Spin- and angle-resolved photoemission study on Dirac-like surface states of ultrathin Bi₂Se₃ films

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<u>Introduction</u>

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 two-dimensional 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 high-resolution spin- and angle- resolved photoemission (SARPES) studies on ultrathin Bi₂Se₃ films to directly verify their spin polarization.

Experiment

The experiments were performed at BL-19A of Photon Factory-KEK. The system has a hemispherical analyzer (SPECS Phoibos-150) equipped with a homemade high-yield spin-polarimeter using spin-dependent very-low-energy electron diffraction (VLEED) [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 Bi₂Se₃ films were fabricated *in situ* in a method similar to the report of Ref. [2]. First, a clean Si(111)-7×7 surface was prepared on an *n*-type substrate (P-doped, 1-10 Ω cm at room temperature) by a cycle of resistive heat treatments. The Si(111) $\beta \sqrt{3} \times \sqrt{3}$ -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 $\beta \sqrt{3} \times \sqrt{3}$ -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 Γ point along the Γ -M direction (x direction) for negative (a) and positive (c) emission angles, respectively, and Figs. 1(b) and (d) show the corresponding spin-polarization 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

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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 $I_{\uparrow} > I_{\downarrow}$ 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 E_F as they pull off from normal emission. We believe that this is representing the helical nature of the spin-split surface states. The obtained spin-polarization values are ~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 I β and/or He II radiation enhancing the background of the spectra [4].

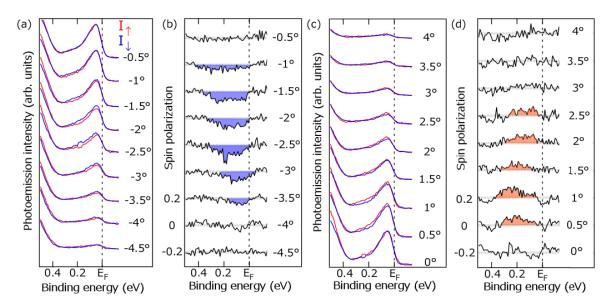


Figure 1: Spin-resolved ARPES spectra of an $8\sim QL$ ultrathin Bi_2Se_3 film along the Γ -M direction (x direction) [(a) and (c)] and the spin polarization curves deduced from them [(b) and (d)]. The spin orientation is along the Γ -M direction (y direction, red (blue) is for +y (-y) direction).

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