Unidirectional spin-Hall and Rashba–Edelstein magnetoresistance in topological insulator-ferromagnet layer heterostructures

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The large spin–orbit coupling in topological insulators results in helical spin-textured Dirac surface states that are attractive for topological spintronics. These states generate an efficient spin–orbit torque on proximal magnetic moments. However, memory or logic spin devices based upon such switching require a non-optimal three-terminal geometry, with two terminals for the writing current and one for reading the state of the device. An alternative two-terminal device geometry is now possible by exploiting the recent discovery of the unidirectional spin Hall magnetoresistance in heavy metal/ferromagnet bilayers and unidirectional magnetoresistance in magnetic topological insulators. Here, we report the observation of such unidirectional magnetoresistance in a technologically relevant device geometry that combines a topological insulator with a conventional ferromagnetic metal. Our devices show a figure of merit (magnetoresistance per current density per total resistance) that is more than twice as large as the highest reported values in all-metal Ta/Co bilayers.
The spin Hall effect (SHE) in non-magnetic (NM) heavy metals originates in their strong spin–orbit coupling (SOC) and has been extensively studied recently. When a charge current flows through a NM heavy metal, the SHE yields a spin accumulation at the interface with a proximal material. If the latter is a ferromagnetic (FM) layer, the spin accumulation at the interface can exchange angular momentum with the magnetic moments and exert a spin-orbit torque (SOT). With certain configurations and sufficient charge current density, the magnetization in the FM can be switched. SOT switching is believed to be potentially faster and more efficient than spin transfer torque (STT) switching that is typically used in magnetic tunneling junction (MTJ) devices for memory and logic applications.

SOT switching devices consist of a current carrying channel with a proximal nanomagnet whose magnetization determines the memory or logic state. Such devices need two terminals for writing the state of the device and an additional terminal, usually an MTJ on top of the nanomagnet, for reading the magnetization state of the device. Since the stable states of the nanomagnet are 180-degree-opposite to each other, symmetry prevents the anti-parallel results in different resistance states and sensing using USMR have both been considered for the same role of USMR. Although both USMR and UMR could play the same role of large unidirectional magnetoresistance (UMR) in NM/FM bilayers, such as Pt/Co and Ta/Sb)2Te3, usually less appealing for memory and logic applications. UMR is observed in Cr–Rh interfaces at very low temperatures (from 2 K up to 30 K).

Results
We observe USMR at temperatures between 20 and 150 K for (Bi,Sb)2Te3 (BST) and Bi2Se3 (BS). The largest USMR among our samples is more than twice as large as the best USMR in Ta/Co samples, in terms of USMR (or UMR) per total resistance per current density. This value is observed in a six quintuple layer (QL) BS and 5-nm-thick CoFeB bilayer at 150 K. The devices studied are fabricated from BST (t QL)/CoFeB (5 nm)/MgO (2 nm) and BS (t QL)/CoFeB (5 nm)/MgO (2 nm) thin film stacks (t = 6 and 10), grown by molecular beam epitaxy (MBE) and magnetron sputtering.

