REVIEWS

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Perspectives on exfoliated two-dimensional spintronics

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Abstract: Magnetic orderings, i.e., the spontaneous alignment of electron spins below a critical temperature, have been playing key roles in modern science and technologies for both the wide applications of magnetic recording for information storage and the vibrant potential of solid state electronic spin devices (also known as spintronics) for logic operations. In the past decades, thanks to the development of thin film technologies, magnetic thin films via sputtering or epitaxial growth have made the spintronic devices possible at the industrial scale. Yet thinner materials at lower costs with more versatile functionalities are highly desirable for advancing future spintronics. Recently, van der Waals magnetic materials, a family of magnets that can in principle be exfoliated down to the monolayer limit, seem to have brought tremendous opportunities: new generation van der Waals spintronic devices can be seamlessly assembled with possible applications such as optoelectronics, flexible electronics, and etc. Moreover, those exfoliated spintronic devices can potentially be compatible with the famed metal-oxide field effect transistor architectures, allowing the harness of spin performances through the knob of an electrostatic field.

Key words: van der Waals magnetic materials; spintronics; two dimensional materials

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1. Introduction

In general, spintronics such as spin valves must involve in their structures multiple layers of magnetic thin films[1–8]. For example, spin-polarized states of magnets can be interfaced into parallel or anti-parallel configurations, giving rise to logic devices thanks to the charge-spin coupling. In the past decades, thin film grown in a vacuum is the main route to fabricating spintronic devices ever since the pioneering discoveries of spin injection and charge-spin interactions[1–9]. However, vacuum-grown thin films require complex facilities, and sometimes stringent lattice match between the target films and the substrates.

Liberating the degrees of freedom of lattice-matching as well as the demanding growth conditions had never been so facile until the day when graphene was exfoliated by simply using a scotch tape[10]. Researchers all over the world rapidly initiated the historical gold mining in the so-called 'wonderland of flatland', i.e., a new yet rational library of more than 5,000 layered compounds on earth[11]. Because of the relatively weak inter-layer van der Waals (vdW) bonding in those materials, they are often referred to as vdW materials. More precisely, most of them are studied at the atomically thin (or the two-dimensional (2D)) limit, in order to reduce the Coulomb screening and to achieve the unprecedented properties that are not possible in their three-dimensional (3D) bulk forms[10, 12–15].

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Magnetic vdW materials, by definition, are consisted of 2D layers with spontaneous spin polarization below their magnetic critical temperatures. A long-range spin ordering was long believed not to be available in 2D as predicted by the early theories based on the isotropic Heisenberg models[16]. Nevertheless, experimentalists, after having exhausted the low-hand fruits in graphene, recently moved their focus to the exfoliation of magnetic vdW compounds, and have claimed unambiguously the observation of magnetism down to the monolayer-limit in several vdW magnets[13–15, 17, 18]. Since the research work on 2D magnetic monolayer reported by two groups almost in the same time in 2017[13, 18], tremendous efforts have been put into discovering new 2D magnetic materials and exploring the spin interactions in them[15, 17–32], making the 2D magnetism one of the most popular issues in both condensed matter physics and material sciences.

Compared to their 3D counterparts, the advantages of magnetic 2D materials can be twofold. Firstly, 2D magnets can be seamlessly assembled with different rotational angles between the layers of different kinds, thus allowing band engineering through tunable moiré super lattices. Secondly, many of the 2D magnets can be implemented into in-plane field effect transistors, as they are semiconducting with Fermi levels gate tunable. It thus largely increases the opportunities for future spintronic applications, in terms of not only the variety of materials but also the emergent physical phenomena that arise from the novel 2D nanostructures. In this review, we will briefly describe the state-of-the-art spintronics based on 2D vdW materials and offer a new perspective by drawing a potential roadmap of vdW spintronics for the next decade.

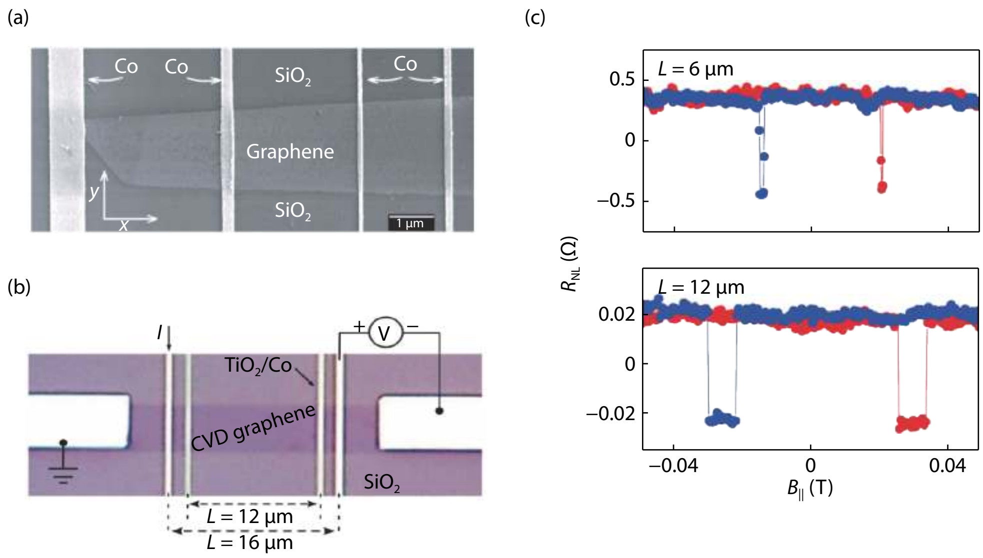

Fig. 1. (Color online) (a, b) Typical spin valve devices made of graphene[33, 34]. (c) The performance of non-local magneto-resistance for CVD graphene spin valve with different channel lengths[34].

