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**Magnetism, spin dynamics, and quantum transport in two-dimensional systems**

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Two-dimensional (2D) quantum materials offer a unique platform to explore mesoscopic phenomena driven by interfacial and topological effects. Their tunable electric properties and bi-dimensional nature enable their integration into sophisticated heterostructures with engineered properties, resulting in the emergence of new exotic phenomena not accessible in other platforms. This has fostered many studies on 2D ferromagnetism, proximity-induced effects, and quantum transport, demonstrating their relevance for fundamental research and future device applications. Here, we review ongoing progress in this lively research field with special emphasis on spin-related phenomena.

**Keywords: Quantum materials, spintronic, magnetic properties, graphene, Topological insulators**

## Introduction

Research on two-dimensional (2D) quantum materials (2DQMs) over the past decade has provided unique insights into condensed matter physics, yielding a multitude of novel effects.<sup>1,2</sup> Two-dimensional QMs, of which graphene is the prime example, include atomically thin materials with metallic, insulating, semiconducting, and magnetic behaviors, and may host intriguing topological properties.<sup>3,4</sup> Remarkable examples of novel phenomena have been continuously reported, such as the quantum spin Hall effect (QSHE) in monolayer WTe<sub>2</sub>,<sup>5</sup> 2D ferromagnetism,<sup>6</sup> strongly correlated states in graphene-based heterostructures,<sup>7</sup> and the observation of unconventional superconductivity in twisted graphene bilayers.<sup>8</sup>

Steady advances in 2D heterostructure fabrication techniques have yielded ultraclean samples providing a highly versatile approach to engineer materials properties. Van der Waals (vdW) heterostructures are a new class of artificial materials formed by a controlled assembly of different atomically thin crystals enabling the emergence of new electronic properties.<sup>1,2,7</sup> Among them, graphene-based heterostructures have attracted increasing attention.

While having high electrical mobility, pristine graphene is a material with low spin-orbit coupling (SOC), which has precluded the observation of the QSHE<sup>9</sup> and the control and manipulation of spin currents. Enhancement of the SOC has been realized by combining it with high SOC layered crystals such as transition-metal dichalcogenides (TMDCs).<sup>10</sup> Such SOC enhancement has been recently proved for these heterostructures,<sup>11–15</sup> featuring a building block for electric-field control of graphene spin properties.<sup>15,16</sup>

Another paradigmatic example of novel materials are topological insulators (TIs), which are characterized by electronic edge states that are protected from backscattering by time-reversal symmetry. The properties of TIs are derived from their bulk band structure following a band inversion driven by their large SOC.<sup>4</sup>

The aim of this article is to examine recent developments for 2DQMs, with particular emphasis on their technological relevance to spin manipulation, spin-orbit-torque memories, 2D ferromagnets, and magnetic sensing.

## **Proximity-induced effects and magnetism in bi-dimensional heterostructures**

### *Graphene-TMDC heterostructures*

Enhanced SOC in graphene interfaced with different TMDCs has been experimentally demonstrated by weak (anti-)localization,<sup>17–20</sup> and recently, by spin precession experiments.<sup>11,12,15</sup> In the latter, proximity-induced SOC in graphene is inferred from the anisotropic spin relaxation and the spin-charge interconversion (SCI) driven by both the spin Hall effect (SHE) and the inverse spin galvanic effect (ISGE), where charge currents are converted into transverse spin currents and non-equilibrium spin densities respectively. In their reciprocals, the inverse spin Hall effect (ISHE) and spin galvanic effect (SGE), spin currents and non-equilibrium spin densities generate transverse charge currents.<sup>13,14,15</sup>

Spin transport in graphene-TMDC heterostructures is well modelled by an effective hamiltonian, that describes the hybridization between graphene and d-orbitals in the TMDC<sup>21,22,23</sup>. In this model, graphene preserves its characteristic linear band structure close to the Fermi level, while the proximity-induced SOC contributions arising from the TMDC and the Rashba type-SOC generated by the lack of space inversion symmetry in the system, open a band gap and remove the spin degeneracy. The strong spin-valley coupling also called valley Zeeman (VZ) interaction produces the out-of-plane spin polarization with opposite directions at the K and K' valleys while the Rashba-type SOC induces a winding in-plane spin component.<sup>11,12,15,23</sup> Such spin textures are imprinted in graphene, having a significant impact on the spin dynamics, resulting in spin lifetimes varying over orders of magnitude for spins pointing in-plane and out-of-the graphene plane.<sup>11,12,15,24</sup>

