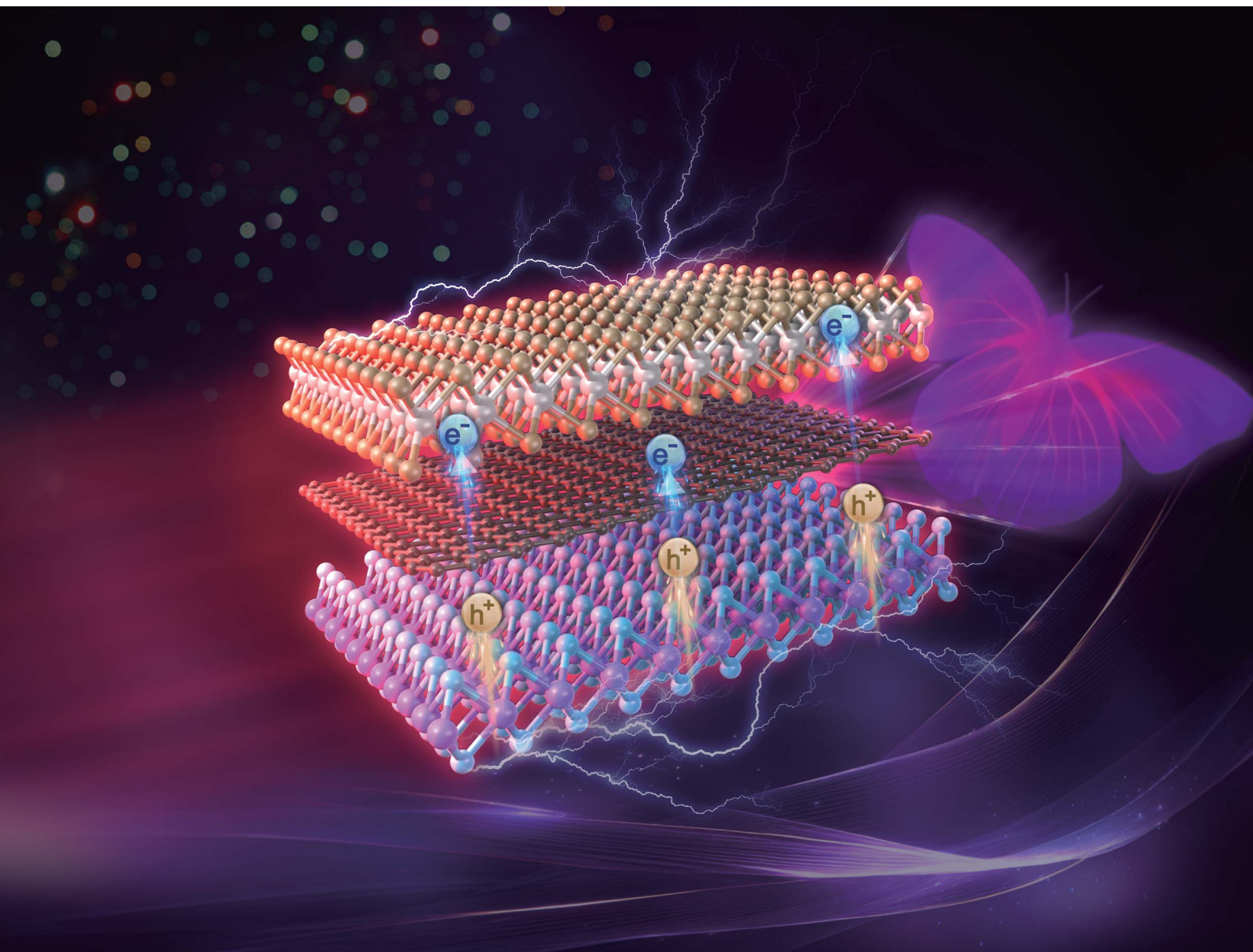


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**PAPER**

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# Electrostatically enhanced infrared absorption in two-dimensional van der Waals structures

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Two-dimensional (2D) semiconductors exhibit strong light absorption, yet their large bandgaps limit broadband photodetection. While van der Waals (vdW) heterostructures of 2D materials enable infrared excitation through narrowed interlayer bandgaps, achieving efficient control over these interlayer transitions remains a fundamental challenge. Through first-principles simulations and electrostatic engineering approaches, this study establishes a direct correlation between interfacial charge redistribution and enhanced interlayer excitations in 2D vdW structures. The results demonstrate that versatile strategies such as external electric field application, substitutional doping, and graphene interlayer integration effectively reduce interlayer bandgaps by several hundred millielectronvolts while significantly increasing interfacial charge exchange. These engineered heterostructures achieve exceptional absorption coefficients greater than  $10^5 \text{ cm}^{-1}$  spanning visible to mid-infrared wavelengths. These findings provide essential design principles and offer engineering strategies for developing broadband photodetectors using 2D vdW structures.

