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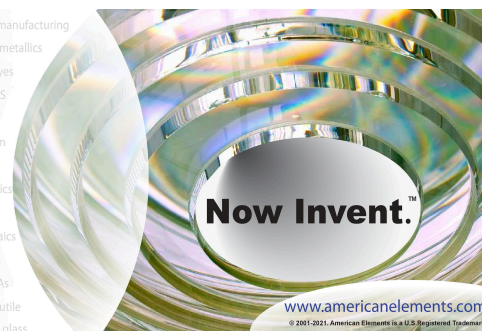
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ABSTRACT

The interconversion between spin and charge degrees of freedom offers incredible potential for spintronic devices, opening routes for spin injection, detection, and manipulation alternative to the use of ferromagnets. The understanding and control of such interconversion mechanisms, which rely on spin–orbit coupling, is therefore an exciting prospect. The emergence of van der Waals materials possessing large spin–orbit coupling (such as transition metal dichalcogenides or topological insulators) and/or recently discovered van der Waals layered ferromagnets further extends the possibility of spin-to-charge interconversion to ultrathin spintronic devices. Additionally, they offer abundant room for progress in discovering and analyzing novel spin–charge interconversion phenomena. Modifying the properties of van der Waals materials through proximity effects is an added degree of tunability also under exploration. This Perspective discusses the recent advances toward spin-to-charge interconversion in van der Waals materials. It highlights scientific developments which include techniques for large-scale growth, device physics, and theoretical aspects.

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I. INTRODUCTION

In the search for continued miniaturization of microelectronic devices, two-dimensional (2D) materials, such as graphene and its siblings, offer opportunities to radically change the technology landscape by enabling the discovery of revolutionary device paradigms.^{1,2} Although the path to technological implementation is long and hard, the extraordinary burst of 2D material synthesis during the last decade allowed for the fabrication of a wide variety of 2D materials,^{1,3} opening thrilling avenues for the design of innovative atomically flat devices. 2D materials possess strong covalent bonds between atoms in the plane, but their out-of-plane

interaction is mediated by 1000 times weaker van der Waals (vdW) forces, which enables an easy delamination of these layered materials. The interfaces obtained are defect-free and extremely sensitive to their environment.⁴ This offers a unique playground of study, as the properties of one material can be transferred to its neighbor by proximity effects when stacking different materials in complex vdW heterostructures. While the device performance is yet to challenge the existing traditional technology,⁵ their ability for unconventional operation nurtures the search for innovative concepts, such as the inclusion of the spin, pseudospin, and valley degrees of freedom.^{6,7}

To address the issues related to the energy loss in devices (e.g., gate leakages, interconnects, and refreshing non-volatile

memories), spin angular momentum can be used to store, transmit, and manipulate information with high efficiency in terms of energy consumption.^{2,8–11} In fact, highly efficient spin transport^{12–14} and long spin diffusion^{15–21} can be achieved in graphene and transition metal dichalcogenides (TMDs),^{22,23} enabling the fabrication of spin-field effect transistors^{24–26} and novel spin filtering devices.^{27,28} However, traditional spin-devices require spin injection from magnetic elements, usually made of transition metal materials such as NiFe or CoFe, and the application of magnetic fields to define their magnetization orientation. This is a challenge because good interface quality between the ferromagnet (FM) and the 2D vdW substrate is of critical importance and difficult to achieve.^{12,13,29} Furthermore, the use of magnetic fields to switch the magnetization limits the density of magnetic elements. These limitations severely hindered the performance of flat spintronic devices despite the apparent appeal of 2D systems. However, they may be circumvented and new opportunities may arise by adopting different strategies. These range from the use of recently discovered 2D magnetic materials (CrSe₂, VSe₂, etc.) to the exploitation of the spin–charge interconversion (SCI) that takes place in materials with large spin–orbit coupling (SOC), among which we find TMD semimetals and topological insulators (TIs). The SCI enables electrical generation of spin currents or spin densities, which in turn can be used to generate a torque on an adjacent FM, thus allowing for all-electrical manipulation of the magnetization direction in the FM.

SOC is the responsible interaction for the locking between the spin and the orbital angular momenta and results in a large number of physical phenomena, such as magnetic anisotropy and damping,^{30,31} spin relaxation,³² and the spin Hall effect (SHE).^{33,34} The latter is of particular importance for spintronic technologies because it offers a route for the electrical generation of spin currents, one of the most crucial limiting factors in current devices. This is why in recent years much attention is drawn toward materials presenting large SOC and inversion symmetry breaking, both needed for most of the spin–orbit phenomena of interest. In addition, a particular type of SOC that enables the direct coupling between the spin of the electron and its linear momentum can be used for the electrical generation of non-equilibrium spin density, offering a different path for the electrical manipulation of magnets through the spin. Such an effect is referred to as the inverse spin galvanic effect (iSGE) or the Rashba–Edelstein effect.

Recently, researchers proposed to use the iSGE^{35,36} and SHE^{37,38} to generate current-driven spin–orbit torque (SOT), enabling the electrical manipulation of ultrathin magnets.^{39–41} The recent observation of current-driven magnetization switching in inversion symmetry broken transition metal systems (Pt/Co,⁴² Ta/NiFe,⁴³ etc.) initiated a massive amount of effort in this direction. Ultrafast switching was demonstrated,^{44,45} and SOT is currently considered a promising method for the next generation of magnetic random-access memories, as explicitly stated in the international roadmap for devices and systems and currently being pursued by major industries.⁴⁵

Among the vast catalog of novel 2D materials, TMDs stand out as a versatile platform for the advancement of disruptive flat microelectronics and spintronics.^{5,6,11,46–48} Indeed, in contrast to graphene, TMDs have a large SOC^{49–51} that was successfully exploited for gate-controlled spin manipulation in graphene/TMD interfaces.^{25,26,52} In addition, some TMDs, such as WS₂ and MoS₂,

possess a large bandgap that promotes optoelectronic operation^{53,54} and the realization of light-driven spin–valley coupling.^{55–60} Beyond the most commonly found semiconducting phase, the TMD family also exhibits a wide variety of electronic behaviors ranging from a Weyl semi-metallic state^{61–63} to superconductivity^{64,65} as well as magnetism.⁶⁶ These features are particularly appealing for the realization of vdW heterostructures^{50,51} that could combine all of them.^{25,26}

Another important class of vdW materials concern bismuth-antimony chalcogenides that display topological properties. TIs have an insulating bulk and spin–orbit coupled surface states.^{67,68} The large spin–momentum locking of their surface states makes them ideal candidates for SCI. Recent progress includes the realization of current-driven SOT in systems using Bi-chalcogenide TIs (Bi₂Se₃/NiFe)^{69,70} and the realization of current-driven switching at room temperature^{71–73} with record low currents required. Note that SOT also was realized using TMD monolayers^{74,75} but with a much weaker efficiency as compared to TIs.

In this Perspective, we present the current state of the art and perspectives on the control of SCI in vdW materials toward the realization of ultra-compact spintronic devices. The complexity of the studied phenomena and the continued surge of new materials make synergy between experiment and theory particularly useful in developing improved large-scale growth methods of such vdW materials to facilitate their implementation in upcoming technologies.

II. GROWTH AND MANIPULATION OF 2D MATERIALS AND VDW HETEROSTRUCTURES

A. Chemical vapor deposition (CVD)

Currently, most 2D materials for fundamental research are produced by mechanical exfoliation, which ensures high crystal quality but with limited yield and size.⁷⁶ Since the report on the synthesis of graphene using chemical vapor deposition (CVD),⁷⁷ in which volatile precursors react or decompose, this method developed into a popular and reliable technique for the synthesis of different 2D materials, such as h-BN, MoS₂, WSe₂, CrI₃, CrTe, MoS₂Se, and 2D vdW heterostructures^{50,78} (Fig. 1).

