Spin-polarized surface bands of a three-dimensional topological insulator studied by high-resolution spin- and angle-resolved photoemission spectroscopy

Akinori Nishide¹, Yasuo Takeichi¹, Taichi Okuda², Alexey A Taskin³, Toru Hirahara⁴, Kan Nakatsuji¹, Fumio Komori¹, Akito Kakizaki¹, Yoichi Ando³,⁴ and Iwao Matsuda¹,⁵

¹ Institute for Solid State Physics, the University of Tokyo, Japan
² Hiroshima Synchrotron Radiation Center, Hiroshima University, Higashi-Hiroshima 739-0046, Japan
³ Institute of Scientific and Industrial Research, Osaka University, Ibaraki, Osaka 567-0047, Japan
⁴ Department of Physics, School of Science, the University of Tokyo, Japan
E-mail: y_ando@sanken.osaka-u.ac.jp and imatsuda@issp.u-tokyo.ac.jp

New Journal of Physics 12 (2010) 065011 (14pp)
Received 2 October 2009
Published 17 June 2010
Online at http://www.njp.org/
doi:10.1088/1367-2630/12/6/065011

Abstract. The spin-polarized surface band structure of the three-dimensional (3D) quantum spin Hall phase of Bi₁₋ₓSbₓ (x = 0.12–0.13) was studied by spin- and angle-resolved photoemission spectroscopy (SARPES) using a high-yield spin polarimeter equipped with a high-resolution electron spectrometer. The spin-integrated spectra were also measured and compared to those of Bi₁₋ₓSbₓ with x = 0.04. Band dispersions of the edge states were fully elucidated between the two time-reversal-invariant points, Γ and M, of the (111) surface Brillouin zone. The observed spin-polarized band dispersions at x = 0.12–0.13 indicate an odd number of the band crossing at the Fermi energy, giving unambiguous evidence that this system is a 3D strong topological insulator, and determine the ‘mirror chirality’ to be −1, which excludes the existence of a Dirac point in the middle of the Γ–M line. The present research demonstrates that the SARPES

⁵ Author to whom any correspondence should be addressed.
measurement with energy resolution $\leq 50\,\text{meV}$ is one of the critical techniques for complementing the topological band theory for spins and spin currents.

Content

1. Introduction 2
2. Experimental details 4
3. Results and discussion 6
4. Concluding remarks 13
Acknowledgments 13
References 13

1. Introduction

Spin–orbit interaction induces various spin properties in solids, and has attracted academic and technological interest. Recently, the spin-Hall effect, which makes it possible to produce spin currents without a magnet, has attracted a great deal of attention for its potential impact on spintronics [1, 2]. The effect has also stimulated physicists to extend the theoretical frameworks to the ‘quantum spin Hall’ (QSH) effect [3–14], which is realized in a topologically nontrivial electronic state, as in the case of the quantum Hall effect. Unlike the spin itself, the spin current is time-reversal invariant. Hence these modes carry spin currents without breaking time-reversal symmetry. Furthermore, the edge modes are robust against disorder or modest changes of boundary conditions, such as surface roughness or impurities.

The theoretical framework of the QSH effect is based on the $Z_2$ invariants of band structures and this topological property has enabled one to seek materials of the QSH phase. In three dimensions there are four $Z_2$ invariants ($\nu_0; \nu_1\nu_2\nu_3$), representing time-reversal-invariant band structures [9, 10]. When $\nu_0 = 1$, the system is a three-dimensional (3D) strong topological insulator, where the ‘edge states’ (i.e. 2D surface states) form spin currents flowing on a surface and are robust against disorder [11, 13, 14]. The spin lies within the surface plane and is perpendicular to the momentum (wavevector) of the electron. The existence of such spin current is in sharp contrast to the trivial insulator, where $(\nu_0; \nu_1\nu_2\nu_3) = (0; 000)$. Recently, the semiconductor alloy Bi$_{1-x}$Sb$_x$ ($x \sim 0.1$) was predicted [13] to be a strong topological insulator.

