Intercalation effect of layered topological insulator studied by photoelectron diffraction

Mao Ye[1†], Eike Schvier[1], Munisa Nurmamat[2], Kaito Shirai[2], Tomohiro Matsushita[3], Makoto Morita[4], Satoshi Kitagawa[4], Ryo Ishii[4], Masayoshi Fujita[4], Kaoru Yasuda[4], Laszlo Toth[4], Hiroshi Daimon[4], and Taichi Okuda[1]

[1] Hiroshima Synchrotron Radiation Centre, Hiroshima University, 2-313 Kagamiyama, Higashi-Hiroshima 739-0046, Japan.

Graduate School of Science, Hiroshima University, Kagamiyama 1-3-1, Higashi-Hiroshima 739-8526, Japan

[3] Japan Synchrotron Radiation Research Institute (JASRI), SPring-8, 1-1-1 Kouto, Sayo-cho, Sayo-gun Hyogo 679-5198, Japan.

[4] Nara Institute of Science and Technology (NAIST), 8916-5 Takayama, Ikoma, Nara 630-0192, Japan.

A novel class of quantum materials, called topological insulators (TIs) [1, 2], has provoked much research interest. A number of materials that hold nontrivial spin-polarized metallic surface states have been intensively studied. Owing to the time-reversal symmetry, topological surface states are protected from backscattering in the presence of a weak perturbation, which is important for the realization of dissipationless spin transport in novel quantum devices. However, such surface state is not protected from scattering by arbitrary angles, so the shielding of this state from impurities is an important problem.

Additionally, most of the already discovered TIs are layered materials that are constructed several-atomic-layer slabs with van der Waals (vdW) force, such as, Bi_2Se_3 and Bi_2Te_3 [3]. Such materials are ineluctably suffered from impurities that intercalated in the vdW gaps. On the other hand, we have previously discovered that such intercalation effect may result in a relocation of topological edges states below the intercalated interface [4]. Nevertheless, the existence of intercalated impurities and the modification on the topological materials are still poorly understood due to the accessibility of the structural information of the intercalated Ag atom beneath the surface (Fig.1).

In this research, we utilized x-ray photoelectron diffraction (XPD) method to study the structural modification of a prototype TIs, Bi_2Se_3 when Ag atoms are absorbed. The experiments are performed at the soft x-ray beamline BL07LSU of SPring-8 using the Display-type Ellipsoidal Mesh Analyzer (DELMA) spectrometer [5]. The single crystal of Bi_2Se_3 is cleaved *in-situ* to obtain contamination-free surface. Ag atoms are deposited from the tungsten filament basket when keeping the Bi_2Se_3 at room temperature. All measurements are conducted at room temperature.

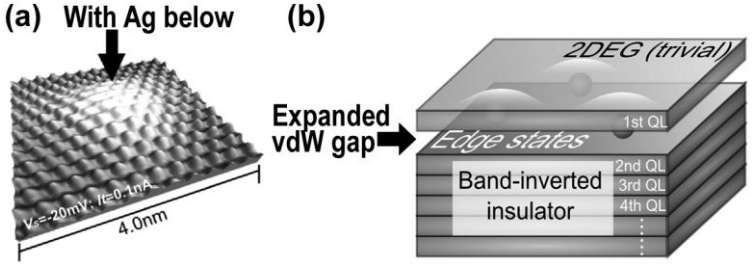

Fig1 (a) STM image of Ag-intercalated Bi_2Se_3; (b) Schematic model of relocated topological states due to the intercalated Ag atoms in the van der Waals gap.

