Tunable room-temperature spin galvanic and spin Hall effects in van der Waals heterostructures

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Spin–orbit coupling stands as a powerful tool to interconvert charge and spin currents and to manipulate the magnetization of magnetic materials through spin-torque phenomena. However, despite the diversity of existing bulk materials and the recent advent of interfacial and low-dimensional effects, control of this interconversion at room temperature remains elusive. Here, we demonstrate strongly enhanced room-temperature spin-to-charge interconversion in graphene driven by the proximity of WS₂. By performing spin precession experiments in appropriately designed Hall bars, we separate the contributions of the spin Hall and the spin galvanic effects. Remarkably, their corresponding conversion efficiencies can be tailored by electrostatic gating in magnitude and sign, peaking near the charge neutrality point with an equivalent magnitude that is comparable to the largest efficiencies reported to date. Such electric-field tunability provides a building block for spin generation free from magnetic materials and for ultra-compact magnetic memory technologies.

In this work, we demonstrate large, room-temperature gate-tunable SCI driven by both the SHE and ISGE (and their reciprocals) in monolayer graphene–TMDC heterostructures with the TMDC being WS₂. The spin Hall and spin galvanic effects are found to coexist in a narrow energy region near the charge neutrality point (CNP), enabling the control of both phenomena by tuning the carrier density n. The SCI mechanisms manifest themselves in the presence of large spin transport anisotropy, as a consequence of the inherited spin textures of the graphene states. The observed (I)SHE conversion efficiency as a function of n and temperature is well reproduced by theoretical calculations of the spin Hall conductivity.

The measurement scheme and optical image of a typical device are shown in Fig. 1b,c, respectively (see Methods for fabrication details and Supplementary Fig. 1 for full device schematics). A carefully designed experimental protocol based on non-local detection and combined with spin precession allows us to isolate the proximity-induced SCI in the modified graphene (graphene–WS₂) from both spurious phenomena and competing SCI in the bulk of WS₂. The device consists of a patterned graphene Hall cross with a WS₂ flake along one of the arms and ferromagnetic injector/detector electrodes (F1, F2, F3) across the other. An electric field E along the graphene–WS₂ arm (and associated current I, Fig. 1b) generates a spin current and spin accumulation due to the SHE (red arrows) and a non-equilibrium spin density conveyed by the ISGE (blue arrow, see Fig. 1a). The spins, carrying information on the SHE and ISGE, diffuse in the graphene and are detected by measuring the non-local voltage V_{NL} at F1. Alternatively, a spin current in graphene can be generated by applying I in F1. In this case, the spin current and non-equilibrium spin density that reach the graphene–WS₂ are converted into a voltage V_{F1}^{E=0} by the reciprocal effects.

Electrodes F1 and F2 are equivalent and either of them can be used to investigate the SCI. In addition, F2 and F3 (in combination with F1) are used to independently characterize the spin...
For the (spin) transport to occur only in the modified graphene, it is crucial to verify the insulating character of WS2. Otherwise, a current in WS2 could generate both out-of-plane and in-plane spins through the SHE in WS2, which can be mistaken with those associated with proximity effects. Alternatively, in the case of reciprocal effects, WS2 could act as a spin sink and the absorbed spin current could then lead to a measurable charge current (and voltage) through the ISHE in WS2 (ref. 32).

Figure 2a shows the current $I_{ds}$ versus $V_{gs}$ when a constant driving voltage $V_{ds}$ is applied between graphene and WS2. $I_{ds}$ increases sharply at $V_{g}^2 > 12$ V, marking the onset above which WS2 becomes conducting. Therefore, to avoid spin transfer to (from) WS2, SCI measurements must be acquired at $V_{g} < V_{g}^2$. Figure 2b shows the gate-dependent resistance $R$ of the graphene–WS2. The fact that the modified graphene is n-doped with $V_{g}^{2(CNP)} = -10$ V $< V_{g}^2$ allows us to investigate the proximity-induced SCI for both electron and hole transport.

