# Introduction

Quantum spin Hall (QSH) insulator [1, 2] featuring helical edge states immune from backscattering by nonmagnetic impurities, is of great interest for the next-generation low-dissipation quantum electronic and spintronic devices. Material systems that can be electrically tuned between the normal insulating and the QSH insulating phases can be of particular interest, providing a mechanism of field effect transistors where the gate, instead of tuning the carrier density in a fixed conducting channel, simply creates/eliminates the topologically protected conducting channel in a bulk gap [3–5]. Concerning such desired electrical switchability, van der Waals heterostructures of two-dimensional (2D) materials can provide a unique approach towards QSH insulators, where the band alignment between the constituent layers can be most effectively tuned in the double gate geometry [6].

Theoretical study has predicted that heterobilayers of transition metal dichalcogenides (TMDs) can be an electrically switchable QSH insulator [5]. The TMD monolayers feature spin-valley locked massive Dirac cones located at the corners of hexagonal Brillouin zone [7–9]. The interlayer hybridization of the spin-valley locked Dirac cones leads to either a topological band inversion or a trivial avoided band crossing, depending on the interlayer atomic registries. This points to an intriguing platform for exploring QSH effect, with versatile tunability by interlayer bias and stacking registries. The sensitive dependence of the topological properties on the stacking configuration, arising from the valley nature of the band edges, further suggests unprecedented possibility for quantum engineering of topological superstructures in the moiré pattern of spatially varying local atomic configurations [5, 10]. Theoretical study has further shown that the strong Coulomb interaction in the 2D...
geometry can turn the QSH insulator into a quantum anomalous Hall (QAH) insulator [11], which features chiral edge states that are completely lossless [12–14].

In TMDs, the spin–orbit coupling is so strong that it gives rise to a spin-valley coupling of hundreds of meV. The size of the topological gap from the interlayer hybridization is essentially determined by the strength of interlayer hopping between the two monolayer TMDs [15]. At the spin–valley locked band edges, the electronic states are predominantly from $d$-orbitals of the transition metal atoms [16–22]. The large separation of the active orbitals in the TMDs heterobilayers limits the interlayer hopping magnitude and hence the size of the topological gap. Moreover, the large band gap in the experimentally explored TMDs heterobilayers requires a large electric field to bring their type II alignment into the inverted regime. These are the two key issues lying between the intriguing concept of gate switchable topological superstructures in moiré and the experimental observation as well as practical applications.

To address these outstanding issues, we consider here a heterobilayer formed by a monolayer hexagonal boron arsenide (BAs) and a TMD, with WSe$_2$ as an example, that features: (1) a much larger interlayer hopping because of the smaller interlayer separation between the band edge orbitals; (2) a small type II band gap that can be inverted by moderate gate bias [23]. Symmetry analysis shows that the conduction band edges of BAs monolayer are nearly spin-degenerate massive Dirac cones at the Brillouin zone corners. Their hybridization with the spin-valley locked valence band edges in TMD leads to topological band inversion depending on the stacking registry. In the non-interacting limit, the double spin degeneracy of BAs leaves an unhybridized conduction band inside the hybridization gap, so the heterobilayer is a spin–valley locked metal. With Coulomb interaction accounted in the double-layer geometry, the interaction with the hybridization induced electric dipole shifts this unhybridized conduction band upwards in energy, giving rise to a sizable global QSH gap, as illustrated in figure 1. Using mean-field theory, we study the phase diagram in lattice-matched heterobilayers of TMD/BAs as function of dielectric constant, interlayer bias, and interlayer translation, which determine the relative importance of Coulomb interaction, the $s$-wave interlayer hopping, and the $p$-wave interlayer hopping in the band inversion. The competition of these factors in a long-period moiré pattern can lead to rich possibilities of electrically tunable superstructures consisting of large gap QSH insulators, excitonic insulators [24] and exciton superfluids.

