

Spin- and angle-resolved photoemission study of MAX phase compound V_2AlC

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MAX phases ($M_{n+1}AX_n$, where M is an early transition metal, A belongs to groups 13-15 and X is either C or N, $n = 1 - 3$) have recently attracted much attention due to their possible application to the production of a new class of two-dimensional (2D) systems called MXenes [1]. However, the bulk electronic structure of MAX phases has been studied mostly through *ab initio*, DFT calculations, mainly due to a lack of single crystalline samples. We have performed angle-resolved photoemission spectroscopy (ARPES) on several MAX phase single crystals to directly investigate the electronic structure of these systems [2,3]. Among the MAX phases, V_2AlC has been expected to be categorized as a high-symmetry point semi-metal with crossing point with some Dirac-like properties referred as “Dirac point (DP)” at the Fermi level (E_F) along ΓM line together with “nodal-line (NL)” around 0.2 eV from DFT calculation [3, 4]. To elucidate the electronic structure, especially, the existence of DP and spin-polarized states, we have performed spin - and angle - resolved ARPES (SARPES) with using a quasi - continuous - wave laser [5].

As a result, we have found the formation of the Dirac cone is consistent with DFT calculation (Fig.1). Furthermore, the SARPES clearly demonstrates spin-polarized branches (S+ and S-) in the larger electron pocket (eL), where the spin directions are opposite to each other and tangential to the Fermi surface along the ΓM line (Fig.2). Furthermore, missing of the out-of-plane spin polarization as well as inversion symmetry breaking at the observed

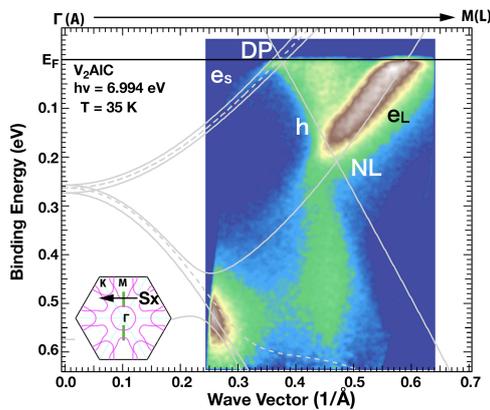


Fig. 1: Band structure along ΓM line of V_2AlC . Gray solid (dashed) lines are obtained from DFT calculation along the ΓM (AL) line. The definition of spin polarization axis in Fig. 2 is indicated by arrow on the DFT Fermi surface in the inset.

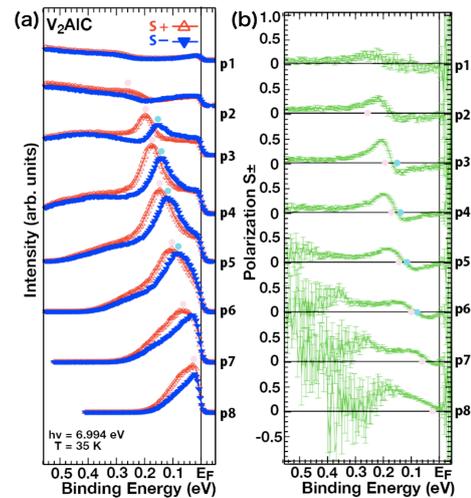


Fig. 2: SARPES spectra (S+: Δ ; S-: \blacktriangledown) (a) and spin-polarization (b) of V_2AlC at the momentum cut from p1 to p8 in Fig. 1.

spin-polarization with respect to the Γ point have been elucidated. There are at least two types of mechanism to understand the origin of the observed spin-textures. One is the existence of helical spin texture around the \bar{M} point with anti-parallel spin splitting due to Rashba effect [6]. The other is the formation of spin-polarized “drum-head surface states” connecting NL [7]. We expect that the present observation of the spin-textures on V_2AlC might suggest the potential of layered MAX phases as new series of topological materials.

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SPIN-ORBITAL TEXTURE IN QUASI-ONE-DIMENSIONAL GIANT RASHBA SYSTEM ON BI/INAs(110)-(2×1)

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Low-dimensional electronic structures with helical spin-polarization due to spin-orbit interaction (SOI) have been extensively studied for not only basic physics but also applications to spintronic devices [1-3]. Recently, it is revealed that the spin texture of many spin-polarized two-dimensional (2D) surface states is not a simple helical one, but entangled with the orbital characters and symmetry operations of the ground states [4-6]. In such case, the spin polarization orientation of the photoelectrons drastically changes, even inverts its sign, depending on the polarization of the incident photons. So far, such spin-orbital entanglement effects have been studied in 2D systems but not extended in one-dimensional (1D) or Q1D systems yet.

In this study, we observed detailed spin-orbital texture of spin-polarized Q1D surface state on Bi/InAs(110)-(2×1) by using laser-based spin-resolved photoemission system (laser-SARPES) at ISSP. Since the surface state of this sample shows one of the largest sizes of Rashba-type spin splitting among Q1D systems [7], it is a suitable material to investigate the role of spin orbital entanglement in Q1D systems.

Figures 1 (a) and (b) show the band dispersions of the Bi/InAs(110)-(2×1) surface along Q1D chain direction measured by conventional (spin-integrated) channel of the laser-SARPE setup with p- and s- polarized photons, respectively. The pair of parabolic bands in both Figs 1 (a) and (b) lie slightly below the Fermi level. The photoelectron intensity of the parabolic band is strong overall the parabola in Fig. 1 (a), whereas its intensity decays around the centre of the surface Brillouin zone in (b), reflecting the difference in the orbital symmetry composing the parabolic band.

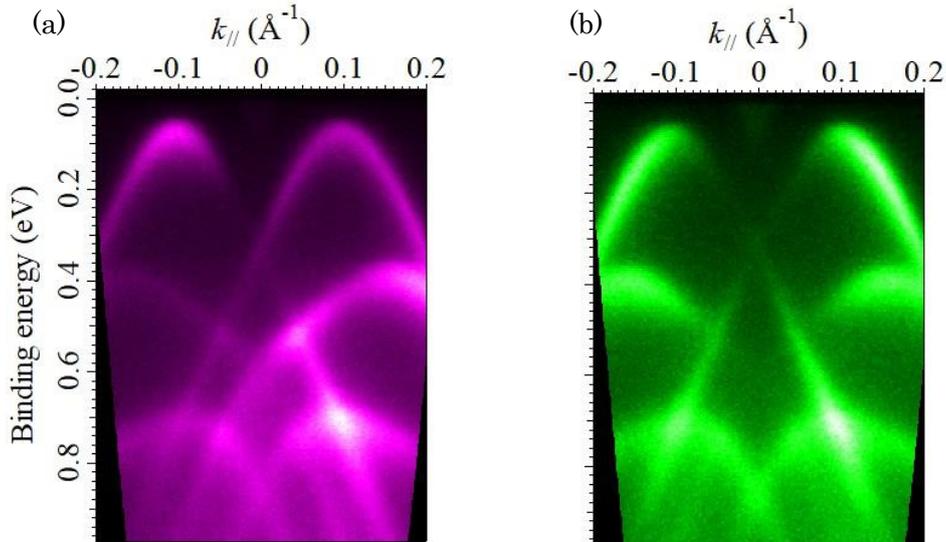


Figure 1. ARPES intensity plots of Bi/InAs(110)-(2×1) along $\bar{\Gamma}-\bar{X}$ (parallel to the surface Bi chains) measured with (a) p-polarized and (b) s-polarized photons at 45 K.

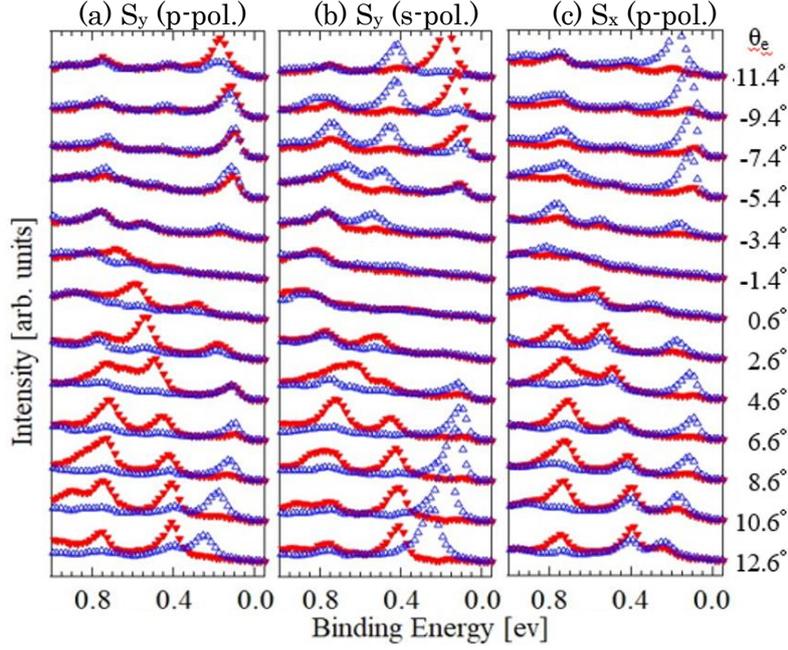


Figure 2. Spin-resolved energy distribution curves of Bi/InAs(110)-(2 \times 1) along $\bar{\Gamma}$ - \bar{X} (parallel to the surface Bi chains). The solid (open) triangles correspond to spin polarization parallel (antiparallel) to (a-b) [001] and (c) $[\bar{1}10]$ direction. $\theta_e = 10^\circ$ corresponds to $k_{||} = 0.14 \text{ \AA}^{-1}$.

