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

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Spin–momentum locking in the Dirac surface state of a topological insulator (TI)1,2 offers a distinct possibility for highly efficient charge-to-spin current (C–S) conversion compared with spin Hall effects in conventional paramagnetic metals3–13. For the development of TI-based spin current devices, it is essential to evaluate this conversion efficiency quantitatively as a function of the Fermi level position \(E_F\). Here we introduce a coefficient \(q_{\text{CS}}\) to characterize the interface C–S conversion effect by means of the spin torque ferromagnetic resonance (ST-FMR) for (Bi\(_{1-x}\)Sb\(_x\))\(_2\)Te\(_3\) thin films as \(E_F\) is tuned across the bandgap. In bulk insulating conditions, the interface C–S conversion effect via the Dirac surface state is evaluated as having large, nearly constant values of \(q_{\text{CS}}\), reflecting that \(q_{\text{CS}}\) is inversely proportional to the Fermi velocity \(v_F\) and/or instability of the helical spin structure. These results demonstrate that fine tuning of \(E_F\) in Ti-based heterostructures is critical in maximizing the efficiency using the spin–momentum locking mechanism.

Three-dimensional topological insulators (TIs) possess metallic surface states in which the spins of carriers are locked orthogonal to their momenta as a result of the time-reversal invariant. This feature is called ‘spin–momentum locking,’ and has been employed as the principal mechanism to induce spin accumulation in the surface states of TIs (refs 1,3–5,14–17). Conceptually, the charge current can fully contribute to the spin current via spin–momentum locking; a C–S conversion efficiency \(\theta_{\text{CS}}\) of 100% is expected at the non-TI/TI heterointerface. This highly efficient C–S conversion can be widely applicable to spintronic devices. However, the C–S conversion efficiency deduced from the spin torque measurement can exceed 100% for TIs with \(E_F\) located in the bulk band, leading to mixed contributions from the surface and bulk bands3–4, when the efficiency is defined as \(\theta_{\text{CS}} = J_S / J_C\), where \(J_S\) is the spin current density (A m\(^{-2}\)) and \(J_C\) is the charge current density (A m\(^{-2}\)) in the entire TI layer. Here we isolate the contribution of the Dirac electrons in the C–S conversion process and clarify the role of the Fermi level \(E_F\) and the Fermi velocity \(v_F\) by employing TI samples with various \(E_F\) positions. Accordingly, we define the interface C–S conversion coefficient \(q_{\text{CS}}\) as \(q_{\text{CS}} = J_S / J_C\), where \(J_C\) is the surface charge current density (A m\(^{-2}\)). Based on the concept of spin–momentum locking, the magnitude of \(J_S\) is governed by that of \(J_C\), which is linked with the conductivity of the surface states on the TI layer depending on the Fermi energy and Dirac dispersion: the Fermi velocity \(v_F\) and the Fermi wavevector \(k_F\) (refs 15–17). In this study, we quantitatively evaluate the interface C–S conversion effect by means of the ST-FMR technique for 8-nm (Bi\(_{1-x}\)Sb\(_x\))\(_2\)Te\(_3\)/8-nm Cu/10-nm NiFe\(_{20}\) (Py) trilayer films, as shown in Fig. 1a. Systematic control of the Fermi levels by varying \(x\) in (Bi\(_{1-x}\)Sb\(_x\))\(_2\)Te\(_3\) (BST) thin films enables us to investigate the relationship between \(q_{\text{CS}}\) and the transport properties at the surface state.

