Topological Surface States with Persistent High Spin Polarization across Dirac Point in Bi$_2$Te$_2$Se and Bi$_2$Se$_2$Te

K. Miyamoto$^1$, A. Kimura$^2$, T. Okuda$^1$, H. Miyahara$^2$, K. Kuroda$^2$, H. Namatame$^1$, M. Taniguchi$^1,2$, S. V. Eremeev$^{3,4}$, T. V. Menshchikova$^4$, E. V. Chulkov$^{5,6,7}$, K. A. Kokh$^8$, and O. E. Tereshchenko$^{9,10}$

$^1$Hiroshima Synchrotron Radiation Center, Hiroshima University, 2-3-1 Kagamiyama, Higashi-Hiroshima 739-0046, Japan
$^2$Graduate School of Science, Hiroshima University, 1-3-1 Kagamiyama, Higashi-Hiroshima 739-8526, Japan
$^3$Institute of Strength Physics and Materials Science, Tomsk State University, 634050, Tomsk, Russia
$^4$Tomsk State University, 634050, Tomsk, Russia
$^5$Departamento de Física de Materiales UPV/EHU and Centro de Física de Materiales CFM and Centro Mixto CSIC-UPV/EHU, 20080 San Sebastián/Donostia, Basque Country, Spain
$^6$Donostia International Physics Center (DIPC), 20018 San Sebastián/Donostia, Basque Country, Spain
$^7$Centro de Física de Materiales CFM-Materials Physics Center MPC, Centro Mixto CSIC-UPV/EHU, Edificio Korta, Avenida de Tolosa 72, 20018 San Sebastián, Spain
$^8$V.S. Sobolev Institute of Geology and Mineralogy, Siberian Branch, Russian Academy of Sciences, Koptyuga pr. 3, Novosibirsk, 630090 Russia
$^9$Institute of Semiconductor Physics, Siberian Branch, Russian Academy of Sciences, pr. Akademika Lavrent’eva 13, Novosibirsk, 630090 Russia and
$^{10}$Novosibirsk State University, ul. Pirogova 2, Novosibirsk, 630090 Russia

(Dated: May 5, 2014)

Helical spin textures with the marked spin polarizations of topological surface states have been firstly unveiled by the state-of-the-art spin- and angle-resolved photoemission spectroscopy for two promising topological insulators Bi$_2$Te$_2$Se and Bi$_2$Se$_2$Te. The highly spin-polarized nature is expected to be persistent across the Dirac point in both compounds. This novel finding paves a pathway to extending their utilization of topological surface state for future spintronic applications.

PACS numbers: 73.20.-r, 79.60.-i, 71.70.Ej.

Three-dimensional topological insulators (3D TIs) with massless helical Dirac fermions at the surface in a bulk energy gap induced by a strong spin-orbit coupling have attracted a great attention as key materials for revolutionize current electronic devices [1–4]. A spin splitting have attracted a great attention as key materials with massless helical Dirac fermions at the surface in 3D TIs [5, 6].

Helical texture of topological surface state (TSS), where the electron spin is locked to its momentum, is a manifestation of significant efforts to realize the surface isolated transport, the progress has thus been hampered by too small surface contribution in the total conductance [24–27] because the uncontrolled bulk carrier doping takes place in Bi$_2$Se$_3$ and Bi$_2$Te$_3$. The absence of such a topological nature at TSS below $E_D$ restricts its variety of spintronics applications. Futhermore, in spite of significant efforts to realize the surface isolated transport, the progress has thus been hampered by too small surface contribution in the total conductance [24–27] because the uncontrolled bulk carrier doping takes place in Bi$_2$Se$_3$ and Bi$_2$Te$_3$ due to the Se vacancy and the Bi-Te antisite defect.

Recently, one of the ternary tetradymite compounds, Bi$_2$Te$_2$Se, where the central Te layer is replaced with the Se layer in Bi$_2$Te$_3$, was shown to be a 3D TI by the

---

*Electronic address: kmiyamoto@hiroshima-u.ac.jp*
ARGES measurement [28, 29]. Importantly, the suppression of the bulk conductivity is anticipated because the well-confined Se atoms in the central layer are expected to suppress the Se vacancy as well as the antisite defects between Bi and Te atoms. Actually, a highly bulk resistive feature in this compound has successfully led to the observation of its surface-derived quantum oscillations in the magnetotransport experiment [30]. Another ternary compound Bi$_2$Se$_2$Te has also been predicted to be a 3D TI but not experimentally verified yet [31].