For wafer-scale synthesis of 2D materials, a number of approaches were developed, including precise control of the growth process,^{79–81} substrate engineering,^{82–84} chemical doping,^{85–87} transfer,^{88–90} surface morphology,^{91–93} and twisted bilayers.^{94–96} Interestingly, adlayer-free large-area single-crystal graphene was realized on single-crystal Cu foil,⁹⁷ which was obtained from commercial Cu foil by the contact-free annealing method.⁹⁸ The CVD growth of h-BN, a popular 2D insulator resembling the hexagonal crystal structure of graphene, and 2D heterostructures of h-BN with graphene and TMDs attracted great interest.^{99–101} For example, large-area high-quality h-BN was synthesized on single-crystal Cu.¹⁰²

Many 2D materials with excellent semiconductor characteristics and high current on/off ratios, such as MoS₂, MoSe₂, WS₂, and WSe₂, were fabricated using CVD.^{78,103,104} In particular, stable and controllable growth of large-scale MoS₂ films was achieved by a number of research groups.^{105,106} A growth and transfer method of large-area high-quality single-crystal MoS₂ was also reported.¹⁰⁷ Using S and MoO₃ as the growth source and Ar and Ar/O₂ as

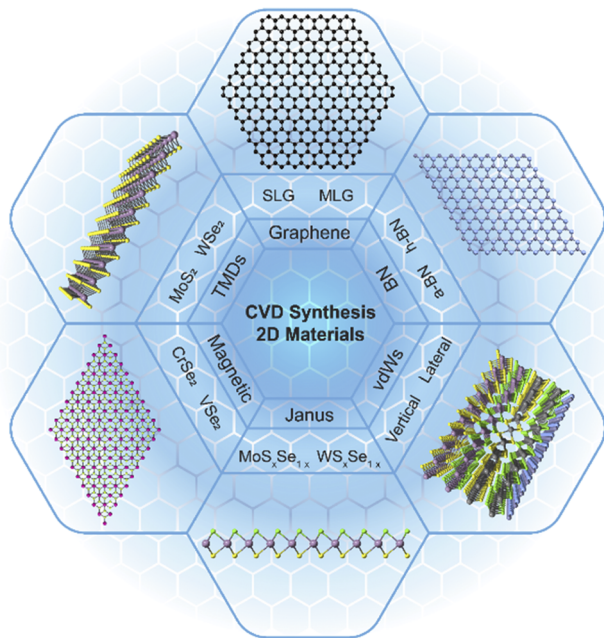


FIG. 1. 2D materials synthesized by the CVD method: graphene, BN, TMDs, magnetic 2D materials, 2D Janus materials, and 2D van der Waals heterostructures.

the carrier gas, the growth was successfully achieved at 930 °C on the sapphire substrate. In addition to MoS₂, there were attempts to grow MoSe₂ by CVD.^{108,109} A promoter-assisted liquid-phase CVD method was reported to synthesize high-quality large-area monolayer MoSe₂.¹¹⁰ A method to grow wafer-scale monolayer WS₂ used H₂S gas instead of sulfur powder, as the sulfur source reacts

after decomposition with WO₃ powder, eventually leading to a 4-in. wafer-scale single-layer WS₂ film.¹¹¹ Going beyond individual TMDs, synthesis and precise control of 2D vdW heterostructure arrays was also achieved.¹¹²

For applications in the field of spintronics, potential magnetic 2D materials include FeX₂, CoX₂, NiX₂, VX₂, CrX₂, and MnX₂ (X = I, Br, and Cl).¹¹³ For the synthesis of large-scale 2D transition metal tellurides by CVD, the use of a mixture of Cr, CrCl₃, and Te was reported to lead to a CrTe film.¹¹⁴

B. Molecular beam epitaxy (MBE)

An alternative method to grow large-scale 2D materials is molecular beam epitaxy (MBE), a physical deposition technique based on co-evaporation of individual elements under ultra-high vacuum on a crystal surface held at a controlled temperature (Fig. 2). Directional molecular beams are produced by thermal evaporation in effusion cells or by electron-beam evaporation. MBE yields materials with high crystalline quality and centimeter-scale areas. The ultra-high vacuum environment ensures minimal contamination and offers the possibility to use surface analysis techniques to characterize *in situ* the properties of the films. The film composition, doping, and thickness can be accurately controlled, thanks to the low fluxes (typically 1–10 Å/min). Heterostructures consisting of a stack of vdW materials can be formed with abrupt interfaces and an arbitrary number of layers. The growth of layered materials by MBE is based on the method of vdW epitaxy, consisting in film growth either on a passivated surface with a very low density of dangling bonds [e.g., F-terminated BaF₂(111)] or on a layered vdW substrate (e.g., SiC/graphene or mica).¹¹⁶ The weak interaction between the epilayer and the substrate largely releases the constraint of lattice matching and leads to the formation of fully relaxed layers. The advantage of vdW epitaxy is the potential to arbitrarily combine vdW materials with atomically sharp interfaces, free from contaminants.

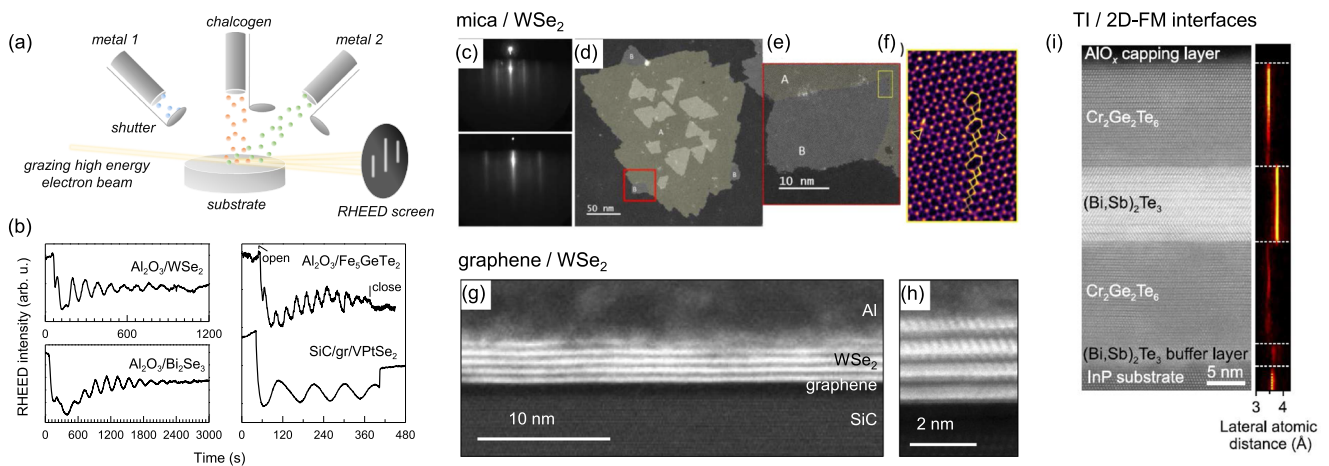


FIG. 2. (a) Schematics of the MBE deposition technique. (b) Reflection high-energy electron diffraction (RHEED) intensity oscillations recorded during the epitaxy of various vdW materials, showing layer-by-layer growth. (c)–(f) Characterization of a WSe₂ monolayer grown by MBE on mica. (c) RHEED patterns in two azimuths showing the single crystallinity of the film. (d)–(f) Plane-view transmission electron microscopy near the boundary between in-plane rotated twin domains (marked A and B). (g) and (h) Cross-sectional high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) image of a multilayer WSe₂ film grown on SiC/graphene. (i) Cross-sectional HAADF-STEM image of a Cr₂Ge₂Te₆/(Bi,Sb)₂Te₃/Cr₂Ge₂Te₆ epitaxial heterostructure [reproduced with permission from Mogi *et al.*, Phys. Rev. Lett. **123**, 016804 (2019). Copyright 2019, American Physical Society].¹¹⁵

