Interface electronic structure at the topological
insulator–ferrimagnetic insulator junction

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An interface electron state at the junction between a three-dimensional topological insulator (TI) film of Bi$_2$Se$_3$ and a ferrimagnetic insulator film of Y$_3$Fe$_5$O$_{12}$ (YIG) was investigated by measurements of angle-resolved photoelectron spectroscopy and X-ray absorption magnetic circular dichroism (XMCD). The surface state of the Bi$_2$Se$_3$ film was directly observed and localized 3$d$ spin states of the Fe$^{3+}$ state in the YIG film were confirmed. The proximity effect is likely described in terms of the exchange interaction between the localized Fe 3$d$ electrons in the YIG film and delocalized electrons of the surface and bulk states in the Bi$_2$Se$_3$ film. The Curie temperature ($T_C$) may be increased by reducing the amount of the interface Fe$^{3+}$ ions with opposite spin direction observable as a pre-edge in the XMCD spectra.

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I. INTRODUCTION

Topological insulators (TIs) are notable materials currently attracting a wide interest in both fundamental and applied research. Although TIs show bulk insulating performance, they exhibit Dirac-like gapless bands at their surfaces. The surface state is ensured by time-reversal symmetry (TRS) and the spin polarization of the surface state electrons is locked to its momentum. Because these properties are resistant to non-magnetic external perturbations, TIs are expected to be promising materials for new spintronic devices.

By breaking TRS, TIs exhibit a number of interesting features, such as the gap-opening at the Dirac points, the half quantum Hall effect, the quantum anomalous Hall effect, and the topological magnetoelectric effect. There are two methods for breaking the TRS: one is by doping magnetic impurities (Cr, Fe and Mn) and the other is by connecting TIs to magnetic materials (such as Fe, Co and EuS). However, with the objective of device applications, magnetic metals in contact with TIs are not appropriate because the TI surface state is short circuited by the metallic material. Furthermore, the Curie temperatures ($T_C$) of these TIs that are doped by magnetic impurities or contacted with the magnetic insulator (EuS) are typically below 35 K and, therefore, raising $T_C$ is required for practical applications.

Recently, it was suggested that a ferrimagnetic insulator, yttrium iron garnet (YIG, Y$_3$Fe$_5$O$_{12}$) with $T_C \sim 550$ K, has the potential to be an underlayer for magnetic TI films. It was reported, from magnetotransport and magneto-optical measurements, that $T_C$ of the Bi$_2$Se$_3$/YIG system reaches $\sim 130$ K due to the proximity effect. Furthermore, $T_C$ of Cr-doped Bi$_2$Se$_3$ on YIG was found to be higher than that on a nonmagnetic substrate through magneto-transport and X-ray absorption magnetic circular dichroism (XMCD) measurement. Understanding the mechanism of the proximity effect between TI and YIG is required to realize $T_C$ above room temperature (RT).

In this paper, we present results of angle-resolved photoelectron spectroscopy (ARPES) and XMCD for Bi$_2$Se$_3$ films on YIG. We have successfully observed the TI surface state in this Bi$_2$Se$_3$/YIG system and obtained direct evidence that the 3$d$ electrons of Fe in YIG induce the proximity effect at the interface between TI and YIG. This result represents a potential advance for the optimization of the properties of the magnetic layer to realize practical new devices.

II. EXPERIMENT

Figure (a) shows a schematic drawing of the sample. YIG (111) thin films (8.4 nm thick) were prepared on gadolinium gallium garnet (GGG) (111) substrates. The details of YIG growth can be found in the Supplemental Material. Bi$_2$Se$_3$ films were then grown on YIG/GGG using Bi and Se conventional effusion cells. At first, 2 quintuple layers (QLs) Bi$_2$Se$_3$ films were grown on YIG which was maintained at 150°C. The samples were...
Figure 1 presents the RHEED pattern of the 6 QL Bi₂Se₃/YIG sample. The symbol a* represents the reciprocal lattice constant of Bi₂Se₃, then annealed at 300°C, followed by further Bi and Se deposition at 250°C. The thickness of the Bi₂Se₃ films was controlled from 3 to 6 QL by observation of reflection high-energy electron diffraction (RHEED) intensity oscillations, as shown in Figure 1(b). After the deposition of the Bi₂Se₃ films at 250°C, the samples were further annealed at 250°C for 15 min to realize a better crystalline quality. Figure 1(c) presents the RHEED pattern of the 6 QL Bi₂Se₃/YIG sample. Referred from the (00)-rod, streaks are identified at 2a* and √3×a* where a* represents the reciprocal lattice constant of Bi₂Se₃. The pattern indicates that the Bi₂Se₃ films have a multi-domain structure such as a ⟨111⟩-oriented texture structure. The sample surface was capped with a 30 nm-thick Se layer and the sample wafer was transferred in air to the ARPES measurement chamber.

