### **REVIEW SUMMARY**

#### MAGNETISM

# **Two-dimensional magnetic crystals and emergent heterostructure devices**

Cheng Gong and Xiang Zhang\*

BACKGROUND: The electron can be considered as a tiny magnet, with two opposite poles defining its magnetic field associated with the spin and orbital motion. When such minuscule magnets are collectively aligned as a result of the inherent coupling, ferromagnetism emerges. However, ferromagnetism had long been believed to hardly survive in two-dimensional (2D) systems because of the enhanced thermal fluctuations revealed by the Mermin-Wagner theorem. The recent discovery of 2D magnetic crystals showed that magnetic anisotropy could stabilize the long-range magnetic order by opening up an excitation gap to resist the thermal agitation. Two-dimensional magnetic crystals constitute ideal platforms to experimentally access the fundamental physics of magnetism in reduced dimensions. In contrast to the traditional magnetic thin films, 2D materials largely decouple from the substrates. allow electrical control, are mechanically flexible, and are open to chemical functionalization. These attributes make 2D magnets accessible, engineerable, and integrable into emergent heterostructures for previously unachieved properties and applications such as atomically thin magneto-optical and magnetoelectric devices for ultracompact spintronics, on-chip optical communications, and quantum computing.

**ADVANCES:** Magnetism has been explored in 2D materials for more than a decade. Magnetic moments have been created through defect engineering based on vacancies, adatoms, boundaries, and edges; band structure engineering, assisted by density functional theory calculations, has raised possibilities of 2D magnetism in, for instance, gated bilayer graphene and doped GaSe; the proximity effect has been applied to imprint spin polarization in 2D materials from magnetic substrates. However, these prior efforts centered on extrinsically induced magnetic response.

In early 2017, the first observations of longrange magnetic order in pristine 2D crystals were reported in Cr<sub>2</sub>Ge<sub>2</sub>Te<sub>6</sub> and CrI<sub>3</sub>. Both are magnetic insulators, yet with distinct magnetic properties. In contrast, 2D Fe3GeTe2 was recently proven to be a magnetic conductor. Itinerant magnets and magnetic insulators possess diverse application perspectives. Molecular beam epitaxial growth of 2D magnets has been reported for Fe<sub>3</sub>GeTe<sub>2</sub>, VSe<sub>2</sub>, MnSe<sub>x</sub>, and Cr<sub>2</sub>Ge<sub>2</sub>Te<sub>6</sub>. The typical Curie temperatures of 2D magnets are much lower than those of their 3D counterparts. However, this does not fundamentally exclude the possibility of hightemperature 2D magnets. Efforts toward this goal have shown promise.





When van der Waals (vdW) magnets contact nonmagnetic materials, time reversal asymmetry could be introduced in the original nonmagnets, likely leading to, for example, valley polarization in transition metal dichalcogenides or quantum anomalous Hall states in topological insulators. However, it should be noted that 2D magnets' properties are susceptible to the contacting materials. Stacking vdW magnets with dissimilar materials could enrich the landscape of emergent phenomena by causing, for example, heterostructure multiferroicity, unconventional superconductivity, and the quantum anomalous Hall effect.

Two-dimensional spintronic and magnonic devices have begun to emerge. Spin-orbit torque has been generated while spin-polarized current is injected from 2D materials (e.g., WTe<sub>2</sub>) into magnetic substrates; conversely, a spin wave has

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been pumped from magnetic substrates into 2D materials for spin-charge conversion. Magnetic tunnel junctions with 2D magnets (e.g., CrI<sub>3</sub>) as tunneling barriers exhibit giant tun-

neling magnetoresistance at low temperatures. New concepts of spin field-effect transistors based on 2D magnets have been reported as well.

**OUTLOOK:** Most currently available 2D magnets rely on mechanical exfoliation and only work at low temperatures. The wafer-scale synthesis of 2D magnets that operate above room temperature is a prerequisite for the development of practical applications. In the longer run, the monolithic integration of such 2D magnets with other functional materials is crucial for practical scalability. Spintronic devices require efficient electrical modulation of 2D magnets, long-distance transport of spins or spin waves, and efficient tunneling and injection of spins at various junctions. The practical

development of low-power spintronic devices needs to be compatible with the existing complementary metal-oxide semiconductor technology (e.g., impedance match and affordable power supply). Furthermore, the exotic spin textures, quantum phases, and quasiparticles in 2D magnetic crystals and hetero-interfaces could lead to new ways of computation and communication. We envision that successive breakthroughs in 2D magnets could usher in a new era of information technologies with exciting applications in computing, sensing, and data storage.

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#### MAGNETISM

# **Two-dimensional magnetic crystals and emergent heterostructure devices**

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Magnetism, originating from the moving charges and spin of elementary particles, has revolutionized important technologies such as data storage and biomedical imaging, and continues to bring forth new phenomena in emergent materials and reduced dimensions. The recently discovered two-dimensional (2D) magnetic van der Waals crystals provide ideal platforms for understanding 2D magnetism, the control of which has been fueling opportunities for atomically thin, flexible magneto-optic and magnetoelectric devices (such as magnetoresistive memories and spin field-effect transistors). The seamless integration of 2D magnets with dissimilar electronic and photonic materials opens up exciting possibilities for unprecedented properties and functionalities. We review the progress in this area and identify the possible directions for device applications, which may lead to advances in spintronics, sensors, and computing.

or centuries, humans had been puzzled about the magic attraction of lodestones to iron, and perhaps even more about the fascinating ability of birds, fish, and insects to navigate between destinations of thousands of miles apart. In early times before the development of electromagnetism and quantum mechanics, it was hard to imagine that these intriguing phenomena may share a common magnetic origin. Magnetism is fundamentally rooted in the moving charges and spin of elementary particles; hence, it is as ubiquitous as the electron itself. It has found broad applications in living organisms as well as in energy harvesting, data storage, and medical diagnosis. When the infinitesimal "electron magnets" are spontaneously aligned, the magnetic order constitutes a fundamental phase of matter, giving rise to a host of functional devices including electric generators and motors, magnetoresistive memories, and optical isolators. The ability to knit such magnetic order in atomically thin flatlands would foster vast opportunities for integrated, flexible, and biocompatible devices; however, such two-dimensional (2D) magnets are not easily attainable because of a fundamental hindrance.

