Electronic and Optoelectronic Applications Based on 2D Novel Anisotropic Transition Metal Dichalcogenides

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With the continuous exploration of 2D transition metal dichalcogenides (TMDs), novel high-performance devices based on the remarkable electronic and optoelectronic natures of 2D TMDs are increasingly emerging. As fresh blood of 2D TMD family, anisotropic MTe$_2$ and ReX$_2$ (M = Mo, W, and X = S, Se) have drawn increasing attention owing to their low-symmetry structures and charming properties of mechanics, electronics, and optoelectronics, which are suitable for the applications of field-effect transistors (FETs), photodetectors, thermoelectric and piezoelectric applications, especially catering to anisotropic devices. Herein, a comprehensive review is introduced, concentrating on their recent progresses and various applications in recent years. First, the crystalline structure and the origin of the strong anisotropy characterized by various techniques are discussed. Specifically, the preparation of these 2D materials is presented and various growth methods are summarized. Then, high-performance applications of these anisotropic TMDs, including FETs, photodetectors, and thermoelectric and piezoelectric applications are discussed. Finally, the conclusion and outlook of these applications are proposed.

1. Introduction

Since the discovery of graphene decades ago, 2D materials have been studied by many research groups because of their excellent mechanical, electronic, and optoelectronic properties.[1–4] 2D transition metal dichalcogenides (TMDs) show great promise for the following reasons. First, their monolayers are bonded via van der Waals (vdW) interactions. In the construction of heterostructures based on TMDs, mismatches between the thermal and lattice coefficients of different materials can be avoided.[5,6] Second, the high mobility and 2D nanostructures of TMDs endow them with great electronic performances, including enhanced integration levels and suppressed short-channel effects.[2,7–10] Third, the indirect–direct bandgap transition between the bulk and monolayer in some TMDs affords various optoelectronic applications, from photodetectors to light emitters.[5,11–13] The common chemical formula of TMDs is MX$_2$, where M is a transition metal (group IVB–VIIB; M = Mo, W, Re, and so on) and X is a chalcogen (group VIA; X = S, Se, Te).[14,15] The sandwich structure of TMDs leads to excellent electronic and optoelectronic properties.[14–18] Currently, TMDs exhibit satisfying properties in various applications.[19–22] Ultralow standby power dissipation was realized in single-layer MoS$_2$-based field-effect transistors (FETs), with a high on/off ratio of 10$^8$.[2] Furthermore, ultrasensitive photodetectors based on monolayer MoS$_2$ were demonstrated by Lopez-Sanchez et al.[23] And high efficiency light-emitting diodes (LEDs) based on monolayer WSe$_2$ p–n junctions via electrostatic doping of gate were investigated by Baugher et al.[24] Ross et al.[25] and Pospischil et al.[26] respectively. In addition, the transparent and flexible nature of TMDs makes them ideal candidates for flexible electronic devices.[9,15,17] However, the studies of current 2D materials...
such as black phosphorus, group IV monochalcogenides (SnS, SnSe, GeSe, GeS), and anisotropic TMDs (WTe$_2$, ReX$_2$) are starting to focus on their anisotropic properties.[1] It is beneficial to control variability and uniformity of device performance by manipulating crystal orientation.[1,27] In addition, this novel physical degree of freedom can be applied for polarized light detection devices and potential valleytronics.[28–30] Herein, we discuss the recently developed MTe$_2$ and ReX$_2$ materials, which exhibit anisotropic features originating from their unique in-plane atomic arrangement.

The strong anisotropic properties of the MTe$_2$ and ReX$_2$ materials discussed in this paper originate from their distorted octahedral phase, specifically the distorted 1T phase for MoTe$_2$ and WTe$_2$, and the strong metal-metal bond (Re–Re bond) in the 1T’ phase for ReS$_2$ and ReSe$_2$. Researches based on Raman spectroscopy, polarization-resolved reflectance spectroscopy, and photoluminescence spectroscopy have demonstrated the strong anisotropic characteristics of these materials.[31–33] In addition to their anisotropic properties, these materials have many other fascinating properties. The ultrahigh theoretical carrier mobility (1000–2000 cm$^2$V$^{-1}$s$^{-1}$) of MTe$_2$ and ReX$_2$ facilitates the ultrahigh-speed conversion of photons into electrical signals, providing an ultrafast response time in optoelectronic devices.[8] Furthermore, monolayer MTe$_2$ and few-layer ReX$_2$ possess direct energy gaps from the near-infrared to the visible spectral region, as their bandgap ranging from 1–1.6 eV.[1,34–40] Unlike other symmetrical 2D materials, such as graphene and MoS$_2$, MTe$_2$ and ReX$_2$ have asymmetrical 2D crystal lattices. Inplane orientation-dependent electron and phonon properties are introduced into the system, which may inspire new concepts and ideas in angle-resolved semiconductor devices.[1,41–43]

Zhang et al. predicted that the valley degree of freedom, a key factor in valleytronics, can be tuned in MoTe$_2$ due to its anisotropic structure, as determined by first-principle calculations.[29] In addition, a new linear dichroic photodetector based on ReS$_2$ with a high responsivity of 1000 A W$^{-1}$ was reported by Liu et al.[44]

In this review article, we give a comprehensive review of the recent progress in anisotropic MTe$_2$ and ReX$_2$, with particular focus on their various applications. First, the crystalline structure and the origin of the strong anisotropy of these materials were analyzed using various characterization techniques. In addition, we discuss the preparation of these 2D materials, highlighting relevant mechanical exfoliation and chemical vapor deposition (CVD) methods. Following this, we discuss FETs, photodetectors, thermoelectric and piezoelectric applications based on these anisotropic TMDs. Finally, we present the challenges, opportunities, and outlook of these applications based on current views.

