Two-dimensional magnetic transition metal chalcogenides

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Abstract
The field of two-dimensional (2D) magnets has expanded rapidly during the past several years since the first demonstration of intrinsic 2D magnetism in atomically thin CrI₃ and Cr₂Ge₂Te₆ in 2017. 2D transition metal chalcogenides (TMCs), a class of strongly correlated materials, have exhibited a wide variety of novel electronic and optical properties, and more recently magnetism. Here, we review recent experimental progress achieved in the growth of 2D magnetic TMC materials using chemical vapor deposition and molecular beam epitaxy methods. Outstanding examples include the demonstration of room temperature intrinsic and extrinsic ferromagnetism in monolayer VSe₂, MnSe₂, Cr₃Te₄, V-doped WSe₂, and so on. A brief discussion on the origin of the exotic magnetic properties and emergent phenomena is also presented. Finally, we summarize the remaining challenges and future perspective on the development of 2D magnetic materials for next-generation spintronic devices.

KEYWORDS
chemical vapor deposition, molecular beam epitaxy, transition metal chalcogenides, two-dimensional magnets

1 | INTRODUCTION

The realization of magnetism in two-dimensional (2D) systems can potentially revolutionize the development of information technology.¹⁻⁷ However, long-range magnetism is difficult to manifest in 2D crystals due to enhanced thermal fluctuations, as predicted by the Mermin–Wagner theorem.⁸ Until 2017, intrinsic 2D magnetism was experimentally achieved in atomically thin layers of CrI₃⁹ and Cr₂Ge₂Te₆¹⁰ showing that the long-range magnetic ordering can be stabilized by magnetic anisotropy with an excitation gap opening. Subsequently, studies have revealed exotic magnetic properties in several other 2D materials, for example, Fe₂GeTe₂,¹¹ FePS₃,¹² and MnSe₂,¹³ which are tunable with the layer thickness,⁹,¹⁴ electric field,¹⁵,¹⁶ strain,¹⁷⁻¹⁹ light,²⁰ and so on. These properties offer attractive opportunities to explore emergent phenomena in these materials and their heterostructures, including the quantum Hall effect, quantum spin Hall effect, quantum spin liquids, and the fractionalization of quasi-particles.⁴ Challenges remain in the large-area controllable growth of 2D magnets with room temperature magnetic ordering, and the validation of their novel properties and emergent phenomena in the 2D limit for applications in next-generation memory and information technology, spintronics, and quantum computing.

The family of binary transition metal chalcogenides (TMCs), including transition metal monochalcogenides,
transition metal dichalcogenides, and other stoichiometries with the general formula of $M_nX_n$ (M refers to transition metal, and X represents S, Se, and Te), represents an emergent class of layered and nonlayered materials with a wide range of robust electronic, optoelectronic, and magnetic properties.\textsuperscript{21–23} On the basis of the first-principles calculations, it was predicted that intrinsinc long-range magnetic ordering can exist in many TMCs with $M = V$, Cr, Mn, Fe, Co and Ni.\textsuperscript{17,24,25} Recently, increasing research effort has focussed on the fabrication of large-scale 2D magnetic TMCs via methods such as chemical vapor deposition (CVD)\textsuperscript{23} and molecular beam epitaxy (MBE).\textsuperscript{26–28} In particular, the successful growth of monolayer (ML) MnSe\textsubscript{2},\textsuperscript{13} VSe\textsubscript{2},\textsuperscript{27} and Cr\textsubscript{3}Te\textsubscript{4}\textsuperscript{29} with Curie temperature ($T_c$) close to and even above room temperature has been demonstrated. Extrinsic magnetism can be also introduced into the otherwise nonmagnetic 2D TMCs by proximity coupling,\textsuperscript{30} defects,\textsuperscript{11,32} doping,\textsuperscript{33,34} and so on. Elegant examples include the long-range magnetic ordering induced by Pt vacancies in ultrathin PtSe\textsubscript{2} films,\textsuperscript{35,36} and V-dopants in ML WSe\textsubscript{2}.\textsuperscript{37,38} These results inspire further research interest in the study of 2D magnetic TMCs.

In this review, we highlight recent research progress achieved in the growth of 2D TMCs, particularly, ML TMCs with room-temperature ferromagnetic ordering. We present a brief overview of the successful fabrication of mono- and few-layer vanadium dichalcogenides, chromium chalcogenides, and so on, using CVD and MBE methods. We next discuss the strategies to induce and control the magnetic properties in nonmagnetic 2D materials (e.g., PtSe\textsubscript{2} and WSe\textsubscript{2}) via defect and dopant engineering. A fundamental understanding of the origin of the magnetism is also presented. Finally, we offer our insight on the challenges and future perspectives of the field of 2D magnetic materials.

