Unconventional Mass Acquisition of Surface Dirac Fermions in a Topological Insulator

A topological insulator is a novel quantum state of matter where an insulating bulk hosts a linearly dispersing surface or edge state, which can be viewed as a sea of massless Dirac fermions protected by time-reversal symmetry. By tracing the surface and bulk electronic states using angle-resolved photoemission spectroscopy, we found that the solid solution system TlBi($S_{1-x}Se_x$)$_2$ goes through a quantum phase transition from the topological to the non-topological insulator. Furthermore, we observed that the massless Dirac state in $TlBiSe_2$ switches to a massive state before it disappears in the non-topological phase. This result suggests that Dirac particles at surfaces acquire mass without explicitly breaking time-reversal symmetry.

In three-dimensional (3D) topological insulators, insulating bulk hosts gapless topological surface states with a Dirac-cone energy dispersion [1, 2] which appear within the bulk excitation gap generated by the large spin-orbit coupling. In topological insulators, time-reversal symmetry and spin-orbit coupling characterize the spin-helical Dirac fermions which are immune to backward scattering by nonmagnetic impurities or disorder and which carry dissipationless spin current, holding promise for exploring fundamental physics, spintronics, and quantum computing [1, 2]. Strong spin-orbit coupling can lead to an inversion of the character of valence- and conduction-band wave functions, resulting in an odd Z_2 invariant that characterizes the topological insulator. All known topological insulators are based on this band-inversion mechanism, but the successive evolution of electronic states across the quantum phase transition from trivial to topological has not been well studied in 3D topological insulators due to the lack of suitable materials. TlBi($S_{1-x}Se_x$)$_2$ is the first system where one can investigate the topological quantum phase transition in 3D topological insulators. The advantage of this system is that it always maintains the same crystal structure irrespective of the S/Se ratio.

To elucidate the evolution of the surface and bulk electronic states in TlBi($S_{1-x}Se_x$)$_2$, we performed high-resolution angle-resolved photoemission spectroscopy (ARPES) at BL-28A and Tohoku University [3, 4].

Figure 1(a) shows the near-E_F ARPES intensity around the Brillouin-zone center for a series of x values including $TlBiSe_2$ ($x = 1.0$) and $TlBiS_2$ ($x = 0.0$). One can immediately see that the surface state is seen for $x \geq 0.6$, whereas it is absent for $x \leq 0.4$, which points to the topological quantum phase transition occurring at $x_c \approx 0.5$. In fact, the bulk band gap estimated from our data approaches zero on both sides of the phase transition, suggesting that a band inversion takes place across the phase transition, in accordance with the natural expectation.

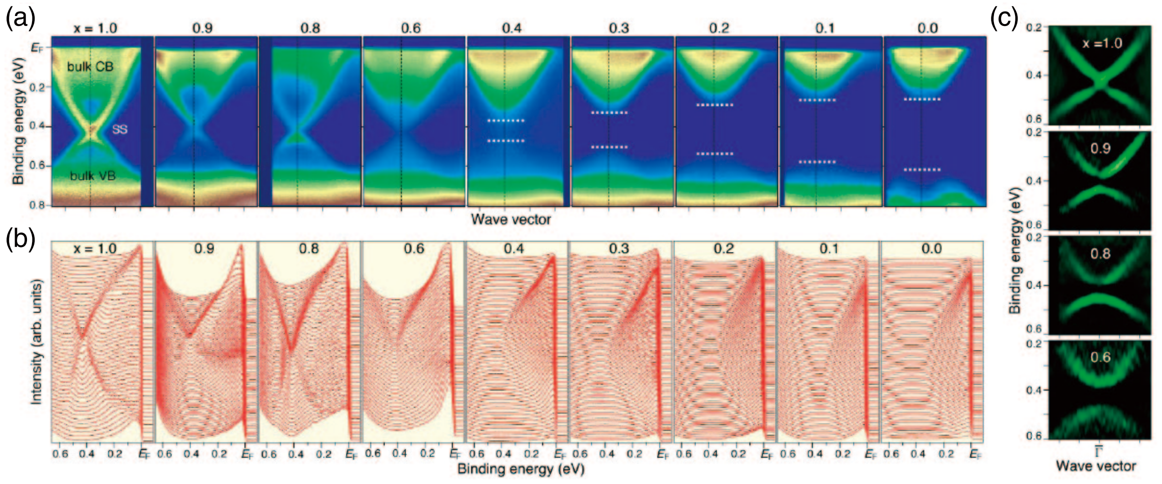

Figure 1
(a) ARPES intensity and (b) corresponding energy distribution curves of TlBi($S_{1-x}Se_x$)$_2$ showing the mass acquisition of surface Dirac fermions. (c) Second-derivative intensity for $x = 1.0$–0.6.