2. The 'pre-history' of vdW magnets spintronics

Many of the vdW materials are non-magnetic. However, even in the early days of the 2D materials research, spin-related phenomena have already attracted vast attentions. Indeed, spin injection from magnetic electrodes into non-magnetic substrates can work as an in-plane configuration of spin valves[1–8, 33, 34]. Because of the weak spin-orbit coupling and the resulting long spin diffusion length of up to tens of micron-meters[34, 35], graphene was demonstrated to be an ideal platform for such applications (Figs. 1(a)–1(c)). Transition metal disulfide (TMD) such as MoS_2 is also reported to show similar effect but with a much shorter spin diffusion length[36].

Except for direct spin injections, MoS_2 was also found to be a candidate for spin manipulations in an optical manner due to the spin-valley locking in this specific type of 2D material[37, 38]. Recent experimental evidence showed that a band ferromagnetic state can be achieved with the assistance of a Zeeman gap[39, 40]. Although this ferromagnetism-like behavior cannot occur without a perpendicular magnetic field, it does shed light on the possibility of obtaining the novel type of ordered spins in such 2D systems. When exposed to high magnetic fields, spin-related physics can also be realized in graphene, such as the observation of tunable quantum spin Hall effect[41, 42]. It is noteworthy that, as a semimetal, graphene itself can be implemented for molecular spintronics[43], which in principle is feasible with other 2D materials.

As stated above, before the isolation of 2D intrinsic vdW magnetic materials, spin-related phenomena have been widely studied in a broad range of vdW materials, which can be categorized in the volume of the 'pre-history' of intrinsic vdW magnets.

3. Exfoliated intrinsic vdW magnets spintronics

2D vdW magnets are not at all new materials, while they were just for unknown reasons not quite exfoliated before 2017. Actually, the bulk forms of layered magnetic compounds have been well characterized in terms of crystallographic and spin structures[44–62]. Early few-layered devices were also fabricated and tested, but without systematic examinations as a function of the number of layers down to the monolayer limit. Since 2017 when magnetism in the 2D limit was reported experimentally[13, 18], 2D exfoliated vdW magnetic materials have received renewed research interest from worldwide, opening a new era of long-range spin orderings at 2D.

To date, experimental examinations together with theoretical predictions show that most spin exchange interactions in the 3D scenario (including direct Ising and *XY* interactions, and other indirect interactions) can prevail down to the 2D limit[13, 15, 17–19, 49, 73–93]. According to the existing literatures, we here list the most studied families of 2D magnetic materials, with their fingerprints (such as bandgap, magnetic order, synthesis method, type of spin exchange interactions, critical magnetic temperature, etc.) labeled in Table 1.

The successful exfoliation of vdW magnets is just the beginning of the game, like every new topic in condensed matter physics — people have to find new physics, as well as new applications out of them. In the coming sections, we will discuss a couple of examples of such efforts.

4. Spin valves based on exfoliated vdW magnets

Spin valves have planar configurations as indicated in Section 1 and in Fig. 2(a), as well as vertical configurations that sometimes take the advantage of electron tunneling by sandwiching a tunneling insulator between two ferromagnetic (FM) layers, as shown in Fig. 2(b). This FM-insulator (I)-FM structure is well known as tunneling magnetoresistance (TMR)[2], with maximum magnetoresistance defined as $2P_1P_2/(1-P_1P_2)$ in the Julliere model, while P_1 and P_2 are spin polarization ratios of each layer, respectively.

The first attempt of the vertical spin valve using vdW magnetic materials was realized in 2015, with two pieces of few-layered Fe-doped TaS_2 as the FM layers, while the tunnel layer was oxides formed naturally between their interface (Figs. 2(d) and 2(e)[94]). TMR was reported to be about 7% in this structure, shown in Fig. 2(f). Intrinsic vdW ferromagnet Fe_3GeTe_2 was recently used to serve as FM layers in this very configuration, but with a better-controlled tunnel layer by intercalating an ultrathin hexagonal boron nitride (h-BN) between two Fe_3GeTe_2 flakes. Indeed, TMR can reach as high as 160% at 4.2

Table 1. A list of typical 2D vdW magnetic materials and their magnetic fingerprints.