## 2. Anisotropic Crystalline Structure

### 2.1. Crystalline Structure

TMDs possess a layered crystal structure with individual layers bonded together via weak van der Waals interactions.[17,45–50] Typically, for TMDs, six chalcogenide (X) ligands are covalently bonded to one transition metal atom (M) center such that no dangling bonds appear in each monolayer. Due to their atomically flat surface, TMDs are ideal materials for application in novel FETs in which the short-channel effects are...
optimized.[49,51–54] Under ambient conditions, most well-studied group VI TMDs, such as MoS$_2$, have an isotropic hexagonal phase (2H) and octahedral phase (1T) crystal structure. This structure consists of alternating stacks of single-layer trigonal prisms formed by X atoms around the M atoms (Figure 1a). In general, the physical properties of TMDs are closely related to their lattice structures and the in-plane configuration of different atoms.

The lattice structures of some TMDs that have recently joined this family, specifically MTe$_2$ and ReX$_2$, can be quite different. Unlike MoS$_2$, these four materials exhibit a distorted structure relative to the 1T phase. The monolayer of the distorted 1T phase still has an X–M–X structure, where the upper X atoms are rotated by 180° with respect to the lower X atoms, forming an M-centered octahedral.[33,55,56] The center M atoms experience a shift in the layer plane along the perpendicular direction, forming strong metallically bonded X pairs. Additionally, the X atoms are no longer coplanar, but instead exhibit a zigzag structure varying along the atomic positions of the perpendicular direction. Small variations in the displacement and stacking lead to two different phases: the 1T’ phase (ReS$_2$, ReSe$_2$, and MoTe$_2$ in their natural state) and the Td phase (WTe$_2$ in its natural state).[40,46,48,54,57–62] We show the 2H, 1T, 1T’, Td crystal structures in Figure 1a. And the parameters of typical TMDs were shown in Table 1. This symmetry-reducing effect strongly enhances the anisotropy in the unit cell. Thus, compared with the well-studied TMD materials, these four 2D materials show astonishing characteristics in optoelectronic and electronic applications.

Moreover, the crystalline structures in these strongly anisotropic TMDs are not static. For MTe$_2$ and ReX$_2$, the 1T’ or Td phase of the atomic system is stable for the ambient environment. However, under certain conditions, the 1T’ (or Td) and 1T phases interconvert, as the energy gap of the two phases can be bridged mechanically or thermally.[63,64] However, though the rhenium atom contains an extra electron in the d orbital, there are still some differences between polytelluride TMDs (MoTe$_2$/WTe$_2$) and group VI TMDs with rhenium atoms (ReSe$_2$/ReS$_2$) in the crystalline structure, which is discussed in detail in the following section.

![Figure 1.](image)

**Figure 1.** a) The four crystalline phases (2H, 1T, 1T’, Td) of 2D TMDs. M represents a metal atom, X represents a chalcogen compound, and all three phases are composed of the X–M–X layer structure. Reproduced with permission.[33] Copyright 2014, Nature Publishing Group. Reproduced with permission.[45] Copyright 2015, American Chemical Society. Reproduced with permission.[65] Copyright 2015, American Physical Society. b) Perspective drawing of a primitive cell of monolayer ReS$_2$. The blue shows the strong metallic Re–Re bond. Reproduced with permission.[66] Copyright 2015, American Physical Society. c) Crystal structure of monolayer ReS$_2$, the red arrows represent the direction of a and b axes. d) Band structure of monolayer, trilayer, and five-layer ReS$_2$. Reproduced with permission.[67] Copyright 2015, the authors, published under CC-BY-4.0 license.
2.1. Anisotropy of Group VI TMDs with Rhenium Atoms

In contrast to traditional TMDs, such as MoS₂, the chalcogen atoms are not all equally displaced above and below the Re plane in polytelluride TMDs. In this special 1T’ crystal-line structure, the extra electron in the d orbital of the rhenium atom promotes the construction of a strong metallic Re−Re bond (Figure 1b). The distance between these dimerized Re atoms can be even shorter than that in rhenium single crystals; thus, the total energy and symmetry of the system are reduced. This is a major origin of the strong anisotropic properties of ReSe₂ and ReS₂ validated by Liu et al. The in-plane anisotropic properties of monolayer and few-layered ReS₂ are shown in Figure 1c. ReS₂ has two principal axes, (a axis, 118.97°; b axis, 61.03°). In accordance with the angles of the a and b axes, a quadrilateral shape with an angle of ≈60° or 120° is observed in thin, exfoliated ReS₂, and the large anisotropic ratio of mobility of ReS₂ along the two axes (up to 3.1) is the highest of all studied layered 2D materials. Meanwhile, the weak interlayer interaction exhibited in ReS₂ and ReSe₂, which is different from other TMDs, also affects the strong anisotropic properties. As determined by measuring the layer thickness, the ReS₂ crystal possesses an interlayer coupling weaker than most TMD crystals, such as MoS₂ and WS₂. The interlayer decoupling originates from the Peierls distortion of the initial 1T ReS₂ revealed by Zhong et al. As a result of the disordered stacking and minimized wavefunction overlap, this interlayer decoupling provides an ideal platform to characterize disordered stacking and minimized wavefunction overlap, this interlayer decoupling provides an ideal platform to characterize disordered stacking and minimized wavefunction overlap, this interlayer decoupling provides an ideal platform to characterize disordered stacking and minimized wavefunction overlap, this interlayer decoupling provides an ideal platform to characterize disordered stacking and...
been extensively used for the characterization of anisotropic 2D materials. In TMDs, Raman spectroscopy can be applied to verify the crystal phases and distinguish the layer numbers. In addition, the Raman response of in-plane anisotropic TMDs is related to the relative relations between the crystalline orientation and the polarizations of the incident laser and Raman scattered photons. In this section, we discuss the Raman spectra of 1T′ (or Td)-phase TMDs, which show the anisotropy of the TMDs. Taking MoTe₂ as an example, the Raman spectra of the 2H phase (black) and the 1T′ phase (red) of MoTe₂ are shown in Figure 3a. 2H MoTe₂ exhibits E₂g and A_g modes at 235 and 174 cm⁻¹, while 1T′ MoTe₂ does not exhibit an E₂g mode, but instead new peaks appear at 124, 138, and 272 cm⁻¹. The appearance of these new peaks indicates that the 1T′ phase has relatively lower in-plane symmetry than the 2H phase. The lower symmetry is fundamental in the anisotropic properties of MoTe₂. The Raman spectra of mono- to few-layer TMDs in the 1T′ (or Td) phase (MTe₂, and ReX₂) are shown in Figure 3b–d.