Figure 1 presents the band structures of the 2D surface states for topologically trivial and such nontrivial cases. Figure 1(a) shows the partially and completely occupied surface bands of insulating (semiconducting) bulk crystals, which is the trivial case. The surface states are formed within the bulk band gap and, for most of the clean and metal-covered semiconductor surfaces, for example, they are spin degenerate [15, 16]. Figure 1(b) depicts the surface-state bands of materials with a large spin–orbit interaction. Usually in the bulk, the time-reversal symmetry ($E(\mathbf{k}, \uparrow) = E(-\mathbf{k}, \downarrow)$) and the space-inversion symmetry ($E(\mathbf{k}, \uparrow) = E(-\mathbf{k}, \uparrow)$) lead to the Kramers degeneracy. However, at the crystal surface, the degeneracy will be lifted due to the loss of the space-inversion symmetry and the 2D electronic bands are spin split (the Rashba effect) [17]. Recently, such spin-split bands have been measured on the crystal surfaces of heavy metals [18, 19], such as Bi and Au, and figure 1(b) illustrates the surface band structure of such a semimetal crystal. The two bands with opposite spin orientations degenerate at the
Figure 1. Schematic representations of surface-state (edge-state) band structures between two time-reversal-invariant momenta, $\Lambda_a$ and $\Lambda_b$. (a) Typical surface band structure with spin-degenerate surface-state bands formed in the bulk band gap of the trivial bulk insulator. (b,d) Trivial cases of the spin-split surface-state bands for the bulk semimetal. (c,e) Nontrivial cases of the spin-split surface-state bands of the bulk topological insulator. The $Z_2$ invariants are (a,b,d) zero and (c,e) nonzero. The projected bulk band structures are drawn as shaded regions.

In the case of the topologically nontrivial system [13] (such as Bi$_{1-x}$Sb$_x$ [20]) (figure 1(c)), the electronic structure is completely different from those of the trivial cases (figures 1(a) and (b)). For the topological insulator, the edge-state bands (the surface-state band), connecting bulk valence and conduction bands, change the spin orientations at $\Lambda_a$ and $\Lambda_b$. Such distinctions in band structures lead to differences in a number of spin-resolved bands crossing the Fermi level, $E_F$: even for the trivial case and odd for the nontrivial case [13]. Figures 1(d) and (e) show further examples that the spin-split surface-state bands, being filled at $\Lambda_a$ and $\Lambda_b$, may cross each other above $E_F$. The presence of such a crossing point (Dirac point) accordingly changes the spin characters of filled bands, and it is classified as ‘mirror chirality’ that has been argued on the theoretical front of topological band theories [21]. Examples of figure 1 demonstrate that, depending on the topological parameters, there are varieties of spin-dependent surface electronic structure. In order to clarify the topological band theory of these systems, it is essential to determine the band dispersion and the spin characters of all the surface states (edge states) in the surface Brillouin zone.
Recently, photoemission studies have mapped the band dispersion curves of the (111) surface states of the semiconductor alloy Bi$_{1-x}$Sb$_x$ ($x \sim 0.1$) and have measured the spin orientations at one of the time-reversal-invariant momenta, $\bar{\Gamma}$, to find a trace of the predicted topological band structure [22, 23]. Despite the necessity to determine spin-resolved band dispersion in the whole-surface Brillouin zone, spin characters of the adjacent time-reversal-invariant momenta, $\bar{M}$, have not been determined yet [23]. Therefore, it is required to conduct direct spin- and angle-resolved photoemission spectroscopy (SARPES) measurements of all the spin-polarized surface-state bands on Bi$_{1-x}$Sb$_x$ crystals.

In the present work, we have developed a new spin-polarized photoemission spectrometer based on spin-dependent very low-energy electron diffraction (VLEED). Taking advantage of the high spin-polarimetry efficiency and the high-energy resolution [24], we have determined all the spin-polarized bands of Bi$_{1-x}$Sb$_x$ ($x = 0.12–0.13$) along the $\bar{\Gamma}$–$\bar{M}$ line. Our results provide direct evidence for the QSH phase in the 3D topological insulator and clarify the new topological parameter, mirror chirality [21], settling the essential topological structure of the surface states of this material.

2. Experimental details

Bi$_{1-x}$Sb$_x$ ($x = 0.04, 0.12, 0.13$) crystals were grown from a stoichiometric mixture of 99.9999\% purity Bi and Sb elements by a zone melting method. Electrical resistivity measurement of a Bi$_{1-x}$Sb$_x$ ($x = 0.13$) sample grown in the same batch was performed before the spin-resolved photoemission experiments. The measured resistivity below 100 K increased monotonically with decreasing temperature, signifying the presence of a bulk band gap. The bulk of the present Bi$_{1-x}$Sb$_x$ ($x = 0.13$) sample was insulating, consistent with the published data in the literature [22, 25].

The crystals were cleaved along the (111) plane at 123 K below $3 \times 10^{-8}$ Pa. The SARPES measurements were performed at BL-19A (KEK-PF, Japan) with a high-resolution hemispherical analyzer (SPECS Phoibos-150) equipped with a homemade high-yield VLEED spin polarimeter [24]. The measurement setup is schematically drawn in figure 2 and the details are described below. Irradiating VUV photons on the sample, photoelectrons are emitted from the sample. The electrons are analyzed for their energy in the electron spectrometer and recorded at the detector, resulting in photoemission energy spectra. When such spectra were recorded at various emission angles, a series of angle-resolved photoemission spectra (ARPES) can directly be converted to an in-plane band diagram through the conservation rules of in-plane momentum and energy [26, 27]. Thus, ARPES techniques have been the most powerful experimental tools to determine band dispersions of surface-state and bulk-state bands. When the electron detector is replaced by the spin polarimeter, the electrons, being resolved in energy and momentum (angle), are further resolved to their spin orientations.