Material	Bandgap	Magnetic orderings	Way to get	Measurement techniques	Exchange interactions	Critical temperature T_C/T_N	Tunability
CrI_3[13, 14, 20, 63]	1.2 eV	Intralayer/FM Interlayer/AFM FM/bulk	Exfoliated	Magneto-optic Kerr effect (MOKE)	Ising/direct Double-exchange/ super-exchange	64 K/bulk 45 1L	Thickness Gate/ionic liquid electric field
$CrBr_3$[21–23, 46]	2.1 eV/bulk	FM/bulk FM/2D	HQ graphene provided/bulk Exfoliated/1L	Magnetic circular dichroism (MCD)	Heisenberg/direct	35 K/bulk 37 K/3L 36/2L 27/1L	Not available (NA)
$CrCl_3$[24, 57, 64, 65]	3.1 eV/bulk	Intralayer/FM Interlayer/AFM AFM/bulk	Chemical vapor transport(CVT)/bulk Exfoliated/2L	Tunneling	XY/direct	14 K/bulk 17 K/few-layer 16/2L	Thickness
$Cr_2Si_2Te_6$[62, 66–69]	0.4 eV/direct-bulk 1.2 eV/indirect/bulk	FM	Self-flux/bulk Exfoliated/2D	Heisenberg/direct Double-exchange/ super-exchange	32 K/bulk 80 K/2D	Thickness	
$Cr_2Ge_2Te_6$[15, 19]	0.45 eV	FM	Exfoliated	MOKE	Heisenberg/direct	45 K(bulk)	Gate/ionic liquid
Fe_3GeTe_2[17]	0	FM	$A1_2O_3$ assisted exfoliated	Anomalous Hall Effect (AHE)	Ising/direct Itinerate/super-exchange	180 K/bulk 20 K/1L	Thickness Ionic liquid
$FePS_3$[25, 70]	1.5 eV	AFM	CVT	Raman + DFT	Ising/direct	123 K/bulk 118 K/1L	NA
$MnPS_3$[25, 26, 47]	2.4 eV	AFM	CVT/bulk Exfoliated/2D	Physical property measurement systems (PPMS)/bulk Raman	Heisenberg/direct	78 K/bulk	Liquid gating
$NiPS_3$[27, 71]	1.6 eV/indirect >2.4 eV/direct	AFM	CVT/bulk Exfoliated/2L	Raman	XY/direct	155 K/bulk 130 K/2L	NA
VSe_2[17, 29]	0	FM/1L AFM/2L Paramagnetic/bulk	Molecular beam epitaxy(MBE)	MOKE AHE	NA	>300 K	Thickness Electric field
$CrTe_2$[72]	0	FM	Oxidation of $KCrTe_2$	Squid	Itinerate/super-exchange	310 K/bulk	NA
V_5S_8[30]	0	AFM/bulk FM/3.2 nm	Chemical Vapor Deposition (CVD)/10 nm Exfoliated/3.2 nm	AHE	NA	32 K/bulk 2 K/3.2 nm	Thickness
CrSe[31]	NA	FM	CVD	PPMS	NA	208 K	NA
Cr_2S_3[32]	NA	FM	CVD	PPMS	NA	120 K	NA

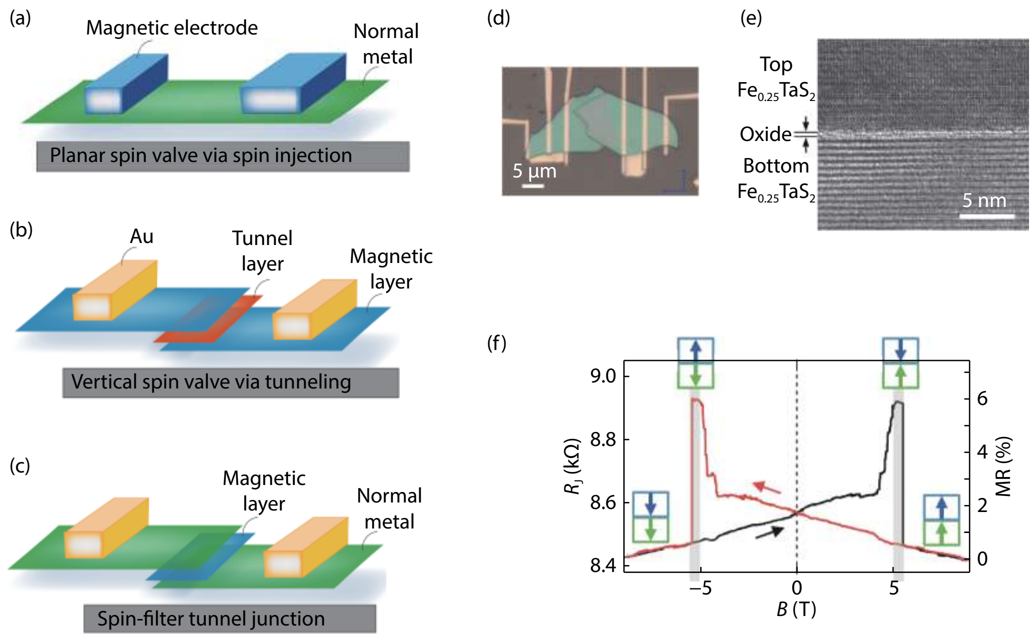

Fig. 2. (Color online) (a, b) Schematics of configurations for 2D spin valve devices, and (c) 2D spin filter tunnel junction (sf-TJ). (d–f) The first spin valve demonstrated using 2D vdW magnetic (Fe-doped TaS_2) materials[94].

K[95].

