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Spin Transport at the Frontier of Layered Spin-Orbit Coupling and Magnetism

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ABSTRACT

The discovery of an ever increasing family of atomic layered magnetic materials, together with the already established vast catalogue of strong spin-orbit coupling (SOC) and topological systems, calls for some guiding principles to tailor and optimize novel spin transport and optical properties at their interfaces. Here we focus on the latest developments in both fields that have brought them closer together and make them ripe for future fruitful synergy. After outlining fundamentals on van der Waals (vdW) magnetism and SOC effects, we discuss how their coexistence, manipulation and competition could ultimately establish new ways to engineer robust spin textures and drive the generation and dynamics of spin current and magnetization switching in 2D materials-based vdW heterostructures. Grounding our analysis on existing experimental results and theoretical considerations, we draw a prospective analysis about how intertwined magnetism and spin-orbit torque (SOT) phenomena combine at interfaces with well-defined symmetries, and how this dictates the nature and figures-of-merit of SOT and angular momentum transfer. This will serve as a guiding role in designing future non-volatile memory devices that utilize the unique properties of 2D materials with the spin degree of freedom.

Key points:

- Fabrication of 2D vdW magnetic systems offers unprecedented opportunities for controlling magnetism and spin transport phenomena down to the monolayer limit.
- Many vdW magnetic systems possess a low-symmetry crystalline structure, providing an array of exotic spin-orbit Hamiltonians, together with added richness arising from interface phenomena driven by layer-to-layer registry.
- Understanding the intertwined contribution of spin-spin interaction and interfacial symmetries is crucial for reaching the upper limit of SOT.
- This exciting research field of spin transport at the frontier of layered spin-orbit coupling and magnetism will lead to discoveries of new materials, novel transport effects, topological phenomena and unconventional electron correlation physics.

Website summary: We present a foresight on how the spintronic properties of the emerging class of layered materials combining magnetism and strong spin-orbit coupling can be tailored by proper optimization of chemical interactions and structural material symmetries. This perspective draws a route to achieve best performing material design for reaching the upper limit of spin-orbit torque efficiency in switching magnetization.

Overall context of magnetism and spin-orbit coupling materials

The realm of two-dimensional (2D) materials has considerably extended recently with the demonstration of magnetic orders in vdW material systems¹⁻⁶. Weak mechanical bonding of vdW gaps in these materials offers unprecedented

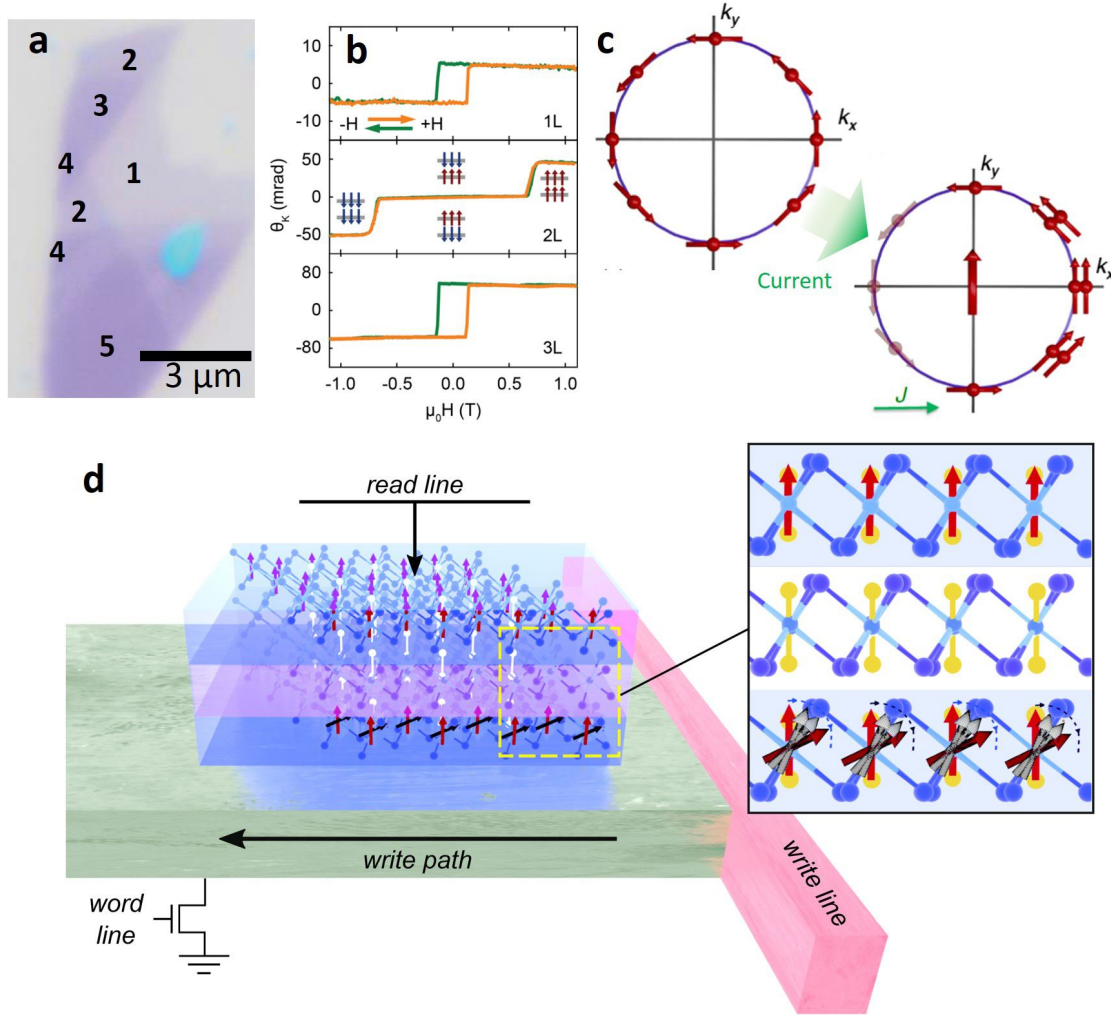


Figure 1. **a** Microscope image of an exfoliated CrI₃ flake. The numbers in the image represent expected monolayer numbers calculated by the image contrast¹. **b** Magneto-optical Kerr rotation angle measurements applied to the different monolayer number regions while sweeping magnetic field. The monolayer and trilayer regions exhibit a ferromagnetic response, whereas the bilayer region shows an antiferromagnetic behaviour. **c** Rashba spin texture in 2D momentum (reciprocal) space k_x and k_y . For the sake of simplicity, we only show one spin-subband with electrons at each Fermi surface (Left panel). When we apply an electric current along the x direction, electrons are redistributed and as a result, they become spin-polarized due to the spin texture. This is a source of SOT from the Edelstein effect. **d** Schematic of a single-cell of an SOT memory device with vdW magnetic materials. An information bit is stored by the relative orientation of two vdW magnets, which can be read by e.g. magnetoresistance through a vertical current through this stack. An external current is sourced by the write line/path electrode which is a low-resistive material such as copper. This current causes magnetization switching due to SOTs in the bottom vdW magnetic layer as highlighted in the zoom-in figure. The current-supplying bottom electrode could be another vdW material that acts as an additional spin source for boosting the switching efficiency. Figure 1a-b are adapted and modified from Ref.¹.

control of individual layer separations/fabrications by exfoliation, from which we are able to create truly monolayer magnets^{1,2} (See Fig. 1a&b as an example) and their heterostructures. This capability in sample fabrication is not readily within reach by other established thin-film growth techniques such as molecular beam epitaxy and sputtering, where the formation of grown layers significantly depends on surface energetics and kinetics together with lattice-matching between crystal structures of seed and grown layers. In contrast, exfoliation of vdW materials allows to stack different vdW monolayers without any thermodynamic constraints, enabling to access an unlimited

number of stack combinations to explore fascinating, novel electronic structures⁷⁻⁹. This will provide tremendously exciting opportunities to generate new magnetism, spin-transport and electron-correlation physics by using 2D building blocks and currently intensive and extensive research activities on vdW magnetic systems are taking place worldwide. This class of new layered vdW materials is particularly vast, and includes transition metal dichalcogenides^{10,11} (TMDs), such as CrSe₂, VSe₂, CrTe₂, transition metal trihalides¹²⁻¹⁴ (CrI₃, CrCl₃) and transition metal phosphorous trichalcogenides¹⁵ (NiPS₃, MnPS₃, etc.). 2D magnetism has been measured at low temperatures in Cr₂Si₂Te₆¹⁶, CrI₃¹ and Cr₂Ge₂Te₆² mono- and bilayers, or in VSe₂¹⁷, Fe₃GeTe₂^{4,18} and up to room temperature (RT) in single layer MnSe₂^{19,20}. In all those materials, the existence of stable magnetic properties for few layered systems is believed to be strongly related to their own magnetic anisotropies as well as the interaction with the substrate that prevents charge density wave instabilities²¹⁻²⁵.

In spintronic applications, magnetic moments are informational bits such as those in hard-disc drives and we process the bits by reversing the moment's direction from one to the other between their equilibrium states²⁶⁻²⁸. Since electronic devices operate either with current- or voltage-regulated inputs, efficient control of magnetic moments by an electric means is therefore the primary interest for developing future commercially-viable functional spintronic devices such as spin random access memory^{29,30}. The spin-orbit interaction fits to this perfectly because it microscopically couples the electron motion (i.e. current) and the spin degree of freedom, hence making it possible to manipulate spins by electric currents (Fig. 1c). The electrically-generated spin polarization can exert spin torques on local magnetic moments via the exchange interaction. This phenomenon in solid states is termed as spin-orbit torque (SOT) which has been discovered and studied mainly in magnetic multilayers with 3D crystals³¹⁻³³ and new SOT memory architectures³⁴ have been proposed and examined in the past several years. Microscopically, SOT originates from two leading effects, the spin-Hall effect³⁵ and Edelstein effect³⁶. The efficiency and mechanisms of SOT are determined by the electronic states of materials as well as interfaces, in particular inversion symmetry breaking in combination with other crystalline symmetries³⁷. In this regard, 2D vdW materials and heterostructures hold great potential to produce unique SOT phenomena from their exotic electronic structures which can be in principle modified by interface engineering. Furthermore, the efficiency of magnetization switching by SOT is proportional to magnetic volume³⁷ and so the smaller the better, ideal to the 2D limit since the volume is minimised along one dimension. In this extreme monolayer regime, it is important to stabilize magnetic orders against thermal fluctuation. This calls for detailed understanding of magnetism at that limit in terms of their driving mechanisms and the strength of them, which can be designed and controlled by material choice and external stimuli. When we have full understanding of both magnetism and SOTs in novel vdW heterostructures, we can design highly-efficient spintronic memory cells based on vdW materials such as one shown in Fig. 1d. In this cell, an information bit is stored by the relative orientation of two vdW magnetic layers separated by another vdW non-magnetic system in the middle. Tunnelling magneto-resistance effect would be best in terms of outputting a large voltage difference as a read-out. Writing actions for this cell is carried out by a horizontal current which will produce SOT effects to exert magnetic switching of the bottom layer. The efficiency of switching, in terms of electric power, is a key figure of merit since the size of a transistor to drive a current for each cell is determined by this efficiency (a large current implies bigger transistor size) and the cell density is currently limited by the transistor size. This directly means that maximising the SOT efficiency leads not only to low-power consumption of memory cells but also to the high density of data processing. In this sense, detailed understanding of physical origins and mechanisms of SOTs in any novel materials is of direct relevance of future ultracompact and low-power spin memory devices. It is important to note that non-magnetic high-spin-orbit vdW materials, such as WTe₂, could be inserted as an additional spin source for SOT-driven magnetization switching. In this regard, a whole variety of vdW heterostructures should be extensively studied for the development of novel vdW spin memory devices.

