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## Magnetic-field-induced topological phase transition in Fe-doped (Bi,Sb)<sub>2</sub>Se<sub>3</sub> heterostructures

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#### Abstract

Three-dimensional topological insulators (3D-TIs) possess a specific topological order of electronic bands, resulting in gapless surface states via bulk-edge correspondence. Exotic phenomena have been realized in ferromagnetic TIs, such as the quantum anomalous Hall (QAH) effect with a chiral edge conduction and a quantized value of the Hall resistance  $R_{yx}$ . Here, we report on the emergence of distinct topological phases in paramagnetic Fe-doped (Bi,Sb)<sub>2</sub>Se<sub>3</sub> heterostructures with varying structure architecture, doping, and magnetic and electric fields. Starting from a 3D-TI, a two-dimensional insulator appears at layer thicknesses below a critical value, which turns into an Anderson insulator for Fe concentrations sufficiently large to produce localization by magnetic disorder. With applying a magnetic field, a topological transition from the Anderson insulator to the QAH state occurs, which is driven by the formation of an exchange gap owing to a giant Zeeman splitting and reduced magnetic disorder. Topological phase diagram of (Bi,Sb)<sub>2</sub>Se<sub>3</sub> allows exploration of intricate interplay of topological protection, magnetic disorder, and exchange splitting.

#### I. Introduction

Three-dimensional topological insulators (3D-TI) are a class of matter that composed of gapless surface states and insulating bulk [1-3]. In such material systems, various types of topological phase transitions (TPTs) are expected to appear as a result of structural modifications and application of external fields [4-13]. For instance, an exchange gap formation in the surface states engenders a TPT, which leads to the quantum anomalous Hall (QAH) phase, i.e., to the Chern insulator. In this phase, similarly to the conventional quantum Hall effect (QHE), charge transport proceeds via a dissipationless chiral edge channel while the origin is completely different [14-16]; the QAHE is driven by the formation of an exchange gap and the QHE by Landau level splitting. To materialize the QAH phase in TIs, the surface electronic states should be modified by the exchange gap owing to some mechanisms (1) coupling between surface state and ferromagnetically aligned spins of transition metal impurities [15-17] or aligned spins by external magnetic field in the antiferromagnetic topological insulator [18,19], (2) proximity coupling effect with adjacent magnetic moment [20-22], or (3) band crossing driven by a Zeeman splitting [17,23]. To date, experimental observations of the QAH effect have been accomplished for Cr-doped or V-doped (Bi,Sb)<sub>2</sub>Te<sub>3</sub> (BST) 3D-TI films, where a perpendicular spontaneous magnetization develops below the Curie temperature

(type (1)) [15,16,24-28], and for a proximitized (Bi,Sb)<sub>2</sub>Te<sub>3</sub>/ferromagnetic insulator heterostructure (type (2)) [29]. In contrast, type (3) of QAH has not been observed in paramagnetic (Bi,Sb)<sub>2</sub>Se<sub>3</sub>-based TIs, whereas the quantization of the Hall resistance is accomplished by the application of a magnetic field to a 4 quintuple layer of Cr-doped (Bi,Sb)<sub>2</sub>Te<sub>3</sub>[30], Ti-doped (Bi,In)<sub>3</sub>Te<sub>2</sub> [31], and MnBi<sub>2</sub>Te<sub>4</sub> [18,19].

As another TPT, the surface state can be gapped when the 3D-TI film is sufficiently thin to activate hybridization of states from the opposite surfaces [1,2]. Thin films of Bi<sub>2</sub>Se<sub>3</sub>-type 3D-TIs are known to be 2D trivial insulators [4,32-34] when the hybridization is sufficiently strong, as presented in Fig. 1(a). Considering this trivial electronic band as an initial state, the giant Zeeman splitting generated by an external magnetic field produces a specific band crossing (Fig. 1(b)) with formation of an exchange gap (Fig. 1(c)) [17,23] at this crossing point, equivalent to that driven by type (1) and (2) in coupling with ferromagnetic systems, inducing a conceptually different type (3) of QAH. To illustrate the topological phase transition from the hybridized insulator to the QAH phase driven by the Zeeman splitting, we have calculated the band structure using a 3D-TI slab geometry. Figures 1(d)-1(f) demonstrate the band modification of 3D-TI by the inter-surface hybridization and the Zeeman effect. When  $\Delta_{hy}$  is larger than  $\Delta_{\text{Zeeman}}$ , the hybridized insulator is stabilized without surface gapless states (Fig. 1(d)). The  $\Delta_{\text{Zeeman}}$  exceeding  $\Delta_{\text{hy}}$  forms a non-trivial exchange gap with one-dimensional chiraledge channel as shown in Figs. 1(e) and 1(f), with gap size proportional to the magnetic field. In Fig. 1(f), red (blue) thick curve shows the chiral edge mode at one (the other) edge.

