Signature of band inversion in the antiferromagnetic phase of axion insulator candidate EuIn$_2$As$_2$

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We have performed angle-resolved photoemission spectroscopy on EuIn$_2$As$_2$ which is predicted to be an axion insulator in the antiferromagnetic state. By utilizing soft-x-ray and vacuum-ultraviolet photons, we revealed a three-dimensional hole pocket centered at the Γ point of the bulk Brillouin zone together with a heavily hole-doped surface state in the paramagnetic phase. Upon entering the antiferromagnetic phase, the band structure exhibits a marked reconstruction characterized by the emergence of an “M”-shaped bulk band near the Fermi level. The qualitative agreement with first-principles band-structure calculations suggests the occurrence of bulk-band inversion at the Γ point in the antiferromagnetic phase. We suggest that EuIn$_2$As$_2$ provides a good opportunity to study the exotic quantum phases associated with a possible axion-insulator phase.

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I. INTRODUCTION

The interplay between magnetism and topology is now becoming an exciting topic in condensed-matter physics. Magnetic order that spontaneously breaks the time-reversal symmetry induces various exotic quantum phenomena which are originally absent in time-reversal-invariant $Z_2$ topological insulators (TIs), as represented by the quantum anomalous Hall effect in magnetically doped TIs [1–6], the unusual electron dynamics in axion TIs [7,8], and the chiral Majorana fermions in superconductor hybrids [9,10]. A well-known strategy to realize such fascinating topological phases is the introduction of magnetic impurities into TI crystals or fabrication of heterostructures. However, the former inevitably introduces disorder in the crystal and the latter requires complex steps to achieve desired quantum properties. Thus, the realization of intrinsic magnetic TIs where the magnetic order is inherently incorporated into the crystal is highly desired, because it would be a more promising platform to realize “cleaner” topological quantum effects.

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FIG. 1. (a) Crystal structure of EuIn$_2$As$_2$. (b) Bulk hexagonal BZ and corresponding surface BZ. (c) Energy distribution curve (EDC) in a wide $E_B$ region measured with $h\nu = 90$ eV in the PM phase ($T = 30$ K). Inset shows the near-$E_F$ EDC measured with $h\nu = 427$ eV. (d) ARPES-intensity mapping at $E_F$ as a function of in-plane wave vectors ($k_x$ and $k_y$) at $k_z \sim 0$ ($h\nu = 427$ eV) measured at $T = 30$ K. (e) ARPES-intensity mapping at $E_F$ as a function of $k_x$ and $k_y$ measured with $h\nu = 360–480$ eV at $T = 30$ K, highlighting the bulk holelike Fermi surface centered at $\Gamma$ (dashed rectangle). (f)–(h) ARPES intensity as a function of $k_\parallel$ and $E_B$ measured along cuts A–C in (e), respectively.

bulk-band structure at the bulk BZ center. As a result of nontrivial topology associated with the parity-based invariant $Z_4 = 2$ [27], an axion-insulator phase would emerge in the AFM phase. There are two metastable magnetic structures in EuIn$_2$As$_2$; i.e., the magnetic moment aligns parallel to the $a/b$ axis (in plane) or the $c$ axis (out of plane), called here AFM\|$b$ or AFM\|$c$ states, respectively. Intriguingly, the AFM\|$b$ state is predicted to host an axion-insulator phase which coexists with a topological-crystalline-insulator (TCI) phase whose high-index crystal surfaces can host hinge states. Moreover, the AFM\|$c$ state also hosts an axion-insulator phase, but it coexists with a higher-order TI state with chiral hinge mode [27]. Despite such intriguing theoretical predictions, the electronic state of EuIn$_2$As$_2$ has been scarcely investigated by experiments [21,22]. It is thus highly desired to clarify the band structure of EuIn$_2$As$_2$.

In this article, we report an angle-resolved photoemission spectroscopy (ARPES) study of EuIn$_2$As$_2$ bulk single crystal. By complementary use of soft-x-ray (SX) and vacuum-ultraviolet (VUV) photons, we have separately identified three-dimensional (3D) bulk and two-dimensional (2D) surface bands. Temperature-dependent ARPES measurements further revealed an intriguing reconstruction of bulk-band structure associated with the AFM transition. The observed band structure in the AFM phase is consistent with the band-structure calculations, pointing to a possibility that EuIn$_2$As$_2$ in the AFM state hosts the axion-insulator phase.

