

Research Activities

– Synchrotron Radiation Experiments –

Unveiling orbital characteristics of surface states in HfSiS

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Keywords: orbital characteristics, Rashba-type surface state.

Recently the non-symmorphic Dirac nodal-line semimetals WHM ($W = \text{Zr, Hf, La}$; $H = \text{Si, Ge, Sn}$, and Sb ; $M = \text{S, Se, Te, O}$) have been theoretically and experimentally investigated. HfSiS is the most representative compound because of the emergence of spin-split surface states (SSs) due to a strong spin-orbit coupling at the $\bar{X}$ point with C_{2v} symmetry near the Fermi level [1, 2]. Thus, HfSiS could be a good candidate to realize exotic orbital and spin texture due to spin orbital fields on the surfaces. However, until now, there has been no systematic investigations on the orbital configuration of these surface states.

We have directly investigated the orbital configurations of the surface-derived states in HfSiS by means of angle-resolved photoelectron spectroscopy (ARPES) employing variable linear-polarized light at HiSOR BL-1 as well as theoretical calculations within density functional theory. To observe electronic states in detail we study the Fermi surface map (FSM) of HfSiS using the p - and s -polarization geometries. The prominent feature in the FSM is the olive-shaped electron pocket around the $\bar{X}$ point [marked by the green rectangle in Fig. 1(i)]. Considering the experimental geometry and the matrix elements for the dipole transition, one can observe initial-state orbitals having even symmetry with respect to the k_x - k_z mirror plane ($p_x, p_z, d_{xz}, d_{x^2-y^2}, d_{z^2}$) in the p -polarization geometry, whereas only initial-states having odd symmetry are excited (p_y, d_{xy}, d_{yz}) in the s -polarization geometry. As seen from Fig. 1, the shape and distribution of the spectral weight in FSM around the $\bar{X}$ -point are significantly different for the p - and s -polarization geometries indicating clearly directional alternation of the orbital symmetry [3].

The calculated Fermi surface spectral function for the p - and s -polarization geometries [Fig. 1 (ii) and (iv)] matches very well the experimental results. Our analysis shows that the Hf-derived $d_{x^2-y^2}$, d_{z^2} , and d_{xz} orbitals contribute to the spectral weight mainly along the $\bar{\Gamma}\bar{X}$ -line when the p -polarized geometry is used. In contrast, the data acquired with the s -polarization geometry [Fig. 1 (iii)] clearly highlights additional bands being part of the SSs with the odd p_y, d_{yz} and d_{xy} orbitals [3].

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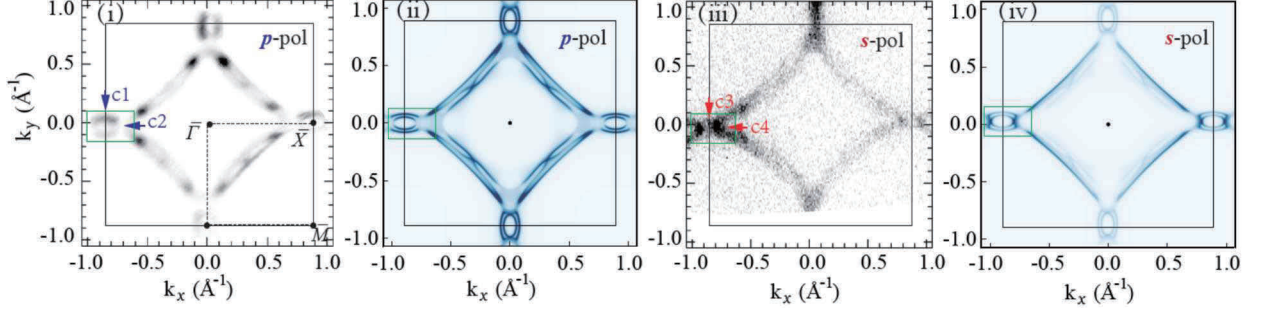


FIG. 1 Experimental Fermi surface measured at $h\nu=60$ eV in p -polarization geometry (i) and s -polarization geometry (iii). (ii) and (iv) Corresponding calculated Fermi surfaces for the p - and s -polarization geometries, respectively.

Effect of spin-orbit coupling on bulk bands of the non-symmorphic semimetal $\text{Hf}_{1-x}\text{Zr}_x\text{SiS}$

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Keywords: non-symmorphic semimetals, spin-orbit coupling (SOC), SOC induced energy gap

In recent years, non-symmorphic semimetals MSiS ($M=\text{Zr, Hf}$) has flourished because of the discovery of many materials that exhibit highly exotic physical properties that dominate the transport properties with extremely high carrier mobility [1, 2]. They share the same crystal structure but with a different spin-orbit coupling (SOC) strength. It is noted that the exotic state is governed by the electronic structures originated from the crystal symmetry with variable SOC strength [3-6]. However, the systematic studies of the electronic structures affected by the effect of SOC have yet to be reported for MSiS systems.

In this study, we systematically studied the bulk electronic states of $\text{Hf}_{1-x}\text{Zr}_x\text{SiS}$ using high-resolution ARPES. VUV-ARPES measurement with p -polarized light with a photon energy ($h\nu$) of 60 eV was performed at BL-1 of HiSOR. The typical energy and momentum resolutions were set at 10 meV and 0.1° , respectively. To understand the effect of SOC on the electronic structures, we begin the systematical analysis of the band structures with changing the x value in $\text{Hf}_{1-x}\text{Zr}_x\text{SiS}$. The VUV-ARPES spectra with p -polarized light of $\text{Hf}_{1-x}\text{Zr}_x\text{SiS}$ along the $\bar{M}\bar{\Gamma}\bar{M}$ -line are shown in Figs. 1(a)-1(c). A hybridization gap opens at the band crossing point halfway of the $\bar{M}\bar{\Gamma}\bar{M}$ -line, where the gap size is the largest in HfSiS [~ 90 meV, see Fig. 1 (a)] than those in $\text{Hf}_{0.6}\text{Zr}_{0.4}\text{SiS}$ [~ 70 meV, see Fig. 1(b)] and ZrSiS , [~ 30 meV, see Fig. 1 (c)], indicating that we can quantitatively adjust the strength of the spin-orbit coupling by controlling the x composition in $\text{Hf}_{1-x}\text{Zr}_x\text{SiS}$.

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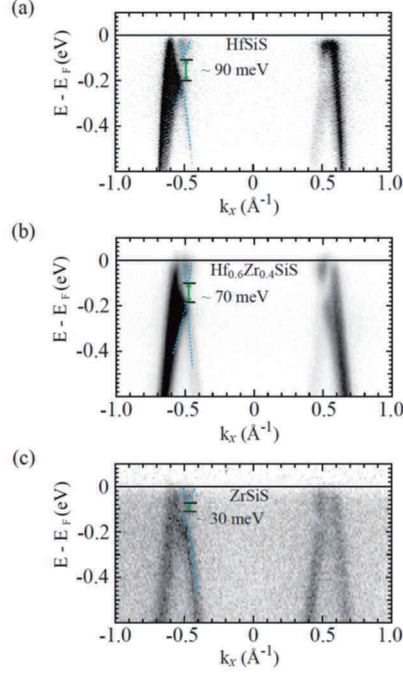


FIG. 1 VUV-ARPES dispersion intensity maps along the $\bar{M}\bar{\Gamma}\bar{M}$ -line acquired with p -polarized light at $h\nu=60$ eV of (a) HfSiS, (b) Hf_{0.6}Zr_{0.4}SiS, and (c) ZrSiS.

Effect of spin-orbit coupling on surface states of the non-symmorphic semimetal $\text{Hf}_{1-x}\text{Zr}_x\text{SiS}$

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Keywords: non-symmorphic semimetals, spin-orbit coupling (SOC), Rashba-type spin splitting

In recent years, non-symmorphic semimetals MSiS ($M=\text{Zr, Hf}$) has flourished because of the discovery of many materials that exhibit highly exotic physical properties which dominate the transport properties with extremely high carrier mobility [1, 2]. They share the same crystal structure but with a different spin-orbit coupling (SOC) strength. It is noted that the exotic state is governed by the electronic structures originated from the crystal symmetry with variable SOC strength [3-6]. However, the systematic studies of the electronic structures affected by the effect of SOC have yet to be reported for MSiS systems. We systematically studied the surface electronic states of $\text{Hf}_{1-x}\text{Zr}_x\text{SiS}$ using high-resolution ARPES.

VUV-ARPES measurement with p -polarized light with a photon energy ($h\nu$) of 60 eV was performed at BL-1 of HiSOR. The typical energy and momentum resolutions were set at 10 meV and 0.1° , respectively. The SOC also plays an important role in causing surface states splitting. The VUV-ARPES band intensity maps along the $\bar{M}\bar{X}\bar{M}$ -line show a ‘V’ shaped band dispersion at a photon energy of 60 eV in Fig. 1(a). These bands are assigned to the surface states (SSs) since they are located inside the gap of the projected bulk-band. The SSs around the $\bar{X}$ points are formed by distinctly separated bands in HfSiS as well as in $\text{Hf}_{0.6}\text{Zr}_{0.4}\text{SiS}$ [see Fig. 1(a)]. To further evaluate the size of the splitting, we extracted the momentum distribution curves (MDCs). The ARPES energy band dispersions of ZrSiS along the $\bar{M}\bar{X}\bar{M}$ -line exhibits a much smaller or no splitting as shown in Fig. 1(a) in comparison with those of HfSiS and $\text{Hf}_{0.6}\text{Zr}_{0.4}\text{SiS}$. The corresponding MDC shows almost no splitting [Fig. 1(b)]. This certainly stems from the lighter atomic mass and thus weaker SOC of Zr compared to that of Hf.

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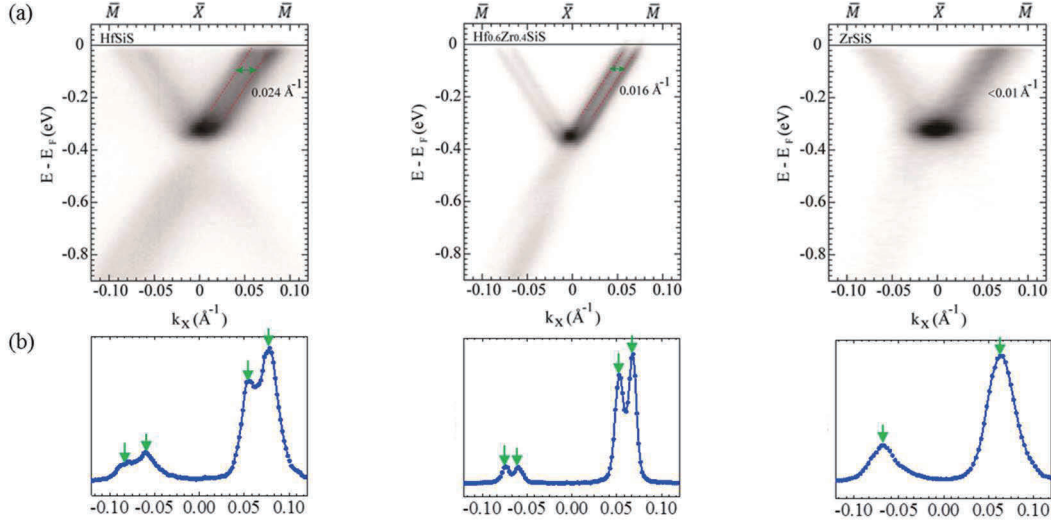


FIG. 1 (a) ARPES dispersion intensity maps for HfSiS, Hf_{0.6}Zr_{0.4}SiS and ZrSiS with p -polarized light along the $\bar{M}\bar{X}\bar{M}$ -line of the surface BZ. (b) The corresponding momentum distribution curve (MDC).

ARPES study of the electronic structure of HfSiS with variable polarized light

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Keywords: non-symmorphic semimetals, spin-orbit coupling (SOC), Rashba-type spin splitting, orbital configuration

VUV-ARPES measurement with *s* and *p*-polarized light with a photon energy ($h\nu$) of 60 eV was performed at BL-1 of HiSOR for one of the non-symmorphic semimetals HfSiS. To understand the electron orbital configuration, we focus on the high symmetry lines of HfSiS as shown in Figs.1(a) and 1(b). We find that the Rashba-like surface states (SSs) with a quasi-linear dispersion along the $\bar{M}\bar{X}$ -line indicated as SS1 [Fig.1 (a)(i)], which is visible with the *p*-polarization geometry. Its degeneracy point is located at 320 meV below E_F at the $\bar{X}$ -point. We should note that the Rashba-type split SSs can be reproduced by *ab initio* calculations only when the SOC is included (*not shown*).

Assuming the Rashba term to be the source of this state, we can extract the Rashba coefficient α as ~ 3.125 eV·Å, which is in accordance with previous reports [1]. Based on the quasi-linear Rashba-like dispersion, the derived Fermi velocity v_F can be evaluated as 4.55 eV·Å (6.9×10^5 m/s), which is comparable to previous reports on Rashba-type spin-split surface states [1,2]. Under the *s*-polarization geometry, the Rashba-like surface spectral weight is strongly suppressed as shown in Fig.1 (a)(ii), indicating the even orbital character. The *p*-polarized band dispersion along the $\bar{\Gamma}\bar{X}$ -line [Fig.1 (b)] shows a quasi-linear Dirac-like band dispersion, with the crossing point located around 500 meV being protected by the non-symmorphic symmetry. In addition, the M-shaped states along the $\bar{\Gamma}\bar{X}$ -line around 300 meV are connected to SS1 along the $\bar{M}\bar{X}$ -line, which emerge from the bulk states at lower binding energies. In contrast, ARPES result for the *s*-polarization geometry in Fig.1 (b)(ii) exhibits surface state (SS2) having a steep dispersion forming the terminal arc-like Fermi surface piece of the electron pocket around the $\bar{X}$ point (*not shown*), which shows a discernible spectral weight from the bulk states compared to the *s*-polarization, demonstrating that the SS2 is dominated by odd symmetry orbitals. Meanwhile, the intensity of the bulk states close to Fermi level is well suppressed under the *s*-polarization geometry. We deduce that the dx^2-y^2 , dz^2 and dxz orbitals of Hf make the main contribution to the spectrum intensity of the body region with *p*-polarized light. Instead, the data acquired with *s*-polarized light [Fig.1 (b)(ii)] clearly indicates the ARPES appreciable intensity at the

two terminals along the $(\bar{\Gamma}\bar{X})$ line of the olive-shaped electron pocket, while the corresponding intensity is strongly suppressed for the p -polarized light.

As is expected, the M-shaped SS1 with the even initial states also show almost vanishing spectral weight under the s -polarized light, which is in accordance with the behavior of the Rashba-like surface states along the $\bar{M}\bar{X}$ line, revealing the real symmetry characteristic of the initial states.

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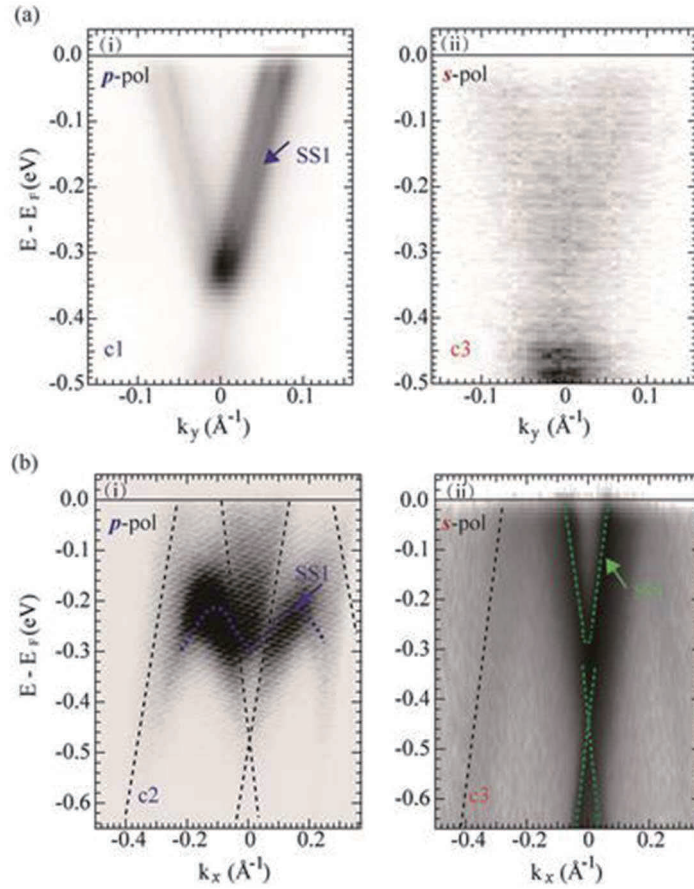


FIG. 1 (a) and (b) Experimental electronic structures of HfSiS recorded by ARPES at $h\nu=60$ eV with p - (i) and s -polarized light (ii) along $\bar{M}\bar{X}$ line and $\bar{\Gamma}\bar{X}$ line of the surface BZ, respectively. The black and green dot lines are guided for the eyes to trace the bulk and surface bands. Notations c1, c2 and c3 correspond to the measured ARPES band dispersions.

Superconductivity and Fermi-surface nesting in the candidate Dirac semimetal NbC

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Keywords: superconductivity, Fermi surface nesting, NbC, ARPES, first-principles calculations

Transition metal carbides (TMCs) are a large family of materials that have held industrial research interest for more than 100 years. In TMCs, carbon atoms are incorporated into the interstitial sites of the parent metals and form strong metal-carbon bonds [1–3]. This unique structure gives TMCs coexisting ionic, covalent, and metallic bonds, leading to metallic conductivity combined with excellent mechanical properties, including extreme hardness, high melting points, excellent corrosion resistance, and interesting catalysis properties [1–3]. Therefore, the TMC family of materials has been widely used in a variety of areas, including cutting tools, energy storage devices, and supercapacitors.