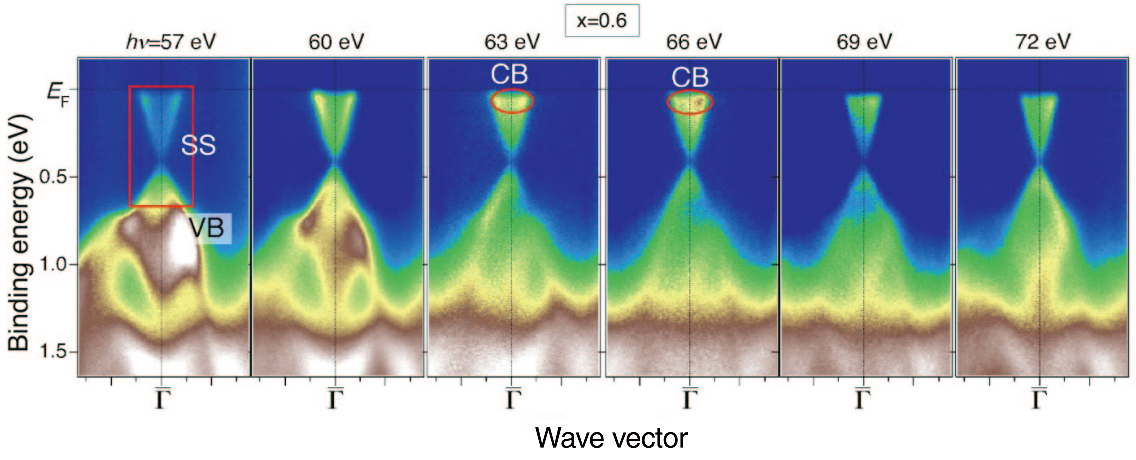

Figure 2
Photon-energy dependence of the ARPES intensity for TlBi($S_{0.4}Se_{0.6}$)$_2$.

The unexpected physics manifests itself at the Dirac point. The bright intensity peak at ~0.4 eV at $x = 1.0$ is a fingerprint of the Kramers degeneracy at the Dirac point, but it is no longer visible at $x = 0.9$ and is markedly suppressed at $x = 0.6$, suggesting that the Kramers degeneracy is lifted upon S substitution while the surface state is still present. The surface-state nature of this band was confirmed by its stationary nature of the energy position with respect to the photon energy as shown in Fig. 2, so this band evidently represents the massive Dirac fermions on the surface. Interestingly, the surface band gap, called here the Dirac gap, grows as x decreases (less than 0.1 eV at $x = 0.9$ and 0.8, and larger than 0.1 eV at $x = 0.6$), indicating that the S content is closely related to the magnitude of the Dirac gap. Taking into account that all the elements contained in TlBi($S_{1-x}Se_x$)$_2$ are nonmagnetic and also that the sample shows no obvious magnetic order, our result strongly suggests that the substitution of Se with S in TlBi($S_{1-x}Se_x$)$_2$ leads to an unconventional mass acquisition of the surface Dirac fermions *without* explicitly breaking the time-reversal symmetry. The present Dirac gap can be much larger than that of the magnetically-doped topological insulator Bi_2Se_3 [5] and is tunable with the S/Se ratio, making the TlBi($S_{1-x}Se_x$)$_2$ system a prime candidate for device applications of topological insulators that require a gapped surface state.

REFERENCES

[1] M.Z. Hasan and C.L. Kane, *Rev. Mod. Phys.*, **82** (2010) 3045.
[2] X.-L. Qi and S.-C. Zhang, *Rev. Mod. Phys.*, **83** (2011) 1057.
[3] T. Sato, K. Segawa, K. Kosaka, S. Souma, K. Nakayama, K. Eto, T. Minami, Y. Ando and T. Takahashi, *Nature Phys.*, **7** (2011) 840.
[4] S. Souma, K. Kosaka, T. Sato, M. Komatsu, A. Takayama, T. Takahashi, M. Kriener, K. Segawa and Y. Ando, *Phys. Rev. Lett.*, **106** (2011) 216803.
[5] Y.-L. Chen, J.-H. Chu, J.G. Analytis, Z.K. Liu, K. Igarashi, H.-H. Kuo, X.L. Qi, S.K. Mo, R.G. Moore, D.H. Lu, M. Hashimoto, T. Sasagawa, S.-C. Zhang, I.R. Fisher, Z. Hussain and Z.-X. Shen, *Science*, **329** (2010) 659.

BEAMLINE

28A

S. Souma[1], T. Sato[1], K. Kosaka[1], M. Komatsu[1], A. Takayama[1], K. Nakayama[1], T. Takahashi[1], K. Eto[2], T. Minami[2], M. Kriener[2], K. Segawa[2] and Y. Ando[2] (Tohoku Univ., [2]Osaka Univ.)