Large-area fabrication of 2D layered topological semimetal films and emerging applications

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ABSTRACT

Topological semimetals represent a new class of topological materials, which are highly desirable for both physics frontier and electronics applications owing to their nontrivial band structures and topologically protected surface states. The large-area fabrication of high-quality topological semimetal films is the prerequisite step to realize their practical applications. Its progress has located in its infant period. In this mini-review, we summarize several typical techniques for the fabrication of large-area 2D layered topological semimetal films. The recent progress in these large-area films for electronics, optoelectronics, terahertz, and spintronics applications is briefly reviewed. It is anticipated that with the rapid development of scalable, reliable, and low-cost production techniques and improved functional realization, large-area 2D layered topological semimetals would find the wide commercial applications in electronics, energy and beyond.

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1. Introduction

Materials are traditionally classified into three categories of conductor, semiconductor, and insulator according to their electronic properties [1]. Topological materials, as a new category of condensed matter, have evidenced new discovered quantized electrical transport phenomena such as quantum Hall effect (QHE) [2] and quantum anomalous Hall effect (QAHE) [3,4]. The topological nontrivial band structures of topological insulators (TIs) or topological semimetals (TSMs) endow them with novel surface states beyond the bulk ones. In TIs, the conduction and valence bands are inverted by strong spin–orbit coupling (SOC) [5], forming gapless surface states with spin-momentum locking [6]. Such surface states were firstly predicted and demonstrated to exist in HgTe/CdTe quantum wells [7,8] and later found in other TIs like Bi₂Se₃ [9], Bi₂Te₃ [10] and Sb₂Te₃ [11]. The band structure of TIs is topologically protected and can be described by topological invariant $Z₂$ [12].

Studies [13–18] of Dirac semimetals (DSMs) and Weyl semimetals (WSMs) boosted significantly after the exciting discoveries in TIs. In a DSM, two doubly degenerate bands cross in the Brillouin zone, forming a fourfold degenerate Dirac point at the cross point with linear dispersion in all three directions of the momentum space, as shown in Figure 1(a). Dirac fermions, acting as the low-energy excitation of DSM, can be described by the Dirac equation, as seen in Na₃Bi [19] and Cd₃As₂ [20,21]. Either broken time-reversal symmetry or spatial inversion symmetry can change a Dirac point into two Weyl points. Such Weyl points always come in pairs with opposite chirality and can be alternatively defined as monopoles and anti-monopoles of Berry curvature [22]. Bands related to topological surface states in WSMs display Fermi arc between two Weyl points, as shown in Figure 1(a). DSMs/WSMs can be divided into two types according to the shape of the Dirac/Weyl cone, which is constrained by the lattice symmetry [23]. For instance, Weyl cones in type-I WSMs are untitled or weakly tilted, accompanied with a point-like Fermi surface when the energy is placed at the type-I Weyl point. In contrast, type-II WSMs host strongly tilted Weyl cones with a Fermi surface consisting of electron and hole pockets that touch at the type-II Weyl points [24], as shown in Figure 1(b). In addition to DSMs and WSMs, nodal-line semimetals have been proved to possess 1D Dirac line or loops close to Fermi level in the momentum space, such as ZrSiX ($X = S, Se, and Te$) family [25–27]. Recently, two dimensional (2D) layered TSMs have gained intense attention due to their exotic electronic properties and controllable crystalline structures [28,29]. In layered structures, metal layers are sandwiched by adjacent chalcogenide layers and these sandwich structures stack along the c axis and interact with van der Waals forces, as shown in Figure 1(c). According to different stacking modes, layered structures can be divided into hexagonal (2H), trigonal (1T),
orthogonal ($T_d$) and distorted trigonal ($1T'$) structures. The rich crystal structures for different TSMs, such as $T_d$ WTe$_2$ [30,31], $T_d$ and $1T'$ MoTe$_2$ [32,33], $1T$ PtTe$_2$ [34] and $1T$ PdTe$_2$ [35], have provided an excellent platform for studying superconductivity [36,37], spintronics [38–42] and valley-optoelectronics [43].

