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First-principles investigations of the controllable electronic properties and contact types of type II MoTe₂/MoS₂ van der Waals heterostructures

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Two-dimensional (2D) van der Waals (vdW) heterostructures are considered as promising candidates for realizing multifunctional applications, including photodetectors, field effect transistors and solar cells. In this work, we performed first-principles calculations to design a 2D vdW MoTe₂/MoS₂ heterostructure and investigate its electronic properties, contact types and the impact of an electric field and in-plane biaxial strain. We find that the MoTe₂/MoS₂ heterostructure is predicted to be structurally, thermally and mechanically stable. It is obvious that the weak vdW interactions are mainly dominated at the interface of the MoTe₂/MoS₂ heterostructure and thus it can be synthesized in recent experiments by the transfer method or chemical vapor deposition. The construction of the vdW MoTe₂/MoS₂ heterostructure forms a staggered type II band alignment, effectively separating the electrons and holes at the interface and thereby extending the carrier lifetime. Interestingly, the electronic properties and contact types of the type II vdW MoTe₂/MoS₂ heterostructure can be tailored under the application of external conditions, including an electric field and in-plane biaxial strain. The semiconductor–semimetal–metal transition and type II–type I conversion can be achieved in the vdW MoTe₂/MoS₂ heterostructure. Our findings underscore the potential of the vdW MoTe₂/MoS₂ heterostructure for the design and fabrication of multifunctional applications, including electronics and optoelectronics.

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1 Introduction

The advent of two-dimensional (2D) materials, characterized by their atomic structures and unique properties, has ushered in a new era of exploration and innovation in condensed matter physics and materials science. This burgeoning class of materials, with graphene¹ as a pioneering example, has captivated both fundamental and industrial researchers. Following the success of graphene, a plethora of 2D materials have been exfoliated and investigated, both in terms of fundamental understanding and practical applications. Recently, the most extensively investigated 2D materials are transition metal mono- and di-chalcogenides,^{2–5} MXenes^{6–8} and graphitic carbon

nitrides.^{9–11} The versatility in the electronic and optical properties of 2D materials makes them promising candidates for multiple applications, ranging from advanced electronics to quantum computing and beyond.^{12–15}

More interestingly, the special versatility of 2D materials lies in their potential for creating 2D vdW heterostructures by stacking them together.^{16–18} The 2D vdW heterostructures based on 2D materials offer an intriguing platform for tailoring and enhancing material properties, unlocking novel phenomena, and paving the way for practical applications in next-generation devices.^{19–21} Recently, a plethora of 2D vdW heterostructures have been synthesized experimentally and explored computationally, for instance, TMD heterostructures,^{22,23} MXene heterostructures,^{24,25} MA₂Z₄ heterostructures^{26,27} and phosphorene heterostructures.^{28–30} Among them, the exploration of vdW heterostructures between different 2D TMD materials has received much more consideration and interest. Many 2D TMD-based vdW heterostructures have been successfully fabricated and explored, such as MoS₂/WSe₂,^{31,32} HfS₂/MoS₂,³³ MoS₂/WS₂,³⁴ MoTe₂/ReS₂ (ref. 35) and black phosphorus/MoS₂.³⁶ One can find that the 2D vdW TMD heterostructures can be synthesized in experiments by various strategies, including top-down³⁷ and bottom-up^{38,39} strategies.

Recently, a novel 2D vdW heterostructure based on MoTe₂ and MoS₂ TMD materials has been successfully fabricated in

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experiments by various methods, such as one-step CVD,⁴⁰ mechanical exfoliation^{41,42} and direct imprinting.⁴³ Using the one-step CVD technique, Ding *et al.*⁴⁰ fabricated a MoTe₂/MoS₂ heterostructure and demonstrated that the photodetector based on such a heterostructure exhibits outstanding photoresponsivity and external quantum efficiency. Lately, Ji *et al.*,⁴¹ utilizing mechanical exfoliation, fabricated a 2D vdW MoTe₂/MoS₂ heterostructure. These findings proved that such a heterostructure can be considered as a promising candidate for optoelectronic devices and integrated photonics. In addition, the MoTe₂/MoS₂ heterostructure can also be fabricated by combining the mechanical exfoliation and transfer methods⁴² or direct imprinting.⁴³ All these experimental findings highlighted the potential applications of MoTe₂/MoS₂ heterostructures for multifunctional devices, including electronics and optoelectronics. Despite experimental successes in the fabrication of MoTe₂/MoS₂ heterostructures, a comprehensive computational investigation into the depth of their atomic structure, electronic properties and the formation of contact types is notably lacking. Therefore, in this work, we perform first-principles calculations to design a MoTe₂/MoS₂ heterostructure and investigate its structures and electronic properties and the formation of type II band alignment. The impact of external conditions is also explored to examine the potential applications of the MoTe₂/MoS₂ heterostructure for multifunctional devices. Our findings underscore the potential of the vdW MoTe₂/MoS₂ heterostructure for the design and fabrication of multifunctional applications, including electronics and optoelectronics.