II. EXPERIMENT

High-quality EuIn$_2$As$_2$ single crystals were synthesized by the flux method. ARPES measurements were performed with synchrotron light at the SX-ARPES end station of the ADDRESS beam line at the Swiss Light Source, Paul Scherrer Institute, Switzerland [30], and the HR-ARPES end station of the I05 beam line at the DIAMOND light source, United Kingdom [31]. The first-principles calculations were carried out by a projector-augmented wave method implemented in the Vienna \textit{Ab initio} Simulation Package with generalized gradient approximation [32–36]. We have also calculated the topological SS by means of the Green’s function method by using the WANNIER90 and WANNIERTOOLS packages [37,38]. For details, see the Appendixes.
III. RESULTS AND DISCUSSION

We at first discuss the band structure of EuIn$_2$As$_2$ in the PM phase. Figure 1(c) displays the energy distribution curve (EDC) in a wide energy range measured at a photon energy ($h\nu$) of 90 eV at $T = 35$ K. One can recognize core-level peaks which are assigned to the As 3d, In 4d, and Eu 4f orbitals. The sharp spectral feature and the absence of core-level peaks from other elements confirm the clean sample surface. As shown in the EDC near $E_F$ measured at $h\nu = 427$ eV with enhanced photoionization cross section of the Eu 4f orbital [39], the Eu 4f states are located at a binding energy ($E_B$) of $\sim 1.7$ eV, consistent with the previous band-structure calculations of EuIn$_2$As$_2$ [27] and photoemission data of other Eu-based compounds [14,15]. To elucidate the bulk surface-topology (FS) topology, we have mapped out the ARPES intensity as a function of in-plane wave vector with $h\nu = 427$ eV in the $k_z \sim 0$ ($\Gamma$KM) plane of bulk BZ, as shown in Fig. 1(d). The increase of the photoelectron mean free path compared to the VUV energy range reduces, by the Heisenberg relation, the intrinsic uncertainty of the out-of-plane wave vector $k_z$ and thus allows accurate 3D band mapping [40]. One can recognize a circular intensity spot centered at $\Gamma$ which follows the periodicity of the hexagonal BZ. This is attributed to the hole pocket, as seen from the ARPES intensity in Fig. 1(f) measured along a cut crossing the $\Gamma$ point (cut A in Fig. 1(d)), which signifies a holelike band crossing $E_F$. This hole pocket has a 3D character, as visible from the ARPES-intensity mapping as a function of $k_y$ and $k_z$ in Fig. 1(e) [$k_y$ is between the $\Gamma\overline{M}$ and $\Gamma\overline{K}$ cuts of surface BZ; see solid line in Fig. 1(d)], which reveals a strong intensity around $\Gamma$ but not around $A$. Such an intensity variation as a function of $k_y$ is associated with the 3D nature of the FS, and is further corroborated with the absence of $E_F$ crossing of bands along a cut passing the A point (cut C in Fig. 1(b)). We have estimated the size of the hole pocket to be $1\%$ of the bulk BZ (about $7 \times 10^{19}$ cm$^{-3}$) of hole carriers, in agreement with the measured positive Hall coefficient and hole concentration of $6 \times 10^{19}$ cm$^{-3}$ estimated from our Hall measurement (see Appendix B for details).

To clarify the existence of the SS, we performed ARPES measurements with surface-sensitive VUV photons. Figure 2(a) displays the ARPES-intensity mapping at $E_F$ as a function of in-plane wave vector $k_y$ as measured in the PM phase ($T = 35$ K) at $h\nu = 90$ eV. One finds a circular pocket centered at $\Gamma$, similar to the case of SX photons [Fig. 1(d)]. However, a careful look one can find that this FS is much bigger than that of the bulk FS at $k_z \sim 0$ (dashed circle), suggestive of its surface origin. In fact, the ARPES intensity shown in Fig. 2(b) as a function of $k_z$ and $k_y$ displays a fairly straight FS segment unlike the case of the 3D bulk FS (dashed curves), supportive of its 2D nature. As shown in Fig. 2(c), this SS forms an outermost linearly dispersive holelike band with the Fermi vector ($k_F$) robust against $h\nu$ variation. On the other hand, one can recognize a much weaker intensity inside this holelike SS. This feature exhibits a finite $h\nu$ variation and its $k_F$ point (white arrows in cut C) is consistent with that of bulk observed with SX photons. The observed large difference in the $k_F$ points between the bulk and surface states suggests extra hole doping to the surface, likely caused by the abrupt termination of crystal at the surface [14,15].

