

# Charge-spin conversion in layered semimetal TaTe2 and spin injection in van der Waals heterostructures

Downloaded from: https://research.chalmers.se, 2025-07-01 22:51 UTC

Citation for the original published paper (version of record):

Hoque, A., Khokhriakov, D., Karpiak, B. et al (2020). Charge-spin conversion in layered semimetal TaTe2 and spin injection in van der Waals heterostructures. Physical Review Research, 2(3). http://dx.doi.org/10.1103/PhysRevResearch.2.033204

N.B. When citing this work, cite the original published paper.

research.chalmers.se offers the possibility of retrieving research publications produced at Chalmers University of Technology. It covers all kind of research output: articles, dissertations, conference papers, reports etc. since 2004. research.chalmers.se is administrated and maintained by Chalmers Library

# Charge-spin conversion in layered semimetal TaTe<sub>2</sub> and spin injection in van der Waals heterostructures

Anamul Md. Hoque<sup>®</sup>, Dmitrii Khokhriakov, Bogdan Karpiak, and Saroj P. Dash<sup>®</sup> Department of Microtechnology and Nanoscience, Chalmers University of Technology, SE-41296, Göteborg, Sweden

(Received 5 December 2019; accepted 17 June 2020; published 6 August 2020)

A spin-polarized current source using nonmagnetic layered materials is promising for next-generation allelectrical spintronic science and technology. Here we electrically created spin polarization in a layered semimetal TaTe<sub>2</sub> via the charge-spin conversion process. Using a hybrid device of TaTe<sub>2</sub> in a van der Waals heterostructure with graphene, the spin polarization in TaTe<sub>2</sub> is efficiently injected and detected by nonlocal spin-switch, Hanle spin precession, and inverse spin Hall effect measurements. Systematic experiments at different bias currents and gate voltages in a vertical geometry prove the TaTe<sub>2</sub> as a nonmagnetic spin source at room temperature. These findings demonstrate the possibility of making an all-electrical spintronic device in a two-dimensional van der Waals heterostructure, which can be essential building blocks in energy-efficient spin-orbit technology.

DOI: 10.1103/PhysRevResearch.2.033204

#### I. INTRODUCTION

Spintronic technology was mainly governed by the generation of spin-polarized currents by using the exchange interaction of conduction electrons in magnetic materials, which require a magnetic field to control their spin orientation [1–4]. However, the demand for faster and smaller spintronic technologies for memory, logic, and artificial intelligence requires efficient and spontaneous spin-polarized current sources using nonmagnetic materials, where all the device operations can be controlled by electrical means [5,6]. Recently, the electronic generation of spin-polarized currents was realized in nonmagnetic materials via the spin Hall effect (SHE), Rashba-Edelstein effect (REE), and spin momentum locking (SML) phenomenon [7–9].

Due to their potential for spintronic applications, the SHE and REE were extensively studied in heavy metals [10,11], semiconductors, oxide heterostructures [7], and more recently, on transition metal dichalcogenides (TMDs) [12], Weyl semimetals [12,13], Rashba spin-orbit materials [14], and their heterostructures with graphene [12,13,15–18]. Topological insulators (TIs) were shown to possess SML characteristics in the Dirac surface states, which could be detected using potentiometric measurements [19–21] and the charge-spin conversion process [22–24]. However, the spin injection from TIs into graphene is so far restricted to very low temperatures due to contributions from trivial bulk states [25,26]. Utilizing the SHE in Pt metal thin films, spin injection and detection has been realized in a graphene channel

[10,11]. However, the direct deposition of metallic layers on graphene can modify its electronic properties [27,28]. Recently discovered two-dimensional (2D) magnets can be potential candidates for spin injection into graphene; however, the low Curie temperature of the 2D magnets at this stage limits their room-temperature applications [29]. Therefore, finding a nonmagnetic layered material as a spin source in a van der Waals (vdW) heterostructure with graphene is necessary for newly emerging 2D spintronics science and technology [30].

Using our recently developed vertical measurement geometry [13], we demonstrate the electronic creation and injection of in-plane spin polarization using a layered semimetal tantalum ditelluride (TaTe<sub>2</sub>) due to efficient charge-spin conversion processes. In a hybrid spintronic device, the spin polarization generated in TaTe<sub>2</sub> is efficiently injected into the graphene channel and subsequently detected by a ferromagnet using a sensitive nonlocal spin-valve and Hanle spin precession measurements. Detailed measurements using different bias currents and gate voltages provide insight into the currentinduced spin polarization in TaTe<sub>2</sub> at room temperature.

## **II. RESULTS AND DISCUSSION**

The choice of the layered monoclinic (1T') semimetal TaTe<sub>2</sub> is motivated by its high spin-orbit interaction (SOI) originating from 5*d* electrons along with the potential for use in charge-to-spin conversion technologies [7,31], and it can be exfoliated into thin 2D layers. Mainly, TaTe<sub>2</sub> is an excellent candidate for the generation of pure spin current due to SHE [7,32] and REE [9,33,34], as shown schematically in Fig. 1(a). Figure 1(b) shows the fabricated device picture, where a vdW heterostructure of TaTe<sub>2</sub> is prepared with a graphene channel to use its excellent spin transport properties [35,36] together with ferromagnetic Co/TiO<sub>2</sub> (FM) injector/detector tunnel contacts (see Appendix A for detailed fabrication processes). The characteristic Raman spectrum of a TaTe<sub>2</sub>

<sup>\*</sup>Corresponding author: saroj.dash@chalmers.se

Published by the American Physical Society under the terms of the Creative Commons Attribution 4.0 International license. Further distribution of this work must maintain attribution to the author(s) and the published article's title, journal citation, and DOI. Funded by Bibsam.



FIG. 1. Characterization of TaTe<sub>2</sub> and spin transport in the graphene-TaTe<sub>2</sub> heterostructure. (a) Schematic representation of the spin Hall effect in a symmetric system, where a spin current ( $I_s$ ) is created due to transverse charge current ( $I_c$ ). The adjacent diagram shows the current-induced spin polarization in Rashba spin-split bands near the Fermi level. (b) Optical micrograph with false colors of a device consisting of graphene (black), TaTe<sub>2</sub> (blue), and FM contacts (gray). The scale bar is 4 µm. (c) Raman spectrum of TaTe<sub>2</sub> at room temperature. (d) Magnetoresistance (MR%) of TaTe<sub>2</sub> at room temperature and 25 K. (e) Nonlocal spin-valve (NLSV) signal without and with spin absorption in the high- $R_{int}$  and low- $R_{int}$  conditions, respectively. The inset shows the NLSV measurement geometry.

flake is shown in Fig. 1(c) using a 638-nm laser, where three prominent peaks at 87, 116, and  $135 \text{ cm}^{-1}$  are visible [31]. In the transport measurements, TaTe<sub>2</sub> shows semimetallic properties (see Appendix B) and a large magnetoresistance ( $MR \ \% = \frac{R_B - R_0}{R_0} \times 100$ ) [Fig. 1(d)], showing MR ~30% at room temperature and ~120% at 25 K at a magnetic field of -0.8 T [37].

Standard nonlocal (NL) spin-valve measurements were performed across the graphene-TaTe<sub>2</sub> heterostructure channel in different graphene-TaTe<sub>2</sub> interface resistance ( $R_{int}$ ) conditions [Fig. 1(e)]. The spin-polarized electrons were injected and detected by FM contacts in the NL geometry while measuring a  $\Delta R_{nl} = (\frac{\Delta V_{al}}{l})$  as a function of magnetic field sweep ( $B_y$ ) to achieve the parallel and antiparallel magnetization of FM electrodes. As shown in Fig. 1(e), while a large spin transport signal  $\Delta R_{nl}$  of 272 m $\Omega$  was measured for high  $R_{int}$  of 16 k $\Omega$ , for very low  $R_{int}$ , tentatively < 100  $\Omega$  (see Appendix C), the  $\Delta R_{nl}$  drastically reduced to 16 m $\Omega$  due to strong spin absorption in the low- $R_{int}$  regime.