ARPES measurements for the 6 QL Bi₂Se₃ film were performed on the VUV-Photoemission beamline at Eletra. After removing the Se capping layer by annealing at 190°C, p-polarized light was incident onto the sample. Measurements of XMCD were performed on 3 and 6 QL Bi₂Se₃ films on the YIG films at RT and 20 K at the BL07LSU beamline at SPring-8.

III. RESULT AND DISCUSSION

A. Angle-resolved photoelectron spectroscopy

Figure 2(a) shows the momentum (k∥) distribution curves (MDCs) of the ARPES spectra around the Γ point, taken at hν = 52.4 eV, at T = 30 K and RT. The MDCs obtained at T = 30 K have slightly narrower peaks than those at RT due to reduction of the thermal blurring. In the figure, it can be seen that the two peaks in the MDCs at the Fermi level (E_F) approach with binding energy (E_b) and overlap each other at E_b = 0.38 eV, followed by separation at higher E_b. These results unambiguously indicate the band-crossing. The MDC peaks in Figure 2(a) were curve-fitted by two peaks and the peak positions are plotted in the photoelectron band diagram in Figure 2(b, c). At T = 30 K and RT, the surface state band shows the Dirac-cone dispersion around the Γ point with the Dirac point at E_F = 0.38 eV. The band-dispersion curves were assigned to those of the Dirac surface state bands of the Bi₂Se₃ film as reported previously. The Fermi velocity of this system is v_F = 5.1×10⁵ m/s and this value agrees with previous studies. For comparison, the photoelectron band diagram, taken at hν = 23.1 eV, is also shown in Figure 2(d). The observed band between E_F and 0.2 eV is assigned to the bulk conduction band of Bi₂Se₃ and it crosses the E_F, indicating the n-type doped nature. The electronic structures of the surface and bulk states are essentially similar to the previous ARPES results of Bi₂Se₃ films on different substrates. Due to the TI nature of Bi₂Se₃ film, the existence of the surface state at the film/vacuum interface suggests its presence also at the junction (interface) with the YIG film. Moreover, Figure 2(d) implies that the Bi₂Se₃ bulk conduction band crosses E_F at the Bi₂Se₃/YIG interface. It is of note that the surface state band structure does not change with the temperature across T_C ≈ 130 K, as shown in Figure 2.

B. X-ray absorption magnetic circular dichroism

Figure 3(a) shows the XMCD measurement configuration. A magnetic field of 0.24 T was applied by a retractable permanent magnet. The XMCD was measured at the Fe L_2,3-shell absorption edge of YIG by the total electron yield (TEY) mode. XMCD spectra were derived from the difference between the two adsorption spectra obtained by circularly polarized light of opposite helicities, where the beam direction was set parallel to the magnetic field orientation and to the surface normal direction. Figures 3(b, c) show the Fe 2p X-ray absorption spectra (b) and XMCD (c). The spectral shapes are mostly in agreement with those of the Cr-doped Bi₂Se₃/YIG sample reported by Liu et al. Since the probing depth of the present XMCD measurement is about 5 ~ 10 nm not much different from the thickness of 3 nm (3 QL) and 6 nm (6 QL) Bi₂Se₃, the XMCD signals are thought to be essentially resulting from the Fe atoms near the Bi₂Se₃/YIG boundary. The positive and negative XMCD peaks at the L_3-edge suggest the opposite spin direction in the Fe atoms, at two different sites in the YIG crystal, octahedral (2 per formula unit) and tetrahedral sites (3 per formula unit), as expected for the ferrimagnet. In the present measurement configuration, the macroscopic magnetic direction of the ferrimagnetic YIG film shows a negative peak for
the tetrahedral Fe site\(^{24}\). While the overall Fe 2p X-ray absorption spectra and XMCD are similar in spite of the different thickness and temperature, we notice a slight difference at the pre-edge structure that may depend on thickness and temperature as described below.

