Fermi-level-dependent charge-to-spin current conversion by Dirac surface states of topological insulators

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1. Charge current and Oersted field distribution in BST/Cu/Py tri-layer film

**Figure S1 | Calculated current density and Oersted field distribution by using a finite element method (FEM).** a, b, 8-nm (Bi$_x$Sb$_{1-x}$)$_2$Te$_3$ (BST)/8-nm Cu/10-nm Py tri-layer structure discretized into mesh grids with assuming a 1-nm-thick conductive surface layer of TI. Calculated sample size is 1 × 3 $\mu$m$^2$. The applied rf power is 13 $\mu$W. c, Color code and yellow arrows respectively show the charge current density and resulting Oersted field distribution.

We calculated the charge current density and Oersted field by using following setting in finite element method (FEM) with COMSOL Multiphysics simulator [ref. S1]. Mesh grids consist of triangular segments about 10-nm on a side. Input rf frequency and power is 7 GHz, 13 $\mu$W ($\approx$ 25 mV), respectively. The setting rf power is 20 times smaller than actual input power, because calculated sample area is 20 times smaller than actual sample size. Resistivity of Py and Cu are 60 and 10 $\mu\Omega$cm, respectively.

From this analysis, it is concluded that the most of charge currents flow in Cu layer in this tri-layer structure as shown in Fig. S1c. Oersted fields originating from the charge currents in Cu and surface state of BST are given by the relation $H_{\text{rf}} = (J_{C}^\text{Cu} t_{\text{Cu}} + J_{C}^\text{surf})/2$, where $J_{C}$ and $t$ is charge current density and layer thickness.
However in an actual system, there is a Py layer on Cu/BST films, causing a slight change in $H_{rf}$ such that the peak of charge current density shifts toward the Py layer. In order to evaluate the influence of Py layer, we estimate the reduction factor $\xi$ as the ratio $H_{rf}^{\text{FEM}} / (J_{C}^{Cu} + j_c)/2$, yielding $\xi = 0.96$.

We estimated $j_c$ with an assumption of the width of surface conducting layer as 1-nm for bulk insulating BST. In addition, for bulk conducting condition, we calculated $j_c$ with 6-nm bulky conducting layer and 1-nm top and bottom surface layer assuming the same conductivity for both layers. Then, $q_{ICS}$ and $\sigma_S$ for bulk conducting Bi$_2$Te$_3$ and Sb$_2$Te$_3$ shown in Fig. 3 in the main text were characterized with $j_c$, while both $q_{ICS}$ and $\sigma_S$ strongly depend on the above two assumptions, i.e., surface thickness and conductivity relationship between bulk and surface layers. In contrast, the estimate for bulk insulating condition is quite robust; the surface thickness only affects the Oersted field made by the TI layer, which hardly modifies the reduction factor $\xi$ and hence $q_{ICS}$. The estimated charge current density is summarized in Table S1 in Section 2.

### 2. Charge current distribution dependence of $q_{ICS}$

![Figure S2](image)

**Figure S2** | Sb composition dependence of $q_{ICS}$ in various bulk contributions in charge current.
Figure S2 shows the Sb composition (x) dependence of interfacial charge-to-spin current conversion efficiency $q_{ICS}$ in case of various bulk contributions in charge current. These values were estimated for different current distributions in bulk and surface states (see Table S1 below). We found that the value of $q_{ICS}$ increases with increasing the bulk contribution, because $q_{ICS}$ is defined as $q_{ICS} = J_S^\text{Py}/J_C$ in the main text page8. Thus it can be seen clearly that the estimated values in case of 1.00:0.00 are the lower limit. Additionally, the error bar does not affect our conclusion even in 0.5:0.5 and 0.25:0.75 by our calculating conditions. Therefore, when $E_F$ is located in the bulk band gap $x = 0.5, 0.7$ and 0.9, we can discuss the essence of $q_{ICS}$ without taking into account the bulk contribution.

**TABLE S1. Charge current density (A/m$^2$) of each layer deduced from the FEM calculation.** Surface charge current density $J_C$ (A/m) is calculated as $J_C^\text{TIsurf} \times 1$-nm.

