Large and Robust Charge-to-Spin Conversion in Sputtered Weyl Semimetal WTe$_x$ with Structural Disorder

Xiang Li$^{1,2*}$, Peng Li$^3$, Vincent D.-H. Hou$^4$, Mahendra DC$^1$, Chih-Hung Nien$^4$, Fen Xue$^1$, Di Yi$^3$, Chong Bi$^2$, Chien-Min Lee$^4$, Shy-Jay Lin$^4$, Wilman Tsai$^4$, Yuri Suzuki$^3$, and Shan X. Wang$^{1,2**}$

$^1$Department of Materials Science and Engineering, Stanford University, Stanford, California 94305, USA
$^2$Department of Electrical Engineering, Stanford University, Stanford, California 94305, USA
$^3$Department of Applied Physics, Stanford University, Stanford, California 94305, USA
$^4$Corporate Research, Taiwan Semiconductor Manufacturing Company, Hsinchu, Taiwan

Email address: *xiangsli@stanford.edu, **sxwang@stanford.edu
Abstract:

Topological insulators have recently shown great promise for ultralow-power spin-orbit torque (SOT) devices thanks to their large charge-to-spin conversion efficiency originating from the spin-momentum-locked surface states. Weyl semimetals, on the other hand, may be more desirable due to their spin-polarized surface as well as bulk states, robustness against magnetic and structural disorder, and higher electrical conductivity for integration in metallic magnetic tunnel junctions. Here, we report that sputtered WTe$_x$ thin films exhibit local atomic and chemical structures of Weyl semimetal WTe$_2$ and host massless Weyl fermions in the presence of structural disorder at low temperatures. Remarkably, we find superior spin Hall conductivity and charge-to-spin conversion efficiency in these sputtered WTe$_x$ films compared with crystalline WTe$_2$ flakes. Besides, the strength of unidirectional spin Hall magnetoresistance in annealed WTe$_x$/Mo/CoFeB heterostructure is up to 20 times larger than typical SOT/ferromagnet bilayers reported at room temperature. We further demonstrate room temperature field-free magnetization switching at a current density as low as 0.97 MA/cm$^2$. These large charge-to-spin conversion properties that are robust in the presence of structural disorder and thermal annealing pave the way for industrial production of Weyl semimetals. Our results open up a new class of sputtered Weyl semimetals for memory and computing based on magnetic tunnel junctions as well as broader planar heterostructures containing SOT/ferromagnet interfaces.
Introduction

Recently, topological insulators (TIs) [1, 2] emerge as a promising class of materials for converting charge current to spin-polarized current. Owing to their strong spin-orbit coupling and spin-momentum locked surface states [3, 4], TIs promises larger charge-to-spin conversion efficiency ($\xi_{ST}$) than spin-transfer torque (STT) filtering through a ferromagnetic metal (FM) or spin-orbit torque (SOT) arising from a heavy metal (HM). However, bulk states that dominate electron conduction in some TIs such as Bi$_2$Se$_3$ might not convert charge to spin as efficiently as surface states.[5-8] Also, when interfacing TIs with ferromagnetic metal (FM) in a magnetic tunnel junction (MTJ) configuration, magnetic impurities diffused out of the FM layer may drive the TI into a trivial state,[9] while TI’s helical surface states may be destroyed due to hybridization with metal bands,[10] making the TI/FM interface vulnerable to annealing or thermal effects. Lastly, for practical applications such as SOT-Magnetic Random Access Memory (MRAM), most of the current and power divert into the FM layer due to TIs’ much higher resistivity (> 1000 $\mu\Omega \text{cm}$)[5, 6, 11] than FM such as CoFeB (~130 $\mu\Omega \text{cm}$), while TIs also introduce large parasitic resistance in series with the MTJ. As the tunneling magnetoresistance of a MTJ is rather limited (< 600%)[12], an insulating SOT write line would significantly degrade the MTJ readout signal, which precludes TIs’ integration in future generations of MRAM.

In contrast, Weyl semimetals (WSMs),[13, 14] such as type-II WSM WTe$_2$, shows great prospect to overcome the above challenges. Both the surface Fermi arcs and bulk Weyl nodes are spin-polarized in WTe$_2$. [15-17] WSMs’ Weyl nodes and spin-momentum-locked surface states persist under broken time reversal and inversion symmetries, and are immune to disorder-induced Anderson localization.[18, 19] Another advantage of using WSMs to generate SOT compared with TIs is their higher conductivity ($\sigma_{xx}$) arising from its semi-metallic band structure and high carrier mobility [20], resulting in smaller parasitic resistance and current shunted into the FM layer.