The insets of Figures 2c, d show typical $R_{ds}^{1/1} = V_{ds}^{1}/I$ versus $B_{x}$ and $B_{y}$, respectively, with $M_1$ antiparallel ($V_{g}^{1/1}$) and parallel ($V_{g}^{1/1}$) to $\hat{y}$ (see Fig. 1b). The current $I$ is applied at F1 and $V_{ds}^{1}$ measured in the graphene across the graphene–WS2 arm. The orientation of $M_1$ is prepared before the measurements by applying a magnetic field along $\hat{y}$ with a magnitude exceeding the F1 coercive field. The inversion of the magnetic-field dependence when $M_1$ reverses demonstrates the spin-related origin of the signal. The main panels of Figures 2c, d show $\Delta R_{ds} = R_{ds}^{1/1} - R_{ds}^{1/1}$. By calculating the difference between $R_{ds}^{1/1}$ and $R_{ds}^{1/1}$, any non-spin-related component in $V_{ds}^{1}$ is eliminated.

Measurements in Figure 2c implement the configuration represented in Fig. 1e; therefore, $\Delta R_{ds}$ corresponds to the ISHE signal $R_{ISHE}$. Similarly, measurements in Figure 2d implement the...
configuration in Fig. 1d and thus $\Delta R_{\text{SGE}} \equiv R_{\text{SGE}}$ (for SHE and ISGE measurements, refer to Supplementary Fig. 2). The data in Fig. 2c,d exhibit an antisymmetric spin precession line shape. The spins injected by F1 are parallel to $\mathbf{y}$ and, consequently, only when the magnetic field generates a $\mathbf{z}$ ($\mathbf{\hat{x}}$) spin component, a non-zero $R_{\text{SGE}}$ ($R_{\text{ISHE}}$) is detected. At low magnetic fields $R_{\text{ISHE}}$ and $R_{\text{SGE}}$ are approximately linear; they reach maximum magnitude at about $\pm 50 \text{mT}$ and then decrease. The extrema in $R_{\text{ISHE}}$ and $R_{\text{SGE}}$ indicate an aggregate spin precession angle of $\pi/2$ when reaching the Hall cross. The asymptotic decrease to zero at larger magnetic fields ($>50 \text{mT}$) is associated with spin dephasing.

The SCI efficiencies can be obtained from the spin precession line shapes in Fig. 2c,d. To reduce the number of unknown parameters, the spin dynamics outside and inside the graphene–WS$_2$ region is independently characterized. Figure 2e shows typical non-local spin precession measurements $r_{\text{nl}} = V_{\text{nl}}^1 / I$ in pristine graphene using electrodes F1 and F3. Measurements are performed for antiparallel (\{1\}) and parallel (\{\uparrow\\uparrow\}) configurations of $\mathbf{M}_{\text{nl}}$ versus $B_\parallel$ and $B_\perp$ (see Supplementary Fig. 3 for $B_\parallel$ measurements). By fitting the results to the solution of the Bloch equations, the spin transport properties of the pristine graphene are extracted (Supplementary Fig. 3). Figure 2f shows the spin precession response in graphene–WS$_2$ versus $B_\parallel$ using electrodes F1 and F2. The results highlight the anisotropic character of the spin transport\cite{17,22,23,37}. As $B_\parallel$ increases, the corresponding $r_{\text{nl}}$ becomes much larger than its value at $B = 0$, which demonstrates that the spin lifetime for spins in the plane of graphene–WS$_2$, $\tau_\parallel^1$, is much smaller than that for spins pointing out of plane, $\tau_\perp^1$. Modelling the spin diffusion in the graphene and the graphene–WS$_2$, with the parameters obtained from Fig. 2e, yields $\tau_\parallel^1 = (6 \pm 1) \text{ps}$ and $\tau_\perp^1 = (52 \pm 10) \text{ps}$ while the anisotropy ratio, defined as $\zeta = \tau_\parallel^1 / \tau_\perp^1$, is $\zeta = 8 \pm 3$ (Supplementary Section II).

