OBSERVATION OF TOPOLOGICAL SUPERCONDUCTIVITY ON THE SURFACE OF AN IRON-BASED SUPERCONDUCTOR

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Topological superconductors are predicted to host exotic Majorana states that obey non-Abelian statistics and can be used to implement a topological quantum computer. By using high-resolution spin-resolved and angle-resolved photoelectron spectroscopy at ISSP, we find that the iron-based superconductor $FeTe_{1-x}Se_x$ (x = 0.45; T_c = 14.5 K) hosts topological surface states at the Fermi level; the surface states exhibit an s-wave superconducting gap below T_c. These results demonstrate that that the surface states of $FeTe_{0.55}Se_{0.45}$ are topologically superconducting, providing a simple and possibly high-temperature platform for realizing Majorana states.

Three evidences are necessary to experimentally prove that $FeTe_xSe_{1-x}$ ($x \sim 0.5$) is a topological superconductor: (i) Dirac-cone–type surface states; (ii) helical spin polarization of the surface states, which locks the spin direction perpendicular to the momentum direction; and (iii) an s-wave superconducting gap of the surface states when $T < T_c$. All these three evidences are observed in our ARPES experiments.

The observation of the Dirac-cone–type surface states. The overall band structure from the high resolution ARPES is summarized in Fig. 1A. We obtained clear parabola-like band together with Dirac-cone type band. Compare with the theory calculations [2], we conclude that the Dirac-cone–type band is the topological surface band, and the parabolic band is the bulk valence band.

The helical spin polarization of the Dirac-cone–type band. To further prove the topological nature of the Dirac-cone–type band, we measured their spin polarization. Two EDCs at the cuts indicated in Fig. 1B were measured. If the Dirac-cone–type band comes from the spin-polarized surface states, the EDCs at cuts 1 and 2 should show reversed spin polarizations. Indeed, the spin-resolved EDCs in Fig. 1CE, show that the spin polarizations are reversed for cuts 1 and 2, whereas the background shows no spin polarization (Fig. 1DF). These data are consistent with the spin-helical texture, which is the direct consequence of “spin-momentum locking” of topological surface states.

The s-wave gap for the topological surface band. Since iron-based superconductors generally have isotropic s-wave superconducting gaps, it is natural that the surface states also open an s-wave gap, due the proximity effect from bulk. Indeed, we observed clear s-wave gap on the surface band [3].

When the spin-polarized topological surface states open an s-wave gap, the corresponding superconducting states are topologically non-trivial. Thus, when an external magnetic field is applied, a pair of MBSs is expected to appear at the two ends of the vortices. Furthermore, if a magnetic domain is deposited on the surface, destroying superconductivity within that domain, there should be itinerant Majorana modes along the domain edge. It should be fairly easy to produce MBSs and Majorana edge modes. The relatively high T_c and facile growth of high-quality single crystals and thin films make Fe(Te,Se) a promising platform for studying MBSs and may further advance research on quantum computing.

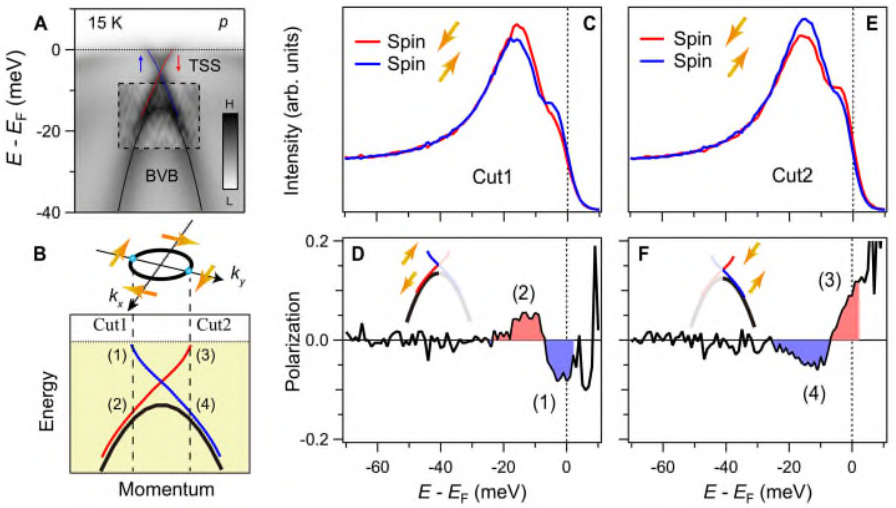

Figure 1 Dirac-cone–type surface band and its spin-helical texture. (A) Band structure measured from high-resolution ARPES. (B) Sketch of the spin-helical FS and the band structure along the sample GM direction. (C) Spin-resolved EDCs at cut 1. (D) Spin polarization curve at cut 1. (E and F) Same as (C) and (D), but for EDCs at cut 2. The measured spin polarizations are consistent with the spin-helical texture illustrated in (B).

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SPIN-POLARIZED ELECTRONIC STRUCTURE OF A SN TRIANGULAR LATTICE ATOMIC LAYER

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Spin-spit bands at solid surfaces have been extensively studied not only because of fundamental interests but also spintronic applications. Symmetry of the crystal surface plays an important rule in determining the nature of the spin splitting and the spin texture of a two-dimensional band [1]. Especially, a K point of the surface with three-fold symmetry is of interest due to the emergence of peculiar spin-polarized bands. For example, a Zeeman-type spin splitting with out-of-plane spin polarization appears at the K point of Tl/Si(111)-(1x1), where the surface structure belongs to the plane group of *p*3*m*1, and thus the symmetry of the K point is C_3 [2]. On the other hand, in the case of Bi/Si(111)-($\sqrt{3}\times\sqrt{3}$)R30° that belongs to the plane group of *p*31*m*, a Rashba-type spin-splitting was found at the K point having the symmetry of C_{3v} [3]. Here we point out that only the symmetry of the crystal lattice has been considered for interpreting the spin splitting of the bands so far. In the present study, we report presence of the both types of spin-spit bands at a K point of a triangular lattice atomic layer (TLAL) consisting of Sn atoms.

Recently, we synthesized the Sn TLAL by intercalation into the interface between graphene and SiC(0001) substrate. In this system, the intercalated Sn atoms are located at T_1 sites on the SiC(0001) surface and covered by monolayer graphene [4]. The Sn TLAL shows a (1×1) periodicity with respect to the SiC(0001) substrate. We investigated the spin-polarized electronic band structure of the Sn TLAL by (spin- and) angle-resolved photoelectron spectroscopy [(S)ARPES] at the Institute for Solid State Physics, the University of Tokyo [5].

