Electrical detection of spin-charge conversion in spin-valves of 2D materials

Yu-Hsuan Lin,1 Manuel Offidani,2 Chunli Huang,3 Miguel A. Cazalilla,1 and Aires Ferreira2

1Department of Physics, National Tsing Hua University and National Center for Theoretical Sciences (NCTS), Hsinchu 30013, Taiwan
2University of York, Department of Physics, YO10 5DD, York, United Kingdom
3Department of Physics, The University of Texas at Austin, Austin, Texas 78712, USA

Bilayers of 2D crystals are among the strongest contenders for the efficient control over the electron’s spin because of their gate-tunable electronic structure and strong interfacial spin-orbit effects. Here, we propose an electrical detection scheme of spin-charge conversion in spin-valves of 2D materials, which enables simultaneous measurement of prototypical spin-orbit phenomena i.e. the inverse spin Hall effect (ISHE) and the spin galvanic effect (SGE). A suitable detection geometry in applied oblique magnetic field is shown to provide direct access to spin-charge transport coefficients through a simple symmetry analysis of the output nonlocal resistance. The scheme is robust against tilting of the spin-injector magnetization and can used be to investigate a wealth of spin-to-charge conversion effects and their Onsager reciprocal in both spin-valve and hybrid optospintronic devices.

Understanding emergent spin-orbit effects at interfaces and surfaces is a major quest towards the effective control over the electron’s spin degree of freedom [1–3].

Heterostructures of 2D crystals have recently emerged as highly-controllable testbeds for exploring interfacial spin-orbit coupling (SOC) phenomena due to their gate-tunable charge carriers and strong interplay between spin and lattice-pseudospin degrees of freedom [4–10]. The excellent electronic properties of graphene and transition metal dichalcogenides (TMDs) have enabled the realization of proximity-induced SOC within atomically thin crystals, opening up intriguing possibilities for both fundamental research and spintronic applications [11–18]. A large breadth of SOC phenomena has been demonstrated and envisioned, including all-optical spin-current injection [19, 20], large spin lifetime anisotropy [22–25] and co-existence of spin Hall effect (SHE) and Rashba-Edelstein effect in 2D heterostructures with noncoplanar spin texture in momentum space [26, 27]. Theoretical studies of coupled spin-charge diffusive transport have unveiled unconventional spin-orbit scattering mechanisms, including a direct magneto-electric coupling (DMC) effect—arising from quantum interference between distinct components of the single-impurity SOC potential—which generates nonequilibrium spin polarization, a “random SOC” counterpart of the familiar Rashba-Edelstein effect [9].

Further progress in this flourishing field will require a detailed understanding of emergent SOC effects in broken symmetry conditions [10]. The phase-coherent behavior of charge carriers in graphene/TMD bilayers, with weak-antilocalization visible at low temperatures, has been attributed to proximity-induced SOC up to 10 meV [13–16]. While SOC on the order of the quasiparticle broadening offers realistic prospects for observing spin-charge conversion effects, the interplay between intrinsic and extrinsic spin-orbit mechanisms is expected to be highly complex owing to the pivotal role played by lattice-pseudospin degrees of freedom [8, 9, 26, 27].

The abundance of microscopic spin-orbit mechanisms in heterostructures of 2D materials motivates the search for device geometries that can enable electrical detection of spin-charge interconversion effects. The H-bar scheme in Refs. [11, 12] employs SHE for spin-current generation together with its Onsager reciprocal (inverse spin Hall effect (ISHE)) for electrical read-out. This method can be further extended to the detection of SGE, the Onsager-reciprocal of current-induced spin polarization, as shown in Ref. [28]. However, the two-step process, at the heart of the quadratic dependence of the output nonlocal resistance with the spin-charge conversion rates, has serious drawbacks. First, it makes the H-bar approach prone to noise and variability, as demonstrated by the controversial experimental reports on graphene with ad-atoms [29–33]. Second, it precludes the accurate evaluation of the spin Hall angle (θSHE) and the SGE efficiency (θSGE) in samples where these effects compete to establish a steady state.