Previous studies on 2D TSMs mainly based on individual films or heterostructures produced by mechanical exfoliation and stacking, which are limited to the small lateral size with poor yielding and not suitable for practical application. To resolve this problem, a variety of techniques have been developed to realize scalable production of large-area, high-quality 2D layered TSM films with controllable thickness. Chemical vapor deposition (CVD) is a universal method to prepare 2D layered films. Using metal films as precursors, centimeter-scale films of various TSMs can be facilely achieved, including WTe$_2$ [44], MoTe$_2$ [45–48] and PtTe$_2$ [49]. In 2017, Tang et al. [50] verified a quantum Hall state in monolayer $1T'$-WTe$_2$ grown by molecular beam epitaxy (MBE).
Moreover, other methods, such as pulsed laser deposition (PLD) [51–53], sputtering [54], atomic layer deposition (ALD) [55] and metal-organic CVD [56,57] are also developed to fabricate the high-quality 2DTSM films and related heterostructures.

Note that a couple of recently published reviews have fully described the development of topological semimetals [23,24,58,59]. In this mini-review, we shall provide an overview of recent progress in large-area 2DTSM films. We firstly introduce several techniques to fabricate large-area films. Secondly, we summarize the novel electronic properties and potential applications of 2DTSMs. Finally, we will briefly discuss the existed problems and point out the prospects in the future applications of 2D TSMs.

2. Large-area fabrication methods

Although tremendous efforts have been devoted to preparing large-area 2D material films in the last decade, the scalable production of TSM films has not achieved enough attention until more recently. Thanks to numerous successful precedents, the large-area fabrication of TSM films is developing rapidly. At the same time, some unique problems are encountered. In this section, we will extendedly discuss the chemical and physical methods that have been widely employed, and give a brief introduction to the strategies which are still in their infancy to prepared large-area 2D TSM films.

2.1 Chemical vapor deposition

In traditional CVD method, powder precursors sublime into gas state and react on the surface of substrates to form films. Although numerous layered material films have been synthesized by this method, it is difficult to grow large-area aligned films. Since most layered TSMs contain Te element, it is a facile method to prepare large-area TSM films by tellurizing a pre-deposited metal or metallic oxide film in a CVD furnace. The pre-deposited films can be prepared by sputter, EBE, ALD or thermal evaporation. 1T’ and 2H MoTe$_2$ films can be selectively synthesized by carefully choosing metal film precursor and controlling the growth condition. Zhou et al. [46,47] reported that 1T’ MoTe$_2$ films tended to be obtained under conditions of insufficient Te and with better uniformity from MoO$_3$ precursors. The transformation process from MoO$_3$ to 1T’/2H MoTe$_2$ was clearly investigated and could be regulated by controlling the carrier gas flow rate and tellurization temperature [48,60,61]. To enhance the reactivity of Te and make tellurization process more easily, H$_2$ gas was introduced to form intermediate hydrogen telluride [62], as shown in Figure 2(a). Meanwhile, thickness of the films after tellurization were determined by
controlling the initial thickness of precursor MoO$_x$ films [62]. Besides, the Te-rich environment can also be obtained by directly placing Ni$_x$Te$_y$ films [63] or directly depositing Te thin films [64] on the surface of transition metal film. Compared with Te powder sublimation, these contact methods result in a more homogeneous tellurization. Similar to MoTe$_2$, CVD growth of large-area WTe$_2$ films has been widely reported [44,65] by using W films and Te powder as precursors.

PtTe$_2$ and PdTe$_2$ are proved to be Dirac semimetals by ARPES and can be prepared by tellurizing metal films. CVD-grown PtTe$_2$ films tend to be polycrystalline [49]. Further study showed a transition of 2D PtTe$_2$ films orientation with increasing thickness, which could affect the electrical conductivity of the films [66]. Wafer-scale PtTe$_2$ thin films were prepared by annealing pre-deposited Pt films on SiO$_2$ substrates in tellurium vapor as shown in Figures 2(b) and 2(c) [67]. The half-open quartz tubes were utilized to effectively reduce the particle loss and potentially reduce heat loss. High-quality PtTe$_2$ films with giant spin hall conductivity were
obtained by this method. Beside SiO₂, PtTe₂ films were grown on a wide range of substrates [68], such as Si, Al₂O₃, glass, etc., including NaCl which can be further dissolved to obtain free-standing films as shown in Figure 2 (d). Also, the selective preparation of PtTe and PtTe₂ phase can be realized by suitably controlling Te steam and the temperature [68]. Unlike other 2D materials, monolayer PtTe₂ and PdTe₂ films are hard to achieve through exfoliation due to their strong interlayer interaction, but can be prepared by directly tellurizing Pd(111) and Pt(111) single crystals [69].