Understanding the fundamental difference between 2D and 3D magnetism is instructive. The driving force underlying the ordering of electrons' spin magnetic moments in an (anti-)ferromagnet is the "exchange interaction," which was initially dubbed as a "molecular field" by Pierre Weiss in 1907 (1) and was understood to be of quantum mechanical origin in 1926 (2, 3) (Fig. 1A). The effect is a coulombic interaction under the Pauli exclusion principle relating to the electrons' anti-

symmetric wave function. The exchange interaction has been used to estimate the Curie temperatures of 3D ferromagnets, based on the argument that the short-range exchange interaction needs to be overcome by thermal energy to randomize the magnetic moments. Nonetheless, the mean-field picture suitable for 3D systems does not work for the length scale of 2D systems, in which the dimensionality effect comes into play (4). Magnon (i.e., quanta of spin wave) dispersion in 2D systems is reduced with respect to that in the 3D counterparts, corresponding to an abrupt onset of magnon density of states (DOS) in 2D systems and thus an ease of thermal agitations (5, 6). For 2D systems without magnetic anisotropy (Fig. 1B), the spin wave excitation gap diminishes (Fig. 1C). Together with the diverging Bose-Einstein statistics of magnons at zero energy, any nonzero temperatures cause massive magnon excitations and the spin ordering to collapse, as predicted by Mermin and Wagner (4). However, for 2D systems with a uniaxial magnetic anisotropy, a magnon excitation gap opens up and resists the thermal agitations (Fig. 1, D and E), which then lifts the Mermin-Wagner restriction and results in finite Curie temperatures. Meanwhile, the exchange interaction together with the dimensionality dictates the magnon band width and profiles (6). Therefore, the synergy of these factors, as well as the inter(quasi-) particle scattering, which potentially renormalizes the magnon spectrum, determines the upper bound temperature (i.e., Curie temperature) below which a 2D ferromagnet can be found.

The past a few decades have witnessed the role of epitaxial thin films and superlattices as testing grounds for the experimental exploration of 2D magnetic properties and spin entity (i.e., spin-polarized electrons or spin waves) propagations. Seminal phenomena such as giant magnetoresistance (7, 8), the dimensionality ef-

fect (9-11), and oscillating exchange coupling (12) were discovered. But it has been a long-standing challenge to access the intrinsic magnetic properties of ultrathin films as a pure quantum confinement effect of their 3D counterparts; these traditional thin films suffer from various perturbations such as interfacial hybridization, electronic redistribution, reduced coordination with band narrowing, atomic interdiffusion, strain, crystalline reconstruction, finite-size islands (typically tens of nanometers), and irregular shapes (13, 14). Therefore, the properties of such ultrathin films are difficult to precisely control and replicate.

In stark contrast, the recently discovered 2D magnetic atomic crystals (6, 15) provide unique opportunities for both fundamental physics and technological advances. Such magnetically ordered atomic crystals enable unprecedented experimental access to the ground states, fundamental excitations, and magnon dynamics of single-crystalline 2D magnets. Because such 2D magnetic crystals are susceptible to a long list of external stimuli including mechanical deformation, electrostatic doping, light incidence, chemical decoration, and dielectric environment, there is enormous room to engineer 2D magnets for desired properties; the sensitive responses of 2D magnets allow the development of miniaturized, lightweight, flexible, and biocompatible devices based on magnetoresistive, magnetoelectric, magnetostrictive, magneto-optical, and magnetobiological effects. Furthermore, the past decade has witnessed the increasingly skillful handling of individual 2D layers, which could facilitate the unprecedented fabrication of multilayer "designer magnets"; a notable outcome could be the giant cross-layer tunneling magnetoresistance by designing the interlayer magnetic coupling. In heterostructures with electronic and photonic materials, the seamless integration and intricate interplay of distinct physical properties could give rise to emergent interfacial phenomena such as heterostructure multiferroicity, unconventional superconductivity, and the quantum anomalous Hall effect. It is reasonable to envision that a vast range of previously unachieved properties will be discovered in 2D magnetic crystals, derivatives, and heterointerfaces, which could be transformed into a host of applications such as low-power spintronics, on-chip optical communications, and quantum computing.

## Induced magnetic response in nonmagnetic 2D materials

Since the advent of graphene, attempts to create ferromagnetism in nonmagnetic 2D materials have continued apace. One of the mainstream strategies is through introducing vacancies or adding adatoms such as hydrogen and fluorine (*16–22*) (Fig. 2, A and B). Such defect engineering produces local magnetic moments from unpaired electrons, which, for example, could be further correlated through conduction electrons in graphene (itinerant  $\pi$ -magnetism). However, attempts to order these moments in a "long-range"

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Fig. 1. Fundamental physical parameters and spin wave excitations in ferromagnets of different dimensionalities. (A and B) In a collinear magnet, exchange interaction and magnetic anisotropy are fundamental parameters. Exchange interaction arises from electrons' antisymmetric wave function and is governed by coulombic interaction under the Pauli exclusion principle. Exchange interaction between spins can be directly established (red dashed line 1) or indirectly mediated by conduction electrons (green ball with dashed lines labeled 2) or intermediate anions (orange ball with dashed lines labeled 3) such as O<sup>2-</sup>. While spins are aligned, there is usually a preferred orientation, which means magnetic anisotropy. Magnetic anisotropy has a variety of sources such as magnetocrystalline anisotropy, shape anisotropy, and stress anisotropy. (C to F) In a 2D isotropic Heisenberg ferromagnet, there will be massive excitations of magnons at nonzero temperatures because of the absence of a spin wave excitation gap, the abrupt onset of magnon density of states (DOS), and the diverging Bose-Einstein statistics at zero energy; the result is collapse of long-range magnetic order. The presence of uniaxial magnetic anisotropy (UMA) opens up the spin wave excitation gap to resist the thermal agitations of magnons, leading to the finite Curie temperature. As the system evolves from 2D to 3D, the magnon DOS spectrum changes from a step function to a gradually increasing function with zero DOS at the threshold of excitation. Therefore, in 3D systems, UMA (related to the spin wave excitation gap) is not a prerequisite for the presence of finitetemperature long-range magnetic order.

pattern posed overwhelming challenges in material preparation. One report (23) disagreed with the feasibility of these approaches to realizing long-range ferromagnetic order, because the authors only observed the paramagnetic response in graphene with fluorine adatoms or vacancies at liquid helium temperatures, and observed a maximum response of about 1 magnetic moment per 1000 atoms in all tested samples. Despite a number of prior reports on the observation of ferromagnetism in originally nonmagnetic 2D materials, no consensus has been reached (24, 25). The primary debates center on the source of the detected magnetic signals: A chunk of material in a superconducting quantum interference device (SQUID) measurement chamber is typically hundreds of micrometers thick, approximately six orders of magnitude thicker than an atomic sheet. Hence, true signals from the 2D sheet may be overshadowed by ferromagnetic impurities in substrates, even if the impurity concentrations are as low as 10 ppm.

Flat band ferromagnetism has been proposed to be realized via extended defects such as zigzag edges of graphene nanoribbons or grain boundaries of 2D materials (26-28). Such defects cause less-dispersed electronic bands that satisfy the huge density of states in a narrow energy scope. leading to the Stoner instability toward a ferromagnetic phase. However, these chemically reactive defects are vulnerable to passivation by foreign species; also, long-range 1D ferromagnetic order cannot exist in theory (26). Although the strict "long-range" (i.e., infinitely long) ferromagnetic order is not allowed in 1D systems, it is possible to make magnetically ordered chains with finite length and width, so that the finitesize ferromagnets (29-31) behave as spin blocks of superparamagnets with reasonably long spin flipping time, which can provide a practical path toward nanoscale spintronic devices (26, 32, 33).