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## 1 Introduction

Two-dimensional (2D) semiconductors exhibit exceptional light-matter interaction and high carrier mobility within their ultrathin atomic layers, making them attractive for photodetectors that demand both high sensitivity and rapid response.<sup>1,2</sup> Strong photon absorption across a broad spectral range underpins high-performance photodetection at the nanoscale.<sup>3</sup> Vertically stacked heterostructures formed through van der Waals (vdW) assembly of 2D layered materials present several unique advantages for photodetection applications,<sup>4</sup> including atomically sharp interfaces, freedom from lattice-matching constraints, and tunable band alignment properties. These features collectively enable enhanced photodetector performance in the visible spectrum, as demonstrated by high responsivities,<sup>5</sup> fast response speed,<sup>6</sup> low dark current, and self-powered operation.<sup>7</sup>

Typically, vdW structures assembled from 2D materials with differing bandgaps exhibit a staggered (type-II) band alignment. This configuration facilitates charge transfer across the atomically sharp junctions and induces built-in electric fields influencing carrier dynamics.<sup>8,9</sup> In such heterostructures, the interlayer bandgap can be several hundred millielectronvolts (meV) smaller than the intrinsic bandgaps of the constituent monolayer. This reduced optical bandgap has enabled interlayer exciton recombination and subsequent infrared photon emission through ultrafast excitonic processes at cryogenic

temperatures, for example, in heterostructures consisting of large bandgap  $\text{MoSe}_2$  and  $\text{WSe}_2$  when pumped with green lasers.<sup>10–13</sup> In contrast, interlayer absorption as a reverse process, where an incoming photon excites electron-hole pairs spatially separated in two 2D layers, encounters significant inefficiency challenges. Such excitonic absorptions require extremely high incident light intensities, often exceeding  $10^{10} \text{ W cm}^{-2}$ , which are orders of magnitude greater than typical photodetector operating conditions ( $\text{nW cm}^{-2}$  to  $\text{mW cm}^{-2}$ ).<sup>14</sup> This stark contrast in light intensity illustrates the severe inefficiency of exciton-mediated interlayer-bandgap absorption for high-performance infrared photodetection applications.

Furthermore, excitonic effects in 2D materials, which are typically weakly bound and require cryogenic operating temperatures, contribute to sub-bandgap absorption only about 0.1 eV below the band edge. Given that most 2D semiconductors have bandgaps between 1 and 2 eV, this limited spectral extension, combined with the stringent excitation conditions, makes excitonic mechanisms impractical as an engineering route for high-performance photodetection device applications. As a result, the absorption spectra of 2D vdW heterostructures remain largely confined to the visible and near-infrared range, and practical strategies to extend absorption threshold into the mid-infrared *via* interlayer excitons have seen limited progress.<sup>15–17</sup>

These fundamental limitations highlight the need for alternative approaches, such as band engineering through a combined heterostructure design and electrostatic modulation. This makes the strategic design and control of the

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electronic structure of 2D vdW heterostructures essential for achieving tunable and efficient absorption beyond the limitations of intrinsic excitonic processes. Furthermore, efficient interlayer tunneling and momentum conservation are essential for strong interlayer excitations,<sup>18,19</sup> which in turn are vital for high-response photodetection. However, many existing studies rely on multilayer 2D materials with indirect bandgaps, which suffer from momentum mismatch and reduced quantum yield.<sup>20,21</sup> Moreover, their small bandgaps can lead to significant dark current, thereby degrading the sensitivity and signal-to-noise ratio of the photodetector. Moreover, exploring different 2D material combinations alone remains challenging because few materials simultaneously offer transparent tunneling barriers and narrow bandgaps, both of which are critical for efficient infrared photon absorption *via* interlayer excitons. To address these gaps, researchers have turned to interface dielectric engineering and gate biasing to tailor optical responses.<sup>22–25</sup> In particular, interfacial electric fields have emerged as a key lever for boosting photocarrier separation, underscoring the intimate link between interface electrostatics and optical absorption in 2D vdW heterostructures.

This paper adopts a band-structure-based derivation of the absorption process to examine how band structures and interface electrostatics deterministically influence infrared optical absorption in vdW structures composed of well-established 2D monolayer materials for photodetector applications. Through first-principles simulations, this study demonstrates that deliberate tuning of the band alignment effectively modulates interlayer excitations, thereby enabling efficient infrared absorption. In particular, the work examines the impact of electric biasing and doping on regulating the band alignment and interlayer excitation in 2D vdW structures, providing device engineering strategies to optimize infrared absorption for high-performance photodetection applications.