This model also enables quantification of the SCI mechanisms arising from the (intrinsic) SHE and SGE.<sup>25,26</sup> For the SHE,<sup>27</sup> numerical calculations of the spin Hall conductivity  $\sigma_{xy}^z$ , considering low disorder, display a non-zero spin Hall angle, which quantifies SCI efficiency.<sup>25,28</sup> Such numerical calculations predicts large spin Hall angle values for graphene-WS2 heterostructures with asymmetric responses for electron and holes, which is a signature of intrinsic SOC in graphene.<sup>15</sup> Although that SCI strongly depends on the degree of disorder in the structure, calculations performed by Garcia *et al.*<sup>25</sup> indicate that VZ interaction plays a crucial role for non-vanishing SHE as it breaks the valley degeneracy and prevents its cancelation as electrons experiences multiple scatter events at opposite valleys. For the SGE,<sup>29</sup> SCI is mainly driven by the winding in-plane spin texture imprinted in graphene due to the Rashba SOC.<sup>26</sup> In this case, the VZ interaction enhances the band splitting, causing an out-of-plane tilting in the spin textures, but it is found that the overall effect on the magnitude of the SGE is minimal.

Such features were recently demonstrated by spin precession experiments in lateral spin devices<sup>15,30</sup> (**Figure 1a**). In spin precession measurements, an electrical current,  $I$ , is applied through the ferromagnetic (F) injector ( $\text{F}$ ), creating nonequilibrium spin accumulation beneath the contact. This spin accumulation diffuses along the graphene spin channel and is

detected by using a ferromagnetic detector. An external magnetic field applied in an oblique configuration allowed for determination of the spin lifetime for spin precession in-plane and out-of-plane, being one order of magnitude larger for the latter. These experimental results suggest that the spin relaxation in such heterostructures is mostly driven by intervalley scattering, that arises from short-range scattering centers such as structural defects in graphene or vacancies in the TMDC. These results are well reproduced by numerical simulations, which indicate that the in-plane and out-of-plane spin lifetimes follow different relaxation mechanisms.<sup>24,30</sup>

For SCI experiments, the devices in the design of Reference 15 are patterned into a Hall cross geometry, with the TMDC lying along one of the arms (Figure 1b). This Hall cross is contacted on the graphene pads underneath the TMDC with metal electrodes, so that the spin accumulation can be detected by SCI due to the SGE and the inverse spin Hall effect (ISHE), which are a consequence of the modified electron–hole states of graphene by proximity with the TMDC.<sup>13–15</sup>

The ISHE was first reported in heterostructures comprising multilayer graphene-MoS<sub>2</sub> and encapsulated graphene-WS<sub>2</sub> heterostructures (**Figure 2a–b**).<sup>13,14</sup> In contrast to theoretical predictions,<sup>25</sup> the larger SCI efficiency of the former is explained by including additional contributions associated with intrinsic SOC and staggered potentials.<sup>13</sup> More recently, in the case of WS<sub>2</sub> heterostructures,<sup>14</sup> the SHE and SGE contributions were differentiated from the symmetric and antisymmetric features of the experimental signals, which were also contrasted with numerical simulations.<sup>14</sup> Here, additional second harmonic signals suggest the onset of thermal effects. These experiments, however, do not independently monitor the electrical conductivity of the TMDC, thus preventing the full discrimination of SCI from proximity and TMDC-related effects.

In a recent report, proximity-induced SCI has been unambiguously demonstrated at room temperature in graphene–WS<sub>2</sub> heterostructures.<sup>15</sup> Here, the measurement protocol, using the device in Figure 1b, is based on spin precession to separate and quantify the SHE and SGE contributions (Figure 2c–d). Additionally, the SGE and SHE are shown to be tunable by electrostatic

gating, being large near the charge neutrality point, and in agreement with theoretical calculations (Figure 2e–f).<sup>25,26</sup> Notably, the magnitude of the equivalent SCI efficiency at room temperature, which takes into consideration the long spin relaxation length, is among the largest observed to date, holding promise for spin-logic applications.<sup>16</sup> These findings show the potential of graphene/TMDCs heterostructures for generation and manipulation of spin currents in 2D systems, which can be used to act on the magnetization through the spin torque phenomena, providing a promising approach toward ultracompact memory devices.

Despite reasonable agreement between theory and recent experiments, some open questions need to be addressed. For instance, intervalley scattering is predicted to produce a suppression of the SHE while being necessary for spin-lifetime anisotropy.<sup>24–26</sup> It turns out that both effects were measured in a recent experiment on the same device, thus contradicting the proposed theory.<sup>15</sup> Furthermore, the SGE is predicted to be proportional to the conductivity at the Rashba pseudogap,<sup>26</sup> which does not seem the case in the experiments. Besides, the spin-dependent disorder, such as magnetic impurities or vacancies, is believed to enhance the SHE, but it is not clear how this will manifest in the experiments. Finally, numerical calculations discard the complexity of measured device geometries, and while an upper limit is predicted, the interplay between the SCI, the geometry, and relaxation processes remains largely unexplored, and thus limits the optimization of practical devices.