Magnetic vdW materials can be easily exfoliated on a desktop and assembled into spintronic devices, which are free of epitaxial technology and in principle can be mass-produced by means of chemical vapor deposition (CVD) etc. It thus opens up a totally new page and shall give a profound impact on the future of the spin-valve industries. Nevertheless, the experimentally-demonstrated spin valves are, so far, working far below room temperature. The transfer/stacking method is yet to be optimized for batch production, which will also be discussed from a broad perspective in the last section of this review.

5. vdW magnetic tunnel junctions

Besides the FM–I–FM configuration, metal–FM–metal configuration, which has the insulating FM sandwiched between two metals, has also been used as spintronic device, often referred to as spin filter tunnel junction (sf-TJ), as shown in Fig. 2(c). As given in Table 1, many of the vdW magnetic materials (including CrI_3, $CrBr_3$ and etc.) have semiconducting gaps, and usually are quite insulating at low temperature, thus providing a unique chance for the study of 2D vdW sf-TJs and related tunneling physics.

Indeed, several experiments have confirmed that multilayered-CrX_3 (X = I, Br, Cl) sf-TJs have extremely large magnetoresistance up to ~ 106%[21, 23, 24, 96–100]. Magnon-assisted tunnel spectroscopy was also studied in $CrBr_3$ sf-TJ devices[21]. Compared with non-magnetic tunnel barriers within the same structure, a spin-polarized tunnel layer can provide new mechanism for electron tunneling via magnon-emissions instead of phonons or localized state in the former case[101–105].

In addition to the applications stated above, sf-TJs made of vdW magnetic tunnel layers can serve in a new kind of vertical spin-related field effect transistor. In this scenario, transparent few-layered graphene gate electrodes are often equipped within the structure illustrated in Fig. 3(a), in the sense that optical probe can penetrate the graphene gate without catching any parasitic magnetic signals but the ones from the vdW magnetic tunneling layer itself. During measurements, gate voltages can be seamlessly applied, pumping electron in and out of the band structure of the target vdW magnet and sequentially affecting the magnetic parameters including coercivity, Curie temperature, and etc.[15, 107].

It is noteworthy that, so far, investigations on sf-TJs that utilize antiferromagnetic vdW layers as tunnel barrier remain scarce. To some extent, both sf-TJs (Fig. 2(c)) and TMRs (Fig. 2(b)) can be used as logic unit and magnetic sensors[107–109]. Again, magnetic critical temperatures up to room temperature of those 2D vdW magnets are deadly desired at the current stage.

6. Planar vdW magnetic field effect transistors

Not quite similar to the sf-TJ with insulating vdW magnetic tunnel layer mentioned above, a real planar structure that mimics the famed metal-oxide field effect transistor (FET) can also be achieved in vdW magnets. This will require naturally a semiconducting channel, or at least a conducting channel whose electron density of states can be gate tuned, via electrostatic or liquid gate techniques.

A series of vdW materials $Cr_2M_2Te_6$ (M = Si, Ge) compounds were reported to be intrinsic magnetic semiconductors, with reported band gaps varying in the range of 0.4–1.2 eV[15, 19, 62, 66–69]. It thus makes them ideal platforms to investigate FETs based on their exfoliated thin flakes. Several iterations of vdW spin-FET devices are illustrated in Figs. 4(a)–4(c), while the earliest experimental attempt can be traced back to 2016, as shown in Fig. 4(a)[62]. However, due to the air-instability, those devices made in air showed relatively poor performance[62, 110], while h-BN protection in the inert gas atmosphere significantly improved the gate tunability of electron charge and spin, as shown in Figs. 4(e)–4(g)[15].

It was known that even conventional metallic thin Fe, Co films can manifest tunable magnetic parameters via ionic gating[111–113], by two orders of magnitude stronger gate effi-

