Direct Measurement of the Out-of-Plane Spin Texture in the Dirac Cone Surface State of a Topological Insulator

S. Souma$^1$, K. Kosaka,$^2$ T. Sato,$^2$ M. Komatsu,$^2$ A. Takayama,$^2$
T. Takahashi,$^{1,2}$ M. Kriener,$^3$ Kouji Segawa,$^3$ and Yoichi Ando$^3$

$^1$WPI Research Center, Advanced Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan
$^2$Department of Physics, Tohoku University, Sendai 980-8578, Japan
$^3$Institute of Scientific and Industrial Research, Osaka University, Ibaraki, Osaka 567-0047, Japan

(Dated: May 27, 2011)

PACS numbers: 73.20.-r, 79.60.-i, 71.20.-b, 75.70.Tj

We have performed spin- and angle-resolved photoemission spectroscopy of Bi$_2$Te$_3$ and present the first direct evidence for the existence of the out-of-plane spin component on the surface state of a topological insulator. We found that the magnitude of the out-of-plane spin polarization on a hexagonally deformed Fermi surface of Bi$_2$Te$_3$ reaches maximally 25% of the in-plane counterpart, while such a sizable out-of-plane spin component does not exist in the more circular Fermi surface of TIBiSe$_2$, indicating that the hexagonal deformation of the Fermi surface is responsible for the deviation from the ideal helical spin texture. The observed out-of-plane polarization is much smaller than that expected from existing theory, suggesting that an additional ingredient is necessary for correctly understanding the surface spin polarization in Bi$_2$Te$_3$.

The topological insulators (TIs) materialize a new state of quantum matter where an unusual gapless metallic state appears at the edge or the surface of a band insulator due to a topological principle. The surface state (SS) of three-dimensional TIs is characterized by a Dirac-cone dispersion which has been shown to have a helical spin structure where the spin vector points parallel to the spin direction and the protection by the time-reversal symmetry would be a key to better understanding the peculiar SS of the TIs, whereas an experimental verification has not yet been successfully made.

In this Letter, we demonstrate that such an OP spin component is indeed present in Bi$_2$Te$_3$, by determining for the first time the detailed IP and OP spin texture at various $k$ locations of the Dirac-cone SS using the low-energy, spin- and angle-resolved photoemission spectroscopy (SR-ARPES) [10]. We also elucidated that the magnitude of the OP spin polarization is related to the strength of the hexagonal warping of the FS. We discuss the present result in relation to theoretical proposals as well as its implications for potential applications.

High-quality single-crystals of Bi$_2$Te$_3$ were grown by melting nearly stoichiometric mixtures of high-purity (99.9999% pure) elemental shots of Bi and Te in a quartz glass tube at 850°C for 48 h and slowly cooling to 500°C over 48 h. To tune the chemical potential to be located close to the bottom of the conduction band (CB), which was crucial for the success of this work, we used a slightly Bi-rich composition of Bi:Te = 0.34:0.66. ARPES measurements were performed by using an SR-ARPES spectrometer at Tohoku University [10, 11]. We used the Xe I photons (8.437 eV) to excite photoelectrons. Samples were cleaved in-situ along the (111) crystal plane in an ultrahigh vacuum of 5×10$^{-11}$ Torr. The energy resolutions during the SR- and regular ARPES measurements were 40 and 6 meV, respectively. We used the Sherman function value of 0.07 to obtain the SR-ARPES data.

Figure 1(b) shows the FS mapping of an $n$-type Bi$_2$Te$_3$ sample. We clearly find a FS showing a strong hexagonal deformation as in previous reports [8]. This snowflake-like FS originates from an electronlike surface band [Fig. 1(c)] corresponding to the upper branch of the Dirac cone. The Dirac point is situated at the binding energy $E_B$ of 0.25 eV, suggesting that the bottom of the bulk CB is located at slightly above the Fermi level (cf. Ref. [8]); in other words, the chemical potential ($\mu$) lies slightly below the bottom of the bulk CB. This is consistent with the slightly $n$-type character of the crystal indicated by the Seebeck coefficient and is suited for examining the OP spin component because of the large hexagonal warping.
of the FS at this energy position \( \phi \). It should be emphasized here that the absence of bulk CB electrons below \( E_F \) makes it possible to quantitatively determine the net spin polarization solely from the surface band by avoiding the contamination from the bulk band.