Figure 2 (a) and (b) show the spin-resolved energy distribution curves (EDCs) taken with p- and s-polarized lights, respectively. The peak positions of the EDCs agree well with the surface band dispersions shown in Fig. 1. The spin polarizations of the peaks in Figs. 2 (a) and (b) invert together with the sign inversion of the emission angles (θ_e), indicating that these spin polarizations obey time-inversion symmetry. The polarization orientations in Fig. 2 (b) are the same for one side of the emission angles. These spin-polarization is consistent with a typical, helical spin texture expected from the Rashba-type SOI. In contrast, the polarizations in Fig. 2 (a) inverts within the same parabola (e.g. see -5.4° and -11.4°). Based on our density-functional-theory (DFT) calculation, it would be because the fractional ratio of the orbital components changes depending on the $k_{||}$ positions; the p_z orbital of surface Bi is entangled with the opposite spin polarizations to the other orbitals. Figure 2 (c) shows spin-resolved EDCs for spin-polarization along $[\bar{1}10]$, parallel to $k_{||}$. One can find clear polarizations whose sign do not change with the sign of emission angle. Apparently, such behaviour breaks the time-inversion symmetry. Since this sample has no magnetic order, this spin polarization should not from the spin polarization of the initial states but from photoexcitation process, so called final-state effect.

In summary, we have found many unconventional spin textures on Bi/InAs(110), which cannot be understood by a simple helical spin texture expected for Rashba effect. Detailed analysis including DFT calculations and spin-dependent photoexcitation processes are ongoing.

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SPIN-POLARIZED QUASI-ONE-DIMENSIONAL METALLIC SURFACE ELECTRONIC STATE OF BI/GASB(110)-(2×1)

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Spin-polarized low-dimensional electronic structures due to Rashba effect and topological order at surfaces and interfaces are extensively studied for a fascinating playground for various spintronic phenomena [1-3]. Among them, one-dimensional (1D) or quasi-1D (Q1D) systems are expected to exhibit the interesting electronic behaviours such as the spin-dependent density-wave formation [4] and Majorana bound states at the junction with superconductors [5].

Recently, we have reported a semiconducting spin-polarized Q1D surface electronic structure of Bi/InAs(110)-(2×1) with a small bulk bandgap (~0.3 eV) [6]. As a next step for spintronic application and observation of spin-dependent electronic phenomena, to make this Q1D state metallic is desirable. In this work, we have tried to tune the Fermi-level (E_F) of the similar spin-polarized Q1D states on Bi/GaSb(110)-(2×1). The surface electronic structure of Bi/GaSb(110)-(2×1) was observed by laser-based angle-resolved photoelectron spectroscopy (ARPES) and spin-resolved ARPES (SARPES) at ISSP.

The GaSb(110) clean substrates were prepared by the repeated cycles of Ar sputtering (0.5 - 1 keV) and annealing up to 450 °C. After substrates cleaning, a few monolayers of Bi was evaporated. Subsequent annealing at 280 °C resulted in the sharp and low-background (2×1) low-energy electron diffraction patterns, suggesting the well-ordered surface atomic structure.

Figure 1 (a) is a Fermi contour of the Bi/GaSb(110)-(2×1) with the binding energy window of 0 ± 20 meV (E_F). As shown there, the metallic surface state is observed, forming a butterfly-like Fermi surface around the \bar{Y} point. Figure 1 (b) shows an ARPES intensity plot along $\bar{Y}-\bar{M}$. The hole-bands with Rashba-type paired parabolic dispersions were observed. The obtained band dispersion is quantitatively the same as the Q1D surface states on Bi/InAs(110)-(2×1) [5].

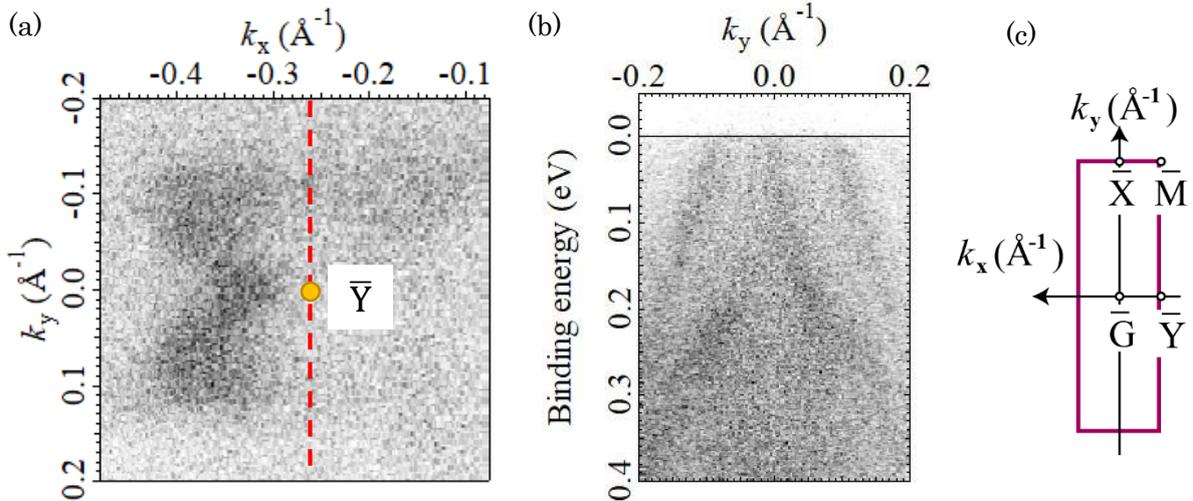


Figure 1. Electronic structure of Bi/GaSb(110)-(2×1) surface measured by ARPES at 14 K. (a) A constant-energy contour at a binding energy of 0 ± 20 meV. (b) Band dispersion along $\bar{Y}-\bar{M}$ ($k_y = 0.26 \text{ \AA}^{-1}$). (c) the surface Brillouin zone of Bi/GaSb(110)-(2×1).

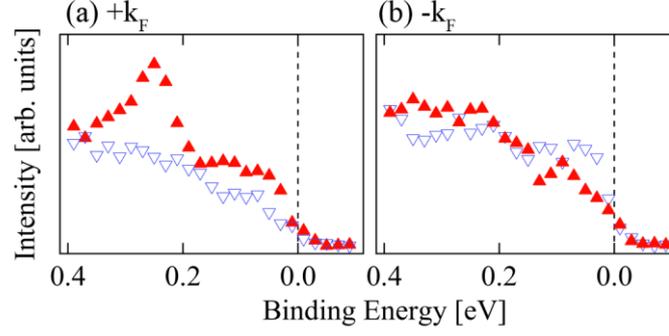


Figure 2. Spin- resolved energy distribution curves along $\bar{Y}-\bar{M}$ at 14 K. (a) $+k_F$ ($k_x = 0.10 \text{ \AA}^{-1}$). (b) $-k_F$ ($k_x = -0.10 \text{ \AA}^{-1}$). The filled (open) triangles correspond to spin polarization parallel (anti-parallel) to [001].

It would be due to the qualitatively common surface atomic structures of Bi/GaSb(110)-(2 × 1) and Bi/InAs(110)-(2 × 1), the “missing – row” model with the zig-zag Bi chains [7, 8].

Figure 2 shows the spin-resolved energy distribution curves (EDCs) at the Fermi wavevector (k_F) along $\bar{Y}-\bar{M}$ taken by SARPES. The spin-polarized peaks in Fig. 2 were clearly observed around E_F . These peak positions are consistent with the band dispersions in Fig. 1 (b). The spin polarization of these peaks inverts together with the signs of the wavenumbers, indicating the time-reversal symmetry, consistent with the Rashba-type spin-splitting.

These results suggest that Bi/GaSb(110)-(2 × 1) surface is the good Q1D candidate for following researches of spin-dependent phenomena, hosting a metallic and widely spin split surface states in Q1D.

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MODIFICATION OF SURFACE STATE OF TOPOLOGICAL INSULATOR Bi_2Se_3 BY ADSORPTION OF ORGANIC DONOR MOLECULE TTN

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Topological Insulators (TI), whose surface states provide various exotic quantum phenomena, attract much attention in quantum matter physics. TI surface states (TSS) have a helical-spin texture and in which spin-locked electrons are topologically protected from back scattering due to the time reversal symmetry attributed by strong spin-orbital coupling. The spintronic devices taking advantage of TI features are now expected to be realized. Thus, controlling the TSS by creating interfaces between TI and other materials is an important problem for applying TIs for spintronics devices. Recently, many researchers have worked on the TI interface with another material [1]. However, few previous researches related to the TI interface with organic materials have been reported [2].

In our research, we aimed to control the TSS of $\text{Bi}_2\text{Se}_3(111)$ surface, where a single Dirac Cone is located at the $\bar{\Gamma}$ point in SBZ, by adsorption of organic donor molecule TTN (Tetrathianaphthacene) onto the TI surface. To this end, we performed ARPES measurement by using angle-resolved photoelectron spectroscopy (ARPES) machine at the Laser and Synchrotron Research Center in the ISSP and studied the thickness dependence of TSS. The experiment was carried out at 140K using s-polarized VUV laser ($h\nu = 6.994$ eV) under ultrahigh vacuum conditions ($\sim 1 \times 10^{-8}$ Pa). The TTN molecule was deposited using Knudsen cell evaporator. The thickness of the molecule layer was estimated by using a quartz-crystal microbalance. The deposition process was performed in situ and at room temperature.

The experimental results are shown in Figure 1. The Bi_2Se_3 single crystal synthesized by Yaguchi group in Tokyo University of Science shows clear Dirac Cone dispersion on its pristine (111) surface, where DP is located at $E - E_F = -0.27$ eV. After TTN deposition of $\sim 5\text{\AA}$, the TSS electronic structure changes dramatically. The DP shifts to higher binding energy ($E - E_F = -0.39$ eV) and the Fermi surface area increases as the circular contour shape turns into the hexagonal warped one [3,4]. At the same time, the bulk conduction band (BCB) state appears inside the TSS with the clear band bottom edge. It appears that n-type doping due to the adsorption of TTN increases the density of surface electrons and consequently leads Fermi level to move into the BCB (away from the DP). Moreover, it is speculated that the surface electrostatic potential is perturbed by n-type doping, which causes triangle-like downward band-bending of the bulk electronic bands close to the surface. We interpret the clear BCB bottom edge as an intensity of the electrons weakly localized near the surface.

At further TTN deposition of 10\AA , the DP shifts to higher binding energy ($E - E_F \approx -0.55$ eV) and Fermi surface area gets larger, which means the TSS is further n-doped by TTN deposition. Besides, we observe the parabolic dispersion attributed to the BCB inside the TSS. This indicates that the relatively strong downward band-bending occurs at the TTN/ Bi_2Se_3 interface where the electrons are accumulated in the potential well forming a two-dimensional electron gas (2DEG) [5,6]. The flat feature lying in the lower energy region is attributed to the HOMO state of TTN. For the TTN deposition of 20\AA , the electronic structure is virtually unchanged compared to 10\AA deposition. It is assumed that the TTN molecular layer achieved 1ML under the 10\AA thickness.