The epitaxial growth of layered TIs by MBE was reported on various substrates [e.g., graphene, Si(111), Al₂O₃(0001), GaAs(111), Ge(111), BaF₂(111), InP(111), and SrTiO₃(111)].^{117–123} Crystals of high quality are commonly produced, but suppressing defects such as chalcogen vacancies or twin boundaries requires fine optimization of the growth parameters.^{118,120,123} It is possible by MBE to adjust in (Bi_{2–x}Sb_x)(Te_{3–y}Se_y) alloys the Dirac point energy through the Te/Se ratio¹²⁴ and the chemical potential through the Bi/Sb relative content,^{119,120} yielding bulk-insulating materials with designed topological Fermion dynamics. Magnetic doping of (Bi_{2–x}Sb_x)Te₃ with Cr and V^{125–128} as well as epitaxy of the antiferromagnets MnBi₂Se₄^{129,130} and MnBi₂Te₄^{131,132} were reported, enabling the experimental realization of the quantum anomalous Hall effect.¹³³ For the large-area growth of TIs, other physical deposition techniques were employed recently, such as sputtering and pulsed laser deposition.^{73,134}

Efforts in the MBE of TMDs are more recent. Epitaxy was reported for a variety of TMDs and TMD heterostructures, including MoSe₂, WSe₂, HfSe₂, PtSe₂, VSe₂, ZrTe₂, MoTe₂, and WTe₂.^{66,135–149} Inert vdW surfaces, such as graphene, mica, or hBN, make it possible to stabilize very flat monolayers of high structural quality, as shown for mica/WSe₂.¹⁴⁹ However, the weak interfacial interaction often results in an in-plane misorientation of crystal grains, which probably remains the main difficulty in the MBE of TMD monolayers. The misorientation is due to atomic position coincidences between the TMD monolayer and the substrate, favoring the formation of grains with an orientation away from the exact epitaxial relationship. These

configurations being metastable, high-temperature growth and/or post-deposition annealing might improve the in-plane crystal quality. Another strategy consists in orienting the grains by nucleation at step edges using vicinal surfaces. In addition, quasi-vdW epitaxy on non-vdW surfaces may promote a better crystal alignment at the expense of a stronger substrate interaction¹⁴⁶ and a possible loss of the intrinsic TMD properties. Recently, substrate-induced strain was used to stabilize the metastable semimetallic 1T' phase in WSe₂¹⁵⁰ and WTe₂.¹⁵¹

Reports on the MBE of 2D-FMs include diluted magnetic TMDs, such as V-doped WSe₂^{152,153} and Mn-doped MoSe₂,^{154,155} intercalated vdW compounds, such as V₅Se₈¹⁵⁶ and Cr₂Te₃,¹⁵⁷ and intrinsic FMs, such as Cr₂Ge₂Te₆,¹⁵⁸ Fe₃GeTe₂,^{159–161} CrBr₃,¹⁶² CrCl₃,¹⁶³ and CrI₃.¹⁶⁴ Intrinsic ferromagnetism was also claimed for MBE-grown monolayer MnSe_x¹⁶⁵ and VSe₂,⁶⁶ although in the latter case, a charge density wave tends to suppress ferromagnetism.^{166,167} TI/2D-FM multilayers with abrupt epitaxial interfaces were achieved by MBE only recently.^{158,161}

III. FABRICATION OF DEVICES

SCI can be probed and exploited in different types of spintronic devices. The two most prominent examples are the non-local Hall cross spin devices and the SOT devices. The non-local Hall cross devices [sketched in Fig. 3(a)] combine different materials with specific purposes. In the case of vdW heterostructures, they consist of a spin channel made of a low-SOC material (typically

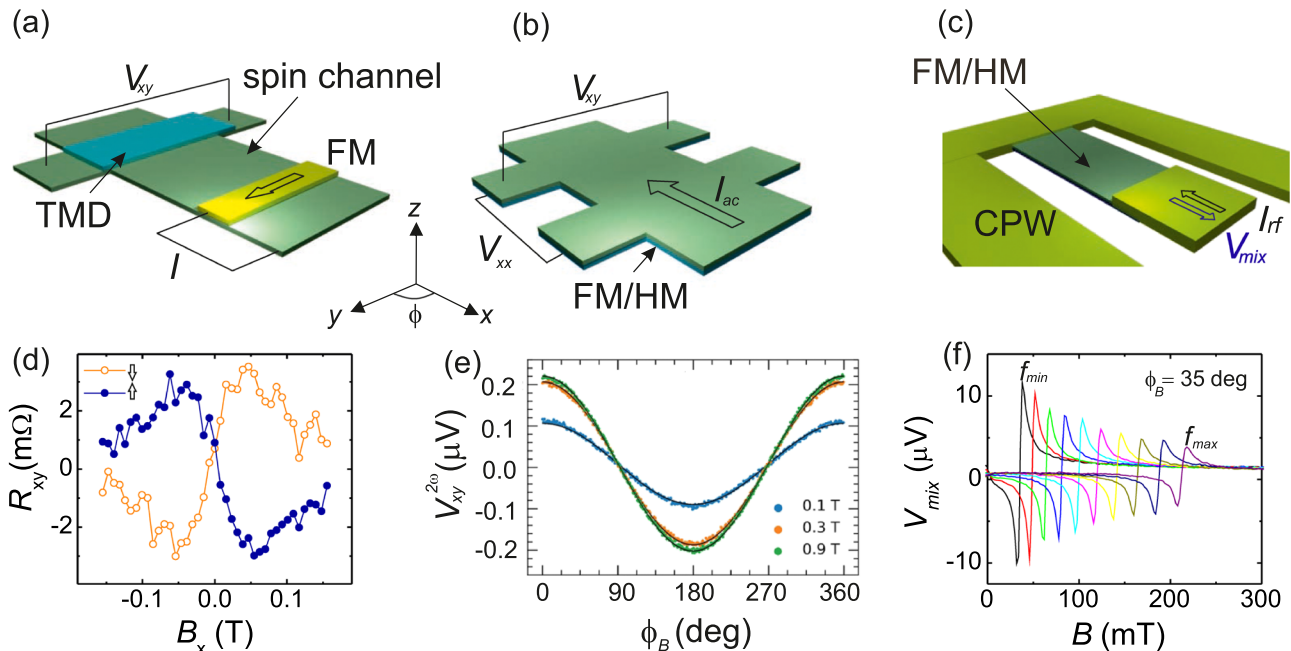


FIG. 3. Schematic illustration of the (a) non-local lateral device, (b) second-harmonic Hall device, and (c) SOT-FMR device. (d) Representative room-temperature non-local resistance ($R_{xy} = V_{xy}/I$) vs magnetic field (B_x) measurements in the iSHE configuration in a graphene-WS₂ device. Open and closed symbols correspond to the magnetization of the FM aligned in the $-y$ and $+y$ directions, respectively. (e) Representative second-harmonic Hall voltage for a TMD/NiFe bilayer as a function of the angle between the in-plane magnetic field and the direction of the current for different magnitudes of the magnetic field. (f) Typical SOT-FMR resonances in a TMD/NiFe bilayer for different frequencies with the in-plane magnetic field applied at 35° relative to the direction of the current. The characteristic resonance frequencies f_{\min} and f_{\max} are in the 1–20 GHz range. (d) Adapted with permission from Benitez *et al.*, Nat. Mater. **19**, 170–175 (2020). Copyright 2020, Springer Nature Limited. (e) Adapted with permission from Gupta *et al.*, Nano Lett. **20**, 7482–7488 (2020). Copyright 2020, American Chemical Society.

graphene) where the spins can propagate over long distances (tens of μm). Spins are injected and detected either through a FM or by taking advantage of SCI in materials with large SOC. Due to the atomically thin nature of graphene, its very weak SOC can be enhanced by proximity effects when stacking the material with other vdW materials, including TMDs, semimetals, and TIs, which are characterized by large SOC.

