BL-28A/2021S2-001
BL-2A/2020G669

μ-ARPES study of Dirac semimetal candidate XMg$_2$Bi$_2$ (X=Ba and Sr)

Daichi TAKANE[1], Yusuke KUBOTA[1], Kosuke NAKAYAMA[1,2,*], Tappei KAWAKAMI[1], Kunihiko YAMAUCHI[3], Seigo SOUMA[4,5], Takemi KATO[1], Katsuaki SUGAWARA[1,2,4], Miho KITAMURA[6], Koji HORIBA[6,7], Hiroshi KUMIGASHIRA[8], Tamio OGUCHI[9,10], Takashi TAKAHASHI[1,4,5], Kouji SEGAWA[11], and Takafumi SATO[1,4,5,12,**]

[1]Dept. Phys., Tohoku Univ., [2]JST-PRESTO, [3]CPIER, Kyoto Univ., [4]CSRN, Tohoku Univ., [5]WPI-AIMR, Tohoku Univ.,[6]IMSS-PF, KEK, [7]QST, [8]IMRAM, Tohoku Univ., [9]CSRN, Osaka Univ., [10]ISIR, Osaka Univ., [11]Dept. Phys., Kyoto Sangyo Univ., [12]SRIS, Tohoku Univ.

1 Introduction

Topological Dirac semimetals (TDSs) materialize a new state of quantum matter which hosts exotic quantum phenomena such as high carrier mobility, giant linear magnetoresistance, and Fermi-arc-mediated surface transport. Recently, it was proposed that a ternary compound $BaMg_2Bi_2$ is a TDS from the automated construction of Wannier functions and high-throughput screening out of non-topological materials. In $BaMg_2Bi_2$ crystal, Ba atoms can be easily replaced with other elements, and its general chemical composition is expressed as XMg$_2$Bi$_2$ where X represents alkaline earth or rare earth metal (Ca, Sr, Ba, Yb, Eu, etc.). XMg$_2$Bi$_2$ crystalizes in the $CaAl_2Si_2$-type structure (space group of *P3m1*, No. 16436 which has the C_3 rotational symmetry with respect to the c-axis. Our first-principles band-structure calculations including spin-orbit coupling (SOC) reveal that $BaMg_2Bi_2$ possesses a Dirac cone close to the Fermi level (E_F) along the ΓA line of bulk Brillouin zone (BZ) due to the bulk-band inversion and the protection by the C_3 symmetry [1]. The predicted simple band structure with no E_F crossings of other bands with topologically trivial origin makes $BaMg_2Bi_2$ an excellent system to search for exotic properties associated with bulk Dirac fermions. Moreover, the XMg$_2$Bi$_2$ family has a high potential to realize different types of topological states. For example, controlling the SOC by replacing the X element and breaking the crystal symmetry e.g. by applying pressure may trigger the topological phase transition to other phases such as TI and ordinary insulator phases. Despite such interesting proposals, the electronic states of XMg$_2$Bi$_2$ family have been scarcely investigated. To clarify the fundamental band structure, we have performed μ-ARPES and SX-ARPES measurements on the single crystals of $BaMg_2Bi_2$ and $SrMg_2Bi_2$ [1].

2 Experiment

XMg$_2$Bi$_2$ single crystals were synthesized by the self-flux method. A 2θ–θ X-ray diffraction scan confirmed that the growth facet is (0001) plane and the obtained c-axis lattice constant is consistent with that of the target phase.

VUV-ARPES measurements with micro-focused synchrotron light were performed with a DA30 electron analyzer at BL28 in Photon Factory [2]. SX-ARPES measurements were performed with a SES2002 analyzer at BL2 in Photon Factory with 230–600 eV photons with horizontal linear polarization. The energy resolutions for VUV- and SX-ARPES measurements were set to be 10–30 meV and 150 meV, respectively. Samples were cleaved *in situ* along the (0001) plane of the hexagonal crystal in an ultrahigh vacuum of 1×10^{-10} Torr, and kept at T = 4.5–40 K during the measurements.