## 2 Band alignment and optical absorption of 2D vdW heterostructures

Band diagrams are essential for understanding the electronic and optical transition properties of semiconductor devices. Key features such as the conduction band minimum ( $E_C$ ), valence band maximum ( $E_V$ ), and the Fermi level ( $E_F$ ) provide insights into barrier distribution and carrier dynamics. In contrast to bulk junctions, vdW junctions assembled from atomically thin 2D layers exhibit negligible band bending. Instead, their band alignment is set directly by interfacial charge transfer, yielding electrostatic and optical properties that differ markedly from those of isolated monolayers. To accurately capture the charge redistribution and its impact on band alignment, this work correlated the first-principles simulation results, including the band structure, projected density of states (DoS), and localized DoS, with a common energy axis to construct the band diagrams of each 2D vdW heterostructure and calibrate the bandgap and band alignment properties with those reported in the literature.<sup>9,17</sup>

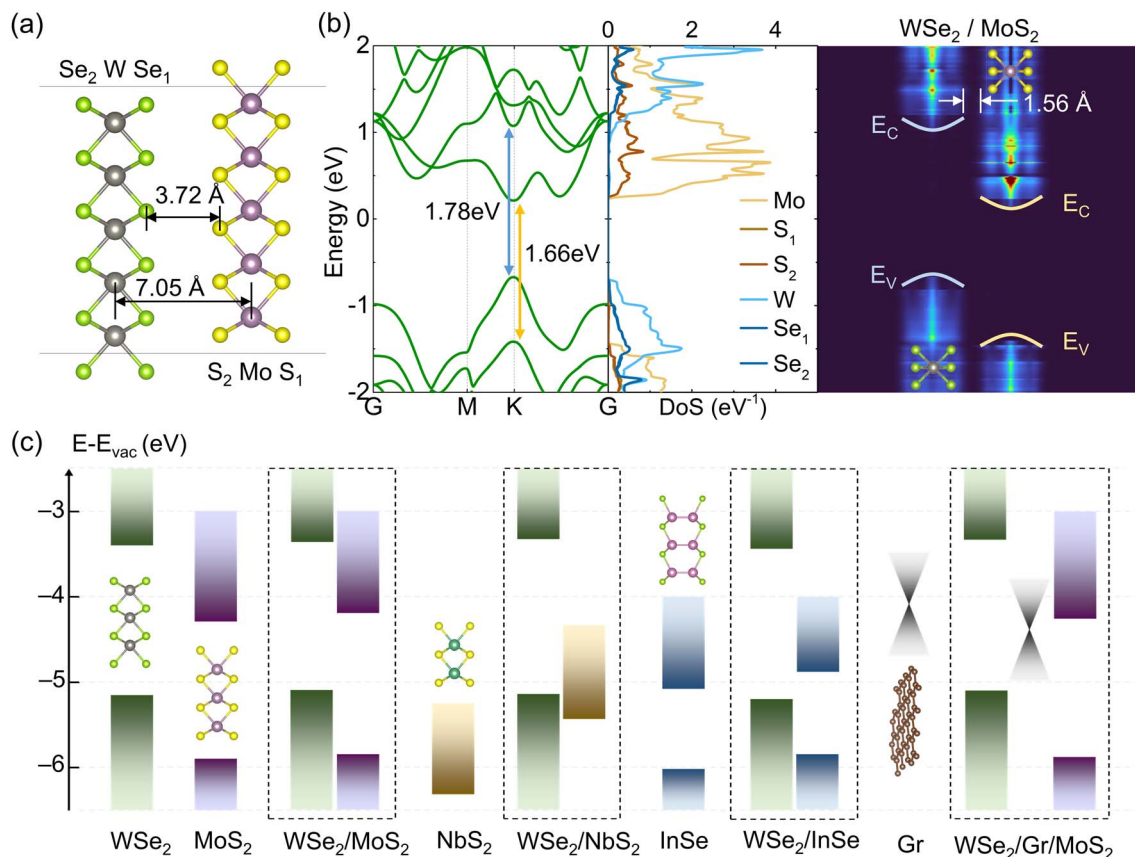
Without loss of generality, the electronic structures of a MoS<sub>2</sub>/WSe<sub>2</sub> vdW structure are investigated as a representative example. Fig. 1(a) displays the relaxed atomic structure. The vdW gap or interlayer distance, measured between the centers of interfacial Se<sub>1</sub> in WSe<sub>2</sub> and S<sub>2</sub> in MoS<sub>2</sub> atoms, is 3.72 Å, while the separation between the Mo and W atom centers is 7.05 Å. Fig. 1(b) presents the band structure, projected DoS spectrum, and localized DoS mapping for the MoS<sub>2</sub>/WSe<sub>2</sub> heterostructure. The results reveal a distinct direct bandgap at the high-symmetry *K*-point of the vdW structure, with  $E_C$  originating from the MoS<sub>2</sub> layer and  $E_V$  from the WSe<sub>2</sub> layer. The spatial separation of conduction and valence bands arises from a staggered-gap alignment and results in a reduced energy gap of the vdW heterostructure for interlayer excitons.<sup>26</sup>