In this paper, we propose a measurement protocol for unbiased direct electric detection of bona fide SOC transport effects in lateral spin-valve devices of 2D materials. We focus on a nonlocal setup comprising a spin-injector, a high-fidelity graphene spin-channel [34–37] and a cross-shaped heterojunction, where graphene is covered by a high-SOC material (see Fig. 1). Alternative device layouts and a hybrid opto-spintronic analogue are also discussed. The general principle is akin to electric detection of ISHE in metals [38]; spin-polarized carriers—.injected by applying a current $I$ at the ferromagnetic electrode—induce lateral charge accumulation, which is detected as a nonlocal voltage at the Hall cross, $V_{nl}$. The possibility to isolate SGE and ISHE contributions to the output nonlocal signal hinges on a fundamental distinction between spin-polarization-density induced Hall effect and spin-current-induced Hall effect. In the former, nonequilibrium spin density, with spin moment collinear with the propagation direction ($\mathbf{J}_i^s = -\mathbf{D}_x \nabla_i s^i$, with $i = x, y$ in 2D materials), generates a diffusive transverse charge current i.e. $J_i = \theta_{SGE} \epsilon_{ij} \mathbf{J}_j^s$. Conversely, in the ISHE,
spin-polarization, spin- and charge-propagation direction are mutually orthogonal i.e. $J_i = \theta_{\text{SHE}} \epsilon_{ijz} J_j^z$. The fundamental role played by the polarization channel ($s^z$ for ISHE and $s^x$ for SGE) is borne out when coherent electron's spin precession is induced by an oblique magnetic field normal to the injector easy-axis, $\mathbf{B} = (B_x, 0, B_z)$. Oblique spin precession is a powerful probing technique of spin relaxation anisotropy in 2D materials [39]. Here, we show that a suitable measurement scheme provides direct access to the charge-spin transport coefficients. Our main result is a linear filtering scheme for the \textit{bona fide} ISHE/SGE nonlocal resistance

$$R_{\text{ISHE(SGE)}} = \frac{1}{2} \left[ \Delta R_{\text{nl}}(\mathbf{B}) \pm \Delta R_{\text{nl}}(\mathbf{B}^*) \right],$$

where $\Delta R_{\text{nl}} = \frac{1}{2} \left( R_{\text{nl};M_y>0} - R_{\text{nl};M_y<0} \right)$ with $R_{\text{nl}} = V_{\text{nl}}/I$ is the output nonlocal resistance difference between opposite configurations of the ferromagnetic injector $M_y \neq 0$, $\mathbf{M}$ is the injector magnetization and $\mathbf{B}^* = (B_x, 0, -B_z)$ is the mirror-image of $\mathbf{B}$. The coefficients ($\theta_{\text{SHE}}, \theta_{\text{SGE}}$) can be accurately determined using a simple model for the spin-injector magnetization tilting in oblique field $\mathbf{M}(\mathbf{B})$.

The optospintronic analogue of this scheme employs a semiconducting TMD to inject spins in graphene [19, 20]. Optically-pumped spin currents are readily polarized out of the plane and ISHE/SGE detection is carried out with standard Hanle measurements in applied in-plane field $\mathbf{B} = (0, B_y, 0)$ as shown below.

\textbf{Theory}.—We provide an intuitive proof of Eq. (1). In the narrow channel limit ($W \ll l_s$, where $l_s$ is the spin diffusion length; Fig. 1), the spin dynamics is captured by the 1D Bloch model,

$$\mathbf{\dot{s}} = D_s \nabla^2 \mathbf{s} + \gamma_L (\mathbf{s} \times \mathbf{B}) - \mathbf{\Gamma s},$$

where $\mathbf{s} = (s_{M_y>0} - s_{M_y<0})/2$, $\mathbf{\Gamma} = \text{diag}(1/\tau^x_s, 1/\tau^y_s, 1/\tau^z_s)$ is the spin-relaxation matrix and $\gamma_L$ is the gyromagnetic ratio. The spin-transresistance $\Delta R_{\text{nl}}$ is proportional to the total current generated at the Hall cross. Dimensional analysis yields:

$$\Delta R_{\text{nl}} \propto D_s [\theta_{\text{SHE}} \nabla^2 s^z(x) + \theta_{\text{SGE}}/l_s s^z] \mid_{x=L}.$$

For a homogeneous ferromagnetic contact, the boundary term $\mathbf{s}(x = 0)$ coincides with the injected $\hat{y}$-spin density $[s_y(0)]_{\mathbf{B}}$ for any field $\mathbf{B} \parallel \hat{y}$. This implies that the solution of the Bloch equation is invariant under the transformation $(B_z, s^z) \to (-B_z, -s^z)$, and thus $\Delta R_{\text{nl}}(B_z) = \pm \Delta R_{\text{nl}}(-B_z)$ for SHE (SGE), proving Eq. (1).