Moreover, for ReX₂, Raman scattering of the linearly polarized exciton also verifies the strong anisotropy. Chenet et al. measured the Raman spectrum of ReS₂ in a backscattering geometry using a laser of 532 nm with a fixed linear polarizer. The strongest modes are present between 120 and 240 cm⁻¹ in unpolarized Raman spectrum for a ReS₂ monolayer. Experimentally, Wolverson et al. found that the ReSe₂ Raman modes occupy the frequency range from 100 to 300 cm⁻¹ and are densely spaced, with the exception of a gap at ≈140 cm⁻¹ (Figure 3e).

In addition, the absorption spectrum further confirms the strong anisotropy of the excitonic transition. The absorption spectrum is anisotropic with respect to the polarization direction of the incident light. For monolayer ReS₂, its low symmetry caused an anisotropic optical response that can be described as the single-particle optical absorption level. As shown in Figure 3f, for the incident light polarized along the Γ-K₂ (x) direction, the single-particle optical absorption spectrum begins near 2.7 eV, while for the Γ-M₃ (y) direction, the spectrum becomes significant at 2.9 eV. Ho and Huang reveal that this optical anisotropy is assigned to the transitions from nonbonding Re 5d t₂g to 5d t₂g* and to antibonding chalcogen p and σ states. Monolayer ReSe₂ exhibits a similar anisotropic optical response, with subtle differences. The optical absorption spectrum direction of
monolayer ReSe₂ is different from the polarization direction of incident light. The lowest-energy absorption peak (~1.4 eV) along the Γ-K₂ (x) direction does not completely disappear for the incident light polarized along the Γ-M₃ (y) direction. Zhong et al. verified that the lowest-energy peak of ReSe₂ consists of two excitons (Eₓ and Eₚ), in contrast to that of ReS₂. Eₓ is dark when the incident light is polarized along the Γ-M₁ (y) direction, such as the lowest exciton in ReS₂. Differently, Eₚ is not completely dark.
along the same direction. Thus, the lowest-energy peak does not have an extremely high polarization anisotropy. The oscillator strength of the excitons ($F_\perp$, $F_\parallel$) shows significant spatial anisotropy. However, the prominent absorption peak at 1.4 eV is the combination of these two excitons, and the overall optical absorption does not show complete anisotropy (Figure 3g).[[65]]

### 3. Preparation Methods

Although the 2D TMDs MTe$_2$ and ReX$_2$ have the abovementioned anisotropic structures, the large-area integration of these materials in the numerous methods that produce single-crystal and uniform-thickness films is limited. Most devices involving isotropic photoreponses have <5 nm layer thickness, that is, the controlled large-area synthesis of few layered TMDs is a prerequisite for utilizing these anisotropic properties. Up to now, many methods have been designed to obtain high-quality anisotropic TMDs. In general, the preparation methods, such as mechanical exfoliation[[109]] and chemical vapor deposition,[[110,111]] can be divided into top-down and bottom-up methods. In this section, we give a brief review of the mechanical exfoliation and CVD methods to obtain MTe$_2$ and ReX$_2$ nanosheets.

#### 3.1. Mechanical Exfoliation

The mechanical exfoliation method (or Scotch-tape method) was first discovered by Novoselov et al., who used the method to obtain 2D graphene.[[112]] This method was a watershed in tomorrow’s technology. Despite its simplicity and crude procedure, the as-cleaved materials provide crystalline samples with extraordinary mechanical and electrical properties.[[112–114]] Similar to graphene, anisotropic materials of MTe$_2$, and ReX$_2$, can also be mechanically exfoliated from a natural crystalline sample to study the fundamental properties of structural anisotropy, and findings suggest that these monolayers may offer unique applications in polarized photodetectors, sensors, and photonic devices, which has attracted a wide range of attention.[[28,40,115–119]] For instance, Octon et al. obtained fast, high-responsivity, few-layer MoTe$_2$ photodetectors by mechanical exfoliation.[[122]] Liu et al. prepared high-responsivity phototransistors based on few-layer ReS$_2$ for weak signal detection by a standard mechanical exfoliation method.[[83]] Lu et al. obtained high-performance ReS$_2$ nanosheet photodetectors via mechanical exfoliation from bulk MoReS$_2$, which showed different performances in different gas environments.[[120]] In particular, to further understand the processes of exfoliation, Golberg et al. systematically investigated the cleavage processes and associated mechanical behaviors via a direct in situ transmission electron microscopy (TEM) probing technique.[[121]] The results showed that the bending behavior of atomic layers is related to the number of layers during exfoliation.[[121,122]] For bulk layered materials, one common deformation mechanism is the formation of kinks.[[123]] However, when the atomic system has <11 layers, the equilibrium shape is determined by non-covalent dispersion forces that constitute the surface energy. Undoubtedly, mechanical exfoliation method has allowed for substantial progress in elucidating the basic properties and applications of TMDs. However, mechanical exfoliation is likely to produce edges and ribbons along well-defined crystalline directions.[[124]] and the layers, morphology, and edges are still not controllable. Additionally, to realize large-area fabrication, the mechanical exfoliation method seems to face significant challenges because of the extremely low yield and low controllability of the layer number and large-area uniformity. Therefore, this technology is expected to have limited commercial high-end applications and to only be used for scientific research.[[125]]

#### 3.2. CVD

CVD is a well-established technology that has been demonstrated as a facile method for synthesizing large-scale monolayer crystals, including graphene, MX$_2$,[113,126–134] and their heterojunctions.[135–140] Compared with the exfoliation method, the direct synthesis of few-layer and monolayer MX$_2$ by CVD is critical to large-scale applications. In this section, we discuss the synthesis of anisotropic materials of MTe$_2$, and ReX$_2$, by CVD methods.