First, the spin-to-charge conversion experiments [measurement geometry in Fig. 2(a)] were performed in TaTe<sub>2</sub> via the inverse spin Hall effect (ISHE), when the  $R_{int}$  is <100  $\Omega$ (see Appendix C). In this condition, the spin current injected from a FM into the graphene channel is strongly absorbed by TaTe<sub>2</sub>, as shown in Fig. 1(e). The spin current in TaTe<sub>2</sub> subsequently gives rise to a transverse charge current due to the efficient spin-charge conversion in TaTe<sub>2</sub>, which is detected as a voltage signal  $(V_{nl})$  across the TaTe<sub>2</sub> [Fig. 2(b)]. By sweeping an in-plane magnetic field along the x axis,  $B_x$ , the  $V_{nl}$  varies antisymmetrically at low field and saturates at  $\pm 0.4 \,\mathrm{T}$  as the magnetization of the injector FM rotates completely toward the x axis. The saturation field range of the FM contacts for the  $B_x$  field sweep has been verified from the spin precession x-Hanle measurements in the graphene channel (see Appendix D). Considering the symmetry of the system with the spin current being absorbed from graphene into TaTe<sub>2</sub> along the z direction, spin polarization ( $\geq 0.4$  T) in the x direction and manifested orthogonal charge current  $(I_s)$  in the y direction can be due to the ISHE or the Edelstein effect (EE) in TaTe<sub>2</sub> [12]. The  $V_{n1}$  signal has been measured at different spin injection bias currents, which scales linearly [Fig. 2(c)] at room temperature.

We observed a large spin-charge conversion (SCC) signal at room temperature in TaTe<sub>2</sub> with signal amplitude  $\Delta R_{SCC} = \Delta V_{nl}/I_{bias} = -1.69 \mu V/-20 \mu A \sim 84 \text{ m}\Omega$ , which is about few times larger than in WTe<sub>2</sub> [12], MoS<sub>2</sub>, and Pt in heterostructure with graphene [10,11,17], one order of magnitude larger than in graphene-MoTe<sub>2</sub> heterostructure [38], and a few orders of magnitude higher than in Pt/Cu heterojunction [39] but comparable to the graphene-TaS<sub>2</sub> heterostructure system [15]. Recently, spin-orbit torque experiments using a TaTe<sub>2</sub>-ferromagnet heterostructure reported a dominating



FIG. 2. Charge-spin conversion effects in TaTe<sub>2</sub> at room temperature. (a) Measurement geometry to detect the inverse spin Hall effect (ISHE) in TaTe<sub>2</sub> by injecting a spin current from the ferromagnet-graphene channel into TaTe<sub>2</sub> by spin absorption and detecting a voltage signal across the TaTe<sub>2</sub> in a NL geometry. (b) ISHE signal  $V_{nl}$  measured with an in-plane magnetic field ( $B_x$ ) sweep for  $I = 50 \ \mu$ A at  $V_g = 40 \ V$  at room temperature in device 1. (c) Bias current dependence of the ISHE signal amplitude  $\Delta V_{nl}$  with a linear fit (solid line). (d) Measurement geometry where TaTe<sub>2</sub> is used as a spin-polarized current source for vertical injection of spins into the graphene channel, which is finally detected by a FM contact in a NL geometry. (e, f) The spin-switch and Hanle spin precession measurements for spin injection from TaTe<sub>2</sub> with a  $B_y$  and  $B_z$  sweep, respectively, with an application of an  $I = -30 \ \mu$ A with  $V_g = 40 \ V$  at room temperature in device 2. The up and down magnetic field sweep directions are indicated by arrows in (e) for spin-switch experiments. The Hanle data is fitted using Eq. (2). A linear background is subtracted from the data.

Oersted field contribution by local measurements [40], which is avoided in our NL measurements. Form our data, it is not possible to calculate the exact spin Hall angle ( $\theta_{SH}$ ) in TaTe<sub>2</sub>, as the spin diffusion length  $\lambda_{TaTe_2}$ , is unknown. Furthermore, the larger width of a TaTe<sub>2</sub> flake (2.6 µm) compared to the spin diffusion length in graphene (~2.5 µm) restrains the use of the available model [10–12]. An estimation of  $\theta_{SH}$  was found to be in the range of 0.2–0.6 using a simple model and considering  $\lambda_{TaTe_2} = 10-110$  nm and  $R_{int}$  of 10–100  $\Omega$ (see Appendix E). Moreover, we have also approximately calculated the inverse Edelstein effect (IEE) of TaTe<sub>2</sub> through IEE length  $\lambda_{IEE} = \theta_{SH} * \lambda_{TaTe_2}$ , and we found  $\lambda_{IEE} \approx 6-60$  nm (see Appendix E for more discussion) as a function of  $\lambda_{TaTe_2}$ (10–110 nm).

Next, we focus on the observation of vertical spin injection (VSI) of in-plane spin polarization from TaTe<sub>2</sub> into a graphene channel at room temperature [see Fig. 2(d) for measurement geometry]. In the spin-switch experiment, we measured the spin signal  $V_{nl}$  as a function of an in-plane magnetic field  $B_y$  sweep, which switches the magnetization of the FM detector contact from parallel to antiparallel orientation with respect to the in-plane injected spin polarization from TaTe<sub>2</sub> [Fig. 2(e)]. The amplitude ( $\Delta R_{nl} = \Delta V_{nl}/I_{bias}$ ) of the spin-to-charge conversion signal for vertical spin injection is about 240 m $\Omega$ . A confirmatory test of the in-plane spin polarization using a Hanle spin precession measurement was performed in the same NL geometry [Fig. 2(d)], where a perpendicular magnetic field ( $B_z$ ) was swept while keeping the magnetization of

the detector FM contact in the in-plane orientation. The  $B_z$ field induces spin precession in addition to diffusion and dephasing, which results in a reduction of the spin signal as the detector probes only the projection of the spin orientation onto the FM contact. Figure 2(f) shows the Hanle data measured at room temperature while injecting spin from TaTe<sub>2</sub> into the graphene channel, and fitting with Eq. (F1), we estimate the spin lifetime  $\tau_s = 195 \pm 26$  ps and spin diffusion length  $\lambda_s = \sqrt{\tau_s D_s} = 1.98 \pm 0.3 \mu \text{m}$ , considering the channel length between the TaTe<sub>2</sub> and detector FM electrode  $L = 3.75 \,\mu\text{m}$ (distance between the center of the TaTe<sub>2</sub> flake to the center of the FM electrode). We found the spin polarization of the FM detector contact is about  $P_{FM} \sim 6.13\%$  (Fig. 10). Subsequently, knowing the  $P_{FM}$ , the lower limit for the currentinduced spin polarization of TaTe2 is calculated to be about  $P_{\text{TaTe}_2} = 1.34\% \pm 0.26\%$  (see Appendix F).

We further investigated the back-gate  $(V_g)$  dependence of the VSI spin-switch signal using TaTe<sub>2</sub> as the spin injector. Figure 3(a) shows a sketch of the graphene-TaTe<sub>2</sub> heterostructure and the change in the Fermi-level position in graphene with an application of  $V_g$  from electron- to holedoped regimes, whereas no change is expected in TaTe<sub>2</sub> due to its metallic state. The measured spin-switch signals  $(V_{n1})$ at various  $V_g$  in the range from -40 to 40 V are depicted in Fig. 3(b). The gate dependence of the magnitude of the spin signal  $\Delta V_{nl}(\Delta R_{nl} \approx 250-400 \text{ m}\Omega)$  is presented in Fig. 3(c) (top panel), which shows that the spin-switch signal direction remains unchanged in the hole and electron transport regime.