With a minor contribution of the ordinary Hall effect under magnetic field owing to Fermi energy ( $E_F$ ) locating in the gap, the magnetic-field-induced anomalous Hall effect plays a critical role for the quantization of the Hall conductivity. This study demonstrates the presence of TPTs between 3D-TI, a two-dimensional (2D) insulator, and QAH phases in paramagnetic Fe-doped Bi<sub>2</sub>Se<sub>3</sub>-based heterostructures by controlling Fe doping and layer thickness and by application of external magnetic and electric fields.

The tetradymite compound Bi<sub>2</sub>Se<sub>3</sub> is a representative 3D-TI, in which a large bulk band gap (approximately 300 meV) hosts gapless surface states [35]. Using angleresolved photoemission spectroscopy (ARPES) of surface states, 50 meV gap formation was detected in paramagnetic Fe-doped bulk Bi<sub>2</sub>Se<sub>3</sub>; it was assigned to breakage of the time-reversal symmetry by exchange interactions [36]. Effects of magnetic impurities upon topological surface states were also studied by *in-situ* deposition of Fe atoms [37-39]. It was found that Fe acts as donor [37,38] if deposited at room temperature but as an acceptor if deposited at 8 K [38]. A question on whether the presence of a Fe surface layer opens a gap or not in the topological states was also examined experimentally and theoretically [37-39].

According to one theoretical proposal, Fe-doped Bi<sub>2</sub>Se<sub>3</sub>-based heterostructures are a preferred platform for observing the emergence of QAH phase [17]. Nevertheless, no QAH effect has yet been observed in magnetically doped Bi<sub>2</sub>Se<sub>3</sub>-based films, probably because of the absence or weakness of the ferromagnetic ordering, and associated difficulties in tuning of the  $E_F$  into a correspondingly small exchange gap [40]. In contrast, a TPT from the TI to the 2D-insulator by film thickness reduction was demonstrated for Bi<sub>2</sub>Se<sub>3</sub> films using ARPES and electrical measurements [4,31-33]. Depending on the experimental method used, the critical thickness for the transition has been reported as 5 [4] or 10 nm [41] in Bi<sub>2</sub>Se<sub>3</sub>. For other 3D-TIs such as Bi<sub>2</sub>Te<sub>3</sub>, (Bi,Sb)<sub>2</sub>Te<sub>3</sub> (BST) and magnetically doped BST films, variations in the strength of the spin-orbit interaction are thought to account for critical thickness changes from 1 to 13 nm [12,42].

#### **II. EXPERIMENTAL DETIALS**

Employing molecular-beam epitaxy (MBE) and a 3-nm-thick *n*-type  $Bi_2Se_3$ buffer layer on a semi-insulating Fe-doped InP (111) substrate, we have grown  $Fe_x(Bi_1-ySb_y)_{2-x}Se_3$  films of thickness *d* ranging from 8 to 30 nm (see Supplementary section 1 [43]). The control parameters are the Sb composition and the gate bias voltage for changing the  $E_{\rm F}$  position [44,45], whereas the Fe concentration and the external magnetic field serve for tuning magnetic disorder and band splitting state. Because Fe doping up to at least x = 0.1 (nominal composition controlled by the beam flux ratio) produces rather minor changes in  $R_{xx}$  and  $|R_{yx}|$  magnitudes (see Supplementary section 2 [43]), we infer that Fe ions are isoelectronic impurities occupying Bi or Sb sites, and assume the same Fe<sup>3+</sup> charge configuration with bulk Fe-doped Bi<sub>2</sub>Se<sub>3</sub> [36] that typically corresponds to the high spin S = 5/2. However, Fe acts not only as a magnetic impurity, but also as an isoelectronic dopant, in a manner similar to Sb, which affects the carrier concentration owing to band shifts with respect to defect formation levels. Moreover, thickness d plays a crucially important role in the control of both (i) hybridization between surface and interfacial topological states and (ii) the width of the depletion layer in the *p*-*n* junction that forms at the interface to the *n*-type Bi<sub>2</sub>Se<sub>3</sub> buffer layer.