There are two main obstacles for the synthesis of WTe$_2$ and MoTe$_2$ by existing CVD or physical vapor deposition methods. First, the electronegativity difference between the transitional metal (W or Mo) and Te (0.4 or 0.3 eV) is low.[[141]] The poor electronegativity difference between the transitional metal (W or Mo) and Te indicates the existence of weak bonding between the metals and Te atoms, which makes the WTe$_2$ or MoTe$_2$ stoichiometry difficult to obtain. Another problem involves synthetic issues of both the precursors and products, including oxidation,[[141]] volatility,[[142]] thermal instability,[[143]] and phase targeting. In general, W(Mo)Te$_2$ single crystals are grown by the chemical vapor transport (CVT) method, and then, few-layer flakes are exfoliated onto SiO$_2$/p++ Si substrates.[[144]] Until now, few papers have focused on the synthesis of telluride, particularly the synthesis of single-crystal monolayers by CVD. In 2015, Park et al. and Zhou et al. synthesized polycrystalline MoTe$_2$ thick films via the tellurization of Mo films deposited on 300 nm thick SiO$_2$/Si substrates with an e-beam evaporator or sputterer.[[63,145]] However, the quality of this as-grown MoTe$_2$ is inferior to that of the mechanically exfoliated sample. This method is difficult to apply to the synthesis of WTe$_2$ due to the low chemical reactivity between W and Te. Gong et al.’s work indicated that Te can facilitate the synthesis of MoS$_2$/WS$_2$ heterojunctions by lowering the melting point of the materials.[146] Inspired by this phenomenon, Zhou et al. demonstrated an effective CVD strategy, as shown in Figure 4a,b using a Te:metal oxide weight ratio of 1:1:1, to directly synthesize few-layer and monolayer WTe$_2$ and MoTe$_2$ on a large scale. As shown in Figure 4c–f, the structures of the as-grown WTe$_2$ and MoTe$_2$ monolayers were characterized by their optical vibrational modes in Raman spectroscopy, and a low defect concentration was confirmed (Figure 4g–j).[[146]] This effective route may lay the foundation for the construction of atomically thin telluride materials, the realization of fundamental properties, and the large-area integration of current silicon substrates.

The melting point of Re is ≈3180 °C, which is one of the highest melting point of all metals, and S and Se have relatively
low melting points (155 and 221 °C, respectively) and high vapor pressures. Thus, the direct crystal growth of ReX₂ is relatively challenging because of the large difference in melting points. Previously, ReX₂ has been synthesized in bulk crystals via CVT or via halogen vapor transport using I₂ or Br₂ as a transport agent. It is inevitable that unintentional background doping (I₂ or Br₂) may change the electrical properties of the materials. The crystals grown by the I₂ vapor transport technique are typically p-type, while the use of Br₂ usually results in n-type materials. Until now, studies on the synthesis of TMDs via CVD are still rare. Very recently, many groups have explored easier and more controlled fabrication methods that give higher yields of ReS₂ and ReSe₂ nanosheets. As shown in Figure 5a,b, Cui et al. introduced a tellurium material based on the Re—Te binary eutectic, whose eutectic point can be lowered to 850 °C or even to 430 °C when the Te—Re weight ratio is 90%. Those novel strategies assist the epitaxial growth of large-area, highly crystalline ReS₂ atomic layers on mica substrates (Figure 5c–i). Similarly, Keyshar et al. fabricated ReS₂ at a low growth temperature (450 °C) with ammonium perrhenate and sulfur as the raw materials on SiO₂/Si substrates. Wu et al. observed the domain architecture and grain boundaries of ReS₂ on sapphire substrates, while Zhai et al. and co-workers achieved the growth of hexagonal single-crystalline ReS₂ flake for the first time. Large-area continuous polycrystalline bilayer ReS₂ films were grown via CVD in a three-zone horizontal tube furnace using ReO₃ and S. A similar route using Se as a precursor...
material on SiO₂/Si substrates was reported by Hafeez et al.\[84\] Interestingly, sapphire was found to facilitate thinner flake or film growth, similar to WSe₂ on sapphire substrates,[141] but the cause is still unclear.

3.3. Other Methods

Chemical, electrochemical, and liquid-phase exfoliation are also effective synthetic methods providing the scalable production of stacked 2D thin films and heterostructures, which have wide application in research fields such as catalysis[154] and energy storage[156] and in FETs.[157] For example, Fujita et al. produced chemically exfoliated ReS₂ nanosheets from bulk powders with a solvent-free method by lithium intercalation. Meanwhile, their semiconducting nature and photocatalytic properties were retained.[158] Recently, Sun et al. reported a novel low-temperature solution synthesis of few-layer 1T’ MoTe₂ nanostructures by the solvothermal method. However, these nanostructures exhibit a lateral lattice compression of ≈1% compared with the bulk analogue, causing light compressive lattice strain.[159] Molecular beam epitaxy (MBE) has the unique advantages over other growth methods of elementally controlled deposition rates and easy switching from one material to another. MBE is also used by some groups to realize the precisely controlled growth of high-quality 2D structures or heterostructures.[160] However, high instrument costs make the commercial viability of MBE technology difficult.