With the spin dynamics fully characterized, the spin and spin current densities can be calculated, at any position, from the analytical solution of the Bloch diffusion equation (Supplementary...
Section II). At this point, the spin precession line shapes in Fig. 2c,d can be inferred from the spin current density \( j_y \) associated with the spins projected in \( \hat{x} \) (ISHE) and the in-plane component of the spin density in \( \hat{x} \) (SGE) in the graphene–WS\(_2\) region (Fig. 1b). The only unknown parameters are scaling factors associated with the SCI efficiencies (Supplementary Section II). The ISHE is quantified with the spin Hall angle \( \theta_{\text{ISHE}} \), which measures the conversion from \( j_y \) to a charge current density \( j_x \). In the case of the SGE, where a spin density is converted into \( j_y \), several figures of merit have been proposed. In our experiments, the spin density leading to \( R_{\text{SGE}} \) derives directly from the spin current density \( j_y \). Therefore, it is possible to adopt an SGE equivalent to \( \theta_{\text{SGE}} \), namely \( \alpha_{\text{SGE}} \equiv j_y/j_x \). Because the latter conversion is from a 2D spin current into a 2D charge current, \( \alpha_{\text{SGE}} \) is dimensionless. This contrasts with the commonly used inverse Edelstein effect length, \( \lambda_{\text{IEE}} \), which quantifies the SGE conversion efficiency from a three-dimensional spin current into a 2D charge current and has the dimension of a length\(^4\).

The solid curves in Fig. 2c,d are fits to the data from which room-temperature \( \theta_{\text{ISHE}} \approx 0.3\% \), and \( \alpha_{\text{SGE}} \approx 0.1\% \) are estimated. The agreement between the model and experiment is excellent considering that only one adjustable (scaling) parameter is used. For comparison purposes, one can convert \( \alpha_{\text{SGE}} \) and \( \theta_{\text{ISHE}} \) into an equivalent efficiency \( \lambda_{\text{IEE}} \). Through \( \lambda_{\text{IEE}} = \alpha_{\text{SGE}} \lambda_{\text{SD}}^{\text{ISHE}} \approx 0.42\) nm and \( \lambda_{\text{IEE}} = \theta_{\text{ISHE}} \lambda_{\text{SD}}^{\text{SGE}} \approx 3.75\) nm, where \( \lambda_{\text{SD}}^{\text{ISHE}} \) and \( \lambda_{\text{SD}}^{\text{SGE}} \) are the in-plane and out-of-plane spin relaxation lengths in graphene–WS\(_2\). Remarkably, these values compare very favourably with those estimated for heavy metals as \( \theta_{\text{ISHE}} \lambda_{\text{SD}}^{\text{ISHE}} = 0.2\) nm for Pt, 0.3 nm for Ta and 0.43 nm for W (ref. 40). Furthermore, \( \lambda_{\text{IEE}} \) is larger than the \( \theta_{\text{ISHE}} \) reported for any material at room temperature, being one order of magnitude larger than in Bi/Ag interfaces, \( \lambda_{\text{IEE}} = 0.2–0.33\) nm (ref. 40), and almost twice \( \lambda_{\text{IEE}} = 2.1\) nm in the topological insulator \( \alpha\)-Sn (ref. 43).

The ISGE and SGE can also be investigated when \( \text{M}_1 \) rotates towards \( \hat{x} \) with sufficiently large \( B_x \) (refs. 7,14). As observed in Fig. 2c, the magnetization rotation is evident for \( B_x > 0.2\) T, with \( \text{M}_1 \) (and \( \text{M}_2 \)) becoming fully aligned with \( B_x \) for |\( B_x | > 0.3\) T. Therefore, a broad step in \( R_{\text{SGE}} \) is expected for \( B_x \) between 0.3 and 0.3 T. Figure 3a shows \( R_{\text{SGE}} \) versus \( B_x \) for three representative \( V_g \) at the CNP (\( V_g = -10\) V) and for electron (\( V_g = -3\) V) and hole (\( V_g = -13\) V) conduction. The SGE signal is clearly observed together with the spin precession response associated with the ISHE (Fig. 2c). The solid curves represent \( M_{1x} \), the projected \( \text{M}_1 \) along \( \hat{x} \), which is obtained using measurements as in Fig. 2e (ref. 7). The agreement between the field dependence of \( M_{1x} \) and the SGE step is excellent. Note that the contribution of the conventional Hall effect induced by stray magnetic fields from the rotating \( \text{M}_1 \) follows the same dependence\(^{21}\). Nevertheless, such a contribution is suppressed in our device design by using a narrow graphene flake and by placing F1 sufficiently far from the Hall cross. Most importantly, the magnitude of the SGE extracted from these measurements matches with that obtained with unambiguous spin precession experiments (Fig. 2d), allowing us to confidently discard any artefacts associated with stray magnetic fields.