Notably, the energy levels at which the DoS of transition metal atoms (Mo and W) diminish correspond to  $E_C$  and  $E_V$ , indicating bandgap values of 1.78 eV for monolayer WSe<sub>2</sub> and 1.66 eV for monolayer MoS<sub>2</sub>, in good agreement with experimental results.<sup>17</sup> The tunnelling layer between MoS<sub>2</sub> and WSe<sub>2</sub>, identified as the interfacial dark gap in the localized DoS map, has a thickness of 1.56 Å. This region contains no allowed states and forms a tunnelling barrier that impedes interlayer charge transfer and exciton transitions, which is a universal feature in vdW structures, typically 1.5 Å to 2 Å wide.<sup>27</sup> These values are consistent with experimental measurements reported in high-resolution TEM studies of the MoS<sub>2</sub>/WSe<sub>2</sub> heterostructure,<sup>28</sup> validating the computational model.

Several 2D materials used for photodetection applications, including MoS<sub>2</sub>, WS<sub>2</sub>, WSe<sub>2</sub>, InSe, NbS<sub>2</sub>, and graphene, in the form of free-standing monolayers as well as combined double-layer and triple-layer vdW structures, were simulated in this work. The lattice parameters for the unit cells are listed in Table 1. As shown in Fig. 1(c), the band diagrams of various 2D vdW heterostructure combinations alongside those of their free-standing monolayers, with calculated energy levels and band alignments consistent with the literature.<sup>9,17</sup> All atomic structures were initially acquired from well-established databases (<https://www.materialsproject.org>) and subsequently rescaled to specific unit cell configurations, with each undergoing lattice constant adjustments of less than 2.5%. For structure modeling, periodic boundary conditions were applied to simulate an extended 2D film, and a vacuum layer exceeding 20 Å was implemented along the *z*-axis (out-of-plane direction) for every simulation unit cell to inhibit inter-cell charge exchange.

The supercells were relaxed using first-principles molecular dynamics with the conjugated gradient method, achieving an energy convergence of 10<sup>−6</sup> eV and a force convergence of 0.02 eV Å<sup>−1</sup>. Self-consistent field (SCF) calculations were conducted using the projector-augmented wave (PAW) method with the Perdew–Burke–Ernzerhof (PBE) exchange and correlation functional within the generalized gradient approximation. A gamma-centered *k*-point grid was adopted for SCF calculations. To further confirm the band alignment of the 2D vdW heterostructures, we additionally verified our simulation results by calculating the band structures using the hybrid functional HSE06, with the results shown in supplementary Fig. S1. These





**Fig. 1** (a) Atomic structure of the MoS<sub>2</sub>/WSe<sub>2</sub> bilayer vdW junction. (b) The band structure, projected DoS spectrum, and localized DoS distribution of the MoS<sub>2</sub>/WSe<sub>2</sub> vdW structure simulated in this work. (c) The simulated band alignment diagrams for the vdW heterostructure comprised widely studied 2D materials.

**Table 1** Lattice constants and *k*-point grids used for the simulation unit cells of 2D vdW structures

| Atomic structures     | MoS <sub>2</sub> /WSe <sub>2</sub> | NbS <sub>2</sub> /WSe <sub>2</sub> | InSe/WSe <sub>2</sub> | MoS <sub>2</sub> /Gr/WSe <sub>2</sub> |
|-----------------------|------------------------------------|------------------------------------|-----------------------|---------------------------------------|
|                       |                                    |                                    |                       |                                       |
| Lattice constant (Å)  | 9.708                              | 6.636                              | 16.485                | 9.558                                 |
| <i>k</i> -point grids | 8 × 8 × 1                          | 12 × 12 × 1                        | 6 × 6 × 1             | 9 × 9 × 1                             |

simulations were performed using DS-PAW and Nanodcal software developed by Hongzhiwei Technology.<sup>29</sup>

The simulation results reveal that most vdW structures exhibit staggered band alignments, with energy level shifts relative to their free-standing forms. For example, the conduction band offset in WSe<sub>2</sub>/MoS<sub>2</sub>, WSe<sub>2</sub>/InSe, and WSe<sub>2</sub>/NbS<sub>2</sub> bilayer heterostructures is reduced by tens to hundreds of meV compared to that of the free-standing monolayers, which is evidence of a built-in potential at the junction interface. When two atomically thin layers come into contact, their Fermi levels equilibrate through charge transfer or charge screening, creating an interfacial dipole. This redistributed charge alters

the Coulomb interaction between layers, directly tuning the interlayer coupling strength. The magnitude of charge transfer is set by the Fermi-level shift ( $\Delta E_F$ ) between the constituent layers in their free-standing forms and their coupled forms in the vdW heterostructure. These energy potential variations are quantitatively comparable to the first-principles simulation results. These findings demonstrate that charge exchange governs interlayer interaction strength in 2D vdW systems, with the degree of coupling determined by the initial Fermi level alignment of the constituent monolayers.