#### **III. RESULT AND DISCUSSION**

#### A. Angle-resolved photoemission spectroscopy (ARPES) and magnetic properties

We have characterized the electronic structure of a 30-nm-thick  $Fe_{0.05}(Bi_{0.34}Sb_{0.66})_{1.95}Se_3/Bi_2Se_3$  heterostructure at 40 K using ARPES. As presented in Fig. 1(g), the topological surface states and the bulk valence band are resolved clearly at the  $\Gamma$  point, indicating that this thick Fe-doped (Bi,Sb)<sub>2</sub>Se<sub>3</sub> layer preserves key features of 3D-TIs and does not engender a substantial Fermi level shift. Linear dispersion of the surface band is more readily apparent when moving the Fermi level up in energy by an aging process that increases the surface electron concentration (see Fig. S3 [43]). Contrary to an earlier report on ARPES at 10 K of bulk Fe-doped Bi<sub>2</sub>Se<sub>3</sub> samples with Fe concentration x = 0.12 and 0.16, which was considered as a consequence of ferromagnetic ordering [36], no sizable gap in the surface states is resolved in the case of our MBE-grown layer with x = 0.05; this indicates the absence of a long range magnetic order at Fe concentration of interest here.

The ARPES data are consistent with the magnetotrasport studies: neither hysteretic nor saturation behavior has been observed in our Hall effect measurements, indicating that Fe ions remain in a paramagnetic phase down to 2 K for x = 0.05. This is in agreement with previous direct measurements of magnetization M(T,H) down to 2 K and up to  $\mu_0 H = 5$  T on bulk samples that showed paramagnetic behavior below x = 0.16[36]. Some hysteresis superimposed on the paramagnetic signal M(H) were detected for  $x \ge 0.16$  at 2 K [36]. The absence of ferromagnetism at low Fe concentrations is consistent with our direct magnetization measurements employing superconducting quantum interference device (SQUID), which do not show any hysteresis. However, a quantitative evaluation of the epilayer paramagnetic signal from our SQUID and electron paramagnetic resonance studies has been hampered by a small thickness of the epilayers and a large magnetic contribution of the Fe-doped InP substrate. The absence of ferromagnetism in the Fe-doped Bi<sub>2</sub>Se<sub>3</sub> is also consistent with *ab initio* studies [46,47].

#### **B.** Magnetotransport properties

The values of the 2D resistivity tensor components, *i.e.*, the sheet resistance  $R_{xx}$ and the Hall resistance  $R_{yx}$ , have been measured in the Hall-bar geometry and using a standard lock-in technique [43]. Figure 1(h) presents a contour plot of  $R_{xx}$  values for  $Fe_{0.05}(Bi_{0.34}Sb_{0.66})_{1.95}Se_3/Bi_2Se_3$  films in B = 0 as a function of layer thickness d and temperature T (see Supplementary Fig. S4(a) for detailed dataset [43]). One phenomenological criterion describing the critical point of the localization transition in the 2D case is the sheet resistance value of  $h/e^2 \approx 25.8 \text{ k}\Omega$  [48,49]. The boundary is pronounced as a white color region in Fig. 1(h). As shown there,  $R_{xx}(T)$  determined for the 30-nm-thick film shows lower values than that of  $h/e^2$ , in addition to weak temperature dependence, implying that this sample is the 3D-TI phase (Fig. 1(g)). With decreasing d at fixed Fe and Sb concentrations (x = 0.05 and y = 0.66),  $R_{xx}(T)$  reaches the critical value of  $h/e^2$  at thickness of d = 14 nm, below which dependence  $R_{xx}(T)$  exhibits insulating behavior and  $R_{xx}$  magnitudes exceed 1 M $\Omega$  at the lowest temperature of 2 K (deep-red region in Fig. 1(h)). Such highly insulating features at d < 14 nm can be ascribed either to the trivial Anderson insulator [50,51] or to the quantum spin Hall insulator [51,52], if the formation of edge states would accompany hybridization of surface states. However, edge conductance would hardly dominate the charge transport in the present Hall-bar geometry with the channels longer than the expected protection length of helical topological channels. The phase coherence length estimated by weak antilocalization analysis of the Bi<sub>2</sub>Se<sub>3</sub> thin films is about 200 nm [45], which is well below the channel length. In principle, ARPES is an effective way to tell the topological phase. However, the high resistance and the corresponding charging effect made it difficult to assess the electronic structure of our thinnest samples. Nevertheless, the critical thickness we found for this 2D-insulator is rather thick compared to the values (5-10 nm) reported for nonmagnetic Bi<sub>2</sub>Se<sub>3</sub> films [4,41]. This point can be explained by the empirical fact (see Supplementary Section 5 [43]) that the substitution of heavier Bi or Sb by lighter Fe reduces the spin-orbit interaction strength, which might increase the critical thickness. Furthermore, the presence of the *n*-type Bi<sub>2</sub>Se<sub>3</sub> buffer layer depletes the *p*-type Fe<sub>0.05</sub>(Bi<sub>0.34</sub>Sb<sub>0.66</sub>)<sub>1.95</sub>Se<sub>3</sub> layer [45], making the effective thickness less than the nominal value.