The above picture can be formalized with a microscopy theory of coupled spin-charge diffusive transport [40]. The charge density $\rho$, charge current density $J_i$, spin polarization density $s^a$ ($a = x, y, z$), and spin current density $J_i^a$ satisfy the coupled spin-charge equations (summation over repeated indices is implied):

$$\partial_t \rho + \partial_i J_i = 0,$$

$$\nabla^a \mathbf{s}^a + \nabla_i J_i^a = -\frac{s^a}{\tau^a_s} - \kappa_i^a J_i,$$

$$J_i = -D_s (\partial_i \rho + \kappa_i^a s^a) + \gamma_{ij}^a J_j^a,$$

$$J_i^a = -D_s \mathbf{\nabla} \rho+ \kappa_i^a s^a + \gamma_{ij}^a J_j^a,$$

where $\gamma_{ij}^a = \theta_{\text{SHE}} \epsilon_{ijz} \delta_{az}$, $\kappa_i^a = l_{\text{DMC}}^{-1} \epsilon_{ia}$. The dimensionless coefficients $\gamma_{ij}^a$ and $\kappa_i^a$, in Eqs. (3)-(5) describe, respectively, the spin Hall angle and DMC efficiency [41]. The charge Herring conversion rates receive contributions from intrinsic and random (impurity potential) SOC, similar to 2D electron gases [42–44]. Homogeneous Bychkov-Rashba (BR) effect in 2D materials with interfacial breaking of inversion symmetry is predicted to enable robust spin-charge conversion irrespective of disorder type [26]. This “intrinsic” GEM-mechanism reproduces the same phenomenon as extrinsic charge-to-spin conversion, and thus it renormalizes the DMC coefficients $\kappa_i^a = \kappa_i^{a,\text{int}} + \kappa_i^{a,\text{ext}}$. The direct coupling between $J_i^a$ and $s^a$ is encoded in the SU(2) covariant derivative [21],

$$(\nabla_i O) = \partial_i O - \epsilon^{a b c} \nu_{F} (A_i^b / v_F) O^c,$$

where $\nu_{F}$ is the Fermi velocity. The internal gauge field $A_i^b$ generates electron spin precession around the spin-orbit field. Its nonzero components for a 2D material read as $A_i^b = -A_{x} = \lambda$ (BR field), $A_i^0 = \lambda_{\nu} \delta_{iz}$ (spin-valley field) and $A_i^0 = \gamma_L B_i$ (Zeeman field) [45, 46].

It will be convenient to parametrize the spin-current coefficients as follows

$$\gamma_{ij}^a = \theta_{\text{SHE}} \epsilon_{ijz} \delta_{az}, \quad \kappa_i^a = l_{\text{DMC}}^{-1} \epsilon_{ia},$$

where $\epsilon(\delta)$ is the Levi-Civita (Kronecker delta) symbol. For typical samples with ($l/l_{\text{DMC}}, \theta_{\text{SHE}}$) \ll 1 [8, 9, 26], where $l$ is the mean free path, the build up of nonlocal voltage in the cross-shaped device involves two independent steps as described below.

\textbf{Step 1: Spin diffusion}.—The Bloch equation describing the steady-state spin accumulation in the channel is obtained from Eqs. (2)-(5) by eliminating the current op-
erators in favor of the spin polarization density. To leading (linear) order in the microscopic spin-charge rates, we find

$$\mathbf{D} \cdot \mathbf{s}(x) + \gamma_L (\mathbf{s}(x) \times \mathbf{B}) = 0 \ ,$$

(7)

with

$$\mathbf{D} = D_s \begin{pmatrix} \partial^2 - l_{s||}^{-2} & 0 & 2l_{R}^{-1} \partial_x \\ 0 & \partial^2 - l_{s\perp}^{-2} & 0 \\ -2l_{R}^{-1} \partial_x & 0 & \partial^2 - l_{s\perp}^{-2} \end{pmatrix} \ .$$

(8)

The diffusion matrix Eq. (8) displays the standard Fick terms (diagonal) and a conspicuous spin-precession term with $l_{R}^{-1} = \lambda v_F / (\epsilon_F D_0)$. Assuming $D_s \approx D_0$, one finds $l_R \approx l x (\epsilon_F / \Delta)$, where $\epsilon_F$ is the Fermi energy and $\Delta = 2\lambda$ is the BR spin gap. If random SOC is significant, $l_{R}^{-1}$ receives an extrinsic contribution that increases the average spin precession rate. In pristine graphene, $l_R$ is too long to be detectable (assuming $\lambda \sim 0.01$ meV [47] and $l_s \sim 10$ l, one has $l_s/l_R \sim 10^{-2}$ for $\epsilon_F \sim 100$ meV). However, BR precession is relevant in inversion-asymmetric channels with short $l_R$. Figure 2 contrasts the Hanle precession curve for an isotropic channel ($l_R = \infty$ and $l_{s||} = l_{s\perp} = l_s$ [39]) and a pure BR channel ($l_R < \infty$ and $l_{s||} = \sqrt{2}l_{s\perp}$ [46]). The role of finite $l_R$ is perceptible when the applied field is tilted ($\phi < 90^\circ$) towards the 2D plane, enabling diffusive spins to precess in the $Oxz$ plane [48, 49]. Interestingly, the BR precession boosts the effective spin diffusion length [40], which explains the enhanced $y$-spin density accumulation away from the detector as compared to a hypothetical BR channel with the same $l_{s||} = \sqrt{2}l_{s\perp}$ but with $l_R = \infty$.

