Electrical Control of the Rashba-Edelstein Effect in a Graphene/2H-TaS$_2$ van der Waals Heterostructure at Room Temperature

Lijun Li$^1$, Jin Zhang$^2$, Gyuho Myeong$^1$, Wongil Shin$^1$, Hongsik Lim$^1$, Boram Kim$^1$, Seungho Kim$^1$, Taehyeok Jin$^1$, Bumseo Kim$^3$, Changyoung Kim$^3$, Johannes Lischner$^2$, Aires Ferreira$^4$*, Sungjae Cho$^1$*

Van der Waals heterostructures are prime candidates to explore interfacial spin-orbit-coupling (SOC) phenomena for both fundamental spintronics research and applications. Proximity-induced SOC and spin dynamics engineering have been recently achieved in graphene/semiconducting dichalcogenide bilayers. However, the emergence of spin-momentum-locked 2D Dirac fermions in a van der Walls material, pivotal for all-electrical control over the electron’s spin moment, has remained elusive. Here, we report current-induced spin polarization, a direct consequence of spin-momentum locking due to broken mirror symmetry, in a semimetallic graphene/2H-TaS$_2$ bilayer. Spin-sensitive electrical measurements unveil full spin polarization reversal by gate voltage (i.e., spin switching) at room temperature. The on-demand electrical generation and control of nonequilibrium spin polarization, not previously observed in a nonmagnetic heterointerface, is an elegant manifestation of unconventional 2D Dirac fermions with robust spin-helical structure. Our findings, supported by first-principles calculations, establish a new route to design low-power spin-logic circuits from layered materials.

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$^1$Department of Physics, Korean Advanced Institute of Science and Technology (KAIST), Daejeon, Korea  *email: sungjae.cho@kaist.ac.kr
$^2$Departments of Materials and Physics, Imperial College London, SW7 2AZ, United Kingdom
$^3$Department of Physics, Seoul National University, Seoul, Korea
$^4$Department of Physics, University of York, YO10 5DD, United Kingdom  
*email: aires.ferreira@york.ac.uk
The enhancement of spin-orbit effects at interfaces holds unique prospects for spin-logic technologies that can offer high-speed operation with reduced energy consumption\textsuperscript{1,2}. Graphene is considered a promising 2D material for next-generation spintronics, owing to micrometer-length spin transport\textsuperscript{3,4} and electrically tunable electronic structure\textsuperscript{5,6}, with potential for integration with Si-based technologies\textsuperscript{7}. Recently, the family of transition metal dichalcogenides (TMDs)—high-SOC layered crystals that cover a broad range of properties, from insulators to superconductors\textsuperscript{8}—has enlarged the breadth of accessible spin-orbit phenomena in graphene-based heterostructures, to include all-optical spin injection\textsuperscript{9,10} and proximity-induced SOC up to 1 meV\textsuperscript{11,12,13,14,15,20}, 20 times larger than graphene’s weak spin-orbit gap\textsuperscript{16}.\n
Microscopically, TMD substrates can induce different types of SOC in graphene. On the one hand, the enhancement of $\mathbf{z} \rightarrow -\mathbf{z}$ mirror-symmetric interactions (Kane-Mele SOC and spin-valley coupling\textsuperscript{14,15}) is predicted to induce spin Hall effect (SHE)\textsuperscript{11,12}, whereby an applied charge current $\mathbf{J}$ is partly converted into a transverse spin current $J_z^s = (\hbar/2e) \theta_{SH} \hat{\mathbf{z}} \times \mathbf{J}$, where the $\hat{\mathbf{z}}$-axis is normal to the 2D plane and $\theta_{SH}$ is the spin Hall angle\textsuperscript{2}. On the other hand, the interfacial breaking of inversion symmetry in 2D heterostructures containing heavy elements can induce a sizeable Bychkov-Rashba interaction\textsuperscript{1}. The emergence of $\mathbf{z} \rightarrow -\mathbf{z}$ asymmetric SOC (hereafter, referred to as Rashba SOC) is predicted to entangle spin and SU(2)-sublattice-pseudospin degrees of freedom, endowing 2D Dirac states in graphene with a Fermi-energy-dependent helical spin texture in momentum space, a van der Waals counterpart of spin-momentum-locked surface states in topological insulator thin films\textsuperscript{17,18}.

Interfacial states with spin-helical structure allow efficient charge-to-spin conversion via the Rashba-Edelstein effect (REE). In this phenomenon, an applied charge current magnetizes the 2D conduction electrons, generating a nonequilibrium spin density $\delta \mathbf{S} = (\hbar/2e) \beta_{EE} \hat{\mathbf{z}} \times \mathbf{J}$, where
\( \beta_{\text{EE}} \) is the microscopic Edelstein efficiency parameter\(^{18} \). REE has been observed in metallic bilayers\(^{19,20} \), oxide heterostructures\(^{21,22} \), and topological insulator/metal heterostructures\(^{23} \), but has not yet been observed in a van der Waals material. Discovery of a graphene-based heterostructure with sizeable Rashba SOC would open up novel opportunities to study and control relativistic SOC transport effects\(^{17,18} \). In particular, the gate-tunability of charge-to-spin conversion rates enabled by the 2D Dirac nature of interfacial electronic states would substantially boost the prospects of utilizing layered materials in energy-efficient spintronic devices.