To clarify this mechanism, this work explicitly compares the InSe/WSe<sub>2</sub> and NbS<sub>2</sub>/WSe<sub>2</sub> structures, which exhibit staggered-

gap (type-II) and broken-gap (type-III) band alignments, respectively. Fig. 2(a) and (d) display the changes in effective potential ( $\Delta V_{\text{eff}}$ ) and redistribution of charge density ( $\Delta\rho$ ) in the constituent 2D layers of the vdW junction. The InSe/WSe<sub>2</sub> heterostructure, composed of two 2D monolayers with closely matched charge neutrality levels, exhibits a weak interfacial dipole, suggesting suppressed interlayer charge exchange. In contrast, the NbS<sub>2</sub>/WSe<sub>2</sub> heterostructure exhibits a pronounced interfacial dipole that generates a large built-in potential, shifting the conduction band of NbS<sub>2</sub> upward by approximately 0.9 eV, resulting in p-type doping of the WSe<sub>2</sub> layer.

The linear potential drop across the vacuum region of the unit cell arises as a secondary effect of charge redistribution within the 2D heterostructure, and it follows Poisson's equation. For instance, in the WSe<sub>2</sub>/NbS<sub>2</sub> heterostructure, charge transfer leads to accumulated positive charge in WSe<sub>2</sub> and negative charge in NbS<sub>2</sub>, and such charge density can penetrate through the monolayers and cause residue charge density on the top and bottom surface of the vdW structure. Due to the use of periodic boundary conditions in all spatial directions in simulation, the residue negative charges in NbS<sub>2</sub> generate an electric field that extends across the vacuum region, terminating at the residue positive charges in WSe<sub>2</sub> in neighboring unit cells. To aid understanding, we have included in the supplementary document an illustration of the periodic boundary conditions used in the atomistic simulations, along with a schematic showing the redistributed charges, the induced electric field, and the resulting changes in the potential profile, presented as supplementary Fig. S2. Since the vacuum

region contains no free charges, the electric field remains constant within it. Notably, because of the thick (>20 Å) vacuum spacer, this electric field does not cause significant dielectric screening or perturb the intrinsic electronic structure of the vdW heterostructures.

These interlayer interactions are further illustrated in the band structures presented in Fig. 2(b) and (e). The band structures, projected onto WSe<sub>2</sub>, InSe, and NbS<sub>2</sub> (colored in blue, yellow, and orange), reveal significant joint dispersion relations near the high-symmetry Gamma-point in both heterostructures. These correlated  $E$ - $k$  dispersion relations provide additional paths for momentum-conserved carrier exchange and interlayer excitations. For example, the NbS<sub>2</sub>/WSe<sub>2</sub> heterostructure not only supports the interband transition of WSe<sub>2</sub> from  $E_V$  to  $E_C$ , but also creates an interlayer excitation pathway at the high-symmetry  $K$ -point, from the  $E_V$  of WSe<sub>2</sub> to the conduction band levels of NbS<sub>2</sub>. A similar effect is observed for the InSe/WSe<sub>2</sub> heterostructure. The interband transitions occur inherently within monolayer WSe<sub>2</sub> and InSe, as indicated by the excitation paths from  $E_V$  to  $E_C$  of each layer, while an additional allowed excitation pathway arises at the Gamma-point due to the joint dispersion relations.

Fig. 2(c) and (f) compare the absorption spectra of the NbS<sub>2</sub>/WSe<sub>2</sub> and InSe/WSe<sub>2</sub> heterostructures with those of their free-standing layers. Both vdW heterostructures exhibit high absorption coefficients ( $>10^5 \text{ cm}^{-1}$ ) in the ultraviolet and visible spectral ranges, arising from inherent intralayer excitations from  $E_V$  to  $E_C$  within the same material, and show notably enhanced absorption in the infrared spectral range due to

