In van der Waals heterostructures of 2D transition-metal dichalcogenides (2D TMDs) electron and hole states are spatially localized in different layers forming long-lived interlayer excitons. Here, the influence of additional electron or hole layers on the electronic properties of a MoS$_2$/WSe$_2$ heterobilayer (HBL), which is a direct bandgap material, is investigated from first principles. Additional layers modify the interlayer hybridization, mostly affecting the quasiparticle energy and real-space extend of hole states at the $\Gamma$ and electron states at the $Q$ valleys. For a sufficient number of additional layers, the band edges move from $K$ to $Q$ or $\Gamma$, respectively. Adding electron layers to the HBL leads to more delocalized $K$ and $Q$ states, while $\Gamma$ states do not extend much beyond the HBL, even when more hole layers are added. These results suggest a simple and yet powerful way to tune band edges and the real-space extent of the electron and hole wave functions in TMDC heterostructures, potentially affecting strongly the lifetime and dynamics of interlayer excitons.

1. Introduction

2D transition-metal dichalcogenides (TMDs) have been intensively studied in the past decade, not only as single layers,[1,2] but, more recently, also as bilayer (2L) van der Waals heterostructures (vdW HS).[3–11] Where two different monolayers (1L) are vertically stacked, forming ubiquitous moiré patterns, due to the lattice mismatch and twist angle between individual layers.[12–14] Such vdW HS offer bandgaps smaller than those of parental single layers and interlayer excitons, which are longer-lived, because of the type-II band alignment, in which electrons and holes reside in different layers.[15–19] These interlayer excitons have been observed in different TMD vdW HS with either crystallographic twist angles (0° or 60°) or with arbitrary, random stackings.[20,21] Moreover, 1L TMDs and vdW HS obey different optical selection rules.[23,24] Thus, one can speak of new physical phenomena in these materials and they are being considered for a wide range of applications, ranging from ultrafast charge separation, (opto)electronics through quantum information processing or valleytronics.[17,19,25–30]

A large variety of 2L vdW HS has recently been investigated on the basis of experiments and theory,[11,13,15,16,18,19,22,30–35] including these constituting of layers from Group 6 TMDCs.[5–7,11,16,36–38] For instance, Kim et al.[39] reported ultra-long valley lifetime in MoS$_2$/WSe$_2$ heterostructures, which was explained by the spatial confinement of electrons and holes in different layers. Park et al.[40] investigated the gate response of such vdW HS and reported ambipolar or unipolar transport behavior depending on the extent of charge depletion in the heterojunction. The key parameter, the interlayer coupling, which predominantly dictates the properties of vdW HS, can be tuned, e.g., via twist angle between the layers or by careful selection of individual layers. For instance, Tongay et al.[41] demonstrated that the interlayer coupling of TMD vdW HS can be tuned by vacuum thermal annealing, where light emission spectrum gradually evolves from single layers, contributing separately to the novel coupled spectrum as function of the interlayer distance.

Up to date, the majority of research focuses on vdW HS made of just two different TMD layers. However, band structure engineering by systematically extending the number of layers of the heterostructure has been much less explored.[39,40] Prominent questions to be addressed are: (i) Can we exploit interlayer coupling by mixing different number of layers to tune the multi-valley, electronic structure near the band edges? (ii) Will additional electron or hole layers delocalize or localize the interlayer excitons in the vdW HS? (iii) For how many layers of the multi-stack, would the bandgap be direct?