There have been developments of various types of electron spin polarimeters to measure the spin polarization of photoelectrons [24, 28–35]. Among these, the Mott detector has been the most popular for electron spin detection [28–32]. The detector utilizes Mott scattering, which is the spin–orbit interaction of high-energy electrons (25–100 keV) and nuclei of the target. In the spin polarimeter, one can determine the spin polarization $P$ of the electron by measuring the scattering asymmetry ($A$) of spin-polarized electrons at the nuclei of heavy atoms. The polarization ($P$) and the observed asymmetry ($A$) are related as $P = A/S_{\text{eff}}$. Here, $S_{\text{eff}}$ is the effective Sherman function, representing the spin-resolving power of the spin detector. For
Figure 2. Schematic drawing of the high-resolution SARPES measurement. Spin-polarized electrons are depicted as up (red) and down (blue) arrows. Spin currents are also depicted for the edge states in the 3D QSH phase, from which the spin-polarized photoelectrons are emitted into a high-resolution electron spectrometer and the high-yield VLEED spin polarimeter.

Table 1. A list of specifications of various spin polarimeters. $|S_{\text{eff}}|$ and $I/I_0$ are the effective Sherman function and the scattering probability, respectively. $\epsilon$ is the figure of merit or the efficiency of spin detection.

| Interaction            | Energy   | $|S_{\text{eff}}|$ | $I/I_0$  | $\epsilon$   |
|------------------------|----------|-------------------|----------|---------------|
| Conventional Mott [28] | Spin–orbit         | 100 keV       | 0.20     | $2.9 \times 10^{-3}$ | $1.1 \times 10^{-4}$ |
| Compact Mott [29]–[31] | 25–40 keV | 0.14–0.15         | 9.7 $\times 10^{-3}$ | 1.9–5.6 $\times 10^{-4}$ |
| SPLEED [33]            | 150 eV   | 0.19               | $2.2 \times 10^{-3}$ | $8.0 \times 10^{-5}$ |
| Diffuse scattering [34]| 150 eV   | 0.19               | $9 \times 10^{-3}$ | $1 \times 10^{-4}$ |
| VLEED (Fe surface) [35]| Spin-exchange     | 12 eV         | 0.4      | 0.1           | $2 \times 10^{-2}$ |
| VLEED (O/Fe surface) [24]| 6 eV      | 0.3–0.4           | 0.12     | $1.9 \times 10^{-2}$ |

The Mott detector, $S_{\text{eff}}$ is 0.1–0.2. At the typical condition for the Mott detector [28–31], the scattering probability $I/I_0$ is $\sim 10^{-2}$ and the efficiency of the spin detection, defined by the figure of merit (FOM) ($\epsilon = S^2 I/I_0$), is order of $10^{-4}$. Here, $I$ and $I_0$ are the numbers of reflected and incident electrons, respectively (table 1).

In the present SARPES measurements, we have developed a high-yield spin polarimeter [24]. In the new detector, the spin-exchange interaction of an electron is applied to measure the spin polarization. That is, the spin-dependent electron reflectivity of a very low-energy electron (below 10 eV) at the surface target of a magnetized ferromagnetic crystal is utilized. Figure 2 shows a schematic drawing of our SARPES measurement system. We have used an Fe(100) crystal surface passivated by an atomic O/Fe layer. Surface contamination is
fatal for the surface scattering of a very low-energy electron, and the oxidized atomic layer prevents the surface from degradation during the measurements. In the present system, the target survives more than two weeks without any treatment. In our performance test, \( S_{\text{eff}} \) reaches 0.4 \( \pm \) 0.02 and FOM was determined as \( \epsilon = 1.9 \pm 0.2 \times 10^{-2} \). Table 1 lists the specifications of various spin polarimeters, and one can see that the \( \epsilon \) of our detector is much higher than that of a conventional Mott detector. Because of this very high spin-efficiency of the VLEED detector, we have combined it with the high-resolution electron spectrometer as shown in figure 2. In the performance test, we have made SARPES measurements with energy and angle resolutions of 30 meV and \( \pm 0.7^\circ \), respectively. Further details of this new spin-polarized photoemission spectrometer with very high efficiency and energy resolution can be found elsewhere [24].