Here a question arises for these compounds: how are these spin polarized natures maintained in their TSSs? Realization of the TSS with high spin polarization in the wide energy range across the Dirac point is crucial for the ambipolar gate control of the TI devices [32, 33]. One can also manipulate the spin orientations by tuning the electron filling level in the TSS (Fig.1(a)) as expected for the dual gate TI device [34]. In this Letter, we have unambiguously clarified for the first time by the second variation method [35], where interaction between the ion cores and valence electrons was described by the projector augmented-wave method [36]. The generalized gradient approximation was used to describe the exchange correlation energy. The Hamiltonian contained the scalar-relativistic corrections, and the spin-orbit coupling was taken into account by the second variation method.

Figures 2(a) and 2(c) show the ARPES energy dispersion curves of the TSS along $\Gamma_2$-$\Gamma_{2nd}$-$\Gamma$ direction for Bi$_2$Te$_2$Se and Bi$_2$Se$_2$Te taken with He discharge lamp ($h\nu$ = 21.22 eV). We find that the TSSs for both compounds are pronounced rather at $\Gamma_{2nd}$ than at $\Gamma_{1st}$ (not shown). Here, $\Gamma_{2nd}$ ($\Gamma_{1st}$) denotes the $\Gamma$ point in the second (first) surface Brillouin zone (SBZ). Figures 2(b) and 2(d) highlight intensity maxima of one TSS branch obtained from momentum distribution curves in the limited momentum space. One can see that the $E_D$ is located at the binding energy, $E_B$ of 415 meV in Bi$_2$Te$_2$Se and 425 meV in Bi$_2$Se$_2$Te. The observed dispersions at $E_B < E_D$ are almost perfectly linear as demonstrated in Figs. 2(b) and 2(d). On the other hand, the dispersions at $E_B > E_D$ are less steep. The band dispersions for both Bi$_2$Te$_2$Se and Bi$_2$Se$_2$Te are in excellent agreement with that produced by the first principles calculation as shown in Fig.2 (b) and 2 (d).
TABLE I: Binding energy at Dirac point (E_D), group velocity at E_D and at Fermi energy (E_F) of Bi_2Te_2Se and Bi_2Se_2Te.

| E_D [meV] | v_g (E_D) × 10^5 (m/s) | v_g (E_F) × 10^5 (m/s) |
|-----------|-------------------------|-------------------------|
| 415±3     | 4.2±0.4                 | 5.8±0.2                 |
| 425±3     | 3.7±0.2                 | 6.1±0.2                 |

The group velocity at the Fermi energy (Fermi velocity) is estimated with using the formula 1/h(∂E/∂k) to be (5.8±0.2)×10^5 m/s in Bi_2Te_2Se and (6.1±0.2)×10^5 m/s in Bi_2Se_2Te. Also, the group velocity (v_g) near E_D is larger for Bi_2Te_2Se than for Bi_2Se_2Te as listed in Table I. The value at E_D for Bi_2Te_2Se is close to v_g=4.6×10^5 m/s above 130 meV from E_D estimated by the magnetotransport measurement [30]. Here, the v_g near E_D is larger than that for Bi_2Se_3 (v_g=2.9×10^5 m/s), while the Fermi velocity is smaller (v_g=6.6×10^5 m/s) [10, 16]. The TSSs in the existing TI materials are usually deviated from a linear dispersion in going away from E_D. The ratios of v_g at E_F with respect to that at E_D for Bi_2Te_2Se and Bi_2Se_2Te are found to be 1.4 and 1.6, respectively. These values are smaller than those of the other 3D TIs (1.8 for TIBiSe_2 and 2.3 for Bi_2Se_3 though their Dirac points are deeper than in the present case), which means that the surface band dispersions for Bi_2Te_2Se and Bi_2Se_2Te possess the wider energy range where the linearly dispersive feature is maintained above E_D.

To unravel the spin character and make a quantitative analysis of spin polarization in these TSSs, we have performed the SARPES measurement. Figures 3(a) and 3(b) show the spin-resolved energy distribution curves (EDCs) of Bi_2Te_2Se and Bi_2Se_2Te. Here, the spin-up and spin-down spectra are plotted with triangles pointing-up and -down, respectively. Let us first take a look at the spin-resolved EDCs of Bi_2Te_2Se. A spin-up peak near E_F at θ=49.5° shifts to higher E_B with increasing θ. A spin-down peak at 0.57 eV emerges from θ=51.5° and move to lower E_B with increasing θ. These spin-up and -down peaks are merged at θ=56° corresponding to Γ_2nd (E_B=0.4 eV). This result clearly shows that the TSS is spin split and the spin orientations are antisymmetric with respect to the Γ point. For the peak near E_F for θ = 53.5° - 58.5°, the spin-down intensity is slightly larger than that in the spin-up channel, which might originate from final state effect [23]. The observed features of band dispersion and the spin polarizations in Bi_2Se_2Te are in agreement with those in Bi_2Te_2Se as shown Fig. 3(b). The similarity in these compounds with different stoichiometry is somewhat surprising, whose origin will be further investigated in near future. In Fig. 3 (c) and 3 (d), we have summarized the observed spin-up and -down peaks in Figs. 3 (a) and 3 (b) with triangles pointing-up and -down, respectively.