Here, in order to address these questions, we investigate the influence of additional electron or hole layers (nL, with n = 1–4) on the electronic properties of a MoS$_2$/WSe$_2$ heterobilayer (HBL). Our results show that adding additional WSe$_2$ layers to the...
HBL (forming 1L-MoS$_2$/(n+1)L-WSe$_2$ HS) will mostly affect the valence (hole) states, while adding MoS$_2$ layers to the HBL (forming (n+1)L-MoS$_2$/1L-WSe$_2$ HS) will affect the conduction (electron) states. We find that the heterostructures of up to 1L-MoS$_2$/4L-WSe$_2$ or 3L-MoS$_2$/1L-WSe$_2$ stay direct gap semiconductors (at the $K$ point), while more layers in both HS would open new transition channels in the $Q$ and/or $\Gamma$ valleys. We, therefore, provide the energy differences between $k$ valleys in the conduction ($Q-K$) and the valence ($K-\Gamma$) bands, values which can be obtained experimentally, e.g., from carrier dynamics measurements.$^{[21]}$ The calculated partial charge densities (PCD) for the electron and hole states, analyzed for each relevant $k$ point of the band edges, are either delocalized throughout all the layers of the same type in the stack or localized in the HBL or just the heterolayer, depending on the stoichiometry of vdW HS. Here, we apply a simple two-step approach of analyzing electron and hole states in large incommensurate HS using single unit cells, where we minimize the effect of the tensile strain or compression, resulting from the differences in the lattice vectors of constituent layers.

2. Results and Discussion

First, electronic structures of bulk and 1L MoS$_2$ and WSe$_2$ were obtained (see Figure S1 in Supporting Information). Good agreement with previous works supports the choice of our simulation methods (HSE06 with SOC, see Computational Details in Section 4).$^{[41,42]}$ While 1L-MoS$_2$ is a direct bandgap semiconductor (at $K$, here 2.21 eV), 1L WSe$_2$ is an indirect gap material (here 1.70 eV),$^{[43]}$ with the corresponding direct bandgap at $K$ being only 50 meV higher in energy. Both 1L systems exhibit spin splitting in the valence band at the $K$ point of 197 and 606 meV for MoS$_2$ and WSe$_2$, respectively, due to strong SOC. It is well known that HSE06 overestimates the spin splitting for TMDs, however, GW calculations performed on top of HSE06 would lower these values to match experiments better.$^{[44]}$ The experimental values are 1.92 eV (1.75 eV optical) for bandgap and 160 meV (430 meV) for spin splitting in MoS$_2$ (WSe$_2$).$^{[45,46]}$

Next, we have calculated band structures of vdW HS, HBL/nL-MoS$_2$ and HBL/nL-WSe$_2$ (with $n$ = 1–4; see structural models in Figure 1), using the fully relaxed structures (method I) and our proposed 2-step approach (method II) for comparison (see Computational Details in Section 4 for detailed explanations on employed methods). In method II, we perform two different calculations to analyze the electron and hole states separately. By using different lattice constants for the electron and hole calculations, our approach removes the problematic tensile strain or compression, which arises from the incommensurability of the layers. This approach is valid for our systems, because the HS exhibit type-II band alignment. Because with method I both $H_h^h$ and $R_h^M$ stackings result in very similar electronic structures (see Figures S2–S5 in the Supporting Information) and energy differences, we have only used the $H_h^h$ systems for method II. Here, we first shortly discuss the results from approach I and then more detailed results are shown for the approach II.

While the stacking arrangement ($H_h^h$ or $R_h^M$) results in very similar band structures, more significant differences are observed for the number of layers ($nL$) in the multistack: 1) the bandgap increases (decreases) for HBL/nL-MoS$_2$ (HBL/nL-WSe$_2$); 2) when $n$ is varied for HBL/nL-MoS$_2$ (HBL/nL-WSe$_2$), the conduction (valence) bands are mostly affected;

![Figure 1](image-url). The van der Waals heterostructures studied in this work. a) A MoS$_2$/WSe$_2$ heterobilayer (HBL) with additional electron or hole layers ($nL$ = 1–4) of either MoS$_2$ or WSe$_2$, respectively. The stacking polytype of the HBL is $H_h^h$ or $R_h^M$ and the one of the $nL$ and its interface with HBL is $H_{nL}^h$. b,c) Top and side views of the considered stacking polytypes ($H_h^h$, also known as 2H, and $R_h^M$, also known as 3R) in HBLs with 0° and 60° twist angles.
3) for HBL/nL-MoS2 (HBL/nL-WSe2), the valley at the Q point in the CB (at the Γ point in the VB) decreases (increases) in energy with number of layers nL. These trends are valid for both methods I and II. The variations are noticeable when discussing the energy differences and the direct versus indirect bandgap characters. To understand these differences, we used method I and calculated atom-resolved band structures (so-called fatbands) for exemplary systems (HBL/2L-MoS2 or HBL/2L-WSe2), which are shown in Figure 2. Both systems are type-II band alignment HS, meaning that the valence band maximum (VBM) is composed mostly of the WSe2 (hole) states and conduction band minimum (CBM) is dominated by the MoS2 (electron) states. Both HS have direct bandgaps at the K point.