Band structure engineering may represent a vital route to create 2D ferromagnetism from originally nonmagnetic 2D materials, without the assistance of structural imperfections. For instance, such band ferromagnetism was predicted to exist in electrically biased bilayer graphene (34, 35) (Fig. 2F) and doped GaSe (36, 37). In bilayer graphene biased by electric fields perpendicular to the basal plane, the different electrostatic potentials experienced by the two layers

induce a gap opening and a Mexican-hat dispersion at low energy. Such Mexican-hat bands are often concomitant with the itinerant ferromagnetism. Nonetheless, the anticipated ferromagnetic phase has not been experimentally observed thus far, likely because of the limited total magnetic moments or low Curie temperatures. The predicted magnetism in doped GaSe is based on the similar strategy of Mexican-hat band dispersion. However, ab initio calculations show the necessity of hole-doped GaSe up to 10<sup>13</sup> to 10<sup>14</sup> carriers/cm<sup>2</sup> to reach the van Hove singularity. Given the large quasiparticle band gap, 3.7 eV (36), it would be challenging to realize this high level of hole doping by conventional electrostatic schemes. Similar spontaneous valley polarization was observed in 2D electronic systems, such as the inversion layer in silicon in the 1970s (38). More recently, giant paramagnetism in monolayer  $MoS_2$  was reported (39), with an unconfirmed yet possible spontaneous ferromagnetism proposed.

The magnetic proximity effect is a scheme to make nonmagnetic 2D materials magnetic by borrowing properties from adjacent magnetic materials. A graphene sheet, which was transferred on yttrium ion garnet (YIG) (40) (Fig. 2D) or upon which EuS was deposited (41) (Fig. 2E), exhibits an anomalous Hall effect. The sole evidence from an anomalous Hall signal does not suffice to conclude that a ferromagnetic phase in 2D materials exists, because other effects such as spin-dependent interfacial scattering or ferromagnetic impurities may result in similar observations (42). However, the quantification of the 14-T interfacial exchange field (41) and the observation of the quantum Hall effect at a much lower external magnetic field shed light on the presence of interfacial exchange fields. More direct evidence of the interfacial exchange field was subsequently obtained on the basis of spin current transport in lateral graphene spin valves on YIG (43, 44). Interestingly, a spin dephasing mechanism due to the temporal and spatial fluctuation of interfacial exchange fields was revealed (44), which highlights from a new angle the critical role of interfacial quality in the spintronic transport properties of proximity systems.

#### Magnetism in pristine 2D materials

The first two reported 2D magnetic atomic crystals are chromium compounds:  $Cr_2Ge_2Te_6$  (6) (Fig. 3, A to C) and  $CrI_3$  (15) (Fig. 3, D and E).  $Cr_2Ge_2Te_6$  is a 2D Heisenberg ferromagnet with small magnetic anisotropy (i.e., collectively aligned spin moments can be oriented toward all directions with small energy difference), whereas  $CrI_3$  is probably a 2D Ising A-type antiferromagnet (i.e., spin moments oriented normal to the basal plane, intralayer ferromagnetism, and interlayer antiferromagnetism).

The slight distortion of the Cr-Te<sub>6</sub> octahedral cage, together with spin-orbit coupling on Cr ions, leads to a small out-of-plane magnetocrystalline anisotropy in Cr<sub>2</sub>Ge<sub>2</sub>Te<sub>6</sub>. The nearly isotropic Heisenberg 2D ferromagnet mimics the ideal Mermin-Wagner condition, on the basis of which the external magnetic field has an



**Fig. 2. Schemes to induce magnetism in nonmagnetic 2D materials.** Point defects such as vacancies and adatoms in 2D materials are accompanied by defect states and local magnetic moments. (**A**) STM topography of graphene with carbon vacancies induced by Ar<sup>+</sup> ion irradiation (17). Scale bar, 5 nm. (**B**) Schematic of local magnetic moments in graphene decorated by an individual hydrogen adatom (small white ball at center) (18). The same spin-polarized state extends a few nanometers in carbon sites of the same sublattice, but the opposite spin-polarized state occupies the other carbon sublattice. (**C**) Magnetization versus magnetic field parallel to the fluorinated graphene planes (23). Dots are experimental data; solid

lines are fitting curves based on Brillouin function. No trace of ferromagnetism was found in both fluorinated graphene and defective graphene with vacancies at liquid helium temperatures. (**D**) Schematic of a graphene field-effect transistor fabricated on YIG, a magnetic insulator (40). (**E**) Schematic of a graphene field-effect transistor covered by a deposited thin film of EuS, a magnetic insulator (41). Nonmagnetic 2D materials can be made magnetic by physically contacting magnetic materials through proximity effect. (**F**) Calculated Mexican-hat band dispersion in electrically biased Bernal stacked bilayer graphene (34). The diverging electronic DOS at the edge of the Mexican hat potentially results in ferromagnetic Stoner instability.

unprecedented effect on the observed transition temperatures of 2D  $Cr_2Ge_2Te_6$ . Despite possessing van der Waals (vdW) spacing, interlayer magnetic coupling is appreciable, as evidenced by the strong dimensionality effect in transition temperatures of  $Cr_2Ge_2Te_6$  of different thickness. An isostructural compound,  $Cr_2Si_2Te_6$  (45), has a larger easy-axis magnetic anisotropy and a lower bulk ferromagnetic phase transition temperature  $T_C$  at 33 K. The Curie temperature of the hypothetical isostructure  $Cr_2Sn_2Te_6$  is theoretically predicted to be higher (46), but the successful synthesis of this crystal has not yet been reported.

The sizable magnetic anisotropy in  $CrI_3$  was suggested to arise from the exchange anisotropy due to the spin-orbit interaction of iodine species that mediate the superexchange between Cr ions (47). Graphite-encapsulated few-layer  $CrI_3$  shows interesting layer-contrasting magnetic properties and was suggested to be an A-type antiferromagnet. Further investigations are needed to identify the origin of the exotic properties of 2D  $CrI_3$  that are contrary to those of its bulk counterpart [bulk  $CrI_3$  is a ferromagnet (48)], although the altered interlayer registry in few-layer  $CrI_3$ was invoked as a tentative explanation (49–52). Possible extrinsic causes relate to graphite capping, partial degradation (CrI<sub>3</sub> is extremely unstable to the moisture and light) (53), and unintentional doping and strain (especially for cases where CrI<sub>3</sub> samples were exfoliated on polymer and later transferred). The isostructural magnet CrBr<sub>3</sub> in bulk is an easy-axis ferromagnet with  $T_{\rm C}$  = 33 K, and CrCl<sub>3</sub> in bulk is an easy-plane A-type antiferromagnet. Magnetic anisotropies and phase transition temperatures of bulk CrCl<sub>(3-x)</sub>Br<sub>x</sub> alloys are tunable by changing stoichiometries (54).