### **Two-dimensional ferromagnets**

The recent discovery of intrinsic 2D ferromagnetism (2D-FM) in pristine vdW materials has opened new venues for spintronics, valleytronics, and quantum transport in layered heterostructures.<sup>31,32</sup> Long-range magnetism was first observed below room temperature in single and few layers exfoliated crystals of CrI<sub>3</sub>,<sup>6</sup> Cr<sub>2</sub>Ge<sub>2</sub>Te<sub>6</sub>,<sup>33</sup> and Fe<sub>3</sub>GeTe<sub>2</sub>,<sup>34</sup> and later at room temperature in epitaxially grown TMDCs such as VSe<sub>2</sub><sup>35</sup> and MnSe<sub>x</sub>.<sup>36</sup> Additional 2D-FMs include magnetically doped TMDCs,<sup>37,38</sup> the FePS<sub>3</sub> materials family,<sup>39</sup> and intercalated layered materials such as V<sub>5</sub>S<sub>8</sub>.<sup>40</sup> These materials present wide electronic and magnetic properties—CrI<sub>3</sub> is a 2D Ising FM with strong

anisotropy and interlayer antiferromagnetic ordering,<sup>6</sup> Cr<sub>2</sub>Ge<sub>2</sub>Te<sub>6</sub> is an insulating Heisenberg FM with weak anisotropy,<sup>33</sup> Fe<sub>3</sub>GeTe<sub>2</sub> is a semimetallic itinerant 2D Ising FM,<sup>34</sup> whereas doped TMDCs are diluted FM semiconductors.<sup>37</sup>

This wealth of attributes allows us to experimentally test models of magnetism in low dimensions by tuning the anisotropy and long-range interactions, both essential aspects for the emergence of 2D magnetism. For instance, the Curie temperature of Fe<sub>3</sub>GeTe<sub>2</sub> was found to be strongly sensitive to the shape anisotropy.<sup>41</sup> In addition, the low-dimensional nature of 2D-FMs introduces new degrees of freedom acting on the magnetic order. Interestingly, the Curie temperature of Fe<sub>3</sub>GeTe<sub>2</sub> can be increased above room temperature by electrostatic gating with an ionic liquid.<sup>34</sup> Also noteworthy is electric-field control of the interlayer magnetic coupling, which was also achieved in CrI<sub>3</sub> where the ferromagnetic/antiferromagnetic transition can be triggered electrically.<sup>6,42</sup> Two-dimensional-FMs can further be employed as building blocks of spintronics devices. Tunneling through insulating CrI<sub>3</sub> has been used as a probe of the magnetic order, yielding large magnetoresistance as a result of the controlled tunneling of spin polarised currents, which depends on the specific magnetic arrangement of the layers (spin filtering effect). Such behavior, shown in **(Figure 3a-c)** has been recently reported using electrical and optical means.<sup>43-45</sup>

Spin-orbit torques and current-induced magnetization switching were also reported in Pt/Fe<sub>3</sub>GeTe<sub>2</sub>.<sup>46,47</sup> A large anomalous Hall effect arises in Fe<sub>3</sub>GeTe<sub>2</sub> due to its band topology and large Berry curvature,<sup>48</sup> effective magnetic field generated by the symmetries and high SOC of the system, and in Cr<sub>2</sub>Ge<sub>2</sub>Te<sub>6</sub> proximitized to a TI.<sup>49</sup> Two-dimensional-FMs are additionally expected to enable substantial progress in valleytronics, which aims at exploiting the electronic valley degree of freedom in information processing. The magnetic proximity effect in TMDC/2D-FM bilayers could lift the valley degeneracy in TMDCs. Alternatively, a “ferrovalley” ground state could be achieved in ferromagnetic TMDCs such as 2H-VSe<sub>2</sub><sup>50</sup> or diluted magnetic TMDCs.

The practical use of 2D-FMs will however require higher Curie temperatures and the development of scalable fabrication methods. Various



proximity effects still demand experimental validation, requiring smart design of 2D-FMs and vdW heterostructures.