2.2 Molecular beam epitaxy

MBE is a versatile growth technique to prepare large-area and high-quality crystalline 2D films. Compared with CVD, it offers high flexibility in chemical components, precise control of the film thickness and the ability to grow heterostructures and superlattices with abrupt interfaces. Ultrahigh vacuum (background vacuum up to 10⁻¹⁰ mbar) in the growth chamber during the growth process ensures the purity of the film. Besides, multiple characterization techniques can be integrated with MBE to in situ monitor the growth process and investigate electronic structures, such as reflection high-energy electron diffraction (RHEED) and angle-resolved photoemission spectroscopy (ARPES). Because of these advantages, MBE can be employed in the growth of almost all TSMs like PtTe₂ [70–72], PtSe₂ [73], PdTe₂ [74] and ZrTe₂ [75], etc.

Among WSMs, MoTe₂ is an interesting material with its hexagonal (2H) phase as a semiconductor and monoclinic (1T’) phase as a type-II WSM. The small energy difference (35 meV per unit) between these two phases makes the selective synthesis of pure-phase films very laborious. 2H-MoTe₂ is slightly more stable under ambient conditions and easier to achieve compared with 1T’-MoTe₂. Figure 3(a) is a schematic illustration of MBE growth of MoTe₂. High-purity Mo and Te molecular beams are evaporated by an e-beam evaporator and a Knudsen cell, respectively [76].

MBE growth of 2H MoTe₂ was reported on a variety of substrates, such as c-Al₂O₃(0001) [77], MoS₂ [78], CaF₂ [79] and GaAs [79]. The similarity of 2H and 1T’ MoTe₂ often results in a mixture of these two phases. Monolayer films of mixed 2H and 1T’ MoTe₂ were grown on highly oriented pyrolytic graphite (HOPG) substrates [80]. The ratio of 2H to 1T’ phase can be tuned by changing the growth temperature and Te fluxes. Besides, lateral hetero-phase homojunctions of 2H and 1T’ MoTe₂ were observed in MBE grown films, which may have potential in lateral homojunction electronic devices [81]. Later, Tang et al. [82] reported a successful growth of high quality monolayer 1T’ MoTe₂ on bilayer graphene terminated 6H-SiC(0001) and confirmed its semimetal properties by ARPES. The 1T’ MoTe₂ will normally transfer to the Tₐ phase at a low temperature below 250 K. However, Tₐ
MoTe$_2$ can be stabilized at room temperature by growing on InAs(111)/Si (111) substrates [83]. Theoretical and experimental results show that the strain caused by the substrate plays an important role in phase stabilization and band structure engineering [84]. Another key factor in stabilizing the structural phase is the growth temperature [85]. According to He et al. [76], the 2H MoTe$_2$ was observed at a growth temperature between 345–365°C, whereas the 1T’ MoTe$_2$ appeared at a higher temperature between 380–450°C, as shown in Figure 3(b) –(c).

WTe$_2$ is another type-II WSM and has attracted much attention on its MBE growth. High quality 1T’ WTe$_2$ films have been successfully grown on 2D (Bi$_2$Te$_3$, MoS$_2$, and graphite) [86] and 3D (c-Al$_2$O$_3$) [87] material substrates. Besides, monolayer 1T’ WTe$_2$ was predicted and confirmed to be a quantum spin Hall insulator with topological protected edge states and its epitaxial growth has also been widely reported [88–90]. Figure 3(d) shows the RHEED patterns of the graphene substrate and monolayer WTe$_2$ grown by MBE [50], demonstrating the layer-by-layer growth mode. A pure 1T’ WTe$_2$ phase with a small angle distortion is verified by further angle-integrated core level photoemission combined with scanning tunneling microscopy characterization [50], as shown in Figure 3(e) –(f).