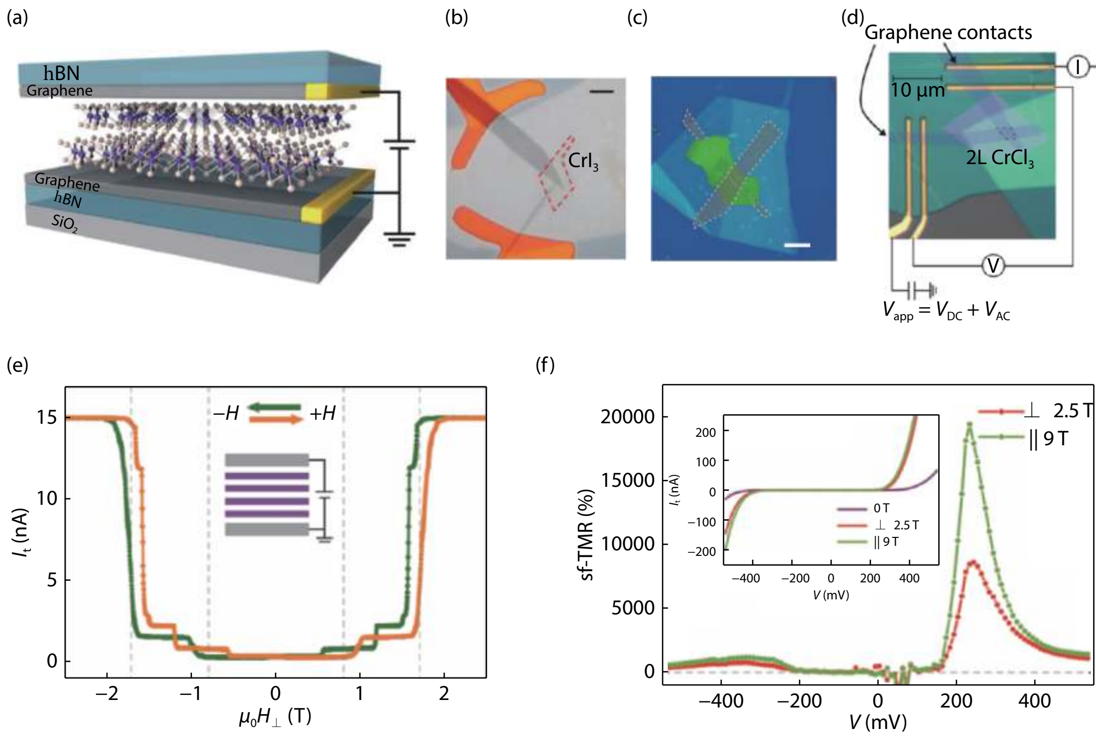

Fig. 3. (Color online) (a) Schematics of CrI_3 sf-TJ[96]. (b-d) Optical images of several iterations of vdW 2D sf-TJ devices since 2017[96, 99, 106]. Notice that all of them have very small junction area possibly to reduce the number of magnetic domains. (e, f) The magneto-tunneling current and spin-filtered magnetoresistance for a four-layered CrI_3 sf-TJ device[96].

ciency compared to the Si gate[15]. Similar effect was observed in a metallic phase few-layered Fe_3GeTe_2 down to the monolayer limit[17]. Strikingly, Curie temperature of a four-layered Fe_3GeTe_2 can be enhanced to over 300 K using the ionic liquid gating technique[17]. Great promises are therefore held that room-temperature vdW magnetic transistor may be a key to revolutionize spintronics in the future. To note that recent reports have shown geometry effect (nano-patterning) can also enhance the magnetic ordering temperature up to room temperature in thin flakes of Fe_3GeTe_2[114].

Up to now, studies on vdW magnetic semiconducting FETs are still on the go. There is plenty of room to improve the performances, including the working temperature, and the interface between FM and AFM vdW semiconductors, and etc.. While vdW FM semiconductors are of great interest for planar FETs, it is noteworthy that there is the family of vdW AFM semiconductors, which are also promising for planar FETs, as well as opto-electronic applications.

7. Current-driven switching of vdW magnets

About three decades after the theoretical predictions[115, 116], it was experimentally confirmed that due to the spin-orbit coupling (SOC), a transverse spin current can be generated in a non-magnetic system via an unpolarized electrical current, which is referred to as the famed spin Hall effect (SHE)[117, 118]. Spin Hall and inverse spin Hall effects are known to have fueled many important manifestations on spintronic devices probed by both electrical and by optical detection techniques[119, 120].

When interfaced with a magnetic layer, the SHE in the non-magnetic layer with large SOC can exert an orbit torque (SOT) on the magnetic layer that can switch the direction of magnetization, thus giving rise to a current-driven spin flip. During the switching process, a small external magnetic field colinear with the current is required[122], unless the lateral structural symmetry is broken[121]. The SOT devices show much-improved energy efficiency as compared to spin transfer torque (STT) technique, and thus are of great importance for magnetic memory applications. As discussed at the beginning of this review, owing to the advantages of 2D materials, it is believed that the vdW magnets can serve as a new material-base for the SOT multi-layer heterostructures[123].

Indeed, the reports in 2019 indicate that it is absolutely feasible to replace the conventional magnetic film with vdW ferromagnets in the SOT structures[124, 125]. As shown in Figs. 5(a), 5(b), and 5(e), few-layered Fe_3GeTe_2 (FGT) was chosen to be interfacially coupled to a metallic Pt thin film, thus giving rise to a non-magnetic/vdW-magnetic interface. With the help of a small in-plane magnetic field, the out-of-plane magnetization of FGT can be switched at a critical current, indicated in Figs. 5(c), 5(d), 5(f), and 5(g)[124, 125].

At the current stage, SOT devices using vdW magnets as a platform are still far from mature to meet the applicational standard, such as free of an external magnetic field, low critical switching current density, and etc. Apparently, room temperature operation is needed, and the SOC layers are so far still conventional metallic thin films. There is thus a long way to go to address the above points to push the vdW SOT devices towards real applications.