2 Computational methods

In this work, the first-principles calculations are performed using the Quantum Espresso simulation package.^{44,45} The geometric optimization process and electronic property calculations are performed in the framework of the Perdew–Burke–

Ernzerhof (PBE) functional⁴⁶ within the projector augmented-wave (PAW) pseudopotential.⁴⁷ A cut-off energy of 510 eV and a Monkhorst–Pack (9 × 9 × 1) *K*-point mesh are employed for all the processes and calculations. The hybrid Heyd–Scuseria–Ernzerhof (HSE) functional⁴⁸ is also employed to get a more accurate band gap value of materials. The weak vdW interactions that may occur in layered materials can be described by adding the long-range dispersion correction of the Grimme DFT-D3 method.⁴⁹ The convergence threshold for the force and energy in all the calculations is set at 0.01 eV Å^{−1} and 10^{−6} eV, respectively. A vacuum thickness of 27 Å is applied along the *z* direction of materials to avoid any unnecessary interlayer interactions. A dipole correction is also employed for all the calculation processes.

3 Results and discussion

We first investigate the atomic structure and electronic properties of MoX₂ (X = Te, S) monolayers. The atomic structures of MoX₂ are depicted in Fig. 1. The MoX₂ monolayer consists of an X–Mo–X layer, where a Mo atom is sandwiched between two X atoms on different sides. Similar to graphene, the MoX₂ monolayer shows a hexagonal atomic structure. The lattice parameters of MoTe₂ and MoS₂ monolayers are calculated to be 3.50 and 3.16 Å, respectively. These values are consistent with the experimental measurement.⁵⁰ The electronic band structures of MoS₂ and MoTe₂ monolayers are depicted in Fig. 1(c) and (d). Both MoS₂ and MoTe₂ monolayers exhibit semiconducting behavior with a direct band gap. The valence band maximum (VBM) and conduction band minimum (CBM) are located at the *K* point for both the MoS₂ and MoTe₂ monolayers. The calculated band gaps of the MoS₂ and MoTe₂ monolayers are 1.78/2.26 and 1.19/1.65 eV given by the PBE/HSE functional, respectively. It is obvious that the traditional PBE functional underestimates the band gap of 2D materials, while the hybrid HSE06 can provide a more accurate band gap. However, both