This Review particularly focuses on two fundamental aspects of 2D vdW properties for future spintronic applications. The first is on the exchange interaction as a source of magnetic order by which 2D vdW magnetic systems display different exchange interaction mechanisms depending on their crystalline structures and transport properties from insulators to metals. Understanding of these underlying mechanisms is useful to visualize different magnetic orders at play in a variety of materials and provides a good guide for designing monolayer magnets that are magnetically stable above RT. After this, we also review recent development on electric and other control of equilibrium magnetic states, which is the current state-of-the-art in the relevant field. We then focus on the potential and opportunities of SOC effects arising within 2D vdW magnetic materials as well as in cases when they are interfaced to an additional material such as substrates. Their low-symmetry point groups exhibit a colorful picture of SOT effects and by applying high-throughput point group analysis, we generate and summarize the expected symmetry of SOTs in all the class currently available in the literature. This will aid future SOT experiments using listed materials as well as those newly discovered but possessing the same crystalline symmetry as listed. We further

categorize field-like and damping-like torques existing in these materials as well as more exotic anisotropy-like torques arising from their low-symmetry properties. These two fundamental aspects have not been the focus of prior review articles on 2D vdW magnetic systems^{38–44}, which extensively cover the recent development of 2D vdW magnetic systems as well as spintronics with non-magnetic vdW materials^{45–48}.

Different exchange interactions to stabilize magnetic orders in van der Waals systems

We here briefly summarize that the magnetic order driven by the exchange interaction of electron spins has different mechanisms, depending on the conductivity regime of materials. This is very relevant to vdW magnetic systems since their conductivity spans across a wide range, from insulating to metallic regimes, possessing a very colorful nature of magnetism due to their conductivity as shown in Fig. 2. This is one of the unique potentials of vdW magnetic systems as research topics for current/future materials science for nano-magnetism and spintronic applications. The origin of long-range magnetic orders in condensed matter arises from the microscopic exchange interaction between individual spins. The exchange interaction between an array of n spins is usually captured by $H_{\text{ex}} = -\sum_{i,j}^n J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j$ where the dot product of $\mathbf{S}_i$ and $\mathbf{S}_j$ represents the relative orientation between the two spins. This Hamiltonian describes how much the spin system gains/loses their total energy by ordering their spin orientations as a whole.

The super-exchange coupling in insulators

The exchange interaction model is better suited to describe spins that are well-isolated, such as in the case of magnetic insulators and low-conducting semiconductors, where isolated spins interact with each other via non-magnetic ions (ligands) such as oxygen in their crystal lattices. Such indirect exchange coupling is often called the super-exchange mechanism⁴⁹ and a schematic to highlight this is shown in Fig. 2. This can be explained by the virtual excitation of electrons in a magnetic cation and subsequent transmission to the neighbouring cation through the non-magnetic ligand within a timescale of Heisenberg uncertainty. This virtual process effectively lowers the total energy of the system when a long-range magnetic order is present. Magnetic ions for the super-exchange interaction possess the same ionic charge states.

Many of vdW materials exhibit semiconductor/insulator properties and this extends to those possessing long-range magnetic ordering. CrX_3 ($X = \text{I, Br and Cl}$) exhibits good insulating properties with a typical bandgap energy of 1.2–3.1 eV^{1,13,14,50}. In a single layer of CrX_3 , each Cr atom is surrounded by X atoms in an octahedral configuration and the Cr atoms form a honeycomb lattice. This crystal field splits the Cr d states into e_g and t_{2g} manifolds where three electrons in their Cr^{3+} state occupy the fully-polarized t_{2g} states with $S = 3/2$ in the valence bands, following the first Hund's rule. These Cr^{3+} moments are coupled through the super-exchange mechanism via an X atom with an approximately 90 degree bonding angle^{51,52}. It is noted that due to the presence of finite antiferromagnetic coupling between Cr-Cr direct exchange mechanisms in these materials, the total exchange coupling strength (hence the size of T_C) is relatively weak⁵³. A comprehensive study for T_C of few layers of CrX_3 has been carried out by Kim et al.⁵⁴. $\text{Cr}_2\text{X}_2\text{Te}_6$ ($X = \text{Ge, Si}$) exhibits semiconductor/insulator properties due to their electronic bands with about a 0.4 eV gap^{2,16}. Very much like the case of CrX_3 , a honeycomb Cr^{3+} network stabilizes their ferromagnetic order owing to the super-exchange interaction via Te atoms. Another key group of magnetic vdW insulators is those of MXPS_3 ($M = \text{V, Mn, Fe, Co, Ni, or Zn}$)¹⁵, with a bandgap of 1.3 – 3.5 eV^{55–57}. Transition metals X host magnetic moments within the honeycomb lattice, showing either antiferromagnetic or ferromagnetic order.

The double-exchange coupling in semiconductors

When magnetic ions in a lattice have two types of ionic states, e.g. Mn^{3+} and Mn^{4+} , an exchange of the two ionic states depends on the spin state of two magnetic ions. This exchange is generated by one mobile carrier (either electron or hole) hopping between the two ions. This hopping is possible when spin states of the two magnetic ions are parallel, leading to delocalisation of the mobile carriers and reduction of their total kinetic energy. This is the double-exchange mechanism⁵⁸ that stabilises a long-range magnetic order. A prototypical example of this is $(\text{La}_{1-x}\text{Ca}_x)(\text{Mn}_{1-x}^{3+}\text{Mn}_x^{4+})\text{O}_3$ ⁵⁹. In this material series, only one type of Mn ionic states exists for both ends of their compositions ($x = 0, 1$), which are good insulators, and the super-exchange interaction stabilises antiferromagnetism in these limits. In their intermediate states, Mn^{3+} and Mn^{4+} coexist (effectively Mn^{4+} being considered as Mn^{3+} and one hole) and a ferromagnetic order outperforms the previously favored antiferromagnetic one via the double-exchange mechanism. In this regime, the presence of mobile carriers is confirmed by the conductivity which becomes several orders greater than those of the insulating compositions.

The double-exchange mechanism has not been discussed extensively in 2D vdW magnetic systems so far, although this mechanism should be commonly present for magnets with moderate electric conduction. As

discussed later, $\text{Cr}_2\text{Ge}_2\text{Te}_6$ with chemical and electric doping^{60,61} is one example where the exchange coupling strength is modified by the double-exchange mechanism, exhibiting large enhancements of the Curie temperature (T_C) compared to undoped $\text{Cr}_2\text{Ge}_2\text{Te}_6$.

The band magnetism in metals

When conduction electrons are fully delocalized and well hybridized with magnetic ions, we enter the itinerant regime where band magnetism is used to describe the magnetic order. In this mechanism, the total energy of the whole system with respect to magnetic order is discussed by the energy gain/loss of electrons close to the Fermi energy with the Pauli paramagnetism picture⁶². Based on this, together with the mean-field model⁶³, the Stoner criterion $D(E_F)U \geq 1$ (here $D(E_F)$ and U are the density of states (DOS) at the Fermi level in a non-spin-polarized state and the electron correlation energy) can be applied^{64,65}. This essentially means that when the Coulomb energy associated with the electron exchange is strong enough, spontaneous exchange-energy splitting takes place to drive ferromagnetism. This model is often employed to discuss the emergence of a magnetic order in metallic ferromagnets⁶².

Magnetic vdW materials with electron conductivity are also available, such as VX_2 ($X=\text{Se}, \text{Te}$)¹⁷, MnSe_2 ¹⁹, V_5Se_8 ⁶⁶, Fe-Ge-Te compounds^{4,18,67,68}, $\text{Fe}_{0.25}\text{TaS}_2$ ⁶⁹ and Cr-X ($X=\text{Te}, \text{Se}$) compounds⁷⁰⁻⁷². As shown in Fig. 2, these metallic 2D vdW materials tend to display higher T_C due to the direct, stronger exchange interaction between individual moments. Some of them already possess an above-RT T_C which is relevant to spintronic device applications, although a much higher T_C than RT is welcomed for stable device operation in any of these applications. The Stoner criterion is indeed discussed for some of these materials, for example Fe_3GeTe_2 ⁷³ was calculated to satisfy the inequality by $D(E_F)U = 1.56 \times 0.71 \geq 1$.

A final remark of this section is on the Mermin-Wagner theorem¹⁰¹ in which an absence of long range magnetic order in spin-rotational invariant systems is introduced. This could be statistically accurate but in reality it is difficult to achieve this since as soon as we make a real magnet, it automatically possesses any sort of magnetic anisotropies, be it from the spin-orbit interaction, anisotropic exchange interaction or magnetic dipole interaction, all of which opens up a gap for magnon excitations that suppress thermal fluctuations.

Control of magnetism and magnetic states by external stimuli

As already summarized in the aforementioned section, the transport nature in magnets is strongly associated with the type of leading exchange interaction terms and their strength. The exchange interaction can be modified by the carrier concentration as well as the distance between magnetic ions. Here we summarize two external control of magnetism and magnetic states, electric field and strain, applied to 2D vdW magnetic systems.

Electric field

By fabricating a field-effect-transistor (FET)-type device with a thin-film magnet, an external control of magnetism by the charge accumulation at the surface is possible, and leads to a modulation of the Fermi level as well as surface carrier density. Therefore, this approach is a promising way of controlling the magnetic properties *in-situ*, which has clear relevance to not only achieving high- T_C materials but also gate-tunable spintronic devices. Electric field control of magnetism and magnetic states has been of interest within the field of spintronics where atomically-thin magnetic metals, dilute magnetic semiconductors and magnetic oxides are fundamental and instrumental materials¹⁰⁴. 2D vdW magnetic systems can offer added values to this already-developed research domain as summarised below.