The paper is organized as follows. In section 2, we first study the non-interacting properties of the TMD/BAs heterobilayer, taking WSe$_2$/BAs heterobilayer as an example, based on the $\mathbf{k} \cdot \mathbf{p}$ model from symmetry analysis. Relevant parameters are obtained by fitting the band structure from first-principles calculations. In section 3, we study the Coulomb effect on the band inversion. Mean-field theory is used to self-consistently take into account the effect of Coulomb interaction in the double-layer geometry. Interesting phases like exciton superfluid and QSH/QAH insulators are found. The corresponding results are presented in section 4, including the phase diagrams for AA-type and AB-type heterobilayers as functions of dielectric constant, interlayer bias, and interlayer translation, where the dependence on the last leads to gate tunable topological superstructures in long-period moiré. Section 5 discusses the relevant issues unexplored in detail in section 4.

### 2. Heterobilayer without Coulomb interaction

Although yet to be synthesized, 2D hexagonal BAs monolayer is predicted to be stable in energetic, dynamic, thermal, and mechanical properties, by extensive first-principles calculations [25–27]. Besides, the stable structure of hexagonal BAs consists of B and As atoms located on the same plane [25–27]. Here, we consider the heterobilayer formed by monolayer hexagonal BAs and WSe$_2$, with lattice constants $a$ of 3.39 Å and 3.325 Å, respectively. The small lattice constants mismatch of $\sim$2% leads to a large moiré pattern with period $\sim$50a, which can be further tuned by a relative strain and/or twisting. In such a large moiré, one can regard each local region as commensurate bilayer with identical lattice constants, while different local regions differ by a relative in-plane displacement vector $\mathbf{R}$, which defines a moiré supercell. The heterobilayer is then described by commensurate bilayers parameterized by a vector $\mathbf{R}$. In BAs, the valence band maximum and conduction band minimum are mainly attributed to the $4p_z$ orbital of As and the $2p_z$ orbital of B, respectively. Therefore, we define AA-type of stacking of WSe$_2$/BAs heterobilayer as B on W and As on Se atoms, as well as other configurations related by interlayer translation; and AB-type of stacking as As on W and B on Se atoms, as well as other configurations related by interlayer translation. The in-plane displacement vector $\mathbf{R}$ is then defined from W to B in AA stacking and from W to As in AB stacking. To model the heterobilayer, in the following we first consider the Hamiltonian of individual monolayer and then add a $\mathbf{R}$ dependent interlayer coupling.

The low energy physics of monolayer TMD is described by the massive Dirac model. TMDs have a strong spin-orbital coupling originating from the $d$-orbitals of the metal atoms. The valence band edge splitting is about 0.15 eV for MoX$_2$ ($X = S, Se$) monolayer and about 0.45 eV for WX$_2$ ($X = S, Se$) monolayer. The conduction band edge splitting is relatively smaller, about a few meV for MoS$_2$ and tens of meV for WSe$_2$. 


WS₂ and MoSe₂. Around the ±K points of Brillouin zone, the Hamiltonian of monolayer TMD is [7]

\[ H = \frac{M_1}{2} \sigma_z + v_1 (\tau k_x \sigma_x + k_y \sigma_y) \]

\[ + \tau \sigma_x (\sigma_0 + \sigma_2) + 2 \lambda (\sigma_0 - \sigma_2) \], \hspace{1cm} (1)

where \( \tau = \pm 1 \) is the valley index and \( \lambda \) is the spin-orbital coupling. The interlayer hopping is tuned from positive to negative by the interlayer bias, \( t_{ij} \). By definition, \( t_{1,1} = t_{r,s} = 1 \), \( t_{r,s} = -1 \).