As the *in situ* growth of vdW heterostructures is still at an early stage, most of the studied systems to date are obtained by stacking mechanically exfoliated crystals. Owing to the nature of the studied effects and stability issues of some of the utilized materials at ambient conditions, the heterostructures are fabricated under controlled atmosphere with a low concentration of H_2O and O_2 (around 0.1 ppm), thus providing at the same time ultra clean interfaces. The stacks are obtained by transferring the 2D materials using a dry transfer method, in which viscoelastic stamps are used to pick-up and release flakes in a controlled manner. Precise alignment between the flakes is achieved by using a micro-positioner stage and an optical microscope. To avoid gas bubbles and blisters and to ensure better attachment between the layers, the finished stacks are usually annealed in an Ar/H_2 atmosphere or high vacuum.

The stacking process is followed by electron-beam lithography and reactive etching steps to define electrical contacts and to pattern the heterostructure with specific shapes and dimensions. Devices for SCI experiments consist of graphene etched in a Hall bar geometry. The graphene channel, typically $1\ \mu\text{m}$ wide and tens of μm long, includes a graphene Hall arm capped with a large-SOC material (see Sec. IV for further details). The device includes a combination of metal and FM contacts. Metal contacts, typically Ti/Au or Ti/Pd, are attached to the graphene Hall cross and can be used either for applying an electric current or for measuring the voltage along the Hall bar. FMs (typically Co) are used to both inject and detect the spin current. Efficient spin injection and detection require the insertion of thin insulating barriers between the FM contacts and graphene to alleviate the conductance mismatch.^{168,169}

SOT devices are made of bilayer heterostructures of FMs and large-SOC materials, the device geometry depending on the measurement technique [Figs. 3(b) and 3(c)]. In the case of SOT ferromagnetic resonance (FMR) experiments [Fig. 3(c)], large-SOC/FM bilayers are embedded into high-frequency coplanar waveguides, whereas for harmonic Hall voltage measurements, the bilayer is patterned in a Hall bar geometry [Fig. 3(b)]. Most of the vdW-based SOT devices studied to date feature heterostructures combining vdW materials with well-known bulk systems. Various studies considered mechanically exfoliated TMD flakes (under vacuum or flowing N_2 , or in a glovebox with inert Ar atmosphere) with a thin film of a FM (typically NiFe due to its low magnetic damping) deposited on top. Further studies spanned from exfoliated crystals to materials grown by CVD (TMDs such as MoS_2 or WSe_2 ^{74,170}) or MBE [TIs such as Bi_2Se_3 ^{69,70} and $(\text{Bi}_{1-x}\text{Sb}_x)_2\text{Te}_3$ ^{171,172}]. The FM is deposited using grazing angle sputtering in various reports, to avoid damaging the underlying TMD, and capped to prevent oxidation. Similarly, vdW FMs, such as Fe_3GeTe_2 and $\text{Cr}_2\text{Ge}_2\text{Te}_6$, obtained from exfoliation methods can be combined with heavy-metal layers of Pt and Ta. Due to the high sensitivity of these 2D FMs to ambient conditions, they are typically rapidly transferred to a deposition chamber and the heavy metal layer is sputtered on top^{173,174} (in

some cases, preceded by a short etching with Ar plasma to clean the interface^{175,176}).

SOT devices (with sizes of up to $20 \times 100\ \mu\text{m}^2$) are commonly patterned either by electron beam or by optical lithography techniques, followed by ion milling and the deposition of metal contacts. However, when investigating magnetization switching mechanisms, it is important to take into account the dimensions of the device; for large devices, switching occurs by domain nucleation and motion, which results in low switching currents. To date, fewer studies were realized for all-vdW SOT devices. These include *in situ* MBE-grown $(\text{Bi}_{0.5}\text{Sb}_{0.5})_2\text{Te}_3/(\text{Cr}_{0.08}\text{Bi}_{0.54}\text{Sb}_{0.38})_2\text{Te}_3$ bilayers with a capping, which were patterned by photolithography and dry etching.¹⁷⁷ More recently, switching of Fe_3GeTe_2 on WTe_2 was reported.¹⁷⁸ The samples were prepared similarly to the above-mentioned lateral structures by dry transfer of mechanically exfoliated flakes inside an inert Ar atmosphere glove box, further protected by electron-beam evaporated Al capping. Progress in the *in situ* growth of vdW heterostructures may greatly simplify the fabrication steps required to preserve the interfaces from degradation due to air exposure.¹⁶¹

IV. SCI: EXPERIMENTS

A. Non-local Hall cross devices

Pioneering SCI experiments were conducted in metallic systems using non-local detection techniques. These devices use a FM contact for injecting spins into a non-magnet, typically Cu and Al, acting as the spin transport channel. Spins diffuse and are absorbed by a heavy-metal, e.g., Pt and Ta, in which a transversal voltage, generated by the iSHE, is measured in a Hall cross geometry.³³ Later, the same geometry is used by replacing the spin transport channel by graphene (see hybrid systems in Table I), providing a highly efficient spin injection that results in enhanced measured SCI efficiencies.

Over the last few years, SCI in lateral spin devices regained attention. The rich variety of vdW materials and possible combinations opened up the possibility to realize SCI in all-vdW systems. When combining graphene, which possesses excellent spin transport properties, with a non-conducting vdW material with large SOC, typically a semiconducting TMD, SCI occurs in graphene due to proximity effects. In the graphene/TMD region, the SOC of graphene is enhanced and the electronic band structure is modified significantly, with the most prominent features being the opening of an energy gap in the meV range at the K and K' points, lifting of spin degeneracy, and imprinting of an out-of-plane spin texture (with the spin orientation depending on the valley and a winding in-plane component far from the charge neutrality point).¹⁷⁹

The device geometry [as sketched in Fig. 3(a)] is slightly different and consists of a graphene Hall cross with one of its arms shrouded with a TMD. The pristine graphene region (not covered by the TMD) is used to transport spin information from the spin injector to the Hall arm and vice versa. A spin current is injected into graphene by means of a planar FM contact with magnetization along its long axis.¹⁰ When diffusing toward the Hall arm region, the spins experience precession under an external magnetic field. The relative direction between the magnetic field and the injected spins is key to distinguish between the different spin-to-charge conversion mechanisms (SGE or iSHE). In the case of the SGE, an out-of-plane

TABLE I. SCI efficiencies in non-local devices based on metallic, hybrid, and all-vdW systems. The parameters are defined as follows: γ^{SH} , α^{SGE} , λ_s^{S} , $\lambda_s^{\text{||}}$ are the spin relaxation lengths of spins precessing in-plane and out-of-plane, respectively; and α^{SGE} , $\tau_{\text{||}}$ being the spin lifetime of the in-plane spins. TMD, v_F being the Fermi velocity, and $\tau_{\text{||}}$ being the spin lifetime of the in-plane spins.