**Figure 3.** MBE growth of MoTe$_2$ and WTe$_2$ films. (a). Schematic diagram of the MBE growth setup for MoTe$_2$. Reproduced from [76]. (b). Growth process control of the MBE growth of 1T’ and 2H MoTe$_2$. Reproduced from [76]. (c). Raman spectra of the MoTe$_2$ films grown at different temperatures. Reproduced from [76]. (d). RHEED patterns of graphene substrate (top) and sub-monolayer 1T’ WTe$_2$ (bottom). Reproduced from [50]. (e). Core level spectra of 1T’ WTe$_2$. Reproduced from [50]. (f). Atomically resolved STM image of 1T’ WTe$_2$. Reproduced from [50].
2.3 Pulsed laser deposition

The PLD technique utilizes a focused high-energy pulsed laser to bombard the target and convert the solid material of a small area into the plasma state, as called the plume. The focused laser pulse can raise the temperature of the micro-region up to ten thousand degrees in a very short time. As such, the PLD technique is very suitable for depositing high-melting-point materials and has no composition selectivity. PLD has many advantages for depositing 2D materials, such as high growth rate, clean fabrication process and relatively low growth temperature [91]. Meanwhile, it allows homogeneous growth of high-quality centimeter-scale films. Besides, the thickness of the films can be readily controlled by changing the number of laser pulses, making PLD very suitable for industrial production.

PLD has promoted the development of photodetectors based on 2D materials like 2H-MoTe$_2$ [51]. In the last few years, several 2D TSM films [92,93] have been produced by PLD. Figure 4(a) shows a two-step synthesis to prepare WTe$_2$ thin films [94]. The amorphous WTe$_2$ films were firstly deposited on cleaned mica by PLD method, and then sealed in a quartz tube with the added Te powder and annealed at 700°C for days to crystallize. The raman spectrum in Figure 4(b) shows good crystallinity of the films. Distinct Shubnikov-de Haas Oscillations [94] and tunable magnetoresistance [52] were observed in these large-area films, even after transferring to other substrates [95]. However, this two-step method is time-consuming and easy to introduce impurities during the annealing process. Very recently, a single-step method was reported. single-crystal-like 1T’ WTe$_2$ films were successfully grown on Bi$_2$Te$_3$ seeded layer by using single-crystal target at a low temperature (210°C) [96]. Clear crystalline structures and abrupt interfaces of multilayered Bi$_2$Te$_3$-WTe$_2$ heterostructures can be seen in the scanning transmission electron microscopic image in high-angle annular dark field (HAADF) mode, as shown in Figure 4(c) –(d). In addition, topological material candidate ZrTe$_2$ films were grown on (110)-oriented SrTiO$_3$(STO) substrates and protected by in-situ deposition of a AlN capping layer [97]. A large magnetoresistance (∼3000% at 2 K and 9 T) was observed in these films, which could benefit their application in magnetoresistance type sensors.

In conclusion, TSM films grown by PLD have developed rapidly in recent years. They exhibit high-quality structures and exotic electronic properties for electronic and optoelectronic devices. Meanwhile, there is still plenty room for developing PLD technique for other TSM films beyond the discovered ones. We believe that with increasing attention, this technique will significantly promote the commercial application of TSMs in the future.
Sputtering is a widely used technique for the deposition of both metal and alloy thin films and has been utilized in numerous industrial productions. During sputtering deposition, glow discharge is first created in inert gas to produce energetic electrons and ions. The electrons will collide with gas atoms and create secondary electrons and more ions to maintain the discharge. Then the created ions of the inert gas are accelerated toward and subsequently bombard the target. As a result, the sputtered species will leave the target and carry enough kinetic energy to reach the substrate and form films. Sputtering deposition is an ideal technique for the fabrication of TSM films due to its high deposition rate and wafer-scale production. Sputtered WTe₂ and MoTe₂ films have been used to study pulsed laser generation [98,99] and spin-to-charge conversion [100,101].
In 2017, Huang et al. [102] reported a successful growth of MoTe$_2$ films by sputtering. As shown in Figure 5(a), with the protection of the SiO$_2$ layer, the as-deposited MoTe$_2$ films with low crystallinity initially form 1T’ structure and convert gradually to 2H structure over a prolonged annealing time at 650°C. The transition process of as-deposited films was also clearly verified in another report [103] as shown in Figure 5(d). Also, co-sputtering technique [54,104] was used to grow 1T’-MoTe$_2$ films under low temperature. As shown in Figure 5(b), Mo and Te atoms were co-deposited onto graphene surfaces at room temperature. The sticking coefficients of Mo and Te were investigated by changing the substrate temperature (Figure 5(c)). Further ex situ thermal annealing in the Te environment could increase the stoichiometry of MoTe$_2$ films [54]. As evidenced by numerous reports, the post-annealing plays an important role in improving the crystallization and stoichiometry of sputtered TSM films.