In the past few years, $\xi_{ST}$ larger than one has been demonstrated in TI/FM heterostructures at room temperature [5, 6, 21, 22]. Lately, research also shows a large damping-like $\xi_{ST}$ up to 0.5 in WTe$_2$,[23] low-temperature enhancement of field-like torque when a current flows along its $b$ axis [24], as well as an out-of-plane damping-like $\xi_{ST}$ around 0.013 when a
current flows along its \(a\) axis.[25] However, most of the spintronics studies using topological materials employ crystalline TIs grown by molecular beam epitaxy (MBE) [5, 6, 21, 22, 26] and exfoliated WSM flakes from bulk crystals [23-25], which are not suitable for large-scale industrial manufacturing. Therefore, it is highly desirable to use industrial deposition techniques, such as sputtering to grow these novel topological materials. Surprisingly, amorphous \(\text{Bi}_x\text{Se}_{(1-x)}\) prepared by sputtering or thermal evaporation exhibits similar topological properties as crystalline TIs, such as high \(\xi_{\text{ST}}\) [11, 27] as well as surface states using angle-resolved photoemission spectroscopy[28]. Moreover, a recent theoretical work shows a topological metal phase in 3D amorphous metals exhibiting similar and even larger Hall conductivity over their crystalline counterparts.[29] However, no work has experimentally investigated the effect of structural disorder in topological semimetals.

Here, we report large and robust charge-to-spin conversion properties in sputtered disordered WSM WTe\(_x\). We first confirm our sputtered WTe\(x\) materials show similar local atomic and chemical structures as crystalline WTe\(_2\). Through low-temperature magnetoresistance and temperature-dependent resistance measurements, we find that WTe\(_x\) film hosts quasi-2D or 3D Weyl fermions depending on its thickness. The increase of structural disorder with WTe\(_x\) thickness also drives a crossover from weak anti-localization to weak localization. We characterize the SOT properties of as deposited WTe\(_x\)/CoFeB bilayers using spin-torque ferromagnetic resonance (ST-FMR), showing large spin Hall conductivity up to \(1 \times 10^5 (\hbar/2e) \ \Omega^{-1} m^{-1}\) and \(\xi_{\text{ST}}\) up to 0.8 at room temperature, which are larger than the corresponding values reported for crystalline WTe\(_2\) flakes. We also find 5-20 times larger unidirectional spin Hall magnetoresistance (USMR) signal in thermally annealed WTe\(_x\)/Mo/CoFeB heterostructure than existing FM/HM or FM/TI bilayers, possibly driven by magnon-scattering of spin-polarized Weyl fermions in WTe\(_x\). We further demonstrate room-temperature field-free magnetization switching at a current density as low as 0.97 MA/cm\(^2\) in this annealed heterostructure using a simple planar USMR device geometry.

**Results**

**Material Properties of WTe\(_x\) Thin Films**
We first confirm the sputtered \( \text{WTe}_x \) films share similar local chemical and atomic structures as crystalline \( \text{WTe}_2 \) via Raman spectroscopy and X-ray Photoelectron Spectroscopy (XPS). Raman spectroscopy is a widely used technique to identify materials based on their unique phonon vibrational mode fingerprints determined by crystal structure and chemical bonding. As shown in Figure 1a, we observe Raman modes \( B_{1g}^{10}, A_{2g}^4, A_{1g}^6, A_{1g}^7, A_{1g}^2 \) in uncapped single layer \( \text{WTe}_x \) samples with different thicknesses (\( t_{\text{WTe}_x} \)). As \( t_{\text{WTe}_x} \) increases, \( A_{1g}^5 \) and \( A_{1g}^2 \) Raman modes show smaller widths, while the \( A_{1g}^2 \) peak frequency decreases (Figure S1), which are consistent with the reports on bulk and flaked \( \text{WTe}_2 \) films.[30, 31]. We further employed XPS to confirm that the chemical bonding state of Te atoms in our \( \text{WTe}_x \) material is the same as that in \( \text{WTe}_2 \). As shown in Figure 1b, the binding energy of Te 3d_{5/2} electron is consistent with \( \text{WTe}_2 \) state reported in exfoliated flakes (572.7 eV), compared with metallic Te (573.0 eV) and \( \text{TeO}_x \) (576.3 eV). [32, 33]

**Figure 1. Material properties of \( \text{WTe}_x \) films with different thicknesses.** a. Raman spectroscopy of uncapped \( \text{WTe}_x \) films with different thicknesses measured the same day when deposited. The
Raman vibrational modes are labeled. b, X-ray Photoelectron Spectroscopy (XPS) profile of Te 3d binding energy region for WTe$_x$ films with different thicknesses, the data were obtained after in situ Ar ion sputtering of the capping layers on WTe$_x$ films. A doublet of Te 3d$_{5/2}$ and Te 3d$_{3/2}$ peaks are separate by 10.4 eV. c, Cross-section scanning transmission electron microscopy (STEM) image of WTe$_x$(5)/CoFeB(4) bilayer (hereafter, all numbers in parenthesis are in nm). d, Cross-section STEM image of WTe$_x$(58)/CoFeB(4) bilayer. e, Cross-section STEM image of WTe$_x$(47)/CoFeB(4) bilayer showing a small crystallite outlined by the green box. The digital diffractogram of the selected crystallite is shown in the green dashed box on top.