The ST-FMR technique has been routinely employed to evaluate the spin current induced via the spin Hall effect in paramagnetic metals3–13. Here we apply this technique to characterize quantitatively the interface C–S conversion effect due to spin–momentum locking in Cu-inserted TI-based trilayer heterostructures, as shown in the top schematic of Fig. 1a. On insertion of a Cu layer between the TI and ferromagnet layers, spin accumulation at the surface states can be separately evaluated owing to suppression of the exchange coupling between the ferromagnet and the surface states of TI (refs 18–20). In addition, the deposition of Cu on BST probably plays a minor role in varying the surface state condition, such as through an energy shift of the Dirac point and valence band maximum3, owing to a similar magnitude between work function of Cu and the electron negativity of BST. A photo of the device and measurement circuit is shown at the bottom of Fig. 1a. To evaluate \(q_{\text{CS}} = J_S / J_C\) by means of ST-FMR, the charge current distribution in the trilayer should be clarified numerically, because \(J_C\) in the TI layer is one of the dominant factors in this evaluation technique. When a radiofrequency (rf) current flows in the trilayer film, FMR is excited in the top Py layer under an external static magnetic field \(H_{\text{ex}}\). Owing to the presence of the highly conductive Cu layer, the peak of the current density is located towards the outside of the Py layer so that homogeneous rf fields \(H_{\text{ex}}\) can be applied to the Py layer (see Supplementary Information 1), providing better conditions for characterizing the C–S conversion effect by means of ST-FMR22. Accumulation of spins takes place simultaneously in the surface state of the TI; these accumulated spins generate a spin current \(J_S\) in the orthogonal direction, diffusing into both Cu and Py layers, and thus exert a spin torque on the Py layer (white arrow in Fig. 1a). Note that the spin pumping effect, as an inverse effect to the charge-to-spin conversion, provides a small contribution in the evaluation of \(q_{\text{CS}}\) (Supplementary Information 8). A typical ST-FMR spectrum is shown in Fig. 1b: the symmetric voltage

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is attributable to the spin torque $\tau_x$ corresponding to the spin current density $J_s$ (details discussed later). By quantitative evaluation of $V_{\text{sym}}$, we can deduce the interface C-S conversion coefficient $q_{\text{ICS}}$.

Figure 2a shows the Hall coefficients $R_H$ obtained for seven films having different Sb composition $x$ at 10 K. The value of $R_H$ is negative for $x=0$, with an increasing magnitude as $x$ is increased to 0.82, indicating n-type conduction and a reduction in the electron density. The polarity of $R_H$ abruptly reverses its sign when $x$ reaches a value of approximately 0.84, revealing that the Fermi energy traverses the Dirac point ($D_P$). In the range $0.88 \leq x \leq 1$, the polarity of $R_H$ is positive, indicating p-type conduction. The charge carrier densities and mobilities shown in Fig. 2b are estimated from the $R_H$ values. Compared to the previous studies, the charge carrier density in the surface state reaches as low as $10^{12}$ cm$^{-2}$, indicating that $E_F$ is finely tuned close to the Dirac point. The mobility $\mu$ in BST films with $x=0.88$ reaches a maximum value of 1.900 cm$^2$/V s, which is comparable to the previous results. These transport properties, and the temperature dependence of the resistivity (Supplementary Information 10) ensure that the Fermi level of BST is systematically varied from n- to p-type across the DP in a controlled manner, as shown in Fig. 2d.

Figure 2c shows the dependence of $V_{\text{sym}}$ on the Sb composition $x$ obtained from an ST-FMR spectrum measured at an rf power of 8 mW. We confirmed that an rf power of 8 mW is low enough to ensure a linear response, with suppression of heating effects (Supplementary Information 5). The sign of $V_{\text{sym}}$ indicates the spin polarization direction of the spin current. We found a positive $V_{\text{sym}}$ in both n- and p-type BST films, which is an ideal feature of the C-S conversion via spin–momentum locking[14]. In the Dirac dispersion shown in Fig. 2e,f, spins on the Fermi circles of n- and p-type surface states of BSTs rotate clockwise and anti-clockwise, respectively. When the electric field $E_z$ is applied in the $-x$-direction, the Fermi circle with the chiral spin structure is shifted from the dashed circles to the solid circles by an amount proportional to $E_z$ along $k_x$, as shown in Fig. 2e. When the Fermi level $E_F$ is above the DP, the surface state of BST films has a higher population of down spins, generating spin polarization of the spin current along the $-y$-direction. When the Fermi level $E_F$ is in the valence band of the Dirac dispersion, up spins with momenta along $+k_y$ are fewer in number than down spins with $-k_y$. Thus the accumulated spin is oriented along the same direction for both n- and p-type BST films. Note that these results are different from the case of a typical semiconductor such as GaAs[15], whose spin Hall effect exhibits a different sign, depending on the carrier type.