Figure 4 shows enlarged images of the X-ray absorption (a) and XMCD (b) spectra in \(h\nu = 705.2 \sim 708.5\) eV. One can notice peak shoulders at \(h\nu \sim 706.5\) eV in the \(\sigma^+\) spectra of the 3 QL Bi\(_2\)Se\(_3\) film at \(T = 20\) K and the 6 QL Bi\(_2\)Se\(_3\) film at RT. Compared with the previous X-ray absorption study\(^{22}\), the spectral features are likely assigned to the Fe\(^{2+}\) state in the YIG crystal. These results indicate the existence of the Fe\(^{2+}\) state at the Bi\(_2\)Se\(_3\)/YIG interface. The pre-edge peak (\(h\nu \sim 706.5\) eV) shows a positive XMCD signal and, thus, it is natural to assume that the Fe\(^{2+}\) possesses an opposite spin direction from the net magnetization of the YIG crystal. It is intriguing to note that the pre-edge feature is seemingly suppressed in the X-ray absorption and XMCD spectra of the 6 QL Bi\(_2\)Se\(_3\) film at \(T = 20\) K. We infer that these Fe\(^{2+}\) states with the opposite spin direction may prevent the ferromagnetism of Bi\(_2\)Se\(_3\) at the interface. While the interface Fe\(^{2+}\) state is considered to be related to the Bi-Fe or Se-Fe interaction at the Bi\(_2\)Se\(_3\)/YIG junction, the proper assignment requires determination of the accurate interface atomic structure and appropriate theoretical calculations.

C. Spin interaction at the interface

From the experimental results above, ferromagnetism of Bi\(_2\)Se\(_3\) at the interface would be associated with the interface spin-polarized states of the Bi\(_2\)Se\(_3\) film and localized spin states of the interface Fe\(^{3+}\) in the YIG film. Thus, the proximity effect would be modeled as their spin exchange interactions at the boundary, as shown in Fig. 5. Such interface interaction has been already calculated for a similar system that is composed of a Bi\(_2\)Se\(_3\) film and an EuS substrate\(^{33}\). When the TI film has a surface state
FIG. 3: (a) A schematic drawing of the XMCD experimental set up. (b) XAS spectra of the 3 and 6 QL Bi$_2$Se$_3$/YIG samples obtained by the total electron yield (TEY) detection at RT and 20 K. The solid red and blue lines represent the spectra taken with circular-polarized light of plus and minus helicity ($\sigma^+$, $\sigma^-$), respectively. (c) XMCD spectra of 3 QL Bi$_2$Se$_3$/YIG at $T = 20$ K (red), 6 QL Bi$_2$Se$_3$/YIG at $T = 20$ K (green), and 6 QL Bi$_2$Se$_3$/YIG at RT (blue).

FIG. 4: Enlarged figures of the (a) XAS and (b) XMCD spectra in Fig. 3. The solid horizontal lines in (b) represent XMCD = XAS ($\sigma^+$) − XAS ($\sigma^-$) = 0. Peaks of the $\sigma^+$ and XMCD spectra in this photon energy region are observed in the data of the 3 QL Bi$_2$Se$_3$/YIG at $T = 20$ K and 6 QL Bi$_2$Se$_3$/YIG at RT, as shown by arrows.

with a gap at the Dirac point by breaking the TRS, these delocalized spins were found to experience an exchange interaction with the localized spins of the 4$f$ electrons in the Eu$^{2+}$ ions\cite{33}. Moreover, it was found that bulk ($p_z$-orbital) states of the Bi$_2$Se$_3$ film also contribute to the spin-coupling between the TI and the ferromagnetic material when $E_F$ locates above the minimum of the bulk conduction band\cite{33}. By analogy, the ferromagnetism of Bi$_2$Se$_3$ at the interface can be understood as an exchange interaction between the spin-polarized electrons in the (gapped) Dirac surface state of the Bi$_2$Se$_3$ film and the localized 3$d$ electrons of the Fe$^{3+}$ in the YIG film\cite{22,23}. Moreover, there is also a contribution from the bulk electrons in the Bi$_2$Se$_3$ film since the bulk conduction band crosses $E_F$, as shown in Fig. 2 (d).
In summary, in the present research, we provide the first evidence of the surface state of the Bi$_2$Se$_3$ film on YIG by ARPES and the significance of the Fe$^{3+}$ state for ferromagnetism of the Bi$_2$Se$_3$ at the interface by Fe $L_{2,3}$-edge XMCD. The origin of the proximity effect is likely described in terms of the exchange interaction between the localized Fe$^{3+}$ 3$d$ electrons in the YIG film and the delocalized electrons of the surface state and the bulk state in the Bi$_2$Se$_3$ film. One may increase $T_C$ for developing future spintronic devices by reducing the amount of Fe$^{2+}$ ions with opposite spin direction that may exist at the interface.

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