We need to perform other experiments to verify whether the charge transfer from TTN to Bi_2Se_3 surface occurs or not and establish the adsorption structure.

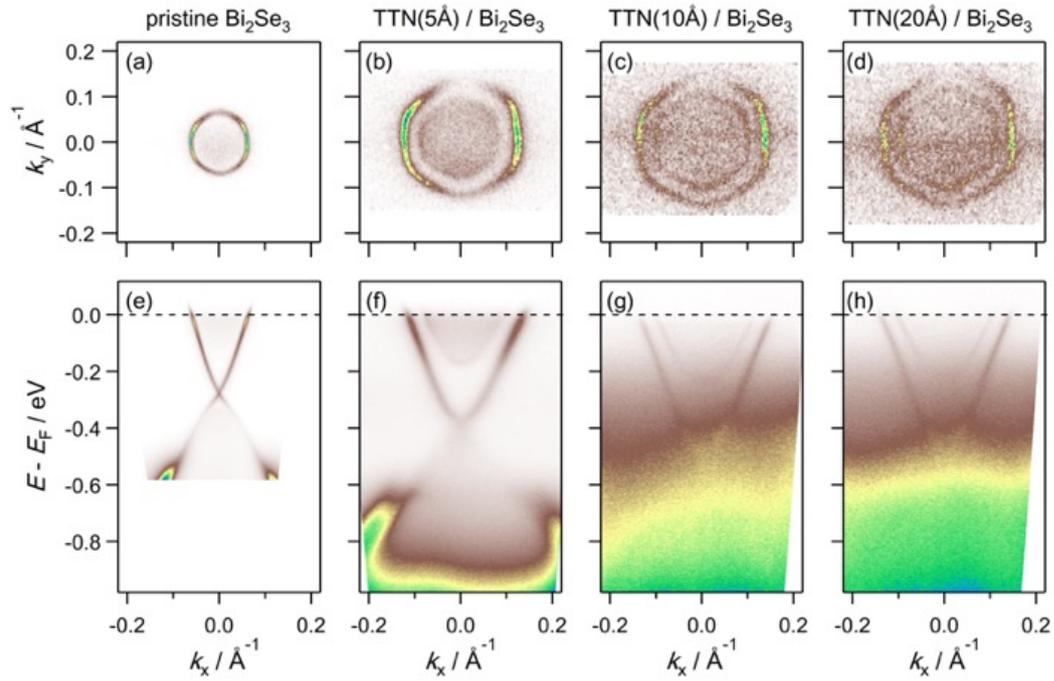


Figure 1. Measured ARPES spectra and Fermi surface maps for (a and e) pristine Bi_2Se_3 , (b and f) $\text{TTN}(5\text{\AA})/\text{Bi}_2\text{Se}_3$, (c and g) $\text{TTN}(10\text{\AA})/\text{Bi}_2\text{Se}_3$, (d and h) $\text{TTN}(20\text{\AA})/\text{Bi}_2\text{Se}_3$ at 140K.

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INVESTIGATION OF TOPOLOGICAL PROPERTIES IN SUPERCONDUCTING TASE₃

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Topological insulators (TIs), which are characterized by the fully gapped bulk states but gapless surface states [1], have attracted a lot of attentions in recent years. Moreover, the proposed topological superconductors [2], in which the Majorana states emerge within the superconducting gap, further boost the progress in this area owing to the potential applications in topological quantum computation. The attractive charge density waves (CDW) in transition-metal trichalcogenides MX₃ (M: Nb, Ta; X: S, Se) with layered quasi-1D chain-like structures [Fig. 1 (a)] have been extensively discussed in the early ages [3]. As one exception, TaSe₃ is appealing in that superconductivity emerges at ultra-low temperatures [4], differently from other analogues which undergo multiple CDW transitions [3]. It is considered that the anomaly stems from the poor nesting conditions in TaSe₃ due to more 3D electronic structures and competing CDW can be induced by Cu doping or in mesowires which reduce the dimensionality, as shown in Fig. 1 (b) [4-6]. Nevertheless, direct demonstrations of the electronic structures are still deficient. More interestingly, the recent first-principle calculations indicate that TaSe₃ actually belongs to a strong TI phase and moreover predict intriguing topological phase transitions among weak TI, strong TI, Dirac semimetal, and normal insulator phases through uniaxial strains, as shown in Fig. 1 (c) [7]. TaSe₃ thus not only offers a simple system to study the interplay between superconductivity and topological surface states (TSSs), but also provides an ideal platform for investigating topological phase transitions. A direct probing of the electronic structures via angle-resolved photoemission spectroscopy (ARPES) is consequently needed to elucidate the novel topological properties. So far, to our knowledge, there is only one direct measurement of the electronic structure of TaSe₃ by ARPES presenting blurred and inconclusive dispersions due to weak intensity and modest energy resolution [8].

In order to clearly reveal the electronic structure of TaSe₃ and confirm its topological properties [7], we have performed high-resolution laser spin-resolved ARPES measurements at the Institute for Solid State Physics, the University of Tokyo [9].

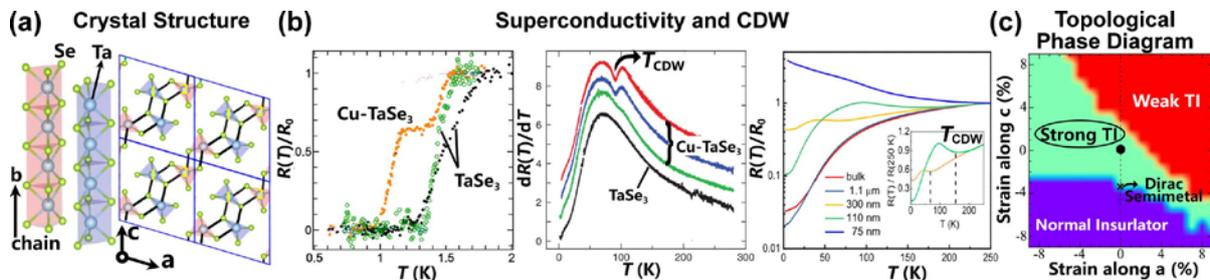


Fig. 1 (a) Crystal structure and unit cell of TaSe₃ [7]. (b) Superconductivity in pure and Cu-doped TaSe₃ (left) [4], CDW signature in Cu-doped TaSe₃ (middle) [5], and direct observation of CDW in mesowire TaSe₃ (right) [6]. (c) Strain-induced topological phase diagram of TaSe₃ including strong topological insulator (TI), weak TI, Dirac semimetal, and normal insulator phases [7].

For comparison with the calculation [7], Figure 2 (a) shows the 3D Brillouin zone and theoretical Fermi surface of TaSe₃, followed by the calculated TSSs at Γ and X points on (-101) surface in Fig. 2 (b). Figure (c) is schematic of the experimental ARPES setup [9] and typical sample size. In Fig. 2 (d) – (f), we show the observed Fermi surface and dispersions at Γ and X points on (-101) surface obtained using 7 eV laser at 10 K. Obviously, the observed band structure is in good agreement with the calculations shown in Fig. 2 (b) [7]. Figure 2 (g) shows one typical spin-resolved result at the cut on the sharp low-energy excitations indicated in Fig. 2 (f) by the dash line. One could clearly resolve the distinct signals from spin up and down states. Spin polarization [9] up to 30% has been detected in the low-energy excitations, which are possibly the TSSs.

In conclusion, we have performed laser spin-ARPES measurements on TaSe₃, the clearly resolved low-energy electronic structure qualitatively agrees with the calculated TSSs. The observation of spin polarization further implies that the sharp low-energy excitations are probably TSSs.

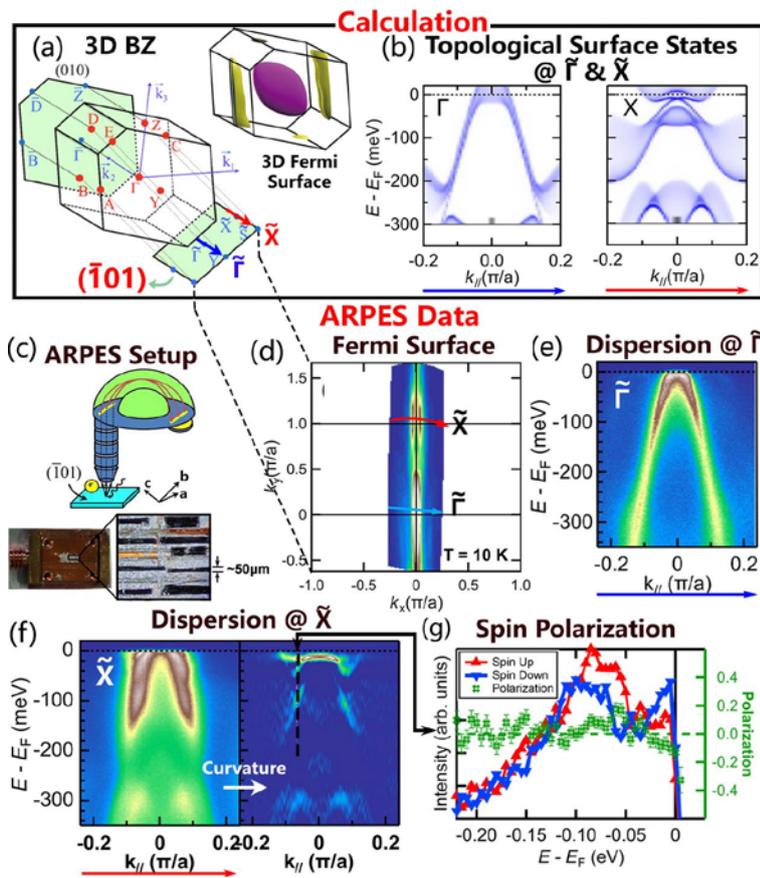


Fig. 2 (a) 3D Brillouin zone and calculated 3D Fermi surface of TaSe₃ [7]. (b) Calculated topological surface states at Γ and X points on (-101) surface [7]. Momentum cut positions are indicated in (a) with blue and red arrows, respectively. (c) ARPES setup. Typical samples size is $\sim 1 \times 0.1 \times 0.01$ mm. (d) Observed Fermi surface on (-101) surface at 10 K. Γ and X points are indicated by blue and red arrows, respectively. (e) Corresponding dispersion at Γ point. (f) Dispersion at X point and its curvature plot enhancing intensity maxima [10]. (g) Spin polarization along X direction at the cut indicated in (f) by a dash line. Red and blue curves are EDCs of spin up and down states, respectively. Up to 30 % spin polarization is detected