A dramatic change of magnetism by electric field, probed by modification of T_C , has been observed in several 2D vdW magnets. Jiang et al.¹⁰² fabricated parallel-plate capacitor devices with monolayer or bilayer CrI_3 together with a ring-shaped local ac-excitation metal patterns (schematic and microscopy images in Fig. 3a). Magnetic responses from the CrI_3 were measured by magnetic circular dichroism as ac susceptibility (Fig. 3b). A clear change of T_C by applying electrostatic voltages was observed, together with other sizable changes of the saturation magnetization (M_S) and coercive field (H_C), as summarized in Fig. 3c. Bi-layer CrI_3 also exhibits a linear magneto-electric (ME) effect due to their magnetic point group with broken spatial-inversion and time-reversal symmetries^{82,105-107}. Together with a relatively weak spin-flip transition in CrI_3 , the linear ME effect can shift the transition field and demonstrate pure electric switch between ferromagnetic and anti-ferromagnetic states^{82,107} when the applied magnetic field is set at the point of transition while the gate voltage is swept. Song et al.¹⁰⁸ demonstrated that tunneling magnetoresistance (TMR) can be electrically modified by up to one order of magnitude in four-layer CrI_3 by using bistable magnetic states with the same net magnetization, yet showing different TMR values.

An electrostatic modulation of magnetism has been also demonstrated in $\text{Cr}_2\text{Ge}_2\text{Te}_6$ by Wang et al.¹⁰⁹ and Verzhbitskiy et al.⁶¹ By applying the gate voltage to a 3.5 nm $\text{Cr}_2\text{Ge}_2\text{Te}_6$ film via a SiO_2 solid-state dielectric, Wang

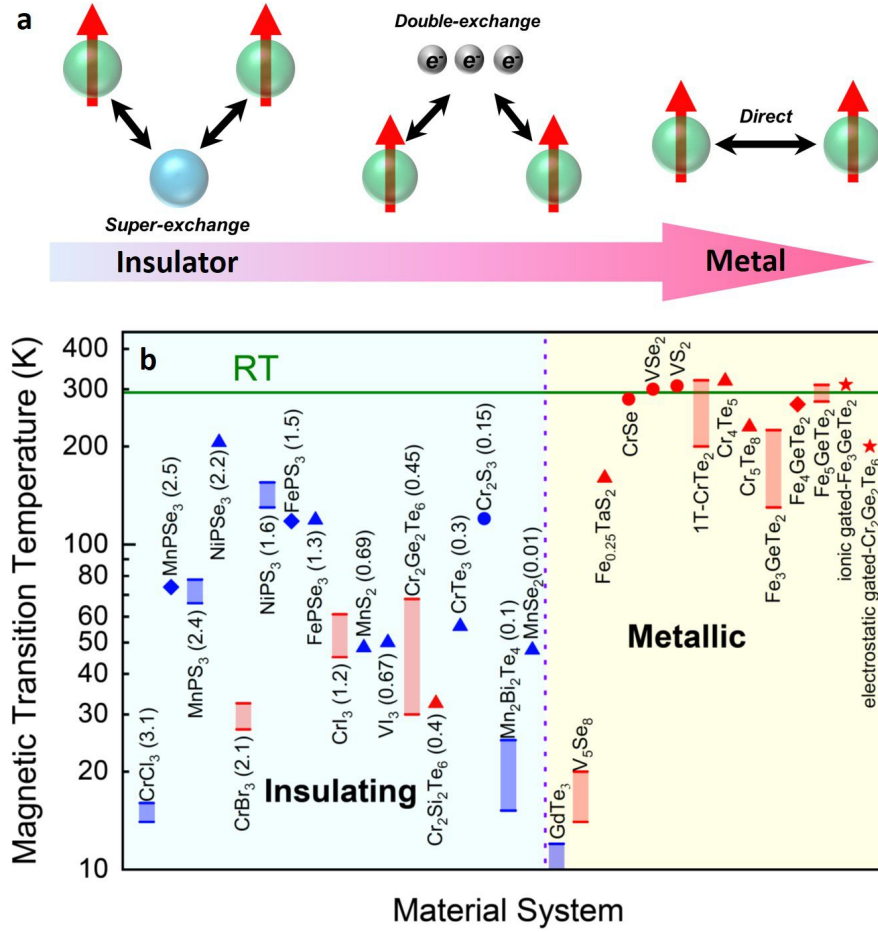


Figure 2. Experimentally validated critical magnetic transition temperature fingerprints for various 2D vdWs magnetic systems. **a** Three different exchange coupling mechanisms of two individual moments as an underlying origin of the long-range magnetic ordering for material systems in the insulating and metallic phases going from left to right. **b** Two-dimensional vdW magnetic materials library showing experimentally determined magnetic transition temperature for materials in insulating and metallic phases. In the insulating phase, number in brackets corresponds to the bandgap value. The dataset is characterised in two-ways using the colours and symbols. Colours: Red (ferromagnetic ordering) and Blue (anti-ferromagnetic ordering). Symbols: Bar (top and bottom points in the bar highlight the transition temperature for bulk and mono/few-layers, respectively), Δ (T_C is reported for the bulk case), $\circ$ (T_C is identified for mono/few-layers), $\diamond$ (T_C is independent of material thickness), and $\star$ (T_C is enhanced by the external techniques). The data points in the plot are taken from Refs. [1,2,14,17–19,55–57,61,66,68–70,73–100](#)

et al. achieved a carrier density of order of 10^{12} cm^{-2} for both electron and hole doping regimes, and observed a moderate change of M_S without any significant change of T_C . Verzhbitskiy et al.⁶¹ attempted a similar FET-type device with 20 nm thick $\text{Cr}_2\text{Ge}_2\text{Te}_6$ with an ionic gel as a dielectric as shown in Fig. 3d. With this scheme, a much stronger doping action has been observed, at the level of 10^{14} cm^{-2} and T_C of $\text{Cr}_2\text{Ge}_2\text{Te}_6$ is enhanced from 64 K (bulk)² to 200 K. This enhancement is explained by the activation of the double-exchange interaction mechanism by introducing a significant number of electrons. In this regime, about one quarter of Cr^{3+} magnetic ions become Cr^{2+} , where hopping of electrons via the Te atoms contributes to the additional exchange mechanism, enhancing the exchange coefficient as well as T_C . With this charge accumulation at the interface, the width of the region where electrons are doped electrostatically becomes comparable to the vdW gap ($\sim 1 \text{ nm}$). Therefore, we can create an artificial two-dimensional electron gas together with magnetism within the 20 nm thick $\text{Cr}_2\text{Ge}_2\text{Te}_6$. Furthermore, the magnetic easy axis of $\text{Cr}_2\text{Ge}_2\text{Te}_6$ is switched from out-of-plane to in-plane by this carrier doping⁶¹.

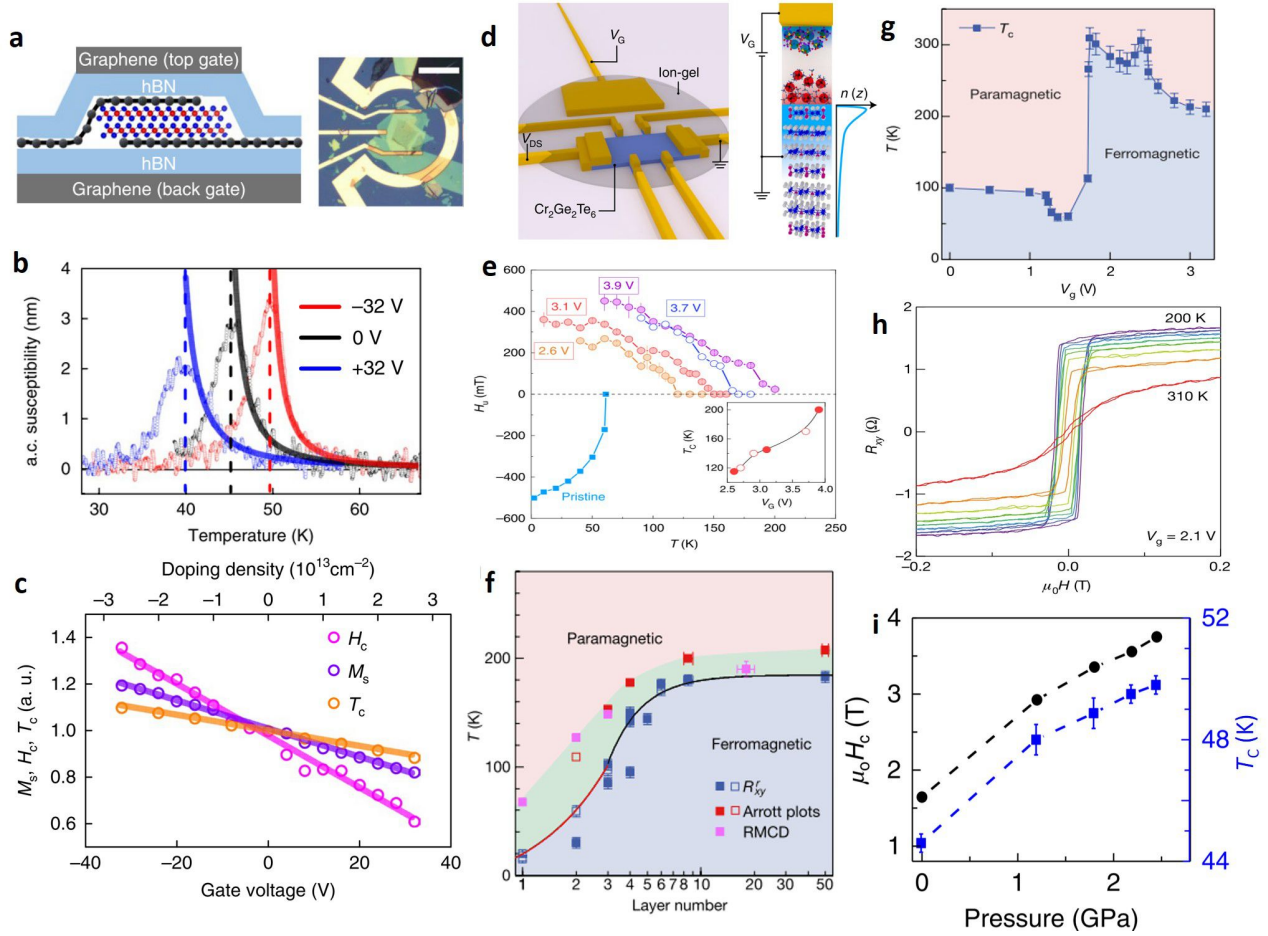


Figure 3. Experimental observation of electric field control of magnetism in different vdW magnets. **a** A schematic side view of a field-effect device with a bilayer CrI_3 encapsulated in few-layer graphene and hBN. The left panel is an optical microscope image for their device with a scale bar of $20\ \mu\text{m}$. **b** Electric field control of magnetism in measured by ac susceptibility with different gate voltages as shown. **c** Coercive force (magenta), saturation magnetization (purple) (both at 4 K) and Curie temperature (orange) normalized by their values at zero gate voltage as a function of gate voltage and induced doping density. **d** Schematics of a field-effect-transistor device with $\text{Cr}_2\text{Ge}_2\text{Te}_6$ and side view of charge accumulation by electric field. **e** Experimentally-deduced out-of-plane anisotropy field in $\text{Cr}_2\text{Ge}_2\text{Te}_6$ as a function of temperature for different gate voltages, with T_C with gate voltages (inset). **f** T_C of Fe_3GeTe_2 with different layer thicknesses measured by different analysis methods shown. **g** Gate voltage dependence of T_C in a trilayer Fe_3GeTe_2 and **h** their hysteresis loops for different temperatures. **i** the extracted critical field for spin-flip transition (black circles) and critical temperature (blue squares) as a function of pressure in a trilayer CrI_3 . Figures are adapted from Ref.¹⁰² (Fig. 3a-c), Ref.⁶¹ (Fig. 3d-e), Ref.¹⁸ (Fig. 3f-h) and Ref.¹⁰³ (Fig. 3i) respectively.