Step 2: Spin-to-charge conversion.—We now turn our attention to the Hall bar region of the spin-valve device, where SOC generates a transverse charge current $J_y$. The nonlocal voltage build-up is determined by the open-circuit condition $V_{nl,\text{total}} = J_y + (\sigma_{2D}/V)\ell_{nl} = 0$, where $\sigma_{2D}$ is the dc conductivity of the heterojunction. Using the relation between $J_y(x)$ and $s^y(x)$ [Eqs. (2)-(5)], with the parameterization in Eq. (6), we find

$$V_{nl}(x) \simeq -\frac{\ell_{nl}}{\ell_{DMC}} \left[ l_{DMC}^{-1} s^x(x) + \theta_{\text{SHE}} \partial_x s^y(x) \right] \ ,$$

(9)

where higher-order (quadratic) terms in the spin-charge rates have been neglected. This result coincides with the one obtained earlier by a dimensional analysis argument.

Detection scheme.—To illustrate the detection scheme, we first consider the device in Fig. 1. The injected spin polarization density at the contact ($s(x = 0)$) is parallel to the quantization axis of the spin-injector, $\mathbf{n} = \mathbf{M} / |\mathbf{M}|$, and thus can be tuned with an external field [37]. First, a field $\mathbf{B}_{nl} \parallel \pm \hat{y}$ is applied to set the injector configuration either “up” ($n_y > 0$) or “down” ($n_y < 0$). Subsequently, the field is removed and an oblique field in the $Oxz$ plane is scanned, $\mathbf{B} = (|B| \cos \phi \ \hat{x} + B \sin \phi \ \hat{z})$, across first and fourth quadrants ($\phi \in [0, 90^\circ]$). The nonlocal resistance difference between opposite configurations of the spin injector, $\Delta R_{nl} \equiv |V_{nl}(x)|_{+n_y} - |V_{nl}(x)|_{-n_y=0}|x=L/2|/2I$ is obtained from Eq. (9) upon solving the effective spin Bloch equation [Eq. (7)] for the spin density profile $s^y(x)$. After a lengthy but straightforward calculation, we find

$$\Delta R_{nl} = R_0 \Im \left\{ \left( \theta_{\text{SHE}} |s_B\rangle \langle s_B| \cos \phi - \frac{l_s}{\ell_{DMC}} \sin \phi \right) e^{-qL} \right\} \ ,$$

(10)

where $s_B \equiv \text{sign}(B)$, $R_0 = |s^y(0)|_B \times (\ell_{DMC}/l_{2D})$, and $q = l_{R}^{-1} \sqrt{1 + B^2 l_s^2 / \lambda^2}$. The complex spin-precession wavevector. The two contributions in Eq. (10) have opposite parity under mirror reflection $B \rightarrow -B$ (or equivalently, $\phi \rightarrow -\phi$). This shows that the bona fide ISHE (SGE) spin-transResistance is given by

$$\Delta R_{nl|\text{ISHE(SGE)}} = \frac{1}{2} \left( \Delta R_{nl|B} \pm \Delta R_{nl|\pm B} \right) \ .$$

(11)

This result deserves a few comments. First, the signal subtraction $\Delta R_{nl} = \frac{1}{2} (R_{nl|+n_y} - R_{nl|-n_y=0})$ ensures that non-spin-related contributions, such as the Hall effect $V_{nl} \propto B_x$, are filtered out and thus it is an essential part of the measurement protocol. Second, the spin-transResistance factor $R_0 = R_{nl}(B)$ defines the maximum achievable nonlocal-signal visibility for a given spin channel. Contacts with low magnetic anisotropy will result in a fast decrease of $R_{nl}(B)$ at large field, thereby shrinking the $\Delta R_{nl}$-lineshape with respect to an ideal spin-injector with magnetization pinned along the easy axis $\mathbf{M} \parallel \pm \hat{y}$. A simulation of typical lineshapes for an isotropic channel with spin-injection from an ideal contact is shown in Fig. 3(a). For field applied perpendicularly to the plane
(φ = 90°), there is no available out-of-plane spin polarization density (as long as \( \mathbf{n} \parallel \mathbf{y} \)) and thus ISHE is absent and the SGE visibility is the highest. When the field is tilted towards the 2D plane (φ < 90°), a symmetric ISHE component appears. Crucially, since \( R_{0,B} = R_{0,-B} \), the symmetry of the ISHE/SGE lineshapes [Eq. (10)] is robust against the tilting of the spin-injector magnetization (see SM below). We briefly discuss an alternative ISHE/SGE detection scheme with oblique field applied in the \( Oyz \)-plane [39]. The nonlocal resistance for a fixed configuration of the spin-injector, \( R_{nl}^{yz} = V_{nl}^{yz}/I \), is found as