One prototypical TMC material is niobium monocarbide (NbC), which crystallizes into a NaCl-type cubic structure. NbC has various outstanding aspects: its melting point is one of the highest among all solid materials, while its hardness (in the 9–10 range on the Mohs scale) is comparable to that of diamond. More interestingly, NbC is a superconductor and has a T_c of approximately 11 K, which is higher than that of niobium. Recent theoretical studies have predicted Fermi-surface nesting in NbC, which will enhance the electron-phonon interactions in the material. Fermi-surface nesting is a subject of intense interest in studies of high-temperature superconductivity and charge/spin density waves. However, the biggest obstacle to the study of NbC is the challenge of synthesizing single-crystal samples. To date, only NbC polycrystals have been synthesized and this has strongly limited further experimental studies. Therefore, only a few experimental works on NbC and NbC_x ($0 < x < 1$) have been performed, although the material was discovered more than half a century ago.

In this work, we have successfully synthesized single-crystal NbC. Transport and magnetic susceptibility measurements confirmed that single-crystal NbC is a type-II superconductor that has a T_c of 11.5 K. Angle-resolved photoemission spectroscopy (ARPES) measurements provided evidence of Fermi-surface nesting, analogous to some iron-based superconductors and charge density wave systems. First-principles calculations showed that NbC hosts type-II Dirac cones near the Fermi level, which qualitatively agrees with our ARPES measurements. We also studied the superconducting gap using high-resolution laser ARPES, and, interestingly, we found that the behavior of the gap deviates from that of conventional Bardeen-Cooper-Schrieffer (BCS) theory.

The crystal structure, Brillouin zone, and XRD characterization of NbC single crystals are shown in Fig. 1(a)-(c), which indicates the high quality of the crystals. From the DFT calculation results in Fig. 1(d) and (e), we found that there is a Type-II Dirac point in NbC, as indicated by the blue cycle.

We then performed ARPES measurements to study the electronic structures of NbC, as shown in Fig. 2. First, we observed strong Fermi surface nesting effects, as indicated by the red arrows in Fig. 2(a). The experimental Fermi surface agree well with the DFT calculation results. Second, we confirmed the existence of Dirac cones and topological surface states. These results show that NbC is an ideal material for exploring topological superconductivity and Majorana fermions.

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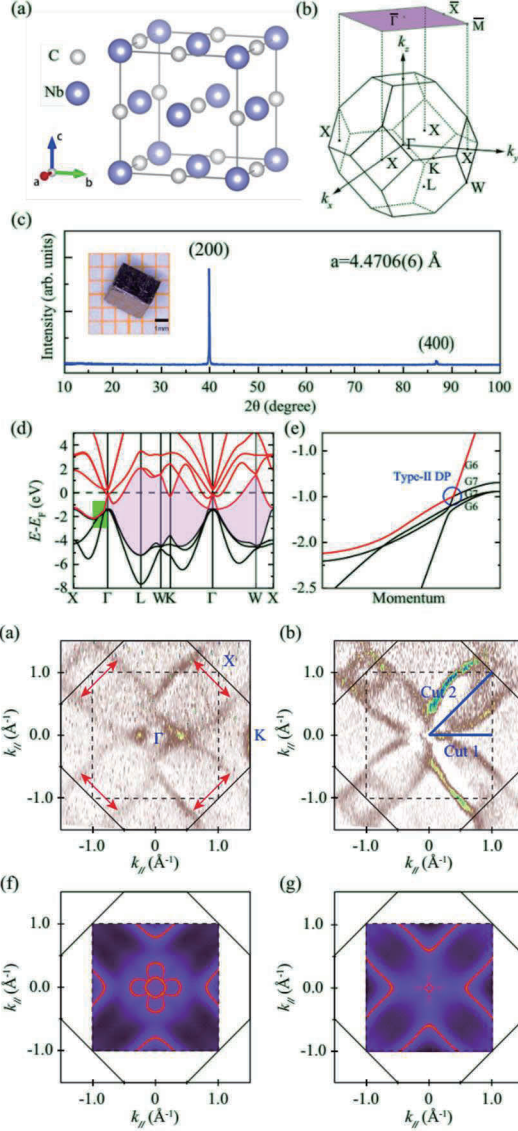


FIGURE 1. Growth of single-crystal NbC and calculations on its electronic structures. (a),(b) Schematic illustrations of the atomic structure and (001)-projected BZ of NbC, respectively. (c) X-ray diffraction pattern of the flat surface of the NbC single crystal. The inset shows a photograph of a typical NbC single crystal. (d) Calculated band structure of NbC, showing that a gap (gray shaded) exists between the black-colored bands and the red-colored bands, with the exception of a gapless Dirac point along X. (e) Magnified view of the band structure in the green shaded region of (d), which indicates the existence of a type-II Dirac cone. This cone is formed by irreducible representations of G6 and G7 under the double group of the C_{4v} symmetry on the line.

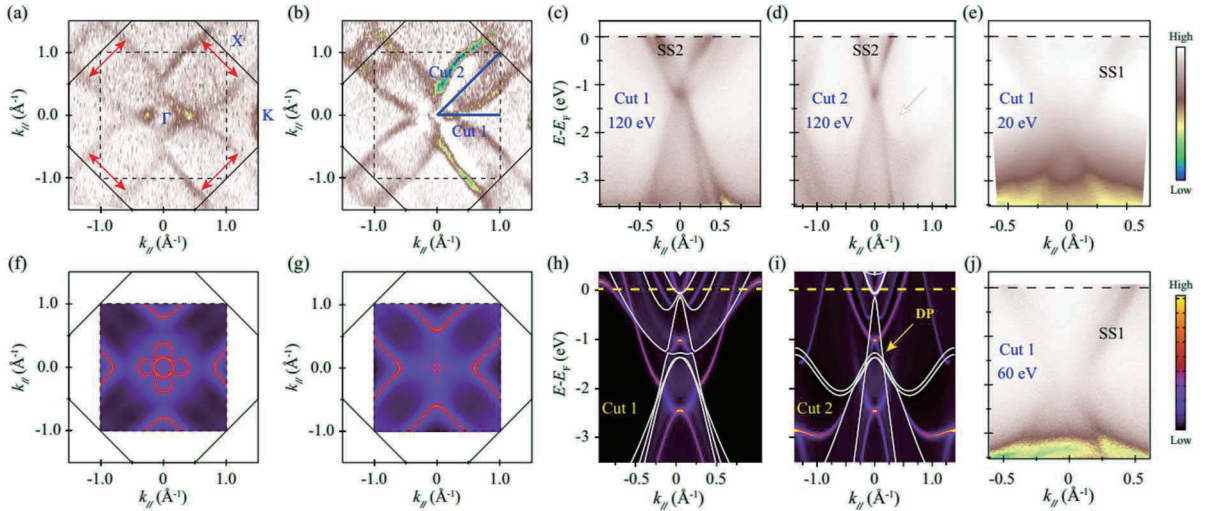


FIGURE 2. Synchrotron ARPES measurements and theoretical calculations of NbC. (a) ARPES intensity map of the Fermi surface of NbC measured using 120 eV photons. Solid and dashed lines indicate the bulk and surface BZs, respectively. Red arrows indicate the possible nesting vectors. (b) ARPES intensity map of the constant energy contour (CEC) at $E_B = 0.6$ eV measured using 120 eV photons. (c),(d) ARPES intensity plots of band structures along K and X measured using 120 eV photons. (e),(j) ARPES intensity plots of band structures along K measured using 20 eV and 60 eV photons, respectively. (f),(g) Calculated bulk bands on the Fermi surface and CEC at $E_B = 0.6$ eV. (h),(i) Purple-yellow colored images are semi-infinite calculation results using the Green's function method along directions that are parallel to K and X, respectively. The surface states can be determined as sharp yellow lines. White lines are the calculated bulk bands along K and X. "SS1" and "SS2" indicate two surface states of NbC.

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Investigations on Angle-resolved Photoemission Spectroscopy and Pressure-induced Transport Properties of BiSbTe₃ Topological Insulator

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Keywords: Topological Insulator, Angle-resolved Photoemission Spectroscopy (ARPES), Pressure-induced Superconductivity

Structural, pressure-dependent resistivity and angle resolved photoemission spectroscopy (ARPES) have been investigated for BiSbTe₃. It has been demonstrated that the Dirac point of the topological surface state (TSS) located exactly at the Fermi level. Additionally, superconductivity emerges under pressure of 8 GPa with a critical temperature of ~ 2.5 K. With further increase of pressure, the superconducting transition temperature (T_c) increases and at 14 GPa it shows the maximum T_c (~ 3.3 K). The investigation indicates that the BiSbTe₃ has robust surface states and becomes superconductor under pressure.

In case of n-type Bi₂Te₃, the Fermi level (E_F) lies in the bulk conduction band (BCB). On the other hand, the E_F of the Sb₂Te₃ TI lies in the bulk valence band (BVB) locating the Dirac point (DP) above E_F . The position of the DP and E_F may be tuned simultaneously by mixing of these two compounds. This may lead to an ideal TI in which E_F and the DP coincides in the insulating bulk band gap, which paves the way to apply topological surface states for future devices. The reported stoichiometric of Bi/Sb for the transition from n to p-type in (Bi_{1-x}Sb_x)₂Te₃ is debatable [1–5].

In this work, we have successfully synthesized a single crystal of BiSbTe₃, and found emergence of the superconducting states under pressure. We have revealed that the DP of topological surface state (TSS) is located at E_F and observed the hole-like bulk conductivity due to crossing of BVB to the E_F .

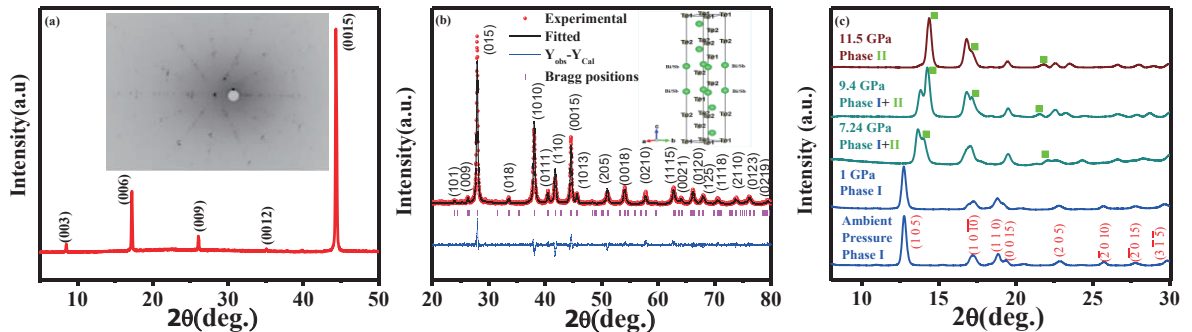


FIGURE 1. (a) Single crystal X-ray diffraction of BiSbTe₃ cleaved along (001) direction, Inset: Laue diffraction pattern (b) Le Bail refinement of powder XRD of as grown BiSbTe₃ single crystal sample using Full Prof software. (c) X-ray diffraction patterns of BiSbTe₃ at various pressures with wavelength 0.7101 Å. The peaks indicated with solid green square are the reflections from high pressure phase.

In order to see the pressure dependency on the structure of the investigated system we have performed XRD experiment with pressure variation using diamond anvil cells. Fig. 1 (c) illustrates the diffraction

pattern of BiSbTe₃ polycrystalline sample under various pressure at room temperature. It is clear from Fig. 1 (c) that evaluation of secondary phase has been started from 7.24 GPa and persisted up to 11.5 GPa. But at 11.5 GP monoclinic phase (secondary Phase) is dominating over rhombohedral phase which is consistent with reported results [6]. Therefore, rhombohedral $R\bar{3}m$ phase is robust up to 9.5 GP as observed from pressure dependent XRD.

In Fig. 2 (a), we can see the variation of the electrical resistance (R) at various pressures, where R is decreased between 2 GPa to 6 GPa. However, no sharp transition in the resistance was observed up to the lowest measured temperature (~ 2 K) (inset: Fig. 2(a)). When pressure was further increased to 8 GPa, a sharp decrease in resistance value is observed with $T_{\text{onset}} = 2.47$ K. We assume that this sharp decrease in the resistance at a particular temperature above 8 GPa can be assigned for the superconducting transition. The superconducting transition is getting more pronounced with increasing pressure and T_{onset} is also shifting toward higher value with applied pressure. The highest T_{onset} (3.36 K) was observed at 14 GPa, above which it becomes almost constant up to 15 GPa.

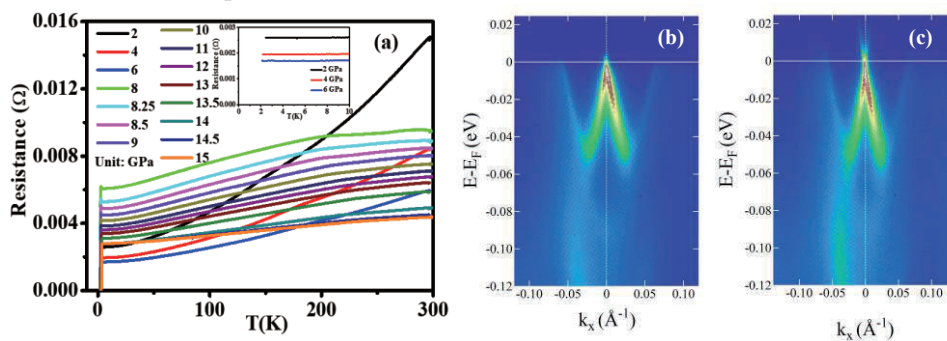


FIGURE 2. (a) Temperature dependent of resistance at different pressures with a superconducting transition around 8 GPa. Inset: Variation of resistance with respect to temperature at 2, 4 and 6 GPa. Band structure of the BiSbTe₃ single crystal measured at (b) $T=20$ K and (c) 60 K.

Figure 2(b) and (c) represent the ARPES spectra taken at 20 K and 60 K, respectively. These images show the BVB of the BiSbTe₃ forming a V-shaped valley like bulk band gap region around the Γ -point in the Brillouin zone and the topological surface state (TSS) residing inside it as observed earlier [7-8]. The sharpness of the linear dispersion of the TSS reveals good quality of the sample as well as the high energy and momentum resolution of the laser ARPES spectra. Note that one can discern the unoccupied part of the Dirac cone (DC) related to the TSS above E_F at 60K due to the thermal broadening of the Fermi distribution function.

In conclusion, we have investigated the external pressure effect on the resistance and the ARPES on BiSbTe₃. It is observed that with increase of pressure resistance decreases and at a value of 8 GPa a sharp drop in resistance is observed which indicates the occurrence of superconductivity. With further increase of pressure, the superconducting transition temperature (T_c) increases and at 14 GPa it shows the maximum T_c (~ 3.3 K). This may indicate that the superconducting transition might be due to the change in the bulk band structure in BiSbTe₃. Furthermore, the ARPES study clearly indicates the Dirac point is located at the Fermi level. This is an ideal condition for the technological exploitation of the topological properties of the TSS.

(#These authors contributed equally to this work. This work has been published in *Physica Scripta* **96**, 055802 (2021))

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Study of the band structure of LaAgSb₂ by angle-resolved photoemission spectroscopy

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Keywords: a square lattice, Dirac electrons, linear magnetoresistance

Dirac electrons form a cone-shaped band structure (Dirac cone) near the Fermi level, which appears on the surface of topological insulators and graphene [1-2], and is known to cause various interesting quantum phenomena. Recently, first-principles calculations have shown that LaAgSb₂, a type of layered rare-earth silver antimonide with a square lattice, forms a Dirac cone due to the intersection of linear bands derived from Sb 5p orbitals. LaAgSb₂ exhibits interesting magnetotransport properties such as very large linear magnetoresistance due to its characteristic electronic state [3].

To explore the band structures of LaAgSb₂, we performed an angle resolved photoelectron spectroscopy (ARPES) at BL1 of HiSOR. Figure 1 shows the experimental band structure along the $\bar{\Gamma}\bar{X}$ line of LaAgSb₂ acquired with *p*-polarized at a photon energy ($h\nu$) of 60 eV and a temperature of 30 K. Here, the vertical axis is the energy (E) from the Fermi level (E_F) and the horizontal axis is the wavenumber ($\text{\AA}^{-1}$). As indicated by the color bar in the upper right corner, the darker color shows the stronger photoelectron intensity. Focusing on the vicinity of the $\bar{\Gamma}$ point at wavenumber $0 \text{ \AA}^{-1}$, we observe an upwardly convex linear band crossing E_F and a downwardly convex band with apex at $E-E_F = -0.8 \text{ eV}$. The two bands intersect at $E-E_F = -0.6 \text{ eV}$, indicating that they are linearly symmetric around wavenumber $0 \text{ \AA}^{-1}$. Focusing on the vicinity of the second Γ point at wavenumber $\pm 1.4 \text{ \AA}^{-1}$, we can see that the upwardly convex band observed near the first $\bar{\Gamma}$ point at wavenumber $0 \text{ \AA}^{-1}$ appears repeatedly. Focusing on wavenumber $-0.9 \text{ \AA}^{-1}$ and $E-E_F = -0.2 \text{ eV}$, we can see that there is a small upward convex band that is differentiated from the upward convex band at the second $\bar{\Gamma}$ point at $-1.4 \text{ \AA}^{-1}$. This band is linear from -0.9 to $-0.7 \text{ \AA}^{-1}$. This linear band around the X point is also observed at wavenumbers from 0.7 to $0.9 \text{ \AA}^{-1}$. The upwardly convex band near the $\bar{\Gamma}$ point is observed with greater intensity at lower wavenumbers, and the band is clearly dense. This can be attributed to the effect of the transition probability of the photoelectron intensity.

The upward convex band dispersions around the first and second $\bar{\Gamma}$ points observed in the experiment are well reproduced by the first-principles calculation. The downward convex band dispersion of the first $\bar{\Gamma}$ point, the small upwardly convex band dispersion observed at wavenumbers -0.9 and $-0.5 \text{ \AA}^{-1}$ and $E-E_F = -0.2 \text{ eV}$ are also consistent with the first-principles calculation.