<table>
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<tr>
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<th>$J_C^\text{Py}$ ($10^9$ A/m$^2$)</th>
<th>$J_C^\text{Cu}$ ($10^9$ A/m$^2$)</th>
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### Supplementary Information

**b.** $\frac{I_{\text{bulk}}}{(I_{\text{surf}}+I_{\text{bulk}})} = 0.05$ (5%):

<table>
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<th>Sb (x)</th>
<th>$J_{C}^{\text{Py}}$ (10^9 A/m^2)</th>
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**c.** $\frac{I_{\text{bulk}}}{(I_{\text{surf}}+I_{\text{bulk}})} = 0.10$ (10%):

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**d.** $\frac{I_{\text{bulk}}}{(I_{\text{surf}}+I_{\text{bulk}})} = 0.50$ (50%):

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**Figure S3** | a, Hall effect measurements for seven BST films at 10  K. b, Sb composition dependence of sheet resistance $R_{\square}$ at 10 K.
Supplementary information

<table>
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<tr>
<th>Sb (x)</th>
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3. Electrical transport properties of BST films

Figure S3 | a, Hall effect measurements for seven BST films at 10 K. b, Sb composition dependence of sheet resistance $R_{\parallel}$ at 10 K.
In Fig. S3a, Hall resistance $R_{yx}$ for BST films measured at 10 K are displayed as a function of perpendicular magnetic field. For BST films with $0 \leq x \leq 0.82$, $R_{yx}$ takes negative values, indicating $n$-type conduction. The slope of Hall resistance as a function of applied magnetic field increases with $x$. On the other hands, for BST films with $0.88 \leq x \leq 1.0$, the $R_{yx}$ are positive, indicating $p$-type conduction. Thus, BST film with $x = 0.88$ have their Fermi levels close to Dirac point. Fig. S3b shows sheet resistance of BST films as a function of Sb composition.

**4. Bulk contribution to spin accumulation in surface state of TI**

![Energy dispersion and Fermi circles of Dirac surface states and bulk bands for bulk conductive $n$-type TI.](image)

Energy dispersion of TI is schematically drawn in Fig. S4a. Gray plane is depicted as Fermi energy buried into the bulk conduction band inside the Dirac surface bands, corresponding to bulk conductive $n$-type condition. In the case of...
bulk conductive TIs, charge currents flow not only along surface channels but also along bulk conduction channels. If bulk channels exhibit Rashba spin splitting like Bi$_2$Se$_3$ (ref. 1), the bulk contribution with opposite spin polarization may cancel the part of surface spin accumulation. Here we consider the contribution of charge current in bulk conduction channels to spin accumulation as shown in Figs. S4b and S3c. Note that, in bulk insulating TI, the spin accumulation is generated only by the shift of Fermi circle with spin momentum locking shown in Fig. 2e in the main text.

Fig. S4b and S4c represent the spin state on Fermi circles without and with electric field, respectively. When electric field is applied to -x direction, the down spin density $\langle \delta S_0 \rangle^D$ (red area) increases in the Dirac surface state. At the same time, the up spin density $\langle \delta S_0 \rangle^R$ (blue area) also increases in the bulk band. Amount of the total spin accumulation in the conduction channels of bulk conductive TI is thus expressed as $\langle \delta S_0 \rangle = \langle \delta S_0 \rangle^D \pm \langle \delta S_0 \rangle^R = q_{ICS}^D j_C^D \pm q_{ICS}^R j_C^R = -\hbar/2e \{ j_C^D/\nu_F \pm (m \alpha_\gamma^R) j_C^R/(2E_F) \}$, where $\alpha_\gamma$ is Rashba parameter in the bulk band. When spin polarization in bulk state with Rashba splitting is opposite direction with that of surface state, $q_{ICS}$ in bulk conductive TI can be expressed as

$$q_{ICS} = -\hbar D/2e \{ j_C^D/\nu_F \pm (m \alpha_\gamma^R) j_C^R/(2E_F) \}/(j_C^D + j_C^R).$$