\[
R_{nl}^{yz} = r_0 \left[ \theta_{\text{SH}} \sin(2\phi) f(q) - \frac{l_s \sin \phi}{l_{\text{DMC}}} e^{-qL} \right]
\]

where \( r_0 = [s^y(0)]_B \times (\hat{W}D/l_s \sigma_{2D}) \) is the typical transresistance factor and \( f(q) = \left[ e^{-L/l_s} - \Re \left( qI e^{-qL} \right) \right]/2 \). The ISHE/SGE components can be extracted via Fourier analysis if the \( \phi \)-dependence of the injected \( \hat{y} \)-spin density \( (r_0) \) is known with good accuracy. Alternatively, for typical long channels (\( L \gtrsim l_s \)), \( \theta_{\text{SH}} \) can be determined directly from nonlocal signal saturation at large field \( R_{nl}^{yz} \to (r_0/2) \theta_{\text{SH}} \sin(2\phi) e^{-L/l_s} \). Once \( \theta_{\text{SH}} \) is determined, the SGE coefficient is easily retrievable by fitting the lineshape to Eq. (12).

Next, we discuss an optospintronic analogue, in which a monolayer semiconducting TMD replaces the ferromagnetic contact as a spin injector [7]. Optical spin-injection has been recently demonstrated in graphene/TMD heterostructures [19, 20]. Since optically-pumped spin currents in graphene are polarized normal to the 2D plane, an in-plane field \( B = B\hat{y} \) can be used to detect ISHE and SGE simultaneously. The spin-transresistance \( \Delta R_{nl}^{yz} \equiv [V_{nl}(x)]_{n_x>0} - [V_{nl}(x)]_{n_x<0} |_{x=L/2I} \) is easily computed as

\[
\Delta R_{nl}^{yz} = r_0^{yz} \Re \left[ (ql_s \theta_{\text{SH}} - d^{-1}_{\text{DMC}}) e^{-qL} \right]
\]

where \( q \) is defined as before and \( r_0^{yz} = s^y(0)\hat{W}D/(l_s \sigma_{2D}) \). Similar to in-plane spin injection configuration, the ISHE and SGE signals have different parity under mirror reflection \( B \to -B \). Typical lineshapes are shown in Fig. 3(b).

**Experimental feasibility.** The measurement protocol is also applicable to spin channels with strongly anisotropic spin lifetime (e.g., TMDs [50]). However, if SOC exceeds the disorder-induced quasiparticle broadening [25], the spin accumulation relaxes on the scale of \( \phi \ll L \), hindering the nonlocal detection of ISHE/SGE. We specialised the discussion around a concrete spin-valence configuration (Fig. 1), however the detection scheme (Eq. 1) has wider applicability. An intriguing possibility is to fabricate the entire spin-valve from a large graphene/TMD flake. In this case, the spin channel can display strong BR effect, due to interfacial of breaking of inversion symmetry. Numerical simulations of non-local resistance for anisotropic channels is provided in the SM [51]. In such case, Eq. (1) ceases to be exact due to the BR precession and a microscopic model to estimate \( l_R \) is required.

Finally, we note that in the modeling of experimental data it is necessary to consider explicitly the response of the ferromagnetic contact to the applied field. As shown in this work, the re-orientation of the magnetization \( \mathbf{M} \) from its preferred easy axis reduces the visibility of the nonlocal resistance by a nonlinear factor given by \( V_{\mathbf{B}} = |s^y(0)|_{\mathbf{B}}/|s^y(0)|_{\text{ideal}} \). The field dependence of the boundary term \( |s^y(0)|_{\mathbf{B}} \) can be accurately described by a Stoner-Wohlfarth model with parameters determined from independent Hanle measurements [34–39]. According to the simulation provided in the SM, the visibility can be as large as 10-20% at low fields \( \sim 0.1 \) T for typical magnetic anisotropy parameters. We expect that such an analysis will enable accurate experimental determination of spin-charge conversion parameters \( (\theta_{\text{SH}}, \theta_{\text{SGE}} = l_s/l_{\text{DMC}}) \) in the near future. We note in passing that a similar scheme can be employed for electrical detection of spin Hall effect and Rashba-Edelstein effect, the Onsager reciprocal of ISHE and SGE, respectively. This could be achieved by injecting a current at the Hall cross and measuring the resulting spin-electrochemical accumulation at the ferromagnetic contact.