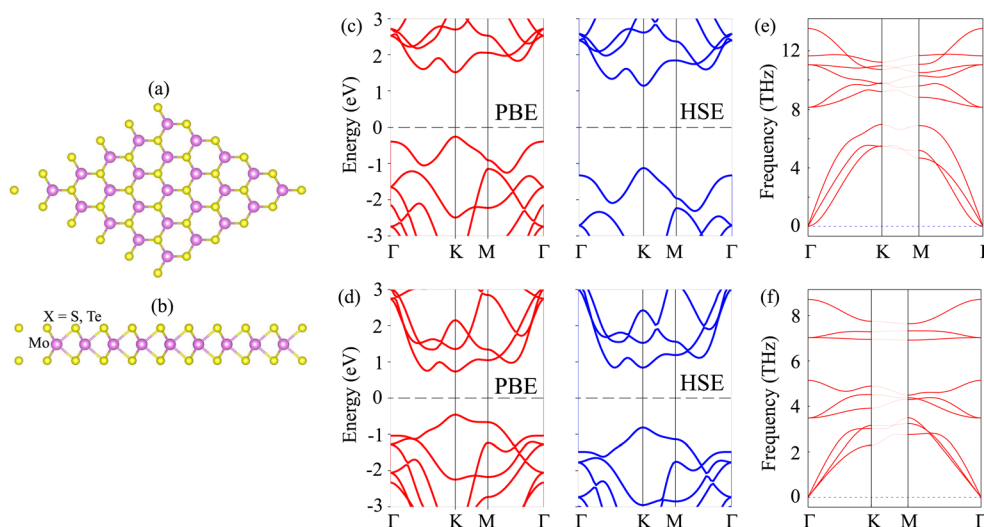


Fig. 1 (a) Top view and (b) side view of the atomic structures of a MoX₂ (X = S, Te) monolayer. Calculated electronic band structures of (c) MoS₂ and (d) MoTe₂ monolayers from PBE and HSE functionals. Phonon spectrum of (e) MoS₂ and (f) MoTe₂ monolayers.



the PBE and HSE06 functionals yield consistent behavior for the MoS₂ and MoTe₂ monolayers. Hence, we employed the PBE functional for the subsequent calculations due to its low computational resource. The phononic spectrum of MoS₂ and MoTe₂ monolayers is illustrated in Fig. 1(e) and (f). One can observe that there are no negative frequencies in the phonon spectrum of both the MoS₂ and MoTe₂ monolayers, predicting that these monolayers are dynamically stable.

The atomic structures of the MoTe₂/MoS₂ heterostructure are illustrated in Fig. 2. The MoTe₂/MoS₂ heterostructure is designed by using (2 × 2) unit cells of a MoS₂ monolayer and (√3 × √3) unit cells of a MoTe₂ monolayer. The lattice mismatch is calculated to be 2.1%, which is small and can be considered negligible. After geometric optimization, the interlayer spacing *d* between the two constituent MoTe₂ and MoS₂ monolayers is obtained to be 3.33 Å. This interlayer spacing is consistent with that in other typical vdW heterostructures^{51–53} and the experimental value.⁴² This indicates that the MoTe₂/MoS₂ heterostructure is characterized by weak vdW interactions. Furthermore, to examine the stability of the MoTe₂/MoS₂ heterostructure, we calculate the binding energy as follows:

$$E_b = \frac{E_H - \sum E_M}{A} \quad (1)$$

Here, *E_H* and *E_M* are the total energies of the MoTe₂/MoS₂ heterostructure and isolated MoX₂ (X = S, Te) monolayers, respectively. *A* stands for the surface area of the heterostructure. The *E_b* of the MoTe₂/MoS₂ heterostructure is obtained to be −32.23 meV Å^{−2}. The negative value of the binding energy indicates that the MoTe₂/MoS₂ heterostructure is structurally stable. In addition, we find that the value of the binding energy is consistent with that in graphite⁵⁴ and other vdW-based systems.^{55,56} All these findings confirm that the weak vdW interactions are mainly dominated at the interface of the MoTe₂/MoS₂ heterostructure. It is noteworthy that the weak vdW interactions keep the MoTe₂/MoS₂ heterostructure stable and can be synthesized in recent experiments by the transfer method⁴² or chemical vapor deposition (CVD) method.⁴⁰

Furthermore, to check the thermal and mechanical stability of the MoTe₂/MoS₂ heterostructure, we perform *Ab initio* molecular dynamics (AIMD) simulation and elastic constant calculation. The fluctuation in the total energy as a function of time steps of the MoTe₂/MoS₂ heterostructure is depicted in Fig. 3(a). It is evident that the change in the total energy of the MoTe₂/MoS₂ heterostructure before and after heating for 6 ps is small. Additionally, there is no distortion in the atomic structure of the MoTe₂/MoS₂ heterostructure after heating for 6 ps. All these findings confirm that the MoTe₂/MoS₂ heterostructure is thermally stable at room temperature of 300 K. The elastic constants *C_{ij}* of the MoTe₂/MoS₂ heterostructure are also calculated to evaluate its mechanical stability. The elastic constants of the MoTe₂/MoS₂ heterostructure are depicted in Fig. 3(b). The elastic constants of the constituent MoTe₂ and MoS₂ monolayers are also calculated for comparison. The calculated *C₁₁*, *C₁₂* and *C₆₆* of the MoTe₂/MoS₂ heterostructure are 264.18, 49.17 and 107.50 N m^{−1}, respectively. One can find that these values of the elastic constants satisfy the Born-Huang criteria,⁵⁷ confirming that the heterostructure is mechanically stable. Furthermore, it is evident that the elastic constants of the MoTe₂/MoS₂ heterostructure exhibit a substantial enhancement compared to those in the constituent monolayers. Besides, the Young's modulus of the MoTe₂/MoS₂ heterostructure is also greater than that of the MoTe₂ and MoS₂