Then we study the evolution of structural disorder with the WTe$_x$ films thickness based on Raman spectroscopy and atomic-scale structural and chemical characterizations. As shown in Figure 1a, two groups of Raman modes co-exist in our WTe$_x$ films, i.e., a first group of $B_1^{10}, A_1^3, A_1^6, A_1^2, A_1^2$ modes and a second group of $B_1^{10}, A_2^3, A_2^5, A_2^2$ modes, which appear at different locations on the same 58 nm thick WTe$_x$ sample (more data in Figure S1). As $t_{WTe_x}$ decreases to 28 nm, modes in the two groups appear together accompanied with a mixture of $A_1^6, A_1^5$ modes. While for the 5 nm thick sample, only $A_1^5, A_1^2$ modes are visible, which is similar as reported in monolayer exfoliated WTe$_2$ flakes [30]. This first group appears when the laser is aligned along the $b$ axis of bulk WTe$_2$ crystal, and the second group along the $c$ axis.[30] The data suggest that as $t_{WTe_x}$ increases, the WTe$_x$ films segregate into small crystalline WTe$_2$ clusters with their $b$ or $c$ axis aligned normal. High-resolution bright-field scanning transmission electron microscopy (STEM) images on the 5 nm thick and 58 nm thick WTe$_x$ films are shown in Figure 1c-d. We see the 5 nm thick WTe$_x$ film exhibits an amorphous structure with sub-nm features, while in the 58 nm sample, ~10-nm features of dark clusters surrounded by white regions emerge. Using energy-dispersive X-ray spectroscopy (EDS), we find that the bright regions are W-rich with Te:W atomic ratio $x$ down to 0.87, while the dark regions are Te-rich with $x$ up to 1.49. The $x$ ratio variation in the 58 nm sample appears to be slightly larger compared with the 47 nm WTe$_x$ sample (Figure S2). Besides, we observe a small crystalline cluster embedded in amorphous WTe$_x$ films, exhibiting diffraction patterns similar as crystalline WTe$_2$ [32, 34], as shown in Figure 1e. Taking together the Raman, STEM, and EDS data, we conclude that uniform amorphous films form when $t_{WTe_x}$ is small (~ 5 nm), while segregation increases when $t_{WTe_x}$ is greater. This increased
segregation may explain the two groups of Raman modes in 58 nm thick WTe\(_x\).

**Quantum Transport in Topological WTe\(_x\) Thin Films**

To study the topological properties of the WTe\(_x\) thin films with different thickness thus segregation disorder, we measure magnetoresistance (MR) and temperature-dependent resistance. We find the increase of disorder and electron-phonon interaction drives a crossover from weak anti-localization (WAL) in a 2D Weyl fermion system (5 nm thick WTe\(_x\)), to weak localization (WL) in a 3D WSM system (41 nm thick WTe\(_x\)). As shown in Figure 2a, the 5 nm thick WTe\(_x\) film shows positive MR with a cusp-shaped perpendicular magnetic field (\(B_z\)) dependence at 4 K. We attribute this behavior to the WAL effect, which describes the destructive quantum interference of electron waves going around a self-intersecting path in opposite directions in a disordered electronic system with spin-orbit coupling (SOC).[35] This WAL behavior has been found in thin WTe\(_2\) flakes [34, 36] and differs from reported classical quadratic MR [20, 36] and positive linear MR in WTe\(_2\) [37, 38] (see Figure S3). In contrast, we find a negative MR in 41 nm thick WTe\(_x\) devices at 4 K, which we attribute to the WL effect driven by electron-phonon interaction. The almost identical negative MR when the magnetic field is aligned along different directions (see Figure S3) differs from chiral anomaly-induced negative MR which appears only when the magnetic field is parallel to the current.[39, 40].

We fit the 5 nm thick WTe\(_x\) MR curves using the simplified Hikami-Larkin-Nagaoka (HLN) equation describing the WAL correction to conductivity \(\Delta \sigma_{xx} = \sigma_{xx}(B_z) - \sigma_{xx}(0)\) under perpendicular magnetic field \(B_z\) in a quasi-2D system with strong SOC [35]:

\[
\Delta \sigma_{xx} \approx -\frac{\alpha_{HLN} e^2}{2\pi \hbar} \left[ \psi \left( \frac{1}{2} + \frac{B\phi}{B_z} \right) - \ln \left( \frac{B\phi}{B_z} \right) \right]
\]

Equation 1

where \(\psi\) is the digamma function, \(B\phi\) is the phase coherence characteristic field, and \(\alpha_{HLN}\) is 0.5 for WAL and -1 for WL. As shown in Figure 2c-d, the HLN equation yields a rather good fit up to \(\pm 9\) T and 50 K. The linear dependence of \(B\phi\) on temperature (\(B\phi \sim T^p, p = 1\)) indicates that electron-electron interactions rather than electron-phonon interactions (\(p = 3\)) contribute to the dephasing of electron waves.[41] We confirm the quasi-2D nature of the 5 nm thick WTe\(_x\) as the effective dephasing length \(l\phi = \sqrt{\hbar/4eB\phi}\) (ranging from 18 nm at 4 K to 5.2 nm at 50 K) is larger.
than the WTe₃ thickness. In contrast, fit based on a formula for the 3D system as proposed in [42] gives rise to \( l_\phi \) and \( p \) values that are not consistent with the 3D system model (see Figure S4). Meanwhile, a similar \( \alpha_{HLN} \) decrease as temperature increases as well as similar \( \alpha_{HLN} \) values between 0 and 0.5 have been observed in Dirac semimetal Cd₃As₂ films in the quasi-2D limit.[43]