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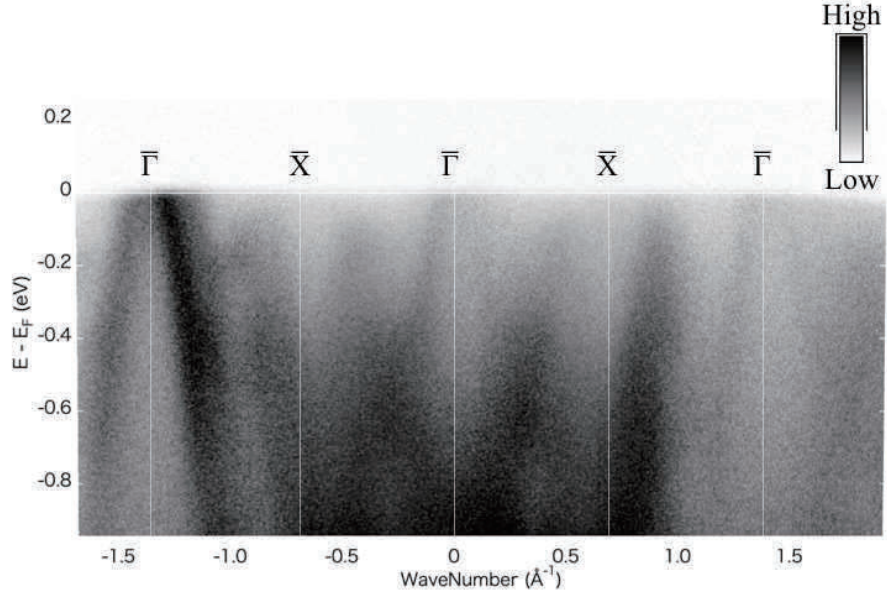


FIG. 1 ARPES band dispersions of LaAgSb₂ acquired with *p*-polarized at a photon energy of 60 eV.

Self-energy Analysis of High- T_c Cuprate Superconductor $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$

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Keywords: cuprate, ARPES.

For uncovering high-temperature superconductivity in cuprates, many-body interactions have been investigated by angle-resolved photoemission spectroscopy (ARPES) [1]. Yet, it had been challenging to obtain high-quality ARPES spectra from three-dimensional crystal systems such as $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (LSCO) due to the roughness of a cleavage plane. On the other hand, in recent years, spectral quality has been improved as developing experimental equipment and performance, enabling us to obtain fine electronic states and study many-body interactions even in three-dimensional systems. Indeed, a recent ARPES study on an overdoped LSCO ($x=0.23$) revealed quasiparticle scattering rate $\Gamma(\omega)=2\text{Im}\Sigma(\omega)$ as expressed by using the imaginary part of the self-energy and claimed its energy dependence deviates from the Fermi liquid behavior [2]. However, since the analysis of the scattering rate is based on only electron-electron interaction, it is necessary to consider other many-body interactions, such as electron-boson interaction (boson: phonon or magnon, etc.) dominating the quasiparticle scattering in the vicinity of the Fermi level. In this work, we have performed a high-resolution ARPES study on an optimally doped LSCO to understand the role of many-body interactions in this system.

High-quality single crystals of optimally doped LSCO ($x=0.155$, $T_c \sim 39$ K) were prepared by the traveling-solvent floating-zone technique [3]. ARPES experiments were measured at the linear undulator beamline (BL-1) of Hiroshima Synchrotron Radiation Center (HiSOR) using the R4000 electron analyzer (Scienta Omicron) [4]. Clean surfaces of the samples were obtained by cleaving *in situ* under ultra-high-vacuum conditions (4×10^{-9} Pa) at a low temperature (~ 27 K). Present ARPES data were taken with the photon energy of 70 eV at 27 K in the *s*-polarization geometry.

Figure 1(a) shows the ARPES image of the optimally doped LSCO taken along the nodal direction, where the circles represent the band dispersions determined by fitting momentum distribution curves (MDCs). To evaluate the many-body interactions, we compared the ARPES dispersion with the calculated dispersion by the tight-binding (TB) model [5], as shown in Fig. 1(a). At first, the TB model dispersion (dashed line) was calculated using TB parameters were first determined to reproduce the band-structure calculations within the local density approximation (LDA). However, we found that the extracted real and imaginary parts of the self-energy do not satisfy the causality, *i.e.*, Kramers-Kronig relation. Thus, we adjusted the TB parameters to satisfy the causality, and the resulting TB model dispersion (solid line) was employed as a non-interacting bare band dispersion in this work. Compared with the TB model dispersion, we found that the experimental dispersions show a slightly larger effective mass in the vicinity of the Fermi level while a smaller one in the higher energy region. Notably, this two-distinct behavior can be characterized by the crossing point between the ARPES and TB model dispersion. At this point, the real and imaginary parts of the self-energy become zero and minimum, respectively. This characteristic behavior is a signature of the high-energy anomaly, widely reported in strongly correlated electron systems, including cuprates [6].

The renormalization effects can be more clearly seen in the extracted real and imaginary parts of the self-energy, as shown in Figs. 1(c) and 1(d), respectively. We found that the imaginary part of the self-energy

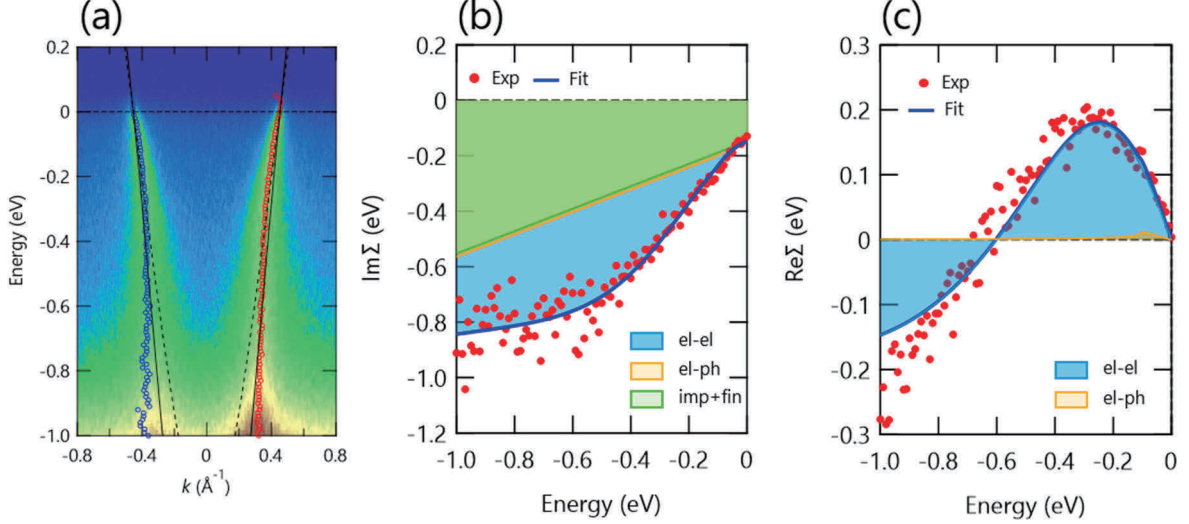


FIGURE 1. (a) APRES image of optimally doped LSCO along the nodal direction. The red and blue circles represent the MDC-derived band dispersion. The dashed and solid lines are the tight-binding (TB) model dispersions based on the different parameters: the parameters were set to reproduce the LDA calculation for the former dispersion, while they were adjusted for the latter dispersion to satisfy the self-consistency between the extracted real and imaginary parts of the self-energy. (b) and (c) Real and imaginary parts of the self-energy, respectively. The imaginary part of the self-energy was fitted with the model self-energy including the contributions from the electron-electron interaction (el-el), the electron-phonon interaction (el-ph), and the impurity scattering as well as the final state effects (imp+fin). Each contribution is indicated by a shaded area with a different color. Note that the TB model dispersion using the adjusted parameters was employed as the bare band for extracting the self-energy.

was well reproduced by the model self-energy assuming the electron-electron interaction ($\text{Im}\Sigma_{\text{el-el}}$) [6], electron-phonon interaction ($\text{Im}\Sigma_{\text{el-ph}}$) [7], and impurity scattering as well as final state effects ($\text{Im}\Sigma_{\text{imp+fin}}$) [8]. Notably, the real part of the model self-energy, obtained via Kramers-Kronig transformation, agrees well with the experimental self-energy. The result thus is validating the assumption of the bare band and quantitative strength of the extracted self-energy. As clearly seen in Fig. 1(b), the imaginary part of the self-energy is dominated mainly by two scattering channels, $\text{Im}\Sigma_{\text{el-el}}$ and $\text{Im}\Sigma_{\text{imp+fin}}$. The significant contribution of the final state effects, giving the energy dependence of $\text{Im}\Sigma_{\text{imp+fin}}$, would be related to a relatively strong three-dimensionality in LSCO. In contrast, the real part of the self-energy is composed of $\text{Re}\Sigma_{\text{el-el}}$ and $\text{Re}\Sigma_{\text{el-ph}}$, as shown in Fig. 1(c), because $\text{Re}\Sigma_{\text{imp+fin}}$ gives zero contribution. The peak maximum of the real part of the self-energy is located at around 0.3 eV, which agrees well with one of the antiferromagnetic spin fluctuations reported by resonant inelastic X-ray scattering (RIXS) measurements [9]. Our result thus indicates that the antiferromagnetic spin fluctuations play an important role in the electronic band renormalization effects in LSCO. Our analysis presented here also allows evaluating the coupling strength of the electron-electron interaction and electron-phonon interaction as $\lambda_{\text{el-el}}=1.2$ and $\lambda_{\text{el-ph}}=0.6$, respectively. It should be emphasized here that they are underestimated or overestimated in the previous works [2, 10]: $\lambda_{\text{el-el}}<0.3$ (experiment), $\lambda_{\text{el-ph}}=1.0-1.3$ (experiment), and $\lambda_{\text{el-ph}}=0.1-0.2$ (theory). The discrepancy between our and previous results must be originated from that only one interaction (el-el or el-ph) was only considered in earlier works. Consequently, our results indicate the importance of considering all the interactions to evaluate the many-body interactions quantitatively.

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Strain Induced Metal-Insulator Transition in Mott Insulator Ca_2RuO_4

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Keywords: Mot insulator, Metal-insulator transition, ARPES

One of the representative Mott-insulators, ruthenate Ca_2RuO_4 , exhibits a dramatic change in its physical properties due to external fields such as temperature, pressure, electric or magnetic field, and element substitution [1-2]. Recently, it has been reported that a weak electric field corresponding to a battery can induce metal-insulator transition and giant anti-magnetism in Ca_2RuO_4 [3], attracted much interest in terms of low electric switching devices. To understand such intriguing physical properties, it is essential to study the electronic structure of Ca_2RuO_4 . In particular, it is most straightforward to observe the electric-field-induced change of the electronic structures using high-resolution ARPES. However, since the electric field may bring unexpected electronic behavior, we have decided to examine the temperature-induced metal-insulator transition of Ca_2RuO_4 at high temperature (~ 350 K) using high-resolution ARPES in this proposal.

High-quality single crystals of Ca_2RuO_4 were prepared by traveling-solvent floating-zone method with RuO_2 self-flux [3]. ARPES experiments were measured at BL-1 and BL-9A of Hiroshima Synchrotron Radiation Center (HiSOR) using an R4000 electron analyzer (Scienta Omicron). Clean surfaces of the samples were obtained by cleaving *in situ* under ultra-high-vacuum conditions ($\sim 10^{-10}$ Torr) at a high temperature (~ 360 K).

Unfortunately, we could not observe clear band dispersion of Ca_2RuO_4 under various experimental conditions (photon energies and polarizations) despite many trials (cleaves). Since the core levels observed in angle-integrated spectra can be well explained by the constitute of Ca_2RuO_4 , we speculate that we were unable to observe the band dispersion due to the small size of samples or surface quality. We will improve the size and/or quality of samples for future studies on this material.

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Experimental observation of Dirac cones in artificial graphene lattices

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Keywords: artificial graphene lattice, ARPES, Dirac cone, Shockley surface state

Graphene is a single layer of carbon atoms with a honeycomb lattice, and it has been intensively studied in the past decade [1,2]. In proximity to the Fermi level, the electrons in graphene behave as massless Dirac fermions. This is the origin of graphene's various exotic properties, such as half-integer quantum Hall effects and the Klein paradox. An alternative way to realize the exotic properties of graphene is by confining the electrons of a two-dimensional electron gas (2DEG) to an equivalent honeycomb lattice, called artificial graphene (AG) [3-5]. Because of the confinement, there exists a Dirac cone at each K point of the Brillouin zone (BZ). As a result, electrons of the otherwise 2DEG behave as massless Dirac fermions, which is analogous to the case of natural graphene. In addition to possessing the novel properties of graphene, artificial lattices possess various tunable parameters, thereby providing an ideal platform for the simulation of quantum behaviors in two-dimensional (2D) Dirac materials [6-9].

Some molecules, such as carbon monoxide and coronene, can serve as potential barriers through which the electrons of the 2DEG are forbidden to travel. Therefore, a hexagonally patterned molecular lattice can confine the 2DEG electrons into an equivalent honeycomb lattice and lead to the realization of AG [3,4], as illustrated in Figure 1. The high energy and momentum resolution of angle-resolved photoemission spectroscopy (ARPES) makes it a powerful technique to directly study the electronic structures of materials. However, a direct experimental observation of the Dirac cones in AG using ARPES has yet not to be reported.

Here, we constructed macroscopic AG systems by growing monolayer C₆₀ molecules on the (111)-terminated surfaces of noble metals, which enabled ARPES studies of the electronic band structure. Our low-energy electron diffraction (LEED) and ARPES measurements show that these AG systems are homogeneous. Therefore, their size is only limited by the scale of the substrates. The Dirac cones at the K (K') points were directly observed by our ARPES measurements. In addition, we performed model calculations on these AG systems, whose results fully supported our experimental observations.

First, we studied C₆₀ monolayers on Cu(111). The C₆₀ molecules form a hexagonal structure with a 4×4 superstructure with respect to the 1×1 lattice of Cu(111) as shown in Fig. S1[10]. Figure 2(b) shows the ARPES intensity of the Fermi surface. The Shockley surface state of Cu(111) almost disappears because of the coverage of the C₆₀ molecules. Instead, a dotlike spectral weight can be seen at each K(K') point. Such features do not exist on pristine Cu(111) because pristine Cu(111) only exhibits Shockley surface states at the BZ center and bulk *sp* bands at the BZ boundary. At deeper binding energies, the dots become circles and their size increases as the binding energies increase, as shown in Figs. 2(c)-2(e).

Figure 2(f) shows the ARPES spectra along the Γ -K direction. One can observe linearly dispersing

bands at the K point, as indicated by the red dashed lines. The fitted crossing point is located approximately 0.1 eV above the Fermi level. Along the K - M - K direction, the dispersion of the bands is also linear, as shown in Fig. 2(g). Combined with the evolution of the constant energy contours, we can conclude that there is a Dirac cone at each K point. In addition, the Dirac bands do not disperse with different photon energies, as shown in Figs. 2(h)-2(j), which agrees with their 2D character. Therefore, the ARPES results confirm that the $C_{60}/Cu(111)$ system is an AG with Dirac cones at the $K(K')$ points of the BZ.

Another prototypical 2DEG system is Au(111), on which C_{60} molecules form a $2\sqrt{3} \times 2\sqrt{3}$ superstructure with respect to the 1×1 lattice of Au(111) as shown in Fig. S1[10]. This different superstructure provides further evidence for the universality of this method. As expected, our ARPES results show the existence of Dirac cones at each $K(K')$ point of the BZ, as shown in Figs. 4(b) and 4(c).

In summary, we demonstrate the synthesis of macroscopic AG by the self-assembly of C_{60} molecules on metal surfaces. Our theoretical calculations and ARPES measurements directly confirm the existence of Dirac cones at the $K(K')$ points of the Brillouin zone.

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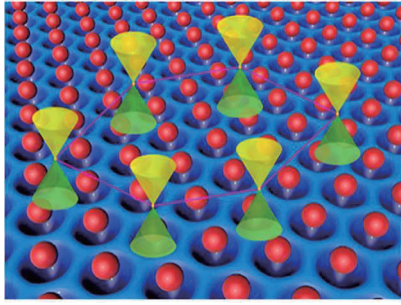


FIGURE 1. Schematic of artificial graphene (AG). Molecular AG system, including molecules that act as potential barriers for electrons (red balls) and the electron density profile (blue surfaces). Band structure of AG. A Dirac cone exists at the $K(K')$ point of the Brillouin zone (BZ), as an analogy to graphene.

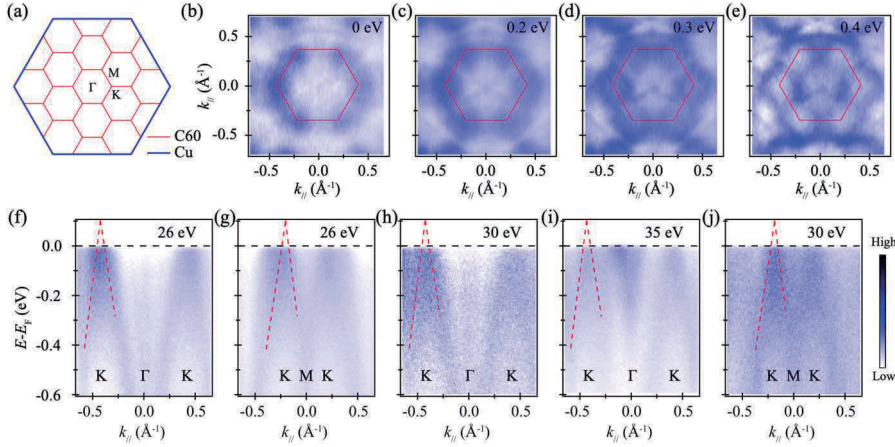


FIGURE 2. ARPES measurements of the C_{60} monolayer on Cu(111). (a) Schematic of the BZs of the C_{60} monolayer (red) and Cu(111) (blue). (b)–(e) ARPES intensity maps of the constant energy contours at different binding energies. (f)–(j) ARPES intensity plots along high-symmetry directions with different photon energy. From Ref. 10.