### **Spin-orbit torques in FM/2D heterostructures**

The large SOC and resulting spin-momentum textures of 3D-TIs and TMDCs make them ideal materials for SCI through the SGE.<sup>29</sup> At the interface with a magnetic layer, the nonequilibrium spin density generated by SGE transfers angular momentum to the magnetization,  $\mathbf{m}$ , and exerts torque on it.<sup>51</sup> This spin-orbit torque (SOT) is of high relevance for technological applications as it can electrically induce magnetization oscillations or magnetization switching. SOT is currently a subject of intense investigation in TI/FM, and more recently in TMDC/FM bilayers.

The phenomenology of SCI in TIs is similar to that in Rashba systems. However, in the latter, split bands possess opposite spin helicities, resulting in partially compensated nonequilibrium spin densities under current flow. In contrast, the surfaces of TIs possess an even number of helical bands, and therefore can, in principle, generate larger net spin densities and SOTs. SOTs have been measured in TI/FM and TMDC/FM bilayers by second harmonic Hall measurements<sup>52,53</sup> and SOT-induced ferromagnetic resonance.<sup>54,55</sup> Some of the most studied TIs have been molecular beam epitaxially grown  $\text{Bi}_2\text{Se}_3$  and  $(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_3$ , which possess a single band at the Fermi energy.

A large damping-like torque (torque odd in  $\mathbf{m}$ ) has been reported in TI/FM bilayers,<sup>54,55</sup> which enabled the achievement of SOT-induced magnetization switching<sup>52,53,56</sup> (e.g., Figure 3d-e) and confirmed the high potential of TIs for spintronics. On the other hand, there is little evidence of a large field-like torque (even in  $\mathbf{m}$ ) that would be expected as the hallmark of Rashba/Dirac SOTs, according to the analytical 2D Rashba and Dirac gas models.<sup>57,58</sup> Several mechanisms giving rise to a damping-like torque have been proposed—spin relaxation in the FM, the SHE due to the finite bulk conductivity of the TI, or the intrinsic contribution from the Berry curvature.<sup>51</sup> However, disentangling their relative contributions in experiments remains challenging.

The SCI efficiency,  $\xi$ , inferred from the measured antidamping SOT is often found to be larger than in conventional heavy metals. However, the

reported values span several orders of magnitudes ( $\xi \sim 0.08 - 52$  as compared to spin Hall efficiencies of  $|\xi| \sim 0.06 - 0.33$  with Pt, Ta, and W).<sup>51</sup> The large scatter of experimental results and the difficulty in unambiguously determining the microscopic origin of the SOTs can be ascribed to several experimental limitations. The determination of  $\xi$  requires knowing the spatial distribution of the charge current, which is made difficult by the use of low resistivity FMs that strongly shunt the current. TI/metal interfaces also suffer from substantial charge transfer and orbital hybridization.<sup>59,60</sup> This strongly alters the energy and spin texture of topological surface states and leads to the formation of a Rashba-split 2DEG coexisting with them.<sup>61</sup>

Moreover, TI thin films are usually defective, with a sizable bulk conductivity that allows a contribution of the SHE to the SOTs. Chalcogenides tend to intermix with metals, forming poorly defined interfaces<sup>62,63</sup> and magnetic dead layers<sup>64</sup> that may strongly alter the SOTs. Most of these drawbacks can be circumvented by the use of FM insulators or vdW FMs with atomically sharp interfaces. So far, SOTs have been measured in  $(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_3/\text{Cr}_y(\text{Bi}_{1-x}\text{Sb}_x)_{2-y}\text{Te}_3$ <sup>52,53</sup> (e.g., Figure 3d–e) and  $\text{Bi}_2\text{Se}_3/\text{BaFe}_{12}\text{O}_{19}$ <sup>65</sup> at cryogenic temperatures, where large SCI efficiency and magnetization switching were reported. Further progress is foreseen by interfacing TIs with 2D-FMs exhibiting strong perpendicular anisotropy and stable ferromagnetism down to the monolayer limit.

The torques in TMDC/FM bilayers are generally weaker than in TI/FM ones, but interestingly, SOTs with unconventional symmetries may be produced due to the reduced crystal symmetry of the TMDC layers. Of relevance for actuating perpendicularly magnetized FMs, an out-of-plane damping-like torque has been observed in  $\text{WTe}_2/\text{Fe-Ni}$ <sup>66</sup> and  $\text{NbSe}_2/\text{Fe-Ni}$ .<sup>67</sup> The underlying mechanisms governing the symmetry and magnitude of SOTs remain to be elucidated. TMDCs further enable the study of SOTs in ultimately thin devices, down to monolayer TMDCs,<sup>68</sup> and potentially, their tuning with light or electrostatic gating.