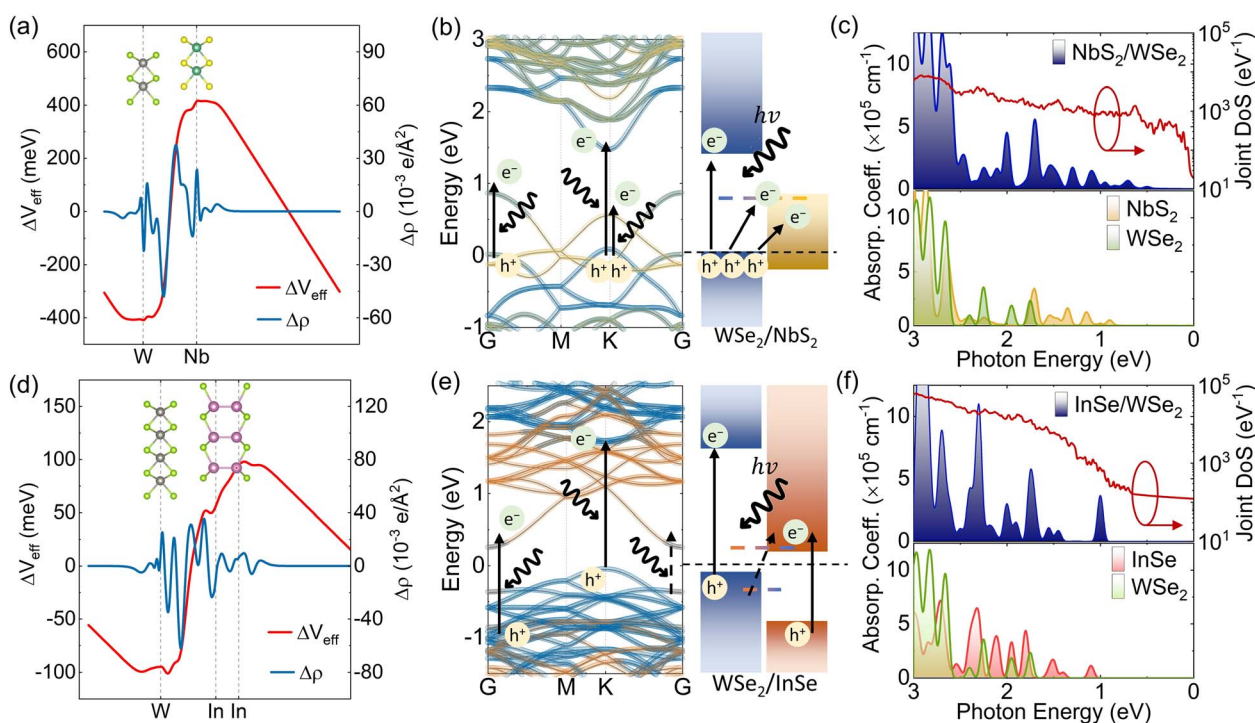


Fig. 2 (a) Effective potential variation ( $\Delta V_{\text{eff}}$ ) and charge redistribution ( $\Delta\rho$ ), (b) band structure and band diagram, and (c) absorption coefficient and joint DoS for the NbS<sub>2</sub>/WSe<sub>2</sub> vdW junctions. (d) Effective potential variation and charge redistribution, (e) band structure and band diagram, and (f) absorption coefficient and joint DoS for the InSe/WSe<sub>2</sub> vdW junctions.

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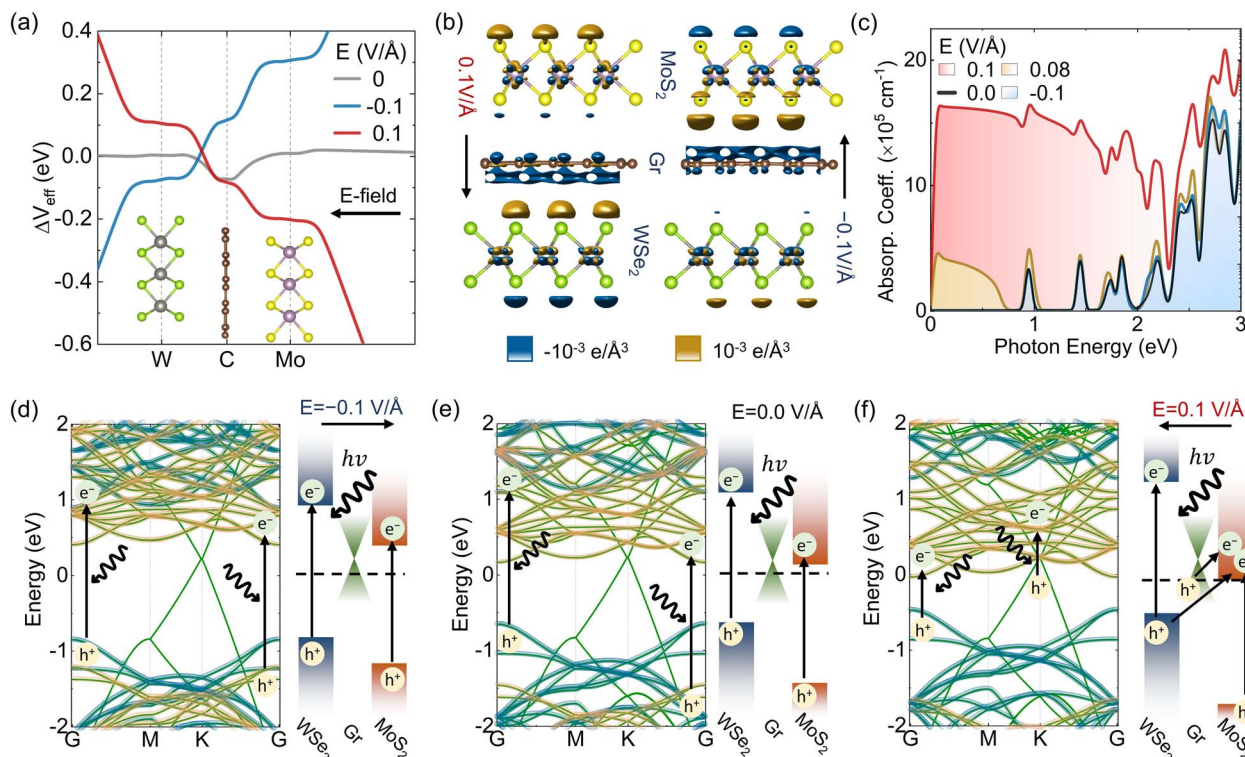