Next, we address a key question regarding how the observed band structure is influenced by the AFM transition. Figures 3(a) and 3(b) show the ARPES intensity at $T = 35$ K (in the PM phase) measured along $k$ cuts crossing the $\Gamma$ point at $k_z \sim 0$ and the A point at $k_z \sim \pi$, respectively, which again confirms an intense holelike SS (called here the S1 band) and a broad bulk band inside the SS. When the crystal was cooled down to 6 K, well below the AFM transition temperature ($T_N = 16$ K), we observed a marked change in the electronic states. As shown by a comparison of ARPES intensity at $k_z \sim 0$ between the PM and AFM phases in Figs. 3(a) and 3(c), a weak broad feature near $E_F$ around $\Gamma$ transforms into a more intense “M”-shaped band (called B1) within 0.1 eV of $E_F$, whereas the S1 band shows no discernible change across $T_N$. Moreover, the M-shaped band does not cross $E_F$ and a finite energy gap opens at $E_F$ [note that such a gap opening can also be visible from a peak shift of EDC at point B; see Fig. 3(i)]. One can also observe another drastic change in the second and third holelike bands called B2 and B3, which degenerate at $\Gamma$ in the PM phase, but apparently split in the AFM phase. At $k_z = \pi$, a similar M-shaped band is resolved in the AFM phase [Fig. 3(d)]. This band becomes a broad holelike feature crossing $E_F$ in the PM phase while its intensity is weak [Fig. 3(b)]. Such a temperature-dependent
change is well visualized in the EDCs at representative \( k \) points [A–C in Fig. 3(c)], where a number of peaks and their location are apparently different at point B (top of the B1 band) and point C (the \( \Gamma \) point) in Fig. 3(i). These results demonstrate a remarkable reconstruction of bulk electronic states associated with the AFM transition. It is noted that the energy dispersion of the S1 band is insensitive to the AFM transition, as visible from no discernible shift in the peak energy at point A (the \( k_F \) point of S1 band) in Fig. 3(i). This suggests a weak interaction between the SS and the localized magnetic moment.

To understand in more detail the observed peculiar electronic states in the AFM phase, we have performed bulk-band-structure calculations for the AFM phase. Since there are two metastable magnetic structures in the AFM phase (AFM parallel \( c \) and AFM parallel \( b \)), and their total energies are very similar to each other \cite{27}, we have calculated the band structure for both cases and display the results in Figs. 3(e)–3(h) [note that the calculation for the PM phase is technically difficult because the standard density functional theory (DFT) codes that deal with the state at \( T = 0 \) K have difficulty in simulating the random spin state at \( T > 0 \) K in the PM phase]. One can notice that the global band structure is very similar between the two phases. For instance, one commonly finds an inverted band structure where the electronlike In 5s conduction band and the As 4p holelike valence band at \( k_z = 0 \) (\( \Gamma \)K cut) largely overlap each other around \( \Gamma \), showing an energy gap of 0.1 eV at the intersection due to the spin-orbit coupling. As a result, the topmost occupied electronic states show an M-shaped band dispersion. Such insensitivity of the low-lying bands to the magnetic structure is associated with the fact that the Eu 4f state is located far away from \( E_F \). As shown in Figs. 3(f) and 3(h), the As 4p bands move away from \( E_F \) at \( k_z = \pi \).

Besides such similarity of calculated band structure between the two phases, one finds some differences between them, such as a small band splitting along the AH cut for the AFM parallel \( b \) phase; this is attributed to the difference in the magnetic space group (\( Cmcm \) vs \( P6_3/mmc' \)) \cite{27}.