Here, we used the layered compound 2H-TaS\(_2\) to induce Rashba SOC in graphene. The work function of 2H-TaS\(_2\) (\( W_{\text{TaS}_2} \approx 5.6 \text{ eV} \)) is very close to the predicted critical value\(^{25} \) where repulsive chemical interactions between the metallic TMD and graphene precisely balance the driving force for charge transfer arising from the work function difference of the two subsystems. This enables strong interface-induced Rashba SOC, while minimizing the Fermi level shift with respect to the unperturbed Dirac points, thus providing ideal conditions to explore the interplay of spin and pseudospin degrees of freedom and its dependence on the charge carrier density in graphene. Figures 1a and b show, respectively, a schematic illustration and an optical image of the van der Waals heterostructure device (see Methods). The spin diffusion channel is made from a five-layer graphene (5LG) flake. This choice provides enhanced spin coherence due to screening of impurity potentials by adjacent layers\(^{26} \). A \( 3.6 \times 6.5 \mu m^2 \) section of 5LG is covered with 15nm-thick 2H-TaS\(_2\) to create a cross-shaped heterojunction to induce REE. The device contains non-magnetic Ohmic electrodes (Cr/Pd, 3/45 nm, labelled NM1 and NM2) and ferromagnetic electrodes (Co/Pd, 50/5 nm, labelled FM1-FM4). Further details on the device fabrication are given in Supplementary Information (SI) 1, 2 and 3.
Figure 1 | Characterization of 5LG/2H-TaS₂ heterostructure device.

a. Schematic illustration of the van der Waals REE device. A pair of ferromagnetic probes FM1 and FM2 (located at distances $l_1 = 5 \mu m$ and $l_2 = 1 \mu m$ from the heterojunction) are used as reference contacts to extract the spin transport characteristics of the 5LG flake not covered by 2H-TaS₂. b. False colored optical image of the device. c. 2D resistivity $\rho_{5LG}$ of 5LG at selected temperatures. The vertical line labelled $V_{\text{CNP}}$ indicates the CNP, where the carrier density in 5LG reaches a minimum. The maximum resistivity at room temperature is 320 $\Omega$ at the CNP ($V_{\text{CNP}} = +7 V$). The temperature dependence shows the expected charge transport crossover from a localized ($d\rho/dT < 0$) to a metallic regime ($d\rho/dT > 0$) at high $V_g$. d. 2D resistivity $\rho_{5LG/2H-TaS_2}$ of heterojunction at selected temperatures. The temperature dependence shows metallic behavior over the entire gate voltage range. e. Nonlocal spin-valve resistance $R_{SV} = (V_+ - V_-)/I$ between the normal contact NM1 and the ferromagnetic contact FM1 in applied magnetic field $\mathbf{B} = B_y \hat{y}$ at $V_g = 0$. The green (black) data correspond to a positive (negative) sweep of the applied field. f. Nonlocal Hanle spin precession measurement with magnetic field $\mathbf{B} = B_z \hat{z}$ applied at $V_g = 0$. Points are experimental results. The diffusion constant $D$ and in-plane spin lifetime $\tau$ of the 5LG spin diffusion channel are extracted from the fit to the Hanle spin precession curve (solid lines). See SI 6 for additional discussions.
We performed charge and spin-sensitive transport measurements to characterize the device components (5LG spin diffusion channel, 5LG/2H-TaS$_2$ and 2H-TaS$_2$; see Fig. 1a). The two-probe resistance of 2H-TaS$_2$, $R_{\text{TaS}_2}$, remains constant under applied back-gate voltage $V_g$ (Fig. S2, SI 4 and SI 5). In contrast, both the 2D resistivity of the spin diffusion 5LG channel, $\rho_{\text{5LG}}$, and the 2D resistivity of the 5LG/2H-TaS$_2$ heterojunction, $\rho_{\text{5LG/TaS}_2}$, exhibit large gate tunability (Figs.1c-d). Moreover, the charge neutrality point (CNP) is seen to shift from $V_g \approx 7$ V in 5LG to $V_g \approx -3$ V in the heterojunction region, confirming that the Fermi level in 5LG/2H-TaS$_2$ remains close to the unperturbed Dirac point. This indicates that 2D Dirac-like states dominate the electronic transport.