While CrI_3 and $\text{Cr}_2\text{Ge}_2\text{Te}_6$ are magnetic insulators in their pristine forms, the electric field modulation of magnetism has been also explored in metallic itinerant vdW magnets such as Fe_3GeTe_2 ¹⁸. Deng et al. performed a detailed study of Fe_3GeTe_2 's T_C with different layer numbers (Fig.3f) and applied electric fields onto a trilayer Fe_3GeTe_2 film using solid electrolyte (LiClO_4 dissolved in polyethylene oxide matrix). T_C is greatly enhanced from 100 K to above 300 K by application of around 2 V, as shown in Fig. 3g&h. This gate-tunability of T_C can be understood by the Stoner model^{164,65} since the energy gain by forming a ferromagnetic phase depends on the size of DOS at the Fermi level. Theoretical calculations suggest that by applying the electric field, the Fermi level can be lifted up to a large DOS peak area in Fe_3GeTe_2 , mainly consisting of Fe d_{z^2} , d_{xz} and d_{yz} orbitals¹⁸. The non-monotonic behaviour of T_C as a function of gate voltage may imply this Fermi level passing by the peak points.

Strain

Strain is also a good external knob to modify the magnetic ground state of 2D vdW magnets. In some of these magnetic materials, the interlayer exchange coupling governs the ground-state spin orientation, leading for instance to antiferromagnetism in CrI₃. Since the exchange coupling is very sensitive to the distance between two spins (in this case, the vdW gap distance), any external perturbation modifying this length enables the control of magnetism, as shown with hydrostatic pressure of few GPa which can induce change of T_C of bulk vdW crystals Cr₂Ge₂Te₆¹¹⁰, CrI₃¹¹¹ and VI₃⁸⁵ as well as the sign change of the two-fold magnetic anisotropy in Cr₂Ge₂Te₆¹¹². In the thin-film regime, Song et al.¹⁰³ and Li et al.¹¹³ both independently applied hydrostatic pressure of a few GPa on a few monolayer of CrI₃ and observed a monoclinic-to-rhombohedral structural transition, driven by interlayer antiferromagnetic or ferromagnetic couplings^{114–117}. Similarly, a ferromagnetic-antiferromagnetic transition under applied pressure, as well as pressure-controlled magnetic state switching between antiferromagnetism to ferromagnetism have been also demonstrated¹⁰³. Such monoclinic-to-rhombohedral structural transition can be also observed in exfoliated CrCl₃ crystals with a variation of the interlayer exchange coupling by up to one order of magnitude, when compared to bulk samples¹¹⁸. This result highlights that sample fabrication processes unintentionally produce strain that greatly perturbs the magnetic properties of exfoliated magnets. Whether this monoclinic phase is thermodynamic ground states or meta-stable states induced by their exfoliation processes is yet to be revealed¹¹⁸. Furthermore, T_C can be modified by application of strain e.g. to a trilayer of CrI₃ as shown in Fig. 3i.

Spin-orbit torques in two-dimensional materials

While the magnetic properties have been successfully controlled in a certain number of 2D vdW magnets, the access to out-of-equilibrium regimes in such materials by using electric currents and other means presents formidable challenges, but also holds paramount potential for future spintronic memory and logic applications^{30,119–121}. This area of research is growing fast but to date only a very limited number of reports are available in the literature^{122–127}. Using electric fields, induced non-equilibrium spin polarization can result in substantial angular momentum transfer and magnetization control to a certain degree. This has been achieved using non-collinearity between two magnets¹²⁸ in magnetic tunnel junctions, or via the intrinsic SOC of materials³⁷. In this section, we start with an overview of the latest experimental demonstrations of magnetization control by electric currents with 2D vdW materials, and then present a theoretical analysis of spin torque physics.

TMDs with high spin-orbit coupling (SOC), such as WTe₂ and NbSe₂, have already been employed to generate spin torques in ferromagnetic metals^{122–124}. In particular, MacNeill and coworkers observed that the torque symmetry arising in NiFe/WTe₂ bi-layers results from the symmetry lowering at the interface¹²², exhibiting the damping-like symmetry with the spin-polarization along the out-of-plane direction which cannot be generated by the spin-Hall effect³⁷. Magnetic moments in vdW ferromagnets have been also manipulated by current-induced spin torques^{125–127}. Interestingly, the spin-Hall effect in Pt was used to generate spin currents into 10 ~ 20-nm-thick exfoliated Fe₃GeTe₂ thin-films, and induce magnetization switching for low current density of 2.5×10^{11} A/m²^{125,126}. This current density is actually comparable to the one needed to switch standard 3d transition metal alloys³⁷. A magnetization switching by the spin-Hall effect has been also discussed in the insulator-Cr₂Ge₂Te₆/Ta system, requiring current density of $\sim 5 \times 10^{11}$ A/m², comparable to the one necessary for Tm₃Fe₅O₁₂¹²⁹. Finally very recently, using current density of about 3.9×10^{12} A/m², a spin-orbit torque induced magnetization switching in an all vdW heterostructure was also reported in Fe₃GeTe₂/WTe₂ stacks¹³⁰.

So far, the majority of studies on current-induced spin-torque manipulations in 2D vdW magnets exploit the properties of conventional heavy metals to generate the spin source. Due to the intrinsic nature of vdW materials, some 2D vdW magnets lack an inversion center in their crystalline structures, hence allowing for spin-orbit Hamiltonian with linear and/or cubic in momentum. This fundamentally means that these materials possess spin textures in momentum space which can be tailored by electric currents for magnetisation control^{36,37}. While spin-orbit torque arising from spin-textures in momentum space has been demonstrated and studied in great detail in 3D crystals^{31,131–133}, including the demonstration of damping-like torques due to the Berry-curvature¹³⁴, to date no such result have been reported in 2D vdW magnets. Since electronic states in 2D vdW materials are inherently very sensitive to any perturbation caused at interfaces, the 2D vdW material family offers a fascinating and promising playground to explore novel and efficient current-induced spin torques with a large number of possible heterostructure combinations. To guide this research development, we here present a summary of possible spin-orbit torque symmetries for different point group crystals, including the contributions emerging at interfaces. We remind the reader that following Neuman's principle, it is the point group which governs the allowed types of linear response tensors, being themselves translation invariant, and that it is the space group of the material which conditions the band structure, which can have an effect on the magnitude of the response¹³⁵. We begin with basic

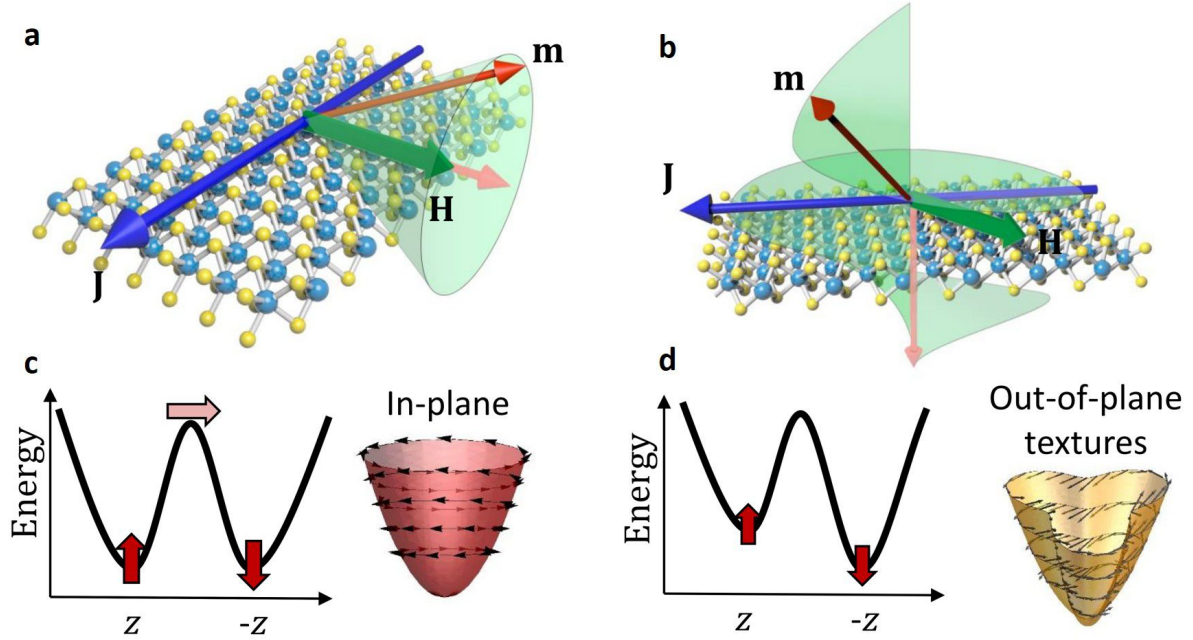


Figure 4. Illustration of spin-orbit torque mechanisms for different spin-textures and their effects on the magnetic energy profile. **a** Magnetization dynamics due to a typical effective SOT field for systems with multiple orthogonal mirror planes. **b** Magnetization dynamics for effective SOC fields with a component parallel to the anisotropy axis. **c** Magnetic energy density for a system with perpendicular magnetic anisotropy and a weak in-plane effective SOC field produced an in-plane spin texture. **d** Magnetic energy density for a system with perpendicular magnetic anisotropy and an effective field with a component parallel to the anisotropy axis, and the spin-textures that enable these kind of fields.

descriptions of the underlying spin physics and then elaborate a broader picture of torque categories, followed by a discussion about the theoretical methodologies needed to explore the nature and strength of the torque phenomena.

