

Core-level photoelectron spectroscopy study of the TlBiSe₂ surface

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Introduction

We have studied the (111) surface of TlBiSe₂ with core-level photoelectron spectroscopy (PES). The material is known to be one of three-dimensional topological insulators. Angle-resolved PES measurements of TlBiSe₂ demonstrated the Dirac-cone surface state in the bulk band gap of ~ 0.3 eV [1, 2]. Such electronic structure is expected to open the application of topological insulators to practicable devices operating at room temperature. On the other hand, a recent theoretical study by ab initio calculation showed that the surface-state band dispersion depends crucially on surface termination, relaxation and reconstruction, and is not reproduced by the simple models with terminations of Tl, Bi and Se [3]. Until now, the atomic structure of the cleaved TlBiSe₂ surface was not experimentally revealed.

Experimental

The core-level PES measurements of Tl 5d, Bi 5d and Se 4d were performed at beam line 18A. A TlBiSe₂ crystal was grown by the Bridgman method. The (111) orientation of the sample was confirmed by X-ray diffraction. The sample was cleaved at pressure below 1×10^{-7} Torr in the preparation chamber and immediately transferred to the analyzer chamber at 2×10^{-10} Torr. Low-energy electron diffraction (LEED) showed a sharp (1 \times 1) pattern with low background intensity. We confirmed the Dirac-cone surface state with valence PES by a SES100 spectrometer. We used an ADES-500 spectrometer to measure the emission-angle and photon-energy dependence of the core levels with a fixed incident angle of 45°. The used photon energy ranges from 50 to 130 eV. All PES measurements were performed at room temperature.

Results and discussion

Figure 1(a) shows the normal-emission core-level spectra with 90-eV photons. It is found that a shoulder appears on the high binding-energy side of each main peak of Tl 5d, in contrast to the Bi and Se peaks with a single component. The relative intensity of the lower-binding-energy component (labeled α) decreases for lower photon energies. With 70-eV photons, the two components of Tl 5d have comparable intensity in normal emission as shown in Fig. 1(b). With increasing the emission angle, it is found that the peak intensity of the component α rapidly decreases.

The TlBiSe₂ crystal has a rhombohedral structure, which is characterized by the stacking sequence of -Se-Bi-Se-Tl-Se- along the [111] direction. Each layer consists of

hexagonally-closed-packed layers of Tl, Bi and Se. The larger interlayer distance of Tl-Se indicates cleavage between the Tl and Se layers. If no surface reconstruction is assumed, Tl or Se terminated surface is likely to appear on the cleaved surface.

Taking into accounts the emission-angle dependence of Tl 5d and the single component peak of Se 4d, it is proposed that the higher-binding-energy component (β) is attributed to the surface Tl atoms. The observed binding energy shift of ~ 0.4 eV indicates that the state of the surface Tl atom is significantly different from that in the bulk. We are proceeding with further analysis of the surface atomic structure in combination with the core-level PES data, dynamical LEED I - V curve analysis and scanning tunneling microscopy observation.

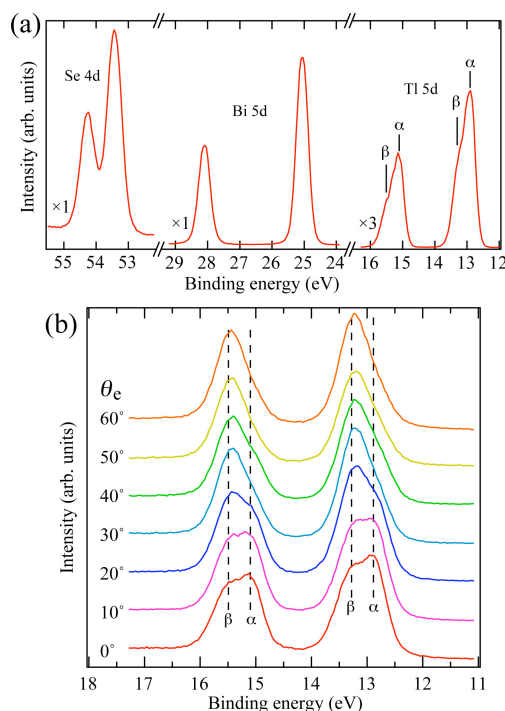


Fig. 1. (a) Normal-emission core-level spectra of the cleaved TlBiSe₂ surface with $h\nu=90$ eV. (b) The emission angle dependence of Tl 5d with $h\nu=70$ eV.

References

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