2.5 Comparison of different fabrication methods

Table 1 is a list of representative reports of fabricating large-area 2D layered topological semimetal films. The growth temperatures are significantly different in different preparation methods. The growth temperature of the CVD method is relatively high, ranging from 500–
800°C, while that of MBE, PLD and sputtering is relatively low, about 300°C. In the choice of the substrate, the CVD method has no special requirements for the substrate, so silicon-based substrates are usually used for industrial compatibility. The MBE method usually uses a substrate with a matching lattice structure to achieve epitaxial growth, such as SiC (0001) and a two-dimensional material substrate. The as-deposited films grown by PLD and sputtering tend to be amorphous, but after annealing at high temperature, the crystalline properties can be significantly improved.

**Table 1. List of large-area growth conditions for 2D layered topological semimetals.**

| Material | Phase | Preparation method | Precursors or target | Growth temperature | Substrate | Ref  |
|----------|-------|-------------------|----------------------|-------------------|-----------|-----|
| WTe₂     | Td    | CVD               | WO₂, film, Te       | 600°C             | SiO₂      | [44]|
| 1T       | CVD   | W film, Ni₃Te₇    | 500°C                | SiO₂              |           | [63]|
|          |       |                   | film                |                   |           |     |
| Td       | CVD   | W film, Te        | 800°C                | SiO₂              |           | [65]|
| 1T       | MBE   | W, Te             | 280°C                | SiC(0001)         | [50]     |
| 1T       | MBE   | W, Te             | 275°C                | Bi₃Te₃, MoS₂, graphite | [86] |
| Td       | PLD   | WTe₂              | 300°C (growth), 700°C (annealing) | mica | [52,92,94] |
| 1T       | PLD   | WTe₂              | 210°C                | Bi₃Te₃            | [96]     |
| amorphous| sputtering | Mo/MoO₃ | Mo, MoO₃, Te | 50–150°C | SiO₂ | [101] |
| amorphous| sputtering | Mo/MoO₃ | Mo, MoO₃, Te | 50–150°C | SiO₂ | [101] |
| MoTe₂    | 2H and 1T' | CVD | Mo/MoO₃, Te | 650–700°C | SiO₂ | [45–47] |
| 2H       | MBE   | Mo, Te            | 450°C                | Mo₅₂             | [78]     |
| 2H and 1T' | MBE  | Mo, Te            | 250–400°C            | SiC(0001)        | [82]     |
| 1T       | MBE   | Mo, Te            | 280°C                | SiC(0001)        | [82]     |
| 2H       | PLD   | MoTe₂             | 300°C (growth), 800°C (annealing) | SiO₂ | [51] |
| 2H and 1T' | sputtering | MoTe₂ | MoTe₂ | 400°C (growth), 650°C (annealing) | SiO₂ | [102] |
| 2H       | sputtering | MoTe₂ | MoTe₂ | 200°C (growth), 700–850°C (annealing) | SiO₂ | [103] |
| PtTe₂    | 1T    | CVD               | Pt film, Te         | 400°C             | SiO₂      | [49,66]|
| 1T    | CVD   | Pt film, Te       | 460°C                | SiO₂              |           | [67] |
| 1T    | CVD   | Pt film, Te       | 450°C                | Al₂O₃, STO, YSZ, mica, SiO₂, etc. | [68] |
|       |       |                   |                      |                   |           |     |
| PdTe₂    | 1T    | CVD               | Pd (111)             | 300°C             | SiC(0001) | [71]|
| 1T    | MBE   | Pd, Te            | 570°C                | SiC(0001)        | [70]     |
| ZrTe₂    | 1T    | CVD               | Zr, Te               | 400°C             | InAs(111)/Si(111) | [75] |
| 1T    | PLD   | ZrTe₃             | 550°C                | STO(110)         | [97]     |
3. Applications