## Quantum spin transport in bidimensional heterostructures

### *Graphene-based devices*

Quantum transport driven by the quantum Hall effect (QHE) appears in high-mobility 2DQMs. In the QHE, the band structure of the 2DQM turns into discretized energy levels usually called Landau's levels (LLs) by the action of a strong magnetic field, leading to dissipationless quantum conduction. The QSHE emerges in the absence of magnetic fields in 2DQMs that are bulk insulators and possess conducting helical spin-edge states protected by time reversal symmetry.<sup>4</sup> The QSHE was initially observed in quantum well semiconductor systems.<sup>69</sup> Owing to the small band gap in these systems, the conductance quantization manifests at ultralow temperatures. This has promoted recent research toward new 2DQMs, where graphene and TMDCs have risen as promising candidates due to their unique and tunable electronic properties.<sup>3,7</sup>

In graphene, early QHE experiments showed a characteristic quantization, arising from its linear band structure, topology, and the fourfold degeneracy of the LLs.<sup>70,71</sup> Interestingly, the energy spectrum driven by the competition between spin and sublattice (valley) interactions displays a zero LL at the Dirac point<sup>72</sup> and Hall conductivity  $\sigma_{xy} = 4e^2/h(N+1/2)$ , where  $e$  is the electron charge,  $h$  is the Planck constant, and  $N$  is the index characterising each LL (e.g., **Figure 4a**). Further studies of QHE on ultraclean monolayer and bilayer graphene devices<sup>73–76</sup> have shown the emergence of a ferromagnetic QH regime, where the bulk ground state is given by the interplay between the Zeeman, Coulomb, and disorder interactions.<sup>7,73</sup> At half filling of the zero LL, temperature dependence studies of the energy gaps showed that valley interactions are dominant, leading to a ground insulating state characterized by a canted antiferromagnetic (CAF) order, where spins at opposite sub-lattices cant respect to each other forming a non collinear spin arrangement. For  $N \neq 0$ , Zeeman interaction dominates, inducing a ferromagnetic state. Notably, at the zero LL, graphene can also host spin polarized counterpropagating modes.<sup>73,74,77</sup> Such an edge mode configuration is analogous to the intrinsic QSHE<sup>9</sup> and can also be modulated from chiral to helical edge modes transport by adding an in-plane magnetic field.<sup>74</sup>

Recently, the fractional quantum Hall effect, consisting on the non-integer quantization of the Hall conductivity ( $\sigma_{xy}$ ) as a result of the interaction between electrons at partially filled LLs, has been observed in dual-gated

encapsulated devices. The ultralow disorder of these encapsulated devices enabled observation of striking QH features at low magnetic fields.<sup>7,78</sup> The quantification of the LL broadening by thermal activation gap measurements is found to be comparable to high-quality GaAs quantum wells.<sup>78</sup> Spin-transport studies through the CAF state in graphene have been recently reported.<sup>75,79</sup> Notably, Stepanov *et al.*<sup>75</sup> demonstrated spin current propagation over channels up to 5  $\mu\text{m}$  long by using dual-gated devices to control the antiferromagnetic order of the channel. The temperature and magnetic field dependence of the signals indicate that spin-transport properties are driven by a spin-superfluid mechanism.<sup>80</sup> Long and tunable spin propagation over the antiferromagnetic (AF) state shows the potential of graphene in the QH regime as a model system for exploring AF and ferromagnetic spintronics.

More recently, evidence of ferromagnetism<sup>81,82</sup> and the quantum anomalous HE (QAHE)<sup>82</sup> have been reported in twisted bilayer graphene superlattices. The QAHE is the quantization of the Hall conductivity  $\sigma_{xy}$  induced by a spontaneous magnetization of the system in the absence of any external magnetic field. Remarkably, the anomalous Hall resistance ( $R_{xy}^{AH}$ ) resulting from enhanced electron–electron interaction displays hysteretic behavior in a narrow range of carrier densities with a sharp peak near  $3/4$  filling of the conduction band (e.g., Figure 4b). Moreover, the current-induced magnetization reversal is found to be orders of magnitude smaller than in conventional systems.<sup>27,51</sup> These findings provide valuable insights into QAHE physics in graphene-based Moiré superlattices with promising applications in low-power magnetic devices.