Next, lateral spin-valve measurements were carried out with reference electrodes FM1 and FM2. Figure 1e shows the measured nonlocal spin-valve resistance $R_{SV}$ as function of the applied in-plane magnetic field along the spin-detector easy axis, $B_y$, at $T = 22$ K and $V_g = 0$ V. The abrupt changes in $R_{SV}$ correspond to the magnetization switching of the two ferromagnetic electrodes. Hanle-type spin precession measurements were performed with a perpendicular magnetic field $B_z$ (see Fig. 1f). By fitting Hanle curves at different $V_g$ to a 1D Bloch model, we have extracted a spin diffusion length $\lambda_G$ in the range 2-3 µm and polarization $P = 4.8\%$ at room temperature (SI 6).

The detection scheme employed to investigate charge-to-spin conversion$^{2,27}$ is depicted in Fig. 2a. An applied charge current $I$ in the 2H-TaS$_2$ strip generates a nonlocal voltage $V_{NL}$ between the Co contact (labelled FM3 in Fig. 1a) and the normal Pd contact on 5LG (labelled NM2 in Fig. 1a). To enable detection of nonlocal signals that originate from REE, a magnetic field $B = -B\hat{\tau}$ is applied in order to tilt the FM3 detector magnetization towards the hard axis direction. We focus initially on measurements for electrons ($V_g > 0$). The driving electric field ($E_y$, along the $+\hat{y}$ direction) shifts the spin-split Fermi surface of interfacial states, thereby producing an excess spin density $\delta S_x$ with spin-moment parallel to the spin diffusion channel (Fig. 2b).
a. The REE measurement protocol: a charge current $I$ is injected along 2H-TaS$_2$ and the nonlocal voltage generated between the ferromagnetic contact and 5LG ($\Delta V_{NL} = V_+ - V_-$) is measured while sweeping the magnetic field $B = -B_x$. b. Schematic illustration of spin-helical Rashba sub-band and REE mechanism. Dashed and solid circles represent, respectively, the Fermi surface of the Rashba sub-band before and after application of a charge current (electric field) along +y direction. The arrows winding around the circle represent the sub-band equilibrium spin polarization vector $s_k$. Spin-momentum locking ($s_k \cdot k = 0$) generates net nonequilibrium spin polarization $\delta S_x$ due to applied electric field $E_y$. c. Magnetic field dependence of REE nonlocal resistance at $T = 293$ K and $V_g = 35$ V. The REE signal is sensitive to the current-induced spin-polarization generated by all spin-split sub-bands in the vicinity of the Fermi level.
The nonequilibrium spin polarization diffuses away from the heterojunction and is detected by the FM3 electrode. In the 1D channel approximation, the spin accumulation $\mu_s = \mu_\uparrow - \mu_\downarrow (\uparrow, \downarrow = \pm \hat{x})$ detected at the contact ($x = L \approx 1.5 \ \mu m$) reads as

$$\Delta \mu_s(V_g, B) \approx \Delta \mu_{\text{REE}}(V_g) \sin(\theta) e^{-\frac{L}{\lambda_G(V_g)}},$$

(1)

where $\Delta \mu_{\text{REE}}$ is the current-induced spin accumulation at the heterojunction ($x = 0$) and $\theta$ is the FM3 magnetization angle with respect to the easy axis (Fig. S2d). Fig. 2c shows the nonlocal REE resistance

$$R_{\text{REE}} \equiv \frac{V_{\text{NL}}}{I} = -P \frac{\Delta \mu_s}{2|e|I},$$

(2)

measured at room temperature for $V_g = +35 \ \text{V}$. The applied field $B$ is swept between $-0.5 \ \text{T}$ and $0.5 \ \text{T}$. We observe an antisymmetric response $R_{\text{REE}}(-B) \approx -R_{\text{REE}}(B)$ characterized by a linear behavior at small magnetic fields, followed by saturation on the scale of $B_{\text{sat}} \approx 0.2 \ \text{T}$ (consistent with the reference spin-valve measurements; SI 6). The data accurately follows the relation $R_{\text{REE}} \propto \sin \theta$, thereby conclusively linking the measured nonlocal resistance to $\hat{x}$-spin-polarized electrons generated electrically by 5LG/2H-TaS$_2$. We contrast this behavior with graphene/semiconducting TMD$^{28}$, where $R_{\text{NL}}$ was found to display a steep decrease towards zero, when the magnitude of the applied field exceeds $B_{\text{sat}}$, indicating that $\hat{z}$-spin-polarized electrons are generated at the interface.