## 2 | INTRINSIC MAGNETISM IN 2D TMCS

### 2.1 | Vanadium dichalcogenides

As a prototype metallic transition metal dichalcogenide, 2D vanadium dichalcogenide (VX\textsubscript{2}, X = S, Se, Te) exhibits a rich variety of interesting physical phenomena, such as predicted room-temperature ferromagnetism, enhanced charge-density wave (CDW) ordering, metal-insulator transition, and valley polarization, stemming from its odd 3$d^1$ electronic configuration.\textsuperscript{17,27,39–42} A ferromagnetic or antiferromagnetic order is predicted in VX\textsubscript{2} ML with both 2H and 1T polymorphs, depending sensitively on the electron correlation of the V d-shell electrons in first-principles calculations.\textsuperscript{17,43–45} However, controversy exists over the intrinsic magnetism in the ML limit.

ML VSe\textsubscript{2} was reported to be one of the first room-temperature 2D ferromagnets by Bonilla et al.\textsuperscript{27} Using superconducting quantum interference device (SQUID) and vibrating-sample magnetometry (VSM), single-layer VSe\textsubscript{2} films grown on graphite and MoS\textsubscript{2} substrates by MBE method demonstrate strong ferromagnetic ordering at room temperature. The magnetic moment was experimentally estimated to be 5–15 $\mu_B$ per formula unit,\textsuperscript{27,41} which is much larger than the calculated values (0.36–0.6 $\mu_B$).\textsuperscript{17,43,44} However, Feng et al.\textsuperscript{46} detected no magnetic signal in X-ray magnetic circular dichroism (XMCD) measurements. Wong et al. have observed traits of spin frustration in MBE-grown ML VSe\textsubscript{2} without long-range magnetic order by means of in situ microscopy and spectroscopic techniques.\textsuperscript{42} Figures 1A and 1B show the atomic model and an atomically resolved scanning tunneling microscopy (STM) image of an ML 1T-VSe\textsubscript{2} on graphite, respectively. Element-specific XMCD spectra (Figure 1C) suggest no exchange-split V electronic bands and ferromagnetically coupled V 3d state observed at 16 K in a 1 T magnetic field. By including a Hubbard on-site repulsion $U$ in DFT + $U$ simulations, the antiferromagnetic state of 1T-phase VSe\textsubscript{2} is more stable than the ferromagnetic state by 0.022 eV per unit cell,\textsuperscript{42} so they proposed that ML VSe\textsubscript{2} is on the verge of thermal instability. In addition, structural distortion-derived CDW orders have been consistently observed by STM and angle-resolved photoemission spectroscopy (ARPES) in ML VSe\textsubscript{2} grown on various substrates.\textsuperscript{41,46,47} As shown in Figure 1D, THE electronic structure of a VSe\textsubscript{2} ML grown on bilayer graphene/SiC reveals a pronounced CDW gap opening at the Fermi level crossing along the $\mathbf{M} - \mathbf{K}$ direction at 20 K.\textsuperscript{48} The CDW gap varies as a function of temperature, with onset at 140 ± 5 K (Figure 1E). It has also been suggested that the CDW transition opens an energy gap at the Fermi level that in turn suppresses the intrinsic ferromagnetic ordering.\textsuperscript{41,46–49} Similarly, MBE-grown VTe\textsubscript{2} MLs on graphite demonstrate a 4 x 4 CDW order at low temperature and XMCD measurements rule out the existence of intrinsic ferromagnetic order.\textsuperscript{50}

The ferromagnetism in ML VSe\textsubscript{2} can be promoted, even at room temperature, after coupling with ferromagnetic Co or Fe thin films.\textsuperscript{51,52} Figure 2A shows a contamination-free Co/VSe\textsubscript{2} interface prepared by in situ thermally deposition of Co onto ML VSe\textsubscript{2} on graphite under ultrahigh vacuum.\textsuperscript{51} The emergence of magnetic polarization on the V atoms upon the deposition of 3 ML Co is confirmed by XMCD hysteresis loops displayed in Figure 2B. The antiferromagnetic coupling of VSe\textsubscript{2} ML
with the Co or Fe thin films induces a magnetic moment of about 0.3 μB and 0.16 per μB per V atom at room temperature, respectively. The observed magnetic transition in the Co/VSe₂ and Fe/VSe₂ heterointerface induced by such a proximity effect provides further evidence for the absence of intrinsic ferromagnetic ordering in ML VSe₂.