FIG. 3. Gate dependence of spin injection signal from TaTe<sub>2</sub>. (a) Measurement setup and the band diagram of TaTe<sub>2</sub> and graphene with an application of a back-gate voltage ( $V_g$ ) across the Si/SiO<sub>2</sub> substrate. (b) The measured NL spin-switch signals at various  $V_g = -40-40$  V for both *n*- and *p*-doped graphene regime in device 3 with a bias current  $I = -20 \,\mu$ A at room temperature. (c) The magnitude of the signal  $\Delta V_{nl}$  as a function of  $V_g$  (top panel). The  $V_g$  dependence of the graphene-TaTe<sub>2</sub> heterostructure channel resistance (bottom panel) along with the Fermi-level position in the graphene band structure.

To compare  $\Delta V_{nl}$  with the heterostructure channel properties, the gate dependence of the graphene-TaTe<sub>2</sub> heterostructure channel resistance is presented in the bottom panel of Fig. 3(c), where the charge neutrality point ( $V_{CNP}$ ) is ~10 V with field-effect mobility of about 1074 cm<sup>2</sup> V<sup>-1</sup> s<sup>-1</sup>. Similar gate dependence of the spin-switch signal is observed for device 1 (see Appendix G). These results rule out any contribution of the local Hall effect produced by the local fringe field from the FM detector contact edges on the graphene, since this effect should change its sign with carrier types in graphene [41].

Next, the electrical-bias-controlled switching of the spin injection from  $TaTe_2$  was measured. Figure 4(a) shows the spin-switch signal  $(V_{nl})$  with injection bias currents of I = $+/-2\mu A$  at  $V_g = 20$  V. Reversing the current direction creates opposite spin polarization in TaTe2 and hence accumulation of opposite spins in the graphene channel, which results in the opposite direction of the hysteretic behavior of the measured spin-switch signal. A full bias-dependent measurement was carried out at larger biases to understand the detailed energy dependence of the spin injection mechanism at the graphene-TaTe<sub>2</sub> junction [see Figs. 4(b) and 4(c)]. In the -I range, we observed an increase of  $\Delta V_{nl}$  with bias currents, where the amplitude of the spin-to-charge conversion signal is about  $\Delta R_{nl} = 300 \text{m}\Omega$ . However, for the +*I*, the  $\Delta V_{nl}$  increases linearly for low biases up to  $I = 8 \,\mu A$ and disappears at larger bias within the measurement noise. One of the reasons for such bias dependence can be ascribed to the energy dependence of spin polarization in TaTe<sub>2</sub> or the availability of spin-polarized density of states near the Fermi level in TaTe<sub>2</sub>. In contrast to local detection of the spin signal [42,43], as the measurements were performed in the NL geometry without any bias across the detector FM contact, any effects originating from the detector can be ruled out. This asymmetric feature of the bias-dependent spin injection effect in graphene-TaTe<sub>2</sub> heterostructure can be correlated to an electronic diode, where instead of controlling the charge current with bias in the conventional diode we control the spin injection in the heterostructure. To further examine the asymmetry of the bias dependence, we also checked the bias dependence of the graphene-TaTe<sub>2</sub> injector interface resistance ( $R_{int}$  [see Fig. 4(d)]. Interestingly,  $R_{int}$ is observed to be asymmetric with bias, which can be the origin of the asymmetry in the spin injection signal due to a conductivity mismatch issue between the TaTe<sub>2</sub> spin source and the graphene channel [35,44].

We discuss the possible mechanisms that engender the generation of current-induced in-plane spin polarization in the TaTe<sub>2</sub> in VSI geometry. First, like the ISHE measurements, we can also explain the VSI signal in TaTe<sub>2</sub> using the symmetry principle [see Appendix H, Figs. 12(a)-12(d)], that the generated field or spin (s) is perpendicular to the applied charge current  $(I_c)$  and spin current  $(I_s)$ . As the TaTe<sub>2</sub> crystals have a relatively smaller thickness (70 nm) compared to their lateral width (2.6-6 µm), as well as a low resistance compared to the graphene and interface  $(R_{int})$ , this makes the electric field within TaTe<sub>2</sub> predominantly in plane. Moreover, considering the in-plane spin injection in graphene, the origin of spin polarization can be attributed to the bulk SHE of TaTe<sub>2</sub> due to an in-plane electric field in TaTe<sub>2</sub>. Similarly, the surface or bulk states of TaTe<sub>2</sub>, if present and spin polarized, can also inject in-plane spin into graphene due to the EE [13] (see Fig. 12). If we consider the lower symmetry of the TaTe<sub>2</sub> crystals [40], both SHE and EE can also give rise to in-plane spin polarization in the TaTe2, as it does not have to follow the orthogonal rule between s,  $I_c$ , and  $I_s$  [45]. On the other hand, proximity-induced SHE in graphene can be excluded as an origin of the signals, as it is expected to align spin in the out-of-plane direction [16–18,46]. Moreover, with the lack of gate-dependent sign change behavior of the spin-switching signal for electron and hole transport regimes, the contribution of proximity-induced REE in graphene can be ruled out [15,16,47–49]. Any contribution of spin polarization from the Te layer can also be ruled out because equal and opposite contributions from both Te surfaces within a single TaTe<sub>2</sub> layer will be canceled out [50].

#### **III. CONCLUSION**

In summary, we demonstrated efficient charge-spin conversion in semimetal  $TaTe_2$  as an efficient spin injector in graphene-based spintronic devices at room temperature by using a vertical spin injection geometry. The advantage of using such a spin source is that the spin polarization direction can be controlled by an electrical bias instead of the magnetic field as conventionally achieved with ferromagnetic materials. Systematic bias and gate-dependent measurements of the spin injection signal indicate that the origin of spin polarization can be mainly because of the SHE or EE in TaTe<sub>2</sub> considering symmetric or low-symmetric spin-charge



FIG. 4. Electrical control and bias dependence of spin injection signal from TaTe<sub>2</sub>. (a) NL spin-switch signals ( $V_{nl}$ ) with bias currents of  $I = + / - 2\mu A$  at  $V_g = 20$  V at room temperature in device 3. (b) The  $V_{nl}$  at different bias currents, measured at room temperature at  $V_g = 20$  V. The data are vertically shifted for clarity. (c) Bias dependence of the magnitude of the signal  $\Delta V_{nl}$ . Insets: Schematic of the nonequilibrium Fermi-level positions for positive and negative applied bias conditions. (d) Graphene-TaTe<sub>2</sub> interface resistance ( $R_{int}$ ) characteristics with bias current (I) measured in four-terminal geometry (inset).

conversion processes. Such a nonmagnetic spin source based on the charge-spin conversion effect shows great potential to replace the ferromagnetic injector in all-electrical 2D spintronic circuits and is suitable for spin-orbit technologies.

#### ACKNOWLEDGMENTS

The authors acknowledge financial support from the European Union's Horizon 2020 Graphene Flagship Core 2 no. 785219 and Core3 no. 881603, 2DTech Vinnova Competence center at Chalmers, the EU FlagEra project (VR No. 2015-06813), and the Swedish Research Council VR project (Grant No. 2016-03658). We acknowledge the discussions and help of Bing Zhao in our research group.