Figure 3a shows the effect of the back-gate voltage on the device output. We observe strong gate-tunability, with a clear antisymmetric behavior $R_{\text{REE}} \propto \sin \theta$ for all $V_g$. A lower bound to the REE efficiency $\gamma_{\text{REE}}$ is found as

$$\gamma_{\text{REE}} = \frac{V_{\text{NL}}}{\rho_{5\text{LG}} l_{5\text{LG}}} \frac{w_{\text{TaS}_2}}{\rho \lambda_G} e^{-L/\lambda_G},$$

(3)
Figure 3 | Gate-voltage characteristics and charge-to-spin conversion efficiency

a. Nonlocal resistance $R_{\text{REE}}$ at room temperature as function of applied magnetic field $B = -B_\times$ at selected values of gate voltage $V_g$. The signal vanishes near the CNP ($V_g \approx -3$ V) for all values of the applied field $B$. Away from CNP, $|R_{\text{REE}}|$ increases monotonically with the applied back-gate voltage up to $|V_g|\approx 40$ V, before it saturates. The $R_{\text{REE}}$-sign is determined by the charge carrier polarity according to $\text{sign} R_{\text{REE}} = p \text{ sign } B$, where $p = 1$ for electrons ($V_g > 0$) and $p = -1$ for holes ($V_g < 0$), as expected for REE originating from spin-helical 2D Dirac states. The $B$-field modulation of the nonlocal signal $R_{\text{REE}}(B)$ follows accurately the relation $R_{\text{REE}} \propto \sin \theta$ for all $V_g$, unambiguously demonstrating that it results from diffusive $\times$-spin polarized currents generated at the heterojunction.  

b. Schematic illustration of the REE-induced spin polarization imbalance under an external electric field. The blue and red sub-bands represent schematic spin-split 2D Dirac states of a graphene-based heterostructure, having opposite spin helicity. Interfacial broken inversion symmetry endows spin-split states with counter-rotating spin textures. The Fermi level lies in the conduction band (left) [valence band (right)]. The applied electric field $E_y$ biases the occupation probability of spin-helical electrons in $k$ space, which results simultaneously in a charge current ($J_y$) and a nonequilibrium spin polarization density with spin moments along the $\hat{x}$ direction ($\delta S_x$) (cf. Fig. 2b). The blue and red filling of the 2D Dirac cones represent the occupied states in applied electric field for majority (counterclockwise spins) and minority (clockwise spins) sub-bands.  

c. Lower-bound charge-to-spin conversion efficiency $\gamma_{\text{REE}}$ as a function of gate voltage at selected temperatures. The applied magnetic field is $B = -0.4 \hat{x}$ (T).
where $I_{5LG}$ is the current flowing through 5LG and $w_{TaS_2}$ is the width of 2H-TaS$_2$ flake (SI 7). The figure of merit ranges from about $-0.1\%$ at $V_g = -50$ V to $+0.2\%$ at $V_g = +50$ V (Fig. 3c). Such a fine degree of electrical control cannot be attributed to the spin transport characteristics of 5LG given the inherent weak $V_g$-dependence of the spin relaxation length; see Fig. S2a and Eqs. (1) and (2). Rather, it shows that $R_{\text{REE}}$ is primarily sensitive to the current-induced spin accumulation at the heterojunction, that is, $\Delta \mu_{\text{REE}}\left(V_g\right)$. The reversal of current-induced spin polarization across the CNP is therefore an unequivocal signature of spin-helical 2D Dirac fermions$^{18}$. The spin texture of interfacial states and associated REE-induced $\mathbf{\hat{r}}$-spin-polarized electrons are illustrated in Fig. 3b. These findings are in contrast to charge-to-spin conversion in graphene/WTe$_2$,$^{29}$ where SHE within WTe$_2$ is the driving SOC transport mechanism. Indeed, the nonlocal resistance in Ref.$^{29}$ is positive (nonzero) at all $V_g$, attaining a maximum around the CNP due to increased spin absorption by WTe$_2$. In our device, SHE in 2H-TaS$_2$ is negligible as borne out by the ambipolar character of the output nonlocal signal, with $R_{\text{REE}}\left(V_g\right)$ vanishing near the CNP (Fig. 3). Further discussions are provided in SI 8 and SI 9.

To elucidate the nature of interfacial spin-orbit interactions in the van der Waals device, we carried out relativistic electronic structure calculations for a representative 5LG/bilayer-2H-TaS$_2$ supercell (see Methods and SI 10). Figures 4a and b show the crystal structure and band structure near the Dirac point of 5LG/2H-TaS$_2$, respectively. The spin-splitting of 5LG Dirac-like states is clearly observed, with spin-gap reaching values as large as 70 meV for holes. Remarkably, the Fermi level shift, with respect to the unperturbed Dirac point, is only about +20 meV, despite the strong interaction between 5LG and 2H-TaS$_2$ (particularly visible in the hole bands).
Figure 4 | Relativistic electronic structure and tight-binding transport calculations.