The values of $q_{\text{ICS}}$ and the spin current conductivity $\sigma_s$ of BST films are summarized as a function of $x$ in Fig. 3. The value of $q_{\text{ICS}}$ can be experimentally evaluated from the ratio of $V_{\text{sym}}$ to $V_{\text{Anti}}$ in the ST-FMR spectrum. By using the conventional evaluation term $\theta_{\text{ICS}} = J_s/J_c$, with assumption of a uniform $J_c$ in the BST film regardless of $E_F$ position, large values of $\theta_{\text{ICS}}$ are obtained for $x=0.5, 0.7$ and 0.9, consistent with previous studies (Supplementary Information 9). Here, we propose a scheme for evaluation of $q_{\text{ICS}}$ making use of $J_c$. In the ST-FMR process, the values of $V_{\text{sym}}$ and $V_{\text{Anti}}$ correspond, respectively, to the spin-induced torque $\tau_x$ and the Oersted-field-induced torque $\tau_z$ generated by charge current flow. These two torques per unit moment on the Py are respectively expressed as $\tau_x = \eta J_c/2 (2eJ_s M_s M_{\text{Py}})$ and $\tau_z = \xi (J_c^2 M_1^2 C_{\text{Py}}^2 + J_c J_z)/2$, where $M_1$, $t$ and $\xi$ are the saturation magnetization, film thickness, and reduction factor of the rf field. Note that $V_{\text{Anti}}$ shows a sin $2\theta$ $\cos \theta$ dependence on the rotation angle of the applied magnetic field (Supplementary Information 6), indicating that $V_{\text{Anti}}$ originates purely from the Oersted field. The value of $\xi$ is calculated numerically by means of a finite element method (see Supplementary Information 1). The value of $q_{\text{ICS}}$ can thus be given by

$$q_{\text{ICS}} = \frac{J_s}{J_c} = \frac{\tau_x}{\tau_z} = \frac{\tau_x}{\tau_z} = \frac{a^2 e^2 J_s e_{\text{C}} M_s (1 + M_1/H_{\text{ext}})^2}{2 \hbar}\left(1 + M_1/H_{\text{ext}}\right)$$

$$= \left(\frac{V_{\text{sym}}}{V_{\text{Anti}}/a}\right)^2 = \frac{a^2 e^2 J_s e_{\text{C}} M_s (1 + M_1/H_{\text{ext}})^2}{2 \hbar}\left(1 + M_1/H_{\text{ext}}\right)$$

where $a$ is the ratio of $I_c^2$ (A m$^{-2}$) to $J_c$. The spin current density into Py $J_s^0$ (A m$^{-2}$) is proportional to the spin accumulation at the surface state of the TI, $\langle \delta S_0 \rangle$, which is expressed as

$$\langle \delta S_0 \rangle = \frac{\hbar}{2} k_T \delta k_x = \frac{e k_T E_z}{2} \tau = \frac{\mu k_T E_z}{2v_F}$$

where $k_T$ is the Fermi wavenumber, $\delta k_x$ is the shift of Fermi circle, and $\tau$ is the relaxation time. In the two-dimensional system, $k_T^2$ is proportional to the carrier density. Therefore, $\langle \delta S_0 \rangle$ reduces to
spin–orbit entanglement in such a material with a strong spin–orbit interaction.\textsuperscript{27} We would also like to note that the product of \( q_{\text{ICS}} \) and the inverse conversion coefficient is expected to be approximately unity for the ideal case where there is no reduction of the in-plane spin polarization in the surface state of the TT.\textsuperscript{28} We now discuss the Fermi-level dependence of \( q_{\text{ICS}} \) shown in Fig. 3a. First, in contrast to the almost constant \( q_{\text{ICS}} \) for bulk insulating BST films with \( x = 0.5, 0.7 \) and 0.9, the values of \( q_{\text{ICS}} \) show a sharp dip around DP \( x \approx 0.82 \) and 0.88; the value of \( q_{\text{ICS}} \) decreases dramatically when \( E_F \) is located close to the DP, or equivalently \( k_x \) becomes approximately zero, originating from the almost zero \( V_{\text{Sym}} \) in our experiments (see Fig. 2c,d). Evaluated from the lowest charge carrier density of approximately \( 10^{12} \text{ cm}^{-2} \), the \( E_F \) position is located within \( \pm 60 \text{ meV} \) of the DP for \( x = 0.82 \) and 0.88, resulting in a small \( \langle \delta S_y \rangle \) due to the small \( \delta k_x \). In such a situation, with \( E_F \) close to the DP, a finite amount of scattering may reduce the generated spin polarization, as reported in experiments with spin-resolved angular-dependent photoemission spectroscopy (ARPES)\textsuperscript{29–31} and scanning tunnelling spectroscopy.\textsuperscript{32} Here we give possible reasons for the reduction in the spin polarization. First, when there are inhomogeneities, such as in the Bi/Sb composition, which can be regarded as analogy to electron–hole puddles in graphene,\textsuperscript{33} in the surface state of BST around the Dirac point, charge current can flow in directions other than the electric field direction. As a result, \( \langle \delta S_y \rangle \) with various spin directions will occur in the surface state of the TI, indicating that the \( \langle \delta S_y \rangle \) in the \( y \) direction will
Finally we show the spin current conductivity $\sigma_S$ defined as $\sigma_S = q_{ICS} \sigma_{ICS}^{bulk}$, where $\sigma_{ICS}^{bulk}$ is the conductivity of the surface state of the TI, as a function of the Sb composition $x$ in Fig. 3b. The values of $\sigma_S$ in bulk insulating BST ($x = 0.5$ and 0.7), excluding the DP and bulk conductive BST, take values close to $1.8 \times 10^4 \, \Omega^{-1} \, \text{m}^{-1}$, which are comparable to those reported for three-dimensional processes originating from the spin Hall effect in paramagnetic metals such as Pt ($3.4 \times 10^4 \, \Omega^{-1} \, \text{m}^{-1}$) (ref. 10) and $\beta$-W ($1.3 \times 10^4 \, \Omega^{-1} \, \text{m}^{-1}$) (ref. 11). This high value of $\sigma_S$ is certainly beneficial, not only for realizing highly efficient magnetization switching, but also for realizing non-volatile spin switching for Boolean and non-Boolean logic initially based on metal spin Hall effects$^{14}$.