More importantly, in our paramagnetic samples, spin disorder scattering by Fe

impurities can be effective for expanding the Anderson insulator range with regard to the thickness and temperature of Fe<sub>x</sub>(Bi<sub>1-y</sub>Sb<sub>y</sub>)<sub>2-x</sub>Se<sub>3</sub> films (Fig. 1(h)), making it wider compared to Bi<sub>2</sub>Se<sub>3</sub>. Temperature dependence of resistivity for samples with d = 14 nm and various x are presented in Fig. S2 [43]. The data show clearly a transition from the weakly localized regime to the strongly localized regime, characterized by a sharp increase of resistivity on lowering temperature, which appears at x > 0.04. As expected in the absence of Landau quantization and in the 2D case [53], this transition has a crossover character, so that it does not obey scaling equations. As presented in Fig. 1(i), the magnitude of magnetoresistance defined as MR =  $[R_{xx}(B) - R_{xx}(0)]/R_{xx}(0)$  exceeds -80% in 9 T and at 2 K in the vicinity of the localization transition. The ordering of Fe spins in the magnetic field reduces magnetic disorder and enhances the spin splitting of electronic states. Consequently, a QAH phase would appear in the thickness region toward type (3) by the application of the magnetic field as schematically shown in Figs. 1(c) and 1(f). In accordance with the expectation that reduction of magnetic disorder occurs for any field direction, we find negative MR to be also present for the in-plane magnetic field (Supplementary Fig. S6 [43]).

Figure 2 presents a dependence of Hall resistance  $R_{yx}$  on the Sb concentration y at T = 2 K and B = 9 T for Fe<sub>0.05</sub>(Bi<sub>1-y</sub>Sb<sub>y</sub>)<sub>1.95</sub>Se<sub>3</sub>/Bi<sub>2</sub>Se<sub>3</sub> with  $d \approx 14$  nm (red circles). This dependence is strikingly different compared to the case of non-magnetic (Bi1- $_{y}$ Sb<sub>y</sub>)<sub>2</sub>Se<sub>3</sub>/Bi<sub>2</sub>Se<sub>3</sub> films with d = 14-20 nm (black squares) [44]. The representative raw data of  $R_{yx}(B)$  for Fe-doped samples are shown in the inset. For non-magnetic (Bi<sub>1-</sub>  $_{y}$ Sb<sub>y</sub>)<sub>2</sub>Se<sub>3</sub>/Bi<sub>2</sub>Se<sub>3</sub>, the sign reversal of  $R_{yx}$  reflects the conversion of the carrier type and therefore the tuning of  $E_{\rm F}$  across the charge neutrality point (CNP), which appears at  $y \cong$ 0.7. In contrast for the Fe-doped films, the positive value of  $R_{yx}$  persists in y > 0.5, indicating that the positive contribution of magnetic-field-induced anomalous Hall resistance is much larger than the negative contribution of ordinary Hall component; empirically  $R_{yx} = R_0 B + R_A M_z$  where  $R_0$  is the ordinary Hall coefficient, B magnetic field,  $R_{\rm A}$  the anomalous Hall coefficient, and  $M_{\rm z}$  out-of-plane component of magnetization. By application of a perpendicular B, B-induced  $M_z$  dominantly contributes to the  $R_{yx}$  with negligibly small ordinary component. Moreover,  $R_{yx}$  is enhanced greatly with a peak at y  $\approx 0.67$ , close to the CNP in non-magnetic films. The behavior of  $R_{yx}$  as a function of y, together with a large magnitude of  $R_{yx}$  at the CNP, demonstrates that  $R_{yx}$  is dominated by the intrinsic anomalous Hall effect, as its amplitude is expected to be dependent on the  $E_{\rm F}$ position while its sign is determined by magnetization direction and exchange coupling [17,19,54]. In fact, the tangent of Hall angle  $R_{yx}/R_{xx}$  approaches unity at values greater than d = 14 nm (see Supplementary Fig. S4(f) [43]) indicating that the condition for the Landau quantization of the density of states,  $R_0B/R_{xx} > 1$ , is not met. It should be noted that such a large  $R_{yx}$  at d = 14 nm is observed also in the insulator region (Fig. 1(h)), where negative MR exists (Fig. 1(i)).