Surface inhomogeneities, for example, defects and patterns, may manifest exotic properties absent in the pristine crystal. Yu et al. have reported that Se vacancies can enhance the ferromagnetism of solution-exfoliated VSe₂, whereas the introduction of thiol molecule layers can improve its air stability and protect the ferromagnetism. The frustrated intrinsic magnetism appears to be lifted by the formation of 1D Se-deficient reconstructions in MBE-grown ML VSe₂. Figure 2C shows an atomically resolved atomic force microscopy (AFM) image of a VSe₂ ML epitaxially grown on MoS₂ after annealing above 400°C, where distinct line features are visible. These 1D defective patterns are identified to be eight-member-ring arrays (Figure 2E), which are formed via a Se deficient reconstruction process. The density of the line features is controllable with the annealing temperature as well as duration. This Se-deficient defective VSe₂ ML displays detectable ferromagnetism with magnetic force microscopy (MFM) and XMCD measurements at room temperature (Figure 2D). DFT calculations confirm the enhanced magnetization upon the formation of the Se-deficient reconstructed patterns, and the suppressed magnetization with the Se-deficient pattern without reconstruction (Figure 2E). Furthermore, the formation of well-ordered Se-deficient line

**FIGURE 1** Monolayer 1T-VSe₂. (A) Atomic model and (B) atomic-resolution scanning tunneling microscopy (STM) image of 1T-VSe₂ monolayer on graphite. (C) Upper panel shows the X-ray adsorption spectra obtained with opposite magnetic field (±1 T) at 30 K, which are highlighted in red and green, respectively, and the total XAS of their sum is in purple. The lower panel shows that the corresponding difference signal (X-ray magnetic circular dichroism [XMCD]) is within the experimental error, indicating the absence of ferromagnetic ordering. Reproduced with permission: 2019, Wiley-VCH GmbH. (D) The electronic structure of monolayer VSe₂ grown on bilayer graphene/SiC measured at 20 K. The orange arrow indicates a pronounced CDW gap opening at the Fermi level. (E) Temperature-dependent CDW gaps derived from ARPES (orange dots) are consistent well with LEED measurements (green triangles). Reproduced with permission: 2018, American Chemical Society.
defects by annealing has also been observed in ML-VSe₂ grown on graphite and demonstrates promising catalytic activity for the hydrogen evolution reaction, comparable to Pt surfaces. Annealing a 2D crystal at elevated temperature is a simple method to engineer the formation of surface defects, which induces ferromagnetism with potential applications in devices.

2.2 Chromium chalcogenides

Chromium chalcogenides (CrₘXₙ, X = S, Se and Te, and m < n), an emergent class of nonlayered and layered materials with robust magnetic ordering, have attracted increasing research interest. Bulk CrₘXₙ is a group of materials with a hexagonal structure similar to NiAs in the P2/m space group (Figure 3). Nonlayered CrₘXₙ (e.g., CrX, Cr₂X₃, and Cr₃X₄) possess a prismatic structure consisting of octahedral CrX₂ layers (an analogy to 1 T MoS₂ ML) connected by intercalated Cr atoms bonding with neighboring X atoms. With different amounts of Cr vacancies in the intercalated Cr layers, a range of CrₘXₙ with different stoichiometric ratios, such as CrX, Cr₂X₃, Cr₃X₄, Cr₄X₅, and CrTe₂, can be formed, and behave as ferrimagnetic, antiferromagnetic, and ferromagnetic. Epitaxial Cr₂S₃, Cr₄Se, Cr₆Te, and Cr₁Te₂ thin films with thicknesses down to several nanometers have been grown on various substrates (mica, SrTiO₃, Si, Bi₂Se₃, and so on) by CVD and MBE, showing high Curie temperature Tᵣ (e.g., ~305 K for 8.7 nm CrTe₂ thin film), and promising electrical transport properties (e.g., unconventional anomalous Hall effect for spintronic devices. The successful synthesis of 2D Cr₄S₃, Cr₂Te₄, and CrTe₂ with ML thickness (referring to the smallest repeating unit along the z-direction) have also been recently reported.