Moreover, as shown in Figure 2b, the change of sheet conductance \( \Delta \sigma_{xx} = \sigma_{xx}(T) - \sigma_{xx}(4K) \) as a function of temperature can be fitted up to 150 K using an equation describing 2D massless Dirac fermions [44], i.e., \( \Delta \sigma_{xx} = (c_{ee} - c_{qi}) \ln(T) \), where \( c_{ee}, c_{qi} \) describe the contributions from electron-electron interaction and quantum interference, respectively. Note that as inversion symmetry is broken in the 5 nm thick amorphous WTeₓ, a Dirac node separates into two Weyl nodes.[14] Hence, the 5 nm thick WTeₓ behaves as a quasi-2D massless Weyl fermion system with electron-electron interaction [45].
Figure 2. Temperature-dependent resistance measurements of WTe$_x$ films with different thicknesses. a, Sheet resistance dependence on perpendicular magnetic field ($B_z$) for WTe$_x$ capped with MgO/Ta, measured at 4K. b, Change in sheet conductance ($\Delta \sigma_{xx}$) dependence on temperature. The 5 nm and 41 nm data below 150 K are fitted using ($c_{ee} - c_{qi}$)$\ln (T)$ and $c_{ee} T^{0.5} - c_{qi} T^{1.5}$ equations, respectively. c, Change in sheet conductance ($\Delta \sigma_{xx}$) dependence on $B_z$, measured at 4, 10, 20, and 50 K. The 5 nm and 41 nm data are fitted based on the 2D Hikami-Larkin-Nagaoka equation [35] and 3D WL/WAL formula proposed in [42], respectively. d, Dependence of phase coherence characteristic field ($B_\phi$) and fit parameters ($\alpha_{HLN}, -c_{1q}^q$) on temperature. The $B_\phi$ values for 5 nm and 41 nm data are fitted using $aT$ and $aT^3+b$ functions, respectively.

In order to understand the mechanisms driving the crossover from WAL to WL as $t_{WTe_x}$ increases to 41 nm,[44] we fit the change in sheet conductance $\Delta \sigma_{xx}$ as a function of $B_z$ using an equation describing WL and WAL in 3D WSM as proposed in [42]:

$$\Delta \sigma_{xx} = \frac{c_1^q B_z^2 \sqrt{|B_z|}}{B_\phi^2 + B_z^2} + \frac{c_2^q B_\phi^2 B_z^2}{B_\phi^2 + B_z^2}$$  \hspace{1cm} \text{Equation 2}

where fitting parameters $C_{1q}^q, C_{2q}^q$ are positive for WL and negative for WAL. As shown in Figure 2c-d, the $B_\phi$ dependence on temperature can be fitted by a $aT^3 + b$ formula, indicating electron-phonon interaction ($B_\phi \sim T^p, p = 3$) is the dominating dephasing process, rather than electron-electron interaction ($p = 3/2$).[41] The non-zero $B_\phi$ value at 0 K indicates surface scattering and/or impurity also play a role in the 58 nm thick WTe$_x$ film.[43, 46] Next, we fit the temperature dependence of $\Delta \sigma_{xx}$ up to 150 K using an equation [44] describing 3D Weyl fermions, $\Delta \sigma_{xx} = c_{ee} T^{0.5} - c_{qi} T^{p/2}$. The good fit using $p = 3$ further confirms the dominant role of electron-phonon interaction. We also verify that the 3D equation is valid as $l_\phi$ values of 3.5 to 6.0 nm are much smaller than the WTe$_x$ thickness (41 nm), while a fit using the equation for the 2D massless Dirac fermions fails (see Figure S5). Therefore, the 41 nm thick WTe$_x$ thin film is a 3D WSM and the segregation discussed above possibly leads to the strong electron-phonon interaction.
Charge-to-Spin Conversion in WTe$_x$/CoFeB Bilayers