Basics of magnetic torques and spin-orbit interaction

The magnetization in ferromagnetic materials $\mathbf{M}$ originates from the spin angular momentum $\mathbf{s}$ of lattice atoms. The exchange interaction enforces the magnetic order but since it is generally isotropic, a source of magnetic anisotropy is required to dictate the direction of the magnetization at equilibrium, and to stabilize the magnetic phase. Typically, this interaction defines two equilibrium positions along the positive and negative directions of the anisotropy axis, which is characterized by two symmetric minima in the system's energy. An external magnetic field with a component parallel to such an axis will then favor a particular minimum and translates into a torque $\mathbf{T}$ on the magnetization, which will align it along the field direction. Still, large fields are required to induce stable and reversible switching, a condition which remains a significant technological challenge. An alternative way is provided by manipulating the SOC to produce electrically driven torques on the magnetization, known as SOT¹³⁶.

SOC is the consequence of the periodic electrical potential due to the nuclei within the crystal, which is felt by the electrons as an effective magnetic field $\mathbf{B}_{\text{soc}}$ proportional to its angular momentum $\mathbf{L}$ ¹³⁷. The characteristics of the coupling between the spin and this effective field dictate the resulting strength and symmetry properties of the dominant SOC terms. Additionally, the spin of the conduction electrons propagating through a magnetic material will couple to its total magnetization through a Zeeman-like interaction $-(2\Delta_{\text{ex}}/\hbar)\mathbf{s} \cdot \mathbf{m}$, where $\hbar$ the reduced Planck's constant¹³⁸, $\mathbf{m} \equiv \mathbf{M}/|\mathbf{M}|$ the normalized magnetization, and Δ_{ex} the effective exchange interaction between the localized and itinerant spins. Therefore, in magnetic systems with SOC, the spin of conduction electrons will be driven by the torque produced by a combination of the exchange field $\mathbf{B}_{\text{ex}} \equiv -(2\Delta_{\text{ex}}/\hbar)\mathbf{m}$ ^{138,139} and the effective momentum-dependent spin-orbit field $\mathbf{B}_{\text{soc}}(\mathbf{p})$ ¹³⁷. Conversely, the action-reaction law states that the magnetization will feel an opposite momentum-dependent torque due to the electronic spin $\mathbf{T}_{\text{soc}}(\mathbf{p})$ and will be then indirectly coupled to spins of the itinerant electrons which are also affected by the SOC¹⁴⁰. We should remember however

that since $\mathbf{B}_{\text{soc}}$ is a relativistic effect, it does not exist in the laboratory frame. As a result, any non-zero torque induced by this field will cancel in equilibrium owing to time-reversal symmetry invariance. Nevertheless, under the action of an external electrical bias ΔV , electrons around the Fermi level E_F are bestowed with extra energy that promotes a reorganization of charge favoring electrons with momentum parallel to the electric field. This non-equilibrium state opens the possibility of a finite non-equilibrium spin density, an effect first discovered by Edelstein³⁶ and referred to as inverse spin galvanic³⁷. From a different point of view, such a non-equilibrium spin density can be understood as stemming from an effective macroscopic SOC field $\mathbf{H}_{\text{soc}}(\Delta V)$, that is a function of the bias and promotes a macroscopic torque $\mathbf{T}_{\text{soc}} = \mathbf{M} \times \mathbf{H}_{\text{soc}}$. In this sense, the bias provides the energy to maintain spins alignment with $\mathbf{H}_{\text{soc}}(\Delta V)$, and it will also affect the magnetization direction through the exchange interaction. The direction and strength of this field will actually depend on the average of the momentum-dependent spin polarization, which is usually termed as spin texture and is computed as the expectation value of the spin operators in Bloch's states. In Fig. 4c, a sketch of this phenomenon is shown, where the surface represents the allowed energies $E(\mathbf{p}, \mathbf{s})$ and the arrows indicate the spin texture pattern. It is clear from the figure that in the chosen situation, the spin texture compensates after summation over all allowed momentum. Alternatively, we could also consider the field $\mathbf{H}_{\text{soc}}$ as an electrically induced non-equilibrium magnetic anisotropy that promotes the alignment along a different orientation¹⁴¹.

In systems with an inversion center, the Bloch's states are doubly degenerated for any given momentum, leading to two superimposed and opposite spin textures that will suppress any possible spin torque¹⁴². This result implies that the SOTs requires a broken inversion symmetry, as typically achieved through the creation of an interface. In layered materials though, the inversion symmetry can be lifted by multilayer stacking^{143,144}, strain¹²³, or even the interaction with the substrate¹²⁵. The second class of highly symmetric 2D systems are those possessing a 4-fold rotation axis or higher. At the Γ point of those systems, only in-plane fields perpendicular to the layer and the current such as the one illustrated in Fig. 4a are allowed by the in-plane spin textures (Fig. 4c) enforced by the symmetries¹⁴⁵. In systems with perpendicular magnetic anisotropy, such a field will gradually reduce the barrier in Fig. 4c without favouring any particular minima, and therefore, cannot produce deterministic switching. However spin-pseudospin coupling¹⁴⁶ and other internal degrees of freedom could lead to nontrivial out-of-plane textures^{147,148}, a possibility that remains largely unexplored in the past. In fact, currently the role of symmetries is under extensive experimental study due to its potential for technological applications^{122,149}. Exploiting this out-of-plane spin texture (as depicted in Fig. 4d) is particularly rewarding since it breaks the energy degeneracy of the moment pointing $\pm z$ directions by an electric current as shown in Fig. 4b&d. This leads to deterministic control of the magnetization switching by an electric current without any needs of external magnetic fields in such spintronic switching devices.

Symmetries of the spin-orbit torques in layered materials

The quantitative determination of spin-textures in momentum space requires knowledge of the microscopic interactions at play, which are complicated to measure. Still, they lead to the SOT which is a macroscopic observable, or equivalently, an effective magnetic field $\mathbf{H}_{\text{soc}}$. The underlying lattice, equilibrium magnetic symmetries and atomic structure fully define the SOC and, as consequence, will also determine the allowed SOT symmetries. Moreover, crystal symmetries will also manifest in the torque's dependence on the enforced current $\mathbf{J}$ and the magnetization direction $\mathbf{m}$ ¹³⁹. In general, $\mathbf{H}_{\text{soc}}$ can be written in terms of the current and magnetization^{139,150–152}.

To illustrate the method, we will limit ourselves only to terms linear in the current and the magnetization. We already determined that a non-equilibrium state is mandatory to produce the spin-orbit fields forcing it to be proportional to current density at lowest order. Its dependence against the magnetization direction is more complicated since there are two possible contributions,

$$\mathbf{H}^e \equiv \mathbf{Y}\mathbf{J}, \quad (1)$$

independent of magnetization and linked to the current density through the 3×3 matrix $\mathbf{Y}$, and

$$\mathbf{H}^o \equiv \mathbf{m}\mathbf{\Lambda}\mathbf{J}, \quad (2)$$

which is proportional to the magnetization and is mixed linearly with the current density through the rank-3 tensor $\mathbf{\Lambda}$. The superscript e/o are included to highlight the even and odd dependence of these fields against magnetization reversal. These two objects comprises over 36 terms that need to be determined to fully describe the torque's symmetries. Through symmetry analysis, these elements can be further reduced to up to two parameters.

To outline how this symmetry analysis work, we will first start by defining a point symmetry $\mathcal{R}$, which is a geometric operation that leaves the positions of the system invariant with respect to a given point, and

are representable as 3×3 unitary matrices if we represent positions as three-dimensional vectors. The main assumptions of symmetry analysis is that a representation of these symmetries will also act on all crystal's physical properties, which is known as Neumann's principle^{139,142}. In this sense, both the current density and magnetization must remain invariant against the action of all system's symmetries $\{\mathcal{R}_1, \mathcal{R}_2, \dots, \mathcal{R}_N\}$, where N the total number of symmetry operations. This will lead to a set of coupled equations that will restrict the number of allowed components. In general, one must evaluate the N symmetries present in the material, but by using group theory it is possible to reduce it to a much smaller minimal set of symmetries known as the generating set, that can be combined to span the whole group¹⁴², and it is sufficient to impose invariance only in that set.

To wrap up and settle the concepts presented here, we provide an example in **Textbox**, which illustrates how to use the symmetry analysis to determine the allowed torques of the C_{3v} point group. This group possesses six symmetry elements but a generating set of only two, which we choose to be a rotation of 120° and a reflection on the yz plane, which are generic for 2D honeycomb structures and serve as a starting point to describe many different materials such as: graphene heterostructures¹⁵³, hexagonal transition metal dichalcogenides, Fe_3GeTe_2 ¹⁵⁴, CrI_3 . Materials belonging to higher symmetry groups such as D_{6h} or C_{6v} will possess fewer allowed torques.

Textbox

The C_{3v} point group of symmetries possess a generating set given by a 120° rotation around the z -axis and a reflection on the yz plane. For any possible symmetries $\mathcal{R}$, the matrices $\mathbf{Y}$ and $\mathbf{\Lambda}$ must satisfy the following set of equations^{139,155} $\mathbf{Y} = \det(\mathcal{R}) \mathcal{R} \mathbf{Y} \mathcal{R}^T$ and $\Lambda^i = \sum_{i'=x,y,z} \mathcal{R}_{i,i'} \mathcal{R} \Lambda^{i'} \mathcal{R}^T$, where $i, j, k \in (x, y, z)$ defining the direction of the axes of a chosen Cartesian system. Below we show how these tensors change after successive application of the two symmetries.

The diagram illustrates the transformation of symmetry tensors $\mathbf{H}_{\text{soc}}^e$ and $\mathbf{H}_{\text{soc}}^o$ under the C_{3v} point group symmetries. It shows three stages: the initial state, reflection along the x -axis, and a 120° rotation around the z -axis.

