Charge-to-Spin Conversion by the Rashba—Edelstein Effect in Two-Dimensional van der Waals Heterostructures up to Room Temperature

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Supporting Information

ABSTRACT: The proximity of a transition-metal dichalcogenide (TMD) to graphene imprints a rich spin texture in graphene and complements its high-quality charge/spin transport by inducing spin–orbit coupling (SOC). Rashba and valley-Zeeman SOCs are the origin of charge-to-spin conversion mechanisms such as the Rashba—Edelstein effect (REE) and spin Hall effect (SHE). In this work, we experimentally demonstrate for the first time charge-to-spin conversion due to the REE in a monolayer WS2-graphene van der Waals heterostructure. We measure the current-induced spin polarization up to room temperature and control it by a gate electric field. Our observation of the REE and the inverse of the effect (IREE) is accompanied by the SHE, which we discriminate by symmetry-resolved spin precession under oblique magnetic fields. These measurements also allow for the quantification of the efficiencies of charge-to-spin conversion by each of the two effects. These findings are a clear indication of induced Rashba and valley-Zeeman SOC in graphene that lead to the generation of spin accumulation and spin current without using ferromagnetic electrodes. These realizations have considerable significance for spintronic applications, providing accessible routes toward all-electrical spin generation and manipulation in two-dimensional materials.

KEYWORDS: Rashba—Edelstein effect, spin Hall effect, Rashba spin—orbit coupling, valley-Zeeman, proximity effect

Spin-orbitronics is a promising field of research that serves the future of spintronic devices, which is based on the manipulation and control of spins and is enabled by spin—orbit coupling (SOC). Graphene is known to be a superior material for long-distance spin transport,1—3 however, it has intrinsically weak SOC.4 The control of the spin signal that is necessary for spin-based devices becomes possible in graphene by inducing SOC that can be realized via the proximity of materials with large SOC. Recent theoretical5—9 and experimental10—18 studies have shown that the proximity of transition-metal dichalcogenides (TMD) can induce SOC with the strength of a few millielectronvolts in graphene.19 This leads to a large spin lifetime anisotropy7,18,16,18 due to the suppression of the in-plane spin lifetime and/or spin absorption.16,21

A few orders of magnitude larger SOC in a monolayer TMD,22 compared with graphene, together with its inversion symmetry breaking, provides this semiconductor with the theoretically predicted large intrinsic Hall angle.23 Moreover, spin-torque24 and spin-pumping25 experiments have shown the possibility of charge-to-spin conversion by the Rashba—Edelstein effect in TMDs. However, for the injection/detection and transfer of the spin information, the short spin relaxation times in TMDs present a major obstacle.

The hybridization of TMD to graphene is an effective way to complement the properties of these materials. Theory predicts that the band structure of graphene in the proximity of TMD is spin-split by the presence of Rashba and valley-Zeeman spin—orbit fields.26—31 These spin—orbit fields are the origin of charge-to-spin conversion mechanisms such as the Rashba—Edelstein effect (REE) and the spin Hall effect (SHE) that generate spin accumulation and spin-polarized currents, respectively.26—31 More importantly, the strength of these spin—orbit fields and also the efficiency of the charge-to-spin conversion mechanisms are dependent on the position of the Fermi energy within the band structure of the TMD-graphene heterostructure. For the first time, we show in this work that this is indeed the case for a WS2-graphene heterostructure where the Rashba—Edelstein effect, in particular, creates a spin accumulation within the graphene channel which is detectable up to room temperature and is tunable by a gate transverse electric field. The Rashba SOC in graphene originates from breaking the out-of-plane symmetry due to the proximity of the TMD.27 The resulting out-of-plane effective electric field (\(\mathbf{E} = \mathbf{E}_p\)) generates an in-plane Rashba spin—orbit field (\(\sim \mathbf{E}_p \times \mathbf{p}\)) that is perpendicular to the momentum (\(\mathbf{p}\)) of the electrons within...
the theoretical prediction of the largest SHE signal, specifically in a 1L WS2-graphene heterostructure, makes the vdW stack of charge-to-spin conversion.9