Figure 1(d) shows spin-resolved energy distribution curves (SR-EDCs) of Bi\(_2\)Te\(_3\) for the IP spin component measured at various \( k \) locations around the FS indicated in Fig. 1(b). We define the IP spin polarization vector to point perpendicular to \( k \) [see, e.g., the thick (red) arrow in Fig. 1(b)] in which the “up spin” points to the clockwise direction. A peak near the Fermi level \( E_F \) in the up-spin EDCs originates from the upper branch of the Dirac cone, while a shoulder feature at \( E_B > 0.2 \) eV is a tail of the bulk valence band. In contrast to the small differences between the up- and down-spin EDCs for the shoulder feature at \( E_B > 0.2 \) eV, the difference for the near-\( E_F \) peak is gigantic: it is dominated by the up-spin component, indicating the basically spin helical nature of the SS where the IP spin vector points to the clockwise direction like in Fig. 1(a).

The important question is whether there is a finite OP spin component perpendicular to the sample surface, \( S_z \). We can directly address this issue with our SR-spectrometer, which allows us to simultaneously measure the OP component with the IP component, and Fig. 1(e) shows the SR-EDCs for the OP spin component. If there is no OP spin polarization, one should observe that the EDCs for \( S_z \)-up and \( S_z \)-down should completely coincide with each other. It turns out that this is not the case, and a clear \( S_z \) dependence of the EDCs was observed. For instance, around the Fermi vector \( (k_F) \) along the \( \Gamma K \) direction corresponding to the FS angle \( \phi = 0^\circ \) (points A and B), the \( S_z \)-down EDC near \( E_F \) is clearly enhanced over to the \( S_z \)-up one around the peak top, although the difference between the two EDCs is much smaller than that in the IP polarization measurements [Fig. 1(d)]. Furthermore, the relative intensity of the near-\( E_F \) peak for the \( S_z \)-down and \( S_z \)-up EDCs is reversed along another \( \Gamma K \) direction (\( \phi = 60^\circ \), which we call here the \( \Gamma K' \) direction) as shown by the EDCs at point C. This demonstrates a sign change of the OP spin polarization within the SS: Spins at the \( k \)-space locations A and B point to the sample side, whereas those at location C point to the vacuum side. Notably, there is no difference between \( S_z \)-up and \( S_z \)-down EDCs along the \( \Gamma M \) direction (point D; \( \phi = 30^\circ \)) within our experimental accuracy, suggesting that the sign change takes place along the \( \Gamma M \) line. Also, one can see in Fig. 1(e) that there is no clear difference between \( S_z \)-up and \( S_z \)-down EDCs at \( k \) near the \( \Gamma \) point (points E and F), suggesting that the \( S_z \) component vanishes as the energy position on the SS approaches the Dirac point and the hexagonal warping weakens.