In summary, we have presented a measurement scheme for unbiased electrical detection of spin galvanic and spin

![in-plane injection](image_url)

**FIG. 3.** Detection of ISHE/DMC contributions for both in-plane and out-of-plane spin injection schemes. Oblique field parameterized as \( B = |B| \cos \phi \hat{x} + B \sin \phi \hat{y} \) (see text for alternative measurement protocol with \( B \parallel \hat{x} \)). Results are for isotropic channel (\( \zeta = 1 \), \( l_R = \infty \)) and junction with \( \theta_{\text{M}} = 0.01 \) and \( l_{\text{DMC}} = 100l_s \). Other parameters as in Fig. 2.
Hall effects in spin valve devices of 2D materials. Our results are relevant for current experimental efforts towards the efficient manipulation of the electron’s spin degree of freedom in graphene-based heterostructures. In compliance with EPSRC policy framework on research data, this publication is theoretical work that does not require supporting research data.

Acknowledgements.– A.F. gratefully acknowledges the financial support from the Royal Society, London through a Royal Society University Research Fellowship. M.O. and A.F. acknowledge funding from EPSRC (Grant Ref: EP/N004817/1).

[1] A. Manchon, H. C. Koo, J. Nitta, S. M. Frolov, and R. A. Duine, New perspectives for Rashba spin–orbit coupling. Nature Materials 14, 871 (2015).
[2] A. Soumyaranarayanan, N. Reyren, A. Fert, C. Panagopoulos, Emergent phenomena induced by spin–orbit coupling at surfaces and interfaces. Nature 539, 509 (2016).
[3] F. Hellman, et al., Interface-induced phenomena in magnetism. Rev. Mod. Phys. 89, 025006 (2017).
[4] 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 (2009).
[5] Z. Y. Zhu, Y. C. Cheng, and U. Schwingenschlögl. Giant spin-orbit-induced spin splitting in two-dimensional transition-metal dichalcogenide semiconductors. Phys. Rev. B 84, 153402 (2011).
[6] D. Xiao, G.-B. Liu, W. Feng, X. Xu, and W. Yao, Coupled Spin and Valley Physics in Monolayers of MoS2 and Other Group-VI Dichalcogenides. Phys. Rev. Lett. 108, 196802 (2012).
[7] R. A. Muniz and J. E. Sipe, All-optical injection of charge, spin, and valley currents in monolayer transition-metal dichalcogenides. Phys. Rev. B 91, 085404 (2015).
[8] A. Ferreira, T. G. Rappoport, M. A. Cazalilla, and A. H. Castro Neto, Extrinsic Spin Hall Effect Induced by Resonant Skew Scattering in Graphene. Phys. Rev. Lett. 112, 066601 (2014).
[9] C. Huang, Y. D. Chong, and M. A. Cazalilla, Direct coupling between charge current and spin polarization by extrinsic mechanisms in graphene, Phys. Rev. B, 94, 085414 (2016).
[10] J. H. Garcia, M. Vila, A. W. Cummings, and S. Roche. Spin transport in graphene/transition metal dichalcogenide heterostructures. Chem. Soc. Rev. 47, 3359 (2018).
[11] A. Avsar, J.Y. Tan, T. Taychatanapat, J. Balakrishnan, G.K.W. Koon, Y. Yeo, J. Lahiri, A. Carvalho, A.S. Rodin, E.C.T. O’Farrell, G. Eda, A.H.C. Neto, and B. Özyilmaz, Nat. Commun. 5, 4875 (2014).
[12] Z. Wang, D.-K. Ki, H. Chen, H. Berger, A. H. MacDonald, and A. F. Morpurgo, Strong interface-induced spin–orbit interaction in graphene on WS2. Nat. Commun. 6, 8339 (2015).
[13] Z. Wang, et al., Origin and Magnitude of ’Designer’ Spin–Orbit Interaction in Graphene on Semiconducting Transition Metal Dichalcogenides, Phys. Rev. X 6, 041020 (2016).
[14] T. Völkl, T. Rockinger, M. Drienovsky, K. Watanabe, T. Taniguchi, D. Weiss, and J. Eroms, Magnetotransport in heterostructures of transition metal dichalcogenides and graphene. Phys. Rev. B 96, 125405 (2017).