We measure large charge-to-spin-conversion efficiency ($\xi_{ST}$) up to 0.8 in as-deposited WTe$_x$/CoFeB bilayers with varying $t_{WTe_x}$ using the spin-torque ferromagnetic resonance (ST-FMR) technique. As illustrated in Figure 3a, by flowing GHz RF current through the WTe$_x$/CoFeB bilayers, the oscillating CoFeB anisotropic magnetoresistance driven by damping-like and field-like torques ($\tau_{DL}, \tau_{FL}$) generates a DC voltage when mixed with the RF current. Based on established methodology (Supplementary Note S6)[47], we fit the mixing voltage $V_{mix}$ using symmetric and asymmetric Lorentzian shapes which originate from $\tau_{DL}$ and $\tau_{FL}$ respectively as in Figure 3b. The extracted $\xi_{ST}$ values for different WTe$_x$ thickness samples are shown in Figure 3c, where each $\xi_{ST}$ value was averaged across 5, 6, 7, and 8 GHz measurements. Notably, the 58 nm thick WTe$_x$ sample shows a large $\xi_{ST}$ value up to 0.8 along with significant variations among different devices on the same sample, as shown in the inset of Figure 3c. This variation is consistent with the two groups of Raman modes due to strong segregation discussed above. An *ab initio* study on WTe$_2$ has also shown that the intrinsic spin Hall conductivity varies when current flows along different axes of WTe$_2$.[48]
Figure 3. Charge-to-spin conversion in WTe\textsubscript{x}/CoFeB bilayers with different WTe\textsubscript{x} thicknesses. a, Schematic of spin-torque ferromagnetic resonance (ST-FMR) experimental setup using fabricated WTe\textsubscript{x}(t_{WTe\textsubscript{x}})/CoFeB(4)/MgO(2)/Ta(2) microstrips. The damping-like and field-like torques are labeled as $\tau_{DL}$, $\tau_{FL}$ respectively. b, Representative mixing voltage obtained in ST-FMR measurement as a function of applied magnetic field $H$ strength for WTe\textsubscript{x} thickness of 58nm measured at 5 GHz RF excitation. The raw data is being fitted by a sum of symmetric and asymmetric Lorentzian. c, $\xi_{ST}$ dependence on WTe\textsubscript{x} thickness from ST-FMR measurements. Inset shows the charge-to-spin conversion efficiency $\xi_{ST}$ for different devices on the 58nm-thick WTe\textsubscript{x} sample, each data point are averaged from ST-FMR measurements at 5, 6, 7, and 8 GHz. d, WTe\textsubscript{x} spin Hall conductivity $\sigma_s$ as a function of longitudinal conductivity $\sigma_{xx}$ for both as-deposited WTe\textsubscript{x}/CoFeB and annealed WTe\textsubscript{x}/Mo/CoFeB heterostructures, measured at room temperature. The $t_{WTe\textsubscript{x}}$ for the as-deposited data decreases from 58 nm to 5 nm from the left to the right. e, Resistivity of WTe\textsubscript{x} single layer extracted from WTe\textsubscript{x}/CoFeB bilayers resistivity as a function of inverse of temperature ($1/T$). The data from 150 to 300 K are fitted using Arrhenius equation $\rho = \rho_0 \exp(E_a/k_BT)$.

The monotonic increase in $\xi_{ST}$ as a function of $t_{WTe\textsubscript{x}}$, as shown in Figure 3c, suggests that bulk spin Hall contributions dominate the charge-to-spin conversion. This bulk origin is consistent with the observation of bulk Weyl fermions in WTe\textsubscript{x} films at low temperatures. We further clarify the origins of the SHE through the evolution of spin Hall conductivity $\sigma_s (= \xi_{ST}/\rho_{xx})$ as a function of conductivity $\sigma_{xx}$, as shown in Figure 3d. The $\sigma_{xx}$ is derived from a parallel resistor model (Supplementary Note S7). According to several theoretical and experimental studies on the SHE in metals [49, 50] and anomalous Hall effects in ferromagnets[51], a crossover from intrinsic metallic regime to dirty metal regime happens when $\sigma_{xx}$ decreases to $10^5 - 10^6 \Omega^{-1}m^{-1}$ ($\rho_{xx}$ increases to 100 – 1000 $\mu\Omega$ cm). In the intrinsic metallic regime, $\sigma_s$ is roughly independent of $\sigma_{xx}$, while in the dirty metal regime, $\sigma_s$ decreases rapidly as $\sigma_{xx}$ decreases. Figure 3d indicates that $\sigma_s$ increases with $\sigma_{xx}$ (when WTe\textsubscript{x} is relatively thick) and then flattens out at $\sigma_{xx} > 2 \times 10^5 \Omega^{-1}m^{-1}$ range (when WTe\textsubscript{x} is relatively thin), which matches the reported crossover very well, suggesting that the increasing segregation disorder as $t_{WTe\textsubscript{x}}$ increases drives WTe\textsubscript{x} towards the dirty metal regime. Notably, the 58 nm thick WTe\textsubscript{x} film with strong segregation is not in the regime
of strong or Anderson localization (see Figure S8), confirming that Weyl semimetals are essentially immune to disorder-induced Anderson localization as predicted by theory.[52]

We further confirm this transition from intrinsic metal to dirty metal regime as \( t_{WTe_x} \) increases based on temperature-dependent resistivity measurements. As shown in Figure 3e, the insulating behavior is consistent with thermal activation-driven electron transport in the dirty metal regime.[51] Fitting of the resistivity data from 150 K to 300 K using a thermal activation model \( \rho = \rho_0 \exp(E_a/k_BT) \) reveals increasing thermal activation energy \( E_a \) as \( t_{WTe_x} \) increases (Supplementary Note S7), which is consistent with our previous finding of increasing degree of segregation disorder as \( t_{WTe_x} \) increases. Research has shown similar insulating behavior in degraded crystalline WTe\(_2\) flakes after exposure to air, consisting of 10 x 10 WTe\(_2\) atoms crystalline clusters embedded in an amorphous matrix, [34] which we also observe in Figure 1e. Note that although we have observed insulating temperature-dependent behavior in our WTe\(_x\) materials, they are still gapless semimetals in the presence of localization because carriers are still present at the Fermi energy as evidenced by the WAL and WL behaviors at low temperature.[36]