To discuss quantitatively the present observation, we plot in Figs. 1(f) and (g) the energy dependence of the IP \( (P_\phi) \) and OP \( (P_z) \) spin polarizations, respectively, for three representative \( k \) points. The spin polarization \( P \) is defined as \( P = (N_\uparrow - N_\downarrow)/(N_\uparrow + N_\downarrow) \) where \( N_\uparrow \) and \( N_\downarrow \) are the intensity of the up- and down-spin states, respectively. One can clearly see that the \( P_\phi \) value increases as \( E_B \) approaches \( E_F \), and it reaches \( \sim 0.6 \) around \( E_F \). In contrast, the magnitude of \( |P_z| \) is much smaller, and at points A or C it reaches only \( \sim 0.15 \), which is about 25% of the maximum \( P_\phi \) value. We have confirmed that this characteristic behavior is reproducible by measuring several samples. In addition, we have found that the exposure of an originally \( p \)-type Bi\(_2\)Te\(_3\) samples to the helium gas in the vacuum chamber leads to the emergence of a similar snowflake-like FS due to the surface band bending [12,13] and that its spin texture is essentially the same as the bulk \( n \)-type samples. It is emphasized here that the clear observation of the OP spin component has become feasible by utilizing the high-flux and low-energy character of the xenon lamp [11] as well as an efficient SR-ARPES spectrometer [11], which enable the high-energy or \( k \) resolution measurement with higher statistics.
To analyze the $k$ dependence of the spin polarization in more detail, we plot in Figs. 2(a) and (b) the spin polarizations along the $x$- and $z$-axes measured in the $k_z=0$ cut [vertical solid (green) line in Fig. 1(b)] obtained by integrating the ARPES intensity at $E_B = 0.08-0.12$ eV; because of the rather large (40 meV) energy resolution of the spin-resolved measurements, the integration in this energy window gives a good representation of the spin polarization of the surface band as a function of $k$. One can see in Fig. 2(a) that the $P_x$ value at $k_y > 0$ ($k_y < 0$) is positive (negative), and its absolute value $|P_x|$ is nearly constant at $\sim 0.6$ for $|k_y| \geq 0.03 \text{ Å}^{-1}$. The sudden sign reversal in $P_z$ near $k_y = 0$ is consistent with the IP spin helical texture. We also found that the $z$-axis spin polarization $P_z$ shows a sign reversal along the same cut [Fig. 2(b)], as expected from the result in Fig. 1 where the $\Gamma K'$ direction at $\phi = 60^\circ$ is identical to the $\Gamma K'$ direction at $\phi = 180^\circ$ due to the three-fold rotational symmetry of the crystal. However, the magnitude of $|P_z|$ stays nearly zero at $|k_y| \leq 0.03 \text{ Å}^{-1}$ and then gradually increases with $|k_y|$, reaching $\sim 0.15$ near the $k_F$ point. This trend suggests that, unlike the behavior of $P_z$, $|P_z|$ grows systematically as the energy position on the SS moves away from the Dirac point at the zone center.

To highlight the OP spin configuration obtained from the present experiment, we illustrate in Fig. 2(c) the OP spin configuration obtained from the present ARPES experiment. The length of the arrow corresponds to the magnitude of $P_z$. The data are symmetrized by taking into account the crystal symmetry. The intensity with gradual shading (red and blue) shows the calculated $P_z$ value based on the theory in Ref. [6].

We also found that the $z$-axis spin polarization $P_z$ shows a sign reversal along the same cut [Fig. 2(b)], as expected from the result in Fig. 1 where the $\Gamma K'$ direction at $\phi = 60^\circ$ is identical to the $\Gamma K'$ direction at $\phi = 180^\circ$ due to the three-fold rotational symmetry of the crystal. However, the magnitude of $|P_z|$ stays nearly zero at $|k_y| \leq 0.03 \text{ Å}^{-1}$ and then gradually increases with $|k_y|$, reaching $\sim 0.15$ near the $k_F$ point. This trend suggests that, unlike the behavior of $P_z$, $|P_z|$ grows systematically as the energy position on the SS moves away from the Dirac point at the zone center.

To analyze the $k$ dependence of the spin polarization in more detail, we plot in Figs. 2(a) and (b) the spin polarizations along the $x$- and $z$-axes measured in the $k_z=0$ cut [vertical solid (green) line in Fig. 1(b)] obtained by integrating the ARPES intensity at $E_B = 0.08-0.12$ eV; because of the rather large (40 meV) energy resolution of the spin-resolved measurements, the integration in this energy window gives a good representation of the spin polarization of the surface band as a function of $k$. One can see in Fig. 2(a) that the $P_x$ value at $k_y > 0$ ($k_y < 0$) is positive (negative), and its absolute value $|P_x|$ is nearly constant at $\sim 0.6$ for $|k_y| \geq 0.03 \text{ Å}^{-1}$. The sudden sign reversal in $P_z$ near $k_y = 0$ is consistent with the IP spin helical texture. We also found that the $z$-axis spin polarization $P_z$ shows a sign reversal along the same cut [Fig. 2(b)], as expected from the result in Fig. 1 where the $\Gamma K'$ direction at $\phi = 60^\circ$ is identical to the $\Gamma K'$ direction at $\phi = 180^\circ$ due to the three-fold rotational symmetry of the crystal. However, the magnitude of $|P_z|$ stays nearly zero at $|k_y| \leq 0.03 \text{ Å}^{-1}$ and then gradually increases with $|k_y|$, reaching $\sim 0.15$ near the $k_F$ point. This trend suggests that, unlike the behavior of $P_z$, $|P_z|$ grows systematically as the energy position on the SS moves away from the Dirac point at the zone center.