The winding Rashba spin–orbit field in the graphene changes sign between the spin-split Dirac cones of the conduction (or valence) band (Figure 1a). Therefore, the current-driven spin densities of the spin-split bands have opposite sign, which reduces the total spin density at the Fermi energy. However, the energy gap between the spin-split Dirac cones is enhanced by the presence of the valley-Zeeman field (calculated to be about 2.2 meV for WS2-graphene6). This results in considerably different magnitudes of the (current-driven) spin densities associated with each of the cones for low-energy states. This avoids compensation of the spin accumulation from the bands with each of the cones for low-energy states. This avoids compensation of the spin accumulation from the bands with opposite spin winding and helps to optimize the efficiency of the charge-to-spin conversion.9

Here we observe clear evidence of the charge-to-spin conversion in TMD-graphene heterostructures due to the SHE, which is accompanied by the SHE. Different directions of the spins generated from these two effects make their contributions distinguishable by their distinct symmetries as a function of the magnitude and direction of the magnetic field in our (oblique) Hanle precession measurements. The SHE has recently been observed in multilayer MoS2/multilayer graphene,18 where the SHE signal in graphene is superimposed by an additional spin-to-charge conversion mechanism which is mainly associated with SHE in the bulk MoS2. However, the measurements in this work are performed on a vdW heterostructure of a single layer of WS2 and graphene by 1L TMD, compared to bulk TMD32 eliminates the vertical charge transport inside the 1L TMD. Therefore, a possible contribution from the SHE in bulk TMD is largely suppressed in our system. Stronger induced SOC in graphene by 1L TMD, as compared to bulk,17 in addition to the theoretical prediction of the largest SHE signal, specifically, in a 1L WS2-graphene heterostructure,30 makes the vdW stack of our sample an optimal choice.

In Figure 1b, we show the device geometry consisting of 1L WS2/1L graphene that is encapsulated between 1L hexagonal boron nitride (hBN) and bulk hBN. The device is fabricated on a 300 nm SiO2/doped Si substrate with Ti/Au and Co electrodes, made by shadow mask evaporation and e-beam lithography, respectively (Methods section). The 1L hBN acts as a tunnel barrier for the spin injection/detection by the Co electrodes. Represented by the white dashed line in the optical image (Figure 1c), the graphene channel is etched into a Hall bar which allows for the nonlocal detection of the induced spin density, generated by both effects. Note that for the fabrication of the vdW stack we do not have control over the crystallographic alignment of the TMD, graphene, and hBN flakes which can affect the strength of the spin–orbit fields as compared to the calculations that assume (super)lattice matching.33,34

**REE and SHE Measurements with an In-Plane Magnetic Field (x–y Plane).** Our main focus in this work is on the TMD-covered graphene region of this device. As shown in the device sketch in Figure 2a, using the Ti/Au contacts on graphene we apply charge current (I), and with ferromagnetic Co electrodes, we measure the nonlocal voltage (\(V_{\text{nl}}\)) as a function of an applied magnetic field (B). With an applied charge current along the y axis and in the presence of REE, one should expect the generation of nonzero spin density polarized along the x axis, \(n_x\). We formulate our theoretical model of coupled charge-spin transport in the presence of REE. By numerically solving Bloch diffusion equations (COMSOL details in section 6 of the SI), we obtain a distribution of \(n_x\) over the full sample shown as a color map in Figure 2a. Using these solutions at any applied field B, one can estimate the signal between the spin-sensitive Co contacts, shown in the bottom right corner of Figure 2a.

At \(B = 0\) T, the magnetization of the Co contacts is along their easy axis (y direction), implying that the nonlocal resistance (\(R_{\text{nl}}\)) should be zero. Applying a magnetic field in the x direction (\(B_x\)) changes the direction of the contact magnetization, in accordance with the Stoner–Wohlfarth model.35 The component of the contact magnetization along the x axis increases linearly with \(B_x\) while the REE-induced spin density stays unaffected. This results in a linear increase in the nonlocal resistance until the contact magnetization direction is completely saturated along the x axis (at \(B_x \approx 0.3\) T, for our Co electrode geometry). A negative magnetic field causes the...
alignment of the contact magnetization in the opposite direction. This results in a negative signal because the polarization of the REE spin density stays unchanged. Therefore, antisymmetric behavior of the nonlocal signal versus $B$ is a direct signature of REE and can be used to extract the REE-related signal from the experimental results.

By the inverse of the REE (IREE), the generation of charge current becomes possible as a result of the nonzero spin density in graphene. In this geometry (shown in Figure 2b), the detection of the nonlocal voltage drop is across the Ti/Au contacts, while Co electrodes are used to apply the current required for the injection of in-plane spins. IREE is the Onsager reciprocal of REE, implying that the detected nonlocal signal should be the same but with a reversed sign of $B$ ($R_{ij}(-B) = R_{kl}(B)$). The measurement is performed at 4.2 K with a charge carrier density of $+1.5 \times 10^{12} \text{ cm}^{-2}$. The inset at the top left shows the symmetric and antisymmetric components of the signal, separately. The inset at the bottom right is the $R_{nl}$ measured in IREE geometry (with $I = 2 \mu$A).