First and second harmonic resistance angular dependence.
Figure 2a shows the definition of the coordinates and rotation planes. Zero angles are at x+, y+ and z+ directions for xy, xz, and yz rotations respectively. The directions of rotation for increasing angle are indicated by the arrows. A 3 T external field is applied and rotated in the xy, xz, and yz device planes while the first harmonic resistance Rxy and second harmonic resistance R2xy are recorded with 2 mA RMS A.C. current. Figure 2b and c shows the angle dependencies of Rxy and R2xy respectively, of the BST (10 QL)/CoFeB (5 nm)/MgO (2 nm) sample at 150 K. The Rxy exhibits typical SMR-like behavior with R2 > R2 > R2. Similar to the
behavior seen in all metallic NM/FM bilayers, the variation of the second harmonic resistance $R_{2\omega}$ with angle is also proportional to the magnetization projected along the $y$-direction. This temperature gradient gives rise to the anomalous Nernst effect (ANE) and spin Seebeck effect (SSE). The ANE is a thermoelectric-effect-driven version of the anomalous Hall effect (AHE) of an FM, which originates from SOC in an FM. The SSE, on the other hand, is the result of the anomalous Hall effect (AHE) of an FM, which originates from SOC in an FM. The ANE/SSE both contribute to the longitudinal second harmonic signal when $\mathbf{M}$ is along the $y$-direction and to the transverse signal when $\mathbf{M}$ is along the $x$-direction. To carefully separate this contribution (denoted as $R_{2\omega}^{\text{ANES}}} + R_{2\omega}^{\text{AD}}$) from the measured $R_{2\omega}$ to verify USRMR, we carried out a series of measurements of Hall or transverse second harmonic resistance with $xy$-plane rotations under various external field strengths. Figure 3a shows the Hall resistance setup. The transverse resistance is measured while the external field is rotated in the $xy$-plane. The second harmonic Hall resistance, $R_{2\omega}^{\text{ANES}}} + R_{2\omega}^{\text{AD}}$, contains contributions from ANE/SSE, field-like (FL) SOT and anti-damping (AD) SOT. The ANE/SSE and AD SOT are proportional to $\cos \phi$ while the FL SOT is proportional to $\cos 3\phi + \cos 5\phi$. Figure 3b shows two examples of $R_{2\omega}^{\text{ANES}}} + R_{2\omega}^{\text{AD}}$ vs angle with 20 mT and 3 T external fields, respectively. Since the AD SOT and the FL SOT perturb the magnetization and thus contribute to $R_{2\omega}^{\text{ANES}}} + R_{2\omega}^{\text{AD}}$ through the AHE and the planar Hall effect, their effects diminish at larger external field. Figure 3b also shows that the data measured under a 20 mT field contain both $\cos \phi$ and $\cos 3\phi$ components, while under a 3 T field, the data exhibit almost no $\cos 3\phi$ component. There are two steps to obtain the $R_{2\omega}^{\text{ANES}}} + R_{2\omega}^{\text{AD}}$. First, by fitting the angle-dependent data, we extract the amplitudes of the $\cos \phi$ and $\cos 3\phi$ components. The FL SOT can then be easily determined and separated. This leaves the contributions of the ANE/SSE and the AD SOT, $R_{2\omega}^{\text{ANES}}} + R_{2\omega}^{\text{AD}}$, to the measured total Hall signal, $R_{2\omega}$. We plot the data corresponding to these contributions vs the reciprocal of total field, as shown in Fig. 3c. In this figure, $B_{\text{dem}} - B_{\text{ani}}$ is the demagnetization field.
expression of the SSE by high external assumption of the ANE/SSE signal being constant since the sample gives its highest USRMR at 70 K, while the USRMR are both temperature dependent. The 10 QL BST/CoFeB mation). At 70 K, BST and BS samples show resistance 4

**Second harmonic resistance components.** Figure 4a and b shows the $R_{2\omega}$, $R_{2\omega}^{\text{TT}}$, and $R_{\text{USRMR}}$ of 10 QL BST (Fig. 4a) and 10 QL BS (Fig. 4b) samples with 2 and 3 mA currents, respectively, at various temperatures. Here, the error bars indicate uncertainty bounds with 95% confidence (see Supplementary Note 8). Temperature affects the chemical potential and the relative contributions to transport from surface and bulk conduction in TIs. As a result, even though the magnetization and resistivity of the CoFeB layer vary little within the range of temperature in our experiments (see Supplementary Note 5 and Supplementary Fig. 5), the charge to spin conversion in TIs and the related USRMR are both temperature dependent. The 10 QL BST/CoFeB sample gives its highest USRMR at 70 K, while the $R_{2\omega}$ and $R_{2\omega}^{\text{TT}}$ keep increasing with increasing temperature up to 150 K. The USRMR of 10 QL BS/CoFeB can only be confirmed within between 50 and 70 K because of larger noise and magnetic-field-dependent signal outside this temperature window (see Supplementary Note 7 and Supplementary Figs. 7–9 for more information). At 70 K, BST and BS samples show resistance $R^2$ of 733 $\Omega$ and 488 $\Omega$, and USRMR per current density of $1.00 \pm 0.11$ and $0.63 \pm 0.10$ m$\Omega$ MA$^{-1}$ cm$^2$, respectively. The ratios of USRMR per current density to total resistance of the two samples are $1.37 \pm 0.15$ and $1.29 \pm 0.20$ ppm MA$^{-1}$ cm$^2$, respectively. These values are slightly better than the best result obtained using Ta/Co bilayers (1.14 ppm MA$^{-1}$ cm$^2$ at room temperature).
a function of temperature (BS\(\{x\}\) or BST\(\{x\}\) are abbreviations of BS or BST samples of \(\{x\}\) QL thicknesses); the error bars indicate uncertainty bounds with 95% confidence (see Supplementary Note 8). These two values also show very similar trends for all samples at various temperatures, except for the comparison between BST6 and BS10 at 70 K, in which BST6 is lower than BS10 in terms of \(\Delta R_{\text{USRMR}}/R\) but higher than BS10 in terms of \(\Delta R_{\text{USRMR}}\). The swap of position is mostly due to the larger total resistance of BST6 compared to BS10, while they show comparable \(R_{\text{USRMR}}\). We could also see that at 20 and 150 K, the BST6 does not show greater-than-zero USRMR reasonably beyond the confidence of the measurement. The largest values of \(\Delta R_{\text{USRMR}}/R\) and \(R_{\text{USRMR}}\) are \(0.90 \pm 0.12 \text{ m\Omega cm}^{-1}\) and \(3.05 \pm 0.39 \text{ ppm MA}^{-1}\) \(\text{cm}^2\), respectively, and both are observed in BS6 at 150 K. These values are more than twice as large as the best reported in the Ta/Co case\(^7\). The USRMR measurements beyond the temperature ranges of the plots of each sample show strong noise and field-dependent signal background as to render the estimations of USRMR unreliable (see Supplementary Note 7 and Supplementary Figs. 7–9 for more information).