To highlight the OP spin configuration obtained from the present experiment, we illustrate in Fig. 2(c) the OP spin configuration obtained from the present ARPES experiment. The length of the arrow corresponds to the magnitude of $P_z$. The data are symmetrized by taking into account the crystal symmetry. The intensity with gradual shading (red and blue) shows the calculated $P_z$ value based on the theory in Ref. [6].

FIG. 2: (color online) (a),(b) Experimental spin polarization along the $x$ and $z$ axes, $P_x$ and $P_z$, of the SS along the $k_z=0$ cut, obtained by integrating the SR-ARPES intensity at $E_B = 0.08-0.12$ eV. (c) Schematic picture of the $z$ component of the spin vector as a function of $k$ in the SS of Bi$_2$Te$_3$ determined from the present ARPES experiment. The length of the arrow corresponds to the magnitude of $P_z$. The data are symmetrized by taking into account the crystal symmetry. The intensity with gradual shading (red and blue) shows the calculated $P_z$ value based on the theory in Ref. [6].

FIG. 3: (color online) (a),(b) Comparison of the FS between TlBiSe$_2$ and Bi$_2$Te$_3$. Solid lines are the simulated FS using the theory in Ref. [6], while the dashed lines are the experimental FS. (c),(d) SR-EDCs measured along the $\Gamma K$ line ($\phi = 0^\circ$) for TlBiSe$_2$ and Bi$_2$Te$_3$, respectively. (e),(f) Comparison of the $P_z$ dependence of $E_F$ for TlBiSe$_2$ and Bi$_2$Te$_3$. (g) Calculated $P_z$ value along the $k_z=0$ cut as a function of $k_y$ for TlBiSe$_2$ and Bi$_2$Te$_3$.
by measuring data at various k points. These results point to an intimate connection between the hexagonal warping of the FS and the magnitude of $P_z$ [18], although it is difficult to rule out all the other possibilities at the moment. To further examine the present observation, we reproduced the observed FS shape of TIBiSe$_2$ and Bi$_2$Te$_3$ by using the theoretical band dispersion with the hexagonal warping term [3] [see the solid curves in Figs. 3(a) and (b)] and calculated the $P_z$ value along the $k_x=0$ cut [dashed lines in Fig. 3(a)] as shown in Fig. 3(g). Apparently, the calculated $P_z$ value in TIBiSe$_2$ is much smaller than that in Bi$_2$Te$_3$ (≈0.2 and ≈0.6 at $k_F$, respectively), which is qualitatively consistent with what we observed.

However, the magnitude of $P_z$ is quantitatively different between theory and experiment, as seen in Fig. 3: For instance, at the $k_F$ point along the $\Gamma K$ direction ($\phi = 0^\circ$) where $|P_z|$ becomes maximal on the FS, the theoretical $P_z$ of ≈0.6 [Fig. 3(g)] is ≈4 times larger than the experimental $P_z$ of ≈0.15 [Fig. 3(f)]. The smaller $P_z$ in the experiment cannot be attributed to extrinsic effects in the experiment such as the instrumental asymmetry of up- and down-spin channel or a nonlinearity in the detector sensitivity to the actual polarization, because the experimental value of $P_z$ is systematically reversed across $k_y=0$ with a similar magnitude relative to $k_x=0$ [see Fig. 2(b)] and the experimental scattering asymmetry was confirmed to be proportional to the theoretical spin polarization by measuring Au(111) as a reference. Therefore, there must be an intrinsic reason for the weakening of $P_z$. One possible explanation is the many-body effects that lead to an electronic order like the spin density wave where the FS is nested with the wave vector connecting parallel segments of the hexagonally warped FS [6, 13]. Another possibility is the presence of an interband scattering channel [13] that degrades the absolute surface spin polarization through the scattering between the surface band and the non-spin-polarized bulk band. It is desirable that a concrete theoretical model is developed to explain the unexpectedly small OP polarization.