It is worth noting that spin Hall effects can be also observed via magneto-optical Kerr measurements as experimentally evidenced in semiconducting TMDs[126]. Therefore, from both fundamental and application points of view, it is believed that almost all-vdW materials-based SOT devices are possible. And thanks to the electronic band gaps often existing in either TMDs or vdW magnets, gate-tunable multi-functional

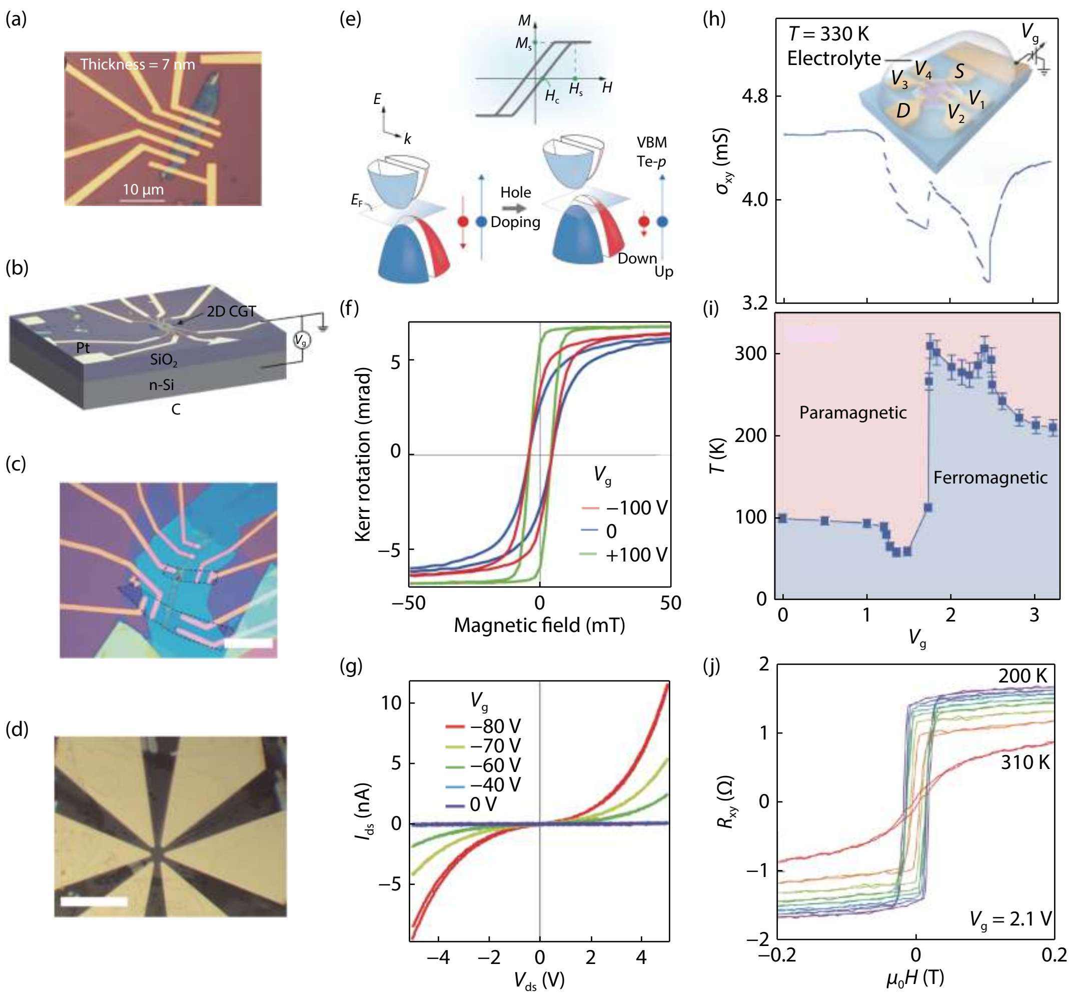

Fig. 4. (Color online) Optical image of several versions of spin-FETs based on magnetic vdW materials (a) semiconducting $CrSiTe_3$[62], (b) semiconducting $Cr_2Ge_2Te_6$[110], (c) h-BN encapsulated $Cr_2Ge_2Te_6$ (red and black dashed lines label the edge of $Cr_2Ge_2Te_6$ and graphene electrodes, respectively)[15], and (d) Al_2O_3-assisted exfoliated 4-layered metallic Fe_3GeTe_2[17], respectively. Scale bars in (c) and (d) are 10 and 100 μm, respectively. (e) Schematic of the tunable Fermi level and simplified spin-polarized band structure of the vdW intrinsic magnetic semiconductor[15]. (f, g) Gate tuned magnetic hysteresis loops and gate-tuned *I–V* curves of the few-layered $Cr_2Ge_2Te_6$ planar FET device[15]. (h, i) Longitudinal conductivity and Curie temperature of the Fe_3GeTe_2 planar FET as a function of ion liquid gate[17]. (j) The anomalous Hall curves of the ionic-gated Fe_3GeTe_2 planar FET at different temperatures[17].

magneto-optotronics and/or memories will have profound impacts in the next generation spintronics.

8. Other possibilities and an outlook of vdW spintronics

In the previous sections a couple of typical vdW spintronic devices such as spin valves, spin filter tunnel junctions, and planar spin FET have been introduced, and we now come to a brief discussion on several other configurations of spintronic devices using vdW magnets as a platform. In Fig. 6, a variety of nanostructures for vdW spintronics are illustrated. It can be seen that spin-related electronic devices can in principle be built via a mechanical stacking method, giving rise to possible applications such as 2D heterostructure of multi-ferroics, vdW magnetic recording, and topological magnetic states, etc.. Up to now, the emerging phenomena in 2D vertical multi-ferroics as well as topological magnetic states in the vdW systems are attracting great interests. We now propose a roadmap for exfoliated spintronics, as indicated in Fig. 7. In short, depending on the development of vdW materials, the future trend of the vdW spintronics can be classified into fundamental- and application-oriented directions.