TSMs have exhibited great potential in new generation electronic devices due to their novel surface states and exotic transport properties. Developments of large-area production further foster their practical application. In this section, we mainly review research on applications of large-area TSM films in field of superconductivity, optical response and spintronics.

3.1 Superconductivity

Plenty of 2D layered TSMs are proven to be superconductive under certain conditions, like WTe$_2$ [37], MoTe$_2$ [36] and Ir$_{1-x}$Pt$_x$Te$_2$ [105]. Interestingly, an unconventional magnetic field enhanced superconductivity was discovered in MBE grown WTe$_2$ [87], which might result from the lifting of inversion symmetry. As for the Pt-based TSMs, PtTe$_2$ has a low $T_c$ of below 0.002 K due to the weak of electron–phonon coupling, which is difficult for applications even in basic sciences [106,107]. There are many methods for improving the $T_c$, e.g. electron doping [108] or alkali-metal intercalations [109]. Although PdTe$_2$ and PtTe$_2$ have similar crystal structures and band structures, PdTe$_2$ has a higher $T_c$ (~ 1.7 K) due to its strong electron-phonon coupling strength [109,110]. Meanwhile, the topological state of the Dirac cone has little influence on the superconductivity state, which is more favorable for the study of the superconductivity characteristics of PdTe$_2$ [111].

After the finding of type-II Dirac fermions in PtSe$_2$-type transition metal dichalcogenides, bulk PdTe$_2$ was proven to possess both superconductivity and tilted Dirac cone. Liu et al. [112] prepared PdTe$_2$ films with various thicknesses from 2-ML to 20-ML on STO substrates by MBE and investigated the electronic and superconducting properties (Figure 6(a)). The monolayer PdTe$_2$ films showed narrow-band gap semiconductor behavior, while multilayer films exhibited good superconductivity. The $T_c$ of the ultra-thin film was significantly higher than that of the classical theoretical formula and could be further increased by Mg intercalation [112], as shown in Figure 6(b). Y. Liu et al. [113] observed abnormal metal states at low temperature in ultra-thin single crystal PdTe$_2$ films compared with traditional superconductor. Their resistance tended to reach a saturation value at extremely low temperature under the condition of high-quality filter measurement (Figure 6(c)). Meanwhile, the superconducting characteristics of the film remained almost unchanged within 20 months. At the same time, they also observed Quantum Griffiths Singularity (QGS) in a 4-ML PdTe$_2$ film under both perpendicular and the parallel magnetic fields [74], as shown in Figure 6(d). However, with the increase of film thickness to
6-ML, the QGS gradually disappeared under a parallel field but still maintained under a perpendicular field, which can promote the further study of novel quantum phase transitions induced by a parallel magnetic field.