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Observation of the interface state for Bi layer deposited on a MnGa thin film

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Spin-orbit torque switching using the spin Hall effect has attracted much attention due to its potential application for ultra-high speed magnetoresistive random-access memories. Recently, spin-orbit torque switching using topological insulator $\text{Bi}_{1-x}\text{Sb}_x$ (BiSb) ($x = 0.1$) has been successfully developed [1]. Here, the BiSb(012) layer as a spin current source is grown on perpendicular ferromagnet $\text{Mn}_{0.6}\text{Ga}_{0.4}$ (MnGa) by molecular beam epitaxy (MBE) method. Since the topological state on the BiSb(012) surface has colossal spin Hall angle and high electric conductivity [1], the spin Hall conductivity of BiSb(012) is higher than that of conventional heavy metals [2-4] and other topological insulators [5]. Actually, the BiSb/MnGa heterostructure has demonstrated magnetization reversal via spin-orbit torque with extra-low current density that is one to two orders of magnitude lower than the current for conventional magnetization reversal. From comparison of spin Hall angles between the BiSb(001) and (012) surfaces, the number of Dirac cones (DCs) crossing the Fermi level (E_F), which is considered to contribute to the large spin Hall effect, in the (012) surface is larger than that in the (001) surface [6].

To understand the origin of the colossal spin Hall angle in topological insulators and ferromagnets, the band structure including the topological surface state of the BiSb/MnGa heterostructure has to be investigated. Therefore, we have performed spin- and angle-resolved photoemission spectroscopy (SARPES) measurements on a Bi/MnGa heterostructure.

A MnGa thin film was grown on a GaAs(001) substrate by the MBE method. The film was capped by amorphous Se layer to protect the surface from oxidization. Details of the film growth are described elsewhere [1]. The film was annealed at 550 °C under ultra-high vacuum better than 10^{-7} Pa to remove the Se capping layer. Bi layer was deposited on the clean MnGa surface at 150 °C with the thickness of about 1 bilayer. Clean surfaces of the thin films before and after Bi deposition were confirmed by low-energy electron-diffraction (LEED) measurements. ARPES with He-I (21.2 eV) and He-II (40.8 eV) light sources and 7eV-laser-based spin- and angle-resolved photoemission spectroscopy (Laser-SARPES) measurements were conducted with the ARPES instrument using DA-30 photoemission analyzer equipped with VLEED spin detector (Scienta Omicron, Inc.) at ISSP, University of Tokyo [7]. The measurements were performed at 30 K in a base pressure below 10^{-9} Pa. The polarization of the laser was linear-horizontal.

Figure 1(a) shows an angle-integrated photoemission spectrum of the Bi/MnGa thin film measured with the He-II source. The core-level peaks of Bi 5*d* (binding energy $E_B \sim 25$ eV) from the deposited Bi layer and Ga 3*d* ($E_B \sim 18$ eV) from the MnGa underlayer are observed. Since the thickness of the Bi layer is not large, photoemission signals from the MnGa layer are also detected. Figure 1(b) shows ARPES spectra along the Γ -X symmetry line of the Bi(012)/MnGa thin film taken with the He-I light source. The concave band around $E_B = 0.1 \sim 0.5$ eV centered at the Γ point originates from the MnGa underlayer. Since the hole-like band crossing E_F near the Γ point appears after the Bi deposition, this band near the Γ point comes from the Bi layer.

The hole-like band near the Γ point has been investigated using Laser-SARPES. Figure 2(a) shows the ARPES spectrum along the Γ -X line taken with the 7-eV laser. While the band

dispersion is unclear with the He-I source, the sharp dispersion can be seen near the Γ point in the Laser-ARPES spectrum due to the higher energy and momentum resolution. The dispersion of this band looks linear, as shown in Fig. 2(b). The SARPES measurements have revealed that the sharp band is spin-polarized along the direction perpendicular to the Γ -X line and the spin polarization is flipped with the momentum direction, as shown in Fig. 2(c). Based on the observations, the band near the Γ point is expected to be the DC state of the deposited Bi layer. Therefore, we have succeeded in observing the topological interface state between the Bi and MnGa layers using Laser-SARPES. Further electronic structure study such as the observations of the entire band structure and DC states near the Brillouin zone boundary will shed light on the origin of the large spin Hall angle of the interface between the topological insulator/ferromagnet heterostructures.

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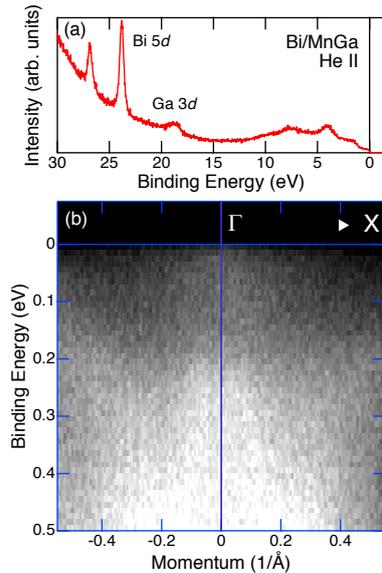


Fig.1. Photoemission spectra of Bi/MnGa thin film. (a) Angle-integrated spectrum taken with the He-II source. (b) ARPES spectrum along the Γ -X line taken with the He-I source.

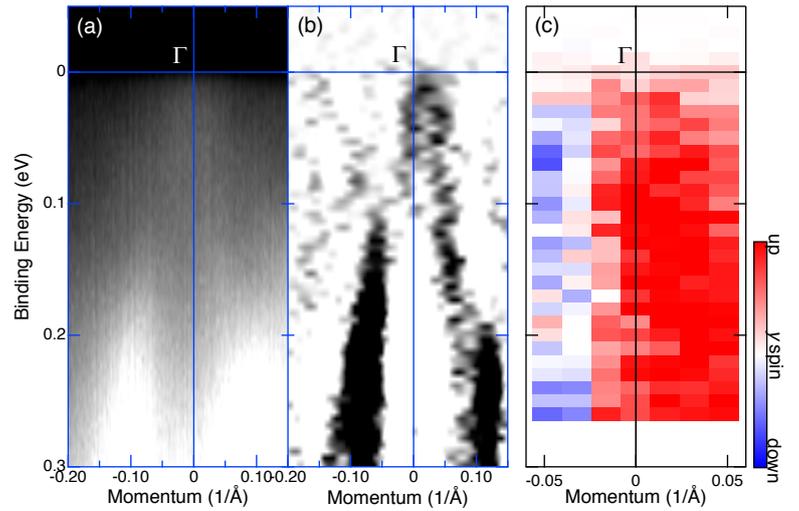


Fig.2. Laser-SARPES spectra of Bi/MnGa thin film. (a) ARPES spectrum along the Γ -X line taken with the 7-eV laser. (b) 2nd derivative ARPES spectrum. The derivation has been performed along the momentum direction. (c) SARPES spectrum. The spin direction is perpendicular to the Γ -X direction (y spin).

SWITCHABLE WEAK TOPOLOGICAL INSULATOR STATE IN QUASI-ONE-DIMENSIONAL BISMUTH IODIDE

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A topological insulator (TI) is characterized by an insulating bulk with conductive edges determined by Z_2 topological invariants. In three dimensions, a TI is classified as either a strong TI (STI) or a weak TI (WTI) [1], and the STI phase was experimentally confirmed rapidly after the theoretical predictions through the observation of topological surface states (TSS) by angle-resolved photoemission spectroscopy (ARPES) [2]. In contrast, the WTI phase has so far eluded experimental verification, since TSSs emerge only on side surfaces, which are typically not cleavable and undetectable in real 3D crystals. In this work, we have investigated the WTI state in β -Bi₄I₄ with naturally cleavable top and side planes, which is necessary for the experimental confirmation of the WTI state.

β -Bi₄I₄ is predicted to be in proximity of three different topological phase by theoretical calculations: a STI (1;110) a WTI (0;001) or a normal insulator (0;000) phase is realized sensitively depending on the calculation parameters [3, 4]. By using laser-SARPES, we have demonstrated that a spin-polarized quasi-1D TSS exist in the bulk band gap of β -Bi₄I₄, indicating the WTI phase in the real crystal. In contrast, α -Bi₄I₄ is predicted to be topologically trivial and no TSS has been detected by ARPES in this compound. From these results, we expect that the phase transition between the WTI phase and the normal insulator phase occurs following the structural transition between the β -phase and the α -phase. However, one may wonder (i) why the observed Fermi surface shown in fig. 1(b) is quite anisotropic along k_y and (ii) whether the topological phase transition really happens in single crystal piece. Here, we discuss these points by using laser-(S)ARPES [5].