The exemplary band structures from method II are shown in Figures 3 and 4 for HBL/2L-MoS2 (or 3L-MoS2/1L-WSe2) and HBL/2L-WSe2 (or 1L-MoS2/3L-WSe2), respectively. Depending on which part of the heterostructure is fully relaxed, 1L or nL, the respective VB or CB are taken into consideration (see Section 4 for details). We have again analyzed the orbital resolved band structures, which consistently show that all HS are of type-II.

The most significant difference between approach I and II is how fast the Q valley in the CB (Γ valley in the VB) decreases (increases) in energy. In other words, the energy differences between the Q and K valleys in the CB and the Γ and K valleys in the VB, indicating direct or indirect bandgaps, differ for both approaches (see Figure 5). Approach I would suggest that both types of HS with at least 6 layers in the multi-stack are still direct-gap materials. However, approach II shows that these changes happen much faster and already HBL/3L-MoS2 and HBL/4L-WSe2 transform to indirect-gap materials.

Additionally, we have calculated the PCD of electron and hole states (Figures 3 and 4; Figures S6 and S7, Supporting Information). All the PCD results are shown together with the percentage of majority contributions (of at least 10%). We selected the band edge states at four relevant k-points, i.e., the K- and Q-valley for the electron states and the K- and Γ-valley for the hole states. The type-II character is also shown by the PCD plots at the K point, where the states are localized in the respective building blocks of the HS, meaning in the 1HL and the (n+1)L stack. Similar results were reported by Kunstmann et al.,[47] for MoS2/WSe2 HBL. Both the Γ and the Q valley states are mixtures of d-orbitals of metal and p-orbitals of the chalcogen atoms, while states at K are dominated by d-orbitals of the metal atoms. There is a vanishing overlap of the states in the K valley, which are separated by at least 6.5 Å (in the bilayer case) in a purely interlayer exciton.

The additional electron or hole layers have different influence on the distribution of the electron and hole states in the HBL or in the whole HS for that matter. As already shown experimentally and theoretically,[19,47] the states in the HBL are such that at band edges (K point), the electrons are localized in the MoS2 layer, while hole in WSe2 layer. Additionally, hole states are distributed nearly equally between both layers at the Γ point. In our study, we find that the same holds for the electron states at the Q point, a fact not discussed previously. Additional electron layers result in the delocalization of electron states throughout all the MoS2 layers in the stack, while hole states do not extend too much beyond these of HBL (maximum to one additional layer at the Γ point), see Figure 3 and Figure S6 (Supporting Information) for exemplary PCD results. This is due to different contribution of orbitals at these k-points.[48] At the QCBM point, electron wave function is distributed over all MoS2 layers, with a maximum in the central layer. Here, both d-orbitals of Mo and p-orbitals of S hybridize. The WSe2 layer breaks, however, the symmetry of the MoS2 stack and leads to an uneven distribution of the wave function weight in the MoS2 layers (32%-44%-20% from top to bottom; see Figure 3c). At the KCBM point, only the Mo dz-orbitals partake, thus, are not affected by HL and, consequently, are equally distributed in all MoS2 layers. The ΓVBM consists mostly of WSe2. Yet, due to the pz contribution from Se atoms and the fact that MoS2 states at Γ are close in energy to these of WSe2 at ΓVBM (see Figure 3b,d), there is a hybridization, which is, of course, largest for the first MoS2 layer (about 45%), as the bands of the second MoS2 layer are further away in energy. Different behavior is found when adding hole layers to the HBL. In this case, all the states are still localized to the HBL and one additional layer, see Figure 4 and Figure S7 (Supporting Information).