	Comments	SCI mechanism	SCI (%)	SCI efficiency (nm)	References
Metallic					
Cu/Ta	ρ (Ta) = 333 $\mu\Omega$ cm (10 K) λ_s (Ta) = 2.7 nm (10 K)	iSHE	γ^{SH} (Ta) = -0.37 ± 0.20 (10 K)	$\gamma^{\text{SH}} \cdot \lambda_s$ (Ta) = 0.01 (10 K)	180
Cu/Pt	ρ (Pt) = 17.9 $\mu\Omega$ cm (RT) λ_s (Pt) = 7 nm (RT)	iSHE	γ^{SH} (Pt) = 0.9 (RT)	$\gamma^{\text{SH}} \cdot \lambda_s$ (Pt) = 0.06 (RT)	181
Cu/Pt	ρ (Pt) = 44.1 $\mu\Omega$ cm (10 K) λ_s (Pt) = 0.75 nm (10 K)	iSHE	γ^{SH} (Pt) = 8.5 ± 1.3 (10 K)	$\gamma^{\text{SH}} \cdot \lambda_s$ (Pt) = 0.06 (10 K)	182
Hybrid					
Bi ₂ O ₃ /graphene	Bilayer graphene	iSHE (in proximitized graphene)	γ^{SH} = 0.10 ± 0.05 (RT) γ^{SH} = 0.6 ± 0.1 (10K)	λ_s not measured in the SCI region	183
Pt/graphene	Single-layer graphene ρ (Pt) = 46 $\mu\Omega$ cm (RT) λ_s (Pt) = 5 nm (RT; taken from the literature)	iSHE (in Pt)	γ^{SH} (Pt) = 15 ± 1 (RT)	$\gamma^{\text{SH}} \cdot \lambda_s$ (Pt) = 0.75 (RT)	184
Pt/graphene	Few-layer graphene ρ (Pt) = 134 $\mu\Omega$ cm (RT) λ_s (Pt) = (2.1 \pm 0.4) nm (RT) (taken from the literature)	iSHE (in Pt)	γ^{SH} (Pt) = 23.4 ± 2.5 (RT)	$\gamma^{\text{SH}} \cdot \lambda_s$ (Pt) = 0.50 ± 0.02 (RT)	185
All-vdW					
MoS ₂ /graphene	Few-layer graphene Spin absorption by MoS ₂	iSHE (in proximitized graphene and MoS ₂)	γ^{SH} = -0.33 ± 0.04 (RT) γ^{SH} = -4.5 ± 0.9 (10 K)	λ_s not measured in the SCI region	186
WS ₂ /graphene	Single-layer graphene (Non-conducting) multilayer WS ₂ λ_s^{\perp} (gr/WS ₂) = 1.25 μm $\lambda_s^{\text{ }}$ (gr/WS ₂) = 420 nm	SHE-iSHE (in proximitized graphene) SGE-iSGE (in proximitized graphene)	γ^{SH} = 0.3 (RT) α^{ISGE} = 0.1 (RT)	$\gamma^{\text{SH}} \cdot \lambda_s^{\perp}$ (gr/WS ₂) = 3.75 (RT) $\alpha^{\text{ISGE}} \cdot \lambda_s^{\text{ }}$ (gr/WS ₂) = 0.42 (RT)	187

TABLE I. (Continued.)

	Comments	SCI mechanism	SCI (%)	SCI efficiency (nm)	References
WS ₂ /graphene	Single-layer graphene (Non-conducting) single-layer WS ₂ λ_s^\perp (gr/WS ₂) = 1.58 μm λ_s^\parallel (gr/WS ₂) = 313 nm	SHE-iSHE (in proximitized graphene) SGE-iSGE (in proximitized graphene)	$\gamma^{\text{SH}} = 0.13$ (4 K), vanishes at $T > 20\text{K}$ $\alpha^{\text{*SGE}} = 2.8$ (4 K) $\alpha^{\text{*SGE}} = 0.56$ (RT)	Different definition of $\alpha^{\text{*SGE}}$ (see the table caption)	188
WSe ₂ /graphene	Few-layer graphene Spin absorption by WSe ₂	iSHE (in proximitized graphene)	$\gamma^{\text{SH}} = 1.7 \pm 0.2$ (RT) $\gamma^{\text{SH}} = 2.8 \pm 0.3$ (10 K)	λ_s not measured in the SCI region	189
2H-TaS ₂ /graphene	Few-layer graphene Multilayer TaS ₂ (metallic)	SGE-iSGE (in proximitized graphene and TaS ₂)	$\alpha^{\text{SGE}} = -1.4$ – 4.3 (RT) (Gate-tunable)	λ_s not measured in the SCI region	190
(BiSb) ₂ Te ₃ /graphene	Single-layer graphene Multilayer BST	SGE (in proximitized graphene)	$\alpha^{\text{SGE}} = 0.17$; 2.5; 1; 4.8 (RT)	λ_s not measured in the SCI region	191
MoTe ₂ /graphene	Few-layer graphene Multilayer MoTe ₂	Unconventional SCI (in MoTe ₂)	$\alpha^{\text{SCI}} \geq 0.21$ (RT) (conventional) $\alpha^{\text{SCI}} \geq 0.10$ (RT) (unconventional)	λ_s not measured in the SCI region	192
WTe ₂ /graphene	Single-layer graphene Multilayer WTe ₂ λ_s (WTe ₂) = 8 nm	Unconventional SCI (in WTe ₂)	$\alpha^{\text{SCI}} = 9$ (RT)	$\alpha^{\text{SCI}} \cdot \lambda_s$ (WTe ₂) = 0.72 (RT)	193

magnetic field causes an in-plane spin precession leading to a voltage along the Hall arm driven exclusively by the SGE. In the case of the iSHE, a magnetic field is applied to graphene along the spin diffusion channel. The spins diffusing toward the Hall cross precess out-of-plane, generating a voltage driven exclusively by the iSHE [Fig. 3(d)].

For the reciprocal effect, i.e., charge-to-spin conversion, a charge current flowing along the Hall arm generates a transversal spin current and a non-equilibrium in-plane spin density due to the SHE and the iSGE, respectively. In the case of the SHE, the spins point out-of-plane, whereas in the case of the iSGE, a non-equilibrium spin density with the spins pointing in-plane and perpendicular to the charge current is generated at the graphene/TMD interface. These two spin components (perpendicular to each other) can be probed by a FM acting as a spin detector, with the magnetization perpendicular to both spin components, and are disentangled by varying the direction of the external magnetic field.

SCI experiments in lateral spin devices were initially reported for multilayer graphene combined with MoS₂.¹⁸⁶ Shortly after, spin-charge interconversions driven by both the SHE and SGE were probed in the graphene-WS₂ heterostructure at room temperature.^{187,188} Remarkably, both SCI mechanisms exhibit a gate-tunable magnitude that can be controlled by electrostatic gating. This knob is, in fact, essential to determine whether the SCI occurs exclusively in graphene. The electrostatic gating not only varies the carrier density in the graphene layer but also shifts the Fermi energy in the TMD. The latter may alter the insulating character, resulting in spin absorption and related SCI within the TMD if the Fermi level shifts toward the conduction (or valence) band. More recently, SCI driven only by the SHE was reported for the graphene-WSe₂ heterostructure.¹⁸⁹

Beyond semiconducting TMDs, SCI experiments combining graphene with other large-SOC materials were also reported for Bi₂O₃,¹⁸³ metallic layered 2H-TaSe₂,¹⁹⁰ the layered p-type TI (Bi_{0.15}Sb_{0.85})₂Te₃,¹⁹¹ and the semimetals 1T'-MoTe₂¹⁹² and WTe₂.¹⁹³ Overall, the SCI efficiency in all-vdW heterostructures, defined as $\gamma^{\text{SH}}\lambda_s$ with λ_s being the spin relaxation length in the region where the SCI takes place, compares favorably with fully metallic or hybrid systems based on heavy metals, such as Pt and Ta. Table I summarizes experimental measurements of the SCI efficiency for metallic systems, hybrid systems, and all-vdW heterostructures, specifying which SCI mechanism takes place and where. We note that some studies consider an effective λ_s^{eff} of the whole channel (not only the SCI region, i.e., include a large contribution of regular graphene). These values are not included in Table I, as they could overestimate the SCI efficiency.

B. SOT devices

Aside from the non-local Hall cross, two other methods are commonly employed to study the SCI in TMD/FM and TI/FM bilayers [Figs. 3(b) and 3(c)]. The second-harmonic Hall (2ω -Hall)¹⁹⁴ and SOT ferromagnetic resonance (SOT-FMR)³⁷ techniques both measure the current-driven SOT in the FM, from which the charge-to-spin conversion efficiency is inferred. Additionally, the spin pumping effect, based on the reciprocal effect of spin transfer, can be used to measure the SCI efficiency.¹⁹⁵ In 2ω -Hall and SOT-FMR

experiments, the current-driven SOT induces precession of the magnetization, which results in time-dependent changes in the longitudinal and transverse resistances. Frequency mixing between the resistance and current oscillations leads to measurable direct current and second-harmonic voltages [Figs. 3(e) and 3(f)]. The magnitude, direction, and nature (conservative/dissipative) of the SOT are deduced from the field and angle dependences of these voltages. In spin pumping experiments, magnetization precession is driven by an external microwave magnetic field. Spins aligned with the average magnetization are pumped into the adjacent large-SOC material, where spin-to-charge conversion gives rise to a measurable direct charge current.