First, we investigated the photoemission intensity distribution of the topological surface states on the side surface by conducting light-polarization dependent ARPES. The experimental geometry for the ARPES measurements is shown in Fig. 1(a), where θ is the measurement angle for each ARPES image. With p -polarized light, the photoemission intensity distribution is clearly anisotropic both at the $\bar{\Gamma}$ point and the \bar{Z} point of the (100) surface Brillouin zone as shown in Fig. 1(c) and 1(e), where one branch of the Dirac dispersion ($-k_F$) is clearly observed while the photoemission intensity of the other part ($+k_F$) is almost vanishing. In contrast, with s -polarized light, the photoemission intensities from both branches are almost the same as shown in Fig. 1(d) and 1(f). These results can be explained by the interference of photoemission

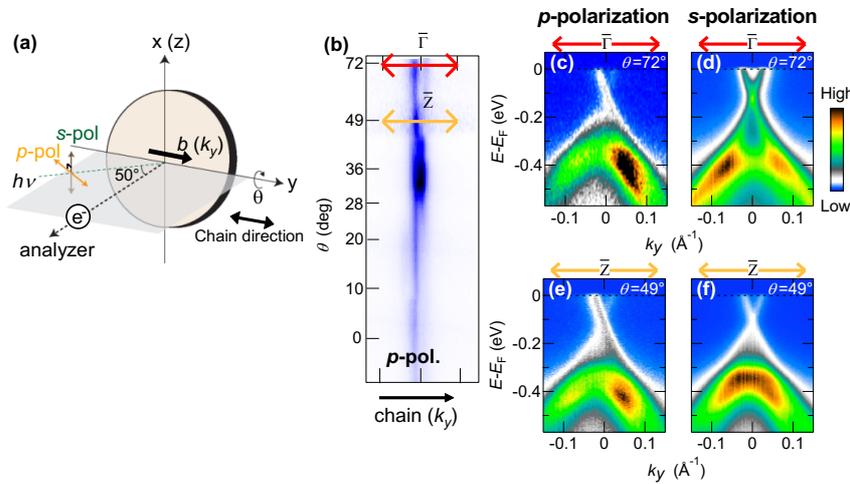


Fig. 1 (a) Experimental geometry of laser-ARPES. (b) The Fermi surface mapping obtained with p -polarized light. (c, d) ARPES band maps across the $\bar{\Gamma}$ point of the (100) surface taken with p -polarized light and s -polarized light, respectively. (e, f) Same as (c) and (d), but for the momentum cut across \bar{Z} point of the (100) surface.

processes. Since the p -pol. light has both of in-plane and out-of-plane components [Fig. 1(a)], it excites electrons with different orbitals at the same time if the TSS is composed of multiple orbitals. Generally, TSS consists in Bi-based topological insulators of Bi $6p_{x,y,z}$ orbitals due to the strong spin-orbit coupling of bismuth [6], and hence the observed photoemission intensity anisotropy is attributed to the result from the interference of photoelectrons from Bi $6p_y$ (in-plane) and $6p_x$ (out-of-plane) orbitals excited by p -pol. light. In the case of ARPES with s -pol. light, the interference effect becomes smaller because s -pol. light has smaller out-of-plane component and excites only Bi $6p_x$ orbital.

Second, we show that the topological phase transition between the WTI phase and normal insulator phase follows the structural phase transition between β - and α -Bi₄I₄, and thus the topological phase is switchable in this material. The procedure for demonstrating the phase transition in single piece of Bi₄I₄ crystal is shown in Fig. 2(a) and 2(d). To unambiguously investigate the effect of the structural transition, the same crystal pieces have been cleaved twice after mild cooling and after rapid cooling. As shown in the ARPES images, quasi-1D TSS observed after rapid cooling [Fig. 2(b)] disappeared after mild cooling [Fig. 2(c)], showing the phase transition from the WTI phase to the normal insulator phase. In addition, the band gap observed after mild cooling [Fig. 2(e)] was gapped with the appearance of TSS after quenching [Fig. 2(f)], reflecting the phase transition from the normal insulator phase to the WTI phase. From these results, it is concluded that the topological phase transition in fact follows the structural phase transition and hence the topological phase in Bi₄I₄ can be controlled by temperature.

In conclusion, these results support the switchable WTI phase in Bi₄I₄ observed in our previous measurements. Since the bulk of Bi₄I₄ is insulating, the TSSs can be probed not only by ARPES but also by transport measurements. Importantly, the capability of controlling the topological phase using structural phase transition is beneficial to making devices such as spin-transistor. Our work will thus stimulate further in-depth study on exotic properties of the WTI state and put the foundation for future WTI-based spintronic applications.

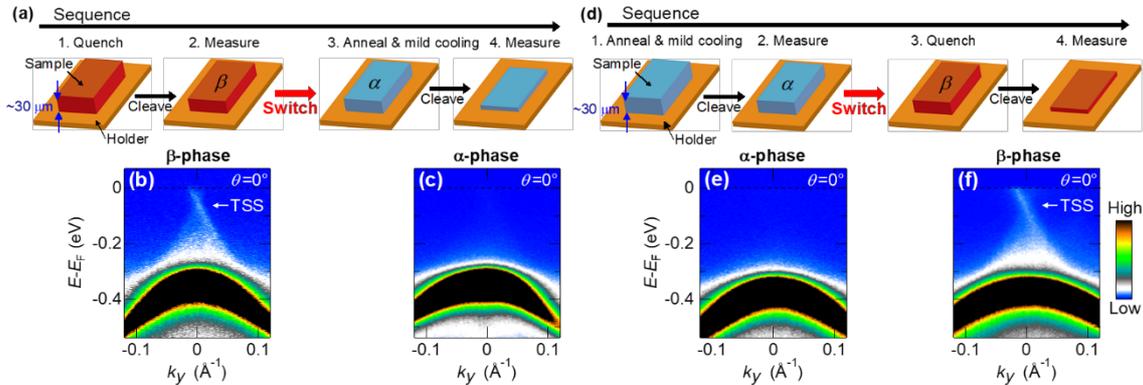


Fig. 2 (a) Experimental sequence to demonstrate the topological phase transition following the structural phase transition from the β -phase to the α -phase using the same crystal piece. (b, c) ARPES images at $\theta=0^\circ$ after quenching (b) and after mild cooling (c) for the same crystal piece. (d) Same as (a), but from the α -phase to the β -phase. (e, f) ARPES images at $\theta=0^\circ$ after mild cooling (e) and after quenching (f) for the same crystal piece.

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Electronic and spin structure of heavy element thallium single crystal thin films

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Introduction

In solid bulk, the two bands with opposite spins are usually degenerated due to the presence of both the time-reversal and space-inversion symmetries. However, in case of surface or interface, where the space-inversion symmetry is broken, the spin degeneracy is lifted by the spin-orbit coupling (SOC). This is called the Bychkov-Rashba effect or simply the Rashba effect [1]. In the early studies, the observation of giant Rashba effects on surface alloys formed by the adsorption of 1/3 ML of high-Z elements Bi or Pb on the Ag(111) surface [2,3] has attracted a lot of interest. The spin-split was discussed based on a simple Rashba effect in these studies. More recently, several peculiar Rashba spins have been observed, and their origins were discussed based on e.g., the symmetry of the surface [4]. Tl atomic layers formed on Si surfaces are one of such exotic systems. Together with this interesting spin-related phenomenon, Tl is also known to exhibit superconductivity at below 2.4 K in crystal phase [5]. However, due to the difficulty to create a single Tl crystal that is large enough to perform photoemission measurements, there is no experimental report on its band structure so far. Moreover, since a Tl single crystal film would show spin-split bands and thus has the possibility of becoming a topological superconductor [6], we planned to create a Tl single crystal film and measure its band structure.

Experimental details

The Ag(111) surface was first sputtered with Ar⁺ with an accelerating voltage of 500 V in high vacuum (5×10^{-3} Pa) to obtain a clean surface. The quality of the Ag(111) surface was confirmed by the observation of sharp LEED spots and a clear surface state. The Tl single crystal film was then prepared by the deposition of 2 ML of Tl on the Ag(111) clean surface. The LEED pattern of the Tl/Ag(111) surface is shown in Fig. 1. Clear and sharp spots originating from the Tl double-layer single crystal are observed. ARPES and SARPES were performed using the laser-SARPES machine developed at ISSP [7]. All PES measurement was carried out at a sample temperature of 35 K under ultra-high vacuum (ca. 1×10^{-8} Pa).

Results and discussion

As shown in Fig. 1, Tl double-layer single crystal thin film has two domains that are rotated $\pm 4^\circ$ from the symmetric axis of Ag(111). This makes the Fermi surface and band structure complicated as shown in Figs. 2 (a) and (b). By considering the number of surface states observed in the band dispersion along the $\bar{\Gamma} - \bar{K}_{\text{Ag}}$ direction, we found that the Tl single crystal film has two metallic bands (as shown in Fig. 2 (a) the bands of Tl are degenerated in the $\bar{\Gamma} - \bar{K}_{\text{Ag}}$ direction). Of the two metallic bands, the one with lower binding energy at the \bar{K} point of the Tl Brillouin zone shows a clear split in the energy direction. In order to investigate the origin of this splitting, we performed ARPES measurement, and found that the split at the \bar{K} point is a spin split, and the spin-polarization vector points along the wave vector directions. This means that the origin of this spin-split is not an ordinary Rashba effect, and we are now tackling this problem by considering other possible origins.

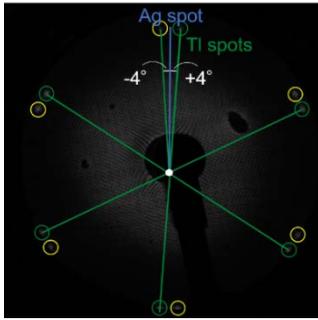


Fig. 1 LEED pattern of Tl/Ag(111) obtained with an electron energy of 85 eV.

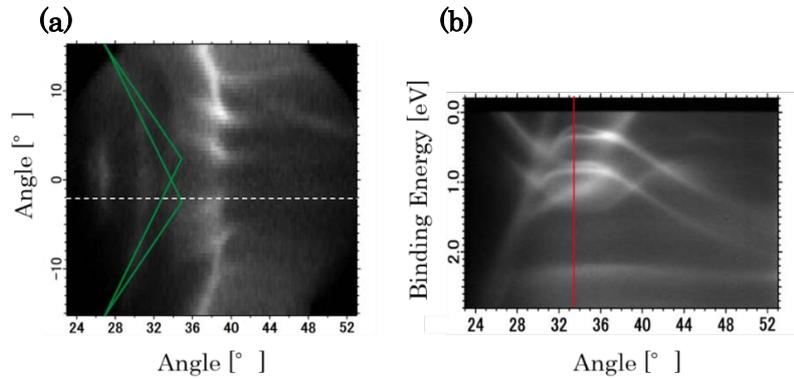


Fig. 2 (a) Fermi surface around the \bar{K} point of the Brillouin zone of Tl. Green lines indicate the Brillouin zones of Tl from the two domains. (b) Band structure cut at the white dashed line in (a).