Figure 3 shows various 2D nonlayered CrₘXₙ samples grown by ambient pressure chemical vapor deposition (APCVD) on mica substrates reported by He’s group. Figure 3A–D show CrS₂ single-crystal behaves as a ferrimagnetic semiconductor with good air-stability, with thickness down to one-unit-cell (~1.78 nm)
and lateral size up to 200 μm. The as-grown Cr₂S₃ possesses a Néel temperature \( T_N \) of 120 K, and a saturation magnetic momentum of 65 μemu (Figure 3C,D). It also demonstrates a p-type transport behavior with an on/off ratio higher than 10³. 2D ferromagnetic Cr₂S₃ with thickness down to ML limit has been synthesized via APCVD by Cui et al. They found that the transport characteristics of the Cr₂S₃ nanoflakes are thickness-dependent, varying from p-type to ambipolar and then to n-type as the thickness increases from 2.6 to 4.8 nm and to over 7 nm. The controllable synthesis of ultrathin CrSe crystals, with thickness down to 2.5 nm and lateral size up to submillimeter scale is demonstrated in Figure 3E–I, showing ferromagnetic ordering at a temperature below 280 K and strong perpendicular magnetic anisotropy. Furthermore, room-temperature ferromagnetism has also been observed in 2D Cr₂Te₃ (Figure 3J–M), confirmed by anomalous Hall effect measurements (Figure 3L). Interestingly, the transition temperature of the ferromagnetism phase \( T_c \) increases from 160 K in the Cr₂Te₃ thick film (40.3 nm) to 280 K in the thin film (7.1 nm). Although the synthesis of 1- and 2-unit-cell Cr₂Te₃ has been achieved (Figure 3K), precise characterization of their magnetic properties is lacking due to poor stability. The origin of the enhanced ferromagnetism in 2D Cr₂Te₃ is still unclear and has been postulated to be due to doping or reconstruction of the 2D Cr₂Te₃.

Chua et al. reported the successful fabrication of Cr₃Te₄ MLs on graphite with room temperature ferromagnetism. It is found that the morphology of the non-layered Cr₃Te₄ crystals significantly depends on the substrate temperature during the MBE growth. As shown in Figure 4A–D, the 2D Cr₃Te₄ islands grown at high substrate temperature (e.g., 400°C and 300°C) demonstrate regular hexagonal shape, whereas the ones grown at low temperature (e.g., 100°C) show irregular dendritic shape. Before full coverage of the substrate is achieved,
the average thickness of the Cr$_3$Te$_4$ flakes decreases gradually with the substrate temperature, as revealed by the lateral profiles in Figure 4E–H. For the samples grown at 100°C and 200°C, the height of the first layer is only 0.93 ± 0.2 nm, indicating the successful growth of 2D Cr$_3$Te$_4$ with ML thickness. Figure 4I shows the experimental atomically resolved STM image of Cr$_3$Te$_4$ ML on graphite, revealing a quasi-hexagonal lattice structure with unidirectional 1D modulation. This is consistent with the simulated STM image shown in Figure 4J, based on the ML Cr$_3$Te$_4$ model (50% Cr vacancies in the intercalated layer and Te-terminated layers in both sides, and thus a Cr:Te stoichiometry of 1:2 is obtained) presented in Figure 4K. The 1D modulation observed both experimentally and theoretically, is ascribed to the surface fluctuation as well as the bond distortion arising from the periodic Cr vacancies in the Cr$_3$Te$_4$ ML.

The M–T curves extracted from XMCD measurements (Figure 4L) reveals that the Cr$_3$Te$_4$ ML possesses an out-of-plane magnetic easy axis and $T_c$ of ~344 K. Thicker Cr$_3$Te$_4$ films (~7 nm) have an in-plane easy axis and a lower $T_c$ (~240 K). DFT calculations suggest that the transition of the magnetic easy axis and the enhancement of ferromagnetic coupling in the Cr$_3$Te$_4$ ML is ascribed to interfacial charge transfer at the ML Cr$_3$Te$_4$/graphite interface. This study sheds light on the scalable growth of 2D nonlayered materials with room temperature ferromagnetism for new magnetic and spintronic devices.

In particular, the low temperature (~100°C) growth process is compatible with Si CMOS technology, opening new opportunities for hybrid 2D–Si systems.