### Transition-metal dichalcogenide heterostructures

$\text{MX}_2$  materials possessing the  $1T'$  crystallographic structure, consisting of an octahedral geometry with M atoms forming a 1D zigzag chains, where  $M = (\text{W}, \text{Mo})$  and  $X = (\text{Te}, \text{S}, \text{Se})$ , have shown to be potential large gap QSH insulators.<sup>3</sup> Among them,  $\text{WTe}_2$  is a very promising candidate due to its natural  $1T'$  ground state structure. Angle-resolved photoemission spectroscopy on epitaxially grown  $1T'$ - $\text{WTe}_2$  showed a modification of the electronic band structure in accordance with a QSH insulator.<sup>83</sup> Remarkably, QSHE experiments on exfoliated crystals of  $\text{WTe}_2$  using transport measurements have

demonstrated quasi-quantized edge conduction states independent of the carrier concentration in the monolayers limit, at temperatures up to 100 K. However, the lack of saturation to the expected quantum ( $e^2/h$ ) plateau suggests the presence of elastic scattering at the edges influenced by the contacts.<sup>84</sup>

QSHE was recently reported in  $WTe_2$  fully encapsulated with hexagonal boron nitride (hBN).<sup>5</sup> By using local top/bottom electrostatic gating integrated in the devices, robust edge conduction was demonstrated to survive over lengths up to 100 nm, being only sensitive to external magnetic fields. Additionally, the electric-field and temperature dependence of the conductance display exponential behavior, in agreement with the expected band gap opening due to Zeeman interaction (Figure 4c). The prevalence of the QSHE up to 100K sheds light toward realization of low-dissipation vdW devices working above cryogenic temperatures.<sup>3</sup>

### **Conclusions and perspective**

In spite of the fast development of the field of 2DQM, several questions remain to be addressed in order to use them as potential components for spintronic and quantum applications. From a practical point of view, large-scale fabrication techniques enabling growth of high-quality 2D crystals with controlled interfaces need to be developed. In vdW heterostructures, enhancement of the proximity-induced SCI mechanisms in order to use them for magnetic manipulation via SOT requires further theoretical and experimental studies to determine the relevance of extrinsic mechanisms and improve the SCI efficiency. Moreover, improvement of interface quality by using vacuum controlled environments and the use of new materials such as  $MX_2$  crystals, which have been predicted to host topological effects, might help to enhance SCI. High-quality fabrication and design of 2D ferromagnet-based vdW heterostructures, possessing sharp interfaces, small magnetic volume, and electric-field control could lead to lower critical currents for magnetization switching. Additionally, the discovery of high Curie temperature 2D magnetic materials having out-of-plane anisotropy will promote their fast insertion for ultracompact memory and storage devices. For

quantum transport, further studies of the QSHE in ultralow disorder vdW devices might lead to long-distance edge conduction at high temperatures.

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## Figure captions

**Figure 1.** (a) Nonlocal spin precession measurement in a typical lateral spin device. (b) Schematic representation of a graphene/transition-metal dichalcogenide Hall cross for studying spin-charge interconversion mechanisms driven by the spin Hall effect (SHE) and spin galvanic effect (SGE). Adapted with permission from Reference 15. © 2020 Nature Publishing Group. Note:  $F_1$ , ;  $F_2$ ;  $\mathbf{B}_\perp$ , ;  $\mathbf{B}_\parallel$ ;  $V_{nl}^+$ , ;  $V_{nl}^-$ ,  $\mathbf{M}_1$ ;  $I_T$ .

Figure 1. (a) Nonlocal spin precession measurement in a typical lateral spin device composed by ferromagnetic (gray wires) and metallic contacts (gold wires). (b) Schematic representation of a graphene/transition-metal dichalcogenide Hall cross for studying spin-charge interconversion mechanisms driven by the spin Hall effect (SHE) and spin galvanic effect (SGE). The blue and red arrows in (b) represent the non-equilibrium spin density and spin accumulation induced by the inverse spin galvanic effect and spin Hall effects

respectively when an electric current  $I_T$  is applied to the device. Adapted with permission from Reference 15. © 2020 Nature Publishing Group. Note:  $F_1$ ,  $F_2$ , are the spin sensitive ferromagnetic injector and detectors respectively;  $B_\perp$ ,  $B_\parallel$  are the out-of-plane and in-plane external magnetic fields used to induce spin precession;  $V_{nl}^+$ ,  $V_{nl}^-$  are the non-local voltage probes with their associated polarities (+,-),  $M_1$ , is the magnetization polarization.

**Figure 2.** (a) ISHE) measurement at 10 K in multilayer graphene-MoS<sub>2</sub> Heterostructures. Reprinted with permission from Reference 13. ©2019 American Chemical Society. (b) Spin Hall effect (SHE) and Rashba Edelstein effects in encapsulated graphene-WS<sub>2</sub> devices at 4.2 K. Adapted with permission from Reference 14. © 2019 American Chemical Society. (c, d) Room-temperature detection of the spin Hall (SH [red]) and spin galvanic effects (blue). (e) Theoretical calculation of the SH conductivity as a function of the carrier density for three different temperature values. The inset shows the schematic representation of the spin bands splitting in graphene, induced by proximity with a transition-metal dichalcogenide. (f) Temperature evolution of the SHE signals as a function of the carrier density ( $n$ ). Adapted with permission from Reference 15. © 2020 Nature Publishing Group. Note:  $R_{nl}$ ,  $R_{SHE}$ ,  $R_{SGE}$ ,  $B_x$ ,  $e$ , electron charge;  $h$ , Planck constant;  $k_x$ ,  $k_y$ ,  $E$ ,  $\epsilon$ , ISHE, inverse spin Hall effect, IREE, .