**Methods**

Methods, including statements of data availability and any associated accession codes and references, are available in the online version of this paper.

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**Author contributions**

Y.O. and Y.T. conceived the project. K.K. made the devices and performed the spin torque ferromagnetic resonance measurements. R.Y. grew the topological insulator thin films and performed Hall measurements. K.K. analysed the data and wrote the manuscript with contributions from all authors. A.T., Y.F., K.S.T., J.M., M.K., Y.T. and Y.O. jointly discussed the results.

**Additional information**

Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to K.K.

**Competing financial interests**

The authors declare no competing financial interests.
Methods

Sample fabrication. We grew 8-nm-thick BST films on semi-insulating InP(111) substrates by molecular beam epitaxy. The detailed growth conditions are described in a previous paper. The Bi/Sb ratio was tuned by adjusting the ratio of the beam equivalent pressures of Bi and Sb. Resistivity and Hall effect measurements were carried out using small chips derived from the same samples as used for the ST-FMR measurements (see Supplementary Information). Thin films of 8-nm Cu/10-nm Ni$_{80}$Fe$_{20}$ (Py)/5-nm Al$_2$O$_3$ were grown on the BST films by e-beam evaporation at a pressure of $5 \times 10^{-5}$ Pa. Al$_2$O$_3$ is used as an insulating capping layer. The resistivities of Cu and Py are measured to be 10 and 60 $\mu\Omega$ cm at 10 K. The BST/Cu/Py trilayer films were patterned into rectangular elements ($10 \times 30, 15 \times 45, 20 \times 60, 30 \times 90, 40 \times 120 \mu m^2$) using optical lithography and an Ar-ion etching technique. A co-planar waveguide of 5-nm Ti/200-nm Au was deposited on both sides of the rectangular elements.

ST-FMR measurement set-up. An rf current with an input power of 8 mW is applied along the long edge of the rectangle by means of a microwave analog signal generator (Keysight: MXG N5183A). An external static magnetic field $H_{ext}$ in the range from 0 to 2.0 kOe is also applied in the film plane at an angle of $\theta = 45^\circ$ with respect to the current flow direction. We demonstrated the rf power dependence of $V^{Sym}$, the heating effect, the dc current dependence of the resonance field, the frequency dependence of the half-width at half-maximum of $\Delta$ for $V^{Sym}$ and the dependence on magnetic field angle of $V^{Anti}$, and concluded that the detected $V^{Sym}$ and $V^{Anti}$ are primarily due to the charge-to-spin conversion effect (Supplementary Information). All the experiments were performed at 10 K to measure the surface-dominant properties of TI.

Data availability. The data that support the plots within this paper and other findings of this study are available from the corresponding author upon reasonable request.