For fundamental research, the future tasks will be looking for new emerging phenomena including topological magnetic states (Skyrmion[127] as an example), magnetic semiconductors and the associated devices such as magnetic p–n junctions, 2D multi-ferroic devices via the stacking of vdW layered materials, the novel devices for superconducting spintronics (such as Pi junction[128], etc.) based on vdW magnets, novel interfacial coupling using layered FM/AFM compounds, as well as NMR/ESR and Rashba effects in 2D vdW systems.

For application research, the top priority will be to find room temperature vdW magnets. And the mission for future application shall include spin torque transfer (or spin Hall related) devices, spin diodes, spin valves, vdW magnetic semiconductors (as compared to the bulk diluted magnetic semiconductors[129–133]), magnetic electronic sensors, data storage devices (such as optical/electrical writing of domains, exchange bias effects that may be gate tunable, magnetic vdW electronics for biological utilities, etc.).

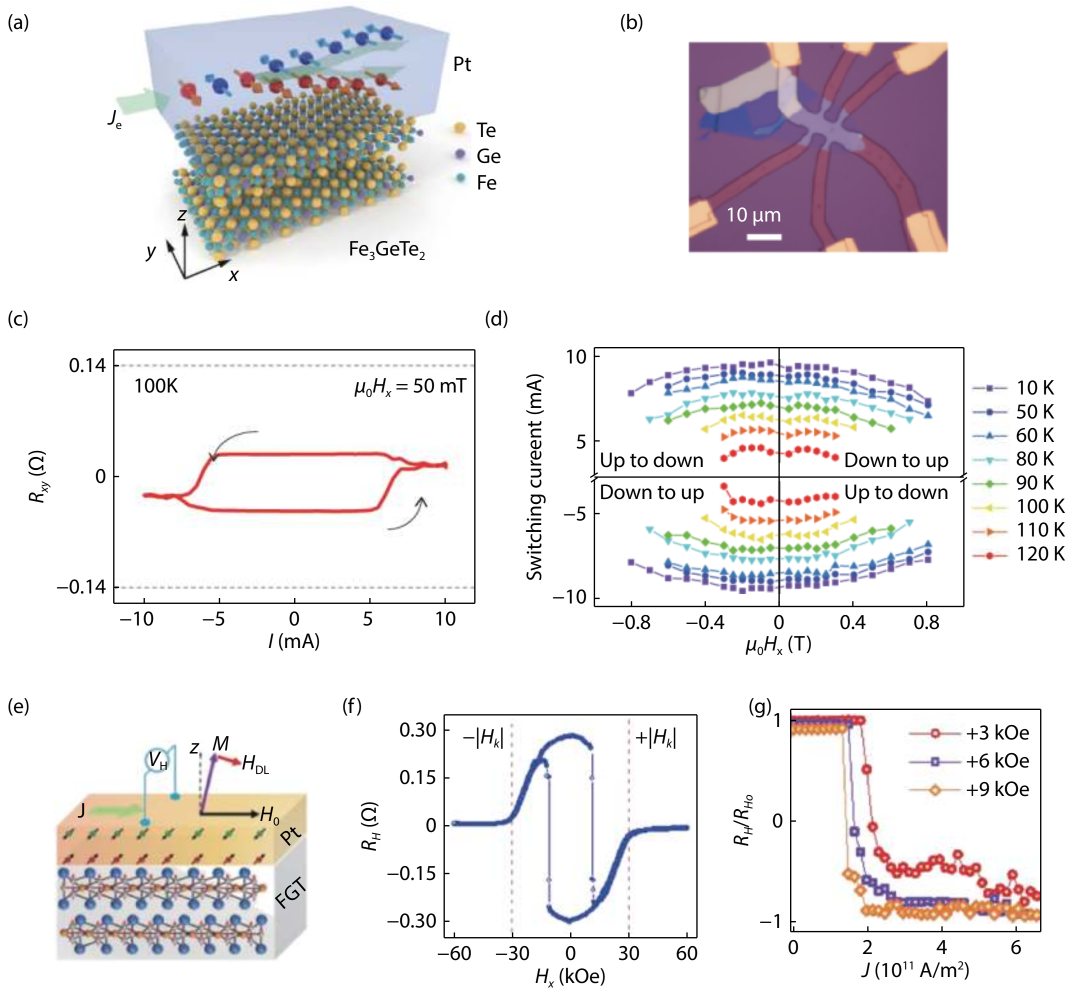

Fig. 5. (Color online) (a, b) Schematic and optical image of a typical Pt/FGT device[124]. (c) Hall resistivity recorded as a function of current flowing in the 2D vdW heterostructure device. A hysteresis loop can be seen, demonstrating the current-driven magnetic switch of the magnetizations in the FGT layer[124]. (d) Switching current as a function of externally applied in-plane magnetic fields at different temperatures[124]. (e) Schematic structure of Pt/FGT device[125]. (f) Anomalous Hall effect curve of the Pt/FGT device[125]. (g) Current-induced magnetic switch at different external magnetic fields[125].