a. Supercell of the heterostructure built from Bernal stacked 5LG and bilayer 2H-TaS$_2$. b. Energy bands near the vicinity of the $K$ point (wavevector path shown in the inset). Here, $a = 0.14$ nm is the lattice scaling of graphene. Spin-split states prominently localized on carbon atoms in 5LG are indicated by red and blue dots. The maximum spin splitting near the Fermi level is on the order of 30 meV (electron sector) and 70 meV (hole sector). 2H-TaS$_2$ bands are shown in background. c. Top panel shows the spin-split Fermi surface and spin polarization texture of electronic states at selected energies ($E = 0.05, 0.1, 0.15,$ and $0.20$ eV) obtained from a minimal tight-binding model of pristine 5LG with proximity-induced SOC (see Methods and SI 12). To aid visualization, only 2 pairs of spin-split states are shown in the top panels (spin-split states mapped out in the left/right panel correspond to the electron states lying closest/second closest to the Dirac point as shown in panel b). The lower panel shows the dimensionless Edelstein efficiency $v_F\beta_{EE}$ of the interfacial carbon layer as function of chemical potential at selected temperatures obtained from the minimal tight-binding model ($v_F = 10^6$ m/s is the Fermi velocity of massless 2D Dirac fermions).
In order to estimate the efficiency of charge-to-spin conversion by spin-split states in 5LG, we carry out tight-binding transport simulations informed by the ab initio electronic structure (see Methods). The Fermi surface of a pristine 5LG with proximity-induced SOC is shown in Fig. 4c, top panel. Because of spin-lattice-pseudospin entanglement\textsuperscript{17}, the spin polarization magnitude $|s_k|$ is sensitive to the Fermi wavevector. This is in contrast to conventional Rashba-split 2D electron gases\textsuperscript{7}, where the polarization is maximal ($|s_k| = \hbar/2$) and in-plane. As such, the $k$-resolved spin texture shows a rich evolution from the CNP, where SOC trigonal warping effects dominate, to the highest accessible energies in the experiment ($|E_F| \approx 0.10 - 0.15$ eV), where well-established counter-rotating spin textures emerge. The Edelstein efficiency $\beta_{EE} = (2e/\hbar)\delta S_x/J_y$ (units s/m) obtained from the tight-binding model is shown in the Fig. 4c (bottom panel). The tight-binding figure of merit increases monotonically from zero at the CNP to its maximum allowed value ($|\beta_{EE}| \approx 0.1/v_F$) when the chemical potential $|\mu|$ is on the order of the average spin gap $\Delta = \langle \Delta_k \rangle$ (see SI \textsuperscript{11}). The ambipolar effect—with the $\beta_{EE}$-sign determined by the charge carrier polarity—is a direct manifestation of spin-pseudospin coupling in an inversion-asymmetric 2D material\textsuperscript{18}. The spin polarization of majority Rashba sub-bands (highlighted in blue in Fig. 4c top panel) rotates anticlockwise, which explains the observed positive (negative) REE sign for electrons (holes) (see Fig. 3c). The observed ambipolar effect is in stark contrast to charge-to-spin conversion by surface states of topological insulators\textsuperscript{23}, where nonequilibrium spins are oriented along the same direction for both $n$- and $p$-type carriers. The slow decay of $\beta_{EE}$ in the high-density regime ($|\mu| \gtrsim \Delta$) signals the onset of minority- (clockwise) and majority-spin (anticlockwise) Rashba sub-bands with large polarization, $|s_k| \approx \hbar/2$. This behavior is little sensitive to the relaxation time $\tau_k$ (e.g. due to static disorder\textsuperscript{18}), and thus provides a useful transport fingerprint of spin helical states.
The microscopic figure of merit $\beta_{EE}$ is linked to the charge-to-spin conversion efficiency $\gamma_{EE}$ accessible in the experiment through the simple relation $\gamma_{EE} = \vartheta_G \times \beta_{EE}$, where $\vartheta_G = D_G/\lambda_G$ is the spin channel efficiency and $D_G$ is the spin diffusion constant of 5LG. The tendency for signal saturation at high gate voltage in Fig. 3c (c.f. Fig. 3a) is thus reasonably explained by the combined effect of a slow decay in $\beta_{EE}$ and an increase in $\vartheta_G$ with applied back-gate voltage (see SI 11). Thermal carrier activation plays an important role in the REE device operation, especially at low chemical potential $|\mu| \lesssim \Delta$, where the energy- and wavevector-dependence of the spin texture are the most prominent (see Fig. 4c, top panel). Charge-to-spin conversion is more efficient at elevated temperatures – with strongest variation near the change of regime $\mu \approx \pm \Delta$ – as expected from a thermal carrier activation scenario (Fig. 4c, bottom panel). Inelastic scattering processes (neglected in our calculations) are expected to contribute significantly to the temperature dependence of the figure of merit. Most importantly, phonon-assisted tunnelling\textsuperscript{30} provides a simple mechanism to enhance nonlocal signals given the small Fermi surface overlap of 5LG and 2H-TaS\textsubscript{2}.