Because of the spin-polarized surface states of TIs, the TI/3D-FM bilayers are among the most widely used SOT systems that include vdW materials. Seminal works showed very large SCI efficiencies for Bi₂Se₃/NiFe⁶⁹ and Bi₂Se₃/CoFeB.⁷⁰ A strong Fermi level dependence of the SOT was found in Bi_xSb_{2-x}Te₃/NiFe,¹⁷¹ and electrostatic gating of the SOT was demonstrated in magnetic Cr-doped Bi_xSb_{2-x}Te₃.¹⁹⁶ A large in-plane damping-like SOT is generally observed, enabling efficient current-driven magnetization switching.^{196,197} Noteworthy, there is less evidence of large field-like SOT, expected to be a clear signature of the iSGE. The reported damping-like SOT efficiency is typically 0.1–1, which is significantly larger than in heavy metals.⁴¹ The corresponding Edelstein inverse length q is in the range 0.1–1 nm⁻¹.^{171,172} On the other hand, the Edelstein length λ measured with spin pumping is typically 0.01–0.1 nm,¹⁹⁸ which is rather low in comparison to other 2D systems (0.2–0.4 nm for the Bi/Ag Rashba interface¹⁹⁹ and 2.1 nm for the surface of the α -Sn TI²⁰⁰).

SOT measurements were reported for various TMD/3D-FM bilayers.²⁰¹ Semiconducting (MoS₂, WS₂, and WSe₂), semimetallic (WTe₂, β -MoTe₂, TaTe₂, and PtTe₂), and metallic (NbSe₂ and TaS₂) TMDs were interfaced with NiFe, Co, or CoFeB. Salient results are the gate-modulation of the SOT with WS₂²⁰² and the generation of unconventional SOT with low-symmetry TMDs.^{203,204} ST-FMR measurements of 1T'-WTe₂/NiFe devices revealed an unusual out-of-plane damping-like SOT whose magnitude depends on the direction of the current with respect to the crystallographic axes, reflecting the reduced symmetry of the bilayer.^{203,204} 2ω -Hall measurements in similar bilayers showed an anisotropic field-like SOT, which was attributed to the contribution of topological Fermi arcs.²⁰⁵ The reported SOTs are generally weaker with TMDs than with TIs. Nonetheless, efficient and field-free current-driven magnetization switching was recently achieved in WTe₂/NiFe.²⁰⁶ Further work is required to clarify the SCI mechanisms at play with TMDs, as contrasting results were reported about the damping-like/field-like SOT ratios and the bulk or interfacial origin of the SOT. Spin pumping studies of TMD/FM systems are still scarce. Spin pumping measurements of MoS₂/YIG yielded an Edelstein length λ of 0.4 nm.²⁰⁷

More recently, the SOT was studied in large-SOC/2D-FM heterostructures, with significant efficiencies and magnetization switching reported in Fe₃GeTe₂/Pt.^{173,175} Single 2D-FM layers are also interesting candidates for the generation of unconventional SOT. For example, the low symmetry of monolayer Fe₃GeTe₂ allows for a SOT akin to a current-driven magnetic anisotropy.^{208,209} Experimental hints of such a SOT were also reported for multilayer Fe₃GeTe₂.²¹⁰

In contrast to lateral spin devices in which the spin injection, transport, and detection are spatially well separated, these processes occur in vertical bilayers within a couple of nanometers near the interface. As recently shown for TI/FM bilayers,¹⁷² the SOT is thus extremely sensitive to the structural, chemical, and electronic properties of the interface, where band bending, orbital hybridization, spin memory loss, material intermixing, and magnetic dead layers were reported.^{211–215} These phenomena are generally detrimental to the SOT, complicate the analysis of the SCI, and are probably responsible for the large experimental spread in the SCI efficiencies. It is expected that all-vdW heterostructures combining 2D-FMs with TIs or TMDs will display enhanced SOT and will constitute model systems, thanks to their sharp and weakly interacting interfaces. Indeed, a recent study found a SOT efficiency of 4.6 in $\text{WTe}_2/\text{Fe}_3\text{GeTe}_2$.¹⁷⁸

Apart from the novel spin phenomena due to proximity effects between different vdW materials, there is promise to solve a challenge that exists in today's magneto-resistive random access memory technology. Here, perpendicular magnetization switching is more attractive because it is faster and more scalable. Efficiently switching a perpendicular magnetization using SOT requires the spins to have a large out-of-plane component (out-of-plane antidamping SOT), which is very difficult to realize by common materials. Recent evidence shows that vdW materials with low-crystalline symmetry, in principle, can be used to control the direction of the current-induced SOT.^{204,216} A summary of experimental parameters describing the SCI in vdW-based SOT devices is given in [Table II](#).

V. SCI: THEORETICAL CONTRIBUTIONS

Successful integration of vdW heterostructures in the technological workflow relies on achieving efficient electrical control. For low-power applications, where a small current flow is essential, linear response theory provides a general framework to determine any observable electrical response. The central figure of merit of the SCI is the spin Hall angle γ_{SH} , which determines the amount of pure spin current generated for an incoming charge current.³³ For quantifying the SOT efficiency, it is usual to evaluate the torque efficiency χ_{τ} , which measures the amount of torque felt by a magnet per unit of current density and can be rewritten as an effective spin Hall angle.⁴¹ The crystal symmetries prescribe the spin orientation via the SOC. In combination with group theory, the theory of invariants enables a relatively straightforward method to determine the allowed directions.^{217,218} First, one identifies the minimum set of point symmetries capable of expanding the whole point group (generating elements) and then imposes invariance of the linear response tensor with respect to those symmetries. By doing so, one finds a set of constraints limiting the number of finite elements and fixes the allowed directions for a given current.

The combination of disorder, exchange interaction, and SOC defines the efficiencies. Therefore, it is essential to properly describe the band structure, spin texture, and exchange splitting of the considered vdW system and evaluate the role of disorder in the diffusive regime, which is the most common experimental situation.^{219–221} Recent reports suggest that the use of symmetry-based models derived from *ab initio* methods provides the most cost-effective way to describe simple systems, such as graphene²²⁰ and WTe_2 .²²²

However, although computationally inefficient, Wannier Hamiltonians are the preferred choice for more complex vdW heterostructures.^{223,224} On the other hand, linear scaling quantum transport methodologies are the only computational approaches capable of addressing the diffusive regime, since the systems involved demand for models containing many millions of atoms.²²⁵ Most of these methods are numerical implementations of the Kubo formula, either in the time or energy domain, allowing us to determine a system's properties for different transport regimes.

For honeycomb-based vdW heterostructures, spin-valley coupling and broken inversion symmetry make the two valleys inequivalent in terms of the spin and Berry curvature, providing a unique platform for efficient SCI.^{186,187,222,226,227} In [Fig. 4](#), we present a sketch of how lowering the symmetries enables modifications in the band structure that enable different spin-related effects. In a highly symmetric system, such as graphene, combination of the inversion and time-reversal symmetries does not allow for spin-momentum locking and, therefore, prevents the Rashba-Edelstein effect and its associated SOTs. In TMDs, the presence of chalcogen and metal atoms lifts the inversion symmetry and enables band-splitting due to the SOC. However, since there is a horizontal mirror plane, the system cannot host a vertical electrical field, which prevents the typical in-plane spin-momentum locking. Yet, the broken inversion symmetry enables a novel torque not related to the Rashba effect that acts as an electrically controlled magnetic anisotropy, which is a promising gateway to magnet-free switching of perpendicular magnetizations.²²⁸ In honeycomb-based heterostructures, the presence of dissimilar layers finally removes the horizontal mirror plane and reduces the symmetry to a C_{3v} point group, which supports the conventional field-like and damping-like torques, as well as an electrically controlled magnetic anisotropy,²²⁸ as demonstrated experimentally in $L1_1$ CoPt/CuPt bilayers.²²⁹ However, this switching depends on the threefold rotation, and systems such as graphene and TMDs, where the Fermi level lies on an isotropic surface, are not expected to display the effect. Recently, tremendous efforts focused on synthesizing a new class of materials, the magnetic Janus TMDs, built by replacing the chalcogen atoms on one side of a TMD with different ones. Janus materials show simultaneously spin-valley coupling and a large Rashba effect due to their intrinsic electrical dipole¹⁷⁸ and magnetism due to the presence of magnetic metal atoms.²²⁴ Although Janus materials are still in their infancy, they combine multiple desirable properties for technological applications and deserve scrutiny. For instance, they are ideal for achieving all-in-one SOT, where the magnetization is electrically switchable in a single material via a current.²²⁴