In the present research, SARPES-spectra were recorded with energy and angle resolutions of 50 meV and \( \pm 1^\circ \), respectively. In the VLEED detector, the spin polarization \( P \) is acquired from intensities of the reflected electrons interacting with the majority-spin (minority-spin) states of the Fe(001)p(1 \times 1)-O target, \( I_{\text{maj}} \) (\( I_{\text{min}} \), by \( P = (1/S_{\text{eff}}) \times [(I_{\text{maj}} - I_{\text{min}})/(I_{\text{maj}} + I_{\text{min}})] \). Here, the effective Sherman function was \( S_{\text{eff}} = 0.32 \pm 0.04 \) and it was determined by the preceding measurement of the polarization of secondary electrons from Fe(001). From \( I_{\text{maj}} \) and \( I_{\text{min}} \), the spin-up and spin-down spectra are obtained by \( I_{\uparrow,\downarrow} = (I_{\text{maj}} + I_{\text{min}})(1 \pm P)/2 \). For comparison, spin-integrated ARPES spectra were measured with a hemispherical analyzer (VG Sciena SES-100) at energy and angle resolutions of 30 meV and \( \pm 0.2^\circ \). All the ARPES measurements were taken at 120–140 K with He I\( ^\alpha \) sources along the \( \Gamma^M \) line in the (111) surface Brillouin zone shown in figure 3(a). In the spin-resolved photoemission experiments, the incident angle of the photon was 45° from the surface normal and the direction was out of a plane defined by the surface normal and the photoelectron detecting directions, as shown in figure 2. On the other hand, for the spin-integrated photoemission experiments, the angle between the directions of incident photons and detecting photoelectrons was fixed at 45°.

### 3. Results and discussion

The evolution of the band structure of the alloy Bi\(_{1-x}\)Sb\(_x\) as a function of \( x \) has been well studied and is summarized in figure 3(a) [13, 20]. At first, as the Sb concentration is increased, the gap between the two bands at the \( L \) point with different parity, \( L_s \) and \( L_a \) bands, decreases. At \( x = 0.04 \), the bands cross and the gap reopens with inverted ordering. Then, the top of the valence band at \( T \) comes down in energy and crosses the bottom of the conduction band at \( x = 0.07 \). At this point, the indirect gap becomes positive, and the alloy is a semiconductor. At \( x = 0.09 \), the \( T \) valence band clears the \( L_s \) valence band, and the alloy is a direct-gap semiconductor at the \( L \) points. As \( x \) is increased further, the gap increases until its maximum value at \( x = 0.18 \). At that point, the valence band at \( H \) crosses the \( L_s \) valence band. For \( x = 0.22 \), the \( H \) band crosses the \( L_a \) conduction band, and the alloy is again a semimetal. The band diagram in figure 3 indicates that Bi\(_{1-x}\)Sb\(_x\) becomes a topological insulator or topologically nontrivial at \( x = 0.07–0.22 \).

In order to trace the evolution of the band feature, we have conducted photoemission band mapping of the Bi\(_{1-x}\)Sb\(_x\) crystal at trivial (\( x = 0.04 \)) and nontrivial (\( x = 0.13 \)) stages. Figures 3(b) and (c) show the spin-integrated gray-scale band diagrams of the \( x = 0.04 \) and \( x = 0.13 \) samples measured along the \( \Gamma^M \) line, respectively. Concerning the band structure of Bi\(_x\)Sb\(_{1-x}\) (\( x = 0.04 \)) shown in figure 3(a), a metallic band, sharply dispersing from the \( \Gamma \) point, can be found. The band denoted as \( \Sigma_2 \) has a Fermi wavenumber, \( k_F \), of \( \sim 0.07 \) Å\(^{-1} \). Labeling

New Journal of Physics 12 (2010) 065011 (http://www.njp.org/)
Figure 3. (a) Schematic representation [13] of the band energy evolution of Bi$_{1-x}$Sb$_x$ as a function of $x$ with the 3D Brillouin zone and its projection onto the (111) surface. High symmetry points in the (surface) Brillouin zones are indicated. The direction of the wavenumber $k_{||}$, which indicates the angle-scanning direction in the present photoemission experiment, is shown as a thick arrow. (b,c) Spin-integrated photoemission band diagram of Bi$_{1-x}$Sb$_x$ crystals with (b) $x = 0.04$ and (c) $x = 0.13$. The brightness in the gray scale corresponds to intensity of the ARPES spectra.

of the surface states in figures 3(a) and (b) was adopted from [21]. Departing from the $\Gamma$ point, two Fermi level crossings can be found at $k_F \sim 0.2$ and $0.3 \, \text{Å}^{-1}$. As shown below, the band feature corresponds to the hole pocket of an electronic state, denoted as $\Sigma_1$. Near the $M$ point, another metallic band can be found. From a comparison with the band structure of Bi [18], the band is assigned to $\Sigma_2$. At the time-reversal-invariant momenta of $\bar{\Gamma}$ and $\bar{M}$, the surface-state bands overlap with the bulk band projection region, as shown in figure 1. Due to matching of
the wave functions of surface-state bands and bulk bands, the photoemission intensity decreases significantly at these regions and it is difficult to trace the dispersions of the surface-state bands to these points.