# APPENDIX A: FABRICATION AND CHARACTERIZATION OF GRAPHENE-TaTe<sub>2</sub> HETEROSTRUCTURE

To fabricate graphene-TaTe<sub>2</sub> heterostructure spintronic devices, first a few layer graphene were mechanically exfoliated from highly ordered pyrolytic graphite (HOPG) onto SiO<sub>2</sub> (300 nm)/*n*-doped Si substrate using the Scotch tape method. Later, TaTe<sub>2</sub> flakes were exfoliated from bulk crystal (from Hq Graphene) on polydimethylsiloxane (PDMS) film and dry transferred onto the few-layer graphene flake under a

microscope using a home-built micromanipulator-controlled transfer stage. Contacts to graphene and TaTe<sub>2</sub> were defined by electron-beam lithography, electron-beam evaporation, and a lift-off process. For the preparation of ferromagnetic tunnel contacts to graphene, a two-step deposition of 0.5 nm of Ti at less than  $3 \times 10^{-7}$  Torr and 30 min *in situ* oxidation at above 30 Torr was carried out, followed by 90 nm of Co deposition. The resistances of ferromagnetic tunnel contacts (TiO<sub>2</sub>/Co) on the graphene channel were in the range of 5–20 kΩs at room temperature. The spin injection measurements were performed in a vacuum cryostat with a magnetic field up to 0.8 T. All the measurements were performed at room temperature using a Keithley 6221 current source, a Keithley 2182A nanovoltmeter, and Keithley 2612B source meter for application of gate voltages.

Figures 5(a) and 5(b) show the optical microscopic picture and atomic force microscopic image of device 1 consisting of exfoliated TaTe<sub>2</sub>, graphene with Co/TiO<sub>2</sub> tunnel contacts. The widths of the TaTe<sub>2</sub> flake and graphene stripe are 2.6  $\mu$ m and 1.8  $\mu$ m, respectively. After bias-dependent measurements of the inverse spin Hall effect, the flake was damaged, presumably due to gate leakage. The thickness of the exfoliated TaTe<sub>2</sub> flake is about 70 nm, scanned along the dotted orange line in the atomic force microscopy image. Figures 5(c) and 5(d) present the optical micrographs of



FIG. 5. (a, b) Optical microscopic picture and atomic force microscopic image of device 1 consisting of exfoliated 70 nm TaTe<sub>2</sub>, graphene with Co/TiO<sub>2</sub> tunnel contacts. (c, d) Optical microscopic images of devices 2 and 3. The green color scale bar in the microscopic images is  $6 \,\mu\text{m}$ .

devices 2 and 3, respectively, prepared by similar fabrication procedures. Dotted lines in the optical micrographs are to highlight the exfoliated multilayer graphene region below the  $TaTe_2$  flake.

## APPENDIX B: ELECTRIC AND MAGNETOTRANSPORT PROPERTIES OF TaTe<sub>2</sub>

Figure 6(a) shows conventional four-terminal (4T) *I-V* characteristics of the TaTe<sub>2</sub> flake with channel resistance 120  $\Omega$  at room temperature (pink line) and 70  $\Omega$  at 25 K (green line). The resistivity ( $\rho = R_L^A$ ) of TaTe<sub>2</sub> is approximated to be ~300 × 10<sup>-8</sup>  $\Omega$ m at room temperature and is ~60 × 10<sup>-8</sup>  $\Omega$ m at 25 K. The TaTe<sub>2</sub> channel length (*L*) is 13  $\mu$ m and the width (*w*) is 11  $\mu$ m; the thickness of TaTe<sub>2</sub> is about 30 nm.

We examined the magnetoresistance (MR%) of TaTe<sub>2</sub> with angle dependence measurements up to  $\pm 0.8$  T from the per-

pendicular ( $\perp$ ) to in-plane field (ll) with respect to the device plane at 300 K with  $I = -150 \,\mu$ A, as shown in Fig. 6(b). Figure 6(c) presents a maximum MR% at different angles at 0.8 T with a cosine fit. It can be seen [from Figs. 6(b) and 6(c)] that MR% is maximum with the out-of-plane field (Angle = 0) but turns to a minimum with an in-plane field.

#### APPENDIX C: INTERFACIAL RESISTANCE OF GRAPHENE-TaTe<sub>2</sub> HETEROSTRUCTURE

The graphene-TaTe<sub>2</sub> interfacial resistance  $(R_{int})$  is estimated by two-terminal (2T) measurement geometry in device 1 [Fig. 7(a)]. At the start of the measurements, the 2T resistance was 35 k $\Omega$  [Fig. 7(b)], and the 4T  $R_{int}$  was about 16 k $\Omega$  [Fig. 4(d)]. In such a high- $R_{int}$  condition, the spin transport in graphene is not much affected (without spin absorption). Such high resistance, in the beginning, can be due to the larger van der Waals gap between graphene and TaTe<sub>2</sub>. After a few hours of measurement, we observed the 2T resistance to decrease and stabilize at  $\sim 5 \text{ k}\Omega$  [Fig. 7(b)]. The 4T measurements could not be performed in the lowinterface condition due to a lack of working contacts on both sides of the TaTe<sub>2</sub> flake. By considering the graphene channel and contact resistances, the low  $R_{int}$  is estimated to be less than 100  $\Omega$ . In this low- $R_{int}$  condition, we observed a drastic reduction of the spin transport signal in graphene due to spin absorption by TaTe<sub>2</sub> [presented in Fig. 1(e)]. The inverse spin Hall effect measurements were performed in low- $R_{int}$  and vertical spin injection measurements that were performed in high-R<sub>int</sub> conditions.

#### APPENDIX D: x-FIELD HANLE IN GRAPHENE-TaTe<sub>2</sub> HETEROSTRUCTURE

Figure 8(a) shows a schematic of the measurement setup for nonlocal xHanle, where the magnetic field is applied in the planner perpendicular direction (*x* axis) to the magnetic easy axis (*y* axis) of the FM contacts [51]. Figure 8(b) shows the measured xHanle spin signal in a condition where TaTe<sub>2</sub> does not absorb the spin significantly. It can be seen that the magnetization of the FM becomes saturated at  $\pm 0.4$  T along the *x* axis.



FIG. 6. Transport properties of TaTe<sub>2</sub>. (a) Four-terminal (4T) *I-V* characteristics of TaTe<sub>2</sub> along with the measurement geometry. (b) Angle-dependent measurements of MR% in TaTe<sub>2</sub> from perpendicular ( $\perp$ ) to the in-plane field (ll) at room temperature. (c) Maximum MR% at different angles at 0.8 T with cosine fit.



FIG. 7. Interfacial resistance in the graphene-TaTe<sub>2</sub> heterostructure. (a) Schematic of the 2T measurement geometry to measure *I-V* characteristics of the graphene-TaTe<sub>2</sub> heterostructure. (b) 2T *I-V* properties of the graphene-TaTe<sub>2</sub> heterostructure, where high resistance, 35 k $\Omega$  (orange line), and low resistance, 5 k $\Omega$  (blue line), stages are shown.



FIG. 8. *x*-field Hanle in graphene-TaTe<sub>2</sub> channel. (a) Schematic representation of the measurement setup to measure nonlocal xHanle, where the magnetic field is applied in the planner perpendicular direction (*x* axis) to the magnetic easy axis (*y* axis) of the FM electrodes. (b) Measured xHanle spin signal at  $I = -200 \,\mu\text{A}$  and  $V_g = 40 \,\text{V}$  at room temperature.

#### APPENDIX E: ESTIMATION OF SPIN HALL ANGLE

We have estimated the spin Hall angle ( $\theta_{SH}$ ) in TaTe<sub>2</sub> by using the spin diffusion model [10–12]. The manifested inverse spin Hall effect (ISHE) signal can be represented using the following equation:

$$\Delta R_{\rm ISHE} = \frac{2\theta_{\rm SH}\rho_{\rm TaTe_2}X}{w_{\rm TaTe_2}} \left(\frac{\hat{I}_s}{I_c}\right). \tag{E1}$$

Here,  $\Delta R_{\rm ISHE}$ ,  $\theta_{\rm SH}$ ,  $\rho_{\rm TaTe_2}$ , X, and  $w_{\rm TaTe_2}$  are the ISHE signal amplitude ( $\Delta R_{\rm ISHE} = \Delta V_{\rm ISHE}/I_{50\mu A} = 43 \,\mathrm{m\Omega}$ ), spin Hall angle, resistivity of TaTe<sub>2</sub> (300  $\mu\Omega$  cm), shunting factor (0.94), and width of TaTe<sub>2</sub> (2.6  $\mu$ m).  $\hat{I}_s$  and  $I_c$  are the effective spin current absorbed by TaTe<sub>2</sub> from the graphene channel due to the shunting effect and converted charge current due to ISHE in TaTe<sub>2</sub>. The shunt factor (X) is estimated by considering the spin signal is in the linear regime with bias current, and from the spin signal presented in Fig. 1(e),  $X = 1 - \frac{16}{272} = 0.94$ , which alludes to 94% spin absorption by TaTe<sub>2</sub>.