As previously mentioned, the magnetization and resistivity of CoFeB vary only by less than 10% throughout the temperature range of our experiments (see Supplementary Note 5 and Supplementary Fig. 5). However, the USRMR, as shown in Fig. 5, varies significantly at various temperatures and between samples. We believe that the amount of current flowing in TIs dictates the USRMR performance of each sample at various temperatures. The basic transport properties of single-layer same-batch TI samples are summarized in Table 1. The resistivity, \(\rho_{3D}\), of TIs is about as low as 5 times (BS6) to as high as 42 times of that of a 5-nm single-layer CoFeB. This major difference of resistivity between the TI layer and the CoFeB layer leads to a small fraction of the charge current flowing in the former, which is then converted to spin accumulation at the interface and results in USRMR. When comparing the USRMR at 70 K across all four samples, we notice that the BS6 exhibits the highest USRMR while being the least resistive and of the largest carrier concentration (average), \(n_{3D}\). Another observation is that the BST6 sample has very low sheet carrier concentration down to \(1.14 \times 10^{12}\) range. This sample, based on its \(n_{3D}\), resistivity, and mobility, is almost an ideal TI. But surprisingly, this sample does not yield high USRMR. And in contrast, the BST10, which has \(n_{3D}\) of an order of magnitude higher, which is considered having considerable bulk conduction, exhibits higher USRMR. Then considering again, among the two BS samples, the one with higher \(n_{3D}\) also yields higher USRMR, we believe that it is very likely that in TI/FM systems, due to the large resistivity mismatch between ideal TI and FM, appropriate amount of bulk conduction on TI could help improving USRMR performance. And with this hypothesis, the overall trend of USRMR in sample BS6 decreased with lower temperature can be interpreted as the freezing of bulk conduction of TI. But this might only be significant when transitioning between a high temperature (70–150 K, for our experiments) to a very low temperature (20 K, for our experiments), where the freezing of bulk carriers is signification.

**Table 1 Summary of transport properties of bare TI samples**

| Sample | BS6 | BS10 | BST6 | BST10 |
|--------|-----|------|------|-------|
| \(\rho_{xx}\) (mΩ cm) | 0.636 | 0.765 | 4.50 | 5.86 |
| \(\mu\) (cm² V⁻¹ s⁻¹) | 246 | 474 | 730 | 58.8 |
| Type | n-type | n-type | p-type | p-type |
| \(n_{2D}\) (cm⁻²) | \(2.40 \times 10^{13}\) | \(1.72 \times 10^{13}\) | \(1.14 \times 10^{12}\) | \(1.81 \times 10^{13}\) |
| \(n_{3D}\) (cm⁻³) | \(3.99 \times 10^{19}\) | \(1.72 \times 10^{19}\) | \(1.90 \times 10^{18}\) | \(1.81 \times 10^{19}\) |

