2D Janus Transition Metal Dichalcogenides: Properties and Applications

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The successful fabrication of Janus transition metal dichalcogenide (TMD) monolayer has sparked extensive research interests in various fields, such as nanoelectronics, optoelectronics, valleytronics, and catalysis. Janus TMDs can not only inherit the advantages of conventional TMDs but also produce novel properties which are different from their counterparts. The breaking of vertical mirror symmetry can induce a variety of novel properties, such as Rashba spin splitting, vertical piezoelectricity, and long exciton lifetime. Moreover, the intrinsic electric field that originates from the vertical asymmetry can serve as a superior platform for tuning the interlayer coupling when forming van der Waals structures. In this mini review, the recent key research progresses of 2D Janus TMDs, including the fundamental properties and potential applications, are briefly summarized, and the existing challenges are also presented.

1. Introduction

Structural symmetry breaking provides a remarkable platform for inducing a wealth of fascinating properties in 2D materials. For instance, inversion symmetry breaking engineered by a vertical electric field (EF) can control the electronic gap of graphene bilayer.[1] Twisted bilayer graphene with inversion asymmetry gives rise to tunable second harmonic generation.[2] Therefore, a large variety of strategies have been proposed to break the symmetry in 2D materials and engineer the electronic/magnetic/optical properties.[3–6] For instance, by surface covalent functionalization, Janus graphene was synthesized in experiments.[7,8] The electronic structures could be modulated by choosing n-type or p-type functional groups.[9] In addition, some other Janus 2D monolayers have also been extensively investigated, such as Janus graphene oxide nanosheets[10] janus silicene,[11] and asymmetric double-sided bilayer WSe2.[12] As a representative class of 2D materials, transition metal dichalcogenide (TMD) monolayers possess intrinsic in-plane (IP) asymmetry, and distinctive electronic and optical characteristics. For instance, the IP asymmetry generates finite Berry curvatures and orbital magnetic moments with opposite signs in two valleys, which lead to novel valley-dependent optical characteristics.[13]

The IP asymmetry, together with strong spin–orbit coupling (SOC) generates Zeeman-type spin splitting as well as valley polarization in TMD monolayers.[14]

Nevertheless, the out-of-plane (OOP) symmetry is well preserved in TMD monolayers, hindering their applications in certain areas. For example, even though TMD monolayers exhibit excellent stability, direct band structures, suitable bandgap, and high carrier mobility, the lack of Rashba SOC and electric-controlled spin precession impedes the application in spin field-effect transistors.[15–17] In addition, owing to the high exciton binding energy, the recombination of photogenerated electron–hole (e–h) is very fast in 2D TMDs. The dramatically low e–h separation efficiency would not facilitate light detection and harvesting applications.[18] As for catalytic performance, the overall catalytic activity of TMD-based catalysts is extremely limited due to the small proportion of active edge sites. Tremendous efforts have been devoted to maximally exposing the active edge sites or activating the inert basal sites, but with little improvement.[19,20]

The successful fabrications of Janus TMDs, an unconventional asymmetry sandwich structure, offer a plethora of new opportunities. The different electronegativity of top and bottom chalcogen atoms produces an internal perpendicular EF. The lack of OOP mirror symmetry is able to unlock novel features for physical and chemical applications.[21–24] In this review, we will first introduce the experimental synthesis of Janus TMD monolayers and highlight their novel properties induced by OOP asymmetry and potential applications. We then focus on the effect of the internal EF on the interlayer interactions in Janus van der Waals (vdW) structures. Finally, a conclusion and the existing challenges will be presented (Figure 1).
2. Recent Progresses of 2D Janus TMD Monolayers