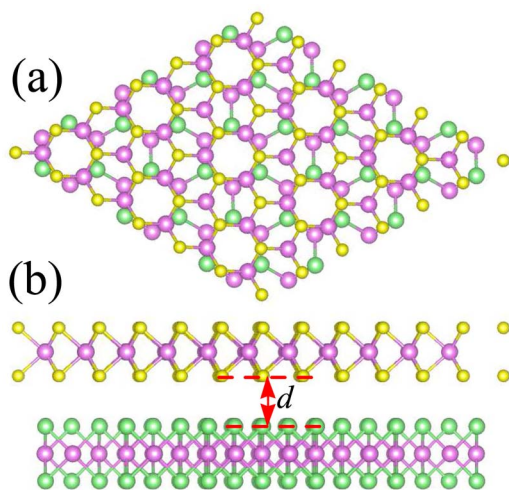


Fig. 2 (a) Top view and (b) side view of the atomic structure of the MoTe₂/MoS₂ heterostructure.

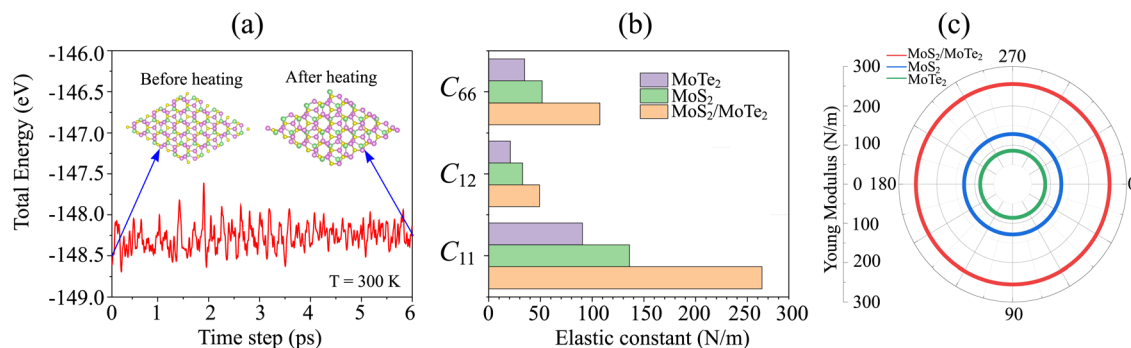


Fig. 3 (a) The fluctuation in the total energy as a function of time steps of the MoTe₂/MoS₂ heterostructure. The insets show the atomic structures of the MoTe₂/MoS₂ heterostructure before and after heating for 6 ps. (b) Elastic constants and (c) angle-dependent Young's modulus of the MoTe₂/MoS₂ heterostructure and the isolated MoTe₂ and MoS₂ constituent monolayers.



monolayers, exhibiting that the construction of the MoTe₂/MoS₂ heterostructure leads to an enhancement in the in-plane stiffness, as depicted in Fig. 3(c).

The projected band structure of the MoTe₂/MoS₂ heterostructure is depicted in Fig. 4(a). The MoTe₂/MoS₂ heterostructure possesses a semiconducting behavior with an indirect band gap. The VBM is located at the Γ point, whereas the CBM is at the K point. The band gap of the MoTe₂/MoS₂ heterostructure is 0.97 eV. Such a band gap is still smaller than that of both the constituent MoTe₂ and MoS₂ monolayers. This implies that the formation of the MoTe₂/MoS₂ heterostructure gives rise to a reduction in the band gap. A narrower band gap corresponds to stronger optical absorption. Therefore, the construction of the MoTe₂/MoS₂ heterostructure could lead to an enhancement in the optical properties. More interestingly, the band edges of the MoTe₂/MoS₂ heterostructure are contributed by distinct layers. The VBM is mainly contributed by the MoTe₂ layer, while the CBM comes from the MoS₂ layer. This finding suggests that the MoTe₂/MoS₂ heterostructure forms a type II band alignment. The formation of a type II band alignment was also observed in previous experiments.^{41,42} Therefore, the type II MoTe₂/MoS₂ heterostructure can be considered as a promising candidate for the design of optoelectronic and electronic devices, such as photodetectors and transistors.