The $2E_F/m \alpha_\gamma^R$ can be estimated $1.5 \times 10^5$ m/s using typical values of $E_F = 0.3$ eV [ref. S3] and $m \alpha_\gamma^R = 0.1$ Å$^{-1}$ [ref. S4]. It indicates that the influence of spin accumulation in bulk state cannot be negligible, because the value of $2E_F/m \alpha_\gamma^R$ is the same order with $\nu_F = 3\sim4 \times 10^5$ m/s. On the other hand, when $q_{ICS}^R$ is the same sign and magnitude with $q_{ICS}^D$, $q_{ICS}$ in the whole tri-layer devices can be expressed as

$$q_{ICS} = -\hbar D/2e \{ q_{ICS}^D j_C^D + q_{ICS}^R j_C^R \}/(j_C^D + j_C^R) \approx -\hbar D/2e \nu_F$$

whose formula is same with that of bulk insulating TI. Therefore the high $q_{ICS}$ even in bulk conductive TI can be obtained.
5. Power dependence of symmetric voltage $V_{\text{Sym}}$ in ST-FMR measurement

![Figure S5](image)

**Figure S5** | a, Microwave power dependence of $V_{\text{Sym}}$ b, Measurement temperature dependence of resonance field shift $\Delta H_r$ c, Microwave power dependence of resonance shift $\Delta H_r$.

Figure S5a shows input microwave power dependence of $V_{\text{Sym}}$ in BST with $x = 0.70$. Sample size is $20 \times 60$ μm. $V_{\text{Sym}}$ is proportional to microwave power. In our ST-FMR measurement, the input power is 8 mW in the linear response region.

In order to evaluate heating effect due to input microwave, we measured the relationship between measurement temperature and resonance field shift $\Delta H_r$ under FMR with $P = 8$ mW as shown in Fig. S4b. The resonance field rises at a rate of 0.049 Oe/K, because the saturation magnetization of Py decreases with increasing the sample temperature.

Figure S5c shows microwave power dependence of resonance field shift at 10 K. The applied rf current power of 8 mW gives rise to $\Delta H_r = 1.7$ Oe; this shift corresponds to the increase in sample temperature of about 35 K estimated from Fig. S5b. Since resistivity of BST weakly depends on temperature below 100 K as shown in Fig. S10 in Section 10, the heating effect of 35 K is rather minor in $V_{\text{Sym}}$, resulting in the above-mentioned linearity between $V_{\text{Sym}}$ and microwave power. The evaluated $V_{\text{Sym}}$ with $P = 8$ mW at $T = 10$ K is therefore valid in our discussion.

6. Magnetic field angle dependence of anti-symmetric voltage $V_{\text{Anti}}$

![Figure S6](image)

**Figure S6** | Magnetic field angle ($\theta$) dependence of $V_{\text{Anti}}$ for BST with $x = 0.70$. Broken line indicates fitting curve with $\sin^2 \theta \cos \theta$.

Figure S6 shows magnetic field angle dependence of $V_{\text{Anti}}$. The angle dependence could be well fitted by $\sin^2 \theta \cos \theta$, indicating that the $V_{\text{Anti}}$ comes purely from the Oersted field. This enables us to precisely estimate $q_{ICS}$.

7. Heating effect due to dc current application

![Figure S7](image)

**Figure S7** | dc current dependence of resonance field measured at 10 K.

In FM/ spin Hall nonmagnetic material (SH-NM) bilayer film, the effective damping constant can be modulated by dc current [ref. S5]. By measuring the effective damping constant at 10 K, we can estimate the heating effect due to dc current application.
6. Magnetic field angle dependence of anti-symmetric voltage $V^{\text{Anti}}$

Figure S6 | Magnetic field angle ($\theta$) dependence of $V^{\text{Anti}}$ for BST with $x = 0.70$. Broken line indicates fitting curve with $\sin 2\theta \cos \theta$.