[15] B. Yang, M. Lohmann, D. Barroso, I. Liao, Z. Lin, Y. Liu, L. Bartels, K. Watanabe, T. Taniguchi, and J. Shi, Strong electron-hole symmetric Rashba spin-orbit coupling in graphene/monolayer transition metal dichalcogenide heterostructures. Phys. Rev. B 96, 041409 (2017).
[16] T. Wakamura, F. Reale, P. Palczynski, S. Guéron, C. Mattevi, and H. Bouchiat, Strong Anisotropic Spin-Orbit Interaction Induced in Graphene by Monolayer WS2. Phys. Rev. Lett. 120, 106802 (2018).
[17] S. Omar and B. J. van Wees, Spin transport in high-mobility graphene on WS2 substrate with electric-field tunable proximity spin-orbit interaction, Phys. Rev. B 97 045414 (2018).
[18] C. K. Säfer, et al. Room-Temperature Spin Hall Effect in Graphene/MoS2 van der Waals Heterostructures, Nano Lett. 19, 1074 (2019).
[19] Y. K. Luo, et al., Opto-Valleytronic Spin Injection in Monolayer MoS2/Few-Layer Graphene Hybrid Spin Valves, Nano Letters 17, 3877 (2017).
[20] A. Avsar, et al., Optospintrons in Graphene via Proximity Coupling, ACS Nano 11, 11678 (2017).
[21] The derivative w.r.t. time coordinate is defined as $\nabla_t O = \partial_t O + \mathbf{u} \cdot \nabla O$.
[22] A. W. Cummings, J. H. Garcia, J. Fabian, and S. Roche. Giant Spin Lifetime Anisotropy in Graphene Induced by Proximity Effects, Phys. Rev. Lett. 119, 206601 (2017).
[23] T. S. Ghiasi, J. I.-Aynès, A. A. Kaverzin, and Bart J. van Wees, Large Proximity-Induced Spin Lifetime Anisotropy in Transition-Metal Dichalcogenide/Graphene Heterostructures, Nano Lett. 17, 7528 (2017).
[24] L. A. Benítez, et al., Strongly anisotropic spin relaxation in graphene-transition metal dichalcogenide heterostructures at room temperature, Nature Physics 14, 303 (2018).
[25] M. Offidani and A. Ferreira, Microscopic theory of spin relaxation anisotropy in graphene with proximity-induced spin–orbit coupling. Phys. Rev. B 98, 245408 (2018).
[26] 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).
[27] M. Milletari, 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).
[28] C. Huang, Y. D. Chong, and M. A. Cazalilla, Anomalous Nonlocal Resistance and Spin-Charge Conversion Mechanisms in Two-Dimensional Metals, Phys. Rev. Lett. 119, 136804 (2017).
[29] J. Balakrishnan, G. K. W. Koon, M. Jaiswal, A. H. Castro Neto & B. Özyilmaz. Colossal enhancement of spin–orbit coupling in weakly hydrogenated graphene. Nature Physics 9, 284 (2013).
[30] J. Balakrishnan et al. Giant spin Hall effect in graphene grown by chemical vapour deposition. Nat. Comm. 5, 4748 (2014).
[31] A. A. Kaverzin and B. J. van Wees. Electron transport nonlocality in monolayer graphene modified with hy-
conversion in disordered two-dimensional electron gases lacking inversion symmetry, Phys. Rev. B 96, 205305 (2017).

[42] K. Shen, R. Raimondi, and G. Vignale, Theory of coupled spin-charge transport due to spin-orbit interaction in inhomogeneous two-dimensional electron liquids. Phys. Rev. B 90, 245302 (2014).

[43] A. A. Burkov, Alvaro S. Núñez and A. H. MacDonald, Theory of spin-charge-coupled transport in a two-dimensional electron gas with Rashba spin-orbit interactions. Phys. Rev. B 70, 155308 (2004).

[44] I.V. Tokatly and E. Y. Sherman, Gauge theory approach for diffusive and precessional spin dynamics in a two-dimensional electron gas, Annals of Physics 325, 1104 (2010).

[45] I.V. Tokatly. Equilibrium Spin Currents: Non-Abelian Gauge Invariance and Color Diamagnetism in Condensed Matter. Phys. Rev. Lett. 101, 106601 (2008).

[46] M. Offidani, R. Raimondi, and A. Ferreira, Microscopic Linear Response Theory of Spin Relaxation and Relativistic Transport Phenomena in Graphene, Condens. Matter, 3 18 (2018).

[47] H. Min, et al. Intrinsic and Rashba spin-orbit interactions in graphene sheets. Phys. Rev. B 74, 165310 (2006).