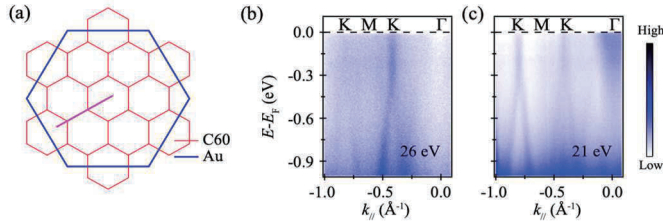


FIGURE 3. ARPES measurements of the C_{60} monolayer on Au(111). (a) Schematic of the BZs of the C_{60} monolayer (red) and Au(111) (blue). (b), (c) ARPES intensity plots along the K - M - K direction. From Ref. 10.

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Topological electronic structure in the antiferromagnet HoSbTe

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Topological quantum materials, such as topological insulators and semimetals, have attracted much attention in the last decade. These materials host topologically nontrivial electronic structures that can be characterized by symmetry protected topological invariants [1-3]. Among the topological quantum materials, Dirac nodal line (DNL) semimetals are of particular interest because they are neighbors of various quantum states. In a DNL semimetal, the conduction and valence bands touch at extended lines in the three-dimensional Brillouin zone (BZ), and the degeneracy of the line nodes is protected by certain symmetries, such as mirror reflection, time-reversal, and spin-rotation symmetries. The breaking of these symmetries can fully or partially gap out the nodal lines, giving rise to rich topological quantum states, such as Dirac semimetals, Weyl semimetals, and topological insulators.

Magnetic topological materials, in which the time-reversal symmetry is broken, host various exotic quantum phenomena, including the quantum anomalous Hall effect, axion insulator states, and Majorana fermions. The study of magnetic topological materials is at the forefront of condensed matter physics. Recently, a variety of magnetic topological materials have been reported, such as Mn_3Sn , $\text{Co}_3\text{Sn}_2\text{S}_2$, and MnBi_2Te_4 [4-6]. Recently, single-crystal HoSbTe, a new member of the ZrSiS (or LnSbTe) family, was synthesized [7]. Because of the unpaired $4f$ electrons of Ho atoms, HoSbTe is an antiferromagnetic (AFM) material with a T_N of approximately 4 K.

Here, we performed angle-resolved photoemission spectroscopy (ARPES) and first-principles calculations to study the electronic structure and topological properties of HoSbTe. Firstly, we performed theoretical calculation. A DNL exists at the $k_z = 0$ and π planes, and the degeneracy of the line nodes is protected by the glide mirror symmetry of the crystal. The degenerate points along the high-symmetry directions are indicated by the red dotted circles in Fig. 1(c). When SOC is taken into account, the DNLs of HoSbTe are fully gapped, as shown in Fig. 1(d). Figures 1(e)-1(h) show the calculated SOC gaps of HoSbTe. One can see that the gap reaches 200 meV along the Z-R direction, which is much larger than the thermal excitation energy. Such a large gap makes the realization of the WTI states in HoSbTe possible in the experimentally accessible regime.

To confirm the intriguing electronic structure of HoSbTe, we performed high-resolution ARPES measurements on the (001) surface of HoSbTe. Figures 2(a)-2(d) and 2(e)-2(h) show the CECs of HoSbTe measured with 55 and 120 eV photons, which corresponds to $k_z = 12.3$ and $17.2 \pi/c$, respectively. Overall,

the data at 55 and 120 eV are quite similar, despite the subtle differences such as the positions of the corners and intensities at the BZ boundary. Along the $\bar{\Gamma}-\bar{M}$ direction, a parabolic band occurs that originates from the gapped DNL, as shown in Figs. 3(a)-3(c).

In summary, we report the observation of topological electronic structures in the antiferromagnetic material HoSbTe. When SOC is neglected, HoSbTe is a DNL semimetal, and its electronic structure is analogous to that of ZrSiS. However, the intrinsic strong SOC fully gaps out the nodal lines and drives the system to be a WTI. Each layer of HoSbTe is a 2D TI, and the bulk crystal can be viewed as a stacking of 2D TIs. The intriguing electronic structure of HoSbTe has been directly confirmed by ARPES measurements. Notably, the SOC gap of HoSbTe is as large as hundreds of meV, which is accessible for most experimental techniques.

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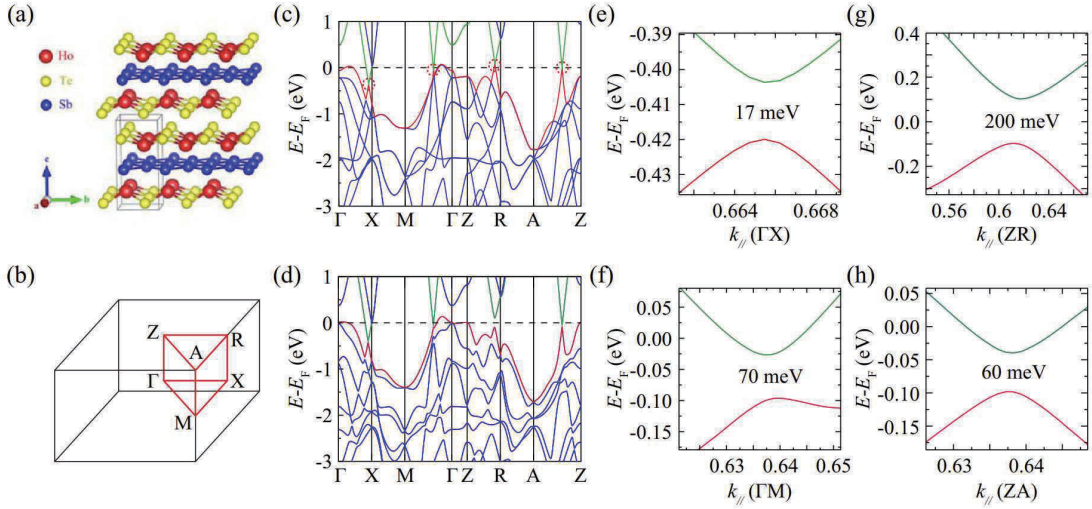


FIGURE 1. (a) Crystal structure of HoSbTe. (b) Brillouin zone of HoSbTe. (c), (d) Calculated bulk bands of HoSbTe along high-symmetry directions without and with SOC, respectively. Red and green lines in (c) highlight the highest valence and lowest conduction bands that form Dirac nodal lines. The red dotted circles in (c) indicate the degenerate points that form DNLs. (e)–(h) Calculated SOC-induced band gaps at the line nodes along high-symmetry directions: -X, -M, Z-R, and Z-A. The sizes of the gaps are indicated in the figure. From Ref. 8.

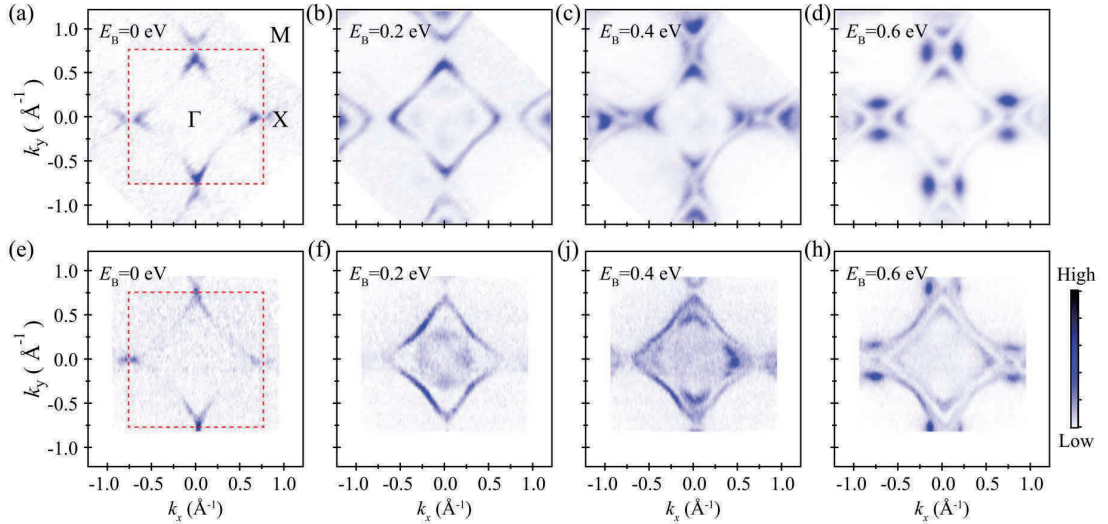


FIGURE 2. ARPES intensity plots of the constant energy contours (CECs) of HoSbTe(001). (a)–(d) CECs measured with 55 eV photons. (e)–(h) CECs measured with 120 eV photons. From Ref. 8.

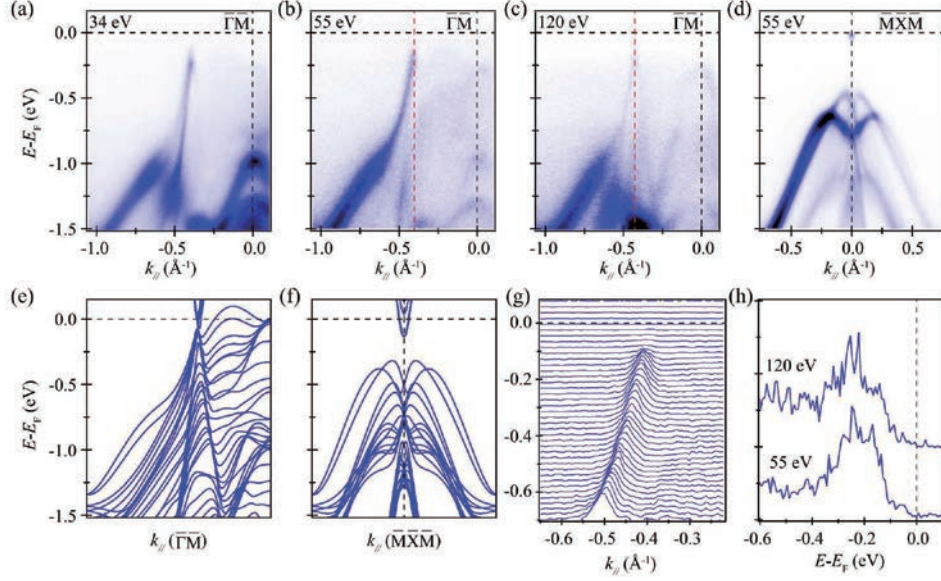


FIGURE 3. (a)–(c) ARPES intensity plots of the band structure of HoSbTe along the $\bar{\Gamma}-\bar{M}$ direction measured with different photon energies. (d) ARPES intensity plots of the band structures along the $\bar{M}-\bar{X}-\bar{M}$ direction. (e), (f) Slab calculation results of the band structure of HoSbTe along $\bar{\Gamma}-\bar{M}$ and $\bar{M}-\bar{X}-\bar{M}$, respectively. (g) Momentum distribution curve plots of (b). (h) Energy distribution curves along the red dashed lines in (b) and (c). From Ref. 8.

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Electronic structure of chiral IrGe₄ studied by angle-resolved photoemission spectroscopy

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Keywords: Kramers-Weyl Semimetal, Chiral material

In chiral materials with strong spin-orbit coupling, Bloch electrons exhibit two-fold Kramers degeneracy at time reversal invariant momentum (TRIM) points. The Kramers degeneracy is lifted except the TRIM points due to the spin-orbit interaction and the space inversion symmetry breaking of the chiral crystal. Therefore, the degenerate point is viewed as a Kramers-Weyl fermion [1,2]. Degree of the degeneracy can be larger than two when the spin-orbit interaction is negligible. CoSi with chiral crystal structure has been studied by means of band structure calculations and angle-resolved photoemission spectroscopy [3-5]. Although the spin-orbit interaction is relatively weak in CoSi, the observation of Kramers-Weyl fermions has been claimed [3-5]. There are many other chiral materials which can harbor the Kramers-Weyl degeneracy at TRIM. Among them, IrGe₄ has trigonal chiral structure just like α -quartz. Since IrGe₄ has no well-defined cleavage plane, it is highly difficult to perform ARPES on it. In the present proposal, we have challenged ARPES observation of the Kramers-Weyl degeneracy and possible surface states in IrGe₄.

ARPES on the single crystals of IrGe₄ were performed at BL-1 of HiSOR using a SCIENTA R4000 analyzer with linearly polarized light. The total energy resolution was set to about 40 meV for $h\nu = 58$ eV. The single crystals were oriented by ex situ XRD measurements. The crystals were cleaved at 20 K under the ultrahigh vacuum and subsequently measured at 20 K. The obtained surfaces were roughly perpendicular to the c-axis of IrGe₄.

Figure 1 shows a Fermi surface map taken at $h\nu = 58$ eV. ARPES intensity is enhanced along the M points of the surface Brillouin zone. This observation is qualitatively consistent with band structure calculations in which Ge 4p bands cross the Fermi level around the M points [6]. However, the observed Fermi surfaces were not clear probably due to surface roughness. Since there is no well defined cleavage plane perpendicular to the c-axis, the obtained surface was not really flat. We need to improve surface quality in order to observe Kramers-Weyl degeneracy in the bulk band dispersions and compare them with the prediction of the band structure calculations [6].

In addition to the bulk Fermi surfaces, we tried to observe helicoidal Fermi arcs which are expected at surfaces of chiral materials. However, we failed to observe surface bands and Fermi arcs in the surfaces obtained by cleaving. It is often claimed that Fermi arcs at surfaces of Weyl semimetals are topologically protected and very robust. However, such surfaces states are rather fragile against surface reconstruction, surface roughness, etc. We need to try various surface treatments to stabilize the Fermi arcs which may have interesting spin polarizations.

In conclusion, we have investigated the electronic structure of chiral IrGe₄ by means of ARPES. Although the observed Fermi surfaces are very broad due to surface roughness, the overall band structure is roughly consistent with the prediction of the band structure calculations [6]. In future, we need to improve surface treatment in order to obtain flat surfaces which can harbor Kramers-Weyl fermions and helicoidal Fermi arcs.

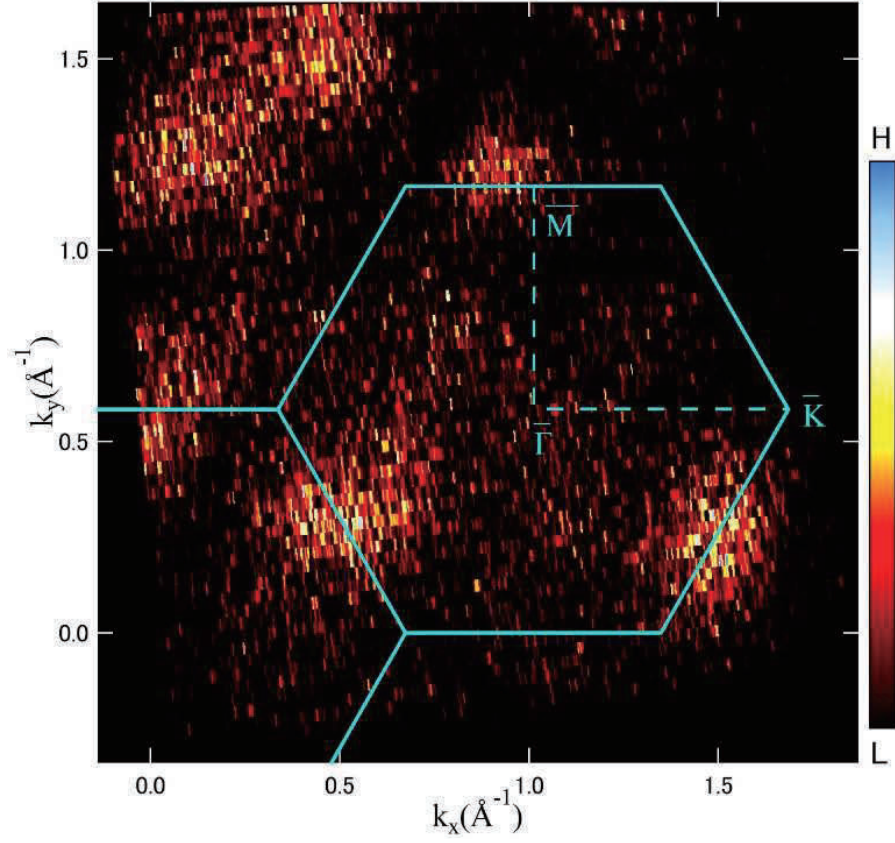


FIGURE 1. Fermi surface map observed at $h\nu = 58$ eV. The solid lines indicate the surface Brillouin zone boundaries.

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Observation of spin-momentum-layer locking in a centrosymmetric crystal

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The spin polarization in nonmagnetic materials is conventionally attributed to the outcome of spin-orbit coupling when the global inversion symmetry is broken. The recently discovered hidden spin polarization indicates that a specific atomic site asymmetry could also induce measurable spin polarization, leading to a paradigm shift in research on centrosymmetric crystals for potential spintronic applications. Here, combining spin- and angle-resolved photoemission spectroscopy and theoretical calculations, we report distinct spin-momentum-layer locking phenomena in a centrosymmetric, layered material, BiOI. The measured spin is highly polarized along the Brillouin zone boundary, while the same effect almost vanishes around the zone center due to its nonsymmorphic crystal structure. Our work not only demonstrates the existence of hidden spin polarization, but also uncovers the microscopic mechanism of spin, momentum, and layer locking to each other.