4. Applications for 2D Optoelectronic and Electronic Devices

Logic devices based on metal–oxide–semiconductor field-effect transistors (MOSFETs) are fundamental in microelectronic circuits.[161,162] As the feature size decreases, the degradation of the MOSFET due to short channel effects is inevitable.[163,164] TMDs are favored for their dangling-bond-free morphology and atomic-scale thickness. As a consequence, its carrier scattering is negligible, while its carrier density can be easily controlled via the gate voltage. In this section, we discuss the recent developments of FETs and photodetectors based on anisotropic TMDs. We first investigate the electronic transport phenomena and application of few-layered MTe₂ and ReX₂. Then, MTe₂ and ReX₂ photodetectors based on photovoltaic effects in the near-IR and visible range for application in optoelectronics are discussed. Notably, polarized light detection is demonstrated in orientation-dependent field-effect photodetectors. Furthermore, we discuss the different optoelectronic behaviors in vdW heterostructures with various composite structures and tuning of the photodetector through intercalation and atmospheric exposure. Finally, we propose prospects for piezoelectric and thermoelectric applications.

4.1. FETs Based on Anisotropic TMDs

FETs, one of the most elementary components in electronics, are widely used in a variety of electronic components. However, the traditional semiconductor FET is close to the miniaturization limit under the constant exploration of many researchers.[7] Sub-10 nm channel lengths lead to multiple challenges in traditional FETs, such as drain-induced barrier lowering and punch through, surface scattering, velocity saturation, impact ionization, and hot electron effects.[163,165] The atomically flat basal planes of 2D TMDs are ideal for electronic device construction. Most MTe₂-based FETs have been reported to possess a carrier mobility of 1–68 cm² V⁻¹ s⁻¹ and large on/off ratios of 10⁵–10⁶.[16,22,75,98,166–168] Pradhan et al. reported hole-doped MoTe₂ field-effect transistors (Figure 6a) with a saturated carrier...
mobility of up to 20 cm$^2$ V$^{-1}$ s$^{-1}$ under a suitable bias at room temperature. This device displayed an on/off ratio over 10$^6$ and typical subthreshold swings of $\approx$140 mV dec$^{-1}$ (Figure 6c). Specifically, a few-layer MoTe$_2$-based phototransistor tuning by a ferroelectric polymer poly (vinylidene fluoride-trifluoroethylene-chlorotrifluoroethylene) (P(VDF-TrFE) top gate reached a current maximum carrier mobility of 68 cm$^2$ V$^{-1}$ s$^{-1}$ among MoTe$_2$-based FET (Figure 6d). This ultrahigh carrier mobility shows that intercalation tuning can efficiently enhance the field-effect transistor performance. When the top gate voltage was swept from $-35$ to $35$ V at room temperature, a high on/off ratio of 10$^6$ is comparable with the highest level achieved with MoS$_2$. In addition, angle-resolved FET has been implemented by regulating asymmetrical monolayer ReS$_2$. Liu et al. observed that the anisotropic ReS$_2$-based FET under a fixed source–drain bias voltage of 100 mV had a large on/off ratio of 10$^7$ and low subthreshold swings of 100 mV dec$^{-1}$ (Figure 6h), when the back gate was swept from $-50$ to $50$ V. The anisotropic mobility ratio $\mu_{\text{max}}/\mu_{\text{min}}$ along two principle axes of this device is 3.1, which is better than that of a device containing few-layer black phosphorus (Figure 6h).$^{[27,41]}$ The direction of lowest mobility (0° or 180°) was defined as the reference direction, and the highest mobility of 15.4 cm$^2$ V$^{-1}$ s$^{-1}$ was measured in a six-layer device in the 120° (or 300°) direction (Figure 6i).$^{[67]}$
4.2. Development of 2D Anisotropic Photodetectors

Light detection is one of the basic components of many optoelectronic applications, including imaging, quantum communication, dynamic capture technology, positioning, and guidance. Modern photodetector devices require small sizes, fast response times, and high detection accuracy and sensitivity over a wide wavelength range. 2D materials are ideal building blocks for photodetectors because of their plasticity, controllability, and excellent physical properties. In the application of 2D TMDs in photodetectors, there are two main photon–matter interaction operating modes: the photoconduction mode and the photocurrent mode, based on the photovoltaic effect. In the photoconduction mode, photoexcited carriers directly increase the device’s conductance. But in the photocurrent mode, photoexcited carriers are transformed into current under an asymmetric built-in electric field. A high responsivity of 880 A W\(^{-1}\) at a wavelength of 561 nm based on the photoconduction effect was found in monolayer MoS\(_2\). It has been reported that the photocurrent in vertically stacked graphene/MoS\(_2\) (16 nm)/graphene heterostructures can be modulated by the gate bias to achieve high quantum efficiency (MAX EQE = 55%, MAX IQE = 85%). Although TMDs such as MoS\(_2\), MoSe\(_2\), WS\(_2\), and WSe\(_2\) have led to considerable progress into current under an asymmetric built-in electric field, a high mobility indicates that the absorbed photons more quickly convert into electrical signals, which can improve the responsivity and the photoresponse time. Octon et al. observed a fast photoresponse time (~160 μs) in few-layered MoTe\(_2\) (6.5 nm) photodetectors based on photoconduction under 685 nm laser illumination with a high responsivity of 6 A W\(^{-1}\) (Figure 7a,b). However, the mobility is restricted due to the high contact resistance between the electrode and MoTe\(_2\). Yin et al. investigated the contact resistance of different electrode materials. This work revealed the emergence of a Schottky barrier in Au-contacted devices. Strikingly enhanced electron injection from the electrode to the channel was observed due to the introduction of a tunneling mechanism. The highest electron mobility of this FET approached 25.2 cm\(^2\) V\(^{-1}\) s\(^{-1}\), with a high photoresponsivity of 2560 A W\(^{-1}\) under the illumination of a 473 nm laser.