Bare TI samples made in the same batches with the BS\(\{6, 10\}\) and BST\(\{6, 10\}\) for USRMR study are referred in this table with the same names, except for the TI transport properties and conditions. The observation of the field sweep and angle rotation data (see Supplementary Note 8 for details).
of USRMR in a TFI/FM system is an important part of the puzzle to build a two-terminal TI-based SOT switching device. Such a two-terminal topological spintronic switching device is potentially more efficient compared to MTJs that use STT switching due to the large SOCM of TIs. The USRMR we observe could enable the read operation of such a device without having to build an MTJ structure on top of the TI. Such two-terminal devices are much more architecture-friendly and more readily embedded in current STT magnetic random-access memory architectures.

Methods

Thin film sample preparation. The Bi2Se3 or (Bi1−xSbx)2Te3 films were grown by MBE on semi-insulating InP (111) substrates. The InP (111) substrate is initially gently etched by argon ion milling (see Supplementary Note 6 and Supplementary Fig. 6 for impact of such etching on TI). Then the CoFeB layer was deposited using a Co0.8Fe0.2B5 target. Finally, an MgO layer was deposited to serve as a protection layer.

Device fabrication. The device fabrication began with photolithography followed by ion milling etching to define the Hall bars. Then, we carried out a second photolithography step and an e-beam evaporation, followed by liftoff to make contacts.

Electrical measurements. The devices were tested in a Quantum Design PPMS that provides temperature control, an external magnetic field, and sample rotation. The AC current at 10 Hz was supplied by a Keithley 6221 current source. A Keithley 6485 or an EG&G 7265 lock-in amplifier was used to supply the AC current at 10 Hz. The AC current was measured by a Keithley 6485 lock-in amplifier.