The valley-driven spin Hall effect is a phenomenon where the electron motion is monitored by a spin-dependent Lorentz force opposite in each valley.^{186,187,227} The broken inversion symmetry of heterostructures can also generate a Rashba interaction, which, when combined with spin-valley coupling, leads to valley-dependent SOT²²⁸ and yields optimal SCI via the Rashba-Edelstein effect.²²⁷ The interplay between spin-valley coupling, Rashba interaction, and magnetism is not extensively studied to date. In magnetic systems with a Zeeman splitting, the valley-Zeeman effect²²¹ leads to valley polarization, as predicted for TMD/2D-magnet heterostructures^{230,231} and recently realized in $\text{WSe}_2/\text{CrI}_3$.²³² Nevertheless, understanding the combined impact of the valley-Zeeman

TABLE II. SCI in vdW-based SOT devices. The parameters are defined in Ref. 41, where additional non-vdW structures are reviewed. For TMDs, a complementary comparative study can be found in Ref. 201.

Fabrication	Method	B_{DL}/j mT/(10 ¹¹ A/m ²)	ξ_{DL}^E Dimensionless	ξ_{FL}^E Dimensionless	ξ_{DL}^E $10^5 \hbar/2e (\Omega\text{m})^{-1}$	ξ_{FL}^E $10^5 \hbar/2e (\Omega\text{m})^{-1}$	q_{REE} nm ⁻¹	λ_{REE} nm	Other torques	References
Non-vdW										
Pt(3)/Co(0.6)/AlO _x (1.6)	Sputtering	-6.9	4	0.13	-0.073	3.5	-2			41
Ta(1.5-5)/CoFeB(0.8-1.1)/MgO(2)	Sputtering	1.3-4.4	-(2-19)	-(0.03-0.11)	0.04-0.47	-(0.14-0.68)	0.22-0.67			
Bi(8)/Ag(5-20)/Py(15)	Evaporation							0.2-0.4		199
α -Sn(30 ML)/Ag(2)/Fe(5)	MBE							2.1		200
TI										
Bi ₂ Se ₃ (8)/Py(8,16)/AlO _x (2)	MBE		2.0-3.5 ^a		2.5-2.8 ^b	1.1-2.0	1.4-1.6			69
Bi ₂ Se ₃ (20)/CoFeB(5)/MgO(1)/SiO ₂ (3)	MBE		0.05-0.11		0-0.1					70
			~0.4		~0.2					
Bi ₂ Se ₃ (10)/Ag(5)/CoFeB(7)/MgO(2)/SiO ₂ (4)	MBE	5.3	3.2	0.5 ^c	0.3 ^b					233
Bi ₂ Se ₃ (7.4)/CoTb(4.6)/SiN _x (3)	MBE	6.1	0.16							71
Bi ₂ Se ₃ (8)/Py(6)/MgO(1)/SiO ₂ (4)	MBE		1.71							72
Bi ₂ Se ₃ (5)/CoFeB(7)/MgO(2)/Al ₂ O ₃ (3)	MBE		1.75				0.82			72
Bi ₂ Se ₃ (20)/CoFeB(7)/MgO(2)/Al ₂ O ₃ (3)	MBE		0.3 ^c				~0.1 ^c			
Bi ₂ Se ₃ (5-35)/Py(20)/SiO ₂ (30)	MBE		0.0093							234
Bi ₂ Se ₃ (5,10)/CoFeB(5)/MgO(2)	+ Ar milling		0.021-0.43							235
Bi ₂ Se ₃ (10)/CoFeB(5)/MgO(2)	MBE		0.3-1.7							235
Ge/Bi ₂ Se ₃ (10)	MBE							-0.026 ^c		236
Bi _x Se _{1-x} (4)/CoFeB(5)/MgO(2)/Ta(5)	Sputtering	99	18.6		1.45					73
	ST-FMR		8.7							
YIG/Bi ₂ Se ₃ (6-60)	MBE							0.035		237

TABLE II. (Continued.)

	Fabrication	Method	B_{DL}/j mT/(10^{11} A/m 2)	B_{FL}/j	ξ_{DL}^j Dimensionless	ξ_{FL}^j	ξ_{DL}^E 10^5 h/2e (Ω m) $^{-1}$	ξ_{FL}^E	q_{REE} nm $^{-1}$	λ_{JREE} nm	Other torques	References
YIG/(Bi,Sb) $_2$ Te $_3$ (6)	MBE	Spin pumping 2 ω -Hall	87	~ 0	3.3					0.017		
MgO(2)/Bi $_2$ Te $_3$ (8)/ CoTb(6)/TaO $_x$ (1.5)	Sputtering	2 ω -Hall										238
(Bi $_{1-x}$ Sb $_x$) $_2$ Te $_3$ (8)/ Cu(8)/Py(10) $x = 0.5, 0.7, 0.9$ $0.8 < x < 0.9$ (E_F near Dirac point)	MBE	ST-FMR			$\sim 0.5^c$ (1-5) a				0.45-0.57 0.1-0.2			171
(Bi $_{0.4}$ Sb $_{0.6}$) $_2$ Te $_3$ (9)/ Py(5)/AlO $_x$ (2)	MBE	ST-FMR	0.02-0.1 mT/(A/m) d	$0 < -B_{FL} < B_{Oe}^c$	(0.88) a				0.19-0.88			172
(Bi $_{0.4}$ Sb $_{0.6}$) $_2$ Te $_3$ (9)/Ag(7)/ Py(5)/AlO $_x$ (2)	MBE	ST-FMR	0.01-0.07 mT/(A/m) d	$B_{FL} \approx -B_{Oe}^c$					0.12-0.53			172
(Bi $_{0.4}$ Sb $_{0.6}$) $_2$ Te $_3$ (9)/Al(6)/ Py(5)/AlO $_x$ (2)	MBE	ST-FMR	0.01-0.06 mT/(A/m) d	$0 < B_{Oe} < -B_{FL}^c$					0.11-0.48			172
Cr $_{0.16}$ (Bi $_{0.54}$ Sb $_{0.38}$) $_2$ Te $_3$ (6)/ (Bi $_{0.5}$ Sb $_{0.5}$) $_2$ Te $_3$ (3)	MBE	2 ω -Hall 1.9 K	48 000 -146 000		140-425 e							177
Cr $_{0.16}$ (Bi $_{0.5}$ Sb $_{0.42}$) $_2$ Te $_3$ (7)/ Al $_2$ O $_3$ (20)	MBE	2 ω -Hall 1.9 K	46 900 b		116 c							196
(Bi $_{0.22}$ Sb $_{0.78}$) $_2$ Te $_3$ (6)/ Py(12)	MBE	Spin pumping								0.075		198
(Bi $_{0.22}$ Sb $_{0.78}$) $_2$ Te $_3$ (6)/ NiO(5)/Py(12)	MBE	Spin- Seebeck effect								0.076		198
TMD												
MoS $_2$ (0.8)/CoFeB(3)/ TaO $_x$ (3)	CVD	2 ω -Hall	~ 0	0.078 b	~ 0	-0.14	~ 0	0.0288				74
WSe $_2$ (0.8)/CoFeB(3)/ TaO $_x$ (3)	CVD	2 ω -Hall	~ 0	0.114 b	~ 0	~ 0	~ 0	0.0552				
MoS $_2$ (0.8)/Py(5)	CVD	ST-FMR	Large									170
WTe $_2$ (1.8-15)/Py(6)/ AlO $_x$ (2)	Exfoliation	ST-FMR/ 2 ω -Hall			0.03 *	~ 0	0.08	0.09			Out-of- plane DL	204
WTe $_2$ (5.6-7)/Py(6)/Ru(4)	Exfoliation	2 ω -Hall RT		$B_{FL} \gg B_{Oe}$				$\sim 1^f$				205
WTe $_2$ (20-31)/Py(6)/Ru(4)	Exfoliation	<50 K 2 ω -Hall RT <50 K						$\sim 2.5^f$ ~ 0				205

TABLE II. (Continued.)