2.1. Synthesis of 2D Janus TMD Monolayers

Janus TMD monolayers were first synthesized in 2017 with two different strategies. One proposed to utilize a modified MoS$_2$ whose top S layers are substituted by H and followed by Se powders vaporizing to achieve selenization. The H$_2$ plasma power and the selenization temperature play key roles in these processes. The other method starts from the MoSe$_2$ monolayer, followed by well-controlled sulfurization of the top Se layer (Figure 2a). To ensure top Se atoms are replaced, while the Se atoms remain intact, appropriate temperature and pressure are essential. Both synthetic approaches result in uniformly distributed asymmetric structure, paving the way for future applications. Recently, pulsed laser deposition (PLD) has been explored to implant Se clusters with low kinetic energy (<10 eV atom$^{-1}$) into WS$_2$ monolayer (Figure 2b). By controlling the kinetic energy, complete top sulfur layer replacement by selenium can be achieved, forming high-quality WS$_2$Se monolayers at 300 °C. These findings provide insights to extend the synthesis of other 2D Janus layers. However, the processing temperatures are relatively high in the abovementioned methods. Very recently, a room temperature method has been proposed to fabricate a variety of high-quality Janus TMD layers and their heterostructures such as vertical MoSSe/WSSe, lateral WSSe/MoSSe, and MoSSe/Mo$_2$S$_2$ by a selective epitaxial atomic replacement (SEAR) process (Figure 2c). This process relies on precursors, which helps efficient removal and substitution of the top chalcogen layer. Compared with previous methods, this technique shows significant advances to synthesize 2D Janus materials, and the mild processing temperature enables the formation of high-quality heterostructures. These breakthroughs will pave the way for exploring the fantastic physical phenomena of 2D Janus TMDs and transferring the novel features into cutting-edge applications.

2.2. Novel Properties of Janus TMD Monolayers

In contrast to traditional TMDs, Janus TMD monolayers have a chemical formula MXY (M = Mo, W; X, Y = S, Se, Te). As shown in Figure 3a, the transition metal layer in Janus TMD monolayer is sandwiched by two different chalcogen layers. Compared to conventional 2H-phase TMDs, the space group is transformed from D$_{3h}$ to C$_{3v}$. The vertical symmetry broken originates from the electronegativity difference of X and Y atoms will give rise to an intrinsic EF. Therefore, a variety of additional electronic, optical, and catalytic features will be unlocked, offering new opportunities for future applications.

2.2.1. Rashba Effect

Strong SOC is one of the most representative characteristics of semiconducting 2D TMDs, which is a relativistic effect that describes the spin splitting under an effective magnetic field. The SOC effect together with IP inversion asymmetry in TMD monolayer generates an effective Zeeman field, leading to opposite spins at the $+K$ and $-K$ points of the momentum space. The valley polarization could be applied to encode information. Due to the unique roles in locking the spin and valley degree of freedom, Ising SOC shows great potential in valleytronics and optoelectronic devices. However, in conventional TMD monolayers, Rashba SOC would not arise due to the protection of OOP mirror symmetry. Interestingly, it can appear in gated...
TMDs or Janus TMDs and lead to a plethora of new discoveries and useful applications. It is commonly expressed with the Hamiltonian $H_R = \alpha_R(\sigma \times k) \cdot \hat{z}$, where $\alpha_R$, $\sigma$, and $k$ denote Rashba parameter, electron's spin, and the electron momentum, respectively. The value of $\alpha_R$ is normally used to measure the Rashba strength. By first-principles investigations, it is found that in addition to valley coupling and Zeeman-type spin, the Rashba-type spin splitting (Figure 3b–d) can be observed around the $\Gamma$ point in Janus TMD monolayers.

The IP spin rotation pattern shown in Figure 3b confirms the Rashba-type spin polarization. The Rashba parameter $\alpha_R$ is sensitive to the SOC strength. Therefore, the splitting magnitudes depend on their constituent elements. For example, the calculated $\alpha_R^2$ values for MoSSe and WSSe are 77 and 158 meV, respectively. This can be understood by the larger SOC of W atom than Mo. Moreover, the Rashba effect could be controlled by external strain, EF, and charge doping. For instance, in WSeTe, it is observed that a small strain (~2%) can substantially change the Rashba SOC (~50%–50%) by modulating the W–Se interaction. The orbital selective external potential method further revealed that the change of W-$d_z^2$ and Se-$p_z$ orbital overlap played a key role in controlling the Rashba SOC.