Significant progress has also been made in the synthesis of 2D layered chromium dichalcogenides (CrX$_2$) by CVD, for example, magnetic CrSe$_2$ and
CrTe₂,⁶⁵ which demonstrate outstanding air stability. Duan’s group reported the synthesis of 2D CrSe₂ nanosheets with tunable thickness down to the ML limit on dangling-bond-free 2D WSe₂.⁶⁴ The thickness-dependent magnetic order, that is, the evolution from weak ferromagnetism in mono-, bi- and tri-layer to strong ferromagnetism in thicker CrSe₂ nanosheets, is largely governed by the charge transfer from the WSe₂ substrate and the interlayer coupling within the CrSe₂ crystals. A Curie temperature \( T_c \) of 65 K is estimated for the ML CrSe₂, which gradually increases to 110 K at the thickness of 16 or more layers. In the other study, Meng et al. grew 1T-CrTe₂ on SiO₂/Si surface with thicknesses ranging from a few to tens of nanometers.⁶⁵ They observed that the magnetic easy axis in the 2D CrTe₂ crystals changes from in-plane to out-of-plane with the decreasing thickness at a critical thickness of ~10 nm. In addition, \( T_c \) increases with decreasing thickness, from ~187 K in the ~130.0 nm film to ~210 K in the ~7.5 nm one. DFT calculations suggest that the occupation of Cr \( d \) orbitals changes significantly with the Coulomb screening in the 2D limit, resulting in many possible magnetic mechanisms with the variation of the thickness. The highly controllable growth, as well as the stability of these CrX₂ materials, are important for fundamental studies and the realization of device applications.

2.3 | Manganese, ion, and cobalt chalcogenides

Room-temperature ferromagnetism has also been observed in ML MnSex grown by MBE (Figure 5A–C).¹³ Figure 5C shows the hysteresis loops for ~1 ML and 40 nm MnSex films grown on GaSe(0001)/GaAs(111) obtained from SQUID measurements, revealing out-of-plane ferromagnetic ordering of MnSex. At room temperature, a saturation magnetic moment of ~4 \( \mu_B \) per Mn
atom is estimated for the ~1 ML MnSe\textsubscript{x} on GaSe and SnSe\textsubscript{2} epilayers, consistent with the DFT predicted value. The magnetic moment is doubled in the 40 nm MnSe\textsubscript{x} film compared to the ML sample. Detailed structural analyses indicate that the ferromagnetism originates from a vdW 1T-MnSe\textsubscript{x} monolayer (Figure 5B) in the ML limit, and could originate from the combination of 1T-MnSe\textsubscript{x} and/or interfacial magnetism of α-MnSe (Figure 5A) in the thick film. This behavior differs from the bulk MnSe\textsubscript{2} (pyrite structure) and α-MnSe (rocksalt structure), which are not ferromagnetic. α-MnTe is another nonlayered p-type semiconductor with antiferromagnetic ordering and a Néel temperature \(T_N\) of 307 K.\textsuperscript{67} It exhibits good stability, high thermal conductivity, and high carrier concentration above room temperature.\textsuperscript{68–70} The controllable growth of high-quality α-MnTe thin films has been realized by MBE\textsuperscript{68,69,71} as well as CVD.\textsuperscript{70}

Iron chalcogenides, another strongly correlated class of materials, have attracted much attention following the discovery of superconductivity in β-FeSe\textsubscript{2} (a PbO-type phase belong to P4/nmm space group). Interesting magnetic properties have also been observed in hexagonal Fe\textsubscript{2}Se\textsubscript{2}\textsuperscript{73} and monoclinic Fe\textsubscript{2}Se\textsubscript{4}\textsuperscript{74} and FeTe.\textsuperscript{66} However, the direct growth of their 2D crystals is lacking.\textsuperscript{75} Liu’s group recently reported a CVD approach to grow layered tetragonal and nonlayered hexagonal FeTe flakes with thickness down to 3.6 and 2.8 nm, respectively (Figure 5D–I).\textsuperscript{66} The phase of FeTe is controlled by the growth temperature (\(T_{\text{growth}}\)), with tetragonal FeTe obtained at ~550°C (Figure 5D–F), and the hexagonal FeTe at ~590°C. The magnetic properties vary significantly with the structural phases. Figures 5F and 5I show the magnetoelectric characteristics of the tetragonal and hexagonal FeTe samples on SiO\textsubscript{2}/Si substrate encapsulated by h-BN capping layers, respectively. The tetragonal FeTe is antiferromagnetic with \(T_N\) gradually decreasing from 70 to 45 K as the thickness decreases from 32 to 5 nm (Figure 5F). In contrast, hexagonal FeTe is ferromagnetic and displays a temperature-dependent anomalous Hall effect. The \(T_N\) of the hexagonal FeTe also decreases with its thickness, from 220 K for 30 nm to 170 K for 4 nm (Figure 5I). Theoretical calculations suggest that the ferromagnetic ordering in hexagonal FeTe is induced by lattice distortion.