In contrast to group VI TMDs, few-layer ReX\(_2\) is a direct bandgap semiconductor with weak interlayer coupling. Thus, ReX\(_2\)-based applications do not require a single-layer structure, reducing the cost and difficulty of preparation. In addition, we achieve an ultrahigh responsivity and external quantum efficiency (EQE) under precise regulation. In the photocurrent operating mode, the regulated crystal orientations of anisotropic materials can tune the photogenerated current very well by changing the built-in electric field at junctions to both improve the responsivity and control the wavelength range of light detection. We provide the performance of various photodetectors in Table 2 and discuss them further in the following sections.

### Table 2. Performance parameters of photodetector (ML: Multilayer, IR: Infrared).

| Devices                  | Responsivity [A W\(^{-1}\)] | EQE [%] | Rise time [ms] | Decay time [ms] | Wavelength       | References |
|--------------------------|------------------------------|---------|----------------|-----------------|-----------------|------------|
| MoTe\(_2\) (4L)          | 6                            | –       | 0.16           | 0.3             | Visible/IR      | [123]      |
| MoTe\(_2\) (ML)          | 2560                         | –       | –              | –               | Visible         | [76]       |
| MoTe\(_2\) (ML)          | 0.024–0.05                   | –       | 1.6            | 1.3             | Visible/IR      | [176]      |
| ReS\(_2\) (ML)           | 16.14                        | 3168    | \(5 \times 10^4\) | \(5 \times 10^4\) | Visible         | [62]       |
| ReS\(_2\) (ML)           | 1000                         | –       | –              | –               | Visible         | [28]       |
| ReS\(_2\) (ML)           | 88 600                       | \(2 \times 10^7\) | –              | –               | Visible         | [97]       |
| ReS\(_2\) (ML)           | 604                          | \(1.5 \times 10^8\) | 2              | 2               | Visible         | [84]       |
| ReS\(_2\) (monolayer)    | 12                           | –       | –              | –               | Visible         | [85]       |
| ReS\(_2\) (ML)           | 10\(^2\)                     | –       | 670            | 5600            | Visible         | [177]      |
| ReSe\(_2\) (ML)          | 95                           | 18 645  | 68             | 34              | Visible         | [88]       |
| Mo:ReSe\(_2\)           | 55.5                         | 10 893  | 96             | 340             | Visible         | [120]      |
| α-MoTe\(_2\)/MoS\(_2\)  | 0.037–0.322                  | 85      | 25             | 25              | Visible/IR      | [16]       |
| P(VDF-TrFE)/MoTe\(_2\)  | 0.0164                       | –       | 1.4            | 1.3             | IR              | [75]       |
| MoTe\(_2\)/graphene      | 0.02                         | –       | 30             | 30              | Visible         | [178]      |
| ReSe\(_2\)/MoS\(_2\)    | 6.75                         | 1266    | 80             | 80              | Visible         | [87]       |
multilayer ReX2 can be used in device construction to improve the light absorbance. Because of this distinct bandgap feature, ReS2-based photodetectors generally have high responsivity (>10 A W⁻¹) and EQE (>1000%) and thus can be applied to detect extremely weak signals. Zhang et al. first fabricated few-layer ReS2 back-gate photodetectors (Figure 7c,d) with a maximum gate-tunable responsivity of 16.14 A W⁻¹ and an EQE of 3168%. [115] The photodetectors were irradiated by a focused laser beam (633 nm, 12.5–1000 nW) under 50 V back-gate bias. In addition, the transport properties were studied at different temperatures using four-terminal back-gated devices, and a maximum mobility of ≈8 cm² V⁻¹ s⁻¹ was acquired at 120 K. Although the responsivity of this device is comparable to that of graphene and MoS2, its applications are limited due to the slow response time (≈500 s) and small on/off ratio of 2.8.

In 2016, a fast-response-time photodetector based on ReS2 flake was constructed by Hafeez et al. In their study, CVD-grown ReS2 film- and ReS2 flake-based photodetectors were illuminated at a 500 nm light of 3.11 mW cm⁻² (Figure 7e,f). Both the rise and decay times of the ReS2 film-based devices are much slower than that of the flake-based devices. The flake-based devices show a fast response time of 2 ms, high responsivity of 604 A W⁻¹, and EQE of 1.50 × 10⁵%. [84] In addition, Liu et al. demonstrated an ultrahigh photoresponsivity of 88600 A W⁻¹, corresponding to a high EQE of 2 × 10⁷% at a wavelength of 532 nm in few-layer ReS2 phototransistors (Figure 7g,h). [83] Such high responsivity is a record for individual 2D photodetectors and is two orders of magnitude higher than that of monolayer MoS2, which can be used for weak signal detection (Figure 7i,j). This high photoresponsivity is attributed to an increase in photon absorption and a photogain mechanism involving trap states in ReS2, where the trap state density was estimated to be 1.96 × 10¹³ cm⁻² by studying the temperature-dependent field-effect mobility.
4.2.2. Polarized Light Photodetector

Polarized light detectors are essential components for controlling the vibration direction and period of light.[179] This photodetector resolves the polarization direction and has enormous potential in communication, remote sensing, and photography.[180] Therefore, polarized light photodetectors with high sensitivity and integration level are necessary. However, current devices are still far from satisfying commercial requirements. Emerging 2D materials have made progress in the detection of polarized light because of their orientation-dependent crystal structures. Liu et al. developed a new method to detect polarized light using 2D anisotropic ReS₂ (Figure 8a). This transistor shows an ultrahigh responsivity of $10^3$ A W⁻¹ in a linear dichroic photodetector with a high electron mobility of $40 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ and on/off ratio of over $10^5$ (Figure 8b–d). This study demonstrated that anisotropic ReS₂ can be applied for light polarization detection with excellent performance, which may open new avenues for polarized light detection.[28] Similar results have also been found in ReSe₂ FETs, where an ambipolar gate-tunable linear dichroism photodetector was exhibited in Figure 8e. High on/off current ratio of up to $10^7$ with stable saturation current was shown in this device (Figure 8f,g). Moreover, the mobility can be tuned by the temperature, and over 500-fold and 100-fold enhancements in the electron and hole mobilities, respectively, were observed at low temperatures.[51]

4.2.3. Photodetector Based on Heterostructures

A photodetector based on the photocurrent operating mode consists of a tunable junction, and the current study of MTe₂ and ReX₂ focuses on out-of-plane heterostructures.[5,12,166,181–183] Heterostructures play a major role in conventional semiconductor technologies because interfaces provide an effective and easy way to control the carrier type, density, and mobility. Mismatches in the lattice and thermal coefficients should be avoided to ensure a satisfying performance of the heterostructures.[184,185] 2D heterostructures can overcome the two aforementioned obstacles resulting from their vertically weak van der Waals bonding characteristics.