Our results suggest that adding more electron layers result in more delocalized electron states, while adding more hole layers, still gives partially localized hole states. The level of (de)localization of states will affect the lifetime and intensity of the resulting interlayer excitons. While the exciton binding energy is expected to reduce with number of layers, at the same time delocalization in more layers and the k-space indirect character of excitons should increase their lifetime. MoS2/WSe2 HBL and multilayers of random twist angles were shown experimentally to have increased lifetime of photogenerated carriers.[19,34] On the other hand, there were also reports on inefficient charge transfer between the constituting layers and tunneling-assisted recombination.[49] These may be dependent on the twist angle between building blocks.
Furthermore, we can also conclude, from Figure 5, that changing the number of layers in nL results mostly in the energy changes of the $Q$ and the $\Gamma$ points: the former is reducing and the latter increasing in energy when increasing nL. This suggests that, even though the materials are direct-gap semiconductors with transitions at the $K$ point from these DFT simulations, a prominent indirect-transitions between $\Gamma$−$Q$ and $K$−$Q$ points should also be present, meaning the materials would have layer-indirect and momentum-indirect excitons, what would result in longer lifetimes, as shown in our recent work on WS$_2$/WSe$_2$ heterobilayers. This type of HS is also promising for phototransistors or photodetectors, as recently shown for heterojunction made of multilayer of both MoS$_2$ and WSe$_2$.

In present work, we avoid discussing the absolute bandgap values. Our 2-step approach can also be used to analyze bandgap values (transitions between valence and conduction bands) if the band edges were calculated with respect to the vacuum energy. From approach I, we have noticed that in HS with nL-MoS$_2$, the bandgaps increase, while in HS based on nL-WSe$_2$, the bandgaps decreased with n (Figures S2–S5, Supporting Information). This subject, however, requires more detailed and very careful investigations and is beyond the scope of this work.

3. Conclusion

We have investigated the influence of additional electron or hole layers (nL, with n = 1–4) on the electronic properties of a MoS$_2$/WSe$_2$ heterobilayer (HBL). We showed that this approach can tune the valleys and the wave functions of the multi-stack in different ways, depending on the stoichiometry of the layers. We have studied two types of systems, namely HBL/nL-MoS$_2$ and HBL/nL-WSe$_2$. Furthermore, we compared a very common approach, where the heterostructure is modelled with small, but strongly strained, unit cells with our simple 2-step
approach, where two calculations are performed to analyze the electron and hole states separately. By using different lattice constants for the electron and hole calculations, our approach removes the problematic tensile strain or compression, which arises from the incommensurability of the layers. It can be used, because the HS are type-II materials, where the electrons reside predominantly in the MoS$_2$ layers and holes predominantly in WSe$_2$ layers.

We have analyzed the energy differences between the band edges at the $K$ valley and other band edge valleys, i.e., the $Q$ valley in the conduction and the $\Gamma$ valley in the valence band. These energy differences can be obtained experimentally, e.g., from carrier dynamics measurements.$^{[21]}$ We show that increasing the number of additional layers, the $Q$-$K$ and $\Gamma$-$K$ energy differences decrease and increase in energy, respectively. Our calculations suggest that the materials stay direct-gap semiconductors for up to HBL/2L-MoS$_2$ and HBL/3L-WSe$_2$.

From partial-charge density calculations of the electron and hole states, we could show that both types of HS differ significantly in the state localization: when increasing the number of MoS$_2$ layers, the CB states at $Q$ and the hole state at $\Gamma$ are delocalized more or less equally throughout all electron layers. The VBM states at $K$ are completely localized in the 1L-WSe$_2$, while at $\Gamma$, it is practically localized in the HBL. This means that $Q$ point offers stronger delocalization when increasing the number of layers, while no significant change is observed for the $\Gamma$ states.

In case of changing the nL of WSe$_2$, all the states are mostly localized to the HBL, except at $\Gamma$, where they cover one additional hole layer. This results in HS with different localization of states, thus, resulting in interlayer excitons of shorter or longer lifetimes. These differences may also affect the exciton dynamics.