When \( E_g \) is tuned from positive to negative by the interlayer bias, the conduction band from BSAs and valence band from WS₂ become inverted and cross at finite \( k \). The interlayer coupling \( t_{ij} \) opens a band gap around the crossing points. The topology of the band gap thus opened is determined by the nature of interlayer hopping. As the \( t_{1,1} \) term is \( k \) independent, the band gap it opens is topologically trivial. On the other hand, the \( t_{r,s} \) and \( t_{v,c} \) terms are linear in \( \eta \) with different chirality, and thus the band gaps opened by these two terms are topologically nontrivial. The topological property of the \( t_{r,s} \) term depends on the stacking. While the band gap is topologically trivial for AB stacking, it is topologically nontrivial with chirality of 2 for AA stacking. However, the band gap is relatively small as it is a high-order process, and may be readily corrected in the presence of other energy bands.

The interlayer hopping matrix element \( t_{ij} \) depends on the interlayer atomic registry, which is characterized by \( R \). By definition, \( t_{r,s} = \langle \Psi_{r,s}^{(3)}(R) | H | \Psi_{r,s}^{(3)}(R) \rangle \), where \( | \Psi_{r,s}^{(3)}(R) \rangle \) denotes the Bloch state at \( \tau K \) point in the WS₂ (BSAs) layer. For these three translation vec-
tors $\mathbf{R} = 0, \frac{2\pi}{3}, \frac{4\pi}{3}$, with $\hat{a}_i, \hat{d}_i$ being the primitive vectors spanning the moiré supercell, the atomic configurations have three-fold rotational symmetry. As $t_{r,ij} = \langle \hat{C}_3 \hat{\Psi}_{ij}^+ | H | \hat{C}_3 \hat{\Psi}_{ij} \rangle = e^{i(\phi_{ij} - \phi_{ji})} \gamma_{r,m}^{i} \gamma_{r,m}^{j}$, with $\gamma_{r,m}^{i} = e^{-\frac{i\pi}{6}}$ and $\phi_{ij} = 0, \frac{2\pi}{3}, \frac{4\pi}{3}$ for rotation centers at W (B) atom, Se (As) atom, hollow center respectively [3], a non-zero interlayer hopping requires $e^{i(\phi_{ij} - \phi_{ji})} \gamma_{r,m}^{i} \gamma_{r,m}^{j} = 1$.

The symmetry allowed interlayer hopping channels between conduction and valence bands of the two layers at $K$ point are listed in the following table. The magnitudes of these interlayer hoppings can be evaluated from the first-principles calculations of band structures. Since it is difficult to evaluate the interlayer coupling constants of a heterobilayer directly, we estimate them from the results in the homobilayers of BAs and WSe$_2$ using the formula $t_{ij} = \sqrt{t_{BAs}^{ij}t_{WSe}_2^{ij}}$. The calculated coupling constants are several times larger than those in TMD bilayers, giving rise to much larger band gaps under the same band inversion.

At a general translation $\mathbf{R}$, the three-fold rotational symmetry is broken. Under the two-center approximation, the $\mathbf{R}$ dependence of $t_{ij}$ can be formulated as

$$
t_{\vec{e}}(\mathbf{R}) \approx \frac{t_{\vec{e} \vec{e}}}{3} \left( e^{i\mathbf{K}\mathbf{R} + e^{i\mathbf{C}_3\mathbf{K}\mathbf{R}} + e^{i\mathbf{C}_3^2\mathbf{K}\mathbf{R}}} \right),
$$

for AA-type of WSe$_2$/BAs heterobilayer, and

$$
t_{\vec{e}}(\mathbf{R}) \approx \frac{t_{\vec{e} \vec{e}}}{3} \left( e^{i\mathbf{K}\mathbf{R} + e^{i\mathbf{C}_3\mathbf{K}\mathbf{R}}} + e^{i\mathbf{C}_3^2\mathbf{K}\mathbf{R}} \right),
$$

for AB-type of WSe$_2$/BAs heterobilayer, where $\mathbf{K}, \mathbf{C}_3 \mathbf{K}$ and $\mathbf{C}_3^2 \mathbf{K}$ denote the wavevectors of the three corners of the first Brillouin zone.