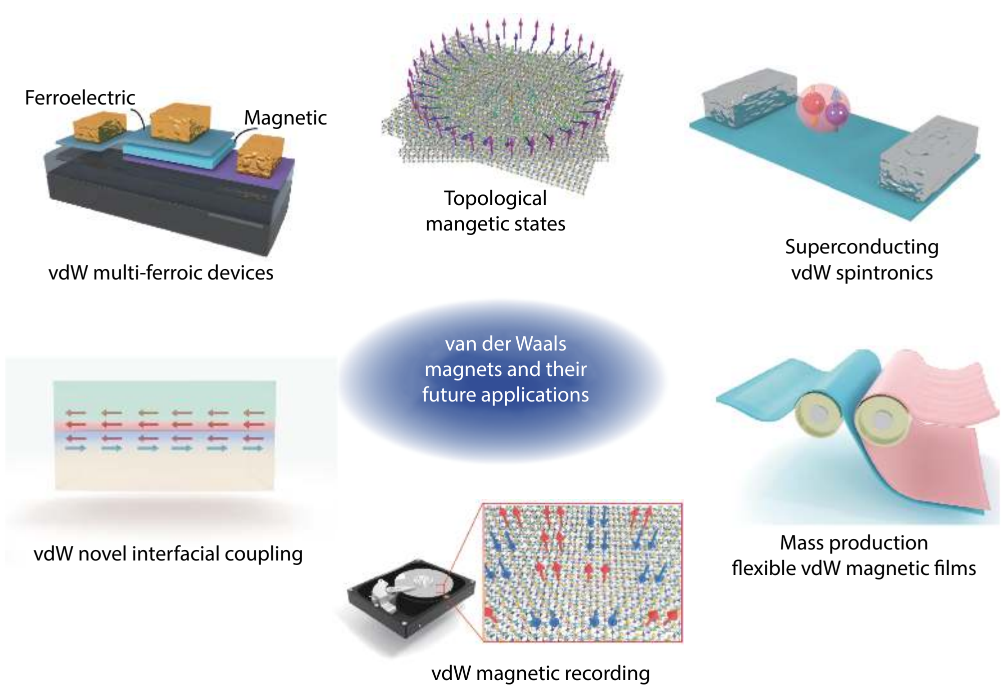

Fig. 6. (Color online) Illustration of different nanostructures for vdW spintronics.

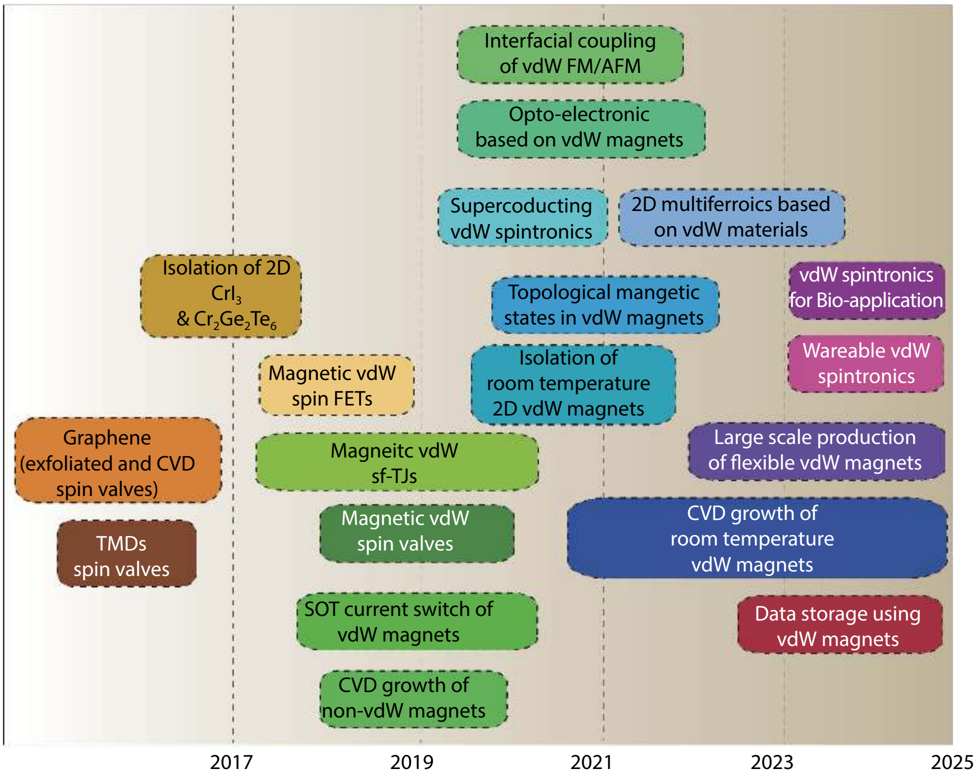

Fig. 7. (Color online) A roadmap for the exfoliated spintronics.

Finally, we would like to recall the fact that, in principle, vdW magnetic materials are compatible with mass production processes such as CVD methods[134]. For example, CVD grown non-vdW magnetic thin films have been demonstrated recently[31, 32]. It is reasonable to believe that flexible spintronics can be realized in the very near future. Layered magnetic materials thus hold great promises for spintronics made in a totally new manner, i.e., atomic layer by atomic layer, with an infinite combination of the rotation angle, and unlimited possibility of assembling sequence, that will truly revolutionize our daily life.

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