In conclusion, we have reported current-induced spin polarization at room-temperature in a van der Waals heterostructure of few-layer-graphene and 2H-TaS\textsubscript{2}. The non-equilibrium spin polarization is readily tunable by a back-gate voltage, with charge-to-spin conversion efficiency ranging from about -0.1% at negative gate voltage ("ON" state with excess spin "$\downarrow$") to +0.2% at positive gate voltage ("ON" state with excess spin "$\uparrow$"). The "OFF" state (no spin polarization) is ideally achieved at charge neutrality. The REE spin-switching effect (that is, electrical reversal of the nonequilibrium spin polarization vector) unveiled in this work paves the way for all-in-one energy-efficient generation and manipulation of spin-based information using nonmagnetic van der Waals materials.
Methods

Crystal Growth. Single crystals of 2H-TaS$_2$ were grown by a chemical vapor transport method using iodine as the transport agent. The temperature was increased to 785 °C in a 120 hours period. The cold end of the tube was kept at 735 °C during growth. Following a stabilization period of 8 days, the high temperature zone was cooled to 570 °C in 100 hours. After synthesis, the tube was left to cool down to room temperature. The size of typical single crystals is 3 x 3 mm$^2$ in the lateral size with a thickness of a few hundred micrometers.

Device fabrication and spin transport measurements. The van der Waals heterostructure was fabricated by vertical assembly of few-layer graphene (prepared from mechanical exfoliation of commercial Kish graphite) and 2H-TaS$_2$ (thickness ≈15 nm). Number of layers and stacking sequence of the graphene flake were determined by Raman spectroscopy. The thickness of 2H-TaS$_2$ was measured via atomic force microscopy. Electrodes were patterned by e-beam lithography and e-beam evaporation of metals in a vacuum chamber of base pressure 10$^{-7}$ Torr. To enhance the spin injection efficiency, a tunnel barrier of oxidized TiO$_2$ was inserted between few-layer graphene and the ferromagnetic contacts. Further details about the device fabrication and characterization are given in the SI 1, SI 2 and SI 3. The transport measurements were performed in a home-built cryostat with rotatable sample stage with a base temperature of 22 K. A room-temperature magnet of maximum field 0.8 T was employed for the magneto-transport study. Electrical measurements were performed using a standard low-frequency lock-in technique.

Ab initio calculations. To gain insights into the atomic and electronic structure of graphene multilayers on TaS$_2$, we carried out ab initio density functional theory calculations. To reduce the
in-plane strain of the multilayer heterostructure, we employ unit cells consisting of a $3 \times 3$ TaS$_2$ supercell and a $4 \times 4$ graphene supercell resulting in a small interlayer mismatch of less than 2 percent. To calculate the ground state energy, we employ a plane-wave/pseudopotential approach as implemented in the computer program VASP (31). In particular, we use the Perdew–Burke–Ernzerhof (PBE) exchange-correlation energy functional, projector-augmented wave (PAW) pseudopotentials, a plane-wave cutoff of 400 eV and a vacuum region of more than 10 Å between periodically repeated slabs. The first Brillouin zone of the heterostructure was sampled using a $3 \times 3 \times 1$ k-point grid. Van der Waals interactions were included. All structures were fully relaxed until the force on each atom was less than 0.01 eV Å$^{-1}$. To visualize the Fermi surfaces, we interpolated the PBE band structure using maximally localized Wannier functions (see SI 11 for further details).

**Tight-binding transport model.** To determine the spin-charge conversion characteristics of the 5LG-2H-TaS$_2$ heterostructure, we carried out accurate tight-binding transport calculations. The graphene multilayer with ABABA stacking was described by the standard Slonczewski-Weiss-McClure model of bulk graphite (32) supplemented with interface-induced Rashba interaction on the two closest layers to 2H-TaS$_2$. The standard graphite tight-binding parameters are adjusted until the spectrum qualitatively reproduces the ab initio energy bands of states predominantly localized on 5LG (Fig. 4b). To determine the response of the heterostructure to a DC electric field, we solve the Boltzmann transport equations incorporating electron scattering processes within the standard relaxation time approximation. The 2D Edelstein efficiency parameter $\beta_{EE}$ is determined from the ratio of charge current-spin density susceptibility $\chi_{xy}(\mu, T) = \delta S_x(\mu, T)/E_y$ to charge conductivity $\sigma_{dc}(\mu, T) = J_y(\mu, T)/E_y$ computed from exact numerical diagonalization of the
optimized tight-binding model for an ideal interface with minimal electron-hole asymmetry (see SI 12 for further details).