The results in Fig. 3a suggest that the ISHE and the SGE follow distinct carrier density dependences. While a SHE signal is clearly distinguished at the CNP, the SGE signal changes sign between electrons and holes, becoming undetectable at the CNP. Figure 3b represents the room-temperature magnitude of the ISHE and SGE versus \( V_g \). The SGE is quantified by the step height, \( R_{\text{SGE}}^{\text{ste}} \equiv R_{\text{SGE}}(0.4\) T) – \( R_{\text{SGE}}(-0.4\) T) and the ISHE by the subtraction \( R_{\text{ISHE}}^{\text{ste}} \equiv R_{\text{ISHE}} \) of the values of \( R_{\text{ISHE}} \) at its two extrema (see Fig. 3a). It is observed that \( V_g \) has a dramatic influence on the SGI. \( R_{\text{SGE}}^{\text{ste}} \) displays a sharp peak with its maximum located near the CNP. In contrast, \( R_{\text{ISHE}}^{\text{ste}} \) is highly asymmetric about the CNP, changing sign between electrons and holes, as concluded from Fig. 3a.

The sign change in \( R_{\text{SGE}}^{\text{ste}} \) stems from the change in the nature of the carriers, as the winding of the spin texture is symmetrical about the CNP\(^{21,25}\). This further confirms our interpretation of the signal as originating from the SGE\(^{24}\). Theoretical calculations for graphene–WS\(_2\) suggest that \( R_{\text{ISHE}} \) also changes sign near the CNP with a behaviour qualitatively similar to that of \( R_{\text{SGE}} \). However, the magnitude of \( R_{\text{ISHE}}^{\text{ste}} \) is expected to be much larger for holes than for electrons\(^{24}\). As a consequence, the smearing effect resulting from the presence of charge puddles and thermal broadening might suppress the extremum in the electron side, leaving only the dominant hole peak by the CNP. The results in Fig. 4a, showing \( R_{\text{ISHE}}^{\text{ste}} \) versus \( n \) as

![Figure 3](image-url)
the temperature $T$ decreases, indicate that this hypothesis is plausible. Indeed, $R_{\text{SHE}}^0$ at room temperature (300 K) presents an incipient electron–hole asymmetry, with a steeper decrease for electron conduction. At 200 K, the decrease becomes steeper as thermal broadening is reduced. At 100 K a negative minimum at $n = 5 \times 10^{11} \text{ cm}^{-2}$ becomes fully developed.

Figure 4b shows the computed spin Hall conductivity $\sigma_{xy}^{\text{SHE}}$ in the weak disorder limit using realistic parameters; $a$ and $b$ are the electron charge and the Planck’s constant, respectively. The insets in a and b show the temperature dependence of $R_{\text{SHE}}^0$ and $\sigma_{xy}^{\text{SHE}}$, respectively, for $n$ at the extrema positions.