Fig. 3 (a) Effective potential variations ( $\Delta V_{\text{eff}}$ ) for the MoS<sub>2</sub>/Graphene/WSe<sub>2</sub> (MGW) junction under electric field biases. (b) Charge redistributions ( $\Delta\rho$ ) under electric field biases. (c) Absorption spectra of the MGW junction and corresponding band diagrams under electric fields of (d)  $-0.1 \text{ V Å}^{-1}$ , (e)  $0.0 \text{ V Å}^{-1}$ , and (f)  $+0.1 \text{ V Å}^{-1}$ .

interlayer bandgap and promotes interlayer charge transfer in 2D vdW heterostructures facilitates stronger interlayer excitations, resulting in enhanced absorption coefficients and broadened infrared spectral bandwidths. In addition, selecting 2D materials that naturally form heterostructures with smaller interlayer bandgaps may allow such effects to be achieved using lower electric fields, enabling more efficient infrared absorption through engineered band alignment. Furthermore, leveraging a positive bias to enhance infrared absorption and a negative bias to suppress unwanted carrier transitions provides a strategic pathway to enhance the spectral range and sensitivity of 2D vdW heterostructure-based photodetectors.

Beyond gate biasing, substitutional doping provides a robust alternative for generating substantial interfacial charge and inducing strong interfacial electric fields in 2D materials. This approach offers advantages such as simplifying device architecture and reducing parasitic effects.<sup>40–43</sup> Therefore, substitutional doping in 2D materials is regarded as a favorable technique and a key building block in the roadmap for advanced device applications.<sup>44</sup> Substitutional doping with Re and Fe (for n-type), and Nb, V and Ta (for p-type), which are compatible with the host lattice, has proven to be an exceptionally efficient approach for modulating charge concentrations in many 2D materials, such as MoS<sub>2</sub>, MoSe<sub>2</sub>, MoTe<sub>2</sub>, and WSe<sub>2</sub> in their monolayer and multilayer forms.<sup>45–48</sup> These extensive experimental and theoretical studies have shown that substitutional doping produces stable materials and enables

precise control over carrier concentrations with negligible lattice distortion, offering excellent material stability, structural compatibility, minimal defect states, and preserved band structures—features essential for engineering device-level functionality.

Fig. 4(a) illustrates the atomic structure of a vdW-stacked p–n junction comprising n-type Re-doped and p-type Nb-doped MoS<sub>2</sub> monolayers, each with a doping concentration of  $10^{14} \text{ cm}^{-2}$ . This doping concentration corresponds to a dopant atomic ratio of less than 2.7%, which is readily attainable through established synthesis techniques.<sup>49,50</sup> The doping concentrations in each layer are kept identical, and lower doping levels can be realized by increasing the size of the simulation supercell while keeping the dopant atom count fixed. Fig. 4(b) shows that an increased doping level simultaneously broadens the absorption spectral bandwidth and enhances infrared absorption, directly correlated with intensified electron exchange, as illustrated in Fig. 4(c).

To clarify the mechanism, Fig. 4(d) and (e) present a comparative analysis of the band diagrams for the doped bilayer MoS<sub>2</sub> p–n junction and the intrinsic bilayer MoS<sub>2</sub>, respectively. While the undoped bilayer MoS<sub>2</sub> exhibits a large indirect bandgap that confines absorption to the visible and ultraviolet ranges, increased doping narrows the interlayer energy gap and intensifies electron exchange. At a doping concentration of  $10^{14} \text{ cm}^{-2}$ , a broken-gap band alignment is established, enabling absorption to extend into the far-infrared