From the (S)ARPES measurements, we found that two different types of spin splittings coexist at the K point of the Sn TLAL: one is a Zeeman-type spin splitting and another is a Rashba-type band crossing. Note that the K point has the C_3 symmetry with taking the crystal structure into account. Therefore, the Zeeman-type is consistent with the symmetry of the lattice while the Rashba-type band crossing is inconsistent. To understand the origin of the unconventional spin degeneracy at the K point, we examined the charge density distribution of the Sn-derived bands by density functional theory calculations. The calculations revealed that the charge density distribution of the Rashba-type band belongs to the plane group of *p*6*m*, meaning that the charge density distribution has the symmetry of C_{3v} symmetry at the K point. The symmetry of C_{3v} allows the Rashba-type band crossing at the K point. We conclude that the symmetry of the charge density distribution governs the nature of the spin-split bands beyond that of the lattice.

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SPIN INTERFERENCE OF PHOTOELECTRONS EMITTED FROM THE HIGH-TC SUPERCONDUCTOR BSCCO

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Copper oxide-based compounds (cuprates) are one of the most studied classes of materials among the unconventional high-T_C superconductors. They have an extremely rich phase diagram and are representative systems for the study of strong electron correlations. As such, they have been probed extensively by angle-resolved photoemission spectroscopy (ARPES), and the cuprate $Bi_2Sr_2CaCu_2O_{8+d}$ (BSCCO 2212) is a well-known example [1-3]. Despite this, there was only one photoemission study of BSCCO with spin resolution, where angle-integrated measurements exploited a resonant process (at Cu L_3 edge, hv = 931.5 eV) in order to determine the Zhang-Rice singlet character of the CuO_2-derived states [4], which are the ones near the Fermi level. We have recently performed a (non-resonant, hv = 50 eV) spin- and angle-resolved photoemission spectroscopy (SARPES) study at the Swiss Light Source [5], with the aim of determining the Eisenbud-Wigner-Smith (EWS) time delay of photoemission. Thanks to an analytical model [6], it is possible to obtain an indirect estimate of time delays from a band in the attosecond (10e-18 s) domain by measuring the variation of spin polarization with binding energy. Since our preliminary results on BSCCO [5] yields time delays sensibly larger than in the non-interacting system Cu(111) [7], the question about influence of electronic correlations on the time scale of photoemission was raised.

In January 2018 we have performed laser-based (hv = 7 eV) SARPES experiments at Kashiwa. We have measured five BSCCO 2212 samples (pure, doped with Ni 10%, and doped with Pr 5% - 7.5% - 10%) with the aim of systematically studying the EWS time delay as a function of doping. The good quality of the samples and of the measurements can be checked in the portion of Fermi surface shown in Fig. 1(a) for the pure BSCCO, where the nodal direction was aligned along the x direction. A comparison of spin polarization for two different samples is shown in Fig. 1(b) with high resolution in a small energy range. The main result is that no clear differences are observed, thus questioning the working hypothesis about influence of correlations on the time delay. However, an unexpected issue makes the interpretation about time delays difficult: the fact that no clear peak of spin polarization is observed in correspondence of the photoemission intensity peak. This is better shown in the energy distribution curve (EDC) in Fig. 2(a), where the y component of the spin polarization is plotted together with the two up and down spin channels. The spin polarization is non-zero, but flat over the whole energy range of 300 meV, even when far from the main photoemission peak at around 20 meV.

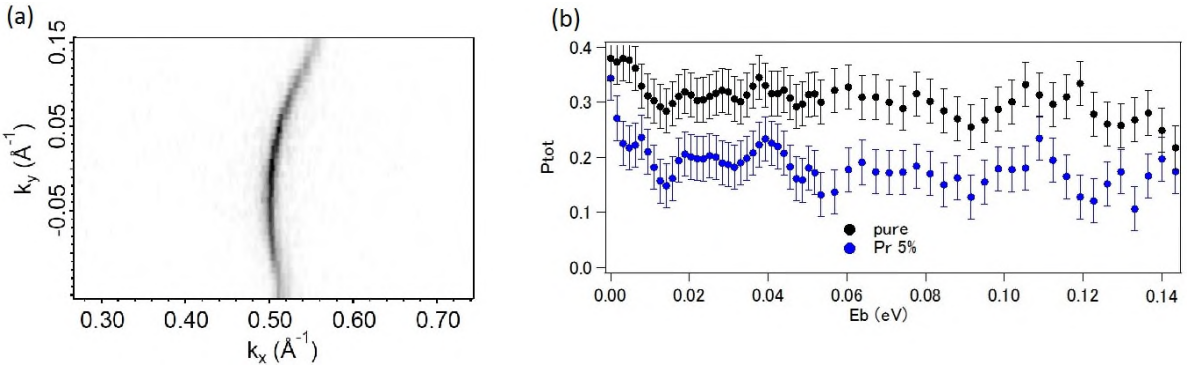

Fig. 1. (a) Portion of Fermi surface of BSCCO 2212. (b) Comparison of spin resolved EDC between pure BSCCO and doped with Pr 5%.

This peculiar flat spin polarization has currently no interpretation and might be related to the incoherent part of the photoemission spectrum. On the other hand, it is also difficult to completely exclude any possible experimental asymmetry that might influence the spin polarization signal.

At this point it is difficult to relate this spin polarization signal to the EWS time delay, and therefore to study the influence of correlations. A possible reason for the absence of a clear peak of spin polarization, as measured at the Swiss Light Source [5], is the use of a different photon energy. At the origin of the spin polarization that is related to EWS time delay lies an interference effect between different partial channels of the matrix element describing the photoemission process, where the two channels are due to different contributions in the initial or final state [6,7]. It could be that at the low photon energy of the laser, which reaches only the very low unoccupied states, only one single channel is allowed. In order to check the orbital composition of the final states and to test this possibility, spin-resolved one-step photoemission calculations are currently being performed by Prof. Jan Minár at the University of West Bohemia, Czech Republic. With the current implementation of the possibility to use 11 eV photon energy we hope to be able to see the interference channel in the near future.