Importantly, when there are clear peak structures in one spin channel, no apparent peak emerges in the opposite spin component over the whole energy range as shown in Figs. 3 (a) and 3 (b). The results clearly show the presence of the surface Dirac fermions with high degree of spin polarizations in the bulk energy gap region. In fact, the TSS in Bi_2Te_2Se (Bi_2Se_2Te) maintains a high spin polarization more than 40% (50%) in a wide energy region across the Dirac point, though a non-polarized inelastic background contributes to the total intensity. Here, the spin polarizations are corrected from the raw data by subtracting the constant and unpolarized background from the originally derived spin-up and spin-down spectra [21]. In particular, the spin polarization of TSS near E_F with the smaller background contribution Bi_2Te_2Se (Bi_2Se_2Te) reaches more than 70% (50%). It was, however, quite difficult to estimate the accurate values of spin polarizations near E_F in case of the measurement with He lamp due to the overlap of additional signal with considerable spin polarizations excited by the higher-energy satellite (β) line (hν=23.08 eV). Therefore,
FIG. 3: (a), (b) Spin-resolved energy distribution curves (EDCs) of Bi$_2$Te$_2$Se and Bi$_2$Se$_2$Te along $\Gamma M$ line obtained with unpolarized He-I$\alpha$ radiation. Spin-up and spin-down intensities are denoted with triangles pointing up (red) and down (blue). (c), (d) E-k mapping ARPES measurements with He lamp. The spin characters derived from spin-resolved spectra in Figs. 3 (a) and 3 (b) are superimposed by triangle pointing up and down. Here, $k_y$ is measured from $\Gamma$$_{2nd}$ point. (e) Spin-resolved energy distribution curves and spin polarizations of Bi$_2$Te$_2$Se at $\theta$ = $\pm$4.2$^\circ$ taken with p-polarized synchrotron radiation ($h\nu$=17.2 eV). (f) Theoretical spin polarization values as a function of wavenumber obtained by the first principles calculation for Bi$_2$Te$_2$Se and Bi$_2$Se$_2$Te.

We have tried to use the synchrotron radiation (SR) to extract accurate values. Figure 3(e) shows the SARPES results in Bi$_2$Te$_2$Se obtained by the monochromatic SR light ($h\nu$=17.2 eV). As shown in the lower panel of Fig. 3 (e), the magnitude of spin polarization for the surface state reaches 87±9% at $\theta$ = -4.2$^\circ$ and -67±3% at $\theta$ = 4.2$^\circ$. The difference of spin polarizations in positive and negative $\theta$ is probably derived from matrix elements for optical transitions. If the final-state spin polarizations are assumed to be equal in positive and negative $\theta$ near $\Gamma$, an averaged spin polarization of 77±5% is regarded as the absolute value of the initial-state spin polarization. Figure 3 (f) shows the theoretical spin expectation values as a function of wavenumber ($k_y$) for Bi$_2$Te$_2$Se and Bi$_2$Se$_2$Te obtained from the first principles calculation. Here, we find that the theoretical spin polarization for Bi$_2$Te$_2$Se (Bi$_2$Se$_2$Te) increases in going closer to $\Gamma$ point and it takes the maximum value of $\sim$75% ($\sim$70%) at $\Gamma$. Note that the spin polarization can not reach 100% because the spin angular momentum is not a good quantum number any more due to the strong spin-orbit entanglement as reported for Bi$_2$Te$_3$ and Bi$_2$Se$_3$ [34]. The experimentally evaluated spin polarization (77%) for TSS in Bi$_2$Te$_2$Se at $k_y$ = ±0.134 Å$^{-1}$ is larger than the theoretical value ($\sim$61%). It is worth noting that the latter is higher in Bi$_2$Te$_2$Se as compared with Bi$_2$Te$_3$, where reverse spin direction was found at outer Te atom [40]. In contrast to that in Bi$_2$Te$_2$Se the layer projected spin analysis revealed identical spin helicity for all atomic layers. Moreover, the spin polarization near $E_D$ is expected to show the higher value than those near $E_F$ as the calculated spin polarization implies although it is difficult to experimentally evaluate the accurate value of spin polarization near Dirac point as mentioned above.