Cobalt chalcogenides are also promising candidates for 2D magnets with multiple structural phases. CoS\textsubscript{2} and CoSe\textsubscript{2} are pyrite-type cubic compounds, and CoS\textsubscript{3} is a ferromagnetic metal with \(T_c\) of 124 K, whereas CoSe\textsubscript{3} is either a Pauli paramagnet or antiferromagnet.\textsuperscript{76,77} Ultrathin CoX\textsubscript{2} crystals have been successfully fabricated by CVD, and the thickness can be controlled from a ML to tens of nanometers.\textsuperscript{78} Magneto-transport measurements reveal that both CoSe\textsubscript{2} and CoTe\textsubscript{2} are paramagnetic in nature, and exhibit linear magnetoresistance.\textsuperscript{78} In addition, CoS and CoSe with anti-PbO type structure are found to be weak itinerant ferromagnets with \(T_c\) of ~10 K.\textsuperscript{79,80} By analogy with FeTe, layered tetragonal and nonlayered hexagonal CoSe can be fabricated by control of the growth temperature during the CVD growth process, and both exhibit strong thickness-dependent electrical properties.\textsuperscript{81} Such multiple-phase 2D materials with tunable properties offer new opportunities for electronic and magnetoelectronic devices but pose challenges in the controlled growth of specific phases.

3 | 2D TMCS WITH EXTRINSIC FERROMAGNETISM

3.1 | Intrinsic defects

Intrinsic defects, such as vacancies, edges, and grain boundaries, which are inevitable in 2D material growth, can induce extrinsic magnetism in otherwise non-magnetic 2D crystals.\textsuperscript{82} For example, it has been predicted that the magnetism can be induced by carbon defects in graphene,\textsuperscript{83} as well as vacancies and grain boundaries in 2D TMCS.\textsuperscript{84–87} Experimentally, robust room-temperature ferromagnetism has been observed in MoS\textsubscript{2} nanosheets, induced by S vacancies intentionally introduced by a hydrothermal method.\textsuperscript{85} Long-range magnetic ordering has also been discovered in bulk semiconducting 2H-MoTe\textsubscript{2} and 2H-MoSe\textsubscript{2} due to antisite defects.\textsuperscript{81}

Thickness-modulated magnetism in ultrathin PtSe\textsubscript{2} crystals induced by Pt vacancies has been recently demonstrated by Kis’ group via magnetoresistance measurements (Figure 6).\textsuperscript{35,36} PtSe\textsubscript{2} is an interesting TMC material due to its unique thickness-dependent electronic properties and undergoes a metal-to-semiconductor transition from thin films (e.g., > 4 nm) to few-layer films (<2.5 nm).\textsuperscript{89} Figure 6A shows the AFM image of the device prepared by transferring mechanically exfoliated PtSe\textsubscript{2} nanoribbon from bulk crystal onto a Si/SiO\textsubscript{2} (270 nm) substrate.\textsuperscript{35} The device with 6.45 nm PtSe\textsubscript{2} shows plateaus with two different resistance values in Figure 6B, which are assigned to the characteristic response of antiferromagnetic ordering. In contrast, a hysteresis loop is observed in the device with an extra layer, revealing a ferromagnetic response of the 7.05 nm PtSe\textsubscript{2} (Figure 6C). As the thickness decreases down to bi- and mono-layer limit, a ferromagnetic-to-antiferromagnetic crossover is observed in the semiconducting PtSe\textsubscript{2} (Figure 6D–F).\textsuperscript{36} DFT calculations suggest that surface Pt vacancies (\(V_{Pt}\)) trigger the thickness-dependent magnetic ordering. In the PtSe\textsubscript{2} ML, a magnetic
The local magnetic moments in the top and bottom Se planes are oriented antiparallel to each other (Figure 6G). For the bilayer case, a net magnetic moment of 1.33 μB is induced by a Pt vacancy, which resides on the outmost Se atoms surrounding the defects, and forms a ferromagnetic configuration as shown in Figure 6H. In the metallic multilayers, only the V_{Pt} defects located at the surface act as magnetic centers with a local magnetic moment of ~1.2 μB. The sign of the magnetic exchange coupling can be switched by adding or removing a single PtSe_{2} layer, thus realizing the ferromagnetic-to-antiferromagnetic transition, which can be explained by Ruderman–Kittel–Kasuya–Yosida interactions (Figure 6I). These findings indicate that defect engineering is promising for introducing magnetism into nonmagnetic 2D materials.