An observation that supports the argument of a single transition channel is the measurement shown in Fig. 2(b), where the photoemission intensity is plotted as a function of linear light polarization angle (with 180 deg corresponding to p polarization). Fitting the data with a function that describes interference of two transitions [8], the obtained weight of one channel is larger than 95%. Since the degree of linear polarization of the used laser is of a similar value, this data is consistent with the single channel hypothesis.

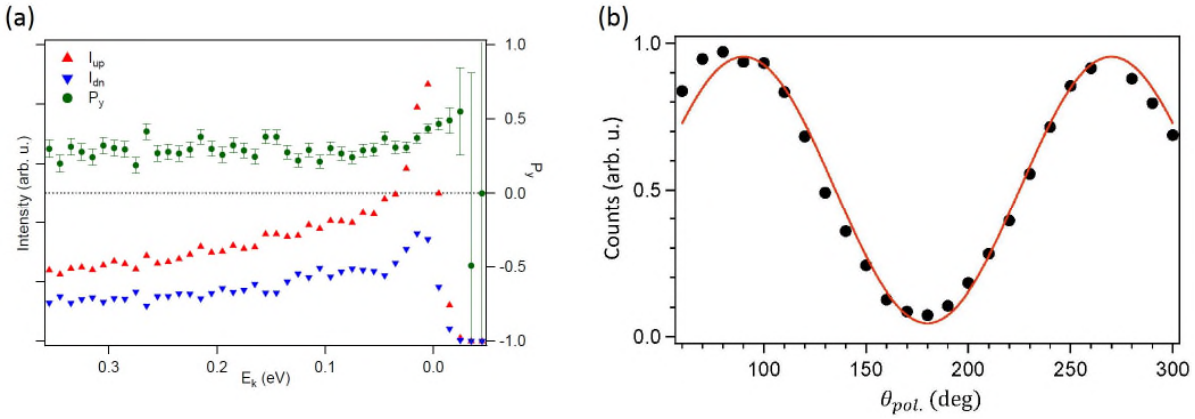

Fig. 2. (a) spin-resolved EDC on pure BSCCO 2212. The y component of spin polarization is flat also far from the photoemission peak. (b) Photoemission intensity as a function of light polarization angle.

Besides these measurements we performed SARPES experiments using different light polarizations on ferroelectric α -GeTe(111) films. High quality data was obtained which is now being prepared for publication together with 1-step photoemission theory.

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STUDIES ON FINITE TEMPERATURE EFFECTS OF HALF-METALLIC FERROMANGETS BY BULK-SENSITIVE HIGH-RESOLUTION SPIN-RESOLVED PHOTOEMISSION SPECTROSCOPY

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Half-metallic ferromagnet (HMF) is a crucial material for spintronics devices because of its 100% spin polarization of the density of states (DOS) at the Fermi level (E_F).[1] CrO_2 was predicted to be a HMF by first-principle calculations.[2] As a notable advantage, at low temperatures, CrO_2 always shows almost 100% spin polarization which is the highest value in candidates for HMFs.[3] However, magnetoresistance studies using magnetic tunnelling junctions reported that the spin polarization decreased drastically with increasing temperature. Accordingly, CrO_2 behaves as 'normal' ferromagnetic metal above 50–100 K.[4] The magnitude of the spin depolarization seen in the magnetoresistance behavior is much greater than that of macroscopic magnetization measured by SQUID. As one of the reasons of the precipitous depolarization, it has been suggested that an electron–magnon interaction broadens bandwidth of conduction minority spin bands and the tail of the minority spin states comes across E_F, which can permit spin-flip scattering of the conducting majority spin electrons into the minority spin states.[1] The minority spin tail state is so-called non-quasiparticle (NQP). Although the indication of existence of the NQP states have been suggested in several tunnelling magnetoresistance studies,[5] there is no direct evidence for the NQP in HMFs at present. In this study, in order to demonstrate the existence of the NQP states, we performed high-resolution spin-resolved photoemission spectroscopy (SRPES) on CrO_2 films with a focus on the close vicinity of E_F with an energy scale of sub-meV.

All of the spin-resolved PES data of $CrO_2/TiO_2(100)$ epitaxial film were acquired by the laser-based spin- and angle-resolved photoemission spectroscopy (ARPES) apparatus at the Institute for Solid State Physics at the University of Tokyo.[6] The photon energy and energy resolution were 6.994 eV and 30 meV, respectively. We magnetized the samples along the magnetic easy axis ([001] direction) by bringing the samples close to a magnet at room temperature.

In Fig. 1(a), a clear Fermi edge was observed in the majority spin spectrum while no states at E_F with an energy gap of 100 meV below E_F were observed. These features are just those of half-metallic ferromagnet. However, in bulk-sensitive SRPES studies, the gap size of the minority spin state is estimated to be 500 meV below E_F at 40 K where the degree of spin polarization is 100 % independent on binding energy.[7] The present minority spin energy distribution curve (EDC) and spin polarization is inconsistent with those of the bulk-sensitive SRPES study. This inconsistency can be attributed to the difference of the surface sensitivity of these measurements. Therefore, in our measurements by the laser, we may observe non-negligible intensity from the surface contaminants in addition to that of CrO_2. Based on this picture, the background seen in the minority spin spectrum at 10 K in Fig. 1(a) can be identified to be the tail of the electronic states of the surface contaminants as Cr_2O_3 which has a peak at 2 eV with no Fermi edge.[8] Since we underestimate the spin polarization when non-negligible unpolarized background exists in a SRPES spectrum, we should remove the

background to obtain intrinsic values of spin polarization.[9] Therefore, we subtracted the smoothed spectrum of the minority spin one at 10 K from both majority and minority spin spectra at any temperatures.

Up to 70 K, the spin polarization decreases in the energy range above 80 meV, with the same binding-energy dependence. Above 80 K, we found that the spin polarization evidently dropped toward E_F. The spectral shape of the spin polarization clearly bends at E_B = 10 meV and the spin polarization drops toward E_F. Such a bending shape of the spin polarization is very evident at temperatures up to 120 K. Above 150 K, the bending point shifts toward the higher binding energy side and the bending structure gets broader. The tendency of the temperature dependence is consistent with the appearance of NQP state. In Fig. 1(b), with increasing temperature, from 20 K to 120 K, while the majority spin states do not change significantly, a finite state appears in the minority spin states at and above E_F. This minority state causes the depolarization near E_F. We observed the fine change of the minority spin state for the first time, by using high-resolution SRPES, which implies that the experimental technique is a powerful tool to investigate the spin-resolved electronic structure of spintronic materials.