**Figure 2.** (a) Inverse spin Hall effect (ISHE) measurement in multilayer graphene-MoS<sub>2</sub> Heterostructures at 10 K. The black and red courbes are similar measurements performed for opposite magnetic configuration. Reprinted with permission from Reference 13. ©2019 American Chemical Society. (b) Spin Hall effect (SHE) and Rashba Edelstein effects in encapsulated graphene-WS<sub>2</sub> devices at 4.2 K. The top left inset displays the resulting symmetric and antisymmetric components of the signal associated to the SHE and REE respectively. The low bottom inset shows the experimental results obtained in the reciprocal configuration, inverse Rashba Edelstein effect (IREE). Results in (a-b) evidenced SCI in graphene-transition dichalcogenides heterostructures at low temperatures. Adapted with permission from Reference 14. © 2019 American Chemical Society. (c, d) Experimental detection of the spin Hall ([red]) and spin galvanic (SG) effects (blue) in graphene-WS<sub>2</sub> devices possessing the geometry shown in figure 1b. The solid black lines in both plots represent the fitting of the experimental data with the solution of the spin diffusion equations (e) Numerical calculation of the SH conductivity versus the carrier density ( $n$ ) for three different temperature values. The inset (left) shows the schematic representation of the energy bands splitting in graphene ( $\epsilon$ ) as a function of the wave vectors ( $k_x$ ,  $k_y$ ), induced by proximity with a transition-metal dichalcogenide. The blue and purple bands represent opposite spin helicities denoted by black arrows; Right, Spin texture and resulting non-equilibrium spin density (blue arrow) due to the SG effect and the action of an electric field ( $E$ ). (f) Temperature evolution of the experimental ISHE signals as a function of the carrier density ( $n$ ). (c-f)

demonstrate room temperature SCI and the carrier-type dependence of the ISHE in correct agreement with existing theoretical models. Adapted with permission from Reference 15. © 2020 Nature Publishing Group. Note:  $R_{nl}$ , non-local spin signals  $R_{ISHE}$ , spin precession signal due to the ISHE;  $R_{SGE}$ , spin precession signal due to SGE;  $B_x$ , in-plane magnetic field;  $e$ , electron charge;  $h$ , Planck.

**Figure 3.** (a) Schematic of a two-dimensional spin-filter magnetic tunnel junction, with bilayer  $\text{CrI}_3$  functioning as the spin-filter sandwiched between few-layer graphene contacts. (b) Tunneling current ( $I_t$ ) as a function of out-of-plane magnetic field ( $H$ ). Green (orange) curve corresponds to decreasing (increasing) magnetic field. (c) Reflective magnetic circular dichroism (RMCD) of the same device. Insets show the corresponding magnetic configuration of the  $\text{CrI}_3$  bilayer. (d) Schematic illustration of current-pulse-induced magnetization switching in  $(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_3/\text{Cr}_y(\text{Bi}_{1-x}\text{Sb}_x)_{2-y}\text{Te}_3$ . (e) Hall resistance measurement ( $R_{xy}$ ) of current-induced magnetization switching. (a–c) Reprinted with permission from Reference 45. © 2018 AAAS. (d, e) Reprinted with permission from Reference 53. © 2017 American Physical Society. Note:  $H_{\perp}$ ,  $\mu_0$ ,  $\mathbf{M}$ ,  $\mathbf{B}$ ,  $\sigma$ ,  $J_{\text{pulse}}$ ,  $j_{\text{pulse}}$ ,  $\mathbf{B}_{\text{DL-SOT}}$ ,  $M_z$ .