2.2.2. Vertical Piezoelectricity

Large vertical piezoelectric polarization is another remarkable feature of Janus TMDs. It is known that 2D piezoelectric materials can offer applications in energy harvesting, actuator, sensors, and medical industries. For instance, the experimental studies demonstrated the strong piezoelectric response in stretched MoS$_2$ monolayer, which is comparable to those conventional piezoelectric materials, such as $\alpha$-quartz, AlN, and ZnO, implying the potential in energy and electric applications. The piezoelectric field, together with exciton properties in MoS$_2$ monolayers, may extend their optoelectronic applications. However, the piezoelectric responses are mainly obtained within the basal plane, ruling out the vertical operation mode. Strikingly, Shenoy et al. unveiled the OOP piezoelectricity in Janus TMD structures (Figure 3e). The piezoelectric effect in Janus MXY can be described by $e_{il}$ and $d_{il}$ tensors:

$$
e_{il} = \begin{pmatrix}
e_{11} & -e_{11} & \cdots & e_{15} \\
-\cdots & e_{15} & \cdots & -0.5e_{11} \\
e_{31} & e_{31} & e_{33} & \cdots
\end{pmatrix}
$$

$$
d_{il} = \begin{pmatrix}
d_{11} & -d_{11} & \cdots & d_{15} \\
-\cdots & d_{15} & \cdots & -2d_{11} \\
d_{31} & d_{31} & d_{33} & \cdots
\end{pmatrix}
$$

The high values of $e_{31}/d_{31}$ in Janus TMD monolayers indicate the remarkable vertical piezoelectric effect. Even though the vertical piezoelectric polarizations are much weaker than that of the IP piezoelectricity for monolayers, as indicated by the values of $e_{15}/d_{15}$, they can be dramatically enhanced in multilayers due to the vertical strain. As revealed by the theoretical results, the enhancement of vertical piezoelectricity could also be realized by interlayer sliding in Janus TMD bilayers.
piezoelectricity in Janus TMD structures will greatly expand the application’s scope.\(^{[40]}\)

### 2.2.3. Photovoltaic Properties

In the Janus TMD monolayer, the built-in vertical EF due to the asymmetry can weaken the excitons’ binding strength. The strong Coulomb screening and reduced wave function overlap between the \(e^{-} - h\) caused by the EF will significantly reduce the recombination process and prolong the exciton lifetime. By time-domain density functional theory (DFT) calculations, Jin predicted that the exciton lifetime in Janus-MoS\(_{2}\) monolayer can be up to 1.31 ns.\(^{[41]}\) Up to date, some appealing features of Janus TMDs as predicted from theoretical simulations have been verified in experiments. For instance, transient absorption results showed that the excitons are \(\approx 30\%\) faster than those in conventional TMDs. In addition, the radiative recombination lifetime of excitons is dramatically longer than their pristine counterparts. The results are originated from the built-in dipole moment, which can enhance electron–phonon interactions and separate the electron and hole wave functions (Figure 3f). These features are beneficial for efficiency improvement and charge collection.\(^{[42]}\)

### 2.3. Promising Applications of Janus TMD Monolayers

#### 2.3.1. Sensors

Due to the intrinsic dipole, which can either enhance or weaken the gas adsorption depending on the polarization direction, Janus TMDs exhibit unique potential in sensors. For example, Jin et al. proposed MoSSe as an ideal material for gas sensing by first-principles calculations.\(^{[43]}\) After examining the adsorption performance of the typical gas molecules, including \(\text{NH}_3\), \(\text{NO}\), \(\text{NO}_2\), \(\text{CO}\), and \(\text{CO}_2\), on Janus MoSSe monolayer, they found that N-based gases and C-based gases prefer to adopt chemisorption and physisorption, respectively (Figure 4a). Compared to S-layer, the binding strengths of these molecules are much stronger on Se-layer. The adsorption behavior can be effectively controlled by tensile strain due to the distinct change of the intrinsic electrostatic potential difference between the S and Se surfaces. Moreover, other works found that the OOP polarization in MoSSe makes it a universal substrate for biomolecule (such as glucose and dopamine) and food adulterant (such as histamine, formalin, and hydrogen peroxide) detecting.\(^{[44,45]}\) These appealing advantages pave the way for the application of sensing and detecting techniques.