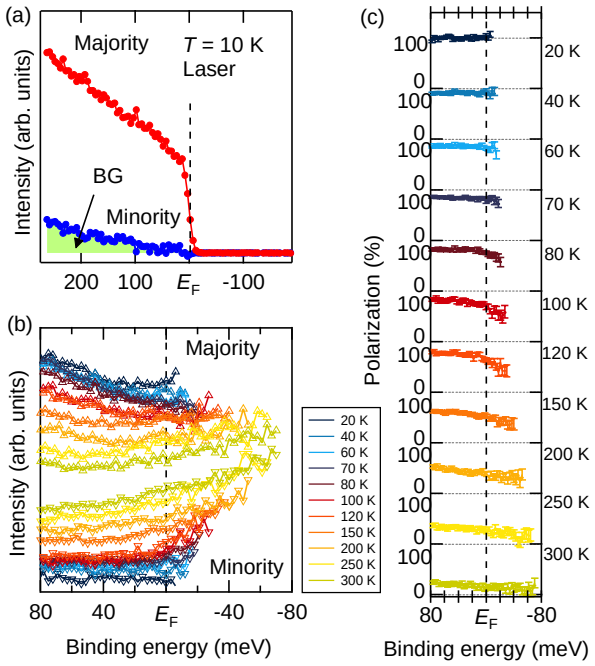

Fig. 1 (a) SRPES spectra before subtracting the background. Red and blue dots with solid lines show the majority and minority spin EDCs, respectively. **(b)** Temperature dependences of spin-resolved spectra divided by the Fermi-Dirac function at measured temperatures after background subtracted. **(c)** Temperature dependence of spin polarization.

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

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Transition metal dichalcogenide compounds with pyrite-type structure show various magnetic and electronic properties.[1] CoS_2 is a ferromagnetic metal with a Curie temperature of 122 K.[2] CoS_2 was predicted to be a half-metallic ferromagnet by first-principle calculations.[3,4] This is supported by a fact that its magnetic moment is 0.92 μ_B/Co which is close to 1 μ_B/Co in a low-spin state of Co^{2+} (S = 1/2 and g = 2).[2] However, it is expected that Co 3*d* electrons are strongly correlated each other, which suggests that the electronic structure can be renormalized and deviated from that predicted from band calculations.[5] Therefore, the electronic structure must be determined experimentally to reveal its half-metallicity. In this report, we demonstrate the spin-resolved electronic structure of CoS_2 revealed by high-resolution spin- and angle-resolved photoemission spectroscopy (SARPES).

All of the SARPES data of a CoS_2 single crystal were acquired by the laser-based SARPES apparatus at the Institute for Solid State Physics at the University of Tokyo.[6] A clean surface for measurements was obtained by *in situ* cleaving of the sample. We magnetized the samples along the [010] direction by bringing the samples close to a magnet below the Curie temperature. *P*-polarized light with $h\nu$ = 6.994 eV was used to excite the photoelectrons. The energy resolutions of ARPES and SARPES measurements were set at 2 meV and 15 meV, respectively.

Figure 1(a)–(c) show the ARPES and SARPES intensity map along Γ–X line measured at 40 K. Highly intense bands are located around 1 eV. In Fig. 1(b) and (c), energy positions of the intense majority and minority spin bands are slightly different, namely, the energy of the majority spin band is lower than that of the minority spin band. The splitting energy is estimated to be 100 meV. These intense bands can be t_{2g} bands. Band structure calculated within LSDA have t_{2g} bands localized between –1.5 eV and –2 eV.[3,4] The energy is 0.5–1 eV smaller than that of our results. Furthermore, the exchange splitting of t_{2g} bands are approximately 1 eV which can be overestimated based on our results.

We also observed clearly spin-polarized bands located between –0.6 eV and E_F: α, β, and γ bands. These bands can be e_g bands because of correspondence to LSDA calculation. In Fig. 1(b), α band is a majority spin band. β and γ bands were observed as minority spin bands in the close vicinity of E_F as shown Fig. 1(e), while no clear majority spin bands are observed near E_F as shown in Fig. 1(d). LSDA calculations indicate that there is no minority spin band along Γ–X line in CoS_2,[3,4] which is inconsistent with our SARPES results shown in Fig. 1(e). This contradiction can be attributed to overestimation of the exchange splitting in LSDA calculations. In LSDA calculations, a band corresponding to α band is located at –0.75 eV at the Γ point, while α band is observed at –0.5 eV.[3,4] If the deviation is attributed to the overestimation, minority spin conduction bands should be move to the lower energy side with

the same energy scale. The studies of the LSDA calculation shows that the bottom of the minority spin conduction band along Γ–X line is located at 0.2 eV; this energy is comparable with the deviation of energy position between the theoretical majority spin band and the experimental one.[3,4] This suggests that the minority spin band may cross E_F, consistent with our results.

In summary, we performed high-resolution SARPES in order to reveal the electronic structure of ferromagnetic CoS_2. We observed a slightly spin-polarized t_{2g} bands and highly spin-polarized e_g bands. We found that a minority spin band crossing E_F exist at 40 K, demonstrating that CoS_2 is not a half-metallic ferromagnet. We also found that LSDA calculations overestimate the exchange splitting of CoS_2. This report implies that the high-resolution SARPES is a powerful tool to investigate strongly correlated ferromagnetic materials.