![Figure 4](https://example.com/figure4.png)

**Fig. 4 | Temperature dependence of the spin-to-charge conversion associated with the SHE.** a, $R_{\text{SHE}}^0$ versus carrier density $n$ at the specified temperatures. The error bars (bottom left) represent twice the standard deviation of the noise distribution at each temperature, as in Fig. 3. b, Spin Hall conductivity $\sigma_{xy}^{\text{SHE}}$ in the weak disorder limit using realistic parameters; $a$ and $b$ are the electron charge and the Planck’s constant, respectively. The insets in a and b show the temperature dependence of $R_{\text{SHE}}^0$ and $\sigma_{xy}^{\text{SHE}}$, respectively, for $n$ at the extrema positions.

incompatible with anisotropic spin relaxation (Fig. 2f) because it signals the presence of intervalley scattering, which effectively reduces the mass of the carriers and leads to the suppression of the SHE. Such discrepancies between experimental results and purely intrinsic effects suggest the presence of extrinsic contributions. A larger localized SOI can be mediated by localized defects, such as sulfur vacancies in WS$_2$, while $\theta_{\text{SHE}}$ could be enhanced by resonant scattering as has been proposed for metallic adatoms.

Our experiments therefore provide valuable insights into the physics underpinning proximity effects in graphene. In addition, they offer strategies to manipulate spin information in ultra-compact van der Waals heterostructures. The electric-field control of the SCI, both in magnitude and sign, opens the door to electrical manipulation of a ferromagnet magnetization with 2D materials and to novel spin-logic circuits.

Note that, after completion of the current research, a work studying the ISHE using magnetization rotation in graphene–WS$_2$ has been reported. The investigation reports SGE modulation at 4.2 K and large $n$ and shows that the SGE signal persists up to room temperature. However, the SGE modulation and the SHE are not observed at room temperature.

### Online content
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Methods

Device fabrication. Van der Waals graphene–WS₂ heterostructures were fabricated by dry viscoelastic stamping. The transfer set-up comprises an optical microscope with large working distance optical objectives (Nikon Eclipse LV 100ND) and a three-axis micrometre stage. Graphene is obtained by mechanical exfoliating highly oriented pyrolytic graphite (SPI Supplies) onto a p-doped Si/SiO₂ substrate. Large-area monolayer graphene is selected by optical contrast after a previous calibration with Raman spectroscopy. To fabricate the van der Waals heterostructure, WS₂ flakes are transferred onto a viscoelastic stamp (Gelpack), which is then transferred on top of the graphene target. After assembling, the stacks are annealed for 1 h at 500 °C in high vacuum (10⁻⁸ Torr). The heterostructure is coated with a polymethyl methacrylate resin mask and patterned into a Hall cross bar using electron-beam lithography followed by oxygen plasma etching. The resin is then removed with acetone. The contact electrodes are defined in two electron-beam lithography steps, one for the normal metal electrodes, Ti(1 nm)/Pd(50 nm), and the other for the ferromagnets, TiOₓ/Co(30 nm). The contact materials are deposited by electron-beam evaporation in a chamber with a base pressure of 10⁻⁸ Torr. The TiOₓ barriers are fabricated by evaporating 4 Å + 4 Å of Ti with 30 min oxidation after each evaporation in an oxygen atmosphere of about 10⁻¹⁵ Torr.

Electrical characterization. The devices are wired to a chip carrier that is placed in a cryogen-free cryostat. Charge transport properties were characterized by means of two- and four-terminal measurements. The contact resistance in the TiOₓ/Co electrodes are larger than 10 kΩ. The typical average electron–hole mobility is in the range of μ = 5,000 cm²V⁻¹s⁻¹ with a residual carrier density of 2.5 × 10¹¹ cm⁻². A back-gate voltage applied to the p-doped Si substrate is used to control the carrier density n in the device.

Data availability

The data that support the plots within this paper and the other findings of this study are available from the corresponding authors on reasonable request.

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Author contributions

L.A.B. and S.O.V. designed the device. L.A.B. and J.F.S. fabricated the device. The measurements were performed by L.A.B. and W.S.T. with the participation of J.F.S. The experimental set-up was implemented by M.T. and M.V.C., who also helped with the measurements. J.H.G. and S.R. carried out the SHE quantum simulations. L.A.B., W.S.T. and S.O.V. analysed the data and wrote the manuscript. All authors contributed to the study, discussed the results and commented on the manuscript. S.O.V. supervised the work.

Competing interests

The authors declare no competing interests.

Additional information

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