Now we discuss the characteristics of observed bands by comparing the band structure between experiments and calculations. Obviously, the S1 band observed by ARPES has no counterpart in the bulk-band calculation because the \( k_F \) point of the S1 band [arrows in Fig. 3(e); \( 0.20 A^{-1} \) from the \( \Gamma \) point] is far from the intersection of the bulk In 5s and As 5p bands [open circles; \( 0.12 A^{-1} \) from the \( \Gamma \) point]. One may argue that such a discrepancy is a consequence of heavy hole doping into the bulk crystal which results in a chemical-potential (\( \mu \)) shift of \( \sim 0.4 \) eV (see vertical dashed lines). In this case, the S1 band may coincide with the bulk band. However, such a large chemical-potential shift is unreasonable when we take into account the tiny hole carriers in the bulk crystal estimated from our Hall measurement (\( 6.5 \times 10^{19} \) cm\(^{-3} \)). The reasonable agreement in the \( k \) position of the M-shaped band top between the experiment and calculations without discernible \( \mu \) shift, together with the apparent inconsistency in the size of bulk and surface-derived FSs in Figs. 1(d) and 2(a), strongly suggests that the observed M-shaped band is of bulk origin and the bulk \( \mu \) shift is not so large in contrast to the highly hole-doped nature of the SS.

Based on the above band assignment, one can further observe some quantitative differences in the location of bands between the experiment and calculations. For instance, the bottom of electronlike In 5s states at \( \Gamma \) is much deeper in the calculation; i.e., the energy overlap between the In 5s and As 4p states is significantly overestimated in the calculation. However, it should be emphasized that, despite such a quan-
tative difference, the experimental band structure still main-
tains the inverted character (i.e., M-shaped feature), which is a
prerequisite for the realization of predicted topological phases
in the AFM state [27]. One can see from a direct comparison
of Fig. 3(c) and Figs. 3(e) and 3(g) that the experimental B2
band is pushed upward by ~0.4 eV compared to the calculated
counterpart topped at $E_F$ ~ 0.8 eV. Such a shift would be
correlated with the shallower In 5$s$ electronlike feature in the
experiment.

As shown by a comparison of the electronic states at $k_z = \pi$
in Fig. 3(d) and Figs. 3(f) and 3(h), the ARPES result and the
calculated band structure share a common feature, i.e., a
characteristic nonparabolic band dispersion called here
B4 [see dashed curve in Figs. 3(b) and 3(d)]. Since such a
nonparabolic band is absent in the experiment at $k_z \sim 0$ as
seen in Fig. 3(c), it is inferred that the VUV-ARPES data
certainly reflect the intrinsic $k_z$ variation in the bulk-band
structure. On the other hand, one can commonly recognize the
M-shaped band near $E_F$ in both Figs. 3(c) and 3(d). A weak
signature of the holelike band that resembles the dispersion of
the B2 band can also be seen in Fig. 3(d) (thin dashed
curve). This is unexpected from the calculated band structure
at $k_z = \pi$ that shows a large gap of ~0.6 eV below $E_F$,
suggesting that the ARPES intensity is influenced by both the
$k_z$-broadened and $k_z$-selective spectral weights. Such a large
$k_z$-broadening effect is likely due to the short escape depth
of photoelectrons excited by VUV photons together with the
small BZ size along $k_z$ (0.35 Å$^{-1}$), as also corroborated by
observation of an M-shaped band in a wide VUV-photon-
energy range ($h\nu = 20–40$ eV). It is also inferred that the
ARPES intensity suffers from a strong matrix-element effect,
judging from the observation that the holelike band which is
obvious in the SX data in Fig. 1(f) is not clearly visible in the
VUV data in Fig. 3(a). The appearance of the M-shaped
band in the AF phase despite the strong intensity suppression in
the PM phase suggests an interesting possibility that the
matrix-element effect is different between the PM and AFM
phases in the region where the strong band reconstruction
takes place. It is also remarked here that the linewidth of the
ARPES spectrum is not sharp enough to distinguish a possible
difference in the band structure expected from the calculations
for AFM$\parallel c$ and AFM$\parallel b$ phases. It is necessary to pin down
the actual magnetic structure from other experiments such as
neutron diffraction.