For the band structure of Bi$_{1-x}$Sb$_x$ ($x = 0.13$), shown in figure 3(c), the spectral features of the $\Sigma_1$ and $\Sigma_2$ bands are identified. The metallic $\Sigma_2$ band has a $k_F$ of $\sim 0.06$ Å$^{-1}$ from the $\bar{\Gamma}$ point. The $\Sigma_1$ band disperses towards $E_F$ and returns to higher $E_B$ without showing a clear $E_F$ crossing. Namely, within the present measurement resolutions, the top of the $\Sigma_1$ band likely lies below $E_F$. Around $M$, a metallic band feature can also be found as in the case of $x = 0.04$. However, the photoemission intensity distribution along the binding energy axis is broader than that of the $x = 0.04$ sample. The feature is clearer when one compares the ARPES spectra as shown in figure 4(a). At two $k_{||}$ points, the widths of the peaks near the Fermi level of the $x = 0.13$ sample are obviously broader than that of the $x = 0.04$ sample. This indicates that there is more than one band around the $\bar{M}$ point for the $x = 0.13$ sample. To extract information on the photoemission peaks, the spectra are fitted by a Gaussian with a background of the Fermi–Dirac function, as shown in figure 4(b). In the present experimental condition, the peak width is determined by the actual instrumental resolution (30 meV). It is clear that there is only one band for the trivial sample ($x = 0.04$) and two bands for the nontrivial sample ($x = 0.13$) near $E_F$ around the $\bar{M}$ point.
Figure 5. SARPES spectra, $I_\uparrow$ (colored in red) and $I_\downarrow$ (colored in blue), of a Bi$_{1-x}$Sb$_x$ ($x = 0.12$) crystal at various emission angles $\theta_e$ and corresponding $k_{||}$ values. Peak positions of the surface states, $\Sigma_1$ and $\Sigma_2$, are indicated by triangles.

Figure 4(b) also shows the up-spin and down-spin spectra of Bi$_{1-x}$Sb$_x$ ($x = 0.12$), and one can see that the two fitted peak positions match individually those of the $I_\uparrow$ and $I_\downarrow$ spectra. This demonstrates that, although the two bands are hardly distinguishable in the spin-integrated spectra, they can be resolved in SARPES spectra. Figure 5 shows typical SARPES spectra of Bi$_{1-x}$Sb$_x$ ($x = 0.12$) at various wavenumbers $k_{||}$, converted from the emission angle $\theta_e$, where spin-dependent spectral features can be distinctively seen in the $I_\uparrow$ (red) and $I_\downarrow$ (blue) plots. Photoemission peaks assigned to the surface states are located at binding energies $E_B$ of less than 0.2 eV and are indicated by triangles. Figures 6(a)–(c) exhibit the SARPES spectra of (a) $I_\uparrow$, (b) $I_\downarrow$ and (c) $P$ near the $\bar{\Gamma}$ point. As shown in figure 3, the $\Sigma_2$ band, dispersing from the $\bar{\Gamma}$ point, crosses $E_F$ at around $k_{||} = 0.1 \text{ Å}^{-1}$. The photoemission intensity of the $\Sigma_2$ band is observed with positive wavenumbers in the $I_\downarrow$ spectra in figure 6(b) and, therefore, $\Sigma_2$ has spin-down polarization. On the other hand, the $\Sigma_1$ band, which shows up in the $I_\uparrow$ spectra (figure 6(a)) and thus has spin-up polarization, approaches $E_F$ from $\bar{\Gamma}$. The $\bar{\Gamma}$ point is the time-reversal-invariant momentum, described in figure 1, and $P$ becomes zero due to the spin degeneracy. Near the $\bar{\Gamma}$ point, $P$ becomes antisymmetric with respect to $k_{||} = 0$. With an increase of $k_{||}$, positive $P$ originating from the $\Sigma_1$ band is clearly identified and negative $P$ of the $\Sigma_2$ band is also found at $E_F$ around $k_{||} = 0.1 \text{ Å}^{-1}$.