Summary

We obtained a Tl double-layer single crystal film on a Ag(111) surface that is large enough to measure its band structure. The presence of two metallic bands and a split of one of them at the \bar{K} point of the Tl Brillouin zone was confirmed by ARPES. This split is a spin-split, and the spin-polarization vector points along the wave vector directions.

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SPIN-POLARIZED BANDS OF AN IN BYLAYER FILM FORMED ON Si(111)

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Introduction

In two-dimensional (2D) systems such as surfaces or heterointerfaces, the electron bands are split due to spin-orbit coupling and the space inversion asymmetry along the direction perpendicular to the plane. This effect is called the Rashba effect [1], which leads the spin-polarization vectors to be parallel to the 2D plane and perpendicular to the wave vector in an ideal 2D system. This Rashba effect was observed on the surface of heavy elements [2], heavy element atomic layers formed on solid surfaces [3] and so on. One of the main motivations of these early studies was to create a system with large Rashba splitting that can be used as a material for spintronics devices. In this project, motivated by the theoretical work reported by Sau *et al.* [4], we focus on the spin-polarized band induced in such atomic layers that has potential to become topological superconductor. The system that we studied is the In/Si (111)-($\sqrt{7} \times \sqrt{3}$) surface, a system that is formed by the adsorption of 2.4 ML of In on top of Si(111) [5] and shows superconductivity below ca. 3 K [6].

Experimental details

Angle-resolved photoelectron spectroscopy (ARPES) and spin-resolved ARPES (SARPES) measurements were performed with the laser-SARPES machine developed at ISSP [7]. A laser light source with a photon energy ($h\nu$) of 6.994 eV was used as the excitation light, and a Scienta Omicron DA30-L analyzer equipped with a VLEED spin detector was used for PES measurements. An n-type doped Si(111) wafer with a resistivity of $< 0.01 \Omega \cdot \text{cm}$ was first annealed at 1520 K to obtain a clean Si(111)-(7×7) surface. Following the adsorption of slightly more than 2.4 ML of In, the sample was annealed at 570 K to obtain the In/Si (111)-($\sqrt{7} \times \sqrt{3}$) surface. The quality of the sample and the presence of a single ($\sqrt{7} \times \sqrt{3}$) domain was confirmed by low-energy electron diffraction (LEED) (Fig. 1) and the observation of clear surface states. All PES measurement was carried out at a sample temperature of 35 K under ultra-high vacuum (ca. 1×10^{-8} Pa).

Results and discussion

By using ARPES, we observed a circular Fermi surface together with a butterfly shape one. Although these Fermi surfaces shows good agreement with those reported previously on In/Si (111)-($\sqrt{7} \times \sqrt{3}$) [8,9], thanks to the high k -resolution of the present setup ($\Delta k = 1.4 \times 10^{-3} \text{ \AA}^{-1}$), we could observed a clear split as shown in Fig. 2 (note that not only the circular Fermi surface shown in Fig. 2 but also the butterfly shape one shows a clear split). Since this split might originate from the Rashba effect, we have performed SARPES measurements to obtain information about the spin polarization of the split Fermi surfaces. Of the two Fermi surfaces, the spin-polarization vector of the circular one was parallel to the surface and perpendicular to the wave vector. This result indicates that the spin-split of the circular Fermi surface might originate from the Rashba effect. In contrast to the circular Fermi surface, the spin-polarization

vector of the butterfly shape Fermi surface was completely different, and cannot be explained by the Rashba effect. The spin-polarization vector of the butterfly shape Fermi surface was parallel to the surface, and also parallel to the wave vector. Since the peculiar spin-polarization vector of the butterfly shape Fermi surface was observed at k point without any symmetry (a k point with symmetry means a point like the Γ point of the Brillouin zone), this cannot be a peculiar spin originated from the symmetry of the surface like those observed in our former studies [10] as well. We are now tackling this problem by considering other possible origins, and hope to get the answer soon.

Summary

By using a high-energy and high- k resolution SARPES system, we succeeded to observe the spin splits Fermi surfaces of In/Si (111)-($\sqrt{7} \times \sqrt{3}$), a system that becomes superconductor at below 3 K. The spin-polarization vector of one of the observed Fermi surfaces cannot be explain by neither a simple Rashba effect nor a symmetry induced peculiar Rashba effect.

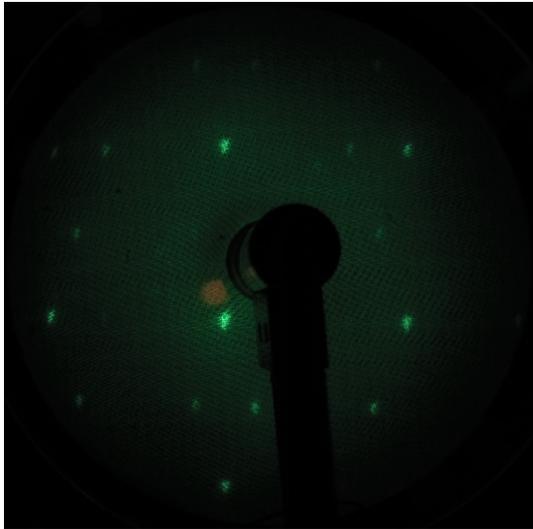


Fig. 1. LEED image of the In/Si (111)-($\sqrt{7} \times \sqrt{3}$) surface obtained with an electron energy of 85 eV.

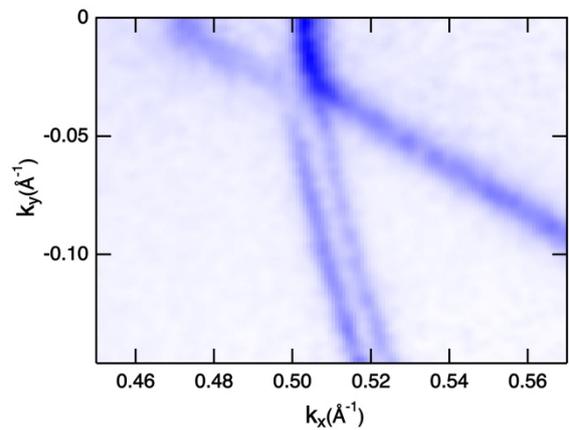


Fig. 2. Fermi surface of In/Si (111)-($\sqrt{7} \times \sqrt{3}$). The circular Fermi surface shows a clear split.

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MULTIPLE TOPOLOGICAL STATES IN IRON-BASED SUPERCONDUCTORS

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Topological materials and unconventional iron-based superconductors are both important areas of study but, to date, relatively little overlap has been identified between these two fields. However, the combination of topological bands and superconductivity promises the manifestation of exotic superconducting states, including Majorana fermions, the central component of topological quantum computation. Here, using high-resolution spin-resolved ARPES, we have identified both topological insulator (TI) and Dirac semimetal (TDS) states near the Fermi energy in different iron-based superconducting compounds, potentially allowing access to several different superconducting topological states in the same material. These results reveal the generic coexistence of superconductivity and multiple topological states in iron-based superconductors, indicating that this broad class of materials is a promising platform for high-temperature topological superconductivity.

The topological insulator and Dirac semimetal states in Li(Fe,Co)As.

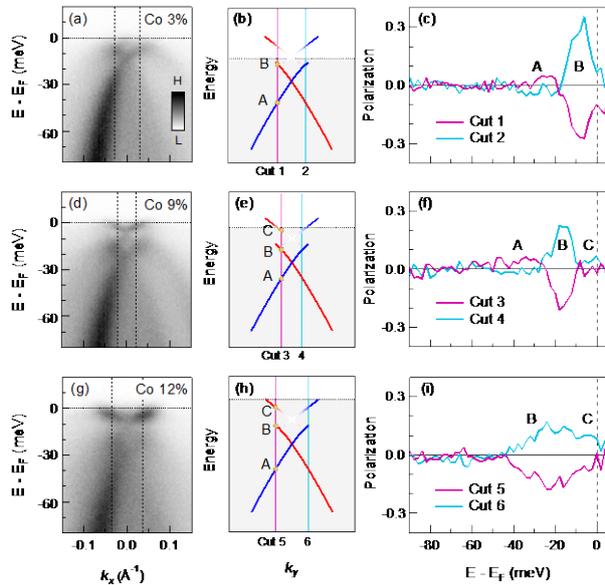


Figure 1 Spin polarization of the Dirac cones from TI and TDS states in Li(Fe_{1-x}Co_x)As. (a) Intensity plot along k_x for $x = 3\%$ sample with a 7-eV laser. (b) Sketch of the spin-polarized surface bands. The blue and red lines stand for opposite spin polarizations, as confirmed by the data in (c). (c) Spin polarization along x direction at Cut 1 and Cut 2, which are indicated by two vertical lines in (a) and (b). (d–f) Same as (a–c), but for samples with $x = 9\%$. (g–i) same as (a–c), but for samples with $x = 12\%$.

We studied a series of Li(Fe,Co)As samples with different Co content by using laser-based ARPES. On the $x = 3\%$ samples, only one Dirac band was observed below Fermi level (Fig. 1(a)), similar to the case of Fe(Te,Se) [1,2]. By doping more electrons, we further observed another Dirac cone in the $x = 9\%$ and 12% samples, as shown in Fig. 1(d,g), which is above Fermi level in the parent compound. The two Dirac cones are consistent with the DFT

calculations: the upper cone comes from the the surface component of the TDS states, while the lower cone comes from the TI surface states. Because of their surface nature, both Dirac cones should be spin-polarized, and the calculated spin patterns are sketched in Fig. 1(b,e,h). We further carried out spin-resolved measurements to check their surface nature. A pair of spin-polarized energy distribution curves along k_y was measured, as indicated in Fig. 1(b,e,h). For both $x = 3\%$ and 9% samples, the spin polarization of the TI Dirac cone is resolved (Fig. 1(c,f)), and consistent with theoretical calculations. For $x = 9\%$ and 12% samples, the spin polarization of the TDS surface cone is observed, as shown in Fig. 1(f,i). Due to the the weak intensity of the upper part of the TDS cone, only the spin polarization of the lower part is resolved.