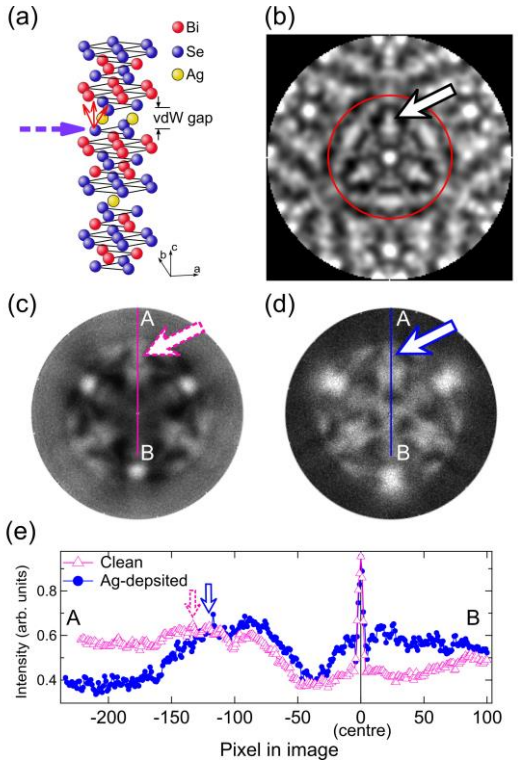

Fig.2 (a) Schematic crystal model of Ag-intercalated Bi_2Se_3; (b) Simulated XPD pattern of Se 3*d* photoelectrons, the red circle indicate the area that is experimentally measured; (c) and (d) XPD patterns of Se 3*d* photoelectrons of Bi_2Se_3 measured before and after Ag deposition (E_k = 600 eV). (e) Intensity profiles along A-B in (c) and (d), see text.

Figure 2(a) shows the schematic crystal structures of Bi_2Se_3 with Ag atoms intercalated in the vdW gaps that are expected to be expanded due to the Ag-atom-intercalation. To experimentally verify such structural modification, we examined the XPD patterns of Se 3*d* photoelectrons. Because the distance between the Se atoms just below and above the vdW gaps may be elongated due to the expansion of vdW gaps, the XPD patterns originated from the forward-focusing-peak (FFP) of Se atoms just below the vdW gaps [blue arrow in Fig.2(a)] should be modified. Figure 2(b) shows the theoretically simulated XPD patterns of pristine Bi_2Se_3 originated from Se 3*d* photoelectrons (E_k = 600 eV), where the intensity resulted from FFP of Se atoms just below the van der Waals gaps is indicated by an arrow.

Figures 2(c) and 2(d) compare the experimentally measured XPD pattern of Se 3*d* photoelectrons before and after Ag deposition. Note that the experimental data taken on pristine Bi_2Se_3 sample in Fig.2(c) keeps good consistence with the simulated pattern in Fig. 2(b). Fig. 2(d) shows the XPD pattern after Ag deposition. The FFP intensity from the Se 3*d* photoelectrons just below the vdW gap is indicated by a purple (blue) arrow before (after) Ag deposition in Fig.2(c) [Fig.2(d)]. In order to find the changes of XPD patterns, the line profiles across the FFP intensity from Se below the vdW gap are shown in Fig.2(e). As indicated by purple and blue arrows, the peak originated from the FFP intensity slightly shifts towards the centre of the image, indicating a possible expansion of vdW gaps after Ag deposition.

This XPD measurement is conducted at SPring-8 with the approval of the Proposal Assessing Committee of Institute for Solid State Physics, the university of Tokyo (JASRI Proposals No. 2012A7427). This work was partly supported by JSPS Research Fellowship (23-7303) and KAKENHI(19340078,23244066), Grant-in-Aid for Scientific Research(B) and (A) of Japan Society for the Promotion of Science.

References

[1] H. Zhang, *et al.*, Nature Phys. **5**, 438 (2009).
[2] M. Z. Hasan, and C. L. Kane, Rev. Mod. Phys. **82**, 3045-3067 (2010).
[3] Y. L. Chen, *et al.* Science **325,** 178 (2009).
[4] M. Ye, *et al.*, arXiv:1112.5869 [cond-mat.mes-hall]
[5] H. Daimon, H. Matsuda, L. Toth and F. Matsui: Surf. Sci. **601**, 4748 (2007).

† Present Address: Shanghai Institute of Microsystem and Information Technology, Chinese Academy of Sciences. 865 Chang Ning Road Shanghai 200050, China. Email: yemao@mail.sim.ac.cn