References

1. Ando, K. et al. Electric manipulation of spin relaxation using the spin hall effect. Phys. Rev. Lett. 101, 036601 (2008).
2. Miron, I. M. et al. Perpendicular switching of a single ferromagnetic layer induced by in-plane momentum rotation. Nature 476, 189–193 (2011).
3. Liu, L. et al. Spin-torque switching with the giant spin Hall effect of tantalum. Science 336, 555–558 (2012).
4. Kim, J. et al. Layer thickness dependence of the current-induced effective field vector in TaCoFeB/MgO. Nat. Mater. 12, 240–245 (2013).
5. Pai, C. F. et al. Spin transfer torque devices utilizing the giant spin Hall effect of tungsten. Appl. Phys. Lett. 101, 122404 (2012).
6. Aradhyia, S. V., Rowlands, G. E., Oh, I., Ralph, D. C. & Bahrami, R. A. Nanosecond-timescale low energy switching of in-plane magnetic tunnel junctions through dynamic oersted-field-assisted spin Hall effect. Nano Lett. 16, 5987–5992 (2016).
7. Nakayama, H. et al. Spin Hall magnetoresistance induced by a nonequilibrium proximity effect. Phys. Rev. Lett. 110, 206601 (2013).
8. Kim, J., Sheng, P., Takahashi, S., Mitani, S. & Hayashi, M. Spin Hall magnetoresistance in metallic bilayers. Phys. Rev. Lett. 116, 097201 (2016).
9. Avci, C. O. et al. Unidirectional spin Hall magnetoresistance in ferromagnet/normal metal bilayers. Nat. Phys. 11, 570–575 (2015).
10. Olejnık, K., Novák, V., Wunderlich, J. & Junghwark, T. Electrical detection of magnetization reversal without auxiliary magnets. Phys. Rev. B 91, 180402 (2015).
11. Avci, C. O. et al. Magnetoresistance of heavy and light metal/ferromagnet bilayers. Appl. Phys. Lett. 107, 192405 (2015).
12. Qi, X.-L. & Zhang, S.-C. Topological insulators and superconductors. Rev. Mod. Phys. 83, 1057–1110 (2011).
13. Hisch, D. et al. A tunable topological insulator in the spin helical Dirac transport regime. Nature 460, 1101–1105 (2009).
14. Neupane, M. et al. Observation of quantum-tunneling-modulated spin texture in ultrathin topological insulator Bi2Se3 films. Nat. Commun. 5, 3841 (2014).
15. Lachman, E. O. et al. Visualization of superparamagnetic dynamics in magnetic topological insulators. Sci. Adv. 1, e1500746 (2015).
16. Li, C. H. et al. Electrical detection of charge-current-induced spin polarization due to spin-momentum locking in Bi2Se3. Nat. Nanotechnol. 9, 218–224 (2014).
17. Tang, J. et al. Electrical detection of spin-polarized surface states conduction in (Bi0.5Sb1.5)2Te3 topological insulator. Nano Lett. 14, 5423–5429 (2014).
18. Tian, J., Miotkowski, I., Hong, S. & Chen, Y. Electrical injection and detection of spin-polarized currents in topological insulator Bi2Te3. Sci. Rep. 5, 14293 (2015).
19. Lee, J. S., Richardella, A., Hickey, D. R., Mkhoyan, K. A. & Samarth, N. Mapping the chemical potential dependence of current-induced spin polarization in a topological insulator. Phys. Rev. B 92, 155312 (2015).
20. Mollnik, A. R. et al. Spin-transfer torque generated by a topological insulator. Nature Nanotech. 11, 449–451 (2016).
21. Jamali, M. et al. Giant spin pumping and inverse spin Hall effect in the presence of surface and bulk spin–orbit coupling of topological insulator Bi2Se3. Nano Lett. 15, 7126–7132 (2015).
22. Deorani, P. et al. Observation of inverse spin Hall effect in bismuth selenide. Phys. Rev. B 90, 094403 (2014).
23. Shiomizu, Y. et al. Spin-electricity conversion induced by spin injection into topological insulators. Phys. Rev. Lett. 113, 196601 (2014).
24. Baker, A. A., Figueroa, A. L., Collins-McIntyre, L. J., van der Laan, G. & Hejedal, T. Spin pumping in ferromagnet-topological insulator-ferromagnet heterostructures. Sci. Rep. 5, 7907 (2015).
25. Rojas-Sánchez, J.-C. et al. Spin to charge conversion at room temperature by Spin pumping into a new type of topological insulator: α-Sn films. Phys. Rev. Lett. 116, 096602 (2016).
26. Wang, H. et al. Surface-state-dominated spin-charge current conversion in topological-insulator-ferromagnetic-insulator heterostructures. Phys. Rev. Lett. 117, 076601 (2016).
27. Fan, Y. et al. Electric-field control of spin–orbit torque in a magnetically doped topological insulator. Nat. Nanotechnol. 11, 352–359 (2016).
28. Mahendra, D. C. et al. Room-temperature perpendicular magnetization switching through giant spin–orbit torque from sputtered Bi2Se1−xTe x topological insulator material. Preprint at https://arxiv.org/abs/1703.05822 (2017).
29. Han, J. et al. Room-temperature spin–orbit torque switching induced by a topological insulator. Phys. Rev. Lett. 119, 077702 (2017).
30. Yasuda, K. et al. Large unidirectional magnetoresistance in a magnetic topological insulator. Phys. Rev. Lett. 117, 127206 (2016).
31. Miyazato, T. et al. Crossover behavior of the anomalous Hall effect and anomalous Nernst effect in itinerant ferromagnets. Phys. Rev. Lett. 99, 086602 (2007).
32. Kikkawa, T. et al. Longitudinal spin Seebeck effect free from the proximity Nernst effect. Phys. Rev. Lett. 110, 067207 (2013).
33. Avci, C. O. et al. Interplay of spin–orbit torque and thermoelastic effects in ferromagnet/normal-metal bilayers. Phys. Rev. B 90, 224427 (2014).
34. Kikkawa, T. et al. Critical suppression of spin Seebeck effect by magnetic fields. Phys. Rev. B 92, 064413 (2015).

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Author contributions

Y.L. and J.P.-W. conceived and designed the experiments. J.S.L., I.K. and N.S. grew TI thin films. D.Z. and M.J. deposited the rest layers of the thin film stacks. Y.L. fabricated...
and tested devices and processed data. All authors reviewed the results. Y.L. and J.-P.W. wrote the manuscript. All authors contributed to the completion of the manuscript.

**Additional information**

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