### 3.2 Optoelectronics

As a type II Dirac semimetal, PtTe$_2$ shows a strongly tilted Dirac cone along some momentum directions, which violates Lorentz invariance and protects the gapless band structure by crystal symmetry [114]. The characteristics of high carrier mobility, ultrafast photo response and low photocurrent make PtTe$_2$ an ideal material for near/mid-infrared (NIR/MIR) and terahertz (THz) detection [115–117]. Wei et al. [72] prepared high-quality PtTe$_2$ films by MBE, which showed high sensitivity at 960 nm wavelength (Figure 7(a)), up to 15.6 mA·W$^{-1}$. Meanwhile, the films have an anisotropic optical response with wavelength dependency. When the scanning wavelength is 10.7 μm, the ratio of anisotropy...
ellipse reaches 8.3, indicating that the optical response of PtTe₂ exhibits stronger anisotropy under low energy excitation [72], as shown in Figure 7(b). This is very important for the study of polarization angle-sensitive detection. To quantify the response speed precisely, a 625 nm wavelength light is used to irradiate the PtTe₂/p-Si device as demonstrated in Figure 7(c) [49]. According to the photocurrent at different optical frequencies, the relative balance changes ~ 5% in the frequency range of 100 kHz and remains ~80.6% even at a very high frequency of 300 kHz (Figure 7(d)). In addition, the rise and fall times of ~1.68 μs and ~ 1.58 μs at an optical frequency of 150 kHz are measured, respectively (Figure 7(e)), which are lower than those of most 2D/2D or 2D/3D materials. Tong et al [118] used lithography to assemble 64-pixel PtTe₂-based NIRPD arrays on SiO₂/Si chips, showing good resolution in near-infrared imaging, as shown in Figure 7(f). The five letters
‘N I R P D’ can be clearly seen due to the sharp contrast with the background, indicating that the PtTe2-based arrays have great potential in the next generation of optoelectronics technology. Besides, PtTe2 also shows a high-performing photodetection system in the THz band [119,120]. Xu et al. [121] fabricated the THz detection devices based on PtTe2 with bow-tie-type planar contacts (Figure 7(g) –(h)), which showed a high photoresponsivity (1.6 A W\(^{-1}\) without bias voltage) with a response time less than 20 µs, while the detectors based on vertically stacked PtTe2 and graphene heterostructures reached responsivity above 1.4 kW W\(^{-1}\) and a response time shorter than 9 µs. Additionally, an impressive THz image of the metal nut inside the envelope is shown in Figure 7(i). Other topologically related optical responses, such as plasmons [44] and nonlinear absorption [92,93] have been observed in large-area WTe2 films and require further realization in other 2D layered TSM films.

### 3.3 Spintronics

With the rapid development of spintronics, magnetic random access memory (MRAM) devices based on spin transfer torque (STT) or spin orbit torque (SOT) have attracted considerable attention due to their fast response, non-volatile memory and low power consumption. Further development of STT/SOT-MRAM devices demands highly efficient spin-to-charge conversion materials. In previous studies, heavy metals were widely used as spin sources. Recently, the discovery of topological quantum materials has opened a new area for the design of spintronic devices. In TSMs, the topological surface states are spin-momentum locking, resulting in highly efficient spin-to-charge conversion. Besides, low symmetric crystal structures of layered TSMs lead to out-of-plane spin polarized current, which can directly switch perpendicular magnetization material without an external field. Studies of spin orbit torque have been reported in WTe2 [38,40,122–124] and MoTe2 [41,42] based on exfoliation flakes. To meet the demand for industrial fabrication, research on large-area TSM films has been carried out.

Fan et al. [100] studied the spin-charge conversion of amorphous WTe2 grown on Co\(_{20}\)Fe\(_{60}\)B\(_{20}\) by sputtering. Spin pumping and second harmonic Hall measurements showed a large field-like (FL) torque contrast to a small damping-like (DL) torque attributed to the spin swapping effect (Figure 8(a) –(b)). However, a large DL torque is found in the sputtered WTe2/CoTb heterostructure. This indicates that the composition of WTe2 may strongly affect the spin-charge conversion [101], as shown in Figures 8(b) and 8(c). In addition, Shi et al [125] estimated in-plane and out-of-plane spin conductivities close to 7.36 × 10\(^3\) (\(\hbar/2e\))
and $1.76 \times 10^3 \ (h/2e) \ (\Omega m)^{-1}$ in a centimeter-scale CVD growth Td-WTe$_2$. Figures 8(d) and 8e are schematic illustrations and images of the Py/WTe$_2$ devices. Magnetization switching was further demonstrated by the magneto-optical Kerr effect (MOKE) [125], as shown in Figure 8(f). The switching current density is as low as $10^5$ A cm$^{-2}$, which is 1–2 orders of magnitude smaller than the typical value of $10^6$–$10^7$ A cm$^{-2}$ in HMs [126]. Compared with other transition metal dichalcogenides (TMDs), PtTe$_2$ exhibits the large spin Hall conductivity because of its high conductivity and strong spin-orbit coupling. Through spin-torque ferromagnetic resonance measurements (Figure 8(g)), PtTe$_2$ presents a larger SOT efficiency ($\xi_{SOT} \approx 0.152$) than Pt ($\xi_{SOT} \approx 0.058$) [67], as shown in Figure 8(h)–(i), which exhibits great potential in wafer-scale spintronic device applications.
Conclusion and outlook

