniguchi, J. Phys.: Conf. Ser. **425**, 032009 (2013).
[20] The He partial pressure in the use of a He discharge lamp was 6×10^{-7} Pa.
[21] The state at $E_B = 1.2$ eV at the $\bar{\Gamma}$ point is the replica state of the one at about $E_B = 3.0$ eV excited by the He I β line.
[22] Since the bottom of the parabola is not clear and determining the Rashba parameter is not possible, we compare only the shift with some other systems.
[23] S. LaShell, B. A. McDougall, and E. Jensen, Phys. Rev. Lett. **77**, 3419 (1996).
[24] M. Hoesch, M. Muntwiler, V. N. Petrov, M. Hengsberger, L. Patthey, M. Shi, M. Falub, T. Greber, and J. Osterwalder, Phys. Rev. B **69**, 241401(R) (2004).
[25] T. Okuda, K. Miyamaoto, Y. Takeichi, H. Miyahara, M. Ogawa, A. Harasawa, A. Kimura, I. Matsuda, A. Kakizaki, T. Shishidou, and T. Oguchi, Phys. Rev. B **82**, 161410(R) (2010).
[26] K. Garrison, Y. Chang, and P. D. Johnson, Phys. Rev. Lett. **71**, 2801 (1993).
[27] C. Carbone, E. Vescovo, O. Rader, W. Gudat, and W. Eberhardt, Phys. Rev. Lett. **71**, 2805 (1993).
[28] M. Donath, Surf. Sci. Rep. **20**, 251 (1994).