 $Fe_3GeTe_2$  is a metallic ferromagnet (55-60) (Fig. 3, F and G). In each layer, three of the quintuple sublayers are iron; the top and bottom sublayers are equivalent and the central one differs. The crystallographic environments of the iron atoms are asymmetric along and normal to the basal plane, leading to the sizable magnetocrystalline anisotropy. The tunable Curie temperatures and coercivities can be realized by varying the iron concentrations. Interestingly, in this 3d electronic system, the coexistence of itinerant ferromagnetism and Kondo lattice was evidenced (61), suggesting the presence of heavy fermions and periodically seated local moments. This constitutes an intriguing 2D material platform, in which itinerant electrons and local magnetic moments coexist and interplay, possibly leading to a plethora of emergent phases and phenomena. Furthermore, for bulk Fe<sub>3</sub>GeTe<sub>2</sub>, evidence from magnetization characterization, electrical transport (62-64), scanning tunneling microscopy (STM) (65), and magnetic force microscopy (63) points to a different magnetic configuration emerging at even lower temperatures (~50 K lower than the Curie temperature of 220 to 230 K). The stripe domain phase in bulk Fe<sub>3</sub>GeTe<sub>2</sub> observed by photoemission electron microscopy (66) indicates the quasi-2D magnetic behaviors, and the thickness-dependent magnetic hysteresis (63, 64) reveals valuable hints that long-range dipolar interaction may play an important role when layers are stacked up to tens of nanometers.

In contrast to ferromagnets, antiferromagnets find limited applications—for example, to stabilize the "fixed layer" in spin valves and magnetic tunnel junctions. Despite the notorious difficulty in using antiferromagnets due to their net vanishing magnetization, antiferromagnets hold promise for high-speed, low-power spintronics because they have magnetic resonance frequencies in the terahertz regime, null stray field for vanishing cross-talk between adjacent bits, and robustness against the external magnetic field perturbation (*67, 68*). Furthermore, antiferromagnets are much more abundant than ferromagnets.



Fig. 3. Representative 2D magnetic crystals. (A to C) Optical image, Kerr image, and dimensionality effect of few-layer  $Cr_2Ge_2Te_6$  exfoliated on SiO<sub>2</sub>/Si (6). Scale bars, 10  $\mu$ m. (D and E) Atomic structure of CrI<sub>3</sub> and thickness-dependent Kerr signal hysteresis loop of graphite-sandwiched 2D CrI<sub>3</sub> (15). In (D), orange arrows represent the ferromagnetically coupled spin magnetic moments. In (E), red and blue vertical arrows represent spin-up and spin-down magnetic moments, respectively. (F and G) Atomic structure of Fe<sub>3</sub>GeTe<sub>2</sub> and thickness-dependent normalized remanent anomalous Hall resistance of 2D Fe<sub>3</sub>GeTe<sub>2</sub> on Al<sub>2</sub>O<sub>3</sub> thin film, which was prepared by thermally evaporating Al in oxygen

Antiferromagnetic vdW materials are diverse in magnetic configurations such as the Ising antiferromagnets FePS<sub>3</sub> and FePSe<sub>3</sub> (69-72), the Heisenberg antiferromagnets MnPS<sub>3</sub> and MnPSe<sub>3</sub> (72, 73), and the XY-type antiferromagnet NiPS<sub>3</sub> (73). Raman spectroscopy could provide nondestructive techniques to indirectly probe the magnetic phase transitions for some antiferromagnets. For example, FePS<sub>3</sub> has a Raman feature peak whose intensity has a sharp upturn when samples are cooled under Néel temperature (74). Methods that can effectively characterize antiferromagnetic order and magnetic excitations in atomically thin micrometersize antiferromagnetic samples would be useful. Reading information from 2D antiferromagnets

steps of transfer and exfoliation (62). (**H** and **I**) STM image of submonolayer VSe<sub>2</sub> grown on HOPG by MBE and magnetic hysteresis of mostly monolayer VSe<sub>2</sub> grown on MoS<sub>2</sub> by MBE (75). Scale bar, 20 nm. (**J** and **K**) Atomic structure of vdW MnSe<sub>2</sub> and out-of-plane magnetic hysteresis of MnSe<sub>x</sub>, one monolayer on average, grown on GaSe by MBE (76). In (J), orange arrows represent the ferromagnetically coupled spin magnetic moments. In (K), red and blue curves represent the half of the hysteresis loop while magnetic field is swept from positive to negative and from negative to positive, respectively.

> would pave the way to taking full advantage of the vdW antiferromagnets for ultrafast low-power spintronics.

> An important consideration for the practical use of 2D magnets is the magnetic phase transition temperature. Substantial work has been devoted to enhancing the Curie temperatures of 2D ferromagnets beyond room temperature, with

a few claims of success thus far. For this ambitious goal, there appears to be no fundamental prohibition as long as the ferromagnetic exchange interaction and uniaxial magnetic anisotropy are strong enough. Yet the practical realization is not that easy. Room-temperature ferromagnetism was reported in single-layer 1T-VSe2 synthesized on both highly oriented pyrolytic graphite (HOPG) and  $MoS_2$  by molecular beam epitaxy (MBE) (75) (Fig. 3, H and I). Two puzzling points remain for this work: the unusually high magnetic moments  $(8000 \text{ emu/cm}^3)$  in single-layer VSe<sub>2</sub> on MoS<sub>2</sub>, and the observed easy-plane anisotropy, which can hardly sustain the long-range ferromagnetic order at room temperature unless a strong uniaxial anisotropy is present within the basal plane. In another contemporary study, the vdW phase of single-layer  $MnSe_x$  was synthesized by MBE (76) (Fig. 3, J and K). Interestingly, such a vdW phase for  $MnSe_x$  only exists in ultrathin layers, but the bulk counterparts prefer rock-salt NaCl or the hexagonal NiAs phase, both of which are antiferromagnetic (77). This observation highlights that 2D materials could exhibit novel structural forms that are not present in bulk, and thus possess unusual physical properties. For MBE samples, the possibly complicated interfaces formed during sample synthesis (e.g., by atomic diffusion, reaction, and alloying) should be carefully examined as the probable causes of the observed magnetic signals. A recent work (78) pointed out the magnetic element in a hot cell as a source of magnetic impurities in MBE synthesis.

In theory, there is not a fundamental restriction to 2D long-range magnetic order at high temperatures, although enhanced thermal fluctuations are always a hindrance. The rule of thumb in designing high-temperature 2D ferromagnets is to strengthen the exchange interaction and uniaxial magnetic anisotropy. Given the extensive prior experience in creating magnetic moments and enhancing spin-orbit coupling in graphene and other nonmagnetic 2D materials, it is safe to envision that thermally robust 2D ferromagnets will be discovered in ever more engineered 2D material systems.