Initial State:

- Coordinate system: x (right), y (up), z (out of page, indicated by a dot in a circle).
- $\mathbf{H}_{\text{soc}}^e$ matrix: $\begin{pmatrix} Y_{xx} & Y_{xy} & Y_{xz} \\ Y_{yx} & Y_{yy} & Y_{yz} \\ Y_{zx} & Y_{zy} & Y_{zz} \end{pmatrix}$
- $\mathbf{H}_{\text{soc}}^o$ matrix: $\begin{pmatrix} \Lambda_{xx}^x & \Lambda_{xy}^x & \Lambda_{xz}^x \\ \Lambda_{yx}^x & \Lambda_{yy}^x & \Lambda_{yz}^x \\ \Lambda_{zx}^x & \Lambda_{zy}^x & \Lambda_{zz}^x \\ \Lambda_{xx}^y & \Lambda_{xy}^y & \Lambda_{xz}^y \\ \Lambda_{yx}^y & \Lambda_{yy}^y & \Lambda_{yz}^y \\ \Lambda_{zx}^y & \Lambda_{zy}^y & \Lambda_{zz}^y \\ \Lambda_{xx}^z & \Lambda_{xy}^z & \Lambda_{xz}^z \\ \Lambda_{yx}^z & \Lambda_{yy}^z & \Lambda_{yz}^z \\ \Lambda_{zx}^z & \Lambda_{zy}^z & \Lambda_{zz}^z \end{pmatrix}$ (Note: The diagram uses color-coding: blue for y -components, red for z -components, and black for x -components).

Reflection along x :

- Coordinate system: x (right), y (down), z (out of page, indicated by a dot in a circle).
- $\mathbf{H}_{\text{soc}}^e$ matrix: $\begin{pmatrix} 0 & Y_{xy} & Y_{xz} \\ Y_{yx} & 0 & 0 \\ Y_{zx} & 0 & 0 \end{pmatrix}$
- $\mathbf{H}_{\text{soc}}^o$ matrix: $\begin{pmatrix} 0 & \Lambda_{xy}^x & \Lambda_{xz}^x \\ \Lambda_{yx}^x & 0 & 0 \\ \Lambda_{zx}^x & 0 & 0 \\ \Lambda_{xx}^y & 0 & 0 \\ 0 & \Lambda_{yy}^y & \Lambda_{yz}^y \\ 0 & \Lambda_{zy}^y & \Lambda_{zz}^y \\ 0 & \Lambda_{zy}^z & \Lambda_{zz}^z \\ 0 & \Lambda_{yy}^z & \Lambda_{yz}^z \\ 0 & \Lambda_{zy}^z & \Lambda_{zz}^z \end{pmatrix}$

120° rotation around z :

- Coordinate system: x (right), y (up), z (out of page, indicated by a dot in a circle).
- $\mathbf{H}_{\text{soc}}^e$ matrix: $\begin{pmatrix} 0 & -Y_{yx} & 0 \\ Y_{yx} & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}$
- $\mathbf{H}_{\text{soc}}^o$ matrix: $\begin{pmatrix} 0 & -\Lambda_{yy}^y & \Lambda_{yz}^y \\ -\Lambda_{yy}^y & 0 & 0 \\ \Lambda_{zy}^y & 0 & 0 \\ -\Lambda_{yy}^z & 0 & 0 \\ 0 & \Lambda_{yy}^y & \Lambda_{yz}^y \\ 0 & \Lambda_{zy}^y & \Lambda_{zz}^y \\ 0 & \Lambda_{zy}^z & 0 \\ \Lambda_{yy}^z & 0 & 0 \\ 0 & \Lambda_{yy}^z & 0 \\ 0 & 0 & \Lambda_{zz}^z \end{pmatrix}$

The matrix $\mathbf{Y}$ is antisymmetric and translate into a cross product between the current and the rotation axis $\mathbf{H}^e = Y_{yx} \mathbf{J} \times \hat{\mathbf{z}}$. The second term is less straightforward but it is, still, fully defined by just five parameters, that reduces to three for 2D materials. This is a substantial reduction in the number of degree of freedom that one would, e.g., use in a phenomenological fit of the experiment. Also, note that a microscopic calculation would automatically give such reduction, which can be used as a check, or simply one can use the symmetry to reduced calculation cost.

A key feature of 2D materials and their combination is the possible variability of symmetry situations, including interface-induced symmetry breaking^{125,153,156–158}. For instance, stacking different 2D materials lift the inversion symmetry and can artificially enhance the SOC and exchange interaction^{143,152,159–162}. Accordingly, it is possible to design and tailor the SOT symmetry by the choice of materials and the engineering of 2D material stacking. There are several studies that show how symmetries affect the magnetization dynamics through a 2D layer inserted at an

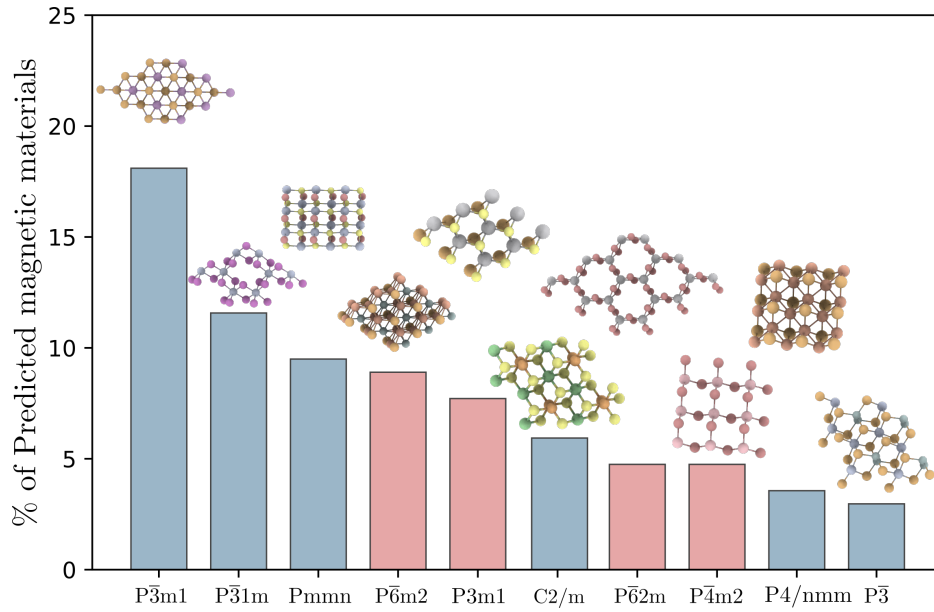


Figure 5. Distributions of magnetic materials predicted using first principle calculations as a function of their Symmetry. The materials family, which requires a substrate-induced inversion symmetry breaking to generate torque components, is identified by the filled blue histograms. Non-centrosymmetric materials which may manifest SOT are outlined by filled red histograms.

interface^{122,123,127,162–166}. However, 2D magnets can also display an internally generated SOT due to their combined SOC, broken inversion symmetry, and exchange interactions, i.e. not needing an interface or spin-injection from an adjacent layer, opening the door for *all-in-one* SOT-memories. To date, the number of synthesized 2D magnetic materials is still very limited, but novel materials and combination emerge regularly. Most of the presently known systems belong to the same spatial group and hence will display the same kind of torques.

Material screening and grouping

We here perform a brute-force screening of all possible 2D magnetic materials in the C2DB database¹⁶⁷ and group them according to their symmetry. To do this, we selected only those that were predicted to be stable in their freestanding form according to the database's stability criteria¹⁶⁷. We also discarded all materials without a point group symmetry since we will restrict the discussion to those materials where symmetry analysis is able to provide physical insights of the torques based on its point group. The final result is presented in Fig. 5, where it is clear that hexagonal and trigonal systems are ubiquitous, in agreement with the trends observed in experimental synthesis. Table I provides allowed torques for the most common space groups and gives examples of representative materials that could display them. The space group is shown in the international notation, and we set the z -axis as the high-symmetry rotational axis and aligned the x -axis to one of the lattice vectors. The table is ordered based on the number of symmetry operators in each group, or equivalently, from the highest-to-lowest symmetric groups. In this sense, it is clear that the lowest symmetry group $C2/m$ requires, in general, 12 parameters to define all possible torques, whenever the highest symmetry group is limited to three parameters. The table also reinforces the result presented in Textbox 1 regarding the stringent role of symmetries concerning the allowed torque components. For example, when there is a two-fold rotation, it is clear there will be different torques pinned to the two perpendicular directions that we choose to be the x and y axes, and in general, one would expect all torques to be defined by a 2×2 matrix. The presence of vertical mirror planes or in-plane rotational axes further halves the torque components quantity. Systems with three-fold rotational symmetry are the best examples to understand the emergence of novel torques upon symmetry breaking. To better appreciate this mechanism, let us focus on the torques odd against magnetization reversals of materials with the spacegroup $P\bar{3}$, which are the simplest to analyse although the result extends to the those with even dependence. These materials are centrosymmetric but lose this property and belong to the reduced $P3$ group when deposited onto a substrate. The remaining three-fold rotational symmetry then permits only two kinds of odd torques, one perpendicular and another parallel to the current. The trigonal structure which only maintains a single vertical mirror plane will then suppress the torque component parallel to

the current. Similar ideas apply to the odd components of the torques. Therefore, the main message here is that within a given class of crystals, new torques will appear when the interaction with the environment starts to disrupt internal crystalline symmetries. These new torque components will be superimposed to the intrinsic ones robust to symmetry breaking effects providing a way to optimize the characteristics of the torque by symmetry engineering of materials. Another interesting result is that following this approach, we recover most of the synthesized 2D magnets such as CrI₃, CrGeTe₃ and Fe₃GeTe₂, but also some unexplored families such as the trihalides and the cubic magnets such as FeTe. In the same table we show the SOT directions allowed by the symmetries for all materials in the freestanding form (blue color) and under a substrate (red color). Most of the system present inversion symmetry and as a result, do not show any torque in their freestanding form and require the presence of a substrate (modelled here by either removing the mirror plane parallel to the layer or the inversion operator out of the generating set). Interestingly, we also identify a set of materials with intrinsic crystal asymmetry leading to an *intrinsic* torque in Fe₃GeTe₂ (P6m2) and certain magnetic trihalides like CrBr₃ (P4m2)*, and magnetic Janus structures such as MnSTe (P3m1)[†]. Additionally, we confirm that the angular dependence of this internally generated torque changes once the substrate is incorporated and breaks additional symmetries, with the only exception of the magnetic Janus TMD^{168,169}, for which the angular dependence remains the same.