![Figure 4. a) The adsorption distance and charge transfer of different gas molecules on Janus MoSSe and MoS\(_2\) monolayers. Adapted with permission.\(^{[43]}\) Copyright 2019 The Royal Society of Chemistry. b) HER volcano including WSSe. c) Schematic illustration of band alignment and crystal field of 2H-TMD and Janus TMD. Reproduced with permission.\(^{[46]}\) Copyright 2018, American Chemical Society. d) Spatial distribution of CBM and VBM in WSSe monolayer. Reproduced with permission.\(^{[47]}\) Copyright 2020, American Chemical Society.](https://www.advancedsciencenews.com/doi/abs/10.1002/pssb.202007734)
2.3.2. Applications in Hydrogen Evolution Reaction

TMDs have been regarded as promising low-cost catalysts. The overall catalytic performance of TMDs is deeply influenced by the edge sites. Most attempts were focused on maximizing edge sites or activating the basal plane, which are quite challenging as they are relying on the precise control of the edge sharp and atomic structures.[46–48] For example, Li et al. proposed to activate the basal layer of strained 2H-MoS2 with the S vacancies for hydrogen evolution. However, precisely controlling either S vacancy concentration or strain remains a significant challenge in the experiment.[49] In this respect, Janus TMDs can provide a unique solution to achieve high catalytic performance. By computationally screening a series of Janus TMDs, Shenoy and coworkers found that WSSe monolayer can be a possible catalyst for the hydrogen evolution reaction (HER) without external strain. The HER catalytic performance could be further improved by the presence of chalcogen vacancies (Figure 4b). The enhanced catalytic performance can be interpreted for the following reasons. First, the intrinsic lattice strain and inside EF due to the Janus asymmetry can change the crystal field and band structure. The energy of certain orbitals is affected by the Jahn–Teller distortion and pushed closer to the Fermi level (Figure 4c). Second, the chalcogen vacancies can be introduced in gap states near the Fermi level. These new freestanding bands enable stable H adsorption, which significantly improves the interaction between H and Janus monolayer and activates the basal plane.[50] From a high-through screening of different metal atoms deposited on defective MoSSe monolayer, Deng predicted the excellent HER performance in single Cd, Co, or Zn atom anchored on defective MoSSe.[51] Defective or non-metal atom (such as B atom) doped Janus TMDs also show superior performance for HER due to the presence of OOP polarizations.[52,53]

2.3.3. Photocatalytic Water Splitting

Photocatalysts with a wide-spectrum solar response and high e–h separation efficiency are highly desirable for water splitting. Janus TMDs are envisaged to be a potential candidate owing to the intrinsic EF, which is beneficial for photogenerated e–h separation. By systematically examining the optical absorption, e–h separation, photoexcited carrier migration, and surface redox reactions with DFT calculations, Ju et al. confirmed that Janus WSSe exhibits excellent performance for overall water splitting.[54] The bandgap of WSSe monolayer (2.13 eV from HSE06 calculations) well meets the requirement for water splitting, and has the optical absorbance within the visible-light spectrum. Especially, in this Janus structure, the conduction band minimum (CBM) and valence band maximum (VBM) are mainly located at the Se side and the S site, respectively (Figure 4d). The spatial separation can suppress the recombination of photoexcited carriers and separate the generated O2 and H2. Due to the benefit of internal EF, the exciton binding energy of WSSe monolayer is smaller than its conventional counterparts (WS2 and WSe2), while the carrier mobility is higher, facilitating carrier separation and transfer. Furthermore, the external strain can greatly improve the catalytic performance by controlling the intrinsic dipole. For example, the uniaxial strain could broaden the absorption to the near-infrared region. It can also lower the binding energy of exciton and tune the adsorption energy of H2O molecules. The potential for photocatalytic water splitting was also theoretically confirmed in Janus MoSSe.[55,56]