BiOI has a tetragonal crystal structure with a centrosymmetric space group $P4/nmm$ containing nonsymmorphic operations of a glide mirror $\{M_z | (\frac{1}{2}, \frac{1}{2}, 0)\}$ and two screw axes $\{C_{2x} | (\frac{1}{2}, 0, 0)\}$, $\{C_{2y} | (0, \frac{1}{2}, 0)\}$. The inversion center is located in the middle of two inequivalent O atoms (site point group D_{2d}), while the Bi and I atoms occupy the noncentrosymmetric polar sites with the site point group C_{4v} . The polyhedrons coordinated by Bi and I atoms are intersected by the O plane. Hence, the quasi-2D unit cell is divided into two sectors α and β , respectively, as shown in Fig. 1(a).

The BZ and DFT-calculated electronic structures of BiOI with SOC are shown in Fig. 1(b) and 1(c). The valence band maximum (VBM) is close to the X point. It is noticeable that at the points X and M, the glide reflection symmetry $\{M_z | (\frac{1}{2}, \frac{1}{2}, 0)\}$ anticommutes with the inversion operator, leading to an extra two-fold degeneracy between two pairs of Kramer's degeneracy, i.e., four-fold degeneracy including the spin. Such a four-fold degeneracy is maintained along the entire X-M line in the absence of SOC. Thus, the band splitting along the X-M line shown in Fig. 1(c) is caused by SOC solely. In analogy to the conventional Rashba/Dresselhaus effect, such a splitting is composed of two sets of spin splitting bands originating from the sectors α and β , respectively [1]. In comparison, the splitting along the Γ -X line is contributed by both of orbital repulsions and the SOC effect, and is thus larger than that along the X-M line. The orbital projection analysis shows that in the vicinity of the points Γ and X, the top two valence bands (designated as VB1 and VB2) are mainly composed of the $p_x + p_y$ and s orbitals, while VB3-VB6 are dominated by the p_z and s orbitals.

The ARPES results measured at a photon energy of 65 eV are shown in Fig. 1(d) [constant energy contours (CECs)], Fig. 1(e), 1(f) (band dispersions) and Fig. 1(g), 1(h) [energy-distribution curves (EDCs)]. A square-like CEC exists at -1.3 eV, with corners located at X points. As the energy decreases, the CEC features at the X point expand and eventually form contours surrounding the M point, merging with those centered at the Γ point. This hole-like behavior is presented in the ARPES spectra along the M-X-M line in Fig. 1(e). From the CECs and spectra results, we have found that the VBM is located around the bulk X point, ~ 1.4 eV below the experimental Fermi level.

By directly comparing the calculated bulk band structure with the ARPES data shown in Fig. 1(e) and 1(f), a good agreement is found, indicating that the surface effect that breaks the global inversion symmetry is relatively weak. The predicted four-fold degeneracy at the X and M points and the splitting two-fold

degenerate branches (VB1 to VB6) away from X and M are all supported by the ARPES measured dispersion. Furthermore, Fig. 1(g) and 1(h) show the EDCs measured along the M-X-M and X- Γ -X directions. At the X point, the degenerate peaks, *i.e.*, $X_{1,2}$, $X_{3,4}$ and $X_{5,6}$ are unambiguously present, while at the Γ point, each of the degenerate peak splits into two individual peaks, *i.e.*, Γ_1 to Γ_6 . Consequently, three pairs of Rashba-like hole-type valence bands are formed at the X and M points, with the band crossing points located around -1.4 eV, -2.1 eV, and -3.0 eV for the X point, respectively (Fig. 1(e)). These results agree well with our calculation, thus confirming the nonsymmorphic feature of the material.

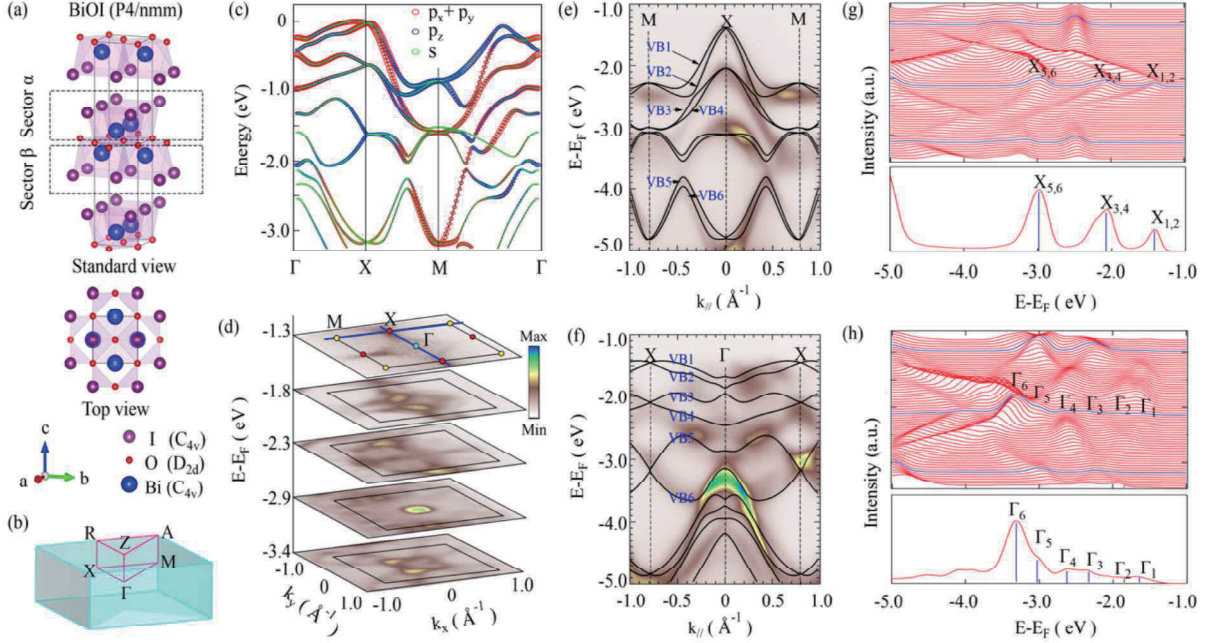


FIGURE 1. (a) The crystal structure of BiOI. The unit cell consists of two BiI layers as inversion partners, labelled as a sector α and β . (b) The bulk Brillouin zone. (c) DFT-calculated bulk band dispersion with orbital projection. (d) ARPES-measured CECs of the valence bands at different energies. (e, f) ARPES-measured spectra along the X-M, and Γ -X high symmetry lines, overlaid by DFT calculated dispersions (solid black lines). (g, h) EDCs corresponding to the spectra shown in (e) and (f), respectively. The lower parts are the EDCs at X and Γ points, respectively, from which one can resolve the spectral peaks corresponding to the top six valence bands.

Figure 2 presents the in-plane spin polarization of BiOI measured by spin-ARPES using photon energies of 65 eV for panels (a, b) and 30 eV for panels (c, d). The wide-ranging measurements involving different photon energies and geometries verified that we have essentially observed the spin polarization and spin textures derived from the initial state. The representative spin EDCs for the three pairs of two-fold degenerate bands VB1-VB6 are shown in Fig. 2(b) and 2(d), with the upper (lower) row showing the spin-resolved EDCs and the corresponding S_y (S_x) spin component. At three time-reversal invariant points M, X and Γ (momentum points ①, ③ and ⑧), the spin-resolved EDCs overlap, indicating negligible spin polarization; this is consistent with the spin degeneracy originating from Kramer's pairs.

When the momenta moved away from the X point, we have observed significant spin polarization (up to 80%) along both k_x and k_y directions (momentum points ②, ④, ⑤ and ⑥). For momenta ⑤ and ⑥, nearly all the six VBs were resolved as the individual polarizations peaked with opposing polarization signs in each pair. This is because the band splitting along Γ -X direction was more significant compared to the splitting along the X-M direction (Fig. 1(c), 1(e), 1(f)). In sharp contrast, the spin polarization surrounding the Γ point was very weak ($<30\%$ for momentum point ⑦), along both k_x and k_y directions.

The detected photoemission signal mainly arises from the topmost sector (α) of the cleaved BiOI single crystal, which is favourable to detect the spin polarization from a local sector. Compared with the previous measurements of HSP materials such as WSe₂ [3], PtSe₂ [4], LaO_{0.55}F_{0.45}BiS₂ [5] and Bi2212 [6] that focus on the spin-momentum locking around a single high-symmetry point, our work revealed the distinct polarization features surrounding different high-symmetry points, *i.e.*, BZ center (Γ) and BZ boundary (X), and observed a sharp contrast between them. Such observations suggest that momentum-dependent spin polarization originates from the HSP rather than merely from the surface potential gradient; further, these observations suggest the key factors affecting the HSP effect, such as the nonsymmorphic symmetry and

orbital characters.

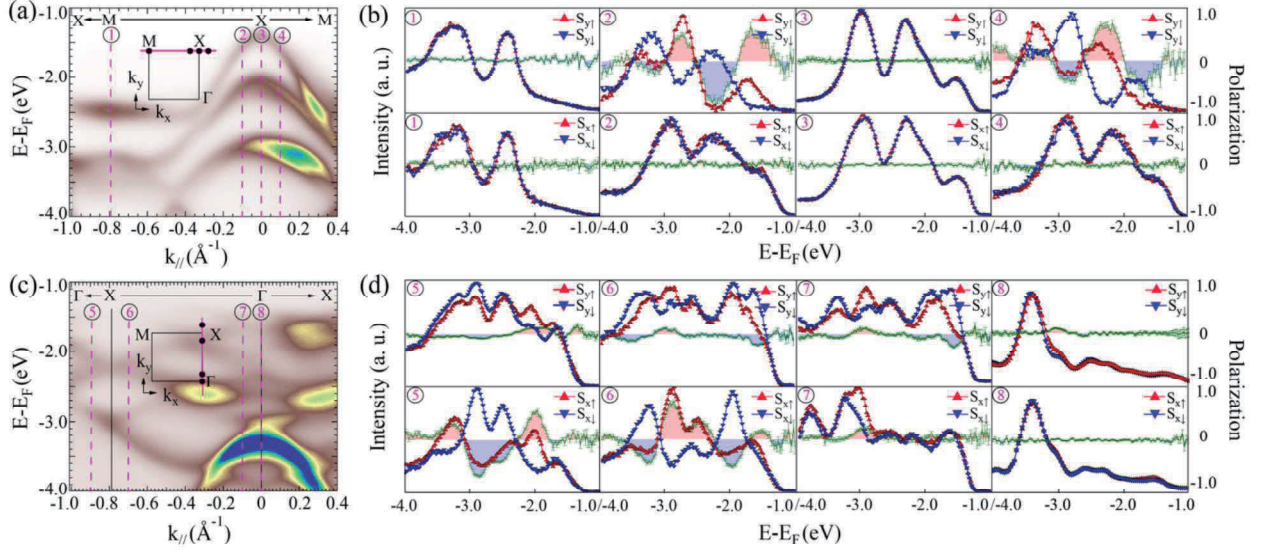


FIGURE 2. (a), (c) Band dispersion along M-X and Γ -X directions, respectively. The inset shows the BZ, with black dots indicating the momentum positions where the spin-resolved EDCs are measured. (b), (d) Spin-resolved EDCs and spin polarizations. Each panel's number corresponds to the momentum point denoted by the pink dashed lines in (a), (c). The green error bars indicate the statistical fluctuation in determining the spin polarizations (proportion $1/\sqrt{N}$; where N is the photoemission intensity).

In summary, combining spin-ARPES measurements and theoretical calculations, we report distinct spin-momentum-layer locking phenomena at different BZ positions in a centrosymmetric material BiOI. The measured spin polarization localized on a specific BiI layer is highly polarized along the BZ boundary but almost vanishes around the zone center due to its nonsymmorphic crystal structure. The layer-resolved spin texture, either Rashba or Dresselhaus type, reflects the symmetry of both real space and k -space. Our findings experimentally demonstrate the existence of the HSP effect and shed light on the design metrics to utilize high spin polarization in centrosymmetric materials by revealing the intimate interplay between spin, orbital, and layer degrees of freedom.

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Low-energy electron-boson couplings in Sr_2RuO_4

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Keywords: Ruthenate, ARPES.

Strontium ruthenate Sr_2RuO_4 is a representative unconventional superconductor ($T_c \sim 1.5$ K) and a strongly correlated two-dimensional electron system [1-2]. To understand the unconventional superconductivity of Sr_2RuO_4 , it is essential to evaluate electron-boson interactions and strong correlation effects quantitatively and separately. In this regard, angle-resolved photoemission spectroscopy (ARPES) is one of the most suitable probes as it can obtain information on many-body interactions. Indeed, many previous ARPES results reported the existence of electron-boson interactions fingerprinted by a “kink” structure in the γ -band derived from Ru $4d_{xy}$ orbital [3-6]. On the other hand, the latest high-resolution ARPES study on the electronic states near the Fermi energy (E_F) reported that the electrons and bosons are not strongly coupled [7]. Such a discrepancy could be attributed to the fact that the existing ARPES measurements only focused on the near E_F region and could not accurately evaluate electron correlations with an energy scale 10 to 100 times larger than that of the boson mode (<100 meV). This suggests that it is necessary to observe not only the band structures in the vicinity of the E_F but also the entire band structure for the accurate identification of coupling components by ARPES. In this regard, to separate and evaluate multiple types of the many-body effects, we have examined the energy and momentum dependence of the many-body effects via high-resolution ARPES measurements in a wide energy range at various momentum locations.

High-quality single crystals of Sr_2RuO_4 were prepared by the floating zone method with a self-flux technique ($T_c \sim 1.36$ K). High-resolution ARPES experiments were measured at the linear undulator beamline (BL-1) of Hiroshima Synchrotron Radiation Center (HiSOR) using an R4000 electron analyzer (Scienta Omicron) [8]. Clean and flat surfaces of the samples were obtained by cleaving *in situ* under ultra-high-vacuum conditions (2×10^{-11} Torr) at a low temperature (~ 20 K). Energy and angular resolution were better than 15 meV and 0.3° , respectively.

Figure 1(a) shows the ARPES images taken at different azimuthal angles ($\varphi = 0, 12, 30$ deg), as indicated in the schematic Fermi surface. Then, we have determined the γ band dispersion by fitting the momentum distribution curves (MDCs) as shown in the blue-filled circles in Fig. 1(b), where a phenomenological model dispersion is also shown. The model dispersion was calculated by including the electron correlation effects with the estimated strength from our soft X-ray ARPES results (not shown). One can see that the experimental dispersion is not reproduced by the model calculations, including the electron correlation effects. This indicates that the electron-boson coupling should be considered to explain the mass renormalization effects in the γ band.

The electron-boson coupling could be more visualized in the real part of the self-energy shown in Fig. 1(c), induced by taking the energy difference between the experimental and model dispersion. We found the characteristic peak at -40 meV and shoulder structures at -60 meV and -20 meV. These structures in the real part of the self-energy correspond to sudden changes of the group velocity, or kinks, in the band dispersion. In our previous ARPES results [5], the structures at -40 meV and -60 meV were also observed, and they can be attributed to the phonon modes, as reported in an inelastic neutron scattering experiment [9]. The remaining structure at -20 meV was not observed in our previous ARPES results. On the other hand, a kink structure having a similar energy-scale at -15 meV was reported for the surface states in Sr_2RuO_4 , and its

origin was discussed in terms of the electron-phonon coupling [6]. To uncover the origin of the kink structure, more detailed self-energy analysis, including the imaginary part of the self-energy, is in progress.

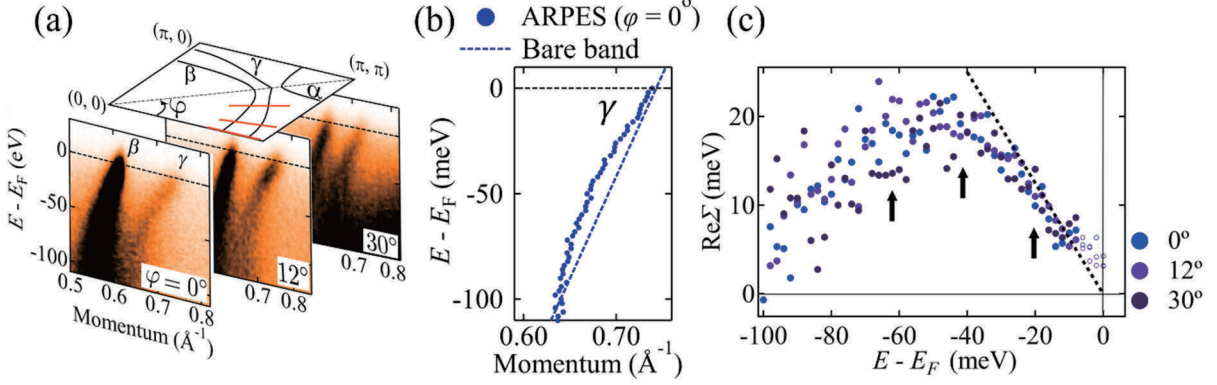


FIGURE 1. (a) High-resolution ARPES measurements on Sr_2RuO_4 as a function of the azimuthal angle ($\varphi = 0, 12, 30$ deg) taken with a photon energy of 65 eV below 20 K with p -polarization geometry. (b) ARPES dispersion determined by fitting MDCs for $\varphi = 0$ deg. The blue dashed line is the phenomenological model dispersion including the electron correlation effects and employed as the bare band to extract the electron self-energy dominated by the electron-boson coupling. (c) Real part of the self-energy as a function of the azimuthal angle ($\varphi = 0, 12, 30$ deg). The arrows indicates the characteristic peak/shoulder structures at -20, -40, and -60 meV.