The conduction band from BAs is nearly spin degenerate, while the valence band from WSe$_2$ is spin up (down) polarized at $K$ ($-K$) valley and separates energetically far away from band of the other spin species because of the large spin-orbital splitting in the valence band. Taking into account the spin degree of freedom, the system is described by a three-band model,

$$
\mathbf{H}_3(\mathbf{K}, \mathbf{R}) = 
\begin{bmatrix}
\varepsilon_{kz} & 0 & 0 \\
0 & \varepsilon_{kx} & h_{k}(\mathbf{R}) \\
0 & h_{k}^*(\mathbf{R}) & \varepsilon_{ks}
\end{bmatrix},
$$

with the basis $\{ |B, s\rangle, |B, s\rangle, |W, s\rangle \}$. Here $s = \uparrow (\downarrow)$ for $\tau = 1 (-1)$, and $\delta$ mean the reverse of $s$. Dispersions of conduction and valence bands are $\varepsilon_{kz} = \varepsilon_{kx} = -\frac{\tau}{M_1} k^2 + \frac{\tau}{M_2} k_y - \frac{\tau}{M_2} k_z$, respectively.

The effective $\mathbf{R}$-dependent hopping matrix element is $h_{k}(\mathbf{R}) = \frac{t_{\vec{e}}}{\tau M_2} (\mathbf{R}) + \frac{t_{\vec{e}}^*}{\tau M_2} (\mathbf{R}) k_x - \frac{t_{\vec{e}}}{\tau M_2} (\mathbf{R}) k_y - \frac{t_{\vec{e}}^*}{\tau M_2} (\mathbf{R}) k_z - \frac{t_{\vec{e}}}{\tau M_2} (\mathbf{R}) k_z - \frac{t_{\vec{e}}^*}{\tau M_2} (\mathbf{R}) k_y$.

In the non-interacting limit, the interlayer hopping induces hybridization of bands with same spin species, while the other conduction band is left unaffected and lies inside the hybridization gap. The resulted phase is a metal with Fermi surfaces of opposite spin polarization in different valleys (figure 1(c)). We will show in the following section that further taking into account the effect of Coulomb interaction can shift the unhybridized conducton band upwards in energy and thus a global band gap is possible. In such a way, this heterobilayer can realize a QSH insulator.

3. The effect of Coulomb interaction

The effect of Coulomb interaction can be taken into account by the Hartree–Fock mean-field theory, which gives reasonable results in treating other bilayer electron-hole systems [28–33]. In mean-field theory, the ground state wave function of the three-band system is $|\Psi\rangle = \prod_{\tau} (u_{\tau k} + v_{\tau k} \hat{a}_{\tau k}^\dagger + v_{\tau k} \hat{a}_{\tau k}^\dagger \hat{b}_{\tau k}^\dagger)|0\rangle$, where $s = \uparrow (\downarrow)$ for $\tau = 1 (-1)$, $\delta$ means the reverse of $s$, and $\hat{a}_{\tau k}$ ($\hat{b}_{\tau k}$) is the annihilation operator of electron with spin $s$ and momentum $k$ in spin degenerate conduction band (spin-valley locked valence band). $|0\rangle$ denotes the insulating state with the valence bands of TMD completely filled. In the limit of vanishing interlayer tunneling, it can be shown that the ground state of the three-band system satisfies the condition $v_{\tau k_{\downarrow}} / v_{\tau k_{\uparrow}} = \beta$, with $\beta$ being a constant independent of $k$ (see appendix). Besides, the ground state energy is degenerate with respect to the value of $\beta$, a manifestation of SU(2) symmetry of conduction band electrons. Consequently, in terms of conduction band basis with spin orientation $|S\rangle$, the three-band wave function becomes an effective two-band one, $|\Psi\rangle = \prod_{\tau} (u_{\tau k} + v_{\tau k} S_{\tau} a_{\tau k}^\dagger \hat{b}_{\tau k}^\dagger)|0\rangle$. Here $\langle S\rangle$ is a function of $\beta$ but independent of $k$, defined as $\langle S\rangle_{\tau} = \frac{1}{2} (v_{\tau k_{\dagger}}^* v_{\tau k_{\dagger}}) \varepsilon_{\tau} (v_{\tau k_{\dagger}} v_{\tau k_{\dagger}})^2 / (v_{\tau k_{\dagger}}^2 + |v_{\tau k_{\dagger}}|^2) \varepsilon_{\tau}$, being the Pauli matrices and $i = x, y, z$. The other conduction band with opposite spin orientation is decoupled from the interaction induced hybridized bands, only shifted upwards in energy due to the interaction with the hybridization induced electric dipole. Without loss of generality, one can choose $v_{\tau k_{\uparrow}} = 0$ in $K (-K)$ valley, in which case $\langle S\rangle_{\tau} \sim \tau z$.