3. 2D Janus TMDs vdW Structures

In conventional TMDs, a direct to indirect bandgap transition will emerge when the thickness increases due to the interlayer orbital hybridization between the chalcogen atoms and metal atoms. Indeed, the interlayer interactions in vdWs structures play vital roles in determining the electronic, optical properties.[57] In analogy to pristine TMD vdW structures, the interlayer coupling also has a great influence on Janus TMD vdW structures. However, the built-in EF of Janus TMDs provides an additional degree of freedom to tune the interlayer coupling between neighboring 2D layers, leading to new physics and applications in Janus TMD vdW structures.[58–60]

3.1. Interlayer Coupling in Janus TMD vdW Structures

It is known that vdW bilayers exhibit versatile properties and provide new possibilities for electronics and optoelectronics. However, the short e–h recombination lifetime as a result of strong interlayer overlapping greatly hinders their applications. In this respect, the Janus-MoSSe bilayer shows a unique advantage. The built-in EF in Janus bilayer MoSSe can effectively separate the centers of the wave function, thus suppressing the strong interlayer coupling (Figure 5a). In this context, the e–h recombination time can be significantly extended. By using the nonadiabatic molecular dynamics (NAMD) technique, it was estimated that the lifetime for Janus bilayer can reach up to 16.5 ns.[61] It is also found that the carrier recombination in Janus-MoSSe bilayer is significantly affected by stacking orientations. Compared to S-Se/S-Se oriented MoSSe bilayers, the opposite dipoles in Se-S/S-Se and S-Se/Se-S oriented bilayers will significantly suppress the charge recombination due to the weaker interlayer interactions.[62] The interlayer coupling tuned piezoelectricity of Janus multilayers is another shining example. The theoretical studies show that the vertical lattice constant and stacking sequences have a significant influence on the piezoelectric coefficient ε33. The dipole–dipole interaction in vdW structures may change the charge distribution of adjacent layers, thus modulating the piezoelectric effect.

The interlayer coupling in Janus heterobilayers is also an interesting research topic. For instance, Janus MoSSe/MoS2 vdW was synthesized from a few-layer MoS2 recently. As revealed by the low-frequency Raman spectroscopy, in MoSSe/MoS2 heterostructure, the interlayer coupling is greatly enhanced (up to 13.2%) as compared to MoS2 bilayer. This phenomenon can be interpreted from the intrinsic dipole and compressive strain effect. On the one hand, the intrinsic dipole moment will induce the charge redistribution, resulting in reduced interlayer distance and extra dipole–Coulombic interactions. On the other hand, the lattice mismatch between two layers would make MoSSe under a compressive strain condition, which also contributes to the enhanced interlayer coupling (Figure 5b).[63]
Inspired by the great achievements in twisted TMD heterobilayers\cite{64,65} and magic-angle graphene,\cite{66} recently much attention has been devoted to manipulating interlayer coupling by twisting the angles between crystal axes. After carefully investigating the effects of twist angle and intrinsic dipole moment in Janus MoSSe/MoS$_2$ heterobilayers, Huang et al. have confirmed the ability of the Janus structure to tune the interlayer coupling and interfacial charge transfer by changing the interface composition and twist angle (Figure 5c).\cite{67}

### 3.2. Potential Applications of 2D Janus TMD vdW Structures

Stacking Janus TMD layers serves as a strategy to obtain ultrathin pn-junctions due to the Janus dipole effect, which presents an opportunity for solar energy harvesting devices. To construct a device, metallic leads are crucial. However, bulk metal would lead to a complete screen of built-in dipole and destroy the pn-junction. Using first-principles calculations, Brandbyge et al. show that the dipole effect is preserved in the stacked Janus-graphene system, and hereby provides a promising method to abrupt pn-junctions (Figure 5d), which has required harsh conditions previously. The upper and bottom graphene are strongly n- and p-doped, respectively. It indicates that the Janus dipole effect could also be employed as a nonintrusive way for graphene doping.\cite{68}