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Ultrathin Ferromagnetic Films on Spin-Orbit-Influenced Metals: Interplay between Exchange and Spin-Orbit Interaction

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Keywords: ARPES, Iron, Tungsten, Electronic Structure, Dirac-cone-like surface state

The W(110) surface exhibits a Dirac-cone-like surface state (DSS). In recent years, this state has been studied extensively [1-3], showing its anisotropic shape due to the two-fold symmetry of the W(110) surface and its topologically nontrivial character [4]. Upon deposition of ultrathin Fe layers the “Dirac-fermion mass” of the DSS was found to be manipulable by varying the film thickness (and thereby the magnetic properties of the Fe film) [5].

Theoretical studies of W(110) predict that the energetic position of the DSS is related to the interlayer distance of the first W layers. For the case of Fe/W(110) the DSS is expected to shift towards higher binding energies, contrary to the findings of Honma *et al.* This has lead us to re-investigate the Fe/W(110) system by means of angle-resolved photoelectron spectroscopy (ARPES) at the BL-1.

Our ARPES results on W(110) and Fe/W(110) are shown in figure 1. We have labeled a parabolic band (“W”) and the DSS and sketched the dispersion of the latter in the bottom panel. For the pristine W(110) surface, the Dirac Point – the band crossing at the Brillouin zone center $\bar{\Gamma}$ – appears at $E_D = -1.25$ eV which is in good agreement with previous experimental and theoretical results. For the Fe/W(110) case, the same features can be recognized with an additional broad feature at about -0.3 eV. When comparing the position of the DSS between the two figures, it appears to have shifted downward in energy by $\Delta E_D = -75$ meV. The same trend was measured for the Co/W(110) case, where the DSS energy shift amounts to $\Delta E_D = -107$ meV. These findings are consistent with our theoretical calculations.

Coming back to our initial motivation on the sample system, we do not find evidence for an opening of a DSS gap. Our results suggest, that the upper part of the DSS gap reported in [5] might be the parabolic band W persisting upon Fe adsorption. Our results are published in [6].

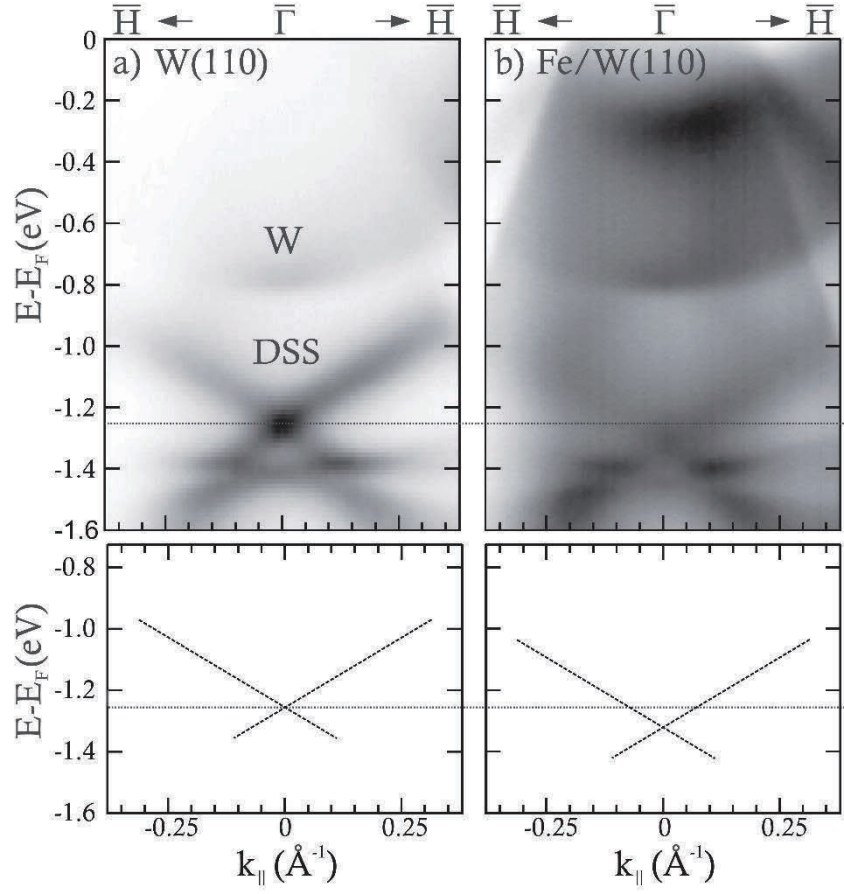


Figure 1. ARPES measurements for the $\bar{H} - \bar{\Gamma} - \bar{H}$ line of W(110) (a) and 0.5 ML Fe/W(110) (b). Below, the dispersion and energetic position of the DSS is depicted. Figure extracted from [6].

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Half-Magnetic Topological Insulator with Magnetization-Induced Dirac Gap at a Selected Surface

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Keywords: axion-insulator, $\text{MnBi}_8\text{Te}_{13}$, angle-resolved photoemission spectroscopy, magnetic topological materials.

The recent discovery of the intrinsic magnetic topological insulator (TI) MnBi_2Te_4 and $\text{MnBi}_2\text{Te}_4/(\text{Bi}_2\text{Te}_3)_n$ ($n=1,2,3,\dots$) have boosted exciting possibilities of producing exotic quantum phenomena by engineering topology and magnetism at the atomic scale [1-4]. Such magnetic TIs are promised to host two exotic quantum phenomena: the “quantum anomalous Hall effect” and the “axion insulating state” [5, 6]. The former has integer-quantized surface Hall resistivity, while the later might present Half-quantized surface resistivity. However, partially because of the lack of a suitable material, it is still not clear if the Half-quantized surface resistivity can be observed in transport measurement, lacking the direct proof of the “axion insulating state”.

In this study, we performed a μ -Laser-angle-resolved photoemission spectroscopy (μ -Laser-ARPES) on an intrinsic ferromagnetic TI, $\text{MnBi}_2\text{Te}_4/(\text{Bi}_2\text{Te}_3)_3$ ($\text{MnBi}_8\text{Te}_{13}$), and demonstrated that $\text{MnBi}_8\text{Te}_{13}$ can be an ideal platform to explore and manipulate the exotic half-quantized surface resistivity. The energy bands show a Dirac gap at the magnetic MnBi_2Te_4 layer but gapless Dirac cone at its nonmagnetic Bi_2Te_3 layers in the opposite side. Remarkably, the magnetic Dirac gap (~ 28 meV) decreases monotonically with increasing temperature and closes right at the Curie temperature (as shown in Figure 1), smoking-gun evidence of magnetization-induced topological surface gap among all known magnetic topological materials.

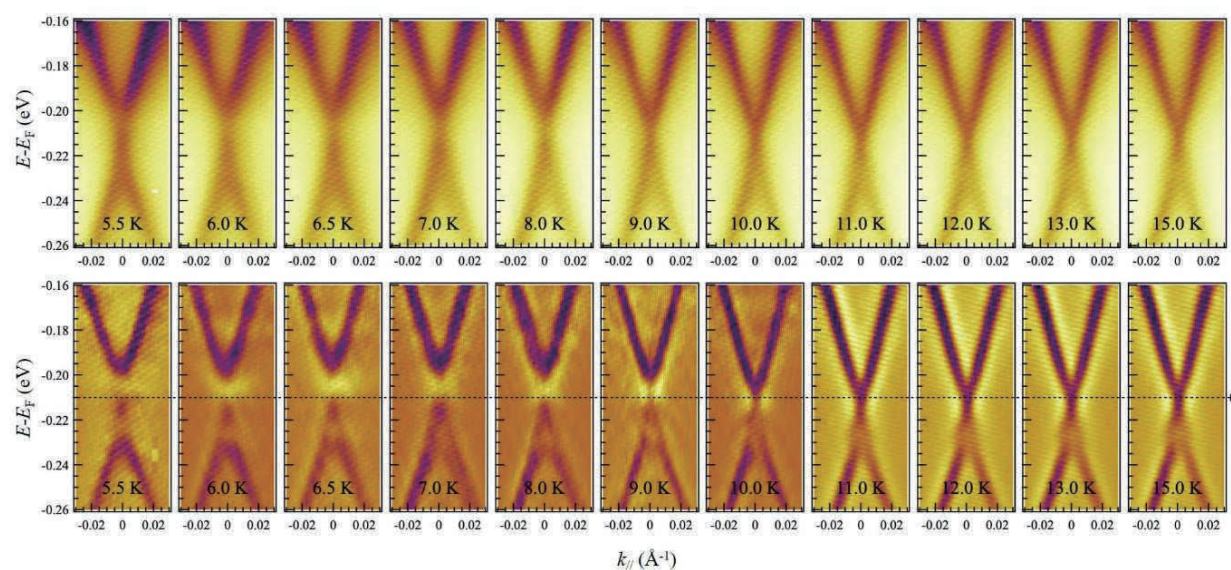


FIGURE 1. ARPES results of the Dirac gap evolution with temperature at the magnetic MnBi_2Te_4 layer of $\text{MnBi}_8\text{Te}_{13}$. Spectra are shown in the form of original ARPES spectra (top row) and 2D curvature spectra (bottom row). Black dashed line indicates the position of gapless Dirac point.

([#]These authors contributed equally to this work. This work has been published in *Physical Review X* **11**, 011039 (2021))

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ARPES Study on low dimensional chiral Dirac material CoNb_3S_6

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Keywords: chiral lattice, Dirac semimetal, CoNb_3S_6 , antiferromagnetic order (AFM), ARPES

The properties two-dimensional materials with layered structure, can be artificially tuned to new ordering by doping, temperature, pressure, and intercalation [1-6]. One significant progress in the 2D material is the realization of two-dimensional magnetism, which can be achieve not only in the systems with intrinsic magnetic order [7-14], but intercalation of magnetic layers in nonmagnetic materials as well [15-19]. As the magnetism can be introduced to layered TMDCs by magnetically element intercalated into the van de Waals gap of TMDCs. Unlike the valley degree of freedom arise from the breaking of inversion symmetry in TMDCs, intercalation of 3d metal can not only breaking the time reversal symmetry and leading to exotic quantum phase of matter, including magnetic topological insulator, QAHE and topological semimetal [15-18], but also triggering crystalline chirality and chiral Dirac fermion as well[19-21].

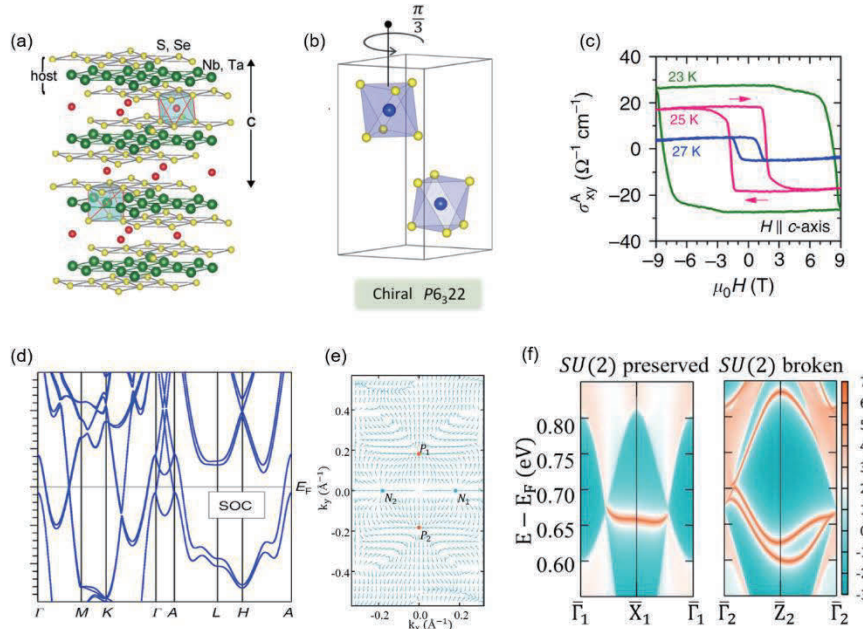


FIGURE 1. (a) Structure of 3d-metal intercalated transition metal dichalcogenides (TMDCs). While red balls represent for 3-d element Co as intercalated layers, yellow and green balls stand for the host TMDCs. Figure (a) is reproduced from [18]. (b) Chirality associated with a specific configuration of octahedra filled in a hexagonal crystal. Figure (a) is reproduced from [20]. (c) Hall conductivity of CoNb_3S_6 as a function of magnetic field. (d) Calculated band structure of CoNb_3S_6 with inclusion of SOC. Figure (c-d) are reproduced from [19]. (e) Distribution of in-plane component of Berry curvature on $k_z = 0$ plane, N_1 , N_2 and P_1 , P_2 denote the source and sink, respectively. (f) Chiral edge states of 2D slice with Chern number of 2. Doubly degenerate edge bands locate at $SU(2)$ -preserved edge, while the nondegenerate ones on $SU(2)$ -broken edge. Figure (e, f) are reproduced from [21]

CoNb_3S_6 is an intercalated transition metal dichalcogenides compound with chiral lattice [18-21]. The intercalated Co atom layers with 3-d electrons sandwiches between layers of 5d transition metal dichalcogenides van de Waals gap, form interlayer antiferromagnet order [18,19,21], and therefore leading to a 1/3 fractional intercalation of NbS_2 layers with chiral lattice ($P6_322$) [21]. Along with its crystalline chirality, the intercalated Co layers contributes magnetic order below 26 K, gives rise to a complex susceptibility for the magnetic field applied along c-axis, large anomalous Hall effect [19] and predicted to host novel state of electrons e.g. chiral fermion (FIG.1) [21]. Although CoNb_3S_6 has been studied via DFT calculation and transport measurements, the experimental studies of its electronic structure still lacking, it is thus crucial to study its electronic structure experimentally [18,19,21].

We performed ARPES measurement on the chiral-lattice antiferromagnet CoNb_3S_6 . Our results revealed the hexagonal Fermi surface, agrees well with existed band calculation [18,19]. Two linear hole-like bands dominate the density of state around the center of Brillouin zone and forms concentric circles centered at Γ point. With increasing the binding energy, the centered hole-like dispersions reach its bottom at both K and M points, in good agreement with DFT calculation. According to the theoretical calculation, CoNb_3S_6 can be a Weyl semimetal with several pairs of Weyl points circled around the Γ point, and the Fermi arcs connect these Weyl points with opposite chirality surrounding Γ point can form a nearly circled feature on its FS. Our observation of the concentric circles feature might partially originate from such multiple Fermi arcs around the Γ point.

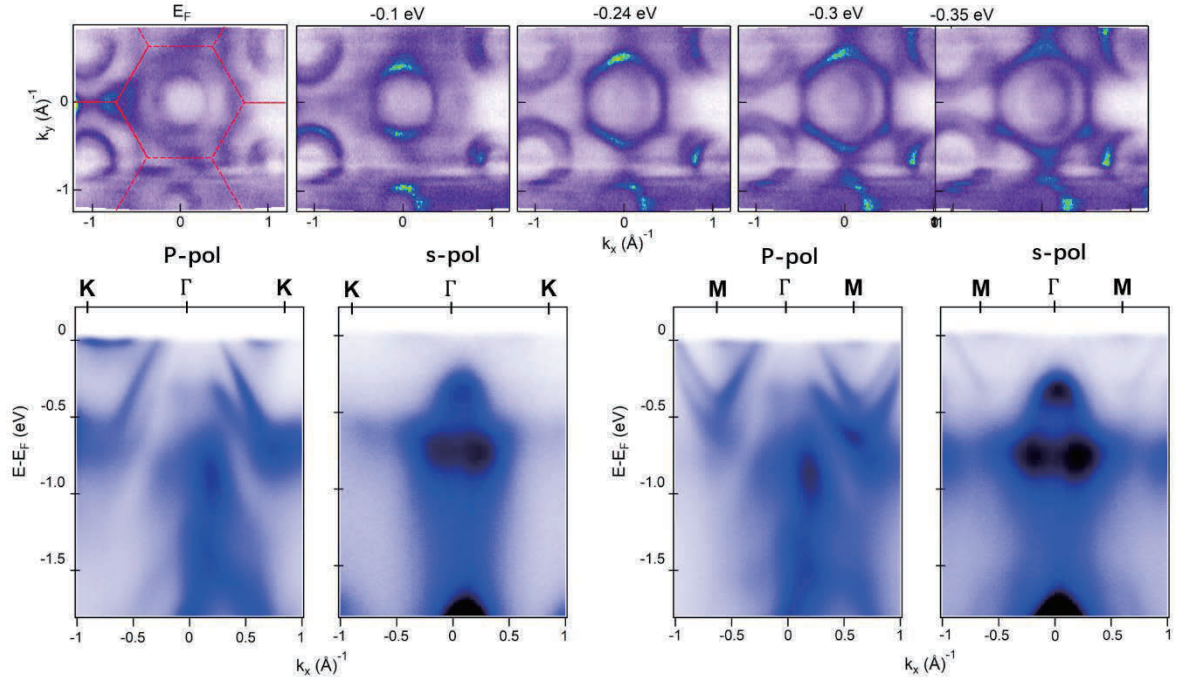


FIGURE 2. Our ARPES intensity map of CoNb_3S_6 single crystal taken @ BL1 with different linear polarized light. The up-panels shows constant energy maps with LH. And the panels downside shows electronic structure along $K - \Gamma - K$ (left) and $M - \Gamma - M$ (right) direction, respectively

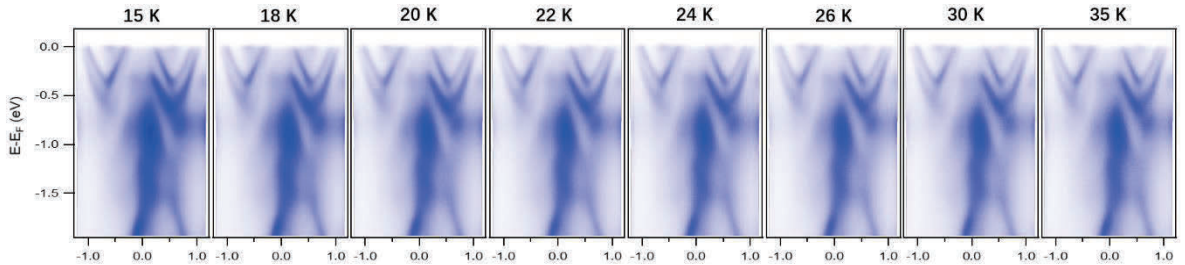


FIGURE 3. ARPES measured electronic structure along $M - \Gamma - M$ direction at different temperature with LH linear polarized light.