The topological Dirac semimetal states in Fe(Te,Se).

The TI surface states in Fe(Te,Se) was observed previously in Ref. [2]. The TDS cone in Fe(Te,Se) cannot be resolved directly, because of the difficulty to dope this sample. However, we may obtain some evidences of the TDS bands in Fe(Te,Se) by measuring the associated surface states, as shown in Fig. 2(a). The spin-resolved intensity plots of Fe(Te,Se) are shown in Fig. 2(b). We measured four cuts with high resolution at high symmetry lines, as indicated in Fig. 2(c). Fig. 2(d-g) display the spin polarization curves for the four cuts. The magnitude of spin polarization of the band is about 50%, indicating coexistence of unpolarized bulk and polarized surface states. The results are consistent with the spin-resolved spectrum from calculations in Fig. 2(a), thus support the existence of the TDS states above E_F in Fe(Te,Se).

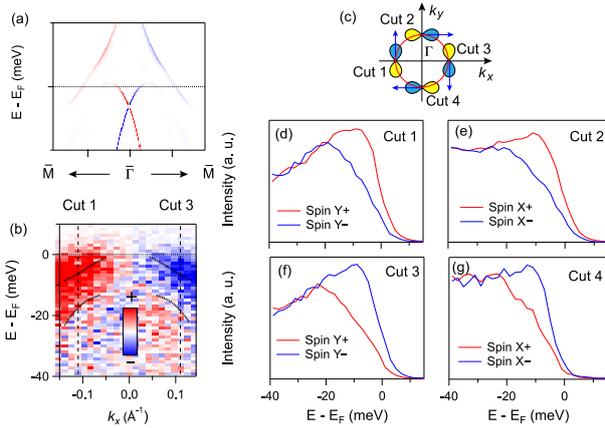


Figure 2 Spin polarization of the band at E_F and its Dirac semimetal origin in Fe(Te,Se).

(a) Spin-resolved (001) surface spectrum from a k - p model calculation. (b) Spin-resolved intensity plot at Γ with a 7-eV laser. (c) Sketch of the spin-orbit pattern from model calculations. (d-g) Spin-resolved energy distribution curves at Cuts 1–4. Because of the matrix element effect, Cuts 1,3 and 2,4 are taken with s -polarized and p -polarized photons, respectively.

Our findings of the TI and TDS states in Li(Fe,Co)As and Fe(Te,Se) prove the generic existence of different types of topological states in iron-based superconductors. Their simple structures, multiple topological states, and a tunable Fermi level make iron-based superconductors ideal platforms for the study of topological superconductivity, Majorana bound states, and as such, potentially, topological quantum computation.

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ELECTRONIC BAND STRUCTURE OF TRANSFER GRAPHENE ON SI(111) SUBSTRATE

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Electronic band structure of graphene, which has a linear dispersion around a K point, has attracted much attention not only because of fundamental interest but also possible applications of their electronic conduction. The epitaxial graphene has been experimentally made on substrates by several methods so far. For example, the graphene grown on Si-terminated surface of SiC(0001) has been well known and established. In contrast to the former studies, in the present study, we have fabricated the single-layer graphene on a Si(111) substrate by transfer method and have investigated the electronic band structure of the transfer graphene by angle-resolved photoemission spectroscopy (ARPES).

We used monolayer graphene film made for transfer that was commercially available. The Si(111) substrate was cut from a wafer. The transfer procedure was as follows: The sacrificial-layer/graphene was put in deionized water. The Si(111) substrate was introduced into the deionized water and scoop the sacrificial layer/graphene from below. After that the sacrificial-layer/graphene was deposited on the Si(111) substrate [Fig. 1(a)]. Then, the sacrificial layer/graphene/substrate was taken out from the water and let it dry 30 minutes in air. Subsequently, the sample is heated in an oven at 150°C for 1.5h. And then, the sample is stored in a vacuum desiccator for 24h to avoid detachment of the graphene from the substrate. The sacrificial layer is removed in acetone for 1h, and then the sample is put into ethanol for 1h. After this procedure, the sample was introduced into the UHV chamber. And then, the sample were annealed at 400C for 30 min. The surface quality was checked by low energy electron diffraction, where we observed a ring pattern due to the multi domain structure of the transfer graphene. Here, the average size of one domain is approximately several micrometers.

ARPES measurements were performed at the Institute for Solid State Physics, The University of Tokyo [1]. The photoelectrons were excited by 21.2-eV photons from a He lamp and were analyzed with a ScientaOmicron DA30-L analyzer. The sample temperature was set to 40K during the measurements.

Figure 1(b) shows the ARPES image of the transfer graphene on Si(111) around the K point. We find a Dirac-cone like band for which the spectral width is broadened due to the multi domain structure of the surface. The Dirac point is located at the binding energy of 0.2-0.3 eV. The doping level of the p-band is smaller than that of the graphene on SiC(0001) substrate [2]. In the present ARPES measurements, the spot size of our He lamp is 0.5 mm on the sample. We could observe a clear band image if we would use the excitation light with the spot size on the order of micrometers.

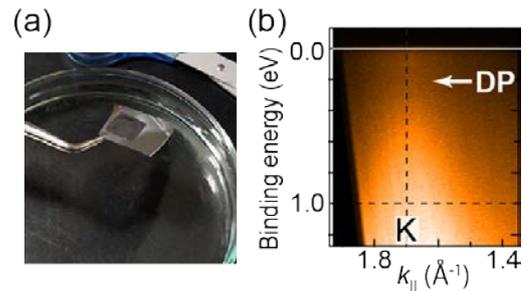


Fig. 1(a) Photo of the sacrificial-layer/graphene scooping on the Si(111) substrate. (b) ARPES image of the transfer graphene on Si(111).

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STUDY ON ELECTRONIC STRUCTURES IN HALF-METALLIC FERROMAGNET CoS_2 BY HIGH-RESOLUTION SPIN- AND ANGLE-RESOLVED PHOTOEMISSION SPECTROSCOPY

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CoS_2 is a ferromagnetic metal whose ordered moment is reported to be approximately $0.9 \mu_B/\text{Co}$ with an easy-axis along the $[111]$ direction [1–3]. The ordered moment is close to $1 \mu_B/\text{Co}$ in a low-spin state of Co^{2+} (configuration: $t_{2g}^6 e_g^1$), suggesting that CoS_2 has a half-metallic electronic structure including completely spin polarized e_g bands crossing the Fermi level (E_F). The half-metallicity has been also supported by theoretical band structure calculated within generalized gradient approximation (GGA) and experimental studies using de Haas–van Alphen (dHvA) effect [4,5]. On the other hand, the results of band calculation within local spin-density approximation (LSDA) and photoemission spectroscopy measurements indicated that CoS_2 has a “nearly” half-metallic electronic structure in which a bottom of the minority spin e_g band touches or slightly crosses E_F near the R point [6]. So far, comprehensive understanding of half-metallicity of CoS_2 has not been obtained. In this work, we performed high-resolution spin- and angle-resolved photoemission spectroscopy (SARPES) measurements on a high-quality CoS_2 single crystal in order to reveal the half-metallicity.

Single crystals of CoS_2 were synthesized by the vapor transport method,[3] and were used for SARPES measurements. Laser-based SARPES experiments were performed at the Institute for Solid State Physics, The University of Tokyo [7]. The p -polarized light with $h\nu = 6.994$ eV was used to excite the photoelectrons. Photoelectrons were analyzed with a combination of a ScientaOmicron DA30L analyzer and a very-low-energy-electron-diffraction (VLEED) type spin detector. The energy and angular resolutions were set to 6 meV and 0.3° (corresponding to ~ 0.005 Å) for spin-integrated ARPES and 10 meV and 1° (corresponding to ~ 0.02 Å) for SARPES, respectively. Calibration of E_F for the samples was achieved using a gold reference. The data were taken at $T = 40$ K. Clean surfaces for all measurements were obtained by *in situ* cleaving of the samples.

Our high-resolution SARPES results are summarized in Fig. 1. By the 7-eV laser, we observed bands located at -1 eV, -0.5 eV, -50 meV, near E_F , and a dispersive hole band. The bands located around -1 eV are assigned to t_{2g} bands based on a photoemission spectroscopy study [6].

For the e_g bands located between -0.5 eV and E_F , we obtained the spin-polarized band structure clearly. We named the e_g bands as shown in Figs. 1 (a) and (d). At -0.5 eV, the SARPES intensity map for the majority spin bands has a clear structure corresponding to the α band, although that for the minority spin bands has no structure at -0.5 eV. In the same way, it is identified that the β band has the majority spin character. The result that the majority spin character of the α and β bands is consistent with the theoretical band structures.[4] On the other hand, the SARPES intensity map for the minority spin bands has clear band dispersion corresponding to the γ and δ bands.

Here, we discuss the half-metallicity of CoS₂. We observed both of the majority and minority spin bands crossing E_F , as shown in Fig. 1, providing spectroscopic evidence for ‘normal’ ferromagnetic metallicity, not half-metallicity, of CoS₂. This result is consistent with claims by a photoemission studies.[6] The non-half-metallicity is also supported by the fact that the ordered moment $\mu_s \sim 0.9 \mu_B/\text{Co}$ at 2 K[3] is not exactly $\mu_s = 1 \mu_B/\text{Co}$ which is the expected ordered moment when the e_g electrons are completely spin-polarized.

In summary, we performed high-resolution SARPES in order to reveal the electronic structure of ferromagnetic CoS₂. We observed t_{2g} bands and highly spin-polarized e_g bands. We observed a minority spin band crossing E_F , demonstrating that CoS₂ is not a half-metallic ferromagnet at least above 40 K. This report demonstrates that the high-resolution SARPES is a powerful tool to identify half-metallicity of theoretically proposed borderline half-metallic ferromagnets.