We further consider the charge redistribution at the interface of the MoTe₂/MoS₂ heterostructure by analyzing the charge density difference (CDD) as follows:

$$\Delta\rho = \rho_{\text{H}} - \sum \rho_{\text{M}} \quad (2)$$

Here, ρ_{H} and ρ_{M} are the charge densities of the MoTe₂/MoS₂ heterostructure and isolated MoX₂ (X = S, Se) monolayers, respectively. The planar-averaged CDD of the MoTe₂/MoS₂ heterostructure is depicted in Fig. 4(b). The dark blue and dark orange regions represent charge accumulation and depletion, respectively. It is evident that the charges are mainly accumulated in the MoTe₂ layer and depleted in the MoS₂ layer. It indicates that the MoTe₂ gains electrons, while the MoS₂ layer loses electrons. The electrons flow from the MoS₂ to the MoTe₂ layer, whereas the holes are transferred in the opposite

direction, *i.e.* from the MoTe₂ to the MoS₂ layer. Bader charge analysis indicates that there is a small amount of charge transfer of about 10^{-3} electrons between the two constituent layers. Furthermore, to confirm the charge transfers in the MoTe₂/MoS₂ heterostructure, we also calculate the work functions of the MoTe₂/MoS₂ heterostructure and the constituent MoX₂ monolayers. The work function of a material can be calculated as: $\Phi = E_{\text{vac}} - E_{\text{F}}$, where E_{vac} and E_{F} represent the vacuum energy and Fermi energy, respectively. The work functions of the MoTe₂ and MoS₂ monolayers are calculated to be 4.76 eV and 4.08 eV, respectively. The lower work function of the MoS₂ monolayer compared to that of the MoTe₂ monolayer confirms that the electrons move from the MoS₂ to the MoTe₂ layer upon the formation of the heterostructure. The work function of the MoTe₂/MoS₂ heterostructure is calculated to be 4.98 eV, which is larger than that of the MoS₂ and MoTe₂ layers. The electrostatic potential of the MoTe₂/MoS₂ heterostructure is displayed in Fig. 4(c). One can find that the difference in the potential of the MoS₂ and MoTe₂ layers is small, verifying a small amount of charge transfer between the two layers. In addition, the potential of the MoS₂ layer is deeper than that of the MoTe₂ layer, confirming that the electrons move from the MoS₂ to the MoTe₂ layer. Such charge transfer leads to the formation of a built-in electric field, pointing from the MoS₂ to the MoTe₂ layer in their combined heterostructure.

Furthermore, we examine how the external conditions impact the electronic properties and contact types of the MoTe₂/MoS₂ heterostructure. Therefore, external electric fields and biaxial strains are applied to the heterostructure. The electric fields are applied along the z direction of the heterostructure, as depicted in the inset of Fig. 5(a). The positive direction of the electric fields is defined as from the MoTe₂ to the MoS₂ layer in their combined heterostructure. It is evident that an electric field can be used to modify the band gaps and change the contact types in the MoTe₂/MoS₂ heterostructure, as shown in Fig. 5(a). The band gap of the MoTe₂/MoS₂ heterostructure increases with applying a negative electric field and decreases with applying a positive electric field. The physical mechanism of such change can be described as follows: the direction of the built-in electric field is opposite to that of the negative electric