3 Results and Discussion

To determine the 3D bulk electronic states of valence band (VB), we performed ARPES measurements at the normal-emission setup with varying $h\nu$ in the SX region (250–522 eV). Figure 1(a) shows the obtained valence band dispersion along the wave vector perpendicular to the sample surface (k_z). One can recognize some energy bands displaying a finite k_z dispersion, e.g., at the binding energies E_B's of E_F-1.5 eV, 2–3 eV, and 3.7–4.3 eV. The observed band dispersions well follow the periodicity of the bulk Brillouin zone; for example, the near-E_F band appears to have the top and bottom of dispersion at Γ and A points, respectively, consistent with the bulk-band calculations including SOC (red curves). A good matching of periodic oscillation in the band dispersion between the

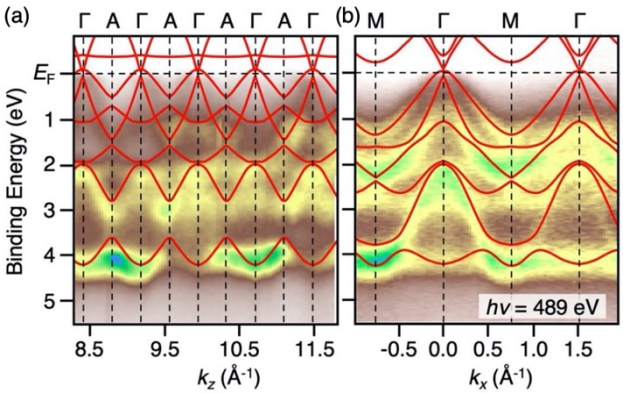

Fig. 1 (a) ARPES intensity of $BaMg_2Bi_2$ measured at T = 40 K along the ΓA cut by varying $h\nu$ from 250 to 522 eV, together with the calculated band structure (red curves). The inner potential was estimated to be V_0 = 15.0 eV from the periodicity of out-of-plane band dispersion. (b) ARPES intensity along the ΓM cut measured at $h\nu$ = 489 eV. Calculated band structure along the ΓM cut is also overlaid [1].

experiment and calculation is seen in the entire E_B region, signifying their bulk origin.

Fig. 1b shows the band-structure map along the in-plane k_x cut crossing the Γ point (ΓM cut) at $k_z \sim 0$. One can recognize several bands whose energy dispersion is reproduced semi-quantitatively by the band calculation along the ΓM cut, suggesting no discernible band-mass renormalization, indicative of a weak electron correlation in $BaMg_2Bi_2$. One can also see two hole bands which rapidly disperse toward E_F with approaching the Γ point. The outer one appears to cross E_F and forms a hole pocket centered at the Γ point.

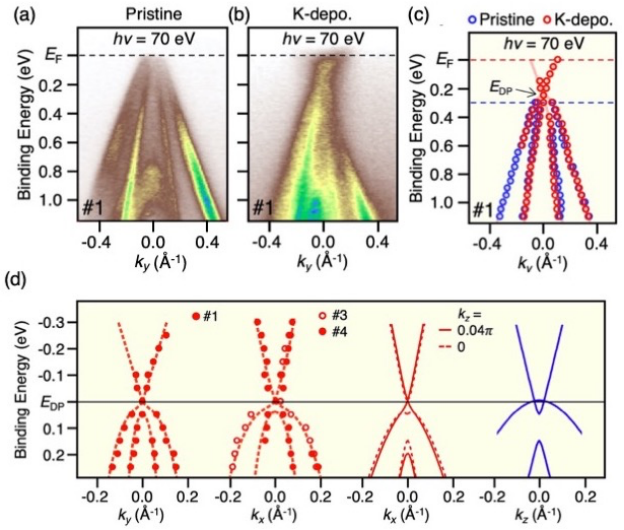

Fig. 2 (a) ARPES intensity in the vicinity of E_F for $BaMg_2Bi_2$ measured nearly along the ΓK cut at $h\nu = 70$ eV. (b) Same as (a) but after K deposition onto the surface of $BaMg_2Bi_2$. (c) Experimental band dispersions for pristine and K-deposited $BaMg_2Bi_2$, obtained by tracing the peak position of MDCs in (a) and (b). (d) Experimental band dispersion (circles and dashed curves) obtained from the MDC analyses along cuts #1, #3, and #4 passing the Γ point, compared with the calculated dispersions at $k_z = 0$ (ΓM cut; red dashed curves) and 0.04 Å−1 (**k** cut crossing the Dirac point; red solid curves), with that along the k_z axis (ΓA cut; blue curves) [1].