Isotropic and anisotropic torques

The Eqs.(1,2) allow us to consider the SOT as a phenomenon produced by a current-induced effective magnetic field prescribed by the symmetries and the SOC. However, despite the apparent simplicity of these expressions, the fields are function of both the magnetization direction and the applied current, and can take very complex functional forms as it is evident from the less symmetric materials reported in Table 1. Nevertheless, given that the functional form of the allowed torques in isotropic systems (i.e, two-dimensional electron gas) resembles the one produced by the external effective magnetic given by $\mathbf{H}_{\text{eff}} \propto \mathbf{J} \times \hat{\mathbf{z}}$, early studies classified them in two components, a field-like torque that produces magnetization precession and is characterized by the parameter τ_{FL} , and a damping-like torque that aligns the magnetization along the field direction and is proportional to τ_{DL} ¹⁷⁰. These two torques defines what we term as *isotropic* torques

$$\mathbf{T}_{\text{iso}} = \mathbf{m} \times [\tau_{\text{FL}} \mathbf{J} \times \hat{\mathbf{z}} + \tau_{\text{DL}} \mathbf{m} \times (\mathbf{J} \times \hat{\mathbf{z}}) + \tau_z m_z \mathbf{J}], \quad (3)$$

given that the strength depends solely on the current magnitude J , allowing the definition of even and odd torques using a single parameter for each. Here, we use three parameters τ_{FL} , τ_{DL} and τ_z to represent the field strength for each symmetry. In fact, by following the procedure outlined in the Textbox 1, it is straightforward to show that such a simple description, where the torque is fully characterized by a *single* effective field, becomes characteristic of systems with two orthogonal vertical mirror planes, including totally isotropic systems. In the less symmetrical 2D systems without these two mirror planes, there will be a structural anisotropy between two orthogonal directions which would translate into an anisotropic electrical response. As a consequence, the current response will be characterized by the two diagonal elements of the conductivity tensor (i.e σ_{xx} and σ_{yy}), while the torque response will be generally described by two or more effective magnetic fields. This means that the strength of the total torque will depend both on the current orientation and magnitude.

To better grasp these concepts, let us consider first the case of system possessing a three-fold rotational symmetry and at least a mirror plane (trigonal). In Textbox 1 we demonstrated how to compute the torque matrices for the C_{3v} point group symmetry and in Table 1 we combined it with Eq.(1) and Eq.(2) to express it in vectorial form, leading to Eq.(3). The first two torques are torques produced by the current-induced spin-polarization along $\mathbf{J} \times \hat{\mathbf{z}}$ with field-like and damping-like mechanisms respectively. Both are described by a single parameter indicating that the strength of the effective field does not depend on the current orientation. The last torque term is parallel to the current, becoming non-negligible only when there is an out-of-plane component of the magnetization m_z . When the mirror plane is removed, a novel field-like torque emerges that is perpendicular to both the current and the conventional field-like torque

$$\mathbf{T}_{\text{FL}}^{\text{ani}} = \mathbf{m} \times [\tau_{\text{FL}}^I \mathbf{J} \times \hat{\mathbf{z}} + \tau_{\text{FL}}^{II} \mathbf{J}]. \quad (4)$$

As a result, this acts as a superposition of two perpendicular in-plane effective magnetic fields and thus leads to an anisotropic response as a function of the current direction.

*Display intrinsic anisotropy-like SOT

[†]Display intrinsic field-like and damping-like SOT

Table 1

Repr. Mat.	Space Group	Crystal System	$\mathbf{H}^e$	$\mathbf{H}^o$
MX_3^*	$\text{P}\bar{6}2\text{m}$ (189)	Hexagonal	$\tau_{\text{FL}} \mathbf{J} \times \hat{\mathbf{z}}$	$\tau_{\text{DL}} \mathbf{m} \times (\mathbf{J} \times \hat{\mathbf{z}}) + \tau_z m_z \mathbf{J} + \tau_{\text{ani}} \nabla_{\mathbf{m}} [J_y m_x m_y + J_x (m_y^2 - m_x^2)/2]$
$\text{Fe}_3\text{GeTe}_2^*$	$\text{P}\bar{6}\text{m}2$ (187)			$\tau_{\text{DL}} \mathbf{m} \times (\mathbf{J} \times \hat{\mathbf{z}}) + \tau_z m_z \mathbf{J} + \tau_{\text{ani}} \nabla_{\mathbf{m}} [J_x m_x m_y + J_y (m_x^2 - m_y^2)/2]$
MnBi_2Te_2	$\text{P}\bar{3}\text{m}1$ (164)	Trigonal	$\tau_{\text{FL}} \mathbf{J} \times \hat{\mathbf{z}}$	$\tau_{\text{DL}} \mathbf{m} \times (\mathbf{J} \times \hat{\mathbf{z}}) + \tau_z m_z \mathbf{J} + \tau_{\text{ani}} \nabla_{\mathbf{m}} [J_x m_x m_y + J_y (m_x^2 - m_y^2)/2]$
$\text{MXY}^\dagger$	$\text{P}\bar{3}\text{m}1$ (156)			
Fe_5GeTe_2	$\text{R}\bar{3}\text{m}$ (147)			
CrI_3	$\text{P}\bar{3}1\text{m}$ (162)			$\tau_{\text{DL}} \mathbf{m} \times (\mathbf{J} \times \hat{\mathbf{z}}) + \tau_z m_z \mathbf{J} + \tau_{\text{ani}} \nabla_{\mathbf{m}} [J_y m_x m_y + J_x (m_y^2 - m_x^2)/2]$
CrGeTe_3	$\text{P}\bar{3}$ (147)	Trigonal	$\tau_{\text{FL}}^I \mathbf{J} \times \hat{\mathbf{z}} + \tau_{\text{FL}}^II \mathbf{J}$	$\tau_{\text{DL}}^I \mathbf{m} \times (\mathbf{J} \times \hat{\mathbf{z}}) + \tau_z m_z \mathbf{J} + \tau_{\text{ani}}^I \nabla_{\mathbf{m}} \left[J_y m_x m_y + \frac{J_x (m_x^2 - m_y^2)}{2} \right] + \tau_{\text{ani}}^II \nabla_{\mathbf{m}} \left[J_y m_x m_y + \frac{J_x (m_x^2 - m_y^2)}{2} \right] + \tau_o^II [(\mathbf{J} \times \mathbf{m}) \cdot \hat{\mathbf{z}}] \hat{\mathbf{z}}$
$\text{FeTe}^\dagger$	$\text{P}4/\text{nmm}$ (129)	Tetragonal	$\tau_{\text{FL}} \mathbf{J} \times \hat{\mathbf{z}}$	$\tau_o \mathbf{m} \times (\mathbf{J} \times \hat{\mathbf{z}}) + \tau_z m_z \mathbf{J}$
CrI_2	$\text{P}\bar{4}\text{m}2$ (115)	Orthorhombic	$\begin{pmatrix} 0 & \tau_{\text{FL}}^{xy} \\ \tau_{\text{FL}}^{yx} & 0 \end{pmatrix} \mathbf{J}$	$(\mathbf{M} \cdot \hat{\mathbf{z}}) \begin{pmatrix} \tau_z^x & 0 \\ 0 & \tau_z^y \end{pmatrix} \mathbf{J} + \mathbf{M} \cdot \begin{pmatrix} \tau_o^x & 0 \\ 0 & \tau_o^y \end{pmatrix} \mathbf{J} \hat{\mathbf{z}}$
CrSBr	Pnmm (59)	Orthorhombic	$\begin{pmatrix} 0 & \tau_{\text{FL}}^{xy} \\ \tau_{\text{FL}}^{yx} & 0 \end{pmatrix} \mathbf{J}$	$(\mathbf{M} \cdot \hat{\mathbf{z}}) \begin{pmatrix} \tau_z^x & 0 \\ 0 & \tau_z^y \end{pmatrix} \mathbf{J} + \mathbf{M} \cdot \begin{pmatrix} \tau_o^x & 0 \\ 0 & \tau_o^y \end{pmatrix} \mathbf{J} \hat{\mathbf{z}}$
NiPS_3	$\text{C}2/\text{m}$ (12)	Monoclinic	$\begin{pmatrix} 0 & \tau_{\text{FL}}^{xy} & 0 \\ \tau_{\text{FL}}^{yx} & 0 & 0 \\ 0 & \tau_{\text{FL}}^{yz} & 0 \end{pmatrix} \mathbf{J}$	$\begin{pmatrix} M_x \tau_o^{xx} + M_z \tau_o^{zx} & M_y \tau_o^{yx} & 0 \\ M_y \tau_o^{xy} & M_x \tau_o^{xy} + M_z \tau_o^{zy} & 0 \\ M_z \tau_o^{xz} & 0 & 0 \end{pmatrix} \mathbf{J} + \mathbf{M} \cdot \begin{pmatrix} \tau_o^{xx} & 0 & 0 \\ 0 & \tau_o^{yy} & 0 \\ 0 & 0 & 0 \end{pmatrix} \mathbf{J} \hat{\mathbf{z}}$

Lower symmetrical systems, such as those with two-fold rotational symmetry, possess a structural anisotropy between two orthogonal crystalline directions. Therefore, the most general relation between the current and the torques should appear in the form of a 2×2 matrix, which is indeed the case reported in Table 1,

$$\mathbf{T}_{\text{FL}}^{\text{ani}} = \mathbf{m} \times \begin{pmatrix} 0 & \tau_{\text{FL}}^{yx} \\ \tau_{\text{FL}}^{xy} & 0 \end{pmatrix} \mathbf{J} \quad (5)$$

where τ_{FL}^{yx} and τ_{FL}^{xy} two independent parameters. Since this field is invariant against two-fold rotation, it is fundamentally different to that in Eq. (4). For example, for $\tau_{\text{FL}}^{xy} = 0$, the field will vanish if the current is applied in the y direction, which will not happen for systems with three-fold symmetry. Therefore, it is evident here that the existence of two-parameters will change the effective field strength with the current direction enabling a general case of anisotropic torques.

Anisotropy-like spin-orbit torques and low symmetry cases

Other systems such as Fe_3GeTe_2 do not have inversion symmetry due to the lack of two orthogonal mirror planes. In these systems, the spin-orbit interaction allows for a SOT that behaves similarly to the one produced by a magnetic anisotropy, which is the reason we call it an anisotropy-like SOT. The axis of the anisotropy is defined by the remaining mirror axis and the direction of the current. It is similar to a magnetic anisotropy in the sense that one can associate the following energy density¹⁵⁴

$$E_{\text{AL}} = J_x m_x m_y - \frac{1}{2} J_y (m_x^2 - m_y^2), \quad (6)$$

which is quadratic in $\mathbf{m}$, and it produces the effective torque through its gradient with respect to the magnetization $\mathbf{T}_{\text{AL}} = -\tau_{\text{AL}} \nabla_{\mathbf{m}} E_{\text{AL}}$.

Certain systems such as monoclinic crystals with just one mirror axis and a two-fold rotation, facilitate the formation of torques possessing components from effective magnetic field proportional to $\mathbf{J} \times \hat{\mathbf{y}}$ and $\mathbf{J} \times \hat{\mathbf{x}}$, which are favorable conditions for switching magnets with an out-of-plane perpendicular magnetic anisotropy.