In conclusion, the helical spin texture and the spin polarizations of TSS in the ternary tetradymite chalcogénides Bi$_2$Te$_2$Se and Bi$_2$Se$_2$Te have been experimentally revealed by the SARPES measurement. The markedly high spin polarization of topological surface states has been found to be $\sim$77% and is persistent in wide energy range across the Dirac point in those compounds. The availability of both upper and lower TSSs promises to extend the variety of spintronic applications, for instance, to the dual gate TI device and the topological p-n junc-
We thank Shuichi Murakami for valuable comments. This work was financially supported by KAKENHI (Grant No. 20340092, 23340105), Grant-in-Aid for Scientific Research (B) of JSPS. K.A.K. and O.E.T. acknowledge financial support by the RSSF and RFBR (Grant No. 12-02-00226).

[1] L. Fu, and C. L. Kane, Phys. Rev. B 76, 045302 (2007).
[2] L. Fu, C. L. Kane, and E. J. Mele, Phys. Rev. Lett. 98, 160803 (2007).
[3] X. L. Qi, T. L. Hughes, and S. C. Zhang, Phys. Rev. B 78, 195424 (2008).
[4] M. Z. Hasan, and C. L. Kane, Rev. Mod. Phys. 82, 3045 (2010).
[5] X. L. Qi, R. D. Li, J. D. Zhang, and S. C. Zhang, Science 323, 1184 (2009).
[6] L. Fu, and C. L. Kane, Phys. Rev. Lett. 100, 096407 (2008).
[7] Y. L. Chen, J. G. Analytis, J.-H. Chu, Z. K. Liu, S.-K. Mo, X. L. Qi, H. J. Zhang, D. H. Lu, X. Dai, Z. Fang, S. C. Zhang, I. R. Fisher, Z. Hussain, Z.-X. Shen, Science 325, 178 (2009).
[8] H. Zhang, C. X. Liu, X. L. Qi, X. Dai, Z. Fang and S. C. Zhang, Nature Phys. 5, 438 (2009).
[9] Y. Xia, D. Qian, D. Hsieh, L. Wray, A. Pal, H. Lin, A. Bansil, D. Grauer, Y. S. Hor, R. J. Cava and M. Z. Hasan, Nature Phys. 5, 398 (2009).
[10] K. Kuroda, M. Arita, K. Miyamoto, M. Ye, J. Jiang, A. Kimura, E. E. Krasovskii, E. V. Chulkov, H. Iwasawa, T. Okuda, K. Shimada, Y. Ueda, H. Namatame, and M. Taniguchi, Phys. Rev Lett 105, 076802 (2010).
[11] B. Yan, C. X. Liu, H. J. Zhang, C. Y. Yam, X. L. Qi, T. Frauenheim, and S. C. Zhang, Euro Phys. Lett. 90, 37002 (2010).
[12] H. Lin, S. R. Markiewicz, L. A. Wray, L. Fu, M. Z. Hasan, and A. Bansil, Phys. Rev. Lett. 105, 036404 (2010).
[13] S. V. Eremeev, G. Bihlmayer, M. Vergniory, Yu. M. Koroteev, T.V. Menschikova, J. Henk, A. Ernst, and E. V. Chulkov, Phys. Rev. B 83, 205129 (2011).
[14] T. Sato, K. Segawa, H. Guo, K. Sugawara, S. Souma, T. Takahashi, and Y. Ando, Phys. Rev. Lett. 105, 136802 (2010).
[15] K. Kuroda, M. Ye, A. Kimura, S. V. Eremeev, E. E. Krasovskii, E. V. Chulkov, Y. Ueda, K. Miyamoto, T. Okuda, K. Shimada, H. Namatame, and M. Taniguchi, Phys. Rev. Lett. 105, 146801 (2010).
[16] Y. L. Chen, Z. K. Liu, J. G. Analytis, J. H. Chu, H. J. Zhang, B. H. Yan, S. K. Ma, R. G. Moore, D. H. Lu, I. R. Fisher, S. C. Zhang, Z. Hussain, and Z. X. Shen, Phys. Rev. Lett. 105, 266401 (2010).