Figure 2(a) shows the ARPES intensity in the vicinity of E_F, measured along the ΓK (k_y) cut at $h\nu = 70$ eV. One can see linearly dispersive inner and outer hole bands, and the latter crosses E_F. Since hole carriers are doped into the crystal due to the slight off-stoichiometry of chemical composition, we were unable to see the bulk conduction band (CB) on the bare surface. To search for a possible band touching of VB and CB predicted in the calculation, we deposited K atoms onto the surface of $BaMg_2Bi_2$ in ultrahigh vacuum. One can clearly see in Fig. 2(b) that the hole bands in K-deposited $BaMg_2Bi_2$ are shifted downward as a whole by ~ 0.3 eV with respect to those in pristine $BaMg_2Bi_2$ due to the electron doping to the surface. Intriguingly, VB in the negative k_y region continuously disperse across $k_y = 0$ towards positive k_y region without losing its intensity, suggestive of the absence of band gap. Taking account of the symmetry of band in momentum space, it is suggested that the VB and CB touch each other at the Γ point and forms the Dirac-cone-like dispersion. We show in Fig. 2(c) the experimental band dispersion obtained by tracing the peak position of MDCs. One can see that, in K-deposited $BaMg_2Bi_2$, the peak for the bulk VB is smoothly connected to that for the bulk CB without obvious discontinuity around the Γ point.

Two panels on the left side of Fig. 2(d) show the experimentally determined energy position of VB and CB from numerical fittings of MDCs at several cuts around different Γ point. We found that outer hole band appears to touch the CB at the Γ point within our experimental uncertainty. The experimental band dispersions (left two panels) show an overall agreement with the calculated band dispersions along the ΓM cut ($k_z = 0$; dashed curves, slightly away from the Dirac point) and off-ΓM cut (k_z = 0.04 Å^{-1}; solid curves, passing through the Dirac point) as shown in the second panel from the right in Fig. 2(d). Although it is difficult to resolve each Dirac point due to the inevitable k_z broadening effect, the overall agreement between the experimental and calculated band dispersions is consistent with the existence of Dirac points around the Γ point, and therefore supportive of the TDS nature of $BaMg_2Bi_2$.

Acknowledgements

We thank Y. Saruta, T. Taguchi, N. Watanabe, and R. Tsubono for their assistance in the ARPES experiments. We thank K. Nagata for his assistance in the preparation of experiments. This work was supported by JST-CREST (No: JPMJCR18T1), JST-PRESTO (No: JPMJPR18L7), JSPS (JSPS KAKENHI No: JP17H01139, JP18H01160, JP19H01845, JP20H01847), Grant-in-Aid for JSPS Research Fellow (No: JP18J20058). D.T. thanks JSPS and Tohoku University Division for Interdisciplinary Advanced Research and Education, T. Kawakami and T. Kato thank GP-Spin for financial support.

References

[1] D. Takane, Y. Kubota, K. Nakayama, T. Kawakami, K. Yamauchi, S. Souma, T. Kato, K. Sugawara, S. Ideta, K. Tanaka, M. Kitamura, K. Horiba, H. Kumigashira, T. Oguchi, T. Takahashi, K. Segawa, and T. Sato, *Sci. Rep.* **11**, 21937 (2021).

[2] M. Kitamura, S. Souma, A. Honma, D. Wakabayashi, H. Tanaka, A.Toyoshima, K. Amemiya, T. Kawakami, K. Sugawara, K. Nakayama, K. Yoshimatsu, H. Kumigashira, T. Sato, and K. Horiba, Rev. Sci. Instrum. **93**, 033906 (2022).

* k.nakayama@arpes.phys.tohoku.ac.jp
** t-sato@arpes.phys.tohoku.ac.jp