**Electronic Structure of Condensed Matter** 

# Spin-resolved photoemission study on Dirac-like surface states of ultrathin Bi<sub>2</sub>Se<sub>3</sub> films

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## **Introduction**

Recently there has been growing interest in topological insulators (3D) or the quantum spin Hall (QSH) phase (2D), which are insulating materials with bulk band gaps but have metallic edge states that are formed topologically and robust against any non-magnetic impurity [1]. In a three-dimensional material, the twodimensional surface states correspond to the edge states (topological metal). Due to the loss of the inversion symmetry, they have spin-filtered properties which are actually the origin of the topological robustness. Since the surface states of Bi2Se3 with Dirac-like linear band dispersion have a very small Fermi wave number, it has been difficult to actually observe the spin-split properties. Therefore in the present study, we have conducted highresolution spin- and angle- resolved photoemission (SARPES) studies on ultrathin Bi<sub>2</sub>Se<sub>3</sub> films to directly verify their spin polarization.

### <u>Experimental</u>

The experiments were performed at BL-19A. The system has a hemispherical analyzer (SPECS Phoibos-150) equipped with a homemade high-yield spinpolarimeter using spin-dependent very-low-energy electron diffraction [1]. A He lamp (He I $\alpha$  =21.2 eV) was used as the excitation source and the SARPES spectra were recorded at 150 K with the energy and angle resolutions of 30 meV and 0.7°, respectively. The effective Sherman function in this experiment was 0.3 as calibrated by the spin-polarized secondary electrons from a standard Ni sample.

The ultrathin Bi<sub>2</sub>Se<sub>3</sub> films were fabricated in situ in a method similar to the report of Ref. [2]. First, a clean Si(111)-7 $\times$ 7 surface was prepared on an n-type substrate (P-doped, 1-10  $\Omega$ cm at room temperature) by a cycle of resistive heat treatments. The Si(111)  $\beta\sqrt{3x}\sqrt{3}$ -Bi surface was formed by 1~ML (7.83×1014 cm-2) of Bi deposition on the 7×7 surface at 620 K monitored by Reflection High Energy Electron Diffraction. Then Bi was deposited on the  $\beta\sqrt{3x}\sqrt{3}$ -Bi structure at ~400 K in a Se-rich condition. Such a procedure is reported to result in a smooth epitaxial film formation with the stoichiometric ratio of Bi:Se=2:3 [2]. It is also known that the minimum film thickness that can be achieved in this method is one quintuple layer (1 QL = 10 Å), and the films can be formed QL-by-QL. We have carefully checked the film thickness by monitoring the spot intensity of the RHEED pattern [3] and the film thickness in the present measurements was 8 QL (80Å).

## **Results and discussions**

Figures 1(a) and (c) show the SARPES spectra  $(I_{\uparrow}, I_{\downarrow})$ near the point along the  $\Gamma$ -M direction (x direction) for negative (a) and positive (c) emission angles, respectively, and Figs. 1(b) and (d) show the corresponding spinpolarization curves P=(  $I_{\uparrow}$ -  $I_{\downarrow}$ )/ ( $I_{\uparrow}$ +  $I_{\downarrow}$ ). The spin orientation is in the +y (red curves) [-y (blue curves)] direction. The photoemission intensity for the bulk states is much stronger than that of the surface states and it is really difficult to resolve the two in the energy distribution curves of Figs. 1(a) and (c). However, if we look carefully at  $I_{\uparrow}$  and  $I_{\downarrow}$  we notice that  $I_{\uparrow} < I_{\downarrow}$  for negative angles,  $I_{\uparrow} = I_{\downarrow}$  just near normal emission, and  $I_{\uparrow} >$  $I_{\!\!\perp}$  for positive angles near the Fermi level. This can also be noticed by the hatched areas in the spin-polarization curves of Figs. 1(b) and (d). The spin-polarized states seem to disperse toward EF as they pull off from normal emission. We believe that this is representing the helical nature of the spin-split surface states. The obtained spinpolarization values are  $\sim 10$  % at maximum, which is very small probably due to the bulk state close by. Another effect may come from a technical problem concerning the He IB and/or He II radiation enhancing the background of the spectra.



Figure 1: Spin-resolved ARPES spectra of an  $8 \sim QL$  ultrathin Bi<sub>2</sub>Se<sub>3</sub> film along ther-M direction (x direction) [(a) and (c)] and the spin polarization curves deduced from them [(b) and (d)]. The spin orientation is along ther-K direction (y direction, red (blue) is for +y (-y) direction).

#### References

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