Our study offers a simple experimental approach to understand and control excitons in TMDC heterostructures.
4. Experimental Section

Computational Details: All simulations were performed using density functional theory (DFT) methods as implemented in the Vienna *ab-initio* simulation package (VASP). The projector-augmented wave (PAW) method was used to describe the interactions between electrons and nuclei. The exchange and correlation potentials were treated using the generalized-gradient approximation (GGA) functional proposed by Perdew, Burke, and Ernzerhof (PBE) for full geometry optimization (lattice vectors and atomic positions). Plane-wave cutoff of 500 eV was used in all simulations. The D3 dispersion correction was included, as proposed by Grimme. Additionally, for exemplary systems with 3L+1HL (either MoS$_2$ or WSe$_2$), we have also tested the optB88 vdW functional and found out that the interlayer distances compared with the D3 cases are consistently larger, but by no more than 1.7%. A vacuum layer of at least 15 Å was added along the out-of-plane direction to avoid spurious interactions with the next periodic images. All structures were relaxed until all the forces acting on each atom were less than 2 × 10$^{-2}$ eV Å$^{-1}$ and the total energy change between two self-consistent steps was less than 1 × 10$^{-4}$ eV. A $\Gamma$-centered 12 × 12 × 1 k-point mesh was used to sample the Brillouin zone during structural optimizations. It is well known that standard GGA DFT functionals underestimated the electronic bandgaps, thus, electronic properties were calculated using the Heyd-Scuseria-Ernzerhof hybrid (HSE06) functional, considering relativistic effects, such as spin-orbit coupling (SOC). Even though, we do not discuss the bandgap sizes in the present work, we have also noticed strong deviations in the shape of bands in band structures between PBE and HSE06, thus, we used the latter for...
the electronic structure analysis. The PCD was, however, calculated using PBE without SO and analyzed for exemplary structures, similar to Kunstmann et al.[47] Structural illustrations and PCD plots were created using the 3D visualization package VESTA.[36]

The systems studied in this work are shown in Figure 1. We consider a MoSe₂/WSe₂ HBL and systematically add one to four additional layers (nL) of either WSe₂ or MoS₂, creating either a 1L-MoS₂/(n+1)L-WSe₂ or a (n+1)L-MoS₂/1L-WSe₂ vdW HS. The (n+1)L multilayers were kept in their most stable stacking arrangement, Hₙ⁺ (also known as 2H). The interface between MoS₂ and WSe₂ is realized either in the H₂ₙ or R₅₃̈ (also known as 3R) stacking fashion, which are among the most stable high-symmetry stacking arrangement observed in 0° and 60° twist angle moiré structures.[24] Other stacking polytypes, e.g., Hₘ or Rₘ, are, of course, also possible,[25,26] but beyond the scope of the present work, because the electronic properties are affected much stronger by the number of layers than the stacking itself.

A lattice mismatch between the building blocks of HS results in an incommensurate moiré superstructure that, in the best case, would need to be approximated by supercells containing hundreds or thousands of atoms (see Figure S8 in the Supporting Information). Such large systems are, however, too demanding for DFT calculations, especially when including SO with hybrid functionals. The most commonly used approach is to relax such a HS in a small unit cell (referred to as method I in the text below), however, this results in strain or compression of the constituent layers, which is an unrealistic representation of the HS. As previously shown in different theoretical and experimental works, tensile strain and compression strongly affect electronic structures of TMDC layers.[37,38] Thus here, we propose a simple 2-step approach (referred to as method II) to separately analyze the electron and hole states of an incommensurate HS by performing two calculations within primitive unit cells. We suggest that, in order to analyze the electron states near the Fermi level, we use a lattice constant (a) of the relaxed (n+1)L-MoS₂, while for the hole states, we use a of the relaxed (n+1)L-WSe₂. This is possible, because of the type-II band alignment, above. We used the same approach in our recent work on the W-based HBLs.[31] In Table S1 in the Supporting Information, we summarized the lattice parameters for (n+1)L-MoS₂ and (n+1)L-WSe₂, and we compare them to the lattice parameters of fully relaxed HS.

**Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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**Conflict of Interest**

The authors declare no conflict of interest.

**Data Availability Statement**

The data that support the findings of this study are available on request from the corresponding author. The data are not publicly available due to privacy or ethical restrictions.

**Keywords**

density functional theory, electron and hole states, electronic structure, multilayer van der Waals heterostructures, transition-metal dichalcogenides

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