In Figure 2c, we show the out-of-plane spin density ($n^z$) produced by the SHE all over the sample. The out-of-plane spins cannot generate a nonlocal voltage across the in-plane-magnetized Co electrodes unless they precess around the applied magnetic field. Therefore, the detected signal develops from zero at $B = 0$ T to a finite value as the spins precess to the in-plane direction along the y axis. Furthermore, the $R_{nl}$ drops back to zero above the saturation field of the Co contact because the Co magnetization and spin alignment are again perpendicular to each other. The sign of the SHE signal depends on the orientation of the Co magnetization (Figure 1c). In our calculations, we assume that the y component of the contact magnetization is oriented along with the y component of the magnetic field direction. (d) $R_{nl}$ versus $B_x$, experimentally measured in the geometry of the REE and SHE (with $I = 5 \mu$A). The measurement is performed at 4.2 K with a charge carrier density of $+1.5 \times 10^{12} \text{ cm}^{-2}$. The inset at the top left shows the symmetric and antisymmetric components of the signal, separately. The inset at the bottom right is the $R_{nl}$ measured in IREE geometry (with $I = 2 \mu$A).
The graphene width is about 1.8 μm. These measurements are performed at 4.2 K with a charge carrier density of $+1.5 \times 10^{12}$ cm$^{-2}$. The observed result contains signals from both REE and SHE effects. The top-left inset shows the antisymmetric and symmetric components that are extracted from the measured data in order to discriminate the spin signal dominated by the REE and SHE, respectively. The magnitude of the measured REE spin signal is $\Delta R_{nl} \approx 200$ mΩ, defined as half of the difference between the $R_{nl}$ values measured at the two saturation levels.

The bottom-right inset shows the IREE spin signal measured with the inverse geometry that shows similar behavior but with reversed sign versus $B$, confirming the spin-to-charge conversion and preservation of the reciprocity in the linear regime. The very small background resistance in these measurements affirms that in our nonlocal geometry the current path is well-separated from the voltage probes.

The magnitude and modulation of the measured spin signal is strongly dependent on the direction of the applied magnetic field. In Figure 3a, we evaluate how the nonlocal resistance changes as we apply the in-plane magnetic field at certain angles with respect to the $x$ axis ($\alpha \approx -75$ to $+75^\circ$). All of the measurements are performed by aligning the contact magnetization at high fields, meaning that the $y$ component of the Co magnetization is always collinear with the $y$ component of the magnetic field. In panel b, we show the corresponding modeled dependences that closely reproduce our experimental results.

The behavior of $R_{nl}$ is understood by considering the precession of the in-plane and out-of-plane spins around the $\alpha$-angled magnetic field and the corresponding symmetries versus $\alpha$. As shown in Figure 3c, REE-induced spins result in the same positive projection on the Co magnetization direction for both $+\alpha$ and $-\alpha$, meaning that the REE spin signal is symmetric versus angle $\alpha$. On the other hand, the precessed out-of-plane SHE spins generate signal projections on the contact magnetization with opposite signs for $+\alpha$ and $-\alpha$, implying the antisymmetric contribution of SHE spins versus the angle. This means that the REE contribution to the signal does not change whereas the SHE contribution changes from peaks (dips) to dips (peaks) when the angle is changed from $+\alpha$ to $-\alpha$.

Specifically, in Figure 3d we show the REE spin signal (antisymmetric vs $B$ and symmetric vs $\alpha$) measured under angled $B$. As expected for the Co magnetization behavior, we observe the shift of the saturation fields under different angles, together with the change in the magnitude of the spin signal. In the following table, we summarize the symmetries for the in-plane Hanle precession measurements as

\[ \begin{array}{ccc}
\text{vs} & \text{REE} & \text{SHE} \\
B & \text{antisym} & \text{sym} \\
\alpha & \text{sym} & \text{antisym} \\
\end{array} \]

Note that in the measured nonlocal signal there is an additional component that does not comply with the symmetries of the REE and SHE. This component is subtracted from the experimental data, resulting in Figure 3a. (For details and discussions, see the SI, section 4).