Recent studies on layered topological semimetals have proven their potential for many applications like superconductive devices, optical devices and spintronic devices. Efforts on the large-area fabrication of 2D layered TSM films further enhance the possibility for practical applications. There is no doubt that TSMs will continuously attract attention and effort from researchers. Here we suggest several topics to be concerned. We expect that 2D layered TSMs can quickly enter commercial applications after addressing the following issues.

(1) Compared with 2D semiconductors which have been intensively studied, experience in preparing TSMs in large area is still lacking. Although various fabrication techniques have been reported, a universal method to produce high-quality and wafer-scale films with low cost and high yield has yet to be developed. Growing epitaxial thin films of certain materials such as MoTe$_2$ and WTe$_2$ is still challenging because of the weak bonding energy and large vapour pressure difference between metal and Te atoms. Mo(W) atoms tend to bond with each other rather than form layered structure with Te, leading to the Te-deficiency formation. Moreover, Mo(W)Te$_2$ will decompose and lose Te as vapour at high temperature [46]. Due to these properties, it is difficult to directly obtain high-quality films by PVD methods, especially methods that rely on vaporizing compound targets. Post annealing in Te atmosphere may both improve the crystallinity and complement Te of as-deposited films [52]. In MBE growth, the flux ratio of Te/Mo(W) is usually kept at tens or even hundreds [77,80,86] to ensure the stoichiometric ratio of films.

(2) Another issue caused by weak bonding energy between Mo(W) and Te is the chemical instability of thin films. Exposure to air may induce surface oxidation of the films, resulting in degradation of electrical properties. For example, amorphous MoO$_3$–TeO$_2$ surface layers were observed in oxidized 1T’-MoTe$_2$ films and significantly enlarged the resistance [127]. In addition, the magnetoresistance of WTe$_2$ thin films increased about 30% after exposure to air for 30 days, which resulted from the p-type doping by surface WO$_x$ layer [128]. Since oxidation layers can remarkably change the intrinsic properties of bottom films, how to keep the prepared film stable in the ambient is the critical issue for practical applications. One feasible approach is to in situ depositing capping layers like AlN [97] and SiO$_2$ [102] that can protect TSM films from moisture and air. Meanwhile, we should explore more novel materials that are chemically stable in the air, like PdTe$_2$, PtTe$_2$, and PtSe$_2$.

(3) Since the critical superconducting temperature of TSMs is generally below 2 K, it seems to be an arduous task for basic research. Improving the T$_c$ of the TSMs appears to be of great importance, which is conducive to understanding the relationship between the topology and the superconductivity of the TSMs. The core of increasing T$_c$ is to increase the carrier concentration...
near the Fermi surface. For Pt-based TSMs, electron doping is one of the most effective means to enhance $T_c$, which can increase the $T_c$ of PtSe$_2$ and PtTe$_2$ up to 2.15 K [109] and 8 K [108], respectively.

(4) Heterostructures consisting of TSMs and other materials could provide a new platform for theoretical research and practical applications. A TSM/semiconductor heterostructure can be used as an electrical contact for FETs based on TMDs. In analogy with TI/ferromagnetic material (FM) structures [129], TSM/FM structures may trigger the new development in studying proximity coupling and spintronics devices. Besides, photoemission and photodetector in the near-infrared to THz range could be promising areas for TSMs. Furthermore, during the preparation of the ferromagnetic heavy metal and TSM heterostructures, the bottom layered materials will inevitably be damaged by the high-energy ion, leading to more spin loss at the interface during the process of spin-charge conversion. Thus, design of the all van-der Waals epitaxial heterojunction is obviously essential.

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Disclosure statement

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