Type-II heterostructures can effectively tune the interlayer gap due to their staggered band alignments.[186,187] Pezeshki et al. investigated photodetectors based on α-MoTe₂ (3l)/MoS₂ (3l) p–n diodes working in photocurrent operating mode (Figure 9a). The calculated type-II interlayer gap between the conduction band minimum of MoS₂ and the valence band maximum of α-MoTe₂ is ~0.35 eV. This device showed a photoresponse time within 25 ms and maintained a stable photovoltaic effect with 1–3 Hz photoswitching dynamics and a good on/off current ratio of $\approx 1 \times 10^{-4} \times 10^3$ (Figure 9b,c). Blue to IR (470–800 nm) light was employed to examine this device, and blue photons gave the highest responsivity of 322 mA W⁻¹ with 85% EQE. The responsivity and EQE of this device decreased with reduced photon energy, where the lowest values were 37 mA W⁻¹ and 6% EQE under 800 nm IR illumination (Figure 9a). Furthermore, Zhang et al. studied IR photodetection in a type-II MoTe₂/MoS₂ heterostructure with strong interlayer coupling by interlayer optical transition (Figure 9d). This device had an interlayer gap of 0.66 eV (=1880 nm) and effectively detected infrared light (1.55 μm) with a distinct photocurrent response (Figure 9e,f). A multilayer ReSe₂/MoS₂ p–n heterostructure-based photodetector operated in photocurrent mode was examined by Wang et al.’s group (Figure 9f). This mobility of the heterostructure device was obviously enhanced (electron, 4 cm² V⁻¹ s⁻¹) and was higher than that of ReSe₂ (hole, 0.145 cm² V⁻¹ s⁻¹) and MoS₂ (electron, 0.226 cm² V⁻¹ s⁻¹). In particular, this device demonstrated an extremely high responsivity of 6.75 A W⁻¹ and EQE of 1266%, which are better than other X/MoS₂-based photodetectors investigated so far.[87]

4.2.4. Photodetector Based on Intercalation and Atmosphere Tuning

Various photodetectors with superb performance based on MTe₂ and ReX₂ were discussed in the previous sections. Most photodetectors were modulated by the applied gate voltage and temperature.[85,176–178,188] However, other tuning methods, such as by intercalation or atmosphere, can also enhance the physical properties of the host materials to enhance the optical-electronic performance.[14] As mentioned above, Huang et al. fabricated a ferroelectric polymer P(VDF-TrFE) top-gate photodetector based on MoTe₂, with a high carrier mobility that was calculated to be 68 cm² V⁻¹ s⁻¹ (Figure 10a,b). Because the remnant polarization of ferroelectrics reduces the dark current, this device performs over a broad photoresponse range (0.6–1.5 mm) with the maximum responsivity and detectivity reaching 16.4 mA W⁻¹ and 1.94 × 10⁶ Jones, respectively, under 1060 nm light.[73] ReSe₂ has similar properties to ReS₂, and ReSe₂-based photodetectors have extremely high responsivity and EQE. In addition, layer-dependent electrical and optoelectronic responses have been observed in ReSe₂-based photodetectors. Yang et al. (Figure 10c,d) showed that a monolayer ReSe₂-based transistor possessed a much higher mobility ($\approx 9.78 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$) than a multilayer device and a red-light-sensitive (633 nm) mobility of $\approx 14.1 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$, Furthermore, because of the enhanced molecular physiosorption due to the low symmetry structure, the photoconduction of ReSe₂ nanosheet photodetectors is significantly affected by the gas atmosphere. This atmosphere-sensitive property allows for an additional tuning approach by gas molecule gating. A high photoresponsivity of 95 A W⁻¹ and an EQE of 18645% were found in a monolayer ReSe₂-based transistor under red light (633 nm) and an O₂ environment (Figure 10e,f).[188] A similar phenomenon was also observed in Mo-doped ReSe₂ nanosheet photodetectors, which showed a photoresponsivity of 55.5 A W⁻¹ and an EQE of 10893% in NH₃ after annealing.[120]

4.3. Prospects for Piezoelectric and Thermoelectric Applications

Piezoelectricity is a direct conversion from mechanical stress force to electricity via accumulating polarization charge in asymmetrical atomic structures, which has been widely applied in micro-electromechanical systems, actuation, sensor, and electronics calls. TMDs have been ideal candidates for piezoelectric applications for the following reasons.[189–192] First, their noncentrosymmetry and low-dimension structure is considered to induce piezoelectricity.[192] Second, TMDs own high crystallinity which can withstand great pressure.[190,191,194] Third, TMDs can retain their single-layer piezoelectric structures without large surface energy, causing thermodynamically unstable lattice
Figure 8. a) The schematic of the ReS₂-based photodetection device under illuminating polarized light by the half wave plate. The photocurrent response as a function of drain voltage under b) different light intensity green light illumination, c) different polarization light illuminations, and d) different drain biases. Reproduced with permission.[28] e) The schematic structure of ReSe₂ photodetectors. The half-wave plate was used to change the polarization direction. f) \( I_{ds}-V_{ds} \) of the photodetector with incident laser power varied from 0 to 20 \( \mu \)W. Inset: Small range of \( I_{ds}-V_{ds} \) with different incident laser powers. g) Scanning electron microscope (SEM) image of the polarization-dependent photocurrent mapping of ReSe₂ photodetector, showing prominent linear dichroic photodetection. Reproduced with permission.[13] Copyright 2016, American Chemical Society.