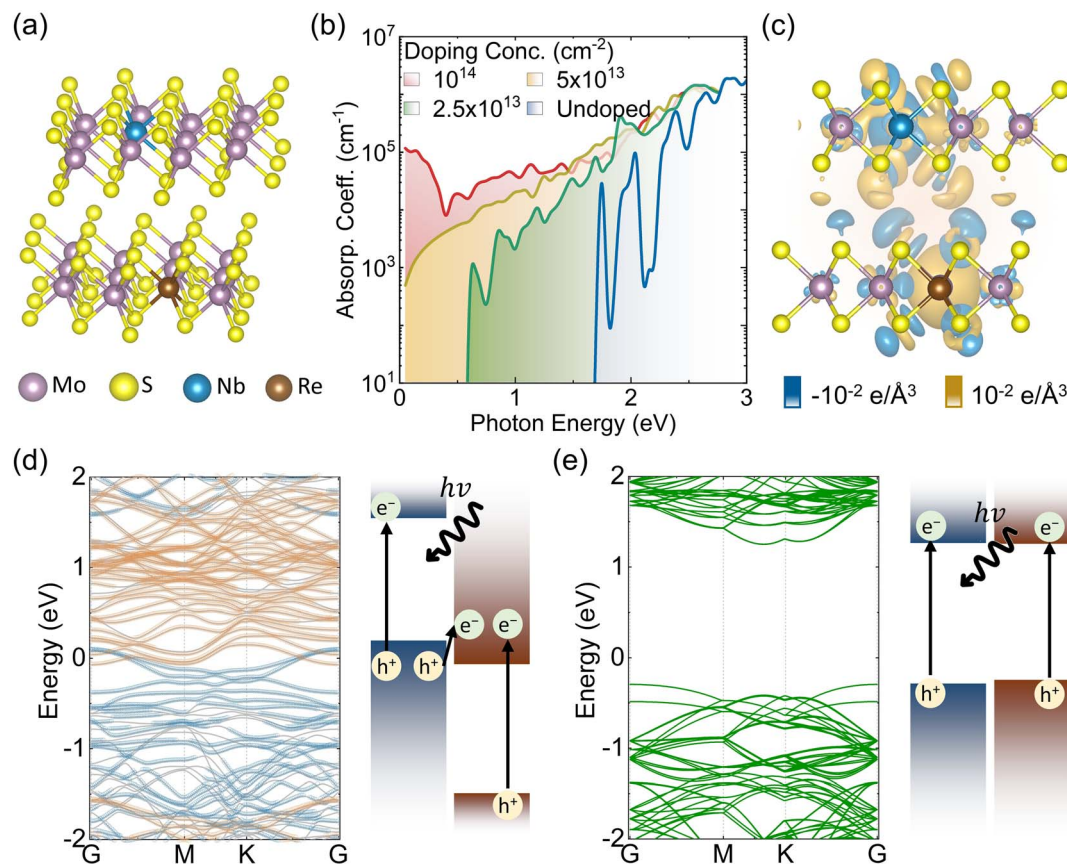


Fig. 4 (a) Atomistic structure of the bilayer MoS<sub>2</sub> vdW p-n homojunction. (b) Absorption spectrum of the MoS<sub>2</sub> p-n junction. (c) Charge redistribution ( $\Delta\rho$ ) for the MoS<sub>2</sub> p-n junction compared with the intrinsic bilayer MoS<sub>2</sub>. Band structure and band diagram for (d) the bilayer MoS<sub>2</sub> p-n junction and (e) the intrinsic bilayer MoS<sub>2</sub>.

region and achieving absorption coefficients comparable to those observed in the visible and ultraviolet ranges. Notably, the enhanced infrared absorption in the bilayer MoS<sub>2</sub> p-n junction arises from strong interlayer charge transfer, driven by the alignment of band edges relative to the Fermi level. This mechanism offers an electrically tunable threshold for interlayer optical transitions. It is important to emphasize that achieving such tunability is inherently challenging in metal-semiconductor Schottky contacts, particularly when 2D materials are involved. In these systems, strong Fermi-level pinning at the metal-semiconductor interface constrains the alignment between the metal Fermi level and the semiconductor band edges, thereby severely limiting the effectiveness of electrostatic modulation of the band structure.

## 4 Conclusions

While 2D vdW heterostructures with staggered band alignment provide optical transition pathways for infrared photodetection, the efficiency has been fundamentally limited by weak interlayer coupling. This work employs first-principles simulations to demonstrate that strategic band engineering through electric field gating and substitutional doping provides effective control over band alignments in 2D vdW structures. These approaches

enable simultaneous achievement of strong interlayer coupling and significantly reduced bandgaps. Systematic simulations reveal substantially enhanced absorption coefficients, in the range of 10<sup>5</sup> to 10<sup>6</sup> cm<sup>-1</sup>, with broad absorption spectral tunability from ultraviolet to mid-infrared regimes, and establish a clear correlation between an enhanced infrared response and improved interlayer charge transfer. This demonstrates that such effects can be effectively modulated through the electrostatic control of band alignment. These findings provide both theoretical insights into the interlayer excitation mechanism and design principles for optimizing high-performance photodetectors in 2D material systems.

## Author contributions

Zichao Ma: conceptualization, data curation, validation, methodology, writing – original draft, writing – review & editing, funding acquisition. Changjian Zhou: conceptualization, investigation, validation, supervision, writing – review & editing, funding acquisition, project administration.

## Conflicts of interest

There are no conflicts to declare.





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