 $\frac{\hat{I}s}{I}$  can be written as below.

$$\begin{split} \frac{\hat{I}s}{I_c} &= \frac{\int_0^{t_M} I_s(z)dz}{I_c t_{\text{TaTe}_2}} \\ &= \frac{\lambda_{\text{TaTe}_2}}{t_{\text{TaTe}_2}} \frac{\left(1 - e^{-\frac{t_{\text{TaTe}_2}}{\lambda_{\text{TaTe}_2}}}\right)^2}{1 - e^{-2\frac{t_{\text{TaTe}_2}}{\lambda_{\text{TaTe}_2}}}} \frac{I_s(z=0)}{I_c} \\ &= \frac{\lambda_{\text{TaTe}_2}}{t_{\text{TaTe}_2}} \frac{\left(1 - e^{-\frac{t_{\text{TaTe}_2}}{\lambda_{\text{TaTe}_2}}}\right)^2}{1 - e^{-2\frac{t_{\text{TaTe}_2}}{\lambda_{\text{TaTe}_2}}}} \\ &\times \frac{2p_i Q_{IF1} \left[ (2Q_{IF2} + 1)(1 - Q_{IM})e^{-\frac{t_{\text{SH}}}{\lambda_{gr}}} - (1 + Q_{IM})e^{-\frac{3I_{\text{SH}}}{2\lambda_{gr}}} \right]}{\left(2Q_{IF1} + 1\right)(2Q_{IF2} + 1)(1 + Q_{IM}) - (2Q_{IF1} + 1)(1 + Q_{IM})e^{-\frac{3I_{\text{SH}}}{\lambda_{gr}}} - (2Q_{IF2} + 1)(1 - Q_{IM})e^{-\frac{-t_{\text{SH}}}{\lambda_{gr}}}} \right]. \end{split}$$

Here,  $R_{gr} = \frac{R_{gr}\lambda_{gr}}{w_{gr}}$ ,  $Q_{IFi} = \frac{1}{1-p^2}\frac{R_{ci}}{R_{gr}}$ , with  $I = 1, 2, R_{ci}$  corresponding to injector and detector contact resistances, respectively.  $Q_{IM} = \frac{w_{gr}R_{ini}}{R_{gr}\lambda_{gr}}$ ,  $R_{int}$ ,  $w_gr$ ,  $R_{gr}$  are the TaTe<sub>2</sub>-

graphene interface resistance, graphene channel width, and square resistance, respectively, and are presented in Table I. As the spin diffusion length in  $TaTe_2$  is an unknown parameter, we estimated spin Hall angle ( $\theta_{SH}$ ) as a function of spin

TABLE I. Parameters of graphene-TaTe<sub>2</sub> for device 3 to estimate the spin Hall angle in TaTe<sub>2</sub> to solve Eq. (E1).

	$D_s$ (m <sup>2</sup> /s)	$\tau_s$ (ps)	$\lambda_{Gr}$ ( $\mu$ m)	$w_{Gr}$ ( $\mu$ m)	$L_{ m SH}$ ( $\mu$ m)	t <sub>TaTe2</sub> (nm)	w <sub>TaTe2</sub> (μm)	$egin{array}{c} R_{sq,Gr} \ (\Omega) \end{array}$	$ ho_{\mathrm{TaTe}_2}$ ( $\mu\Omega$ cm)	$R_{c1}$ (k $\Omega$ )	$R_{c2}$ (k $\Omega$ )
Device 1	0.021	311	2.56	1.8	2.5	70	2.6	339	300	20	15



FIG. 9. Estimation of the spin Hall angle ( $\theta_{SH}$ ) in TaTe<sub>2</sub>. (a) Spin Hall angle ( $\theta_{SH}$ ) as a function of spin diffusion length in TaTe<sub>2</sub> ( $\lambda_{TaTe_2}$ ) considering  $R_{int} = 20 \Omega$ . (b) IEE length ( $\lambda_{IEE}$ ) as a function of  $\lambda_{TaTe_2}$ . (c) Spin Hall angle ( $\theta_{SH}$ ) as a function of  $R_{int}$  considering spin diffusion length  $\lambda_{TaTe_2} = 30 \text{ nm}$ .

diffusion length ( $\lambda_{TaTe_2}$ ) in TaTe<sub>2</sub> by assuming  $R_{int} = 20 \Omega$ [shown in Fig. 9(a)]. The spin Hall angle  $\theta_{SH}$  decays exponentially with increasing  $\lambda_{TaTe_2}$  according to Eq. (E1) and its simplified solution. We found  $\theta_{SH}$  is 0.59 at  $\lambda_{TaTe_2} = 10$  nm; 0.23 at  $\lambda_{TaTe_2} = 30$  nm, and becomes saturated with 0.20. We can also approximately calculate the inverse Edelstein effect (IEE) of TaTe<sub>2</sub> through IEE length  $\lambda_{IEE} = \theta_{SH} * \lambda_{TaTe_2}$ , and we found  $\lambda_{IEE} \approx 6-60$  nm [see Fig. 9(b)] as a function of  $\lambda_{TaTe_2}$  (10–110 nm) by considering  $R_{int} = 20 \Omega$ .

Overall, the calculated range of  $\theta_{\text{SH}}$  for TaTe<sub>2</sub> seems to be consistent with Pt ( $\theta_{SH,Pt} \approx 0.23$ ) [10,11], and other TMDs such as MoTe<sub>2</sub> ( $\theta_{SH,MoTe2} \approx 0.2$ ) [38], MoS<sub>2</sub> ( $\theta_{SH,MoS2} \approx 0.1$ ) [17], but one order of magnitude higher compared to the lower limit of the  $\theta_{SH,WTe2}$  for WTe<sub>2</sub> ( $\theta_{SH,WTe2} = 0.01$ ) [12]. Interestingly, the estimated values of  $\lambda_{\text{IEE}}$  are comparable to topological insulators [52] (2.1 nm), oxide interfaces [53] (6.4 nm), but it seems to be one order of magnitude higher than in heavy metals (0.1–0.4 nm) and other Rashba systems [33,54]. Figure 9(c) depicts  $\theta_{\text{SH}}$  as a function of  $R_{\text{int}}$  by considering spin diffusion length in TaTe<sub>2</sub>,  $\lambda_{\text{TaTe}_2} = 30$  nm at room temperature. The spin Hall angle,  $\theta_{\text{SH}}$ , shows a linear dependence on  $R_{\text{int}}$  and ranges from 0.22 to 0.29 for  $R_{\text{int}} = 1-100 \Omega$ .