Magnetism as a result of many-body interaction suggests electron correlation effects. Spincharge coupling has been evidenced in NiPS<sub>3</sub> (79), an easy-plane antiferromagnet, showing the presence of electron correlations. Antiferromagnetism and strong electron correlations are reminiscent of unconventional superconductors. Interestingly, a recent experiment showed that FePSe<sub>3</sub>, a vdW antiferromagnet, indeed evolves to be superconducting under hydrostatic pressure above 9 GPa (80). For ferromagnetic  $Cr_2Ge_2Te_6$ , the sharp upturn of electrical resistivity when temperature is lowered to a certain point above  $T_{\rm C}$  suggests the spin-charge coupling (81). Strong electron correlations are implied by the enhanced electronic specific heat of Fe3GeTe2 with respect to that of nonmagnetic isostructural Ni<sub>3</sub>GeTe<sub>2</sub>, and are also supported by the comparison between experimental and calculated Sommerfeld coefficients in  $Fe_3GeTe_2$  (57). Therefore, on the basis of all of this evidence, electron correlations appear generally present in 2D magnetic crystals. Such electron correlations enrich the complex phase diagrams, allow the electrical tuning and compositional engineering to switch between multiple phases, and make heterostructures involving 2D magnets fertile grounds for emergent interfacial phenomena.

#### Control of 2D magnetism

Electric control of low-dimensional magnetic systems (82, 83), through either an electric field or electrostatic doping, changes the electron population, orbital occupation, and possibly electrochemical reactions (84), leading to the modification of exchange parameters and magnetic anisotropies and thus the resultant magnetic properties. Electrical control of recently emerged 2D magnets has been achieved. In bilayer antiferromagnetic CrI<sub>3</sub>, both electric fields and electrostatic doping can affect the spin-flipping magnetic field (85-87). One remarkable phenomenon is the almost complete conversion of the few-layer graphene-encapsulated bilayer CrI<sub>3</sub> from interlayer antiferromagnetism to ferromagnetism by electrostatic doping (87). The Curie temperature of single-layer CrI<sub>3</sub> was modulated between 40 K and 50 K by changing doping levels. Electrical control of few-layer Cr<sub>2</sub>Ge<sub>2</sub>Te<sub>6</sub> via ionic liquid gating shows the modulation of coercivity and saturation field (88). In contrast to magnetic insulators, the itinerant nature of ferromagnetism in Fe<sub>3</sub>GeTe<sub>2</sub>, given the magnetism is mediated through conductive electrons, possibly allows more effective tuning of Curie temperatures by controlling the carrier concentrations within. The hysteresis loop of few-layer Fe3GeTe2 was recently reported in anomalous Hall effect measurements at room temperature (62), while the 2D magnet was electrostatically doped via an ionic liquid. Scanning microscopy may provide further information on whether a long-range ferromagnetic order or local magnetic islands are formed, considering that both structural distortion and electrochemical reaction possibly occur when 2D materials meet ionic liquids. Also, the conversion between antiferromagnetism and ferromagnetism was predicted to occur in both electron- and hole-doped MnPSe<sub>3</sub> above  $3 \times 10^{13}$  to  $4 \times 10^{13}$  carriers/cm<sup>2</sup> (89). Such antiferromagnetism-ferromagnetism conversion may prompt a new means of magnetization switching for logics and memories.

Magnetoelectric multiferroics could enhance the efficiency of electric control of magnetism because of the inherent coupling of magnetic and electric orders. Magnetoelectric multiferroics that simultaneously possess ferromagnetism and ferroelectricity hold great promise in nextgeneration spintronics and microwave magnetoelectric applications. However, in the 2D regime, each type of ferroic order has its own hindrance, such as the enhanced thermal fluctuations for 2D ferromagnetism and the strong depolarization fields for 2D ferroelectricity. Despite the recent success in realizing 2D ferromagnetism and 2D ferroelectricity (*90–94*) in different 2D materials, the simultaneous realization of both orders in one 2D material has not been reached. The theoretically predicted 2D multiferroics are primarily of ferromagnetism and ferroelasticity [e.g., monolayer group IV mono-oxide  $\alpha$ -SnO (95)] and of ferroelectricity and ferroelasticity [e.g., monolayer group IV monochalcogenides such as GeS, GeSe, SnS, and SnSe (96) and monolayer ternary compounds such as GaTeCl (97)]. At the time of writing, very few theoretical predictions have been made on 2D magnetoelectric multiferroics (98, 99). An unusual 2D magnetoelectric multiferroics was predicted on the basis of hyperferroelectric CrN (98). The scarcity of single-phase 2D magnetoelectric multiferroics relates to the longstanding challenge in multiferroics physics: Ferromagnetism prefers partially filled *d*-orbitals, whereas ferroelectricity prefers empty d-orbitals for displacive movement of ions.

Another knob for the effective control of magnetism is pressure or strain. Magnetic properties critically hinge on materials' structural parameters. Thus, spin-lattice coupling and magnetostrictions are common phenomena in magnetic materials. Spin-lattice coupling has been experimentally observed or theoretically predicted in layered magnets such as Cr<sub>2</sub>Si<sub>2</sub>Te<sub>6</sub> (45), Cr<sub>2</sub>Ge<sub>2</sub>Te<sub>6</sub> (100), and Fe<sub>3</sub>GeTe<sub>2</sub> (58). While being cooled into ferromagnetic phase, both the in-plane lattice constant and the interlayer spacing of Cr<sub>2</sub>Si<sub>2</sub>Te<sub>6</sub> undergo the increase (45). For Cr<sub>2</sub>Ge<sub>2</sub>Te<sub>6</sub>, frequencies of a few phonons exhibit upturns while being cooled into a ferromagnetic phase, highlighting the spin-phonon coupling (100). A hydrostatic pressure of 1 GPa can reduce the Curie temperature of the bulk  $Cr_2Ge_2Te_6$  by ~9% (101). Furthermore, hydrostatic pressures above 1 GPa can reorient the spins of the bulk  $Cr_2Ge_2Te_6$  from out-of-plane to in-plane (102). As discussed above, pressurized FePSe3 even underwent a transformation from antiferromagnetic insulator to superconductor. These findings showcase the effectiveness of pressure or strain in engineering 2D magnets.

### Heterostructures and interfacial engineering

High spin and valley polarizations in 2D materials provide tantalizing opportunities for efficient spintronics and valleytronics. Placing 2D electronic or valleytronic materials on magnetic insulators offers an effective methodology to spin-polarize and/or valley-polarize 2D materials. Especially if such magnetic insulators are also vdW materials, the seamless integration and interplay of vdW heterostructures could benefit the interfacial exchange interaction due to atomically sharp interfacial registry. Various material systems in which the proximity effect has been used to spin- or valley-polarize 2D materials include graphene on YIG (40), EuS on graphene (41), WSe2 on EuS (103), and WSe2 on CrI<sub>3</sub> (104).