Measurement symmetry with the predicted $\mathbf{H}_{\text{SOC}}$

Experimentally, we are able to detect these predicted torques via low-frequency second-harmonic measurements¹⁷¹ and current-induced FMR experiments^{131,172}. In both detection schemes, we can consider the current-induced effective fields that tilt the static magnetization direction by a free energy change of $\mathbf{M} \cdot \mathbf{H}_{\text{SOC}}$, or that drive magnetic precession with a magnetic torque of $\mathbf{M} \times \mathbf{H}_{\text{SOC}}$. We can substitute the vector expressions of $\mathbf{H}_{\text{SOC}}$ fields in Table 1 to predict the torque symmetry for future magnetization-control experiments. For example, by using $\mathbf{J} \times \hat{\mathbf{z}}$ as $\mathbf{H}_{\text{SOC}}$, we have the torque expression of $\mathbf{M} \times (\mathbf{J} \times \hat{\mathbf{z}})$ for spin dynamics excitation. Assuming that a driving current is flowing along the x direction, i.e. $\mathbf{J} \parallel \hat{\mathbf{x}}$, the torque expression reduces into $\mathbf{M} \times \hat{\mathbf{y}}$. This suggests that FMR amplitudes excited by this $\mathbf{H}_{\text{SOC}}$ display $\cos\phi$ dependence when the magnetization is rotated in-plane with ϕ defined by the angle between $\mathbf{J}$ and $\mathbf{M}$. In general, measured FMR voltages also have $\sin 2\phi$ dependence due to a time-dependent change of resistance arising from anisotropic magnetoresistance and magnetic precession¹⁷³. As a result, we expect $\mathbf{H}_{\text{SOC}} (\propto \mathbf{J} \times \hat{\mathbf{z}})$ produces $\sin 2\phi \cos\phi$ dependence when we perform FMR voltage measurements for in-plane angle rotation of $\mathbf{M}$. We can apply the same vector analysis for other effective fields for FMR experiments as well as the low-frequency second-harmonic experiments.

Strength of the torques

An important point for comparison with experimental data is how to obtain theoretically the strength of the SOT parameters. In 2D materials, the SOT originates solely from the current-induced spin density in the material. Therefore, in the limit of small currents, the best approach to compute these parameters is via the Kubo formula within the linear-response approximation¹⁷⁴. This approach requires knowledge of the magnetization-dependent Hamiltonian $H(\mathbf{m})$ and an effective way to compute its associated retarded Green's functions $G^+(H, E)$. For non-interacting electrons, the spin-orbit torque is determined by the following expression

$$\mathbf{T}^\alpha(\mu, T) = -\frac{2\pi\Delta_{\text{ex}}\mathbf{J}_\alpha}{\sigma_{\alpha\alpha}} \int dE f(E - \mu, T) \text{ImTr} \left[\text{Im}G^+ \hat{\mathbf{J}}_\alpha \frac{dG^+}{dE} (\hat{\boldsymbol{\sigma}} \times \mathbf{m}) \right], \quad (7)$$

where Im Tr represents the imaginary part of the trace of the operators in the brackets, $f(E - \mu, T)$ the Fermi-Dirac distribution for a given chemical potential μ and temperature T , $\hat{\mathbf{J}}$ the current-density operator and $\mathbf{J}$ its macroscopic average, $\sigma_{\alpha\alpha}$ the conductivity, and $\hat{\boldsymbol{\sigma}}$ the Pauli matrices operator. This approach gives access to the SOTs by making no assumptions on their dependence against magnetization. Furthermore, Eq. (7) can be further sliced into two contributions, those resulting from scattering mechanisms of electrons at the Fermi level (proportional to the conductivity), and those arising from a Fermi-sea contribution, which in principle can provide both dissipative and non-dissipative SOTs, and are primarily scattering independent contributions³⁵. The existence of a SOT proportional to scattering times also evidences that disorder plays a major role to determine the system's behavior. In simple systems, the Kubo formula can be evaluated following a semi-analytic solutions based on perturbation theory^{175,176}, where the disorder is usually described by point impurities. For low concentration of impurities and when the single scattering approximation is valid, the Kubo formula reduces to the Boltzmann equation, which offers a simple way to estimate the behavior of the torque parameters. However, a quantitative determination for a generic disorder or a structurally complex system is only attainable through linear scaling quantum transport methodologies¹⁷⁴, that enable reaching the required size scales to study the torques in the diffusive and localized regimes. For ferromagnetic materials, the magnetization can be instead replaced by the exchange-correlation field, facilitating the use of the Kubo formula to determine the torques from first principles simulations^{177,178}.

Beyond the Kubo formula, nonadiabatic time-dependent evolution in realistic systems is also a great challenge, although there are some recent efforts along this direction¹⁷⁹.

The Kubo formula provides a method to compute the torque parameters in units of a torque density (eV/m^3). However, since these parameters depend directly on the driven current density, a current-to-torque conversion efficiency is usually defined. Such an efficiency definition is quite straightforward in systems where the torque is mostly due to the spin Hall effect, so it can be connected to the spin Hall angle which is given by the ratio between the spin and charge conductivities³⁵. Following similar considerations, the torque conductivity σ_τ can be defined as the angular momentum absorbed by the magnet per second per unit interface area per unit applied electric field¹⁵⁶, from which the torque efficiency χ_α^X is derived as:

$$\chi_\alpha^X \equiv \frac{2e}{\hbar} \frac{M_s t}{\gamma} \frac{\tau_X}{J_\alpha}, \quad (8)$$

where M_s the saturation magnetization, X the torque label (FL, DL, etc), t the thickness of the magnet, which is taken as the Van der Waal gap in 2D materials. The former expression defines an efficiency for different kind of torques

classified according to their symmetries, but there is an alternative dimensionless expression which describes the efficiency according to the torque's directions expressed as¹³⁹

$$\chi_{\alpha\beta} \equiv \frac{e\Delta_{\text{ext}}}{\hbar} \frac{T_{\beta}}{J_{\alpha}}. \quad (9)$$

Both expressions allow to quantify a torque efficiency due to a source of spins independently of geometric factors. For a Cobalt/Platinum system $\chi_{\alpha} \approx 0.0075$ for a $t \approx 0.85$ nm¹⁸⁰, while in experiments considering TMD/Py heterostructures the efficiency only reaches values between $10^{-5} - 10^{-3}$ ¹⁶². Another important parameter is given by $\chi_{\alpha}^{\text{X}}/\chi_{\alpha}^{\text{FL}} = \tau_{\text{X}}/\tau_{\text{FL}}$ which measures the relative value of all torques against the dissipative field-like contribution and generally ranges from 1-10. Notice that in 2D materials $M_s t$ is actually the magnetization per unit area or sheet magnetization¹⁰⁷. From all this discussion, it becomes clear that a lot of work remains to be done to obtain realistic and quantitative information about spin torque efficiency in vdW heterostructures combining 2D magnets with strong SOC materials. Beyond well-controlled ab-initio simulations and the elaboration of properly designed SOC-dependent tight-binding models, the use of computational methodologies that can numerically solve the Kubo formula Eq. (7) for large scale models is a grand challenge, particularly when disorder effects affecting the spin physics demand non-perturbative treatments. Brute-force simulations using linear scaling methods demand further developments and implementations, as well as comparison of their predictions with future experiments.

As a final remark, the spin-Hall effect³⁵ should exist in a variety of vdW material systems, both magnetic and non-magnetic, except in the single monolayer limit. In order to exploit such spin-charge phenomena, vdW heterostructures are usually fabricated with one magnetic layer in direct contact with a spin current source, so as to have a close and efficient magnetization manipulation. The symmetry of spin-Hall effect is well-established and the spin-current source layer could be also magnetic, in which case additional magnetization-dependent anomalous components¹⁸¹⁻¹⁸³ might appear.

Future Outlook

The present review has put in perspective the formidable playground and plethora of possibilities to create new hybrid materials with emerging spin-dependent interface properties. Among the vast zoo of novel structures, theory and modelling based on symmetry features of interfaces bring a pathfinder to find the most suitable conditions for best controlling spin transport through spin-orbit or exchange coupling fields. One most urgent quest is the search for the upper limit of the achievable spin-orbit torque efficiency (at RT and with scalable materials), which could ultimately enable low-energy and fast switching of the material magnetization in the frame of SOT memory technologies^{29,34,120,184}. Such is a challenging task which will require proper combination of theoretical and experimental research within a large community of material grower, advanced characterization and simulation tools, as well as optimized device fabrication approaches. The search for materials with high Curie or Néel temperatures is also a key aspect of this research, and efforts to guide experiments such as the recently studied monolayer Fe₃P with predicted T_C of 420 K¹⁸⁵ should be followed. In this regard, the exchange coupling strength by controlling molecular orbital alignments in 2D magnetic semiconductors⁵³ could be an interesting approach where CrWI₆ and CrWGe₂Te₆ are predicted to display ferromagnetic order above RT via the purely super-exchange interaction. Furthermore, high-throughput computation screening using an existing materials database¹⁸⁶ and other material informatics methods could be an effective approach to guide us towards discoveries of novel high- T_C vdW magnetic materials. Here we have focused our attention to the more familiar ferromagnetic cases, but this approach is valid for predicting torques in non-magnetic vdW systems by using the same space group analysis as well as for novel types of torques in antiferromagnetic systems, such as the Néel SOT¹⁸⁷ and other antiferromagnetic specific SOC effects¹⁸⁸ that are ripe for exploration in these exciting 2D magnetic materials class. Looking further for more exotic 2D vdW materials with order parameters, multiferroics is predicted in a certain class of 2D materials¹⁸⁹ such as in a doped CrBr₃ monolayer¹⁹⁰, in a bilayer of halogen-intercalated phosphorene¹⁹¹ as well as in a monolayer ReWCl₆ with magnetic transitions induced by electric field¹⁹². Some vdW magnetic materials, such as MnBi₂Te₄¹⁹³⁻¹⁹⁵, host topological electronic states and offer unique topologically-protected transport phenomena¹⁹⁶. We anticipate that this research domain will be rapidly expanding to explore the topological transport as well as its use in spintronic applications¹⁹⁷. Besides, nontrivial spin transport physics is also expected to develop at complex interfaces of vdW heterostructures, including the generation of skyrmions¹⁹⁸⁻²⁰⁰, the excitation and detection of magnons at the 2D limit²⁰¹⁻²⁰³ and their interaction with other spin-dependent characteristics of interfaced materials, as well as a wealth of proximity effects which could reveal unprecedented manifestations of entanglement and many-body physics, as recently discussed in the context of topological superconductivity or unconventional ferroelectricity in

magic-angle twisted bi/trilayer graphene and related moiré superlattices²⁰⁴. In conclusion, there is a lot of rooms for further exploration of spin transport at the frontier of layered magnetism and SOC materials, and likely many exciting discoveries waiting for emerging to the surface.

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Competing interests

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