Figure S6 shows magnetic field angle dependence of $V^{\text{Anti}}$. The angle dependence could be well fitted by $\sin 2\theta \cos \theta$, indicating that the $V^{\text{Anti}}$ comes purely from the Oersted field. This enables us to precisely estimate $q_{\text{ICS}}$.

7. Heating effect due to dc current application

Figure S7 | dc current dependence of resonance field measured at 10 K.

In FM/spin Hall nonmagnetic material (SH-NM) bilayer film, the effective damping constant can be modulated by dc current [ref. S5]. By measuring the
damping modulation, we can estimate the spin Hall angle of SH-NM. In our sample, however, the most part of charge current flows into the inserted Cu layer and hence it is practically impossible to detect the modulation within a reasonable range of the dc current as discussed below.

Figure S7 shows applied dc current dependence of resonance field. Input rf current frequency is 8 GHz. Red and Blue plots correspond to data with magnetic field in the direction of 45 and -135 degrees, respectively. The fitting curves of these plots consist of linear and parabolic components; the linear components are caused by Oersted field due to dc current, while the parabolic ones may be due to heating effect. The change rate of parabolic curve was about 0.06 Oe/(mA)^2. When the surface conductive layer thickness and \( q_{ICS} \) are assumed 1 nm and 0.4 nm\(^{-1}\), we have to apply the large dc current of about 25 mA to realize the detectable change of half width (~ 0.5 Oe). Considering that the application of such a large dc current will cause the sample heating of about 700 K, it is very difficult to evaluate the \( q_{ICS} \) by using damping modulation measurement.

Figure S8a shows the frequency dependence of half width at half maximum \( \Delta \) at 10 K. From this data, we estimated the effective damping constant \( \alpha_{eff} \) and the spin mixing conductance \[\text{ref. S6}\] for all the samples, as displayed in Fig. S8b. Here we assumed the followings: the surface conductive layer thickness of 1 nm, the spin diffusion length of 1 nm, and reciprocity between the spin-to-charge current conversion efficiency and the charge-to-spin current one. Then we estimated \( \alpha_{eff} \) and \( \alpha_{SP} \) and \( V_{Sym} \) and \( V_{SP} \). d, Sb composition dependence of interfacial charge to spin current conversion efficiency \( q_{ICS} \). Blue plots are \( q_{ICS} \) obtained by removing the influence of spin pumping.
8. Estimation of influence of spin pumping effect

**Figure S8** | a, Input rf frequency dependence of half width at half maximum $\Delta$. b, Sb composition dependence of effective damping constant $\alpha_{\text{eff}}$ (left axis) and spin mixing conductance (right axis). c, Sb composition dependence of $V_{\text{sym}}$ and $V_{\text{SP}}$. d, Sb composition dependence of interfacial charge to spin current conversion efficiency $q_{\text{ICS}}$. Blue plots are $q_{\text{ICS}}$ obtained by removing the influence of spin pumping.

Figure S8a shows the frequency dependence of half width at half maximum $\Delta$ at 10 K. From this data, we estimated the effective damping constant $\alpha_{\text{eff}}$ and the spin mixing conductance $g_{\text{eff}}^{11}$ [ref. S6] for all the samples, as displayed in Fig. S8b. Here we assumed the followings: the surface conductive layer thickness of 1 nm, the spin diffusion length of 1 nm, and reciprocity between the spin-to-charge current conversion efficiency and the charge-to-spin current one. Then we estimated...
the output voltage originating from spin pumping $V^{SP}$. Figure S8c shows the Sb composition dependence of $V^{Sym}$ and $V^{SP}$, indicating that the $V^{SP}$ is about 10-25% in total output voltage ($V^{Sym}$). Figure S8d shows the interfacial charge to spin current conversion efficiency $q_{ICS}$ with and without contribution of spin pump effect $V^{SP}$. We found that the estimation of $q_{ICS}$ is only slightly affected by the spin pumping contribution.

9. Charge to spin current conversion efficiency $J_S/J_C$ obtained by considering the three-dimensional carriers

![Graphs](image)

Figure S9 | Sb composition dependence of (a) antisymmetric voltages, (b) symmetric voltages, (c) charge current density in TI film and (d) charge to spin current conversion efficiency.