Figures 7(a)–(c) exhibit the SARPES spectra of (a) $I_\uparrow$, (b) $I_\downarrow$ and (c) $P$ near the $\bar{M}$ point. The $\Sigma_1$ band, found in the $I_\downarrow$ spectra, disperses back as $k_{||}$ increases towards $\bar{M}$, as found in figure 3(c). In the $I_\uparrow$ data in figure 7(b), a band appears at $E_F$ around 0.5 Å$^{-1}$ and it disperses slightly to higher $E_B$. Since this band has spin-down polarization, it is likely assigned to $\Sigma_2$. Near the $\bar{M}$ point, the $\Sigma_1$ band crosses $E_F$ in the $I_\uparrow$ spectra in figure 7(a). These two metallic
Figure 6. SARPES spectra of (a) $I_{\uparrow}$ (colored in red), (b) $I_{\downarrow}$ (colored in blue) and (c) $P$ for Bi$_x$Sb$_{1-x}$ ($x = 0.12$) near the $\Gamma$ point. Peak positions of the surface states, $\Sigma_1$ and $\Sigma_2$, are indicated by triangles. The positive and negative $P$ regions are painted in red and blue, respectively.

Figure 7. SARPES spectra of (a) $I_{\uparrow}$ (colored in red), (b) $I_{\downarrow}$ (colored in blue) and (c) $P$ for Bi$_x$Sb$_{1-x}$ ($x = 0.12$) near the $M$ point. Peak positions of the surface states, $\Sigma_1$, $\Sigma_2$ and $\Sigma_1'$, are indicated by triangles. The positive and negative $P$ regions are painted in red and blue, respectively.
bands with opposite spin polarizations, $\Sigma_2$ of down-spin and $\Sigma'_1$ of up-spin, converge at the $\tilde{M}$ point. In the $P$ spectra of figure 7(c), positive $P$ of the $\Sigma_1$ band is found as figure 6(c). Between $k_{||} = 0.5$ and $0.6$ Å$^{-1}$, negative $P$ of the $\Sigma_2$ band appears at $E_F$ and $P$ becomes zero as it approaches the $\tilde{M}$ point. The reduction of the negative $P$ is due to overlap of the positive $P$ of the $\Sigma'_1$ band and the two bands finally make spin degeneracy at the time-reversal-invariant momentum of the $\tilde{M}$ point (figure 1).

Figure 8 shows a dispersion plot of the spin-polarized surface-state bands obtained from the SARPES spectra in figures 6 and 7. For comparison, the data are displayed on a grayscale band diagram constructed from the spin-integrated ARPES spectra shown in figure 3(c). While the band structure is essentially similar to the one reported previously [22, 23], it is now found to be basically composed of three surface-state bands, $\Sigma_1$, $\Sigma_2$ and $\Sigma'_1$. The $\Sigma_2$ band, creating an electron pocket at the $\bar{\Gamma}$ point with the Fermi wavenumber $k_F$ of $\sim 0.1$ Å$^{-1}$, has asymmetric spin polarization with respect to $\bar{\Gamma}$. The $\Sigma_1$ band disperses towards $E_F$ and returns to higher $E_B$ without showing a clear $E_F$ crossing. Namely, within the present measurement resolutions, the top of the $\Sigma_1$ band likely lies below $E_F$; this is slightly different from the previous result [22, 23], in which the $\Sigma_1$ band obviously crossed $E_F$ and formed a small hole pocket. The slight shift of the $E_F$ position is likely due to the difference in Sb concentration ($x = 0.09–0.10$ in [22, 23], while $x = 0.12–0.13$ in the present work), which could lead to a chemical potential shift. The $\Sigma_2$ band appears again at $E_F$ near the midpoint of the $\bar{\Gamma}$–$\tilde{M}$ line and forms an electron pocket around $\tilde{M}$. The dispersion of the $\Sigma'_1$ band is very close to that of the $\Sigma_2$ band and the two bands are hardly distinguishable in the spin-integrated spectra, as shown in the