The interlayer tunneling explicitly breaks the SU(2) symmetry of conduction band electrons due to coupling with the spin-valley locked valence band. From an energetic point of view, hybridization induced by both Coulomb interaction and interlayer tunneling should occur between conduction and valence bands with the same spin species to open the largest possible
gap, so the Coulomb interaction coupled conduction band should have the same spin polarization with the spin-valley locked valence band. Therefore, only $v_{xk}(\pm)$ is nonzero in $K - (K)$ valley, and the other conduction band with spin down (up) is still decoupled. This argument is also confirmed by our numerical solution of a fully three-band self-consistent gap equation with both Coulomb interaction and interlayer tunneling.

With the mean-field wave function $|\Psi\rangle = \prod_{k}(u_{ck} + v_{ck}d_{k\uparrow}b_{k\downarrow})|0\rangle$, the band coupling brought by both interlayer tunneling and Coulomb interaction can be described by the mean-field Hamiltonian [11] in the original three-band basis,

$$\hat{H}_\tau = \sum_k \left( \hat{a}_{k\uparrow}^\dagger \hat{a}_{k\downarrow}^\dagger - \hat{a}_{k\downarrow} \hat{a}_{k\uparrow} \right) h_{\tau k} \left( \hat{d}_{k\uparrow} \hat{b}_{k\downarrow}^\dagger \right) T, \text{ and}$$

$$h_{\tau k} = \begin{bmatrix} \xi_{\tau k} & 0 & 0 & \xi_{\tau k}^* \n -\Delta_{\tau k}^* & \tau_{\tau k} \n 0 & -\Delta_{\tau k} & \tau_{\tau k} \n \end{bmatrix}. \quad (9)$$

The pairing gap function $\Delta_{\tau k} = \sum_{k'} \sqrt{V(k-k')} \ n_{\tau k'}^* n_{\tau k'}$, and $\xi_{\tau k} = \xi_{\tau k}(\pm) = \xi_{\tau k}(\pm) = \sum_{k'} U(k-k') |v_{\tau k'}|^2 + \xi_{\tau k}(\pm) = \frac{2\pi e^2}{\epsilon} \sum_{k'} |v_{\tau k'}|^2$. Here $U(k) = 2\pi e^2/(\epsilon k)$ and $V(k) = U(k) e^{-|k|d}$ are the intra- and inter-layer Coulomb interactions, respectively. $\epsilon = \sqrt{\epsilon_{\parallel}/\epsilon_{\perp}}$, where $\epsilon_{\parallel}/\epsilon_{\perp}$ is the intralayer (interlayer) dielectric constant. $d = \sqrt{\epsilon_{\parallel}/\epsilon_{\perp}}$ with $D$ being the geometric interlayer distance. The last term in $\xi_{\tau k}(\pm)$ is the classical charging energy of the bilayer as a parallel-plate capacitor. The ground state wave function $|\Psi\rangle$ shall be solved from the self-consistent gap equation,