In the context of SHE, both intrinsic and side-jump mechanisms contribute to the SHE in the intrinsic metallic regime. Ab initio theory [48] has calculated \( \sigma_s \) to be up to \( 0.5 \times 10^5 (\hbar/2e) \Omega^{-1}m^{-1} \) in WTe\(_2\) due to intrinsic contributions, which is almost half of the \( \sigma_s \) value for our 5 nm thick WTe\(_x\) film \( (0.95 \times 10^5 (\hbar/2e) \Omega^{-1}m^{-1}) \). While the highest \( \sigma_s \) demonstrated in WTe\(_2\) flakes reaches \( 0.88 \times 10^5 (\hbar/2e) \Omega^{-1}m^{-1} \) in exfoliated 120 nm thick WTe\(_2\) flakes.[23] Hence, there is likely contributions from both intrinsic spin Hall effect and side-jump in our 5 nm thick WTe\(_x\) film. As side-jump contribution is proportional to impurity concentration, [53] it might play a significant role in our sputtered WTe\(_x\) films with structural disorder.

**SOT-Induced Switching of WTe\(_x\)/Mo/CoFeB Heterostructures Detected via USMR**

To demonstrate SOT-driven magnetization switching, we synthesized heterostructures composed of 5 nm thick WTe\(_x\) films with low resistivity and large \( \sigma_s \), which is better matched with metallic CoFeB and has higher write efficiency. Integration of sputtered WTe\(_x\) films into CMOS backend processes calls for a stack design sustaining SOT properties after thermal annealing processes. Here, we insert 1-2 nm thick Mo between WTe\(_x\) and CoFeB due to the
excellent thermal annealing stability, while the low $\xi_{ST}$ and large spin diffusion length of Mo will not significantly alter the SOT properties of the heterostructure. [54, 55] We first confirm there is no significant degradation of the WTe$_2$ film’s Raman spectrum after insertion of Mo and after 300 °C annealing (see Figure S9 and S10). Then we utilize second-harmonic Hall measurements to quantify the damping-like SOT in annealed WTe$_2$(5)/Mo(1)/CoFeB(1) heterostructure. Following established analysis methodology, [26, 56, 57] we determine that there are minimal field-like torque, Oersted field, thermoelectric effects (including anomalous Nernst and spin Seebeck effects), and damping-like SOT from the Mo inserttion layer. We also obtain $\xi_{ST}^{WTe_2} = 0.426 \pm 0.004$, $\rho_{WTe_2} = 435 \mu \Omega \text{cm}$, and $\sigma_s$ of $0.98 \times 10^5 (h/2e) \Omega^{-1} \text{m}^{-1}$ in this WTe$_2$(5)/Mo(1)/CoFeB(1) heterostructure, which is consistent with that of $0.95 \times 10^5 (h/2e) \Omega^{-1} \text{m}^{-1}$ in as deposited WTe$_2$(5)/CoFeB(4) bilayer discussed in the last section. This confirms that high $\sigma_s$ is sustained after insertion of Mo layer and thermal annealing. (Supplementary Note S11 and Figure S12)

We next characterize the unidirectional spin Hall magnetoresistance (USMR) effect which can distinguish magnetization along y and $-y$ directions via second-harmonic longitudinal resistance ($R_{2\omega}^{xx}$) measurements [58]. We use 300 °C-annealed WTe$_2$(5)/Mo(2)/CoFeB(1) heterostructures here for magnetization switching experiments as the CoFeB layer in the WTe$_2$(5)/Mo(1)/CoFeB(1) heterostructure has a negligibly small in-plane coercivity (see Figure S13). As shown in Figure 4a, when the magnetization aligns along different transverse orientations ($y$-axis), $R_{2\omega}^{xx}$ signal switches with a coercivity around 1-2 Oe. The $R_{2\omega}^{xx}$ signal vanishes at a large $H_y$ field, as shown in Figure 4a inset, which we attribute to spin-dependent electron scattering between SOT spin current and magnons in the ferromagnet [59-62], rather than spin-dependent electron scattering between SOT spin currents and the magnetization [58, 61, 63]. We also determine the thermoelectric contribution to the USMR effect to be negligible (Supplementary Note S14). Following reference [59], we use total USMR per current density per total longitudinal resistance $\Delta R_{2\omega}^{xx}/JR_{xx}$ to benchmark the strength of USMR across various material stacks and device geometries. Here, $\Delta R_{2\omega}^{xx}$ is defined as the maximal change of USMR when magnetization points to $-y$ or $y$ direction. Notably, the $\Delta R_{2\omega}^{xx}/J_{WTe_2}R_{xx}$ value of 82.2 ppm $\text{MA}^{-1}\text{cm}^2$ in this work is about 5-20 times higher than that in existing FM/HM or FM/FI bilayers (see Table S1). As the 5 nm thick WTe$_2$ film hosts quasi-2D Weyl fermions at
low temperature, the large USMR strength suggests that highly spin-polarized Weyl fermions plays a critical role in converting charge to spin more efficiently than other HMs or TIs.