## APPENDIX F: SPIN TRANSPORT AND SPIN POLARIZATION IN GRAPHENE-TaTe<sub>2</sub> HETEROSTRUCTURE

We characterized the spin transport properties in the graphene-TaTe<sub>2</sub> channel (without spin absorption) to estimate the spin polarization of the detector FM contact. Figure 10 (a) shows the sketch of the measurement geometry for NL Hanle measurement, where the magnetic field is applied in the out-of-plane direction. The measured spin precession signal is fitted with the following Hanle precession Eq. (F1) to approximate the spin diffusion constant ( $D_s$ ), spin diffusion length ( $\lambda_s$ ), and lifetime ( $\tau_s$ ):

$$\Delta R_{\rm NL} \propto \int_0^\infty \frac{1}{\sqrt{4\pi Dt}} e^{-\frac{L^2}{4D_{\rm S}t}} \cos \left(\omega_L t\right) e^{\frac{-t}{\tau_{\rm S}}} dt. \qquad ({\rm F1})$$

Furthermore, the measured Hanle spin signal (purple circles) along with its fit (magenta line) to Eq. (F2) is shown in Fig. 10(b) to extract  $TiO_2/Co$  contact polarization:

$$\Delta R_{\rm NL} = \pm \frac{P_i P_d R_{sq} D_s}{W} \operatorname{Re} \left\{ \frac{1}{2D_s} \frac{e^{-L\sqrt{\lambda_s^{-2} - i\frac{\omega}{D_s}}}}{\sqrt{\tau_s^{-1} - i\omega}} \right\}.$$
 (F2)

Here,  $\omega$  depends on the frequency of spin flips and Larmor precession,  $\omega_L = \frac{g\mu_B}{\hbar}B_{\perp}$  is the Larmor frequency, where  $B_{\perp}$ is an applied perpendicular magnetic field to the easy axis of the FM contacts, g = 2 is assumed. In equation (F1) and (F2),  $D_s$  is the spin diffusion constant, and  $\lambda_s$  (2.56 ± 0.14  $\mu$ m) and  $\tau_s$  (311 ± 16 ps) are the spin diffusion length and spin lifetime, respectively. In device 1,  $R_{sq} = 339 \Omega$  for multilayer exfoliated graphene at  $V_g = 40$  V on Si/SiO<sub>2</sub> substrate. The distance between spin injection and spin detection electrodes is  $L = 4.5 \,\mu$ m, graphene width,  $w = 1.8 \,\mu$ m. Considering the spin polarization of the injector and detector to be equal ( $P_i = P_d$ ),  $P_{\text{Co/TiO2}}$  is estimated to be about 6.13% ± 0.9%.

To estimate the current-induced spin polarization in TaTe<sub>2</sub> in device 2, we have also used the same equation, Eq. (F2), to fit the Hanle signal, as shown in Fig. 2(f). In device 2, using the graphene channel resistance,  $R_{sq} = 1425 \Omega$ , width



FIG. 10. Spin transport in the graphene-TaTe<sub>2</sub> heterostructure. (a) Sketch of the measurement setup for nonlocal Hanle measurement. (b) Hanle spin precession signal (violate circles) without spin absorption by TaTe<sub>2</sub> at back-gate voltage  $V_g = 40$  V with bias current  $I = -30 \,\mu$ A along with the fitted line (magenta line) to Eq. (F2) to estimate the spin polarization of FM contacts.



FIG. 11. Spin injection from TaTe<sub>2</sub> into graphene and spinswitch signal in device 1. (a, b) Spin injection from TaTe<sub>2</sub> into graphene for the electron- and hole-doped regimes at gate voltages  $V_g = 40$  and -50 V with I = -40 and  $-30 \mu$ A, respectively.

 $w = 1.2 \,\mu \text{m}$ , we found that

$$\begin{split} /P_{\text{TaTe}_2} P_{\text{Co/TiO2}} &= 0.0287 \pm 0.0127 \\ P_{\text{TaTe}_2} &= \frac{0.0287^2}{P_{\text{Co/TiO2}}} \pm \frac{0.0127^2}{P_{\text{Co/TiO2}}} \\ &= 0.0134 \pm 0.0026 \,. \end{split}$$

Now, using previously obtained  $P_{\text{Co/TiO2}} = 6.13\%$ , the spin polarization is of TaTe<sub>2</sub> is approximated to be  $P_{\text{TaTe}_2} = 1.34\% \pm 0.26\%$ .

# APPENDIX G: SPIN INJECTION FROM TaTe<sub>2</sub> INTO GRAPHENE IN DEVICE 1

The vertical spin injection results were reproducibly observed at room temperature in three different devices fabricated with similar methods, as presented in this manuscript. Figure 11 shows the spin injection from TaTe<sub>2</sub> into graphene and spinswitch signal in device 1. The possibility of spin transport contribution from the reference FM electrodes on the graphene channel in the injector and detector circuit can be ruled out, as it would have resulted in multiple switches in the measured signal (see Fig. 1(e), where both FM injector and detector contacts are involved in the conventional spin-valve measurements).

#### APPENDIX H: CURRENT-INDUCED SPIN POLARIZATION IN SYMMETRIC AND LOW-SYMMETRIC CRYSTALS

Here we discuss possible origins of in-plane spin polarization in TaTe<sub>2</sub> considering the crystal structure of TaTe<sub>2</sub>. 1T'-TaTe<sub>2</sub> is a centrosymmetric layered material at room temperature where spin current ( $I_s$ ), charge current ( $I_c$ ), and spin polarization (s) are perpendicular to each other for symmetric measurements of the spin Hall effect (SHE) or Edelstein effect (EE). To facilitate our discussion, three-dimensional (3D) axis directions are presented in Fig. 12(a), similar to the schematics shown in Figs. 2(a) and 2(d). Conventional ISHE in the symmetric 3D crystal of TaTe<sub>2</sub> is shown in Fig. 12(b), where  $I_s$ ,  $I_c$ , and s are perpendicular to each other in the z, y, and x directions, respectively, and can explain the measurements depicted in Figs. 2(a)–2(c).

Interestingly, in-plane spin polarization along the y axis in TaTe<sub>2</sub>, presented in Figs. 2(d)–2(f), is possible while injecting charge current ( $I_c$ ) can be along the z axis [Fig. 12(c)] and x axis [Fig. 12(d)], which needs to be perpendicular to the corresponding spin current ( $I_s$ ) to maintain symmetry. These symmetries allow bulk and surface states to inject spin from bulk (via SHE) or surface states (via Edelstein effect) of TaTe<sub>2</sub>. While charge current ( $I_c$ ) and spin current ( $I_s$ ) are along the z axis and x axis, respectively, the spins are accumulated at the edges of TaTe<sub>2</sub> [Fig. 12(c)]. However, the TaTe<sub>2</sub> crystals have a relatively smaller thickness (70–100 nm) and



FIG. 12. (a) The x, y, and z-axis directions similar to the schematics shown in Figs. 2(a) and 2(d) to explain three-dimensional crystal symmetry. (b) Spin to charge conversion due to inverse spin Hall effect (ISHE), shown in Figs. 2(a)–2(c) in the high-symmetry crystal structure, where spin absorption in TaTe<sub>2</sub> renders z-direction spin current ( $I_{s,z}$ ) with the spin direction along the x axis ( $s_x$ ) and measured charge current in the y axis ( $I_{c,y}$ ). (c) Spin polarization in symmetric TaTe<sub>2</sub> to explain the spin-switch measurements shown in Figs. 2(d)–2(f). In this case, injected spin current in graphene is along the y direction, and keeping this in mind, we have two possible scenarios. First, charge current and spin current are out of plane (along the z axis) and in plane (along the x axis), respectively, and spins are accumulated at the edges of TaTe<sub>2</sub>. In the second scenario, shown in (d), where spin along the y direction is accumulated at the top and bottom surface of TaTe<sub>2</sub> and charge current and spin current are in x and z direction. (e, f) Spin polarization and spin current are parallel and along the y axis in lower symmetry situations but perpendicular to the charge current, which can be in the z or x axis, respectively.

negligible edge area in contact with graphene compared to the bottom surface area. Hence, in-plane (y-axis) spin injection into graphene from the accumulated spins at the TaTe<sub>2</sub> edges is improbable. On the other hand, considering charge current ( $I_c$ ) and spin current ( $I_s$ ) are along the x and z axis [Fig. 12(d)], where spin along the y axis can accumulate at the top and bottom surface of TaTe<sub>2</sub>, this can explain our measurements of spin polarization in symmetric TaTe<sub>2</sub> crystal with lateral widths of 2.6–6 µm in contact with graphene and nanometer-range thickness.