$$\Delta_{\tau k} = \sum_{k'} \sqrt{V(k-k')} \ n_{\tau k'}^* n_{\tau k'} = \frac{\Delta_{\tau k'} - \tau_{\tau k'}}{2 \sqrt{\xi_{\tau k}^2 + |\Delta_{\tau k'} - \tau_{\tau k'}|^2}}, \quad (10)$$

where $\xi_{\tau k} = \frac{2\pi e^2}{\epsilon} \sum_{k'} U(k-k') |v_{\tau k'}|^2 + 2\pi e^2 \sum_{k'} |v_{\tau k'}|^2$. The quasiparticle band dispersions are subsequently obtained as $E_{\tau k\uparrow} = \frac{2\pi e^2}{\epsilon} \sum_{k'} U(k-k') |v_{\tau k'}|^2$ and $E_{\tau k\downarrow} = \frac{2\pi e^2}{\epsilon} \sum_{k'} |v_{\tau k'}|^2$. The global gap of the system is given by $\delta = \min\{\min(E_{\tau k\uparrow} - E_{\tau k\downarrow})\}$. Both the single particle dispersion and band hybridization gap are renormalized by Coulomb interaction.

In the process of numerically solving the gap equation, there are parameter regimes where multiple metastable solutions exist, in which case the ground state is the one with lowest energy. Coulomb interaction favors a s-wave interlayer (electron–hole) pairing, and without interlayer tunneling ($\tau_{\tau k} = 0$), the pairing gap function $\Delta_{\tau k}$ is a real function only dependent on $|k|$. The interlayer tunneling can have s-wave/p-wave component or their mixture, depending on the stacking registries. The pairing gap function $\Delta_{\tau k}$ plays the role of an effective interlayer tunneling, and its competition with the bare interlayer tunneling leads to rich possibilities of topological and correlated phases. If interlayer tunneling is p-wave dominant, the ground state can be topological phases like QSH or QAH insulators, or exciton condensates with superfluid properties. If interlayer tunneling is s-wave dominant, the ground state will be a topologically trivial excitonic insulator (EI) without superfluidity. The detailed phase diagrams are presented in the next section.

### 4. Phase diagram in different stackings

#### 4.1. AA stacking

**4.1.1. High symmetry points**

We first study the commensurate case, neglecting the small lattice mismatch in the heterobilayer. At the two high symmetry points $AA_0$ and $AA_1$ of AA stacking as defined in table 1, interlayer tunneling can lead to topological phases upon band inversion. At $AA_0$, only $t_{xy}$ is nonzero among the four interlayer hopping matrix elements. So $t_{xy} = -\frac{\pi D}{2M} t_{vv} (k_x - i k_y)$, with $v_1 (M_1)$ being the Fermi velocity (band gap) in monolayer WSe$_2$. Similarly, at $AA_1$, only $t_{yy}$ is nonzero. Then $t_{yy} = \frac{\pi D}{2M} t_{vv} (k_x - i k_y)$, with $v_2 (M_2)$ being the Fermi velocity (band gap) in monolayer BAs. For these two different stackings, the effective interlayer couplings have different strengths. Coulomb effect on the band inversion and phase diagrams in these two high symmetry points are shown in figure 2.

In the numerical calculations, we fix $\epsilon_{\parallel}/\epsilon_{\perp} = 1.6$ based on first-principles calculations of TMD bilayer [34], $D = 5.7$ Å from first-principles calculations of WSe$_2$/BAs heterobilayer, and parameters in the TMD massive Dirac model are obtained by fitting to first-principles band structures [7].