Figure 4. Pulsed SOT-induced switching in WTe$_x$/Mo/CoFeB-based heterostructure detected via the USMR effect. a, Second-harmonic longitudinal resistance ($R_{2\omega}^{xx}$) as a function of the in-plane magnetic field along the y-axis ($H_y$) under an AC current amplitude of $I = 0.7 \, mA$ for WTe$_x$(5)/Mo(2)/CoFeB(1)/MgO(2)/Ta(2) heterostructure annealed at 300 °C for 30 minutes. The dotted line corresponds to $H_y = 0$. The top-left inset shows $R_{2\omega}^{xx}$ under a wider range of $H_y$ from -6000 Oe to 6000 Oe. The bottom-right inset shows the schematic of the second harmonic measurement with an input of sinusoidal current $I_{sin}(\omega t)$. b, $R_{2\omega}^{xx}$ measured as a function of pulse current amplitude $I_{pulse}$ under zero external field. The current pulse width is 1 ms. The read current amplitude is 0.7 mA. The red and black curves correspond to initialization of magnetization along $-M_y$ and $M_y$ direction respectively.

We then demonstrate room temperature pulsed current-driven switching of in-plane magnetization detected via the USMR effect and deduce a $\xi_{ST}^{WTe_x}$ value that is consistent with previous second-harmonic Hall measurements. First a square-shaped current pulse with a pulse width of $t_{pulse}$ flows through the current channel, and subsequently the $R_{2\omega}^{xx}$ was measured under an AC read current. Similar magnetization switching curves at zero external magnetic field with opposite magnetization initialization directions as shown in Figure 4b confirms that current-
induced SOT drives the switching. The damping-like SOT from the WTe$_x$ layer has an opposite sign to that of Pt [47] (Supplementary Note S15). The analog and gradual switching process indicates that multi-domains form during the switching process due to the small coercivity (< 2 Oe) of the CoFeB layer [64]. While the asymmetric switching current can be attributed to a small remnant field around 0.5 Oe during measurements, as seen from the shift of maximum and minimum $R_{2\omega}^{xx}$ values away from 0 field in Figure 4a. Note that 30.1% of the total current flows through the WTe$_x$ layer based on a parallel resistor model. (see Figure S12) The lowest switching current density $J_{WTe_x}$ achieved is 0.97 and 2.05 MA/cm$^2$ using a 100 ms pulse width.

Discussion

In contrast to the traditional method of detecting in-plane magnetization direction using a MTJ with two in-plane magnetic layers sandwiching a tunnel barrier, the USMR effect used in this work can detect in-plane magnetization switching in a simple planar WSM/FM bilayer geometry.[58] The 5-20 times larger USMR strength in our heterostructure not only shows promise of USMR-based novel bilayer spintronics devices, but also calls for more studies into the role of Dirac electrons/Weyl fermions-magnon scattering in charge-to-spin conversion physics.[59]

We last compare the charge-to-spin conversion properties in sputtered WTe$_x$ films with single crystalline WTe$_2$ flakes and other SOT materials. Remarkably, the $\sigma_s$ of 0.95 – 0.98 [× 10$^5$(h/2e)Ω$^{-1}$m$^{-1}$] in 5 nm thick sputtered WTe$_x$ films exceeds that of 0.88 × 10$^5$(h/2e)Ω$^{-1}$m$^{-1}$ in exfoliated 120 nm thick WTe$_2$ flakes.[23] Moreover, a large $\xi_{ST}$ value up to 0.8 is achieved in 58 nm thick sputtered WTe$_x$ films compared with around 0.2 for WTe$_2$ flakes at a similar thickness of 60 nm, as well as 0.51 in 120 nm thick WTe$_2$ flakes. The $\sigma_s$ in 5 nm thick WTe$_x$ is comparable with $\sigma_s$ in the range of 0.4 – 2 [× 10$^5$(h/2e)Ω$^{-1}$m$^{-1}$] in MBE-grown Bi$_2$Se$_3$ and sputtered Bi$_x$Se$_{1-x}$, though the latter have resistivity values (1755 – 13000 $\mu\Omega$ cm) much higher than the sputtered WTe$_x$ (159 – 435 $\mu\Omega$ cm). Our results are also comparable with sputtered narrow-bandgap TI Bi$_{1-x}$Sb$_x$ with $\sigma_s$ of 0.4 – 1.2 [× 10$^5$(h/2e)Ω$^{-1}$m$^{-1}$] and resistivity of 330 – 1000 $\mu\Omega$ cm.[65] As the switching power in future SOT-MRAM designs is proportional to $\rho_{SOT}t_{SOT}/\xi_{ST}^2$, the sputtered WTe$_x$ films promise up to 20 times lower write
power than WTe₂ flakes, as well as 15% and 2 times lower write power than W and Pt films, respectively (Supplementary Note S16). Our results will stimulate condensed matter theory and experimental studies to go beyond granular and amorphous TIs [11, 28, 66, 67], and to investigate the new class of topological amorphous or disordered semimetals [29] that are robust against time-reversal and inversion symmetry breaking as well as strong localization effects.