**Figure 3.** (a) Schematic of a two-dimensional spin-filter magnetic tunnel junction, with bilayer  $\text{CrI}_3$  functioning as the spin-filter sandwiched between few-layer graphene contacts encapsulated with hexagonal boron nitride (hBN). (b) Tunneling current ( $I_t$ ) as a function of out-of-plane magnetic field ( $H$ ). Green (orange) curve corresponds to decreasing (increasing) magnetic field. Inset shows the electric configuration used to measure the tunneling current. Central purple layers represent  $\text{CrI}_3$  while outer gray layers correspond to the graphene contacts. (c) Reflective magnetic circular dichroism (RMCD) of the same device. Insets show the corresponding magnetic configuration (black arrows) of each  $\text{CrI}_3$  layer, Above (below) 0.5T, the magnetizations are parallel (antiparallel), which is associated to the low (high) resistance in state in (b). These results demonstrate large magnetoresistance in vdW heterostructures. (d) Schematic illustration of current-pulse-induced magnetization switching in  $(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_3/\text{Cr}_y(\text{Bi}_{1-x}\text{Sb}_x)_{2-y}\text{Te}_3$ . (e) Hall resistance measurement ( $R_{yx}$ ) of current-induced magnetization switching. This work shows magnetization switching at low current densities, indicating large spin orbit torque in topological insulator-based heterostructures (a–c) Reprinted with permission from Reference 45. © 2018 AAAS. (d, e) Reprinted with permission from Reference 53. © 2017 American Physical Society. Note:  $H_{\perp}$ , is the out-of-plane magnetic field;  $\mu_0$ , is the magnetic permeability;  $\mathbf{M}$ , is the magnetization;  $\mathbf{B}$ , is the external magnetic field;  $\sigma$ , is the spin current polarization;  $J_{\text{pulse}}$ , is the current pulse amplitude;  $j_{\text{pulse}}$ , is the current pulse density;  $\mathbf{B}_{\text{DL-SOT}}$ , is an effective field induced by the spin orbit coupling in the system that acts on the magnetization;  $M_z$ , is the projection of the magnetization along the z axis depicted in (a).

**Figure 4.** (a) Quantum Hall (QH) effect experiments showing unconventional Landau quantization in graphene-based devices. Left upper inset shows the longitudinal  $R_{xx}$  (black) and transverse  $R_{xy}$  (green) resistances, respectively. The lower left inset shows Landau level splitting for low and strong magnetic fields. Right inset displays the Hall conductivity ( $\sigma_{xy}$ ) near the charge neutrality point. The colors represent different magnetic field values. Reprinted with permission from Reference 72. ©2006 American Physical Society. (b) Temperature ( $T$ ) dependence of the QH conductivity for several carrier concentrations, exhibiting deviation from the quantum Hall regime above 100 K. Inset displays the Hall resistance evolution as a function of the gate for different temperatures. Reprinted with permission from Reference 5. © 2005 AAAS. (c) Zero-field anomalous (red) and ordinary (blue) Hall resistance as a function of carrier density extracted from magneto-transport measurements. Reprinted with permission from Reference 81. © 2019 AAAS. Note:  $R$ , resistance;  $V_g$ , ;  $n$ , ;  $n_s$ , ;  $V_c$ , ;  $R_{xy}^{AH}$ , ;  $R_H$ ,

**Figure 4.** (a) Quantum Hall (QH) effect experiments showing unconventional Landau quantization in graphene-based devices. The colors in the main panel represent different magnetic field values varying from 9T (pink) to 25T (black). Left upper inset shows the longitudinal  $R_{xx}$  (black) and transverse  $R_{xy}$  (green) resistances, respectively. The lower left inset shows Landau level splitting for low and strong magnetic fields. Right inset displays the Hall conductivity ( $\sigma_{xy}$ ) versus backgate ( $V_c$ ) near the charge neutrality point for  $B=9T$  (pink),  $B=11.5T$  (pentagon) and  $B=17.5T$  (hexagon) at  $T=30mK$ . This work evidenced additional QH states, indicating the lifting of the spin and sublattice degeneracy in graphene. Reprinted with permission from Reference 72. ©2006 American Physical Society. (b) Zero-field anomalous (red) and ordinary (blue) Hall resistance as a function of ( $n/n_s$ ) extracted from magneto-transport measurements. These results evidenced ferromagnetic order and current-induced magnetization switching in graphene-based Moire superlattices. Reprinted with permission from Reference 81. © 2019 AAAS. (c) Temperature ( $T$ ) dependence of the edge conductance ( $G_s$ ) for different local backgate values ( $V_c$ ), exhibiting deviation from the quantum spin Hall (QSH) regime above 100 K. Bottom insets represent the transition from purely edge (left) to bulk transport (right). Top inset displays the Hall resistance evolution as a function of an electrical gate ( $V_g$ ) for different temperatures. These results demonstrate high temperature quantum spin Hall in van der Waals heterostructures. Reprinted with permission from Reference 5. © 2005 AAAS Note:  $R$ , is the electric resistance;  $n$ , carrier density;  $n_s$ , superlattice density; , ;  $R_{xy}^{AH}$ , is the anomalous Hall resistance ;  $R_H$ , is the ordinary Hall resistance.

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