Time-reversal symmetry breaking in 3D topological insulators can induce a gap opening in the 2D Dirac surface states, possibly giving rise to quantum anomalous Hall states on 1D edges. The conventional way to realize the quantum

anomalous Hall effect (QAHE) is through magnetic dopants, such as Cr-doped Bi<sub>2</sub>Se<sub>3</sub>. However, magnetic impurities scatter the electrons' traveling and suppress the temperature limit for the realization of QAHE. Given the atomically flat vdW ferromagnet, the sharp interfacial registry may effectively magnetize the surfaces of topological insulators without incurring structural perturbations. Such endeavors have been attempted using metal-organic chemical vapor deposition of  $Bi_2Te_3$  on  $Cr_2Ge_2Te_6$  (105) and MBE growth of Cr<sub>2</sub>Ge<sub>2</sub>Te<sub>6</sub> on (Bi,Sb)<sub>2</sub>Te<sub>3</sub> (106). Another surge of interest toward QAHE was triggered by the recent advances in the synthesis of intrinsic magnetic topological insulators such as MnBi<sub>2</sub>Te<sub>4</sub> and MnBi<sub>2</sub>Se<sub>4</sub> (107-112). These layered materials are not only intrinsically magnetic but also topologically nontrivial. Rather than randomly distributing in the Bi sites substitutionally, Mn self-organizes in the center layer of the septuple layers of, for example, Mn<sub>2</sub>Bi<sub>2</sub>Te<sub>4</sub>, which is an A-type antiferromagnetic topological insulator and may host the quantized topological magnetoelectric effect, quantum anomalous Hall state, and intrinsic axion insulator state. Successful synthesis of Mn<sub>2</sub>Bi<sub>2</sub>Te<sub>4</sub> in both thin films and millimetersize bulk single crystals (108) has been reported.

Placing 2D vdW magnets in contact with other materials not only lends the magnetic properties of 2D magnets to others, but can also modify the 2D magnets via interfacial engineering. A remarkable example is the magnetic phase at the EuS-Bi<sub>2</sub>Se<sub>3</sub> interface persisting beyond room temperature (113). Interfacial engineering represents a vital scheme to tailor 2D magnetic properties: It is based on artificial material systems, which means tremendous freedom relative to the design and synthesis of single-phase materials. Moreover, unlike the modulation of magnetic properties through external stimuli such as electrical field, interface engineering does not require external power sources to maintain the modified properties.

A host of factors make possible the interface engineering of 2D magnets, as summarized in Fig. 4:

1) The interface charge transfer changes the electron concentration and orbital occupation in 2D magnets, leading to the property change.

2) The interface dipole or built-in electric field can modify the electronic structure or crystal field of 2D magnets.

The above two factors were proposed to explain the nonzero remanent magnetization in bilayer  $CrI_3$  sandwiched by few-layer graphene [figure 3a of (87)] and the different magnetic properties between graphite-bilayer  $CrI_3$ -BN and BN-bilayer  $CrI_3$ -BN [figure S4 of (85)]. The interface effects in the resultant 2D magnetic properties were also reflected by the distinct coercivities in different regions of the same monolayer  $CrI_3$  flake sandwiched by graphite [figure 2d of (15)].

3) The interfacial orbital hybridization affects the resultant magnetic property of 2D magnets by impinging on the electronic properties and orbital characters of 2D magnets. 4) The morphology and lattice strain of 2D magnets can be modified when 2D magnets interface with other materials, resulting in the change of 2D magnetic properties.

5) Exchange anisotropy and the superexchange mediated by outermost atoms of 2D magnets are susceptible to the contacted materials.

6) The band structure of a 2D magnet may renormalize when interfacing with a material of a similar lattice constant. For example, graphene band properties can be strongly altered by *h*-BN. Band renormalization of 2D magnets can also be caused by the dielectric screening of adjacent materials.

7) The dielectric environments screen the coulombic interaction (note: in nature, exchange interaction is a coulombic interaction). Such dielectric screening of electron-electron interaction has been well studied for mobility enhancement in transistors (114) and has been shown to effectively weaken the excitonic binding energy in 2D materials [the adjacent single-side bilayer

graphene reduces the excitonic binding energy in  $MoSe_2$  by ~100 meV (*115*)].

8) The spin-orbit coupling proximity can play a role when 2D magnets are in contact with heavy elements, given that the magnetocrystalline anisotropy is intrinsically related to the spinorbit coupling. For example, graphene's spin-orbit coupling strength was enhanced three orders of magnitude by contacting WS<sub>2</sub> (116).

## Device applications: 2D spintronics, magnonics, and spin-orbitronics

The magnetic tunnel junction (MTJ) is a fundamental building block for the state-of-the-art spintronic industry (7, 8, 117–123). One of the obvious advantages in all-vdW MTJs is that the uniform barrier thickness facilitates the all-area tunneling. In contrast, tunneling in nonuniform MTJs preferentially occurs through thinner barrier regions, because the tunneling current is an exponential function of the barrier thickness. Such vdW MTJs have been pursued based on



Fig. 4. Interfacial engineering of 2D magnets. Magnetic properties of 2D magnets can be affected by adjacent materials via different mechanisms. The central structure depicts an interface between a 2D magnet (green) and a dissimilar material (orange). (A) Charge transfer and interfacial dipole. The orange and red balls represent electrons and holes, respectively. (B) Interfacial hybridization. The lower green bar represents a 2D magnet; the upper bar is a dissimilar material. The dumbbells represent electronic orbitals of the two materials, overlapping at the interface to hybridize. (C) Strain effect. The lower solid bar represents a stretched 2D magnet in contact with a dissimilar material; the lower dashed bar represents the relaxed 2D magnet without the top contacted material. (D) Additional superexchange interactions. The orange circles with arrows represent the elements in adjacent materials that provide additional channels to mediate the superexchange interaction between magnetic ions in 2D magnets, which are represented by the red solid balls with arrows. (E) Structural perturbation. The wavy green belt represents 2D magnets that are structurally perturbed because of contact with the adjacent materials. (F) Band renormalization. The solid curves represent the electronic, magnonic, or phononic band dispersions of 2D magnets after band renormalization with contact of the adjacent materials; the dashed curves represent the same band dispersions before band renormalization without contacting the adjacent materials. (G) Dielectric screening. Red balls with arrows represent the exchange-coupled electrons in 2D magnets; orange curves depict the electric field lines connecting electrons. The environment with higher dielectric constant  $\epsilon$  screens the coulombic interaction more. The nature of exchange interaction as a coulombic interaction makes 2D magnets susceptible to the dielectric screening. (H) Spin-orbit coupling (SOC) proximity. By contacting heavy-element materials, the SOC in 2D magnets will be effectively modified, leading to the change of magnetocrystalline anisotropy.