	B_{DL}/j	B_{FL}/j	ξ_{DL}^j	ξ_{FL}^j	ξ_{DL}^E	ξ_{FL}^E	q_{REE}	λ_{REE}	References
	mT/(10 ¹¹ A/m ²)	Dimensionless	10 ⁵ h/2e (Ωm) ⁻¹	nm ⁻¹	nm	Other torques			
	Fabrication	Method							
WTe ₂ (5.8–122)/Py(6)	Exfoliation	ST-FMR coercivity	0.09–0.79 0.15–0.65	~0	0.02–0.6 ^f	Out-of-plane DL		206	
YIG/MoS ₂ (2.4)	Exfoliation	Spin pumping	0.32			0.4		207	
TaTe ₂ (4.5–19.7)/Py(6)/AlO _x (2)	Exfoliation	ST-FMR/2ω-Hall	~0	~0	~0			239	
MoTe ₂ (0.7–14.2)/Py(6)/AlO _x (2)	Exfoliation	ST-FMR			0.058	0.15	Out-of-plane DL	240	
PtTe ₂ (3–20)/Py(2.5–10)	CVD	ST-FMR	0.05–0.15	-0.004	0.2–1.6			241	
NbSe ₂ (0.6–6)/Py(6)/AlO _x (2)	Exfoliation	ST-FMR			0.03	0.40	Out-of-plane DL	242	
TaS ₂ (0.88)/Py(7)/AlO _x (3)	Plasma-assisted sulfurization	ST-FMR	0.25	~0	14.9	~0		243	
WTe ₂ (10)/CoTb(6)/Ta(2)	Sputtering	Magnetic loop shift	0.20					244	
2D FM									
Fe ₃ GeTe ₂ (4)/Pt(6)	Exfoliation	2ω-Hall low T	53.4	24.3				173	
Cr ₂ Ge ₂ Te ₆ (19.6)/Pt(10)	Exfoliation	2ω-Hall 5 K	2.0	~0	0.25	~0		174	
Fe ₃ GeTe ₂ (23)/Pt(5)	Exfoliation	2ω-Hall 180 K		0.14				175	
Cr ₂ Ge ₂ Te ₆ (8)/Ta(5)	Exfoliation	Coercivity	~80					176	
WTe ₂ (12.6)/Fe ₃ GeTe ₂ (7.3)/AlO _x (2.6)	Exfoliation	Coercivity 160 K	4.6		2.25			178	
Fe ₃ GeTe ₂ (6–21)	Exfoliation	Coercivity 2 K					Anisotropy torque	210	

^a Assuming 3D carriers.

^b Inferred from other quantities.

^c Contribution of non-topological Rashba states included in the interpretation.

^d Assuming 2D carriers.

^e Reference 197 later showed that ξ_{DL}^j is largely overestimated in magnetic TIs when the asymmetric magnon scattering is disregarded in 2ω-Hall measurements.

^f Anisotropic.

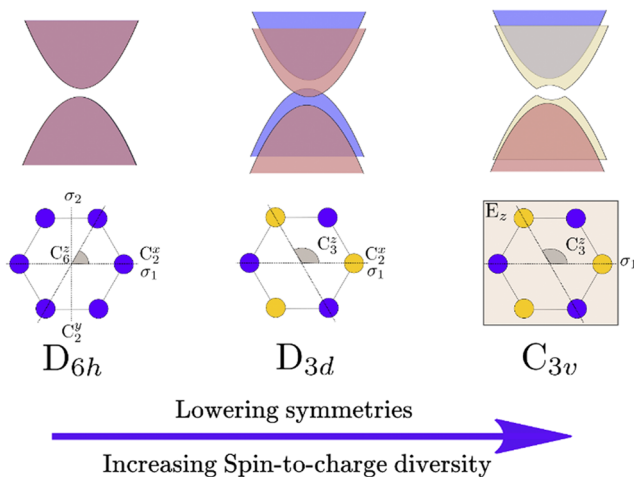


FIG. 4. Transition of the band structure and spin orientation of Dirac materials while lowering the symmetries. The quantization axis is assumed to be out-of-plane. Red and blue colors represent the bands with up and down spin. The D_{6h} point group contains 24 symmetries, including the inversion and time-reversal symmetries, which prevents spin splitting and only allows for the iSHE as the SCI mechanism due to the SOC. The D_{3d} point group contains 12 symmetries, with the inversion symmetry absent, allowing for the SHE and spin-splitting and anisotropy-like SOT. Finally, the C_{3v} point group contains six symmetries and allows for the SHE, anisotropy-like SOT, and Rashba–Edelstein effect.

and Rashba interactions on the magnetization dynamics remains to be achieved and, beyond resolving some inconsistent reports of its symmetries,²⁰¹ can lead to unprecedented features for efficient SOT applications such as zero magnetic field switching, a long sought-after mechanism for improving non-volatile memory technologies.

VI. CONCLUSIONS AND OUTLOOK

In this Perspective, we discussed the main challenges and recent results concerning materials synthesis, device fabrication, and optimization for eventually achieving the highest figures of merit for SCI in vdW heterostructures. On the material growth side, MBE is becoming an essential technique to obtain magnetic layered materials, although CVD-growth approaches and integration of two-dimensional materials in nano-electronics were already demonstrated. The next challenge is to demonstrate efficient integration of vdW materials in spintronic building blocks, such as in magnetic tunneling junctions, stimulating further research toward industry-oriented activities, for instance, in the fields of spin-transfer torque and SOT magneto-resistive random access memories. Current results concerning the SCI in vdW heterostructures combining 2D systems and topological materials are already encouraging, since they evidence high figures of merit even at room temperature in scalable materials. Besides these encouraging experimental results, advanced modeling techniques make it possible to reach a deep level of data interpretation and to predict SOT properties of arbitrary combinations of materials, even prior to their measurement, therefore acting as a pathfinder for more targeted experimental efforts. However, there are still complex theoretical issues to be tackled, such as developing very accurate effective Hamiltonian models

based on first-principles. For instance, Wannier interpolation techniques, which boil down to fitting the band structure using maximally localized Wannier functions, have proven to be very accurate in realistic charge transport simulations. Nonetheless, their extension to the investigation of non-equilibrium spin properties can be problematic, as the Wannier functions do not necessarily map the crystal structure accurately. This limitation is particularly serious for SOT simulations. Indeed, the interplay between magnetism and SOC in vdW heterostructures of interest demands special care in the study of resulting ground state spin textures in the reciprocal space as well as their modification under external electric or magnetic fields. On the experimental side, current fabrication techniques demand specific material treatment to prevent rapid degradation of produced devices, which embed some materials sensitive to oxidation effects. Efforts to synthesize materials and fabricate devices *in situ* are under way, but experimental methods to produce more air-stable heterostructures are an important task to be dealt with in the near future. As a conclusion, the field of SCI in vdW heterostructures is attracting more and more attention from various scientific communities, which gives hope that progress will be achieved in the years to come in the design of ultra-compact spin devices of relevance for non-volatile technology and spin logics.

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DATA AVAILABILITY

Data sharing is not applicable to this article as no new data were created or analyzed in this study.

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