The closest fit to the data (Figure 3b) gives an estimate of the in-plane spin lifetime of $\tau_{\parallel} \approx 3.5$ ps with a spin lifetime...
bulk MoS2-graphene is about 0.05 (with undetermined charge neutrality point at...n-type doping (see supplementary information for details.) We formulate the spin Hall angle with the theoretically predicted decay in the REE efficiency versus the position of the Fermi energy.9

The preservation of the charge-to-spin conversion mechanism at room temperature is a prerequisite for potential applications. We evaluate the temperature dependence of the REE (Figure 4b) and observe that the spin signal generated by the REE and the IREE is preserved up to room temperature; however, it decays by about 80% from 4 K up to RT. This behavior indicates the robustness of the REE charge-to-spin conversion mechanism, which is in agreement with theoretical predictions. We observe that the features associated with SHE in our system, together with the unknown component in the Hanle precession measurements, vanish at temperatures above 20 K. This indicates that the SHE has a stronger dependence on temperature than does the REE.

REE Measurements with an Out-of-Plane Magnetic Field (x−z Plane). We further characterize the spin transport by applying the magnetic field in the x−z plane, under an angle (θ) with respect to the normal to the sample plane (shown in the sketch of Figure 5). First, the θ-angled B brings the contact

Figure 4. Gate and temperature dependence of the REE spin signal. (a) Hanle precession measured with respect to Bx (antisymmetrized Rnl vs B) at gate voltages of −20 to +20 V (at T = 4.2 K). The inset is the magnitude of the REE spin signal versus Vg. (b) Antisymmetric component of Rnl vs Bx measured at different temperatures in the REE geometry. The signal shown at room temperature is measured in IREE geometry. The inset is the temperature dependence of the REE spin signal.

Figure 5. Observation of REE by out-of-plane Hanle precession measurements. The device sketch shows the precession of the in-plane REE spins about the applied magnetic field, angled by θ with respect to the normal to the plane. The curve is the antisymmetric component of the subtraction of the signal measured under angles of θ = ±30° (attributed to the REE spin signal), measured at 300 K. The inset is the antisymmetric component of the nonlocal resistance as a function of Bx measured with respect to the θ-angled magnetic field.
magnetization direction out of plane, and second, it precesses
the in-plane spins in the out-of-plane direction, which are then
detected by the contacts with a tilted magnetization. The
symmetry table for the non-precessing SHE and REE
components for the out-of-plane field measurements is
\[
\begin{array}{ccc}
\text{REE} & \text{SHE} & \theta \\
B & \text{antisym} & \text{antisym} \\
\theta & \text{antisym} & \text{sym} \\
\end{array}
\]

SHE is symmetric vs angle and is antisymmetric vs \( B \), thus
having the same symmetries as the ordinary Hall effect. The
nonlocal sample geometry minimizes the local charge current
contribution to the detection voltage to a large extent. However,
the detection electrodes in our sample are close to the current
path, which results in a (small) Hall effect contribution of an
order of magnitude similar to that of the SHE contribution. This
implies that we are not able to extract the SHE component.
Nevertheless, by antisymmetrizing the measured signal with
respect to the angle we eliminate both SHE and the regular Hall
effect, thus leaving only the REE contribution. In Figure 5, we
show the resulting dependence of the described procedure
which gives an \( \sim 200 \) m\( \Omega \) REE spin signal, measured at RT. Note
that this is considerably larger than the REE-associated spin
signal obtained via the in-plane geometry at RT, which is
consistent with an increase in REE efficiency with a lower carrier
density (estimated for holes from charge transport at 10 K, SI
section 2). More importantly, taking into account the change in
the sign convention of the current source electrodes (compared
with the in-plane measurements, Figure 2a), we observe that the
sign of the REE signal remains the same. This is because of the
fact that the REE spin polarization associated with holes should
have a sign opposite to that of electrons.27

Note that in the nonlocal geometry of the measurements in
such graphene Hall bars there is the possibility for a finite
contribution of a Hall effect, locally generated by the stray field
of the ferromagnetic electrodes, in both in-plane and out-of
plane magnetic field measurements. This contribution of the Co
stray fields in the nonlocal voltage as well as the regular Hall
effect (by the out-of-plane external magnetic field) is dependent
on the current distribution in the graphene channel in the region
in between the Co voltage probes. However, in this sample, we
observe that the linear change in the \( R_{\text{ul}} \) (associated with the
regular Hall effect, shown in the inset of Figure 5) is about 1
order of magnitude smaller than the Hall voltage expected from
the modeled current distribution in the region of Co detectors.
This discrepancy can be related to the inhomogeneities in the
graphene channel that may lead to smaller current flow in the
region of the Co probes. Considering also the possible
uncertainties in the determination of sample geometry, we
quantify the magnitude of the nonlocal voltage generated by the
stray fields to be about 1 order of magnitude smaller than the
measured signal in both in-plane and out-of-plane measure-
ments associated with the REE spins. Therefore, the effect of the
stray field in these measurements is unlikely to explain our
results. (For further details, see SI section 11.)