Figure 9. a) Photoinduced \( I-V \) curves of MoTe₂/MoS₂ heterojunction on a logarithmic scale along. Inset: The schematic and of MoTe₂/MoS₂ photodetector. b) Time domain of short-circuit current (ISC) and under 1-Hz RBG LED illuminations. c) Photoresponse characteristics of responsivity based on MoTe₂/MoS₂ photodetector under RGB LEDs and IR (800 nm) laser, obtained at zero volt. Reproduced with permission.[16] d) The schematic illustrations of type-II interband excitation processes in MoTe₂/MoS₂ vdW heterostructures. Inset: The schematic diagram of a MoTe₂/MoS₂ vDW heterostructure device under infrared light excitation. e) The curves of \( I_{ds}-V_{ds} \) under infrared light illumination of 1550 and 2000 nm. Inset: Photovoltaic effect of the fabricated device under 1550 nm light illumination. Characterization of ReSe₂/MoS₂ heterojunction and device. Reproduced with permission.[166] Copyright 2016, American Chemical Society. f) Output plots of the devices in the dark and under 8.15 mW cm\(^{-2}\) light illumination. Inset: Schematic of the device based on the p-n ReSe₂/MoS₂ heterojunction. Reproduced with permission.[87] Copyright 2015, Springer.
reconstruction under ambient conditions. In 2014, the piezoelectricity of monolayer MoS$_2$ was first observed with a power density of 2 mW m$^{-2}$ and mechanical-to-electrical energy conversion efficiency of 5.08% by Wu et al. Then, Zhu et al. measured the piezoelectricity of monolayer MoS$_2$ with a piezoelectric coefficient of $2.9 \times 10^{-10}$ C m$^{-1}$. Compared with MoS$_2$, MTe$_2$ possesses more intense asymmetry because of distorted octahedral structure. And it is important that piezoelectricity originates from non-centrosymmetry structure. Thus, Duerloo et al. calculated that 2H MoTe$_2$ has the highest piezoelectric coefficients ($e_{11} = 2.98$ (clamped-ion)/$5.43$ (relaxed-ion) $10^{-10}$ C m$^{-1}$) and 2H WTe$_2$ has the lowest piezoelectric coefficients in TMDs ($e_{11} = 1.60$ (clamped-ion)/$3.40$ (relaxed-ion) $10^{-10}$ C m$^{-1}$). Both 2H MoTe$_2$ and 2H WTe$_2$ have the huge $\Delta e_{11}$ bigger than that of MoS$_2$. In addition, the piezoelectric effect of 2H MTe$_2$ may be much more abundant than the theoretical calculation due to its surprising magnitude.

The anisotropic crystal structure may benefit not only the piezoelectric properties as mentioned above, but also the thermoelectric properties. Analogous to piezoelectricity, thermoelectric devices convert heat into electrical energy by the Seebeck effect. The low-dimension materials such as nanowires, superlattices, or quantum dots have achieved excellent thermoelectric figure of merit ($ZT$) via reducing dimensionalities. And bulk SnSe is expected to enhance the thermoelectric efficiency due to anisotropic bonds. Thus, low-dimension and anisotropic structure was considered to increase $ZT$ and thermoelectric efficiency. The strong anisotropic TMDs of MTe$_2$ and ReX$_2$ meet the above requirements and have the potential to exceed anisotropic black phosphorus. Ma et al. have demonstrated strong anisotropic thermal conductivity of monolayer WTe$_2$ by first-principle calculations. WTe$_2$ exhibits thermal conductivity of 9 and 20 W m$^{-1}$ K$^{-1}$ along two principal lattice directions in room temperature, which is higher than thermal conductivity of black phosphorus ($<10$ W m$^{-1}$ K$^{-1}$). In addition, there may be two types of transport in TMDs including in-plane transport in the van der Waals interface and out-plane transport across van der Waals by a tunneling effect. ReX$_2$ possesses weak interlayer coupling, which is different from strong interlayer coupling. The ratio between two transports of ReX$_2$ which may result in an improvement of the $ZT$.

5. Summary and Outlook

Herein, we reviewed the latest knowledge of the crystalline structure, preparation methods, and electronic and optoelectronic applications of novel and strongly anisotropic MTe$_2$ and ReX$_2$ materials. The anisotropy, caused by the distorted octahedral phase, was analyzed using various characterization methods. Mechanical exfoliation from bulk materials and CVD...
are the primary methods to prepare strongly anisotropic TMDs. Other methods, such as chemical and liquid-phase exfoliation, were further discussed. However, the growth of high-quality, large-scale, and uniformly oriented MTe₂ and ReX₂ remains a challenge. In addition, the stacking of different 2D materials requires more in-depth study to upgrade the device performance. Photodetectors containing MTe₂ and ReX₂ based on the photovoltaic effect show better performance than other TMDs in terms of the responsivity and EQE, especially in the detection of polarized light. Intercalation and atmosphere tuning were also demonstrated to improve the device performance. However, this improvement is not sufficient. Linear polarized excitons and selectively tunable optical Stark effects have been theoretically predicted in thin ReS₂, ReX₂-based or MTe₂-based photodetectors and light-emitting diodes are extremely promising devices that still require in-depth examination. It is worth mentioning that magnetic-field-induced valley Zeeman splitting and polarization have been observed in monolayer MoTe₂. Novel valleytronics made of MTe₂ have great potential for next-generation optoelectronic devices. Overall, the recent findings concerning anisotropic TMDs indicate broad promise in electronic and optoelectronic applications.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

2D, anisotropy, electronics, optoelectronics, transition metal dichalcogenides

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