### 3.2 Doping with metal atoms

Doping 2D nonmagnetic materials with magnetic atoms is an alternative route to induce magnetism in the material. It has been theoretically and experimentally demonstrated that robust ferromagnetic ordering can be achieved in 2D TMC systems (e.g., MoS_{2}, MoTe_{2}, WS_{2}, WSe_{2}, and SnS_{2}) through the substituted doping of transition metal atoms (e.g., V, Cr, Mn, Fe, Co, Ti, Ta, and Ni)\(^{37,38,90-97}\). In particular, room-temperature ferromagnetism has been observed in ML V-doped MoTe_{2},\(^{93}\) Co-doped MoS_{2},\(^{95}\) V-doped WSe_{2}\(^{37,38}\), and V-doped WS_{2},\(^{97}\) resulting in dilute magnetic semiconductors. The magnetic properties are tunable with dopant concentration. For instance, the high crystalline quality of V-doped WSe_{2} ML with various V dopant concentrations.
(0.5–8 at%) is revealed by the high-resolution scanning transmission electron microscope images shown in Figure 7A–D, and V aggregation is rarely observed even in the sample with 8 at% of V atoms (Figure 7D). MFM reveals a distinct ferromagnetic signature at 300 K for the V-doped WSe₂ samples, which strengthens as the V concentration increases from 0.5 to 4 at% (Figure 7E–G), and becomes quenched as it further increases to 8 at% (Figure 7H). In another study, Yun et al. reported that the ferromagnetic domains in V-doped WSe₂ manifest stripe-like shapes at low doping concentration (0.1 and 0.3 at%), which transforms to polygon shapes at intermediate concentrations (0.5 and 2 at%) and disappears at high concentration (10 at%). The WSe₂ ML retains its semiconducting characteristic with a high on/off ratio of ~10⁵ at 0.1 at% V-doping concentration. Similar trends are also observed in V-doped WS₂ ML, with optimized ferromagnetism at an intermediate V concentration of ~2 at%, which gradually fades with increasing V concentration. DFT calculations suggest that the ferromagnetism originates from the V dopants, and thus is tunable with the V–V distance. The quenching at high dopant concentration is caused by orbital hybridization at closer V–V distance, where the antiferromagnetic coupling becomes dominant and competes with the ferromagnetic coupling between the moderately separated V-atoms.

2D semiconducting TMC materials also display luminescent and ferromagnetic properties at the same time, with potential applications for novel magneto-electric and magnetooptical devices. Unfortunately, the presence of abundant magnetic dopants and defects in

![Figure 7](https://example.com/figure7.png)

**Figure 7** Ferromagnetism in transition metal chalcogenide monolayer induced by doping. (A–D) Atomic-resolution scanning transmission electron microscopy (STEM) image of WSe₂ monolayers (A) without V dopant, and with (B) 1 at%, (C) 4 at%, and (D) 8 at% V dopants. (E–H) Magnetic force microscopy image of (E) pristine, (F) 1 at%, (G) 4 at%, and (H) 8 at% V-doped WSe₂ monolayer. Reproduced with permission: 2020, Wiley-VCH. (I) Atomic force microscopy image and (J) high-resolution STEM image of (Co, Cr)-incorporated MoS₂ monolayer. (K) Photoluminescence spectra and (L) magnetization vs magnetic field (M–H) curves of single-layered pristine MoS₂, Co-MoS₂, and (Co, Cr)-MoS₂. Reproduced with permission: 2019, Springer Nature Limited.
the nonmagnetic TMCs, which induce magnetism, are usually detrimental to luminescence due to the formation of defect-mediated recombination centers and charge excitons (e.g., trions).98–101 This dilemma was solved through the incorporation of two different transition metals into a TMC ML, for example, a (Co, Cr)-incorporated MoS$_2$ ML (Figure 7I–L).95 Figure 7I,J shows the AFM image and the high-resolution STEM image of a (Co, Cr)-MoS$_2$ ML grown on a SiO$_2$/Si substrate by the CVD method, respectively. Co atoms substitute the Mo atoms to activate ferromagnetic coupling, whereas the Cr atoms are adsorbed atop the Mo sites to provide extra nonradiative recombination channels for excitons. As a result, a 35-fold enhancement of photoluminescence intensity (Figure 7K) and a 90-fold enhancement of saturation magnetization (Figure 7L) are observed in the (Co, Cr)-MoS$_2$ ML compared to the pristine one. From a back-gate field-effect transistor device, a positive magnetoresistance of 35% is obtained at room temperature. This strategy can be utilized to tune both the optical and magnetic properties of other 2D materials, with applications in spintronics, valleytronics, and photoelectric devices.