[17] D. Hsieh, Y. Xia, D. Qian, L. Wray, J. H. Dil, F. Meier, J. Osterwalder, L. Patthey, J. G. Checkelsky, N. P. Ong, A. V. Fedorov, H. Lin, A. Bansil, D. Grauer, Y. S. Hor, R. J. Cava, and M. Z. Hasan, Nature 460, 1101 (2009).
[18] S. Souma, K. Kosaka, T. Sato, M. Komatsu, A. Takayama, T. Takahashi, M. Kriener, K. Segawa, and Y. Ando, Phys. Rev. Lett. 106, 216803 (2011).
[19] S.-Y. Xu, L. A. Wray, Y. Xia, F. von Rohr, Y. S. Hor, J. H. Dil, F. Meier, B. Slomski, J. Osterwalder, M. Neupane, arXiv:1101.3985.
[20] Z.-H. Pan, E. Vescovo, A. V. Fedorov, D. Gardner, Y. S. Lee, S. Chu, G. D. Gu, and T. Valla, Phys. Rev. Lett. 106, 257004 (2011).
[21] C. Joziwak, Y. L. Chen, A. V. Fedorov, J. G. Analytis, C. R. Rotundu, A. K. Schmid, J. D. Denlinger, Y.-D. Chuang, D.-H. Lee, I. R. Fisher, R. J. Birgeneau, Z.-X. Shen, Z. Hussain, and A. Lanzara, Phys. Rev. B 84, 165113 (2011).
[22] P. Cheng, C. Song, T. Zhang, Y. Zhang, Y. Wang, J. F. Jia, J. Wang, Y. Wang, B. F. Zhu, X. Chen, X. Ma, K. He, L. Wang, X. Dai, Z. Fang, X. Xie, X. L. Qi, C. X. Liu, S. C. Zhang, and Q. K. Xue, Phys. Rev. Lett. 105, 076801 (2010).
[23] T. Hanaguri, K. Igarashi, M. Kawai, H. Takagi, and T. Sasagawa, Phys. Rev. B 82, 081305 (2010).
[24] J. G. Checkelsky, Y. S. Hor, M.-H. Liu, D.-X. Qu, R. J. Cava, and N. P. Ong, Phys. Rev. Lett. 103, 246601 (2009).
[25] N. P. Butch, K. Kirshenbaum, P. Syers, A. B. Sushkov, G. S. Jenkins, H. D. Drew, and J. Paglione, Phys. Rev. B 81, 241301(R) (2010).
[26] K. Eto, Z. Ren, A. A. Taskin, K. Segawa, and Y. Ando, Phys. Rev. B 81, 195309 (2010).
[27] D.-X. Qu, Y. S. Hor, J. Xiong, R. J. Cava, and N. P. Ong, Science 329, 821 (2010).
[28] S. Y. Xu, L. Wray, T. Xia, R. Shankar, A. Petersen, A. Fedorov, H. Lin, A. Bansil, Y. Hor, D. Grauer, R. Cava, and M. Hasan, arXiv:1007.5111.
[29] T. Arakane, T. Sato, S. Souma, K. Kosaka, K. Nakayama, M. Komatsu, T. Takahashi, Z. Ren, K. Segawa, and Y. Ando, Nature Comms. 3, 636 (2012).
[30] Z. Ren, A. A. Taskin, S. Sasaki, K. Segawa, and Y. Ando, Phys. Rev. B 82, 241306(R) (2010).
[31] L. L. Wang, and D. D. Johnson, Phys. Rev. B 83, 241309(R) (2011).
[32] J. Wang, X. Chen, B.-F. Zhu, and S.-C. Zhang, arXiv:1202.1579.
[33] K. Segawa, Z. Ren, S. Sasaki, T. Tsuda, S. Kuwabata, and Y. Ando, arXiv:1203.2047.
[34] O. V. Yazhev, J. E. Moore, and S. G. Louie, Phys. Rev. Lett. 105, 266806 (2010).
[35] K. A. Kohk, B. G. Nenasev, A. E. Kokh, and G. Y. Shvedenkov, J. Crystal Growth 275, e2129 (2005).
[36] T. Okuda, K. Miyamoto, H. Miyahara, K. Ueda, A. Kimura, H. Namatame, and M. Taniguchi, Rev. Sci. Instrum. 82, 103302 (2011).
[37] G. Kresse, J. Furthm¨uller, Comput. Mater. Sci. 6, 15 (1996).
[38] G. Kresse, D. Joubert, Phys. Rev. B 59, 1758 (1999).
[39] R. Feder, Polarized Electrons in Surface Physics, World Scientific, Singapore (1985).
[40] S. V. Eremeev et al., Nature Comms. 3, 635 (2012).