The presence of the OP spin polarization provides a novel mechanism for the “mass acquisition” of the Dirac particles on the surface [6], which is a prerequisite of exotic topological phenomena like the half-integer quantum Hall effect and the quantized magnetoelectric effect [20, 21]. When the SS has a purely IP helical spin texture, the gapping of the Dirac cone requires an OP magnetic field $B_z$ or magnetization $M_z$. On the other hand, when the SS has a finite OP spin component $S_z$, application of an IP magnetic field $B_z$ would also lead to a gap opening due to the coupling between $B_z$ and $S_z$ [6]. This way of gapping would be useful for achieving a massive Dirac state since it can essentially avoid the complicated orbital effects which generally couple to the IP k’s and spins [7]. Hence our observation of the OP spin component assures a promising direction for realizing novel quantum state and device applications that require a gapped SS.

In conclusion, our SR-ARPES measurements of Bi$_2$Te$_3$ provide direct and compelling evidence for the OP spin component on the gapless SS, by simultaneously determining the IP and OP spin textures at various k points in the Brillouin zone. Moreover, we found that a stronger hexagonal deformation of the FS leads to a larger OP spin component, by comparing the observed FS and the SR-ARPES data with those of TIBiSe$_2$. Intriguingly, the OP polarization is found to be much smaller than that expected from existing theory in both Bi$_2$Te$_3$ and TIBiSe$_2$, which poses an interesting question for future studies.

We thank T. Arakane for the development of an in-situ sample rotation system for the SR-ARPES measurements and Zhi Ren for the preliminary crystal growth of Bi$_2$Te$_3$. This work was supported by JSPS (KAKENHI 19674002 and the Next-Generation World-Leading Researchers Program), JST-CREST, MEXT of Japan (Innovative Area “Topological Quantum Phenomena”), and AFOSR (AOARD 10-4103).

Note added. Recently, we became aware of a related SR-ARPES study on more highly electron-doped Bi$_2$Te$_3$ [22]. The reported $P_z$ value (0.3) is larger than that of the present study (0.15). This difference is consistent with the k-p theory [6] and is attributed to the difference in the magnitude of the warping factor $w$ between the two cases (1.1 in the present sample, as opposed to 1.6 in Ref. [22]).

---

[1] M. Z. Hasan and C. L. Kane, Rev. Mod. Phys. 82, 3045 (2010).
[2] Y. Xia et al., Nature Phys. 5, 398 (2009).
[3] P. Roushan et al., Nature (London) 460, 1106 (2009).
[4] J. Seo et al., Nature (London) 466, 343 (2010).
[5] Z. Alpichshev et al., Phys. Rev. Lett. 104, 016401 (2010).
[6] L. Fu, Phys. Rev. Lett. 103, 266801 (2009).
[7] H.-J. Zhang et al., Phys. Rev. B 80, 085307 (2009).
[8] O. V. Yazyev, J. E. Moore, and S. G. Louie, Phys. Rev. Lett. 105, 266806 (2010).
[9] Y. L. Chen et al., Science 325, 178 (2009).
[10] S. Souma et al., Rev. Sci. Instrum. 81, 095101 (2010).
[11] S. Souma et al., Rev. Sci. Instrum. 78, 123104 (2007).
[12] D. Hsieh et al., Nature (London) 460, 1101 (2009).
[13] S. R. Park et al., Phys. Rev. B 81, 041405R (2010).
[14] M. Bianchi et al., Nat. Commun. 1 : 128 (2010).
[15] T. Sato et al., Phys. Rev. Lett. 105, 136802 (2010).
[16] K. Kuroda et al., Phys. Rev. Lett. 105, 146801 (2010).
[17] Y. L. Chen et al., Phys. Rev. Lett. 105, 266401 (2010).
[18] The close connection between the hexagonal warping and the magnitude of $P_z$ may also be recognized by looking at the data of Bi$_2$Te$_3$ in Fig. 2(b) where the $P_z$ value is reduced near the $\Gamma$ point as the hexagonal warping becomes weaker near the Dirac point [5, 10].
[19] K. Kuroda et al., Phys. Rev. Lett. 105, 076802 (2010)
[20] X. L. Qi, T. L. Hughes, and S. C. Zhang, Phys. Rev. B 78, 195424 (2008).
[21] X.-L. Qi et al., Science 323, 1184 (2009).
[22] S.-Y. Xu et al., arXiv:1101.3985