Figures S9 summarizes experimental data set in our experiments by ST-FMR. In addition to the discussion of C-S conversion efficiency based on $q_{ICS}$, Figure S9d shows the charge-to-spin current conversion efficiency $J_S/J_C$ obtained by considering the three-dimensional carriers, in other words, the conventional way often applied for the estimation of conversion efficiency [ref. S7]. The primary data
for the analysis in ST-FMR is \( V^{\text{Anti}} \) and \( V^{\text{Sym}} \) shown in Fig. S9a and S9b, those implying constant current density applied in Cu layer and almost zero spin current in Py, respectively. Then, \( J_{C}^{\text{TLSurf}} \) corresponds to the resistivity variation (Fig. S3b) and values listed in Table S1a. In this analysis with these data set, the efficiency in BST with \( x = 0 \) and \( 1 \) is seemingly smaller than that of BST with \( x = 0.5, 0.7 \) and 0.9. However, we can never discuss the charge-to-spin current conversion due to spin-momentum locking in the surface state, because the uniform charge current in the whole area of BST film is assumed in the above analysis. This is indeed the reason why we proposed the interfacial charge-to-spin current conversion efficiency \( q_{ICS} \) in the main text; in Fig. 3(a), we plotted the \( q_{ICS} \) estimated by taking into account a certain current distribution, which is discussed in Section 1 of supplementary information. Moreover, the small values of \( q_{ICS} \) for \( x = 0.82 \) and 0.88 with \( E_F \) closing to DP is experimentally valid so that we discuss the possible origins for this reduction.
10. Temperature dependence of resistance of BST films

Figure S10 | Measurement temperature dependence of sample resistance $R_{xx}$.

Figure S10 shows the temperature dependence of longitudinal resistance $R_{xx}$ of BST for $x = 0, 0.5, 0.7, 0.82, 0.88, 0.9$ and $1$ samples. $R_{xx}$ systematically changes with Sb concentration $x$. For the end compounds of $x = 0$ (Bi$_2$Te$_3$) and $1$ (Sb$_2$Te$_3$), $R_{xx}$ shows a monotonic metallic temperature dependence, indicating the Fermi level is in the bulk band. In contrast, for $x = 0.5, 0.7, 0.82, 0.88$ and $0.9$, $R_{xx}$ exhibits a non-monotonic temperature dependence. As previously reported [S8], the insulating behavior at high temperature is due to the insulating bulk state whereas the metallic behavior at low temperature is originating from the metallic surface state. Combined with the Hall coefficient shown in Fig. 2a in the main text, the Fermi level should be in the bulk band gap around the charge neutral point of $x \sim 0.84$.

Please note that $R_{xx}$ in $x = 0.5$ sample exhibits a weak temperature dependence, indicating that $x = 0.5$ is far from the charge neutral point. The less insulating but not metallic behavior of $R_{xx}$ suggests that the Fermi level is close to the band edge of bulk conduction band and hence $x = 0.5$ sample is not an ideal intrinsic semiconductor. Nevertheless the conductivity of the bulk region is still smaller than...
that of the surface state at 10 K. Therefore, we treated $x = 0.5$ sample in the same manner as other intrinsic ones such as $x = 0.7$, 0.82, 0.88 and 0.9 samples.

11. Frequency dependence of $q_{ICS}$

![Figure S11](image_url)

**Figure S11 | Input rf frequency dependence of $q_{ICS}$ in BST ($x = 0.7$)**

The $V_{Sym}$ can be modulated by the influence of parasitic impedance in the circuit for ST-FMR which may give artificial symmetric voltage contributions. Figure S11 shows frequency dependence of $q_{ICS}$. If there are non-negligible voltages due to parasitic impedance, the $q_{ICS}$ values should depend on input rf frequency. However, the estimated $q_{ICS}$ values were independent on rf frequency. Thus we think that the artificial voltage due to parasitic impedance can be negligible in this ST-FMR measurement.
References:

S1. COMSOL Multiphysics engineering simulation software (www.comsol.com).