4 | SUMMARY AND OUTLOOK

As presented in this review, significant progress has been achieved during the past few years in the realization of intrinsic 2D magnets in ML form. This expands the scope of novel properties and phenomena in the 2D family, which is still rapidly growing. In particular, 2D TMCs exhibit robust electronic, optoelectronic, and magnetic properties with promising potential applications in next-generation magneto-electric and magneto-optical devices. Various synthesis methods, as well as engineering strategies, have been developed to grow high-quality 2D TMC materials with tunable magnetic properties. Intensive research effort is still needed to address the critical issues as discussed below.

Challenges remain in identifying intrinsic ML 2D magnets with room temperature ferromagnetic and ferrimagnetic ordering. Pushing the transition temperature higher above room temperature is imperative for useful applications. Many TMC materials have been predicted to be robust 2D magnets,22 however, their structural stability, environmental stability, and synthesis feasibility require experimental validation. Although the realization of 2D magnetic TMCs with Curie temperatures at or above room temperature has been demonstrated, for example, in ML MnSe$_2$,13 and Cr$_2$Te$_6$,29 further improvement in the sample quality, as well as stability, is still needed. Many of these 2D materials are nonlayered and possess isotropic covalent bonding in all three dimensions, and thus prefer to grow in 3D mode. Advanced fabrication strategies are needed for the large-scale growth and mass production of such nonlayered material with controllable thickness down to ML. Furthermore, their room temperature magnetic strength should be comparable to or exceed that of current magnetic materials (e.g., Fe and Co), so they can be useful for future flexible wearable devices.

Alternatively, 2D magnetism can be controlled by various extrinsic perturbations such as electrostatic gating and fields, strain, light, pressure, proximity effects, moiré patterns, impurities, defects, and dopants. We briefly introduced examples of the control of magnetic states in the otherwise nonmagnetic TMC materials (e.g., PtSe$_2$ and WSe$_2$) via defect and dopant engineering. Compared to the nonlayered magnetic TMCs, layered materials are more stable, and can be synthesized using scalable procedures. More research effort is needed in developing feasible and controllable strategies to induce magnetic properties in intrinsically nonmagnetic 2D materials.

Another future direction is to explore magnetic 2D heterostructures and the interface-induced magnetic phenomena. Placing 2D electronic materials on magnetic materials (2D or bulk) can effectively spin-polarize and/or valley-polarize the 2D crystals via proximity. If magnetic 2D materials are used in all 2D heterostructures, the electronic properties can be expanded to include multi-functionalities for spintronics and valleytronics. For instance, exchange field-induced Zeeman splitting has been observed in EuS on graphene,102 WSe$_2$ on EuS,103 and WSe$_2$ on CrI$_3$,104 providing a new approach to control the valley properties of 2D crystals. Furthermore, the adsorption of organic molecules on 2D magnets could also be used to nondestructively modulate the magnetic properties. Ferromagnetism has been induced in superconducting NbSe$_2$ treated by hydrazine molecules, evidenced by the observation of negative magnetoresistance and the Kondo effect.105 Theoretically, it has been predicted that the adsorption of organic molecules can effectively modulate the magnetic anisotropy energy of 2D CrI$_3$,106 and the spin-dependent tunneling barriers in VSe$_2$.107 It is expected that the organic/2D magnet heterostructure will play critical roles in determining many important spin-related phenomena at the interface, for example, magnetic anisotropy, magnetic ordering, magnetoresistance, spin selectivity, and so on, via the proximity effect as well as interfacial charge transfer.

Moving forward, we believe that the field of 2D magnetic materials is in its infancy, and will flourish in the coming years. Both intrinsic and extrinsic 2D magnetic TMCs have immense potential for both fundamental
science and device applications, and more extensive studies are needed to advance the field.

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CONFLICT OF INTEREST

The authors declare no conflict of interest. [Correction added on 29 June 2021, after first online publication: Conflict of Interest section has been added.]

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