Acknowledgments

We acknowledge support from the Korea NRF under Grant No. 2016R1A5A100818 (S.C.) and Grant No. 2017R1D1A1B03030877 (L.L), the Institute for Basic Science under Grant No. IBS-R009-G2 (B.S.K and C.K.), the Royal Society of London through a University Research Fellowship (A.F.) and a Global Challenges Research Fund award under Grant No. CHGR1\170063 (J.Z. and J.L.).

Author contributions

S.C. supervised the project. L.L. fabricated devices and performed measurements. S.K, G.M., W.S., H.L., B.K. and T.J. assisted with device fabrication and low-temperature transport measurements. G.M. performed Raman measurements. B.K. and C.K. grew 2H-TaS$_2$ single crystal and performed bulk characterization. A. F. conceived the theoretical modelling of the van der Waals interface and performed tight-binding transport calculations. J.L. and J. Z. carried out density functional theory calculations. S.C., A. F., J. L. and L. L. analyzed the experimental data and wrote the manuscript. All of the authors contributed to editing the manuscript.

Competing interests

The authors declare no competing financial interests.
References

1. A. Soumyanarayanan, N. Reyren, A. Fert, C. Panagopoulos, Emergent phenomena induced by spin-orbit coupling at surfaces and interfaces. *Nature* **539**, 509 (2016).

2. J. Sinova, S. O. Valenzuela, J. Wunderlich, C. H. Back, T. Jungwirth, Spin Hall effects. *Rev. Mod. Phys.* **87**, 1213 (2015).

3. N. Tombros, C. Jozsa, M. Popinciuc, J. T. Jonkman, B. J. van Wees, Electronic spin transport and spin precession in single graphene layers at room temperature. *Nature* **448**, 571 (2007).

4. M. Drogeler, F. Volmer, M. Wolter, B. Terres, K. Wantanabe, T. Taniguchi, G. Guntherodt, C. Stampfer, B. Beschoten, Nanosecond Spin Lifetimes in Single- and Few-Layer Graphene–hBN Heterostructures at Room Temperature. *Nano Lett.* **14**, 6050 (2014).

5. J. Xu, T. Zhu, Y. K. Luo, Y-M. Lu, R. K. Kawakami, Strong and Tunable Spin-Lifetime Anisotropy in Dual-Gated Bilayer Graphene. *Phys. Rev. Lett.* **121**, 127703 (2018).

6. J. C. Leutenantsmeyer, J. Ingla-Aynes, F. Jaroslav, B. J. van Wees, Observation of Spin-Valley-Coupling-Induced Large Spin-Lifetime Anisotropy in Bilayer Graphene. *Phys. Rev. Lett.* **121**, 127702 (2018).

7. K. Kim, J-Y. Choi, T. Kim, S-H. Cho, H-J. Chung, A role for graphene in silicon-based semiconductor devices. *Nature* **479**, 338 (2011).

8. Q. H. Wang, K. Kalantar-Zadeh, A. Kis, J. N. Coleman, M. S. Strano, Electronics and optoelectronics of two-dimensional transition metal dichalcogenides. *Nature Nanotechnology* **7**, 699 (2012).

9. A. Avsar, D. Unuchek, J. Liu, O. L. Sanchez, K. Watanabe, T. Taniguchi, B. Ozyimaz, A. Kis, Optospintronics in Graphene via Proximity Coupling. *ACS Nano* **11**, 11678 (2017).

10. Y. K. Luo, J. Xu, T. Zhu, G. Wu, E. J. Macormick, W. Zhan, R. Neupane, R. K. Kawakami, Opto-valleytronic spin injection in monolayer MoS$_2$/few-Layer graphene hybrid spin valves. *Nano Lett.* **17**, 3877 (2017).

11. A. Avsar, J. Y. Tan, T. Taychatanapat, J. Balakrishnan, G. K. W. Koon, Y. Yeo, J. Lahiri, A. Carvalho, A. S. odin, E. C. T. O’Farrell, G. Eda, A. H. Castro Neto, B. Ozyilmaz, Spin-orbit proximity effect in graphene. *Nature Commun.* **5**, 4875 (2014).

12. Z. Wang, D-K. Ki, H. Chen, H. Berger, A. H. MacDonald, A. F. Morpurgo, Strong interface-induced spin–orbit interaction in graphene on WS$_2$. *Nat. Commun.* **6**, 8339 (2015).

13. Z. Wang, D-K. Ki, J. Y. Khoo, D. Mauro, H. Berger, L. S. Levitov, A. F. Morpurgo,
Origin and magnitude of ‘designer’ spin-orbit interaction in graphene on semiconducting transition metal dichalcogenides. *Phys. Rev. X* 6, 041020 (2016).

14. T. S. Ghiasi, J. Ingla-Aynes, A. A. Kaverzin, B. J. van Wees, Large proximity-induced spin lifetime anisotropy in transition-metal dichalcogenide/graphene heterostructures. *Nano Lett.* 17, 7528 (2017).