- W. Han, R. K. Kawakami, M. Gmitra, and J. Fabian, Graphene spintronics, Nat. Nanotechnol. 9, 794 (2014).
- [2] I. Žutić, J. Fabian, and S. Das Sarma, Spintronics: Fundamentals and applications, Rev. Mod. Phys. 76, 323 (2004).
- [3] D. D. Awschalom and M. E. Flatté, Challenges for semiconductor spintronics, Nat. Phys. 3, 153 (2007).
- [4] A. Fert, Origin, development, and future of spintronics (Nobel lecture), Angew Chemie Int. Ed. 47, 5956 (2008).
- [5] A. Manchon, J. Železný, I. M. Miron, T. Jungwirth, J. Sinova, A. Thiaville, K. Garello, and P. Gambardella, Current-induced spin-orbit torques in ferromagnetic and antiferromagnetic systems, Rev. Mod. Phys. 91, 035004 (2019).
- [6] K. Garello, F. Yasin, H. Hody, S. Couet, L. Souriau, S. H. Sharifi, J. Swerts, R. Carpenter, S. Rao, W. Kim *et al.*, Manufacturable 300 mm platform solution for Field-Free Switching SOT-MRAM, 2019 *Symposium on VLSI Technology* (Kyoto, Japan, 2019), pp. T194-T195.
- [7] J. Sinova, S. O. Valenzuela, J. Wunderlich, C. H. Back, and T. Jungwirth, Spin Hall effects, Rev. Mod. Phys. 87, 1213 (2015).
- [8] A. Manchon, H. C. Koo, J. Nitta, S. M. Frolov, and R. A. Duine, New perspectives for Rashba spin-orbit coupling, Nat. Mater. 14, 871 (2015).
- [9] J. C. Rojas-Sánchez and A. Fert, Compared Efficiencies of Conversions between Charge and Spin Current by Spin-Orbit Interactions in Two- and Three-Dimensional Systems, Phys. Rev. Appl. 11, 054049 (2019).
- [10] W. S. Torres, J. F. Sierra, L. A. Benítez, F. Bonell, M. V. Costache, and S. O. Valenzuela, Spin precession and spin Hall effect in monolayer graphene/Pt nanostructures, 2D Mater. 4, 041008 (2017).
- [11] W. Yan, E. Sagasta, M. Ribeiro, Y. Niimi, L. E. Hueso, and F. Casanova, Large room temperature spin-to-charge conversion signals in a few-layer graphene/Pt lateral heterostructure, Nat. Commun. 8, 661 (2017).
- [12] B. Zhao, D. Khokhriakov, Y. Zhang, H. Fu, B. Karpiak, A. Md. Hoque, X. Xu, Y. Jiang, B. Yan, and S. P. Dash, Observation of charge to spin conversion in Weyl semimetal WTe2 at room temperature, Phys. Rev. Res. 2, 013286 (2020).
- [13] B. Zhao, B. Karpiak, D. Khokhriakov, A. M. Hoque, X. Xu, Y. Jiang, and S. P. Dash, Edelstein effect in type-II Weyl semimetal WTe2 up to room temperature, arXiv:1910.06225.
- [14] Z. Kovács-Krausz, A. M. Hoque, P. Makk, B. Szentpéteri, M. Kocsis, B. Fülöp, M. V. Yakushev, T. V. Kuznetsova, O. E. Tereshchenko, K. A. Kokh *et al.*, Electrically controlled spin injection from giant Rashba spin-orbit conductor BiTeBr, Nano Lett. **20**, 4782 (2020).

However, in-plane spin polarization can also emerge in unconventional and lower symmetric TaTe<sub>2</sub> crystal [see Figs. 12(e) and 12(f)], where spin current and spin orientation can be parallel to each other and along the y axis ( $s \parallel I_s$ ) but are perpendicular to the charge current ( $I_c$ ), which could be in either z or x direction in our measurement geometry [45]. Again, considering k-dependent spin polarization and the lateral width of TaTe<sub>2</sub>, which is considerably higher than the thickness, the latter situation is more likely to emanate in such a lower symmetric crystal because of SHE or EE.

- [15] L. Li, J. Zhang, G. Myeong, W. Shin, H. Lim, B. Kim, S. Kim, T. Jin, S. Cavill, B. S. Kim *et al.*, Gate-tunable reversible Rashba-Edelstein effect in a few-layer graphene/2H-TaS<sub>2</sub> heterostructure at room temperature, ACS Nano 14, 5251 (2020).
- [16] T. S. Ghiasi, A. A. Kaverzin, P. J. Blah, and B. J. van Wees, Charge-to-spin conversion by the Rashba-Edelstein effect in two-dimensional van der Waals heterostructures up to room temperature, Nano Lett. 19, 5959 (2019).
- [17] C. K. Safeer, J. Ingla-Aynés, F. Herling, J. H. Garcia, M. Vila, N. Ontoso, M. R. Calvo, S. Roche, L. E. Hueso, and F. Casanova, Room-temperature spin Hall effect in graphene/MoS<sub>2</sub> van der Waals heterostructures, Nano Lett. **19**, 1074 (2019).
- [18] D. Khokhriakov, A. W. Cummings, K. Song, M. Vila, B. Karpiak, A. Dankert, S. Roche, and S. P. Dash, Tailoring emergent spin phenomena in Dirac material heterostructures, Sci. Adv. 4, eaat9349 (2018).
- [19] C. H. Li, O. M. J. van't Erve, J. T. Robinson, Y. Liu, L. Li, and B. T. Jonker, Electrical detection of charge-current-induced spin polarization due to spin-momentum locking in Bi<sub>2</sub>Se<sub>3</sub>, Nat. Nanotechnol. 9, 218 (2014).
- [20] A. Dankert, J. Geurs, M. V. Kamalakar, S. Charpentier, and S. P. Dash, Room temperature electrical detection of spin polarized currents in topological insulators, Nano Lett. 15, 7976 (2015).
- [21] A. Dankert, P. Bhaskar, D. Khokhriakov, I. H. Rodrigues, B. Karpiak, M. V. Kamalakar, S. Charpentier, I. Garate, and S. P. Dash, Origin and evolution of surface spin current in topological insulators, Phys. Rev. B 97, 125414 (2018).
- [22] N. H. D. Khang, Y. Ueda, and P. N. Hai, A conductive topological insulator with large spin Hall effect for ultralow power spin-orbit torque switching, Nat. Mater. 17, 808 (2018).
- [23] A. R. Mellnik, J. S. Lee, A. Richardella, J. L. Grab, P. J. Mintun, M. H. Fischer, A. Vaezi, A. Manchon, E. A. Kim, N. Samarth, and D. C. Ralph, Spin-transfer torque generated by a topological insulator, Nature (London) 511, 449 (2014).
- [24] D. C. Manendra, R. Grassi, J.-Y. Chen, M. Jamali, D. R. Hickey, D. Zhang, Z. Zhao, H. Li, P. Quarterman, Y. Lv, M. Li, A. Manchon, K. A. Mkhoyan, T. Low, and J.-P. Wang, Roomtemperature high spin-orbit torque due to quantum confinement in sputtered Bi<sub>x</sub>Se<sub>(1-x)</sub> films, Nat. Mater. **17**, 800 (2018).
- [25] J. A. Voerman, C. Li, Y. Huang, and A. Brinkman, Spinmomentum locking in the gate tunable topological insulator BiSbTeSe<sub>2</sub> in non-local transport measurements, Adv. Electron. Mater. 5, 1900334 (2019).
- [26] K. Vaklinova, A. Hoyer, M. Burghard, and K. Kern, Currentinduced spin polarization in topological insulator-graphene heterostructures, Nano Lett. 16, 2595 (2016).