In this work, we also observe modulations of the second
harmonic signal \( (V_{\text{ul}}/I)^2 \). The results show a considerable
dependence on the applied magnetic field and gate electric field
which is a signature of thermally driven spin polarization in the
TMD-graphene heterostructure. This observation (results are
shown in SI section 13) could be an indication of spin-Nernst or
spin-Seebeck effects 58 in this system; however, it requires further
studies.

Our experimental observations are unambiguous evidence for
the presence of both Rashba–Edelstein and spin Hall charge-to-
spin conversion mechanisms in a monolayer TMD-graphene
heterostructure. This is the direct proof of the effective imprint
of the Rashba and valley-Zeeman spin–orbit fields in graphene,
while its charge-transport properties are preserved. In this work,
we comprehensively addressed the charge-induced nonequi-
librium spin density, generated by the REE, and we employed
strategies in order to discriminate this effect from SHE by
symmetries of the Hanle precession measurements as a function
of oblique magnetic fields. The ability to address the individual
effects in one heterostructure allows for a valid comparison of
their strengths. Moreover, the observed strong dependence of
the REE spin signal on the position of Fermi energy shows the
efficient tunability of spin generation by a transverse electric
field. This observation, in addition to the fact that the spin signal
remains considerable up to room temperature, confirms that the
monolayer TMD-graphene heterostructure is a promising
choice for the future of two-dimensional spin transistors without
the need for bulk ferromagnetic electrodes.

### METHODS

**Device Fabrication.** The monolayers of WS\(_2\) and graphene
and hBN (1L and bulk) are mechanically cleaved from their bulk
crystals (provided by HQ graphene) on SiO\(_2\)/Si substrates,
using adhesive tapes.59 The monolayer flakes are identified by
their optical contrast with respect to the substrate.40 The
thicknesses of the flakes are verified by atomic force microscopy
(AFM). Using a dry pick-up technique,31 we transfer the
graphene on the bulk hBN flake. By a prepatterned PMMA
mask, we etch the graphene-bulk hBN by oxygen-plasma into an
H-bar geometry. We finalize the fabrication of the vdW stack by
the transfer of the 1L hBN-WS\(_2\) on top of the etched graphene-
bulk hBN. We proceed with the fabrication of electrodes on the
dvW stack by shadow-mask evaporation and an e-beam
litography technique (using PMMA as the e-beam resist).
Because of the complications of the fabrication process, there is
a high chance of breaking the graphene channel. In the sample
studied in this work, the presence of a few cracks in our graphene
channel has caused the TMD-covered graphene region to be
electrically disconnected from the rest of the sample. Therefore,
our analysis is focused only on the TMD-covered graphene
region, shown in the device sketch of Figure 1b.

**Electrical Measurements.** The charge and spin transport
measurements are performed by using a standard low-frequency
(<20 Hz) lock-in technique with an ac current source of 100 nA
to 5 \( \mu \)A. A Keithley source meter is used as the dc voltage source for
the gate. Rotatable sample stages (separate for the in-plane
and out-of-plane measurements) are used to apply the magnetic
field by a (superconducting) magnet in all possible directions.

### ASSOCIATED CONTENT

#### Supporting Information

The Supporting Information is available free of charge on the
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Further details on device fabrication, AFM characterization,
charge transport, (anti)symmetrization of Hanle
precession measurements, contact magnetization behavior,
modelings, fittings and determination of REE and SHE
coefficients, y–z and x–z plane Hanle precession measurements,
the effect of stray fields, gate dependence
of the nonlocal measurements, and second-harmonic measurements (PDF)

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**Notes**

The authors declare no competing financial interest.

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■ NOTE ADDED IN PROOF
During the publication procedure, we became aware of very recent work on a similar system, where the ISHE and IREE are observed in a bulk WS₂-graphene heterostructure. 42