In the parameter regime considered here, we find overall five phases. Different phases are characterized by their different features of pairing gap $\Delta_{\tau k}$ (shown in figure 3) and Hall conductance when the Fermi level lies inside the global gap. For these two stackings, their interlayer couplings both have p-wave component alone but with different strengths, while the Coulomb interaction favors a s-wave interlayer pairing. The phase diagrams are resulted from their competition.

| $R/\sqrt{3}a$ | $t_{cc}$ | $t_{yy}$ | $t_{vv}$ | $t_{xy}$ |
|---------------|---------|---------|---------|---------|
| $AA_0$        | 0       | 33      | 0       | 0       |
| $AA_1$        | 1/3     | 0       | 98      | 0       |
| $AA_2$        | 2/3     | 0       | 0       | 106     |
| $AB_0$        | 0       | 0       | 0       | 81      |
| $AB_1$        | 1/3     | 0       | 0       | 0       |
| $AB_2$        | 2/3     | 46      | 113     | 0       |

Table 1. Interlayer coupling matrix elements $t_i$ (meV) at high symmetry points of AA and AB stackings for K valley. Different stacking registries are characterized by different interlayer translations $R$ along the longer diagonal of the supercell.
exciton binding energy, the exciton density suddenly increases and the system can become a s-wave exciton superfluid (ES) with minor effect brought by interlayer tunneling, or a QAH insulator with spontaneous time reversal symmetry breaking and nonzero Chern number. The former phase appears in the case of AA₀ stacking (figure 2(a)) with relatively weak interlayer tunneling, while the latter appears in the case of AA₁ stacking (figure 2(b)) where the interlayer tunneling is relatively strong. The spontaneous time reversal symmetry breaking in the QAH phase can be understood in the light of attractive intra-valley exciton exchange interaction as adopted by various other researchers [11, 32, 35, 36]. With single particle gap further reduced, the system eventually enters the QSH insulator phase, where interlayer tunneling dominates and Coulomb interaction renders a global gap by shifting the un-hybridized conduction band lies inside the hybridized band gap, resulting in a metal phase (M phase in figure 2(a)).

In the QSH phase, the overall layer hybridized band gap is strongly enhanced by Coulomb interaction compared with non-interacting limit, and sizable band gap along with electrical switchability of topological phases points to potential applications of this heterobilayer as dissipationless spintronic devices.

4.1.2. General translation
The small lattice mismatch between WSe₂ and BAs leads to moiré superlattice in their heterobilayer. For long-period moiré pattern, different regions in the moiré supercell are equivalent to commensurate bilayers with different interlayer translations. Modeling the moiré supercell by spatially varying interlayer tunneling, the phase diagram in the supercell can be subsequently obtained.

For AA stacking, the phase diagram is shown in figure 4, where we fix $\epsilon_\perp = 6$ and the band inversion as $E_g = -100$ meV. In this parameter regime, only

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**Figure 2.** Phase diagrams of AA stacking at two high symmetry points AA₀ (a) and AA₁ (b), as functions of single particle gap $E_g$ and interlayer dielectric constant $\epsilon_\perp$. In the parameter regime shown here, the two phase diagrams both have NI and QSH insulator phases. In phase diagram (a) with relatively weak interlayer tunneling, there are also ES and metal phases, while in (b) with relatively strong interlayer tunneling, there is a QAH insulator phase. The red dashed line marks the boundary of QSH insulator to metal phase transition, at which the global gap first vanishes.

**Figure 3.** Profiles of the ES, metal, QAH and QSH phases ((a)–(d)) for a typical set of parameters, including quasiparticle band structure, magnitude of order parameter $|\Delta|$ and phase angle $\text{arg}(\Delta)$. The shown range of momentum space is $[-\pi \frac{a}{10}, \pi \frac{a}{10}] \times [-\pi \frac{a}{10}, \pi \frac{a}{10}]$. 
one high symmetry local AA₁ can support topological phases.