By taking advantage of changeable measurement configuration of BL1 end-station of HiSOR, it is straightforward to get ARPES measurement with different linear polarized light. While p-polarized light clear revealed the dispersion feature, especially the electron pocket resides along $\Gamma - K$ direction, the s-

polarized light resolved the hole band centered at Γ point with its band maximum located around 200 meV below Fermi level (FIG. 2). By combining the different polarization of incident light, we obtained the complete information CoNb₃S₆ electronic structure. In addition, transport measurement indicates a magnetic transition occurs at ~ 25 K [19]. To uncover its electronic structure evolution upon temperature variation, we performed systematic ARPES measurement upon different temperature (15 K-35 K), and our ARPES data indicates no obvious change in its electronic structure along high symmetry direction (FIG. 3). The anticipated magnetic order transition might lead to more delicate changes such as the spin polarization or Fermi arc length, rather than electronic band structure. Further spin-ARPES and CD-ARPES experiments will be able to resolve such magnetic transition associated spin evolution.

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Termination-Dependent Surface States of Cr₂O₃ on Graphene Covered Ni(111) Substrate

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Keywords: Cr₂O₃, heterointerface, band structure, ARPES

Interfaces between a magnetic material and heavy metal that has a large spin-orbit coupling exhibit a variety of spin-related interfacial phenomena, such as anomalous Hall effect, interfacial Dzyaloshinskii-Moriya interaction. These phenomena have been of great importance in spintronic applications. Investigations of such interfacial properties have been extended to heavy metal/antiferromagnet systems. It has so far been predicted that a nontrivial band gap opening in the graphene Dirac bands asymmetric between K and K' valleys at antiferromagnets Cr₂O₃/graphene interface, which is mediated by symmetry protected boundary magnetization of Cr₂O₃. The successful fabrication of Cr₂O₃ on graphene covered Ni(111) substrate pave the way for verifying the band alignment of Cr₂O₃/graphene heterostructure. A narrowed band gap of ultrathin α -Cr₂O₃ than that of crystal was also observed by STS. While the progresses are impressive, it should be recognized that directly experimental observation of an interfacial band structure of Cr₂O₃/graphene is still lack. Antiferromagnet Cr₂O₃ exhibits a spontaneous ferromagnetic termination (boundary magnetism) which is insensitive to the surface roughness. A general consensus is that the lattice parameter c of the topmost several layers is compressed. In fact, the surface structure of Cr₂O₃ can be quite complex. The variation of fabrication method, oxygen pressure could make peculiar reconstruction and occupation states in ultrahigh vacuum environment. And it is possible that the electronic structure of heterointerface is distinguished to their bulk and forms particular band structure specific to the its termination structure. From this perspective, it is beneficial to direct describe band structure of ultrathin Cr₂O₃/graphene.

In this study, ultrathin Cr₂O₃ thin films are deposited on Gr/Ni(111) by O₂-assisted MBE method. Here, we confirmed O-terminated and Cr-terminated Cr₂O₃ thin films coexistence on graphene covered Ni(111) substrate by ARPES and first principle calculation. The Cr-terminated model shows novel surface states at BE~0.25 eV due to the uncompensated dangling bonds formed a , while O-terminated model shows traditional bulk band characteristic. Besides, the adsorption type of graphene is depending on not only Ni(111) substrate but the Cr₂O₃ termination. Graphene is freestanding type with clear Dirac cone at BE~2 eV O-terminated model, but keeps metallic characteristic in Cr-terminated model.

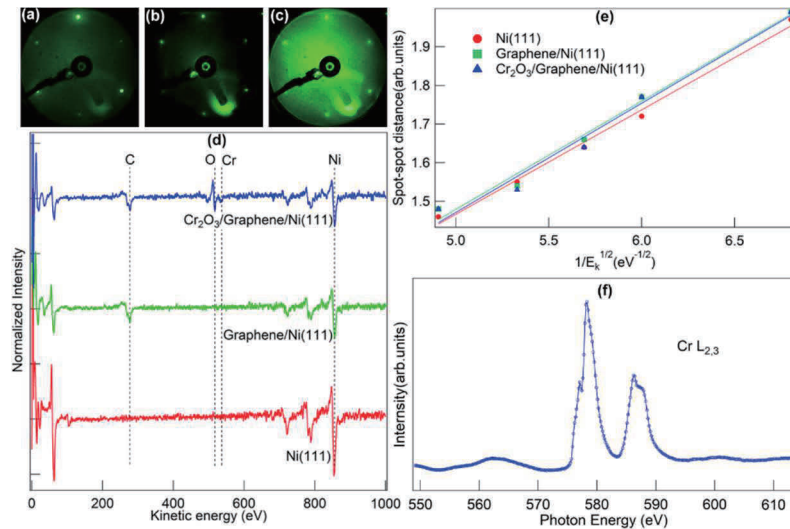


Figure 1 (a)-(c) LEED pattern of Ni(111), Graphene/Ni(111), and 0.5nm-Cr₂O₃/Graphene/Ni(111), (d) corresponding differentiated AES spectra, (e) spot-spot distance of corresponding sample, (f) Cr L_{2,3} XAS spectrum. Red, green and blue curves corresponding to Ni(111), Graphene/Ni(111), and 0.5nm-Cr₂O₃/Graphene/Ni(111).

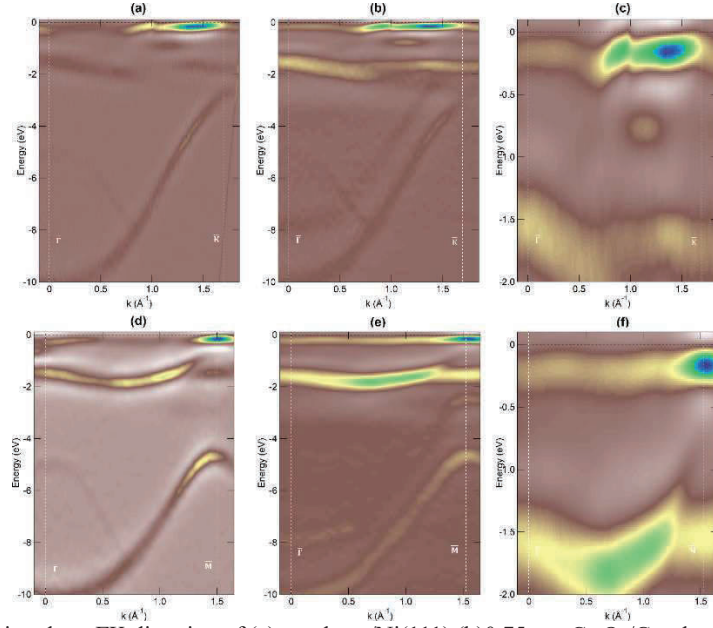


Figure 1 ARPES intensities along ΓK direction of (a) graphene/Ni(111) (b) 0.75 nm- Cr_2O_3 /Graphene/Ni(111) and (c) zoom-in image of (b) 0.75 nm- Cr_2O_3 /Graphene/Ni(111). (d)-(f) are corresponding ARPES along ΓM direction. One can find two type of graphene coexist in Cr_2O_3 /Graphene/Ni(111) interface, Cr_2O_3 3d states not only dominating at BE $\sim$ 1.9 eV, but also shows a flat band around BE $\sim$ 0.25 eV.

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High-resolution ARPES of heavily overdoped Bi2201: evaluation of coupling parameters

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Keywords: High- T_C Superconductor, ARPES, Many-body interactions

High transition-temperature (T_C) cuprate superconductors have attracted much interest since their discovery in 1986. While there have been extensive studies, there still exist many unsettled physical phenomena such as the bad metal state, density waves formation, magnetic fluctuations, and nematic phases [1-4]. These quantum states are emergent from the competing charge, spin and lattice degrees of freedoms. To understand the origin of the physical properties of cuprates, it is desirable to disentangle these competing interactions and quantify each contribution.

To this end, here we focus on the Bi-based high- T_C cuprate, $(\text{Bi,Pb})_2\text{Sr}_2\text{CuO}_{6+\delta}$ (Bi2201). The Fermi surface of Bi2201 is relatively simple because there exists a single CuO_2 plane in the unit cell. While it has been widely believed that the electronic state in the heavily overdoped region is the Fermi Liquid like, Kurashima et al. recently have found a ferromagnetic fluctuation close to the vanishing T_C [3]. Previously Meevasana et al. have done angle-resolved photoemission spectroscopy (ARPES) and examined the self-energy due to the electron-electron and electron-phonon interactions, especially for the heavily overdoped Bi2201 with no superconducting transition [5]. However, each contribution from the electron-phonon (or electron-boson) and electron-electron interactions as a function of hole concentration has not been quantitatively elucidated. Our purpose is, therefore, to systematically clarify the evolution of these interactions as a function of the hole concentrations as well as T_C .

In this study we examined the electronic structure of as grown Bi2201 single crystal with $T_C=6$ K. We have done linear-polarization dependent ARPES experiments on BL-1 [6] to investigate the Fermi surface and energy band dispersion in the wide energy range. To examine the fine details of the electron-phonon interaction near the Fermi level, we have done high-resolution ARPES using focused ultraviolet laser light [7].

Figure 1(a) shows the observed Fermi surface taken at $h\nu=90$ eV at 50 K. In addition to the hole-like Fermi surface, we have observed additional Fermi surfaces moved by $(\pi/2, \pi/2)$, which suggests superstructure formation. To quantitatively analyze the shape of the Fermi surfaces, we have employed the tight-binding (TB) model [8]. The red line shows the best fit result using the two-band TB model. We have also tried the single-band TB model [5], but we could not sufficiently reproduce the shape of the Fermi surface. It may indicate the importance of the hybridization effect.

Figure 1(b) and 1(c) show the band structure along the nodal direction (cut 1) and the anti-nodal direction (cut 2) taken at $h\nu=40$ eV. We measured cut 1 (cut2) using s-(p)-polarization geometry taking into account the dipole selection rule [6,9]. One can see the reduction of the group velocity near the Fermi level compared with the TB model. The deviation is assumed to derive from the self-energy due to electron-electron interaction. We also overlaid the LDA result [10] for Bi2201 in Fig. 1(b). One can see that the deviation of the group velocity is not so strong compared with the two band TB model, which should be examined in more detail in the future.

We have also measured fine details near the Fermi level using the μ -Laser ARPES machine. We have observed a clear kink structure around -80 meV. We have extracted the self-energy due to the electron-phonon and electron-electron interactions referring to the two-band TB model.

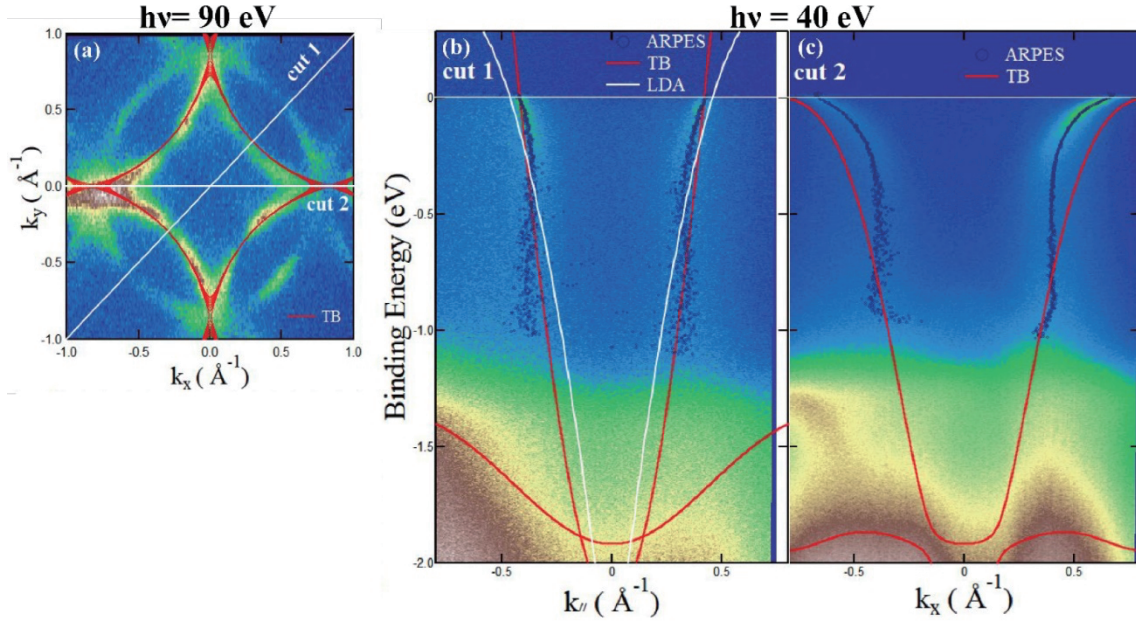


FIGURE 1. (a) Fermi surface of Bi22012 taken at $h\nu = 90$ eV at 50 K. The red lines show calculated Fermi surface using the two-band TB model. ARPES image plot along (b) the nodal direction (cut 1 in (a)) and (c) the anti-nodal direction (cut 2 in (a)) respectively. Red lines in (b) and (c) show the band dispersions calculated by the two-band TB model. The white line in (b) shows a band dispersion obtained from the LDA calculation [10]. Blue dots in (b) and (c) show the band points evaluated by the momentum distribution curve analyses.

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Topological Surface State in Sb, Te, and Se Based Single Crystals

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Keywords: Topological Insulators, ARPES

Three-dimensional (3D) topological insulators (TIs) belong to the new quantum state of matter family, which has attracted great attention since last decade because of their unique physical properties and potential applications to future devices [1]. The 3D TI has an energy gap at the Fermi level (E_F) in the bulk bands but has gapless topological surface states (TSSs). The metallic property of the TSS is robust against perturbations that conserve the topological numbers. Studies on a layered semiconductor Sb_2Te_3 and related compounds have increased tremendously since Sb_2Te_3 has been predicted and demonstrated to behave as a 3D TI. From the recent reports on Sb_2TeSe_2 , however, it is not clear that whether it belongs to the TI family or not. While Zhang *et al.* reported that Sb_2TeSe_2 belongs to the TI family by *ab-initio* calculations in 2010 [2], Menshchikova *et al.* suggested that it is a normal insulator with a 100 meV bandgap in 2013 [3]. Therefore, it is desirable to experimentally clarify the TSSs in these families.

In this study, we examined the electronic band structure of Sb_2Te_3 and $\text{Sb}_{1.9}\text{Sn}_{0.1}\text{TeSe}_2$ single crystals using angle-resolved photoemission spectroscopy (ARPES). These samples behave as p-type semiconductor and have the Dirac point inside the bulk bandgap. The latter property is favorable for dissipationless spin-based electronic devices because the surface to bulk scattering is suppressed [4,5].

We have done ARPES experiments on the linear undulator beamline BL-1 [6] with the *p*-polarization geometry. We cleaved the sample in the ultrahigh vacuum to get clean surfaces. Based on the core-level photoemission spectra, we have confirmed all the elements included in these crystals.

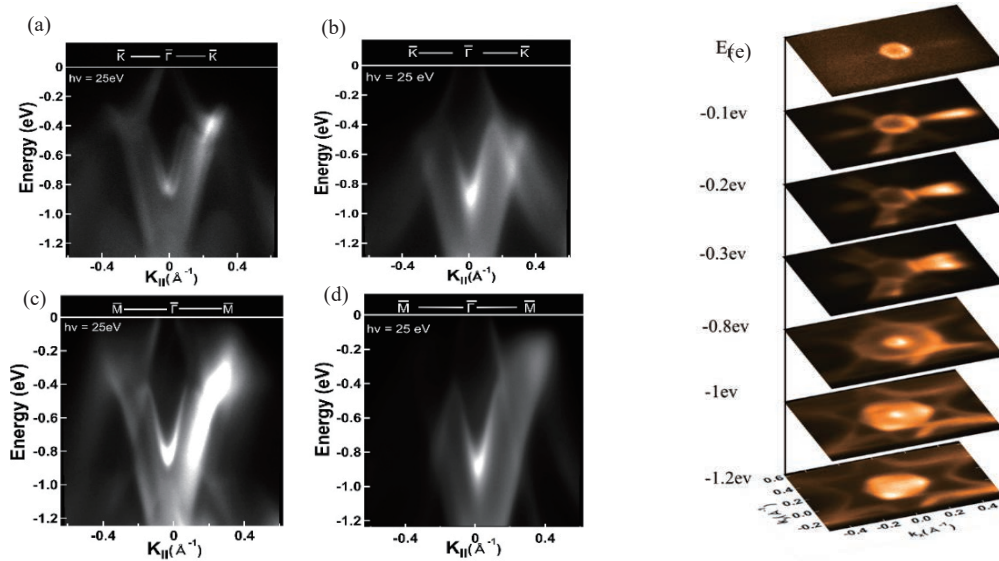


Figure 1. ARPES results of Sb_2Te_3 and $\text{Sb}_{1.9}\text{Sn}_{0.1}\text{TeSe}_2$ taken at $h\nu = 25\text{ eV}$ and at 22 K. (a) and (c) show ARPES image plots of Sb_2Te_3 along $\bar{\Gamma}-\bar{K}$ and $\bar{\Gamma}-\bar{M}$ directions, respectively. (b) and (d) show ARPES image plots of $\text{Sb}_{1.9}\text{Sn}_{0.1}\text{TeSe}_2$ along $\bar{\Gamma}-\bar{K}$ and $\bar{\Gamma}-\bar{M}$ directions, respectively. (e) The constant energy contours of $\text{Sb}_{1.9}\text{Sn}_{0.1}\text{TeSe}_2$.