[48] The role of the BR field was previously noted in Ref. [49], where it was shown that it induces damped oscillations in $s^{\sigma}(x)$ when injected spins are oriented in the $Oxz$-plane.

[49] P. Zhang and M. W. Wu, Electron spin diffusion and transport in graphene. Phys. Rev. B 84, 045304 (2011)

[50] L. Yang et al. Long-lived nanosecond spin relaxation and spin coherence of electrons in monolayer MoS$_2$ and WS$_2$. Nature Physics 11, 830 (2015).

**SUPPLEMENTARY INFORMATION**

We parameterize the field-dependent polarization as

$$\mathbf{n}(B, \phi) = \hat{x} \cos [\gamma(B, \phi)] \sin [\beta(B, \phi)] + p \hat{y} \cos [\gamma(B, \phi)] \sin [\beta(B, \phi)] + \hat{z} \sin [\gamma(B, \phi)],$$

(14)

where $p = \pm 1$ for initial “parallel” (+$\hat{y}$)/”antiparallel” ($-\hat{y}$) magnetic configurations of the ferromagnetic injector. The dependence of the angle variables ($\beta, \gamma$) on the applied field ($B, \phi$) can be accurately described by a Stoner-Wohlfarth model. For our purposes, it suffices to consider a heuristic model for the tilting angles

$$\beta(B, \phi) = \arctan(B_x/B_{sat,x}), \quad \gamma(B, \phi) = \arctan(B_z/B_{sat,z}),$$

(15)

where $B_{sat,i}$ is the saturation field normal to the easy axis $\hat{i} \perp \hat{y}$ ($B_{sat,x} \gg B_{sat,x}$ due to shape anisotropy).

Figure 4 shows the output nonlocal resistance for a hypothetical spin-valve device displaying ISHE and SGE. The bare signal for “parallel” and “antiparallel” configurations is shown in the left panel. The plateaus at large field $|B|$ signal the saturation of the ferromagnetic contact. The middle panel shows the spin-transresistance between parallel and antiparallel configurations. The right panel shows the ISHE/SGE lineshapes isolated using the protocol presented in the main text

$$\Delta R_{\text{ISHE/SGE}} = \frac{1}{2} [\Delta R_{\text{nl}}(B) \pm \Delta R_{\text{nl}}(-B)] \equiv \frac{1}{2} [\Delta R_{\text{nl}}(\phi) \pm \Delta R_{\text{nl}}(-\phi)].$$

(16)

The ISHE/SGE signal has even/odd parity with respect to mirror reflection ($\phi \to -\phi$). This shows that the filtering scheme accurately separates the two independent components. Furthermore, the lineshapes have the same exact form than those presented in Fig. 2, main text (in which the tilting correction had been neglected by setting $R_0 = \text{constant}$).

As shown in the main text, close inspection of the spin Bloch equation reveals that unambiguous detection of ISHE/SGE is also possible in channels with anisotropic spin relaxation ($\tau_\parallel \neq \tau_\perp$), i.e. devices where the spin channel...
FIG. 4. Extraction of ISHE and SGE contributions to the spin transresistance at selected oblique fields ($\phi = 45^\circ, 60^\circ$), in-plane field ($\phi = 0^\circ$) and perpendicular field ($\phi = 90^\circ$) for an isotropic graphene channel with a high-SOC heterojunction. Left: Bare signal $R_{nl}$ for initial parallel (solid) and antiparallel (dashed) magnetic configurations. Middle: Nonlocal resistance $\Delta R_{nl}$. Right: Bona fide ISHE and SGE nonlocal resistances. $B_{\text{sat},x} = 0.2$ T and $B_{\text{sat},z} = 1$ T. Other parameters as in Fig. 3, main text.

(main arm) itself is characterized by strong SOC; see Fig. 5 (middle panel). The filtering scheme is only approximately valid if diffusive spins experience strong BR precession before reaching the Hall bar ($l_R \sim l_s$ in the spin-channel), which can complicate the extraction of spin-charge conversion rates $\theta_{\text{SHE(SE)}}$; see Fig. 5 (right panel).

FIG. 5. Left: ISHE/SGE filtered nonlocal resistance with magnetization tilting correction (solid lines) and without (open symbols). Middle: Simulation of individual ISHE and SGE nonlocal signals (solid lines) and filtered signals using the protocol Eq. (16) for a highly anisotropic spin channel with $l_{\perp} = 10l_{\parallel}$. Right: Same than middle panel but for a channel with $l_{\perp} = \frac{1}{2}l_{\parallel}$ and large Rashba precession parameter $l_R = 5l_s$. Other parameters as in Fig. 4.