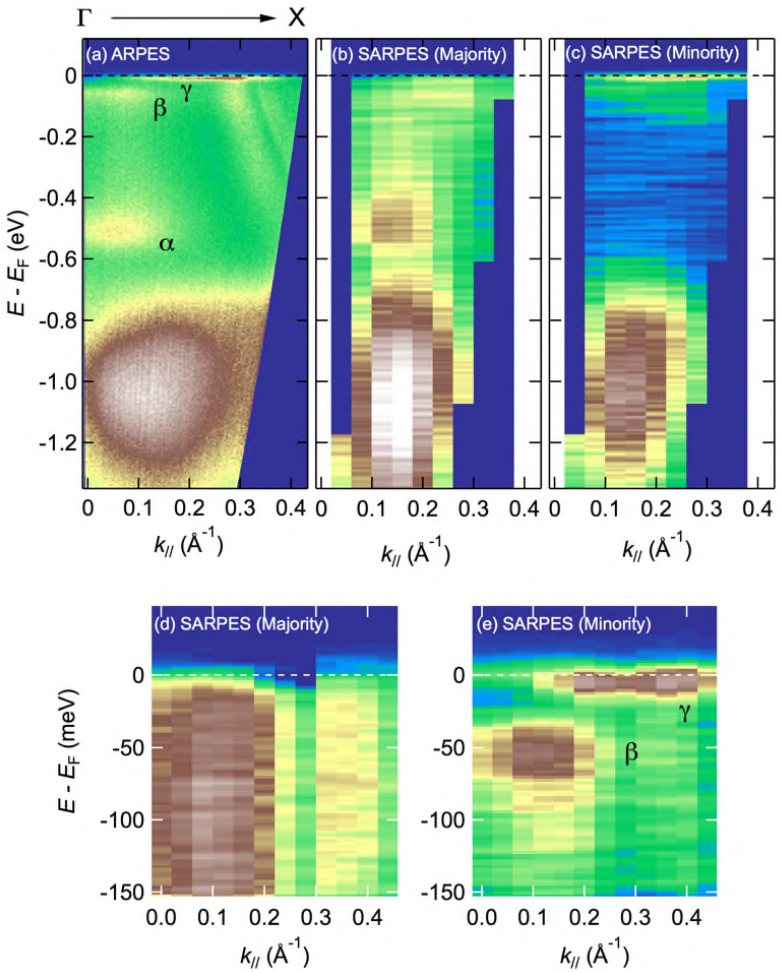

Fig. 1 (a) ARPES intensity map along Γ–X line, (b) SARPES intensity map for the majority spin bands, and (c) that for the minority spin bands of CoS2 (100) surface acquired at 40 K. (d) and (e) SARPES intensity map near E_F for majority and minority spin bands, respectively.

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

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The major breakthroughs in understanding topological materials over the past decade were all triggered by the discovery of the Z_2 topological insulator (TI). In three dimensions (3D), the TI is classified as either "strong" or "weak", and experimental conformations of the strong topological insulator (STI) rapidly followed the theoretical predictions [1-3]. In contrast, the weak topological insulator (WTI) has so far eluded experimental verification, since the topological surface states exist only on side surfaces which are typically undetectable in real crystals. In this work, we reveal the WTI state, for the first time, in β-Bi_4I_4 with naturally cleavable top and side planes, which is necessary for the experimental confirmation of the WTI state [4, 5].

The quasi-1D compounds β-Bi_4I_4 is formed by a stacking of Bi_4I_4 chains. From band calculations, the β-phase is in proximity of three different topological phases [4, 5]: a STI (1;110), a WTI (0;001), or even a trivial phase (0;000), sensitively depending on correction of the band gap. Experimental determination is therefore required through the direct observation

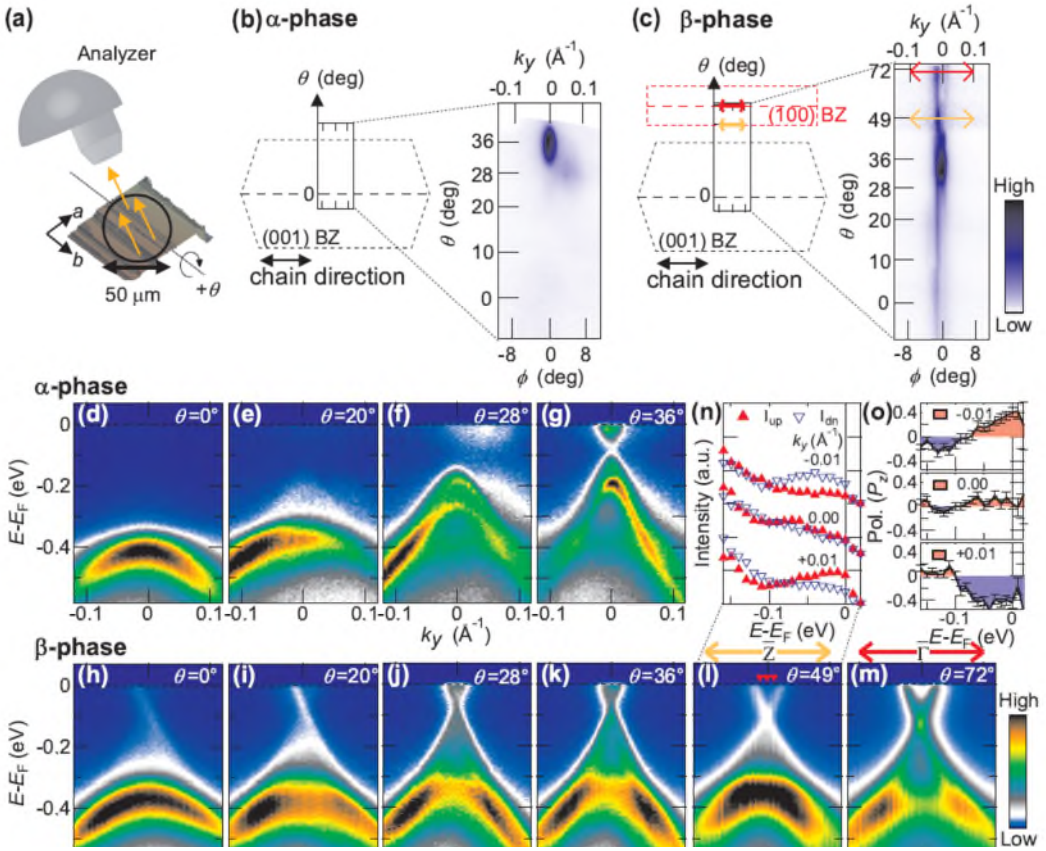

Fig. 1 (a) Experimental geometry of laser-(S)ARPES. We collect the photoelectrons at different emission angle θ with respect to the normal of the top-surface (001). (b, c) Photoelectron intensity distributions at E_F for the two materials. The intensities are integrated within 40 meV. The black and red dashed lines display the surface Brillouin zones of (001) and (100) surfaces, respectively. (d-g) and (h-m) APRES band maps taken at different θ-s in the α-phase and the β-phase, respectively. In (l) and (m), ARPES band mapping along the high-symmetry lines cut at the $\bar{Z}$ and $\bar{\Gamma}$ of the (100) surface BZ. (n, o) Spin-resolved photoelectron intensity and spin-polarizations along *z*-axis (in-plane) at θ=49°. The red triangles in (l) indicate the measurement positions.

of relevant topological surface states (TSS). There is the family material α-Bi_4I_4 with different stacking sequence, which is considered to be topologically trivial. Interestingly, these two-material phases can be selectively controlled just by temperature around room temperature, and thus opening a possible way to switch the band topology.