Together with revealing enhanced values of  $R_{yx}(B)$ , transition behavior is found in  $R_{xx}(T)$  in elevated fields up to 24 T, as depicted in Fig. 3(a). The field-induced insulatormetal transition becomes apparent below 50 K. For B > 12 T, the metallicity increases concomitantly with increasing B down to temperatures as low as 1.6 K. The criterion in B = 9 T for the sign change of  $dR_{xx}/dT$  being close to 0.5  $h/e^2$ . This value for the MIT, compared to 1  $h/e^2$  for MIT driven by structural modifications (cf. Fig. 1(h)), indicates one other type of the TPT. We attributed this field-induced metallization to a TPT from the 2D-insulator to a QAH phase. The insulator at B = 0 with hybridization gap  $\Delta_{hy}$  (Fig. 1(a) and 1(d)) turns to be a state with inverted bands by Zeeman splitting  $\Delta_{\text{Zeeman}}$  when  $\Delta_{\text{Zeeman}} > \Delta_{\text{hy}}$ . With the assistance of spin-orbit coupling, a gap is formed at the crossings of surface bands hosting carriers with opposite spin orientations [17,30], as depicted in Fig. 1(c) and 1(f). Furthermore, at  $\Delta_{\text{Zeeman}} > \Delta_{\text{hy}}$ , gapless disorder-protected chiral channels are formed, in a full analogy to the theoretical proposal for the QAH effect [17].

Having observed the signature of this new TPT, we study  $R_{xx}(B)$  and  $R_{yx}(B)$  at T= 1.6 K employing a field-effect transistor (FET) device for precise tuning of  $E_F$ . The FET layout is depicted in the inset shown with Fig. 3(b) (see also Supplementary Materials [43]) - it consists of a 1-nm-thick Bi<sub>2</sub>Se<sub>3</sub>/18.5-nm-thick Fe<sub>0.05</sub>(Bi<sub>0.33</sub>Sb<sub>0.67</sub>)<sub>1.95</sub>Se<sub>3</sub>/3-nmthick Bi<sub>2</sub>Se<sub>3</sub> trilayer structure. As depicted in Fig. 3(b), by sweeping magnetic field up to 15 T under  $V_{\rm G} = -60$  V, the large  $R_{\rm xx}$  value of 3.92  $h/e^2$  in B = 0 (black line) decreases monotonically to 0.2  $h/e^2$  whereas  $R_{yx}$  (red line) increases, reaching 0.975  $h/e^2$ , i.e., almost the value expected for the ideal QAH phase,  $h/e^2$ . Consequently, the magnetic-fieldinduced transition from the 2D-insulator to the QAH phase can be observed clearly at the high temperature of T = 1.6 K, at which the quantization accuracy of the  $R_{yx}$  value is comparable to that reported for Cr modulation doped (Bi,Sb)<sub>2</sub>Te<sub>3</sub> heterostructures [15,55] and higher than that for Cr-doped or V-doped (Bi,Sb)<sub>2</sub>Te<sub>3</sub> thin films [8,24-28]. Note that this quantization is hardly considered to be QHE due to insulating initial state at B = 0with a negligible ordinary Hall effect. The electrostatic tuning of the  $E_{\rm F}$  position with respect to the gap around the CNP promotes other TPTs as presented in conductivity tensor in Supplementary Fig. S10 [43].