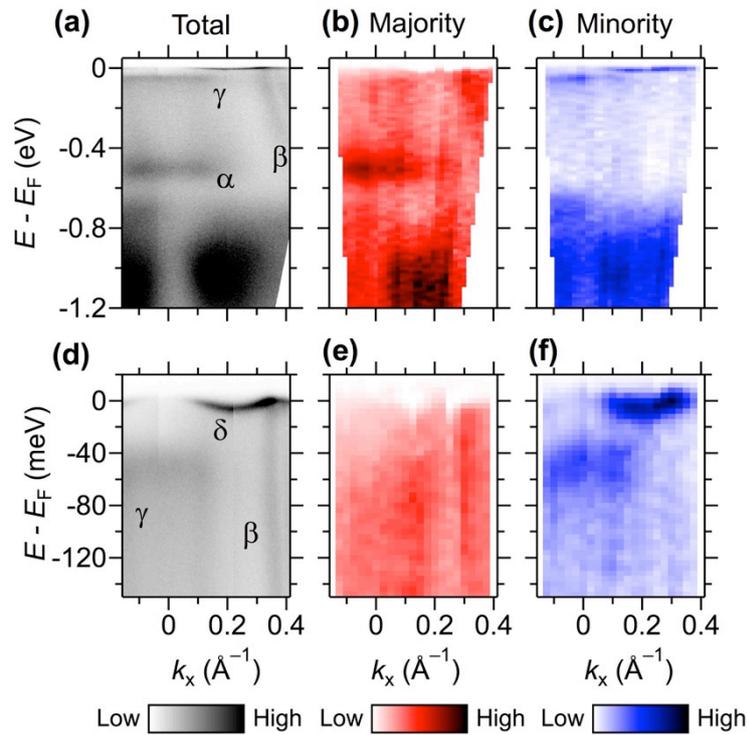


Fig. 1. (a)–(c) ARPES (a) and SARPES ((b) Majority spin bands; (c) Minority spin bands) intensity maps as a function of energy to E_F and k_x . (d)–(f) Same as (a)–(c) but measured in the vicinity of E_F .

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ANGLE-RESOLVED PHOTOEMISSION SPECTROSCOPY STUDY OF TYPE-II WEYL SEMIMETAL WTe₂

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Introduction

So far, no example of a Weyl fermion has been observed in particle physics research. But some topological materials can host electrons behaving as Weyl fermions, which are called Weyl semimetals. In Weyl semimetals, there are bands crossing linearly near Weyl points, which means that electrons near Weyl points have zero-mass [1-3].

In some Weyl semimetals, the hole- and electron-like pockets touch each other at several Weyl points, and these kinds of Weyl semimetals are called type-II Weyl semimetals. Weyl points are topologically protected, and topological surface states (Fermi arcs) connected a pair of Weyl points with opposite chirality. WTe₂ is a kind of quasi-2D type-II Weyl semimetal. The space inversion symmetry along *c*-axis is broken, and the top and bottom surfaces are different. According to theoretical calculation, the surface electronic states of the two surfaces are different [4]. However, so far there is no experimental research showing the differences between the two surface states. On the other hand, some of Weyl points in WTe₂ locate above the Fermi level (E_F), which cannot be observed by angle-resolved photoemission spectroscopy directly, and neither the Fermi arcs above E_F have been observed. On the other hand, it is difficult to say whether the band structure is topological surface state or not by ARPES. Spin-ARPES can provide more evidence.

Experiment

The single crystals of WTe₂ with large domains were grown via the iodine vapor transport method [4]. We cut one piece of WTe₂ sample into two halves and turn one of them upside down. We cleaved them *in situ*.

The experiment was carried out with the spin-resolved laser ARPES (SARPES) at ISSP, the University of Tokyo. The light source is 7 eV laser from KBBF crystal. We measured two kinds of surfaces of WTe₂: surface A and B. We used the 7 eV laser ARPES to map the energy contours and measured the band structures. To observe the topological Fermi arcs are surfaces states which connect hole and electron

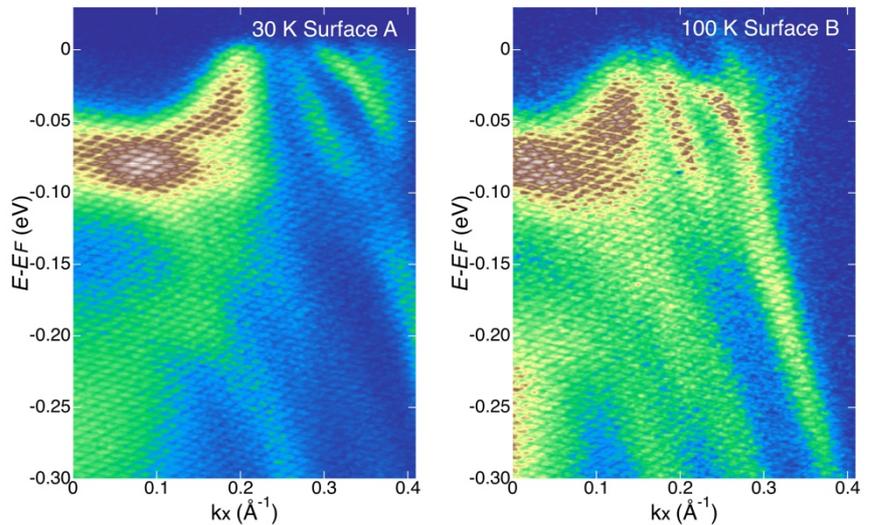


Fig.1 Band dispersion on surface A and B of WTe₂ measured by laser ARPES. The surface states connect the bulk electron- and hole-like pocket.

pockets, and parts of them are above the E_F , we measured the samples at a high temperature (over 150 K) which unoccupied states about 30 meV above E_F could be seen. SARPES measurements on candidate topological surface states were taken after the ARPES measurements.

Results

We measured two kinds of surfaces of WTe_2 : surfaces A and B with 7 eV laser-ARPES. In Fig. 1, we can see the band structures where the surface state connected the hole- and electron-like bulk states on both surface A and B. The surface states of the opposite surfaces are supposed to be different.

Figure 2 and 3 show the SARPES results of the candidate topological surface states. For surface A (Fig. 2), clear spin reversal could be observed for the surface states for both x- and y-direction. The surface state for surface A is spin-polarized. For surface B (Fig.3), the spin-polarization along x-direction could be observed, but no clear evidence showing that the surface state on surface B is spin-polarized along y-direction.

We observed two kinds of surfaces of type-II Weyl semimetal WTe_2 with laser-ARPES. Clear surface states which connect the bulk hole- and electron-like pockets could be seen. By SARPES measurements, we found that the surface states are all spin-polarized, which is important evidence for that the observed surface states are topological Fermi arcs.

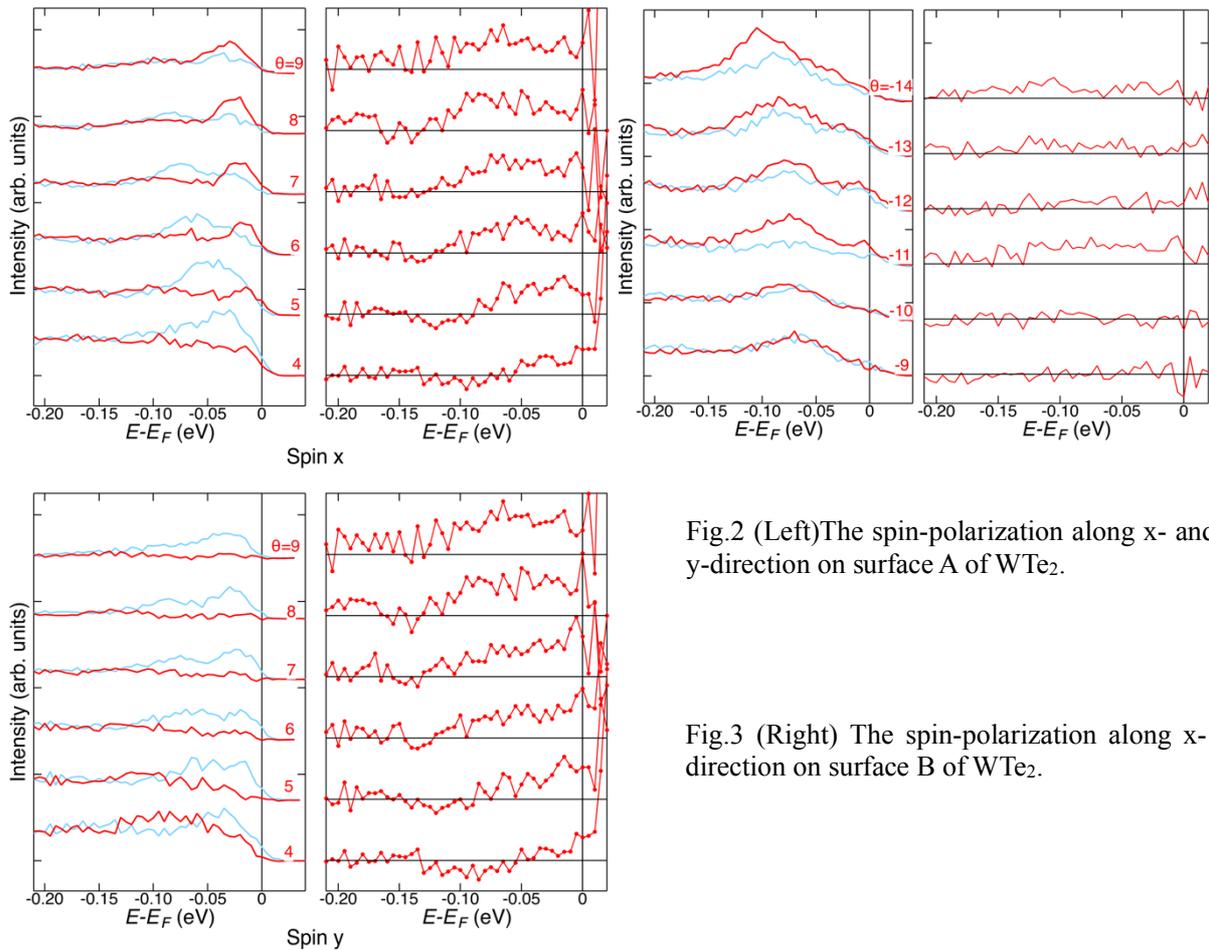


Fig.2 (Left)The spin-polarization along x- and y-direction on surface A of WTe_2 .

Fig.3 (Right) The spin-polarization along x-direction on surface B of WTe_2 .

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