To verify the possible WTI state and the topological phase transition, we perform laser-SARPES at ISSP. Fig. 1(a) shows the experimental geometry Since the incident laser light with 50 μm in spot size illuminates both the terraces and facets exposed on the cleaved surface, the ARPES signals from the (001) and (100) planes are accumulated all together in the experiments. The mapping of photoelectron intensities at the Fermi energy (E_F) exhibits clearly different for the two materials [Figs. 1(b, c)]: a quasi-1D like shape is obtained in β-phase, whereas the island-like intensities are detected only around $\bar{M}$ in BZ of the (001) plane for the α-phase. Figures 1(d-g) display the ARPES maps in the α-phase at different emission angles (θ) with respect to the normal of the top-surface (001). No in-gap TSS is observed at high-symmetry momenta, the $\bar{\Gamma}$ [Fig. 1(d)] and $\bar{M}$ points [Fig. 1(g)], revealing its trivial band topology. In sharp contrast, we find a Dirac-cone like energy dispersion near E_F in the β-phase [Figs. 1(h-m)]. This band shows a quasi 1D character seen in Fig. 1(c), and it is only weakly dispersive in energy along $k_{x/z}$ (or with θ-variation). Since this state is not detected in the trivial α-Bi_4I_4, it should be TSS. Remarkably, the Dirac-state is observed even at θ=0°, or $\bar{\Gamma}$ in the BZ of the (001) surface, whereas no such state is predicted by the DFT calculations for any cases of band topology. The only possible explanation for it is that the observed quasi-1D Dirac-state is derived from the side-surface (100). The nontrivial band topology of either STI or WTI can be determined by comparing the observed surface dispersions with theoretical expectations: a large gap should be observed for a STI at the $\bar{Z}$ point, while no gap is expected for a WTI. In Figs. 1(l-m), we show the ARPES band maps obtained at θ=49° and 72°, which cut across the $\bar{Z}$ and $\bar{\Gamma}$ points along k_y, respectively, in the BZ for the (100) surface. The observed Dirac dispersions are gapless at the both *k* points. Moreover, in-plane spin polarizations are observed by spin-resolved measurements at θ=49°, ensuring the existence of TSS at the $\bar{Z}$ point of the (100) surface [Fig. 1(n, o)]. All of these results validate the classification of β-Bi_4I_4 as a WTI [6].

Our work will stimulate further in-depth study on exotic quantum phenomena of the WTI state as well as provide technological possibilities of functional topological materials using the topological phase transition accompanying the structural transition.

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SPIN-POLARIZED QUASI ONE DIMENSINAL ELECTRONIC STRUCTURE OF BI/INAS(110)-(2×1)

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Spin-polarized one-dimensional (1D) electronic structures are extensively studied as one of the promising platform for the basic research of exotic low-dimensional phenomena such as Majorana bound states [1] and spin-selective density wave formation [2]. In order to realize such spin-split 1D states, Rashba-type spin-orbit interaction (SOI) driven from space-inversion asymmetry in crystal surfaces and interfaces has been gathering attentions in this decade [1-3].

To realize sizable Rashba SOI, surface atomic structures containing heavy elements are regarded as a good candidate. Among them, we focused to the Bi/InAs(110)-(2×1) surface because its surface atomic structure is constituted with heavy Bi atoms forming quasi-1D (Q1D) atomic structure, implying anisotropic surface states [4]. Despite such interesting surface atomic structure, the detailed surface electronic structure, especially its spin polarization, has never been studied so far.

In this work, we have found the spin-polarized Q1D state on Bi/InAs(110)-(2×1) surface. The band structure and spin polarization of the Bi/InAs(110)-(2×1) surface electronic states were measured by laser-based angle-resolved photoelectron spectroscopy (ARPES) and spin-resolved ARPES (SARPES) at the Institute for Solid State Physics [5]. For the SARPES measurements, the photon-incident plane was (001), and the electric-field vector of the linearly polarized photons were normal to the incident plane (001). The InAs(110) surface was prepared by cleaving the side face of the InAs(001) wafer in an ultra-high vacuum chamber. Then, Bi were evaporated at room temperature. After annealing at 563 K for 15 minutes, a (2×1) periodic structure was observed by low energy electron diffraction (LEED), consistent with a previous study [4].

Figure 1 shows the ARPES intensity plot along $\overline{\Gamma}$–$\overline{\mathrm{X}}$. k_x is defined parallel to [$\bar{1}$10] direction. A pair of the hole bands lie slightly below the Fermi level, showing a semiconducting character. Figure 2 displays ARPES constant energy contour: k_y is defined parallel to [001]. The contour shape is highly anisotropic and waving along k_x, indicating that this state is Q1D state with inter-chains interaction. Since such states were not observed on the as-cleaved InAs(110) surface, they should be derived from Q1D Bi chains on the surface.

Figure 3 shows the SARPES energy distribution curves (EDCs) taken along k_x at $\overline{\Gamma}-\overline{\mathrm{X}}$ (k_y =0.00 Å^{-1}) and $\overline{\mathrm{Y}}-\overline{\mathrm{M}}$ (k_y =0.26 Å^{-1}) at 40 K. The filled (open) triangles correspond to spin polarization parallel (anti-parallel) to [001].The peaks of EDC in Fig. 3 disperse downward from $\overline{\Gamma}$. Such dispersions are consistent with the paired hole bands in Fig. 1. The spin direction of EDCs inverts together with the sign of the emission angle. Such paired parabolic dispersion with spin-polarization without breaking time reversal symmetry strongly suggests the Rashba-type spin splitting.