The nature of relevant phases is assessed by examining the renormalization group flow of the conductivity tensor components [ $\sigma_{xx} = R_{xx}/(R_{xx}^2 + R_{yx}^2)$ ,  $\sigma_{xy} = R_{yx}/(R_{xx}^2 + R_{yx}^2)$ ] as a function of temperature [8,25,56]. Figures 4(a) and 4(b) show  $\sigma_{xx}(T)$  and  $\sigma_{xy}(T)$  for various *B* at  $V_G = -60$  V. As *T* decreases, all  $\sigma_{xx}(T)$  values decrease, irrespective of B. However,  $\sigma_{xy}(T)$  goes to  $e^{2/h}$  and to zero, respectively, in high and low fields, with a crossover point of about 0.5  $e^{2/h}$ . These data are shown in the  $[\sigma_{xy}(B), \sigma_{xx}(B)]$  plane in Fig. 4(c). By lowering T from 15 K (open symbols) to 1.6 K (close symbols) under various magnetic fields, the data converge to either  $(\sigma_{xx}, \sigma_{xy}) = (0, 0)$  for the insulator or  $(0, e^2/h)$ for QAH phase. The finding of a converging point in our Fe-doped (Bi,Sb)<sub>2</sub>Se<sub>3</sub> samples clearly evidences the presence of a phase transition driven by the magnetic field, with the critical field of 7–8 T corresponding to the point at which  $\Delta_{\text{Zeeman}}$  becomes comparable to  $\Delta_{\rm hv}$ . Moreover, as shown in Supplementary Fig. S10 [43], the transition driven by an electric field in fixed magnetic fields of 9 and 12 T is visible. Given the experimental observation of the TPT in the accessible magnetic field, the large g-factor of the surface state should be taken into consideration to estimate  $\Delta_{\text{Zeeman}}$ , as discussed in Supplementary Section 9 [43]. The Zeeman splitting (g-factor of Bi<sub>2</sub>Se<sub>3</sub> ~ 18-50 [57,58]) can be enhanced considerably over the band value  $g^* \mu_{\rm B} B$  through the *sp-d* exchange interaction between itinerant carriers and localized magnetic moments, as observed in various dilute magnetic semiconductors, primarily doped with Mn but also with  $Fe^{3+}$  ions [59]. By applying gfactor engineering, the TPT might be shifted to much weaker magnetic fields and higher temperatures. The renormalization group flow for type (3) in paramagnetic (Bi,Sb)<sub>2</sub>Se<sub>3</sub> film (Figs. 1(c) and 1(f)) can be understood in terms of universal QAH phenomenon as previously discussed in type (1) in ferromagnetic (Bi,Sb)<sub>2</sub>Te<sub>3</sub> films [8,25,56].

#### **IV.** Conclusion

We have performed comprehensive study of topological phase transitions based on Fe-doped (Bi,Sb)<sub>2</sub>Se<sub>3</sub> thin films, which are driven by structural modification, doping, and external magnetic and electric fields. We found that topological phase transition occurs from 3D-TI to 2D-insulator by Fe doping owing to localization by magnetic disorder. The 2D-insulator is turned to QAH state by external magnetic field owing to a large Zeeman splitting and suppression of the magnetic disorder. Demonstration of the magnetic-field-induced TPT toward the QAH state in Bi<sub>2</sub>Se<sub>3</sub>-based 3D-TI sheds light on new perspectives of the QAH physics and expands it to a broader class of paramagnetic TI materials with properties that can be controlled by *g*-factor engineering.