**Methods**

**Material deposition**

The WTeₓ and Mo films were deposited using ion-beam sputtering techniques using a stoichiometric WTe₂ target and Mo target. The ion-beam sputtering was conducted using Xe gas under 0.1 mTorr at room temperature. Meanwhile, MgO, Ta, and Co₂₀Fe₆₀B₂₀ magnetron sputtering guns are also integrated into the same vacuum chamber. Hence, we can achieve in-situ growth of the whole stack discussed in the main text without breaking vacuum. The Ta and Co₂₀Fe₆₀B₂₀ layers were deposited using DC sputtering, while the MgO layers were deposited using RF sputtering from an insulating MgO target. The stacks with Mo insertion were annealed at 300 °C for 30 minutes using an All-Win Rapid Thermal Process (RTP) system with Ar ambient.

**Device fabrication**

Temperature-dependent magnetoresistance and resistance data were gathered from Hall bars patterned on MgO(2)/WTeₓ(5)/MgO(2)/Ta(2) and WTeₓ(58)/Ta(2) stacks using a four-point probe method. The WTeₓ/CoFeB(4.4)/MgO(2)/Ta(2) (number in parenthesis is in nm) stacks for ST-FMR measurements were fabricated into 10 μm × 40 μm microstrips using standard photolithography and Ar ion mill techniques. The MgO(2)/WTeₓ(5)/Mo(1 or 2)/CoFeB(1)/MgO(2)/Ta(2) (number in parenthesis is in nm) stacks for second harmonic measurements were fabricated into 10 μm × 130 μm Hall bars using standard photolithography and Ar ion mill techniques. The patterned devices are subsequently covered with Ti(5 nm)/Au(120 nm) as contacts using photolithography and liftoff techniques.

**Film characterization**
Raman spectroscopy: The Raman spectrum was gathered using a Horiba Labram HR Evolution Raman System with a laser of 532 nm wavelength, a grating of 600 l/mm, and objective magnification of 100x. Each spectrum is an average of 10 captures each collected over 10 s. The laser spot size is around 0.3 μm.

X-Ray Photoelectron Spectroscopy (XPS): The XPS spectrum was gathered using a PHI VersaProbe System with Al (Ka) radiation (1486 eV). The focused ion gun used for in situ depth profiling has a sputter rate of around 2 nm/min with a beam energy of 500V 0.6 μA, and a raster size of 1 μm × 1 μm.

Transmission Electron Microscopy (TEM) and Energy Dispersive X-ray Spectroscopy (EDS): Cross-sectional TEM samples were prepared with Focused Ion Beam technique (Thermo-Fisher Helios 460G4). At the final sample thinning stages, Ga ion beam energy was reduced from 30kV to 8kV, 2kV, and then 500V to mitigate ion damages over TEM lamella surfaces. TEM lamellas were imaged with Thermo-Fisher Metrios TEM equipped with probe Cs-correctors operated at 200kV. STEM high-angle annular dark-field (HAADF) imaging is the primary imaging mode used in this study, and the resolution is better than 0.13nm. EDS experiment was carried out in STEM mode with the probe current set to about 600pA. The EDS detector system is of SDD type with a nominal collection solid angle of 0.7 or 0.9 radians (Super-X™ or Dual-X™, respectively). EDS data analysis was performed in Bruker’s Esprit EDS software. Quantification of W:Te ratio in WTeₓ film was obtained by using the built-in Cliff Lorimer factors of the Esprit EDS software. W signal maps (intensity of W M-lines) were processed by Principle Component Analysis (PCA) to isolate W-M lines from Si K-line and Ta-M lines.

Device Electrical Measurements

Spin-Torque Ferromagnetic Resonance (ST-FMR): ST-FMR measurements were performed on WTeₓ/CoFeB microstrips with sizes of 60 x 40 μm² using the setup. The external H field is oriented with a φ angle of 45° with respect to the microstrip thus current flow direction. A GHz signal was generated by an HP 83640B microwave source and was sent through the WTeₓ/CoFeB bilayer through a T-Bias and ground-signal-ground coplanar waveguide.

Second harmonic and pulsed switching measurements: The second harmonic setup consists of a Keithley 6221 current source providing AC current with a frequency of 1.333 kHz, and two Stanford Research SR830 Lock-in amplifiers recording the first and second harmonic signal of
longitudinal or Hall resistance of the Hall bar device. The pulse current used in the switching experiments were generated by Keithley 6221 through the square wave settings. The DC device resistance was obtained using a four-point probe method with Keithley 6221 current source and Keithley 2000 voltage meter.

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Author contributions

X.L. conceived and designed the research with contributions from P.L., M.D., C.B., S.-J.L., W.T., Y.S., and S.X.W.. S.X.W. supervised the study. X.L. deposited the thin films, carried out Raman spectroscopy, XPS, and second harmonic measurements. X.L. and P.L. fabricated the Hall bar and microstrip devices and carried out ST-FMR measurements. X.L., P.L., and D.Y. carried out the SQUID measurements. V.H., C.-H.N., and C.-M.L. carried out TEM and EDS studies. X.L. performed data analysis with contributions from P.L., V.H., M.D., F.X., C.-H.N., S.-J.L., W.T., Y.S., and S.X.W. X.L. wrote and revised the manuscript with input and comments from all authors.

Additional information

Supplementary information is available in the online version of the paper. Correspondence and requests for materials should be addressed to X.L. and S.X.W.

Competing financial interests:

The authors declare no competing financial interests.
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