- [27] B. Zhao, D. Khokhriakov, B. Karpiak, A. M. Hoque, L. Xu, L. Shen, Y. P. Feng, X. Xu, Y. Jiang, and S. P. Dash, Electrically controlled spin-switch and evolution of Hanle spin precession in graphene, 2D Mater. 6, 035042 (2019).
- [28] A. H. Castro Neto and F. Guinea, Impurity-Induced Spin-Orbit Coupling in Graphene, Phys. Rev. Lett. 103, 026804 (2009).
- [29] B. Karpiak, A. W. Cummings, K. Zollner, M. Vila, D. Khokhriakov, A. M. Hoque, A. Dankert, P. Svedlindh, J. Fabian, S. Roche, and S. P. Dash, Magnetic proximity in a van der Waals heterostructure of magnetic insulator and graphene, 2D Mater. 7, 015026 (2020).
- [30] S. Manipatruni, D. E. Nikonov, C.-C. Lin, T. A. Gosavi, H. Liu, B. Prasad, Y.-L. Huang, E. Bonturim, R. Ramesh, and I. A. Young, Scalable energy-efficient magnetoelectric spin-orbit logic, Nature (London) 565, 35 (2019).
- [31] J. J. Gao, J. G. Si, X. Luo, J. Yan, F. C. Chen, G. T. Lin, L. Hu, R. R. Zhang, P. Tong, W. H. Song, X. B. Zhu, W. J. Lu, and Y. P. Sun, Origin of the structural phase transition in single-crystal TaTe<sub>2</sub>, Phys. Rev. B **98**, 224104 (2018).
- [32] M. Milletarì, M. Offidani, A. Ferreira, and R. Raimondi, Covariant Conservation Laws and the Spin Hall Effect in Dirac-Rashba Systems, Phys. Rev. Lett. 119, 246801 (2017).
- [33] J. C. R. Sánchez, L. Vila, G. Desfonds, S. Gambarelli, J. P. Attané, J. M. De Teresa, C. Magén, and A. Fert, Spin-to-charge conversion using Rashba coupling at the interface between nonmagnetic materials, Nat. Commun. 4, 2944 (2013).
- [34] M. Offidani, M. Milletari, R. Raimondi, and A. Ferreira, Optimal Charge-to-Spin Conversion in Graphene on Transition-Metal Dichalcogenides, Phys. Rev. Lett. 119, 196801 (2017).
- [35] M. V. Kamalakar, A. Dankert, P. J. Kelly, and S. P. Dash, Inversion of spin signal and spin filtering in ferromagnet|hexagonal boron nitride-graphene van der Waals heterostructures, Sci. Rep. 6, 21168 (2016).
- [36] D. Khokhriakov, B. Karpiak, A. M. Hoque, and S. P. Dash, Two-dimensional spintronic circuit architectures on large scale graphene, Carbon 161, 892 (2020).
- [37] J. Zhang, B. Yang, H. Zheng, X. Han, and Y. Yan, Large magnetic anisotropy and strain induced enhancement of magnetic anisotropy in monolayer TaTe<sub>2</sub>, Phys. Chem. Chem. Phys. **19**, 24341 (2017).
- [38] C. K. Safeer, N. Ontoso, J. Ingla-Aynés, F. Herling, V. T. Pham, A. Kurzmann, K. Ensslin, A. Chuvilin, I. Robredo, M. G. Vergniory *et al.*, Large multidirectional spin-to-charge conversion in low-symmetry semimetal MoTe<sub>2</sub> at room temperature, Nano Lett. **19**, 8758 (2019).
- [39] S. O. Valenzuela and M. Tinkham, Direct electronic measurement of the spin Hall effect, Nature (London) 442, 176 (2006).
- [40] G. M. Stiehl, D. MacNeill, N. Sivadas, I. El Baggari, M. H. Guimarães, N. D. Reynolds, L. F. Kourkoutis, C. J. Fennie, R. A. Buhrman, and D. C. Ralph, Current-induced torques with Dresselhaus symmetry due to resistance anisotropy in 2D materials, ACS nano 13, 2599 (2019).

- [41] B. Karpiak, A. Dankert, A. W. Cummings, S. R. Power, S. Roche, and S. P. Dash, 1D ferromagnetic edge contacts to 2D graphene/h-BN heterostructures, 2D Mater. 5, 014001 (2018).
- [42] R. S. Patel, S. P. Dash, M. P. De Jong, and R. Jansen, Magnetic tunnel contacts to silicon with low-work-function ytterbium nanolayers, J. Appl. Phys. **106**, 016107 (2009).
- [43] S. Sharma, S. P. Dash, H. Saito, S. Yuasa, B. J. van Wees, and R. Jansen, Anisotropy of spin polarization and spin accumulation in Si/Al<sub>2</sub>O<sub>3</sub>/ferromagnet tunnel devices, Phys. Rev. B 86, 165308 (2012).
- [44] A. Fert and H. Jaffrès, Conditions for efficient spin injection from a ferromagnetic metal into a semiconductor, Phys. Rev. B 64, 184420 (2001).
- [45] D. Culcer and R. Winkler, Generation of Spin Currents and Spin Densities in Systems with Reduced Symmetry, Phys. Rev. Lett. 99, 226601 (2007).
- [46] A. Dankert and S. P. Dash, Electrical gate control of spin current in van der Waals heterostructures at room temperature, Nat. Commun. 8, 16093 (2017).
- [47] D. Khokhriakov, A. M. Hoque, B. Karpiak, and S. P. Dash, Gate-tunable spin-galvanic effect in graphene-topological insulator van der Waals heterostructures at room temperature, Nature Commun. 11, 1 (2020).
- [48] A. M. Hoque, D. Khokhriakov, B. Karpiak, and S. P. Dash, All-electrical creation and control of giant spin-galvanic effect in 1T-MoTe<sub>2</sub>/graphene heterostructures at room temperature, arXiv:1908.09367v2.
- [49] L. A. Benítez, W. S. Torres, J. F. Sierra, M. Timmermans, J. H. Garcia, S. Roche, M. V. Costache, and S. O. Valenzuela, Tunable room-temperature spin galvanic and spin Hall effects in van der Waals heterostructures, Nat. Mater. 19, 170 (2020).
- [50] X. Zhang, Q. Liu, J.-W. Luo, A. J. Freeman, and A. Zunger, Hidden spin polarization in inversion-symmetric bulk crystals, Nat. Phys. 10, 387 (2014).
- [51] J.-C. Rojas Sánchez, P. Laczkowski, W. F. Savero Torres, M. Cubukcu, V. D. Nguyen, L. Notin, C. Beigné, C. Vergnaud, A. Marty, M. Jamet, L. Vila, and J. P. Attané, In-plane and out-of-plane spin precession in lateral spin-valves, Appl. Phys. Lett. 102, 132408 (2013).
- [52] J.-C. Rojas-Sánchez, S. Oyarzún, Y. Fu, A. Marty, C. Vergnaud, S. Gambarelli, L. Vila, M. Jamet, Y. Ohtsubo, A. Taleb-Ibrahimi *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).
- [53] E. Lesne, Y. Fu, S. Oyarzun, J. C. Rojas-Sánchez, D. C. Vaz, H. Naganuma, G. Sicoli, J.-P. Attané, M. Jamet, E. Jacquet *et al.*, Highly efficient and tunable spin-to-charge conversion through Rashba coupling at oxide interfaces, Nat. Mater. 15, 1261 (2016).
- [54] A. Nomura, T. Tashiro, H. Nakayama, and K. Ando, Temperature dependence of inverse Rashba-Edelstein effect at metallic interface, Appl. Phys. Lett. **106**, 212403 (2015).