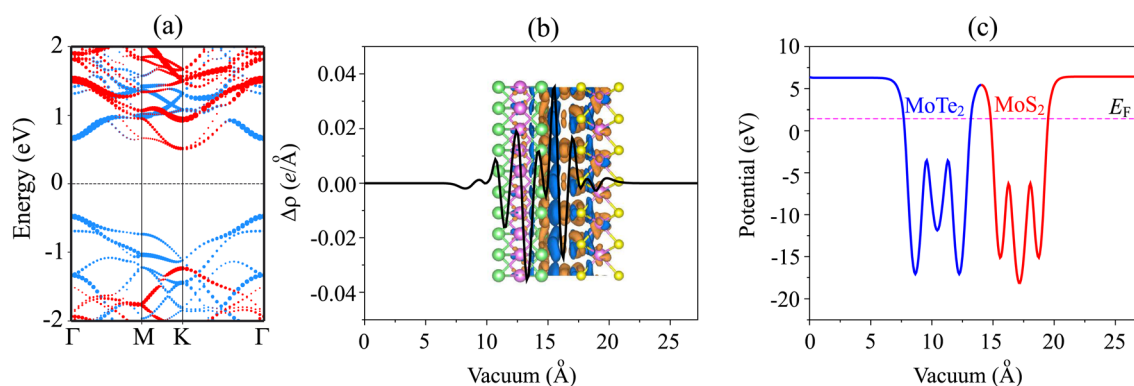


Fig. 4 (a) Projected band structure, (b) planar-averaged charge density difference and (c) electrostatic potential of the MoTe₂/MoS₂ heterostructure. Red and blue balls stand for the contributions of the MoS₂ and MoTe₂ layers, respectively. The dark blue and dark orange represent the charge accumulation and depletion, respectively.



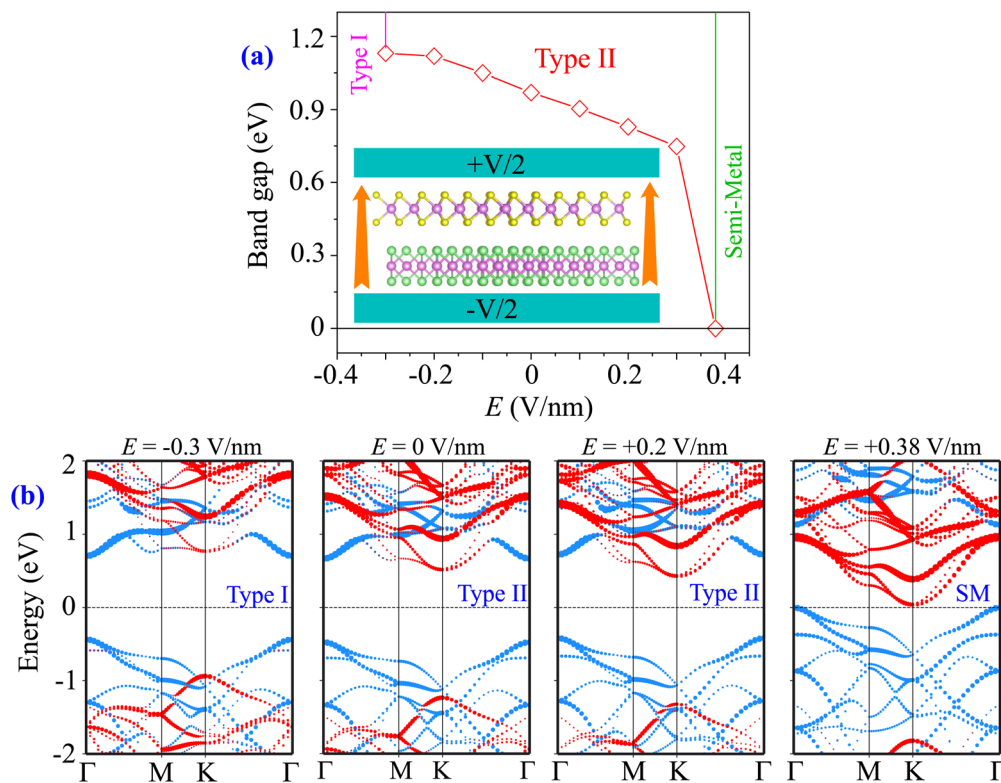


Fig. 5 (a) The variation of the band gaps and (b) the projected band structures of the MoTe₂/MoS₂ heterostructure under electric fields of different strengths. The inset shows the schematic model of applied electric fields along the z direction of the heterostructure.