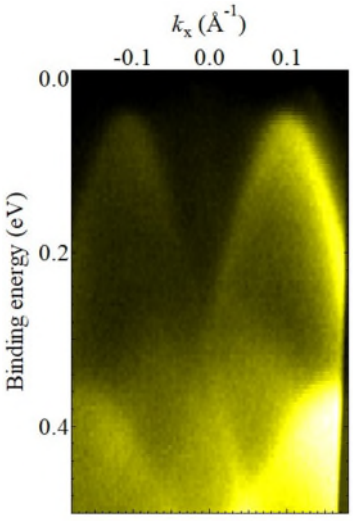

Figure 1. ARPES intensity plot of Bi/InAs(110)-(2×1) along $\overline{\Gamma}$–$\overline{\mathrm{X}}$ (k_y = 0.00Å^{-1}) measured at 40K with circular polarized photons.

Assuming parabolic dispersions with spin splitting along k_x, the size of the Rashba SOI, scaled by the Rashba parameter α_R, is evaluated as 5.5 eVÅ along $\overline{\Gamma} - \overline{X}$. This value is the largest among other 1D and Q1D Rashba systems [6, 7] and is comparable with typical giant Rashba systems such as BiTeI and Bi/Ag surface alloys [8, 9].

In summary, we have reported the spin polarized Q1D surface state on Bi/InAs(110)-(2×1) surface by using laser-SARPES. The surface state of Bi/InAs(110)-(2×1) showed large spin-polarized Q1D state driven by Rashba-type SOI. This Q1D state would be a good template for further research on spin-dependent 1D physics as well as a future application to spintronics [10]. In parallel, detailed research on spin-orbital entanglement of the Q1D states as well as the origin of the giant Q1D Rashba SOI is in progress.

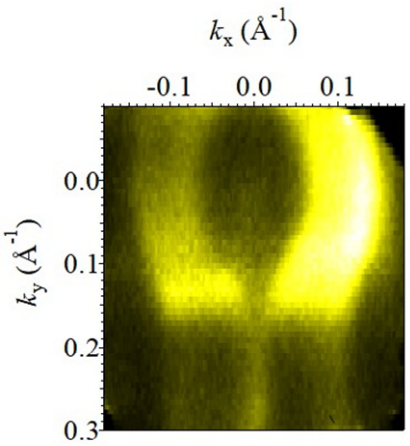

Figure 2. ARPES constant energy contour at a binding energy of 100 meV.

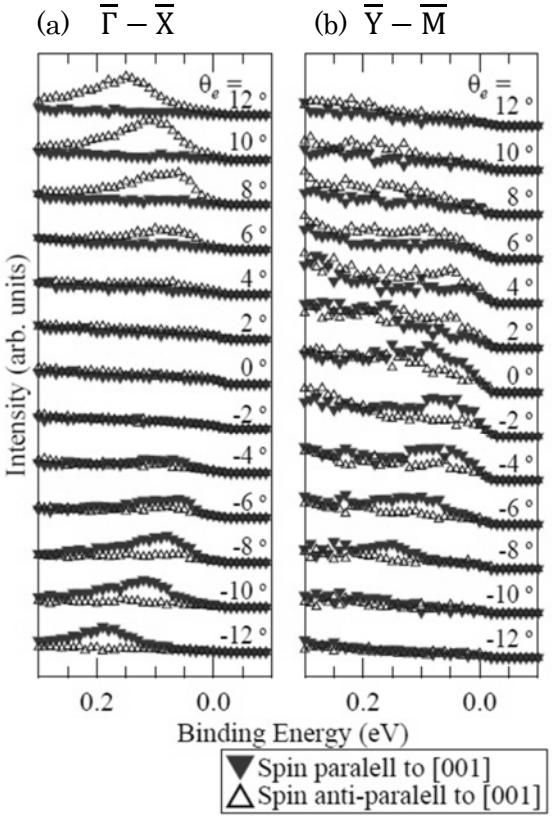

Figure 3. SARPES energy distribution curves taken along k_x at (a) $\overline{\Gamma} - \overline{X}$ (k_y = 0.00 Å^{-1}) and (b) $\overline{Y} - \overline{M}$ (ky = 0.26 Å^{-1}) at 40 K. θ_e = 10 ◦ corresponds to 0.14 Å^{-1}

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Laser SARPES study on spin-orbit coupled surface states of topological insulators: helicity dependence of photoelectron spin

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Introduction:

A central subject in the research for spintronic application is realization of functional capabilities to generate highly spin-polarized electrons and control its spin degree of freedom. As a promising spintronic material, a highly spin-polarized topological surface state (TSS) in three-dimensional topological insulators (TIs) has grabbed particular attentions. The TSS forms Dirac-cone-like energy dispersion and exhibits helical spin texture in momentum space due to the strong spin-orbit coupling (SOC) [1]. The peculiar spin texture protects the TSS electron from backscattering, which makes the TSS robust against perturbations. The TSS therefore can be useful as a novel spin generator and conductor.

Here, the TSS faces a new challenge for optical control over its spin properties. In addition to the helical spin texture, it was proposed that the relativistic SOC plays an important role to blend quantum states with different spin and orbitals, which gives a rise to a "spin-orbital texture" in TSSs [2, 3]. Since electric field of light directly couples with the orbitals, through the entanglement between the spin and orbital wavefunctions, it may enable us to optically manipulate the spin degree of freedom.

Recently, we use laser-SARPES at ISSP, and show a great capability of linear-polarizations to optically control photoelectron's spin in three-dimension (see Fig. 1). This light-polarization evolution of the spin orientation is successfully described by interference between spin-up or spin-down spinor wavefunctions from the initial spin-orbital textures [4, 5]. In this report, we here extend this photoemission model to include circular polarization effect that is considered as a superposed field of *p*- and *s*-polarizations with a relative phase of $\pi/2$. We demonstrate that the rotation of the excited photoelectron's spin follows the coherent optical superpositions.

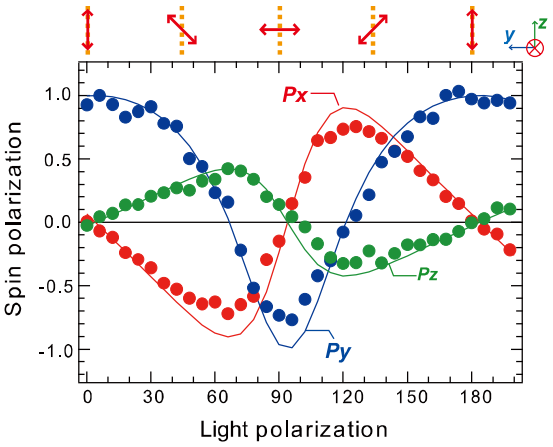

Figure 1: Plots of previously observed spin polarizations for *x*, *y* and *z* axis of the TSS as a function of the linear-polarization angle (θ) with respect to the mirror plane at k_x=−k_F [4]. At θ=0°, the electric field of the laser is aligned in the *x*-*z* plane [see Fig. 2(a)] and thus *p*-polarization. From 0° to 90°, the electric field changes from *p*- to *s*-polarization, and becomes *p*-polarization again at θ=180°.