Fe<sub>0.25</sub>TaS<sub>2</sub>-Ta<sub>2</sub>O<sub>5</sub>-Fe<sub>0.25</sub>TaS<sub>2</sub> (124), graphite-CrI<sub>3</sub>graphite (49, 125-127), Fe<sub>3</sub>GeTe<sub>2</sub>-BN-Fe<sub>3</sub>GeTe<sub>2</sub> (128), and graphite-CrBr<sub>3</sub>-graphite (129). The Curie temperatures of  $Fe_x TaS_2$  are tunable by Fe's stoichiometry with maximum  $T_{\rm C}$  at ~160 K when x = 0.2, and Ta<sub>2</sub>O<sub>5</sub> is a thin non-vdW native oxide (Fig. 5, A and B). The tunneling magnetoresistance is about 6.5% at 5 K (124). Recently, several research groups reported the very large tunneling magnetoresistance based on graphite-CrI<sub>3</sub>-graphite sandwich structures (Fig. 5, C and D), with maximal magnetoresistance amounting to 19,000% at 2 K (125), 550% at 300 mK (126), 10,000% at 10 K (49), and 1,000,000% at 1.4 K (127). The large variance among these reported tunneling magnetoresistance values relates to such detailed experimental conditions as measurement temperature, thickness of the spacing layer, applied bias, and the detailed interfacial quality. The key of this type of MTJ was deemed to make use of the multiple scattering of tunneling electrons across the alternatively spin-polarized CrI<sub>2</sub> lavers. These MTJs were found to be a result of the magnon-mediated tunneling process, in contrast to the conventional phonon- or electronmediated tunneling (126, 129). Another vdW MTJ, Fe<sub>3</sub>GeTe<sub>2</sub>-BN-Fe<sub>3</sub>GeTe<sub>2</sub>, exhibited 160% tunneling magnetoresistance at 4.2 K (128). These proofof-concept studies show the attractive promise of vdW magnets in high-efficiency spintronics or magnonics.

To be balanced, outstanding challenges remain in these vdW MTJs concerning room temperature, nonvolatility, and low-power switching. For example, the current version of CrI<sub>3</sub>-MTJs works at about liquid helium temperatures, is volatile, and requires large magnetic fields for toggling between distinct states (e.g., 2 T for tetralayers). For practical vdW MTJs, efforts need to be directed toward such important issues as high-temperature 2D magnets, perpendicular anisotropy, large remanence, modest coercivity, and robust exchange bias with antiferromagnets. Nonetheless, despite challenges, it is hoped that the smaller magnetic volume in ultrathin vdW MTJs, better electrical control, and enhanced thermal fluctuations in 2D magnets could allow a lower critical current for spin torque magnetization switching relative to the traditional nonvdW MTJs. This lower critical current is the key long-sought goal for spin-transfer torque magnetoresistive random-access memory (STT-MRAM).

Bilayer structures consisting of 2D and magnetic materials provide intriguing opportunities in magnonics and spin-orbitronics. Spin pumping and spin-orbit torque are reciprocal processes. Magnons can be excited in magnetic substrates coherently by microwave. The excited magnons, without the necessity of conducting electrons, then propagate into the adjacent 2D materials. If the 2D materials have large spin-orbit coupling strengths or large inverse Rashba-Edelstein coefficients, they would be capable of efficiently converting the traveling magnons into charge current and detectable voltage. Proof-of-concept experiments on such spin-charge conversion have been conducted in graphene-YIG (*130, 131*) (Fig. 5E),  $MoS_2$ -Al-Co (132), and  $MoS_2$ -YIG (133) heterostructures.

In a reverse manner to spin pumping, spintransfer torque or spin-orbit torque results from an injection of spin current from a 2D material into a magnetic thin-film substrate. A longitudinal charge current in a 2D material with strong spinorbit coupling can deflect electrons of transverse spin orientations toward the directions normal to the bilayer interface, resulting in the injection of a



Fig. 5. Spintronic, magnonic, and spin-orbitronic devices based on 2D magnets or magnetic heterostructures. (A and B) MTJ based on  $Fe_{0.25}TaS_2-Ta_2O_5-Fe_{0.25}TaS_2$  (124). Iron-intercalated  $TaS_2$  is ferromagnetic, and the surface native oxide was used as an insulating spacing layer. (A) Atomic structure of Fe-intercalated  $TaS_2$ . (B) Cross-section transmission electron microscopy image of the MTJ sandwich structure. (C and D) MTJ based on graphite-CrI<sub>3</sub>-graphite (126). (C) Schematic of MTJ. (D) Magnetic field–dependent tunneling conductance. (E) Schematic of graphene-YIG heterostructure for spin-charge conversion based on spin pumping (130). (F) Schematic of the spin-orbit torque measurement system, for which the core material architecture is WTe<sub>2</sub>-permalloy heterostructure (135). Inset is an optical image of the tested device. (G and H) Schematics of a spin field-effect transistor based on a bilayer A-type antiferromagnet and its predicted electrical properties (140).

Chalcogenides	$Cr_2Ge_2Te_6$ , $Cr_2Si_2Te_6$ , $Fe_3GeTe_2$ , $VSe_2^*$ , $MnSe_x^*$	$Fe_{2}P_{2}S_{6}, Fe_{2}P_{2}Se_{6}, Mn_{2}P_{2}S_{6}, Mn_{2}P_{2}Se_{6}, Ni_{2}P_{2}S_{6}, Ni_{2}P_{2}Se_{6}, CuCrP_{2}Se_{6}^{*}, AgVP_{2}S_{6}, AgCrP_{2}S_{6}, CrSe_{2}, CrTe_{3}, Ni_{3}Cr_{2}P_{2}S_{9}, MnBi_{2}Te_{4}^{*}, MnBi_{2}Se_{4}^{*}$		CuCrP <sub>2</sub> S <sub>6</sub>
Halides	CrI <sub>3</sub> *, CrBr <sub>3</sub> , GdI <sub>2</sub>	CrCl <sub>3</sub> , FeCl <sub>2</sub> , FeBr <sub>2</sub> , FeI <sub>2</sub> , MnBr <sub>2</sub> , CoCl <sub>2</sub> , CoBr <sub>2</sub> , NiCl <sub>2</sub> , VCl <sub>2</sub> , VBr <sub>2</sub> , VI <sub>2</sub> , FeCl <sub>3</sub> , FeBr <sub>3</sub> , CrOCl, CrOBr, CrSBr, MnCl <sub>2</sub> <sup>*</sup> , VCl <sub>3</sub> <sup>*</sup> , VBr <sub>3</sub> <sup>*</sup>	$\mathrm{CuCl}_2, \mathrm{CuBr}_2, \mathrm{NiBr}_2, \mathrm{NiI}_2, \mathrm{CoI}_2, \mathrm{MnI}_2$	
			α-RuCl <sub>3</sub>	
Others	VS <sub>2</sub> , InP <sub>3</sub> , GaSe, GaS	$\label{eq:MnX_3} \begin{array}{l} (\mathrm{X}=\mathrm{F},\mathrm{Cl},\mathrm{Br},\mathrm{I}),\mathrm{FeX_2}\;(\mathrm{X}=\mathrm{Cl},\mathrm{Br},\mathrm{I}),\\ \mathrm{MnSSe},\mathrm{TiCl_3},\mathrm{VCl_3} \end{array}$	SnO, GeS, GeSe, SnS, SnSe, GaTeCl, CrN, CrB <sub>2</sub>	