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#### **Figure legends**

FIG. 1 Schematic and calculated electronic band structure. (a-c) Evolution of surface spin subband structure in paramagnetic Fe-doped (Bi,Sb)<sub>2</sub>Se<sub>3</sub> thin films upon thickness reduction and external magnetic field application. (a) Surface states of spin-up (magenta) and spin-down (cyan) subbands with gap  $\Delta_{hy}$  driven by inter-surface hybridization, leading to 2D insulator phase. (b) Under an external perpendicular magnetic field, the spin subband degeneracy is lifted by the Zeeman effect, producing band crossing when the Zeeman energy  $\Delta_{\text{Zeeman}}$  exceeds  $\Delta_{\text{hy.}}$  (c) At crossings of spin-up (magenta) and spindown (cyan) bands, a gap is generated by the spin-orbit interaction, leaving a onedimensional chiral edge channel in the bulk bandgap (red solid line). (d-f) Calculated band structure of three-dimensional topological insulator thin films. The band structure is calculated using the 3D-TI slab for cases in which (d)  $\Delta_{hy}$  is larger than  $\Delta_{Zeeman}$ , (e)  $\Delta_{hy}$ is comparable to  $\Delta_{\text{Zeeman}}$ , and (f)  $\Delta_{\text{Zeeman}}$  is larger than  $\Delta_{\text{hy.}}$  (g) ARPES intensity of the 30nm-thick Fe<sub>0.05</sub>(Bi<sub>0.34</sub>Sb<sub>0.66</sub>)<sub>1.95</sub>Se<sub>3</sub>/3-nm-thick Bi<sub>2</sub>Se<sub>3</sub> film around the  $\overline{\Gamma}$  point measured at T = 40 K. SS and VB respectively represent surface state and bulk valence band.  $E_F$  is dashed line. (h) Longitudinal indicated by white resistance  $R_{\rm xx}$ а of  $Fe_{0.05}(Bi_{0.34}Sb_{0.66})_{1.95}Se_3/Bi_2Se_3$  at B = 0 as a function of the magnetic layer thickness d and temperature T. Colour bar scale is in the units of  $h/e^2$ . (i) Magnetoresistance of  $Fe_{0.05}(Bi_{0.34}Sb_{0.66})_{1.95}Se_3/Bi_2Se_3$  at 2 K as a function of d and B. Colour scale bar corresponds to +20 (blue) to -100 (red) %.

FIG. 2 Hall resistance of Fe-doped and non-magnetic heterostructures against Sb composition *y* at 9 T and 2 K. Hall resistances  $R_{yx}$  of 14-nm-thick Fe<sub>x</sub>(Bi<sub>1-y</sub>Sb<sub>y</sub>)<sub>2-x</sub>Se<sub>3</sub>/3-nm-thick Bi<sub>2</sub>Se<sub>3</sub> (red circles) dominated by the anomalous component and of 14–20-nm-thick non-magnetic (Bi<sub>1-y</sub>Sb<sub>y</sub>)<sub>2</sub>Se<sub>3</sub>/3-nm-thick Bi<sub>2</sub>Se<sub>3</sub> films (black squares) are shown against Sb content *y*. CNP denotes the charge neutral point of non-magnetic samples. The inset shows the Hall effect measurement for Fe<sub>x</sub>(Bi<sub>0.33</sub>Sb<sub>0.67</sub>)<sub>2-x</sub>Se<sub>3</sub>/3-nm-thick Bi<sub>2</sub>Se<sub>3</sub> films for Fe content *x* = 0 (solid black line), 0.04 (orange), and 0.05 (red).

FIG. 3 Magnetic-field-induced insulator-metal transition and quantum anomalous Hall effect. (a) Longitudinal sheet resistance  $R_{xx}(T)$  of 14-nm-thick  $Fe_{0.05}(Bi_{0.33}Sb_{0.67})_{1.95}Se_3/Bi_2Se_3$  heterostructure (inset) in various magnetic fields *B*. (b) Hall and sheet resistances,  $R_{yx}(B)$  (solid red line) and  $R_{xx}(B)$  (solid black line), measured for a FET device (shown in the inset) at  $V_G = -60$  V and T = 1.6 K. The  $R_{yx}$  data below 1 T were not obtained owing to the large value of  $R_{xx}$ . FIG. 4 Magnetic-field-induced insulator-QAH phase transition. (a,b) Temperature dependences of the longitudinal and Hall conductivities ( $\sigma_{xx}$  and  $\sigma_{xy}$ ) at  $V_G = -60$  V for various magnetic fields *B*. (c) Renormalization group flow in the [ $\sigma_{xx}(T)$ ,  $\sigma_{xy}(T)$ ] plane in various *B* from 3 to 15 T. Each curve is extracted from data in (a) and (b) employing the same colour code. Empty and filled circles respectively present data obtained at T = 15 and 1.6 K. The [ $\sigma_{xx}$ ,  $\sigma_{xy}$ ] flow extracted from the data as a function of the magnetic field at T = 1.6 K is also shown (solid black line).



Figure 1 Y. Satake et al.



Figure 2 Y. Satake et al.



Figure 3 Y. Satake et al.



Figure 4 Y. Satake et al.