field. Thus, the negative electric field can give rise to an enhancement in the band gap of the MoTe₂/MoS₂ heterostructure because the total electric field is weakened. On the other hand, the direction of the built-in electric field and positive electric field is the same. The total electric field is strengthened. Thus, the positive electric field causes a reduction in the band gap of the MoTe₂/MoS₂ heterostructure. The negative electric field can also lead to the transition from type II to type I band alignment, while the positive electric field gives rise to the semiconductor to semimetal transition. The underlying mechanism of these transitions can be described by analyzing the projected band structures of the MoTe₂/MoS₂ heterostructure under electric fields of different strengths, as depicted in Fig. 5(b). At a critical strength of the negative electric field of -0.3 V nm^{-1} , the CBM of the MoTe₂/MoS₂ heterostructure is located at the Γ point, as shown in Fig. 5(b). Such a CBM is contributed by the MoTe₂ layer, indicating that the negative electric field gives rise to a shift in the CBM of the MoTe₂/MoS₂ heterostructure from the MoS₂ to the MoTe₂ layer. Meanwhile, the VBM of the MoTe₂/MoS₂ heterostructure remains at the Γ point and is contributed by the MoTe₂ layer. These findings predict that the MoTe₂/MoS₂ heterostructure changes to form a type I band alignment. In addition, when a positive electric field is applied, both the CBM and VBM of the MoTe₂/MoS₂ heterostructure move towards the Fermi level, leading to a reduction in the band gap. At a critical strength of the positive electric field of $+0.38 \text{ V nm}^{-1}$, a transition from semiconductor to semimetal can be achieved in the MoTe₂/

MoS₂ heterostructure, as its VBM crosses the Fermi level. Furthermore, it should be mentioned that a high strength electric field can be generated from the tabletop terahertz source within an electrolyte top gate.⁵⁸ Additionally, a high strength electric field always requires high- k and a back (top)-gated device architecture.⁵⁹ All these findings prove that the electric field can be considered as an effective tool to manipulate the electronic properties and contact type of the MoTe₂/MoS₂ heterostructure, thereby expanding its potential applications in electronics and optoelectronics.⁶⁰

The biaxial in-plane strain is derived from $\varepsilon_b = (a - a_0)/a_0 \times 100\%$, where a and a_0 are the lattice parameters of the MoTe₂/MoS₂ heterostructure with and without the application of the biaxial strain, respectively. The negative and positive values refer to the compressive and tensile strains, respectively. The schematic model of the in-plane biaxial strain is depicted in the inset of Fig. 6. It is obvious that the strain causes a change in both the band gap values and contact types of the MoTe₂/MoS₂ heterostructure. The biaxial strain gives rise to a reduction in the band gap of the MoTe₂/MoS₂ heterostructure, as depicted in Fig. 6(a). The band gap of the MoTe₂/MoS₂ heterostructure can be reduced down to zero under the application of either a compressive strain of -12% or a tensile strain of $+16\%$. This observation indicates that the transition from semiconductor to metal can be achieved in the MoTe₂/MoS₂ heterostructure under the application of biaxial strain. Additionally, the tensile strain can also lead to the transformation between type II and type I band alignment in the MoTe₂/MoS₂ heterostructure.