The excellent properties of Janus TMD vdW structures highlight the potential applications in electronics, optoelectronics, and photocatalysts.\cite{69} As the Rashba strength is dominated by the vertical EF applying on the metal atoms, it is reasonable to expect that the OOP orbital overlap between interfacial X and Y atoms will affect the Rashba effects. Indeed, theoretical simulations confirm that the Rashba effect is influenced by stacking orders and interlayer distance. Take the AA-stacked MoSSe bilayer for instance. In contrast to MoSSe monolayer, the Rashba effect of bilayer vanishes in the range of 3.2–3.7 Å. As the distance further decreases, the Rashba effect appears again (Figure 5e). This phenomenon can be interpreted by the competition between the interlayer and intralayer EF, which influences the net vertical dipole moment on Mo atoms.\cite{70}

Additionally, different stacking configurations in Janus TMD multilayers also have an influence on bandgap, dipole moment, and carrier mobility.\cite{70} Wang et al. theoretically predicted that MoSSe/WSeTe heterostructure can be a direct Z-scheme photocatalytic candidate for HER.\cite{71} DFT calculations indicate that due to the electrostatic potential difference in Janus structure, built-in EF would occur at the MoSSe/WSeTe interface, which would suppress charge recombination and facilitate e–h separation. The NAMD indicates that photogenerated carrier transfer may behave as type-II or Z-scheme path by different stacking. However, surface chalcogen vacancies can switch type-II path to Z-scheme path effectively. More interestingly, the appearance of these defect trap states will increase the time difference between carrier transfer and recombination, making MoSSe/WSeTe vdW structure a highly promising direct Z-scheme photocatalyst. As for MoSSe/WSe$_2$, this heterostructure exhibits not only ultrafast charge separation but also high photoresponsivity in wide ranges of the spectrum. These appealing features make it promising in optoelectronic and photovoltaic applications.\cite{72}
4. Conclusions and Prospects

As one of the most promising 2D candidates, TMDs have received notable attention due to the unique combination of structural stability, direct bandgap, strong SOC, and excellent mechanical and electric properties. The emerging Janus TMDs not only inherit the advantages of conventional TMDs but also unlock some fascinating phenomena such as the Rashba effect and vertical piezoelectricity due to the OOP symmetry breaking. In addition, the intrinsic EF that stems from the electronegativity difference of top and bottom chalcogen atoms can be regarded an ideal way to tune the interlayer interactions when forming Janus vdW structures, which opens new opportunities for applications. Furthermore, the interlayer coupling is also sensitive to the stacking configurations, interlayer distances, or twisting angles, which provide an additional way to control the properties. In this regard, we summarize some of the key theoretical and experimental results of 2D Janus TMD monolayers and present potential utilizations in sensors, HER, and photocatalytic water splitting. The exciting properties of 2D Janus vdW structures have also been summarized, which hold potentials for varieties of applications in nanoelectromechanics, (opto-) electronics, and catalysis.

Despite the great potential in wide ranges of areas, large-scale applications of Janus TMDs remain a problem. To date, the experimental studies of 2D Janus materials are still rare, mainly because of the difficulties in Janus layer synthesis as well as limited material quality. Janus layers need to be synthesized with extreme precision. Therefore most studies are based on theoretical simulations. Up to now, only a few kinds of Janus TMD monolayers (MoS\(_2\)/MoSe\(_2\), WS\(_2\)/WSe\(_2\)) have been fabricated. Therefore, many novel properties predicted in theory have not been confirmed in experiments. Thus, it is highly expected that more high-quality 2D Janus TMDs can be synthesized by optimizing existing conditions or designing new strategies. In practical applications, stability is a fundamental and important factor. Hence, deeper investigation of the stability of 2D Janus TMDs under different conditions is needed. In addition, as we mentioned, the internal dipole moment in Janus TMDs can act as an efficient way to tune the interlayer coupling and broaden their applications. The full potential still awaits further exploration. To sum up, although a number of problems have not been settled, current progress will set important milestones for more exciting discoveries and applications.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords

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