15. L. A. Benitez, J. F. Sierra, W. Savero Torres, A. Arrighi, F. Bonell, M. V. Costache, S. O. Valenzuela, Strongly anisotropic spin relaxation in graphene-transition metal dichalcogenide heterostructures at room temperature. *Nat. Phys.* 14, 303 (2018).

16. J. Sichau, M. Prada, T. Anlauf, T. J. Lyon, B. Bosnjak, L. Tiemann, R. H. Blick, Resonance microwave measurements of an intrinsic spin-orbit coupling gap in graphene: A possible indication of a topological state. *Phys. Rev. Lett.* 122, 046403 (2019).

17. E. I. Rashba, Graphene with structure-induced spin-orbit coupling: spin-polarized states, spin zero modes, and quantum Hall effect. *Phys. Rev. B* 79, 161409(R) (2009).

18. M. Offidani, M. Milletari, R. Raimondi, A. Ferreira, Optimal charge-to-spin conversion in graphene on transition-metal dichalcogenides. *Phys. Rev. Lett.* 119, 196801 (2017).

19. J. C. R. Sanchez, L. Villa, G. Desfonds, S. Gambarelli, J. P. Attane, J. M. de Teresa, C. Magen, A. Fert, Spin-to-charge conversion using Rashba coupling at the interface between non-magnetic materials. *Nat. Commun.* 4, 2944 (2013).

20. H. Nakayama, Y. Kanno, H. An, T. Tashiro, S. Haku, A. Nomura, K. Ando, Rashba-Edelstein magnetoresistance in metallic heterostructures. *Phys. Rev Lett.* 117, 116602 (2016).

21. Q. Song, H. Zhang, T. Su, W. Yuan, Y. Chen, W. Xing, J. Shi, J. Sun, W. Han, Observation of inverse Edelstein effect in Rashba-split 2DEG between SrTiO3 and LaAlO3 at room temperature. *Sci. Adv.* 3, e1602312 (2017).

22. E. Lesne, Y. Fu, S. Oyarzun, J. C. Rojas-Sanchez, D. C. Vaz, H. Naganuma, G. Sicoli, J.-P. Attane, M. Janet, E. Jacquet, J.-M. George, A. Barthelemy, H. Jaffres, A. Fert, M. Bibles, L. Vila, Highly efficient and tunable spin-to-charge conversion through Rashba coupling at oxide interfaces, *Nat. Mater.* 15, 1261 (2016).

23. K. Kondou, R. Yoshimi, A. Tsukazaki, Y. Fukuma, J. Matsuno, K. S. Takahashi, M. Kawasaki, Y. Tokura, Y. Otani, Fermi-level-dependent charge-to-spin current conversion by Dirac surface states of topological insulators. *Nat. Phys.* 12, 1027 (2016).

24. T. Shimada, F. S. Ohuchi, B. A. Parkinson, Work function and photothermal of layered metal dichalcogenides. *Jpn. J. Appl. Phys.* 33, 2696 (1994).
25. G. Giovannetti, P. A. Khomyakov, G. Brocks, V. M. Karpan, J. van den Brink, P. J. Kelly, Doping graphene with metal contacts. *Phy. Rev. Lett.* **101**, 026803 (2008).

26. T. Maassen, F. K. Dejene, M. H. D. Guimaraes, C. Jozsa, B. J. van Wees, Comparison between charge and spin transport in few-layer graphene. *Phys. Rev. B* **83**, 115410 (2011).

27. L. Vila, T. Kimura, Y. Otani, Evolution of the spin Hall effect in Pt nanowires: size and temperature effects. *Phys. Rev. Lett.* **99**, 226604 (2007).

28. C. K. Safeer, J. Ingla-Anynes, F. Herling, J. H. Garcia, M. Vila, N. Ontoso, M. R. Calvo, S. Roche, L. E. Hueso, F. Casanova, Room-temperature spin Hall effect in graphene/MoS2 van der Waals heterostructures. *Nano Letters* **19**, 1074 (2019).

29. B. Zhao, D. Khokhriakov, Y. Zhang, H. Fu, B. Karpiak, A. Md. Hoque, X. Xu, Y. Jiang, B. Yan, S. P. Dash, Observation of spin Hall effect in semimetal WTe2. arXiv:1812.02113 (2018).

30. U. Chandni, K. Watanabe, T. Taniguchi, J. P. Eisenstein, Signatures of phonon and defect-assisted tunneling in planar metal–hexagonal boron nitride–graphene junctions. *Nano Lett.* **16**, 7982 (2016).

31. G. Kresse, J. Furthmuller, Efficient iterative schemes for ab initio total-energy calculations using a plane-wave basis set. *Phys. Rev. B* **54**, 11169 (1996).

32. E. McCann, M. Koshino, The electronic properties of bilayer graphene. *Rep. Prog. Phys.* **76**, 056503 (2013).