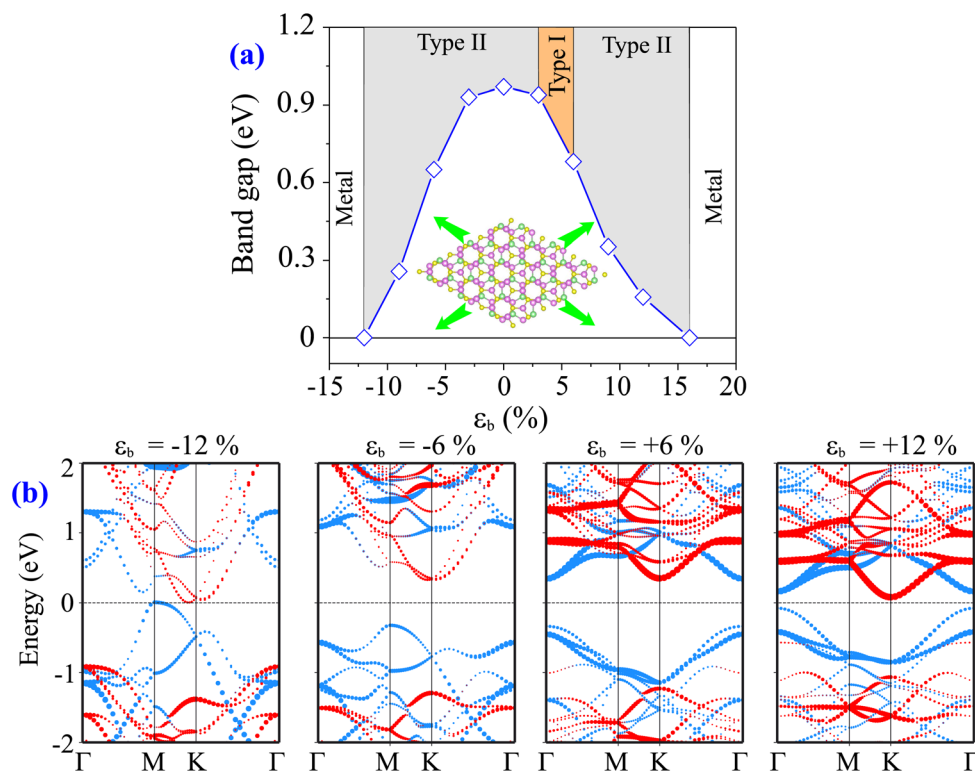


Fig. 6 (a) The variation of the band gaps and (b) the projected band structures of the MoTe₂/MoS₂ heterostructure under different ratios of strain. The inset shows the schematic model of the applied in-plane biaxial strain to the heterostructure.

To have a better understanding of the impact of the strain, we further analyze the projected band structures of the MoTe₂/MoS₂ heterostructure under different strain ratios, as illustrated in Fig. 6(b). When the compressive strain is applied, both the VBM and CBM of the MoTe₂/MoS₂ heterostructure shift towards the Fermi level, giving rise to a reduction in the band gap values. Similarly, the band edges of both the MoTe₂ and MoS₂ layers in the MoTe₂/MoS₂ heterostructure move closer to the Fermi level under the tensile strain. Under a tensile strain of $\epsilon_b = +3\%$, the CBM of the MoTe₂/MoS₂ heterostructure shifts from the *K* to the Γ point. Thus, the indirect-to-direct transition is achieved in the MoTe₂/MoS₂ heterostructure. In addition, both the VBM and CBM of the MoTe₂/MoS₂ heterostructure now come from the MoTe₂ layer, indicating that there occurs a transition from type II to type I band alignment. The type I band alignment is maintained in the MoTe₂/MoS₂ heterostructure under tensile strains ranging from +3% to +6%. When the tensile strain is larger than +6%, the CBM of the MoTe₂/MoS₂ heterostructure is recovered from the Γ to the *K* point, while the VBM is preserved at the Γ point. This recovery indicates that there is a transition from direct to indirect semiconductor and a conversion from type I to type II band alignment because the band edges of the MoTe₂/MoS₂ heterostructure are contributed by the MoTe₂ layer. When the tensile strain is larger than 16%, the band edges of the MoTe₂/MoS₂ heterostructure cross the Fermi level, leading to a transition from semiconductor to metal. Our findings prove that the in-plane biaxial strain can effectively be used to tailor the electronic properties and contact types in the

MoTe₂/MoS₂ heterostructure, thereby expanding its potential applications in multifunctional devices.

4 Conclusions

In conclusion, we have performed first-principles calculations to design a 2D vdW MoTe₂/MoS₂ heterostructure with the formation of a type II band alignment. Our results indicate that the MoTe₂/MoS₂ heterostructure is structurally, thermally and mechanically stable. The weak vdW interactions are found to be dominated at the interface of the MoTe₂/MoS₂ heterostructure and thus it can be synthesized in recent experiments by the transfer method or chemical vapor deposition. The vdW MoTe₂/MoS₂ heterostructure exhibits a staggered type II band alignment, effectively separating the electrons and holes at the interface and thereby extending the carrier lifetime. Furthermore, our findings reveal that the electronic properties and contact types of type II vdW MoTe₂/MoS₂ heterostructures can be tailored under the application of external conditions, including an electric field and in-plane biaxial strain. The semiconductor–semimetal–metal transition and type II–type I conversion can be achieved in the vdW MoTe₂/MoS₂ heterostructure. Our findings underscore the potential of the vdW MoTe₂/MoS₂ heterostructure for the design and fabrication of multifunctional applications, including electronics and optoelectronics.

Conflicts of interest

There are no conflicts to declare.



Acknowledgements

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