Figures 1(a)-(d) show the ARPES image plots of Sb_2Te_3 and $\text{Sb}_{1.9}\text{Sn}_{0.1}\text{TeSe}_2$ single crystals taken at photon energy of $h\nu = 25$ eV at temperature of 22 K along the $\bar{\Gamma} - \bar{K}$ and $\bar{\Gamma} - \bar{M}$ high symmetry directions of the surface Brillouin zone. In Fig. 1(a), one can clearly see a Rashba-like band splitting in Sb_2Te_3 around the $\bar{\Gamma}$ point at the energy of -0.8 eV along the $\bar{\Gamma} - \bar{K}$ direction. However, the splitting is not visible along the $\bar{\Gamma} - \bar{M}$ direction. Based on the analysis of the momentum distribution curves (MDCs), we have evaluated the Fermi wavenumber as $k_F \sim 0.06 \text{ \AA}^{-1}$ and Fermi velocity as $v_F \sim 4.9 \times 10^5$ m/s for Sb_2Te_3 , which is consistent with previous study [4]. On the other hand, in Figs. 1(b) and 1(d), the ARPES linewidth of $\text{Sb}_{1.9}\text{Sn}_{0.1}\text{TeSe}_2$ becomes much broader, and Rashba-like band splitting around the $\bar{\Gamma}$ point is not observed. We found that the evaluated Fermi wavenumber $k_F \sim 0.075 \text{ \AA}^{-1}$ and the Fermi velocity $v_F \sim 5.6 \times 10^5$ m/s were slightly larger than those of Sb_2Te_3 . Along the $\bar{\Gamma} - \bar{K}$ direction, the bulk valence bands do not cross E_F whereas they cross E_F along the $\bar{\Gamma} - \bar{M}$ direction.

Figure 1(e) shows constant energy contour plots in the momentum space at $h\nu = 25$ eV at 22 K. A rounded hexagonal Fermi surface derived from the surface state has been observed at E_F . While the Fermi surface derived from the bulk valence bands are weak at E_F , they are discernible along the $\bar{\Gamma} - \bar{M}$ directions. The bulk valence bands become more visible below energy of -0.2 eV. While the lattice symmetry should be six folded, the distribution of the photoemission intensity looks three-fold symmetric at energies between -0.1 eV to -0.3 eV. Note that the six-fold symmetry of the valence band is clearly visible at energies of -1.0 eV and -1.2 eV. We assume this is due to the matrix element effects.

The states crossing E_F in $\text{Sb}_{1.9}\text{Sn}_{0.1}\text{TeSe}_2$ look like a TSS, as in pristine Sb_2Te_3 in Fig. 1(a). To further confirm the TSS, photon energy-dependent ARPES measurement on the Sn-doped and pristine Sb_2TeSe_2 samples are required.

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Band Dispersion Unique To The Cobalt Intercalation In The Noncentrosymmetric Antiferromagnet CoNb_3S_6

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Keywords: Angle-resolved photoemission spectroscopy (ARPES), magnetic Weyl semimetals.

CoNb_3S_6 [Fig. 1(a)] is a transition metal dichalcogenide NbS_2 with cobalt intercalation. Neutron scattering experiments have revealed that intercalated Co atoms emerge collinear antiferromagnetic structure along in-plane direction [1]. Furthermore, weak ferromagnetism along out-of-plane direction and anomalous Hall effect have been observed recently [2]. However, the anomalous Hall response is giant compared to the weak ferromagnetic moments, and the mechanism of this giant anomalous Hall effect has been unclear.

We studied the electronic structure of CoNb_3S_6 by comparing it with that of FeNb_3S_6 , which is intercalated with Fe instead of Cu to NbS_2 . Single crystals of CoNb_3S_6 and FeNb_3S_6 were synthesized by chemical vapor transport method. Angle-resolved photoemission spectroscopy (ARPES) measurements using the vacuum ultraviolet (VUV) light were performed at BL-1 of HiSOR. We used the 120 eV light as a light source. The measurement temperature was kept lower than 20 K for CoNb_3S_6 and 35 K for FeNb_3S_6 , where both temperatures are lower than T_N of these materials. All samples were cleaved in situ along the (001) direction at an ultrahigh vacuum of 5.0×10^{-9} Pa.

Figure 1(b) shows the band dispersions of CoNb_3S_6 and FeNb_3S_6 . The original material NbS_2 without the intercalation has two hole pockets around the $\bar{\Gamma}$ point and these dispersions were also observed in the intercalated materials, as highlighted by red dashed curves. Furthermore, an additional band dispersion appeared near the Fermi level only in CoNb_3S_6 [blue dashed curves in Fig. 1(b) left panel]. This dispersion seems to come from Co atoms, because such a band dispersion does not exist in the band dispersion of NbS_2 , FeNb_3S_6 , and CrNb_3S_6 [3]. Therefore, we conclude that this electronic structure is unique to the cobalt intercalation, and possibly responsible for the exceptionally large anomalous Hall effect of CoNb_3S_6 .

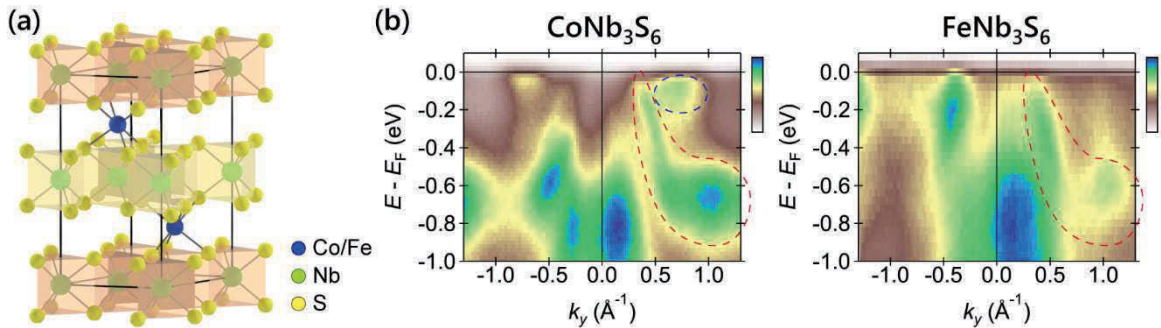


FIGURE 1. Crystal structure and band dispersions of CoNb_3S_6 and FeNb_3S_6 . (a) Crystal structure of CoNb_3S_6 and FeNb_3S_6 . (b) Band dispersions of CoNb_3S_6 and FeNb_3S_6 along the $\bar{\Gamma} - \bar{M}$ direction of NbS_2 . Band structures originating from NbS_2 are highlighted by red curves and an additional band in CoNb_3S_6 is highlighted by blue curves.

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Development of On-site Cleaning Method of Carbon Contamination with Atomic Hydrogen 2

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Keywords: Contamination, Cleaning, Atomic hydrogen.

Carbon contamination of optical elements in synchrotron radiation beamlines is a common and serious problem [1-4] since accumulation of carbon on the surface makes thick carbon layer which cause drastically decrease of intensity even around 1KeV. There is a cleaning method which uses ozone and UV light but the method is not applicable for easily-oxidizable-metal such as Ni, Cr, etc. since Ozone cause surface oxidation. Therefore, the cleaning technique for this kind of mirrors is not established.

Recently, we reported off-site cleaning treatment using the atomic hydrogens (AH) is very effective in removing contamination of mirrors [5]. In response to this, we received requests from many people to develop AH cleaning equipment that can be used “on-site”. Therefore, we began to develop a small on-site cleaning device that can be mounted in a mirror vacuum chamber. Here we report on the tests of the cleaning method conducted at the HiSOR beamline BL-6 of Hiroshima University.

Fig. 1 shows the AH generator apparatus attached to the grating chamber of HiSOR BL6. Fig.2 shows

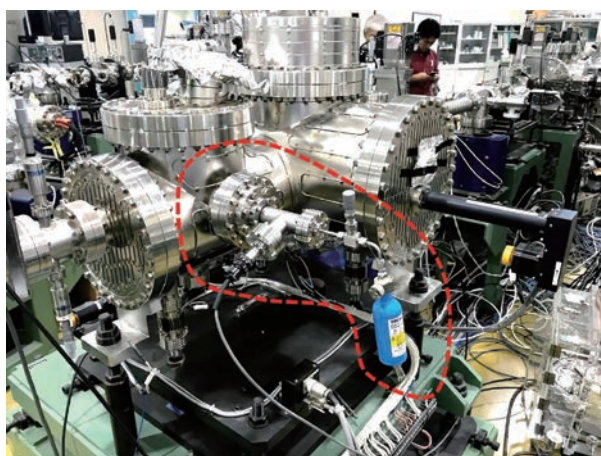


FIGURE 1. Atomic hydrogen generator setup attached to the grating chamber. The blue cylinder is a hydrogen canister using hydrogen storage alloy, which makes safe handling of hydrogen without using high pressure canister.

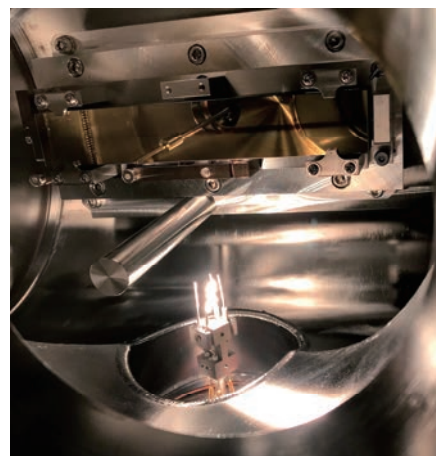


FIGURE 2. Atomic hydrogen generator head in the grating chamber.

Since the filaments is too bright for taking photos, applied current is reduced from typical value when taking photo shot.

the head part of the AH generator apparatus which consists of tungsten (W) filament, current introducing electrode, hydrogen gas introduction piping, hydrogen gas source and flow regulator. These parts can be easily installed in the vacuum chamber of the beamline by fixing/positioning them to the short pipe with 70 mm CF flange. By using a canister with a low-pressure hydrogen storage alloy as the gas source, clean hydrogen gas can be easily supplied into the mirror chamber at the synchrotron radiation experimental hall where it is difficult to introduce a high-pressure dangerous gas cylinder. Hydrogen gas was introduced in the vacuum chamber to contact with W filaments heated to about 1700 °C for generation of AHs by catalytic reaction.

In the test experiment at the beamline in the last year, we cannot confirm cleaning effect and conclude that amount of AH is not sufficient to remove carbons from surface within short time scale. Thus, we have improved generator by increased number and length of filament, an expected surface area of filaments becomes around 3 times compared to the last time. For measurement of intensity of the beam we have newly installed gold mesh before grating which makes easier to monitor beam intensity accurately.

We have tested the atomic hydrogen cleaning of the contaminated M2 mirror of the monochromator in BL-6 of the HiSOR facility. Fig. 3 shows the intensity profile of the monochromator before and after the AH treatment. Overall intensity seems slightly increased, however there is no improvement after repeating AH cleaning, therefore it might be error of measurement.

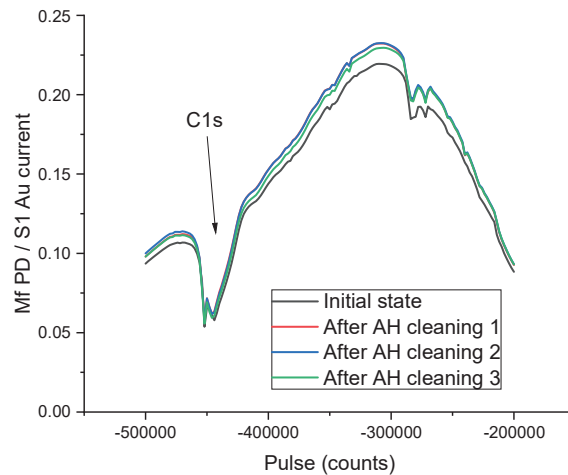


FIGURE 3. Intensity profiles of the monochromator before and after the atomic hydrogen treatment.

Our conclusion is the etching speed of carbon by atomic hydrogen is low and it is not enough to remove the thick layer of carbon which is evidently seen by eye. Recently, oxygen cleaning was introduced to recover from carbon contamination at MAX IV, which dosing oxygen gas around 1E-6 mbar while shining beam around oxygen edge and quickly remove carbons around a day for whole energy region of monochromator. This method works fine, however, weak absorption probably comes from oxidation of surface is observed. Atomic hydrogen is effective to dioxide oxides as we observed in tungsten oxides. Therefore combination of oxygen cleaning with atomic hydrogen cleaning will be a candidate of method to clean surface of mirrors.

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Comparison of soft X-ray absorption spectra of transition metal sulfates

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Keywords: Soft X-ray absorption spectrum, 3d transition metal sulfate, H₂O, SO₄²⁻, bond length

The 3d transition metal (Fe, Co, Ni, Cu, and Zn) sulfates make an octahedral structure with 6 coordination containing some water molecules. Those complexes have various numbers of the coordinated water such as FeSO₄·7H₂O, NiSO₄·6H₂O, and CuSO₄·5H₂O. Only copper combines directly with SO₄²⁻ in five elements. Four H₂O are coordinated in the same plane and two SO₄²⁻ are coordinated out of plane (Figure 1). To investigate how the electronic states of coordinated water molecules are affected by the central metal atoms, soft X-ray absorption spectra have been measured in the energy range of 530–545 eV (O1s) and 690–1050 eV (metal 2p) of the 3d transition metals at BL-6 [1] of the HiSOR.

Figure 2 shows the total electron yield (TEY) spectra of the 3d transition metal sulfates in the O1s edge. The lowest peaks are observed around 532.8 eV and their energies depend on the 3d transition metals. There are two kinds of oxygen atoms (H₂O and SO₄²⁻) in the present complexes. To distinguish them, the TEY spectrum of anhydrous copper sulfate (CuSO₄) is also measured and compared with CuSO₄·5H₂O (Figure 3, top panel). The lowest peak observed only in CuSO₄·5H₂O is thus ascribed to contribution of the coordinated water molecules. The difference spectrum is calculated by normalizing the two spectra with the number of oxygen atoms (Figure 3, middle panel) and compared with the spectrum of a gaseous water molecule [2] (Figure 3, bottom panel). Comparing to the 4a₁ peak of gaseous water molecule, the lowest peak of the coordinated water is shifted to lower energy side by about 1 eV and broadened. The lowest peak energies of the 3d transition metal (Fe, Co, Ni, Cu, and Zn) sulfates at the O1s edge are summarized in Table 1.

TEY spectra are also measured at metal 2p edge. There observed spin-orbit separation peaks of 2p_{3/2} and 2p_{1/2} to 3d transitions with the energy separation of 13–21 eV except Zn, which has no hole in the 3d orbit. Comparing the experimental transition energies of 2p_{3/2} to 3d with the calculated differences of binding energies between 2p and 3d electrons, the largest discrepancy for Cu is considered to be ascribed to direct coordination of SO₄²⁻ to the central metal.

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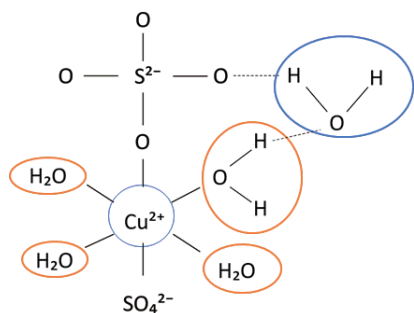


FIGURE 1. Schematic diagram of $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$.

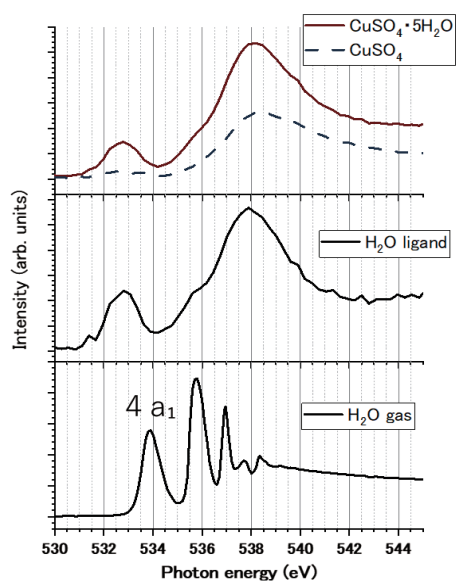


FIGURE 3. TEY spectra of (top) $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ and CuSO_4 , (middle) calculated ligand (H_2O) component, and (bottom) H_2O (gas).

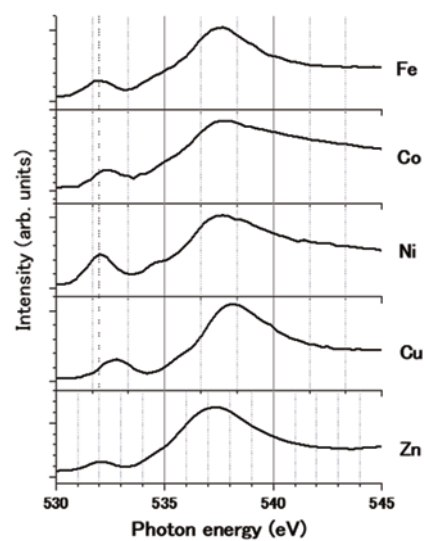


FIGURE 2. TEY spectra of $3d$ transition metal (Fe,Co,Ni,Cu, and Zn) sulfates at the $\text{O}1s$ edge.

TABLE 1 The lowest peak energies of the $3d$ transition metal (Fe,Co,Ni,Cu, and Zn) sulfates at the $\text{O}1s$ edge.

	Peak energy (eV)
$\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$	532.0
$\text{CoSO}_4 \cdot 7\text{H}_2\text{O}$	532.4
$\text{NiSO}_4 \cdot 6\text{H}_2\text{O}$	532.0
$\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$	532.8
$\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$	532.0