**Figure 8.** Spin-polarized band dispersions of the surface states of Bi$_{1-x}$Sb$_x$ ($x = 0.12–0.13$). The spin-resolved data of the $x = 0.12$ sample, from figures 6 and 7, are plotted with open circles and overlaid on a spin-integrated grayscale diagram. Spin-up (spin-down) bands are colored in red (blue) and their dispersion curves are schematically traced by thick lines. For comparison, the data are displayed on a grayscale band diagram ($x = 0.13$) of the spin-integrated ARPES spectra shown in figure 3(c). The bulk band projection, tentatively estimated from the ARPES data, is shown as the shaded purple area.
gray-scale diagram in figure 3(c). However, these two bands have opposite spin polarizations and, therefore, their dispersion can be unambiguously resolved in the spin-revolved spectra and plotted as the two spin-polarized bands in figure 8. While the previous photoemission work [22, 23] reported a degeneracy of the \( \Sigma_2 \) and \( \Sigma_1' \) bands at \( E_F \), they actually intersect with \( E_F \) at different wavenumbers, \( k_F \sim 0.5 \text{Å}^{-1} \) for \( \Sigma_2 \) and \( k_F \sim 0.6 \text{Å}^{-1} \) for \( \Sigma_1' \) (figure 8). The \( \Sigma_1' \) band also creates an electron pocket around \( \bar{M} \) and it converges with \( \Sigma_2 \) at \( \bar{M} \), which is natural because these two bands have opposite spin polarities and the spin polarization \( P \) must disappear at the time-reversal-invariant point \( \bar{M} \), as shown in figure 7(c). As described above, these spin properties demonstrated in figures 6–8 are typical for a compound with a large spin–orbit coupling. The Kramers theorem dictates that the spin-split states become degenerate at the time-reversal-invariant momenta, \( \bar{\Gamma} \) and \( \bar{M} \), and the spin structure naturally becomes antisymmetric with respect to these points.

Based on \( Z_2 \) topology, the band structures of bulk insulators are classified into trivial and nontrivial cases by examining the parity of the valence bands. Furthermore, when the system is nontrivial, it has been shown that counting the number of surface-band crossings at \( E_F \) gives a convenient way of distinguishing a strong topological insulator [13]: an odd number as described in figures 1(c) and (e). If bands are spin degenerate, as in the case of figure 1(a), the number always becomes even. In the case of figures 1(b) and (d), typical Rashba systems, the number of \( E_F \) crossings of surface states is also even. Thus, any one of figures 1(a), (b) and (d) cannot be a strong topological insulator. As shown in figure 3(b), the number of \( E_F \) crossings of \( \text{Bi}_{1-x}\text{Sb}_x \) \((x = 0.04)\) is even. Therefore, the \( \text{Bi}_{1-x}\text{Sb}_x \) \((x = 0.04)\) sample is not a strong topological insulator either, as expected from the band diagram in figure 3(a). On the other hand, the clear spin-polarized band structure shown in figure 8 signifies three \( E_F \) crossings (odd number of times) for the spin-split surface states. Consequently, one can unambiguously conclude that \( \text{Bi}_{1-x}\text{Sb}_x \) \((x = 0.12–0.13)\) is a strong topological insulator, as predicted in figure 3(a) [13]. It should be noted that the previous spin-integrated [22] and partially spin-resolved [23] experiments reached the same conclusion, although the number of \( E_F \) crossings was different. The present study not only disentangled the complicated band structure suggested in [22, 23], but also unambiguously elucidated which of the many bands represents the topologically nontrivial nature; in the present case, it is the \( \Sigma_1' \) band that makes \( \text{Bi}_{1-x}\text{Sb}_x \) a strong topological insulator.

Recently, new topological invariants, mirror Chern number and mirror chirality, have been introduced [21], which is analogous to the spin Chern number defined in the 2D QSH state in graphene [9, 10]. The main distinction is the presence of a crossing point (Dirac point) of the spin-split surface bands, \( \Sigma_1 \) and \( \Sigma_2 \), that are filled at the two symmetry points, \( \bar{\Gamma} \) and \( \bar{M} \) [21]. The mirror chirality \( \eta \), which is derived from the mirror Chern number, is \(+1\) for the crossing case (figures 1(d) and (e)), and \(-1\) (figures 1(b) and (c)) for the non-crossing case [21]. As is clear from figure 8, the \( \Sigma_1 \) and \( \Sigma_2 \) bands do not cross each other, which leads to the conclusion that \( \eta = -1 \) (the mirror Chern number, \( \eta_M \), is \( \eta_M = -1 \)) and the surface band structure of \( \text{Bi}_{1-x}\text{Sb}_x \) \((x = 0.12–0.13)\) correspond to the band diagram of figure 1(c). Intriguingly, this implies that the \( g \) factor is negative in the 3D QSH phase of \( \text{Bi}_{1-x}\text{Sb}_x \), which has also been indirectly inferred for pure Bi [21] from the SARPES data [18]. It is useful to note that the spin-resolved surface band structure elucidated here is consistent with a theoretical approach [12], which considers the hybridization between the topological surface state and other trivial surface states.
4. Concluding remarks

A high-yield spin polarimeter using VLEED was developed and equipped at a high-resolution electron spectrometer to perform SARPES measurement with energy resolution $\lesssim 50 \text{meV}$. With the present experimental system, the spin-polarized band structure of the 3D topological insulator $\text{Bi}_{1-x}\text{Sb}_x$ ($x = 0.12$–$0.13$) was fully elucidated. Between the two time-reversal-invariant points, $\bar{\Gamma}$ and $\bar{M}$ of the (111) surface Brillouin zone, the number of edge-state bands crossing the Fermi level was odd, giving unambiguous evidence for the strong topological insulator phase. The observed spin-polarized band dispersions determine the ‘mirror chirality’ to be $-1$, which excludes the existence of a Dirac point in the middle of the $\bar{\Gamma}$–$\bar{M}$ line and agrees with the theoretical prediction based on first-principles calculations.