Experimental:

Laser-SARPES was performed at laser-SARPES machine at ISSP with high-flux 6.994-eV laser light [6]. The laser-SARPES machine is based on two high-efficient VLEED spin-polarimeters and the hemispherical analyzer with photoelectron deflector function (ScientaOmicron DA30L). This spectrometer can resolve spin polarization components of photoelectrons for in-plane ($P_{x,y}$) and out-of-plane (P_z) orientation. During the measurement, the sample temperature was kept below 20 K, and instrumental energy and angular resolutions were set below 20 meV and 0.7°, respectively. Experimental configurations is shown in Fig. 2(a) where the light incidence plane and the detection plane match the mirror plane of the crystal (*x*-*z* plane). The *p*- (*s*-) polarization excites *even* (*odd*) parity orbitals with respect to the mirror plane [4, 5]. The light polarizations are controlled by half- and quarter-waveplates.

Results:

Figure 2(b) represents spin-integrated ARPES intensity map for Bi_2Se_3 along the Γ-M high

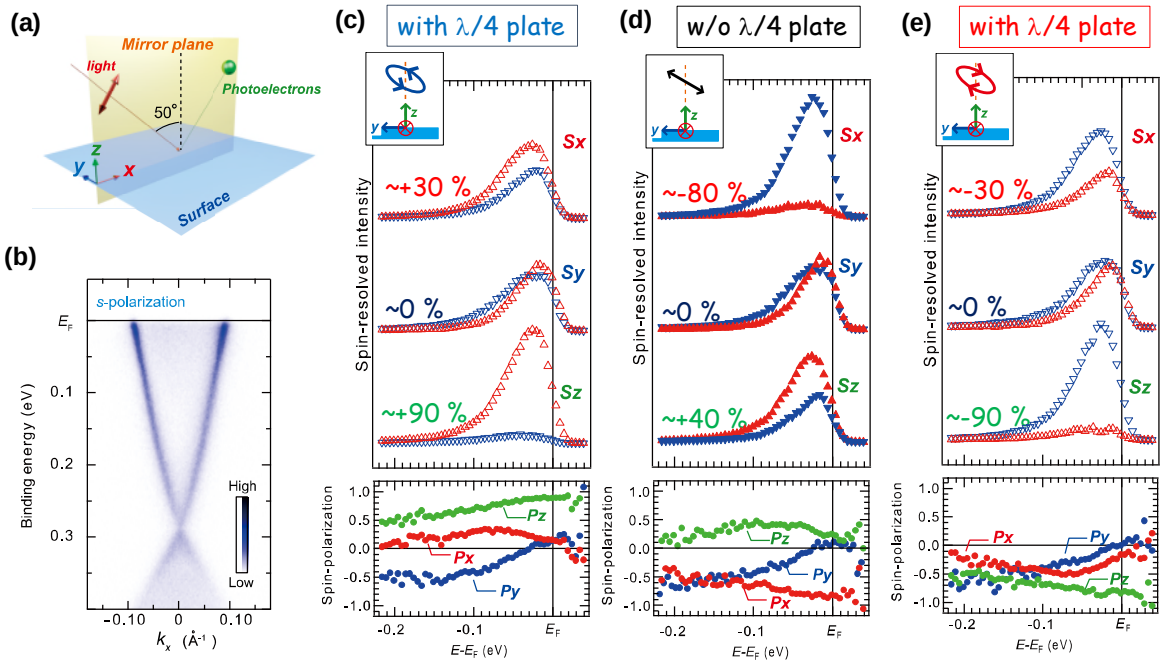

Figure 2: (a) Experimental geometry. In the geometry, according to pure orbital selection rule, *p*- and *s*-polarized lights selectively excite spin-up and spin-down states, respectively. (b) ARPES intensity map, showing Dirac-cone like energy dispersion of TSS in Bi_2Se_3. (c-e) Spin-resolved photoemission intensities and the corresponding spin polarization as a function of binding energy obtained by various light polarizations, (c) left-handed elliptical, (d) tilted linear, (e) right-handed elliptical (see the inset). These light polarizations are obtained by a combination of half- and quarter-waveplates for 7eV laser. The obtained spin axes correspond to those in (a).

symmetry line. The data well resolves the sharp TSS with the Dirac-cone-like energy dispersion of the surface state. All of the SARPES measurements shown below are performed at (k_x, k_y) = (–k_F, 0), which is the same *k* point [4]. By using tilted linear polarization with half-waveplate and without quarter-waveplate, we see almost no-spin polarization along *y* axis while large spin-polarizations for *x* and *z*. In particular, the size of the spin polarization for *x* achieves 80 %. This behavior is quite consistent with the previous result as shown in Fig. 1 (θ~65°).

As a heart of our interests, we now verify helicity dependence, which gives an optically induced "phase" in the photoemission interference process, since the light has own relative phase (–π/2 or +π/2) depending on its helicity. To demonstrate this, we now fix the half-wave plate angle, to produce the tilted polarization, and insert quarter-waveplate. The resulting light field is expected to be elliptical with a long-axis along the tilted linear-polarization [see the inset of Figs. 2(c) and 2(e)]. Depending on the quarter-wave plate angle, we tune its helicity with either left-handed (–π/2) or right-handed (+π/2). By the left-handed polarization [see Fig. 2(c)], one can immediately see the impact of the helicity by looking at the data for *x* and *z* compared to those for the linear polarization [Fig. 2(d)]. Apparently, there is a cross relation in not only the size of the spin-polarization but also the shape of the spectra. This cross relation indicates that the resulting spin-polarization rotates by –π/2 in P_x-P_z plane. Moreover, the similar relation is clearly observed for the right-handed [Fig. 2(e)], and but the resulting spin direction is completely opposite with respect to that obtained in the left-handed [Fig. 2(c)]. These data unambiguously demonstrate that the light helicity induce "phase" in the spin matrix elements which is directly visualized by laser-SARPES.

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