Now we discuss implications of the present results in
relation to the predicted topological phases. As illustrated in
the band diagram in Fig. 3(j), the ARPES data combined with
the band calculations suggest the occurrence of bulk-band
inversion at $\Gamma$ in the AFM phase. The Fermi level is located
close to, but slightly above, the valence-band maximum due to
the hole doping. While the observed S1 band may merge into
the bulk conduction or valence bands, their actual connectivity
is unclear due to the hole-doped nature. Nevertheless, it is
inferred that the S1 band is a trivial SS originating from the
As-terminated surface and would likely be connected to the
valence band [see Fig. 3(j)], since a similar SS was predicted
for the P-terminated surface of a sister compound EuSn$_2$P$_2$
and confirmed by ARPES [15]. We speculate that, besides the
S1 band, another topological Dirac-cone SS that is connected
to the B1 band may emerge in mainly the above-$E_F$ region
in EuIn$_2$As$_2$ (schematically shown by gray curves) [27] since
the band inversion must be accompanied by the Dirac-cone SS
within the spin-orbit gap. The existence of such a topological
SS is also supported by our first-principles band calculations
by means of the Green’s function method (see Appendix C
for details). A careful look at the EDC at $T = 6$ K at point B
shown in the inset to Fig. 3(j) suggests that there exists a Fermi
edge cutoff besides a hump slightly away from $E_F$ originating
from the M-shaped band. This suggests that the observed gap
at point B is not a full gap but an imperfect gap. This may
be consistent with the appearance of a weak topological SS
crossing $E_F$ although this conjecture should be verified by
visualizing the full energy dispersion by experiments such as
two-photon ARPES.

According to the theory [27], the predicted axion-insulator
phase coexisting with the exotic TCI (for AFM$\parallel b$) or higher-
order TI (for AFM$\parallel c$) requires an inverted band structure at $\Gamma$
as well as a gapped Dirac-cone band on some surfaces. Thus,
from the present ARPES result, it can be said that EuIn$_2$As$_2$
in the AFM phase is either a TI or an axion insulator, depending
upon whether or not a finite magnetic gap opens at the Dirac
point in the AFM phase. Clarification of the actual magnetic
structure and observation of the Dirac-cone SS modulated by
the antiferromagnetism are the next important steps to further
investigate the exotic physical properties associated with the
topological characteristics of EuIn$_2$As$_2$.

IV. SUMMARY

In conclusion, the present ARPES study of EuIn$_2$As$_2$ has
revealed a band reconstruction associated with the AFM tran-
sition together with a signature of bulk-band inversion in the
AFM phase, consistent with the band calculation that supports
the axion-insulator phase in EuIn$_2$As$_2$. The present study lays
a foundation for understanding the interplay between AFM
order and nontrivial topology.

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APPENDIX A: SAMPLE FABRICATION, ARPES EXPERIMENTS, AND BAND CALCULATIONS

High-quality EuIn$_2$As$_2$ single crystals were synthesized by the flux method. High-purity Eu (lump), In (shot), and As (lump) were loaded into an Al$_2$O$_3$ crucible with the atomic ratio of Eu:In:As = 1:12:3, and sealed into a quartz tube in a vacuum of $2 \times 10^{-6}$ Torr. The tube was heated up to 1373 K in 20 h and held for 10 h at this temperature, then slowly cooled down to 973 K at a rate of 2 K/h, at which the flux was removed by centrifuge. Shiny crystals with typical size of $2 \times 2 \times 0.2$ mm$^3$ were obtained.

SX and VUV ARPES measurements were performed with energy-tunable synchrotron light at the SX-ARPES end station of the ADDRESS beam line at the Swiss Light Source (SLS), Paul Scherrer Institute, Switzerland [30], and the HR-ARPES end station of the I05 beam line in the DIAMOND light source, United Kingdom [31], respectively. SX- and VUV-ARPES measurements were performed with 320–480 and 20–120 eV photons with SX- and VUV-ARPES measurements were performed with 40–60 and 6–25 meV, respectively. The energy resolutions for SX- and VUV-ARPES measurements were 10–100 eV, respectively. Samples were cleaved in situ along the (001) crystal plane in an ultrahigh vacuum of $1 \times 10^{-10}$ Torr. The tube was heated up to 1373 K in 20 h and held for 10 h at this temperature, then slowly cooled down to 973 K at a rate of 2 K/h, at which the flux was removed by centrifuge. Shiny crystals with typical size of $2 \times 2 \times 0.2$ mm$^3$ were obtained.