Acknowledgments

Shuichi Murakami is gratefully acknowledged for valuable comments and discussions. This work was partly supported by JSPS (KAKENHI 18360018, 19340078, 19674002, 20030004 and 21244048) and by AFOSR (AOARD-08-4099).

References

[1] Murakami S, Nagaosa N and Zhang S-C 2003 Science 301 1348
[2] Sinova J, Culcer D, Niu Q, Sinitsyn N A, Jungwirth T and MacDonald A H 2004 Phys. Rev. Lett. 92 126603
[3] Bernevig B A and Zhang S-C 2006 Phys. Rev. Lett. 96 106802
[4] Sheng D N, Weng Z Y, Sheng L and Haldane F D M 2006 Phys. Rev. Lett. 97 036808
[5] Bernevig B A, Hughes T L and Zhang S-C 2006 Science 314 1757
[6] Konig M, Wiedmann S, Brune C, Roth A, Buhmann H, Molenkamp L W, Qi X-L and Zhang S-C 2007 Science 318 766
[7] Murakami S 2006 Phys. Rev. Lett. 97 236805
[8] Murakami S and Kuga S I 2008 Phys. Rev. B 78 165313
[9] Kane C L and Mele E J 2005 Phys. Rev. Lett. 95 146802
[10] Kane C L and Mele E J 2005 Phys. Rev. Lett. 95 226801
[11] Murakami S 2007 New. J. Phys. 9 356
[12] Zhang H-J, Liu C-X, Qi X-L, Deng X-Y, Dai X, Zhang S-C and Fang Z 2009 Phys. Rev. B 80 085307
[13] Fu L and Kane C L 2007 Phys. Rev. B 76 045302
[14] Fu L, Kane C L and Mele E J 2007 Phys. Rev. Lett. 98 106803
[15] Uhrberg R I G and Hansson G V 1991 Critic. Rev. Solid State Mater. Sci. 17 133
[16] Matsuda I, Hirahara T, Konishi M, Liu C, Morikawa H, D’angelo M, Hasegawa S, Okuda T and Kinoshita T 2005 Phys. Rev. B 71 235315
[17] Bychkov Y A and Rashba E I 1984 JETP Lett. 39 78
[18] Hirahara T et al 2008 New. J. Phys. 10 083038
[19] Hoesch M, Muntwiler M, Petrov V N, Hengsberger M, Patthey L, Shi M, Falub M, Greber T and Osterwalder J 2004 Phys. Rev. B 69 241401
[20] Lenoir B, Cassart M, Michenaud J-P, Scherrer H and Scherrer S 1996 J. Phys. Chem. Solids 57 89
[21] Teo J C Y, Fu L and Kane C L 2008 Phys. Rev. B 78 045426
[22] Hsieh D, Qian D, Wray L, Xia Y, Hor Y S, Cava R J and Hasan M Z 2008 Nature 452 970
[23] Hsieh D et al 2009 Science 323 919
[24] Okuda T, Takeichi Y, Maeda Y, Harasawa A, Matsuda I, Kinoshita T and Kakizaki A 2008 Rev. Sci. Instrum. 79 123117
[25] Taskin A A and Ando Y 2009 Phys. Rev. B 80 85303
[26] Osterwalder J 1997 Surface Rev. Lett. 4 391
[27] Hüfner S 1995 Photoelectron Spectroscopy (Berlin: Springer)
[28] Kisker E, Clauberg R and Gudat W 1982 Rev. Sci. Instrum. 53 1137
[29] Qiao S, Kimura A, Harasawa A, Sawada M, Chung J-G and Kakizaki A 1997 Rev. Sci. Instrum. 68 4390
[30] Petrov V N, Grebenshikov V V, Grachev B D and Kamochkin A S 2003 Rev. Sci. Instrum. 74 1278
[31] Petrov V N, Grebenshikov V V, Andronov A N, Gabdullin P G and Maslevtcov A V 2007 Rev. Sci. Instrum. 78 025102
[32] Hugo Dil J 2009 J. Phys. Condens. Matter 21 403001
[33] Wang G-C 1981 Phys. Rev. B 23 1761
[34] Unguris J, Pierce D T and Celotta R J 1986 Rev. Sci. Instrum. 57 1314
[35] Jungblut R, Roth Ch, Hillebrecht F U and Kisker E 1992 Surf. Sci. 269/270 615