**Fig. 6. Van der Waals magnets library.** Color code: Green, bulk ferromagnetic vdW crystals; orange, bulk antiferromagnets; yellow, bulk multiferroics; gray, theoretically predicted vdW ferromagnets (left), half metals (center), and multiferroics (right), which have not yet been experimentally confirmed; purple,  $\alpha$ -RuCl<sub>3</sub> (a proximate Kitaev quantum spin liquid) (144). Notes (asterisks): VSe<sub>2</sub> has been found only in 1T-VSe<sub>2</sub> form in experiments to date (145), although the magnetic properties of 2H-VSe<sub>2</sub> have been widely studied by density functional theory calculations. MnSe<sub>x</sub> is ferromagnetic and of vdW structure in MBE-synthesized 2D form but is antiferromagnetic in bulk (which could be either rock-salt or hexagonal structure). Crl<sub>3</sub>, although ferromagnetic in bulk, was experimentally suggested to be

spin-polarized current into the magnetic substrate. The injected spin current exerts spin-orbit torques on the magnetic moments of the underlying magnetic substrate, exciting the moments to precess or even switch to the opposite orientation. Spin-orbit torque MRAM, if using 2D materials as the spincurrent sources, has the potential toward the atomic thinness. The other advantage of using 2D materials as a spin-current source is that the majority of the spin current would be at the interface rather than being dissipated in bulk, constituting a tantalizing platform for low-power spintronics. Many types of bilayer structures consisting of 2D materials such as MoS<sub>2</sub>-permalloy (134), WTe<sub>2</sub>-permalloy (135) (Fig. 5F), and (MoS2 and WSe2)-CoFeB (136) have been investigated.

Lastly, it is worthwhile to note the family of half metals. To improve spintronics, high spin polarization at the Fermi level of a material is desirable. Half metallicity, a property arising from the metallic nature for electrons with singular spin polarization and insulating for electrons with the opposite spin, holds the potential for 100% spinpolarized conduction electrons. There have been a few theoretical predictions on the possible 2D half metals, including manganese trihalides (137), FeCl<sub>2</sub>, FeBr<sub>2</sub>, FeI<sub>2</sub> (138), and Janus structure of monolayer MnSSe (139). Half metallicity could also be electrically induced in bilayer A-type antiferromagnets (e.g., in 2H-VSe<sub>2</sub>) (140). This electrically induced half metallicity potentially prompts a new type of spin-field-effect transistor in which both switching and reversal of spin polarization can be realized by a single knob: gate voltage, in analogy to the conventional semiconductor transistors (Fig. 5, G and H).

#### **Conclusions and outlook**

The recently discovered 2D magnetic crystals provide ideal platforms to study the ground state, fundamental excitations, dynamics, and frustrations of spin ensembles under strong quantum confinement. Such intrinsic 2D magnetic crystals distinguish themselves from traditional magnetic thin films and nonmagnetic 2D materials, in terms of both properties and application prospects. The interplay of dimensionality, correlation, charge, orbital character, and topology makes 2D magnetic crystals and heterostructures extremely fertile condensed matter systems with a large reservoir of exotic properties. The 2D materials' outstanding feature of being susceptible to a large variety of external stimuli makes the versatile control of 2D magnetism possible by electrical, mechanical, chemical, and optical approaches. Furthermore, because of the unrivaled compatibility for heterostructure constructions, vdW heterostructures present attractive opportunities for designer 2D magnetic, magnetoelectric, and magneto-optical artificial heteromaterials. Those materials that contact 2D magnets will affect the 2D magnetic properties. Novel spintronic, magnonic, and spinorbitronic devices have started to emerge.

As evident in Fig. 6, a large variety of magnetic vdW materials are available (141-148), most of whose 2D counterparts remain to be studied. The family of 2D magnetic crystals is rapidly growing, magnetic heterostructures consisting of 2D materials are being actively expanded, and new device concepts are being developed. One of the most critical needs concerns the material-level realization of room-temperature 2D ferromagnets (in contrast to sustaining a high Curie temperature with external supplies such as electrical, optical, and mechanical means), which ideally would be air-stable. Furthermore, the wafer-scale synthesis (149) of such crystals has to be accomplished for practical mass production. Advances in both metallic and insulating 2D magnets, both 2D ferromagnets and antiferromagnets, promise various aspects of applications based on their distinct properties.

Fundamental spintronic parameters—including spin Hall angle, spin diffusion length, magnon

an A-type antiferromagnet in the 2D regime. CuCrP<sub>2</sub>Se<sub>6</sub> does not host the electric order while being cooled down to 10 K according to experimental data (147), but the calculated ground state of CuCrP<sub>2</sub>Se<sub>6</sub> is multiferroic with antiferroelectricity (99). MnBi<sub>2</sub>Te<sub>4</sub> and MnBi<sub>2</sub>Se<sub>4</sub> may exhibit ferrimagnetic features as a result of uncompensated odd-layer A-type antiferromagnets or surfaces of antiferromagnetic topological insulators. MnCl<sub>2</sub> has a magnetic structure that has not been completely determined, which could be either antiferromagnetic or helimagnetic (142). Bulk VCl<sub>3</sub> and VBr<sub>3</sub> have been inferred to be weak antiferromagnets on the basis of experimental data, although detailed magnetic structures have not been determined; however, monolayer VCl<sub>3</sub> was calculated to be ferromagnetic (146).

damping coefficient, spin injection efficiency, and spin mixing conductance-must be evaluated in 2D magnets and across interfaces. There is a need for in-depth understanding of the relation between spintronic transport metrics and material parameters, along with the corresponding strategies for optimization. Furthermore, exotic spin textures and topologically protected spin configurations (e.g., magnetic skyrmions) in 2D magnets or heterostructures remain open to explore by rationally weighing the material constituents, crystal symmetry, spin-orbit coupling, Rashba effect, and spin (re-)orientations. New quantum phases and quasiparticles could be found, leading to new ways of computation and communication. Such advances in understanding and control of 2D magnetic crystals and emergent heterostructure devices will foster a widespread range of applications (150) including low-power spintronics, quantum computing, and optical communications.

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#### Two-dimensional magnetic crystals and emergent heterostructure devices

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#### The ultimate in thin-film magnetism

The alignment of the magnetic properties of atoms gives rise to a wealth of simple and exotic properties that can be exploited. As the dimension of the material is reduced, such that the atoms are in a single monolayer, it was widely believed that thermal fluctuations overwhelm and prevent magnetic ordering. Gong and Zhang review the developments that have followed the recent discovery of magnetism in two-dimensional materials. Recognizing that magnetic anisotropy can be used to induce stable magnetism in atomic monolayers, they provide an overview of the materials available and the physical understanding of the effects and then discuss how these effects could be exploited for widespread practical applications.

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