SX and VUV-ARPES measurements were performed with 320–480 and 20–120 eV photons with SX- and VUV-ARPES measurements were performed with 40–60 and 6–25 meV, respectively. Samples were cleaved in situ along the (001) crystal plane in an ultrahigh vacuum of $1 \times 10^{-10}$ Torr. 

The first-principles calculations were carried out by a projector-augmented wave method implemented in the Vienna Ab initio Simulation Package (VASP) [32] with generalized gradient approximation (GGA) [33] and the Perdew-Burke-Ernzerhof (PBE) [34] type exchange-correlation potential. Fully optimized lattice constants of $a = b = 4.2951$ Å, and $c = 17.9583$ Å, were used for the further calculations. The spin-orbit coupling was included self-consistently. A uniform grid of $13 \times 13 \times 5$ was used for sampling the BZ [35]. The energy cutoff was set as 400 eV. To better process the on-site Coulomb interactions, the Hubbard $U$ parameter was set as 5 eV for Eu 4$f$ electrons in the GGA+$U$ calculations [36].

APPENDIX B: HALL MEASUREMENT

Figure 4 shows the magnetic field dependence of $\rho_{yx}$ at $T = 2$ K (black) and 300 K (red). One can see that the $\rho_{yx}$ shows a linear dependence on magnetic field regardless of temperature. The positive slope indicates that the dominant carrier is a hole in this system. The hole density $p$ at $T = 2$ K was estimated to be $6.5 \times 10^{19}$ cm$^{-3}$ from the relationship $p = 1/eR_H$, where $R_H$ is the Hall coefficient and $e$ is the elementary charge.

APPENDIX C: BAND-STRUCTURE CALCULATIONS FOR THE TOPOLOGICAL SURFACE STATE

To numerically simulate the topological SS, we have carried out first-principles band-structure calculations by means of the Green’s function method by using the WANNIER90 package [37] to generate the maximally localized Wannier functions for $d$ and $f$ orbitals of Eu, $s$ and $p$ orbitals of In, and $p$ orbitals of As. By using the WANNIER90 package [38], we have calculated the energy dispersion of the topological SS on the (001) surface for two types of AFM order (AFM$|b$ and AFM$|c$). As shown in Figs. 5(b) and 5(c), the calculated ARPES intensity profile around the $\Gamma M$ point for the Eu-terminated surface [red line in Fig. 5(a)] signifies a broad intensity distribution from the M-shaped bulk valence band below $E_F$ and the W-shaped bulk conduction band above $E_F$ (white and light red region), consistent with the bulk-band calculation shown in Figs. 3(e)–3(h). One can recognize in Fig. 5(b) a couple of sharp dispersive bands (red curves) in the projection gap of the bulk bands (blue region) that traverse the gap between bulk valence and conduction bands. These bands are attributed to the topological Dirac-cone SS associated with the bulk-band inversion. For AFM$|b$, a largely deformed gapless Dirac-cone band is found to be shifted along the in-plane wave vector [Fig. 5(b)], while, for AFM$|c$, a finite energy gap opens at the Dirac point. This is qualitatively similar to the previous report [27], and theoretically supports the axion-insulator phase in the AFM$|c$ state of EuIn$_2$As$_2$. To examine the influence of surface termination on the calculated band structure, we have also carried out the calculation for
and the As-terminated surface, and show the result in Figs. 5(d) and 5(e). One can recognize that the shifted and gapped Dirac cones for the AFM$|b|$ and AFM$|c|$ phases, respectively, are also seen in this calculation, whereas the Dirac point for both AFM$|b|$ and AFM$|c|$ phases sinks well below $E_F$ relative to that of the Eu-terminated surface [Figs. 5(b) and 5(c)]. This suggests that the energy position of topological SS relative to that of the bulk band is highly surface-condition dependent.

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