For regions close enough to the high symmetry local, the weight of s-wave component is relatively small and the QSH phase is still preserved. Away from the high symmetry local AA₁, the strength of s-wave tunneling gradually increases, tending to turn the QSH phase into topologically trivial EI phase with the phase of pairing gap completely pinned and superfluidity lost. At the boundary of QSH to EI phase transition, band gap closes as in non-interacting limit (The finite band gap at the boundary here is due to the finite interval in numerical discretization of k and decreases for denser k-point meshes). Note that ES phases are also present for regions close to the high symmetry local AA₀ due to vanishing s-wave tunneling there, as further discussed in detail in section 5.

4.2. AB stacking
4.2.1. High symmetry points
Next, we consider the AB stacking case in lattice-matched heterobilayer. Only one high symmetry point AB₂ can support topological phases. At AB₂,  

\[ t_{\tau k} = \frac{1}{\sqrt{M}} \tau^\ell_{\mu} (k_x - i k_y) - \frac{1}{\sqrt{M}} \tau^c_{\nu} (k_x + i k_y) \]

The phase diagram at AB₂ stacking is shown in figure 5. The other parameters adopted in the numerical calculations are the same as AA stacking and there are overall three phases found.

The general feature of the phase diagram resembles that of figure 2(b). At large band gap, the exciton density is extremely small and Coulomb interaction has little effect on the NI phase. As the band gap is reduced below the exciton binding energy, the system enters the QAH insulator phase with spontaneous time reversal symmetry breaking, and finally turns into the QSH insulator phase as the band gap is further reduced. All these three phases are anisotropic in pairing gaps and quasiparticle dispersions due to the anisotropy of interlayer coupling. However, the qualitative feature of pairing gap \( \Delta \) in each phase is the same as figure 3.

4.2.2. General translation
Similarly, in the moiré supercell of AB stacking, the phase diagram is shown in figure 6. For regions close to the high symmetry local AB₂, the system lies in the QSH insulator phase, with anisotropic pairing gap. Far away from the local AB₂, the s-wave component of interlayer coupling dominates and the system enters EI phase. At the boundary of this topological phase transition, band gap should close. Here also due to

![Figure 4.](https://example.com/figure4.jpg)
finite discretization interval of $k$-points in numerical calculations, the minimal gap is nonzero.

5. Discussion

Type II band alignment forms between monolayer 2H-MoTe$_2$/2H-WSe$_2$/2H-Te$_2$ and hexagonal BAs, among which 2H-WSe$_2$ is the best choice from the aspect of lattice match with BAs [23], so that their heterobilayer consists of a moiré pattern with long period. However, the main features predicted here also apply to other choices of TMD/BAs heterobilayer. The local approximation works well in predicting the local physics in moiré superlattices of long period, but is incapable of giving the moiré miniband structure.

At the high symmetry locals where the $s$-wave component of interlayer tunneling vanishes, the phase of $s$-wave exciton condensate is unrestricted.
and counterflow superfluidity is preserved. As an unique feature, here the ES phase is also associated with an in-plane electric dipole [11], as the wave function of electron-hole relative motion is anisotropic due to $s$/$p$-wave component mixing. In other words, the overall electron-hole dipole tilts away from the out-of-plane direction. More specifically, analogous to cooper pair in Bardeen–Cooper–Schrieffer (BCS) theory of superconductivity [37], the exciton wave function in momentum space is $F_k = u_k v_k$. Fourier transform of which gives the exciton wave function in real space $F(r)$, with $r$ being the in-plane component of electron-hole separation vector. The in-plane electric dipole per exciton is then calculated as $d_{ex} = e \int dtF(r) i \tau F(r) / \int dt|F(r)|^2$. Decomposing the pairing gap function in terms of $s$- and $p$-wave components, $\Delta_{\pm k} = \tau \Delta_s(k) e^{-i r \theta} - \tau \Delta_p(k) e^{-i r \phi(k)}$, with $\phi(k)$ being the azimuth angle of $k$ and $\theta$ being the unrestricted condensate phase, the orientation of the in-plane electric dipole is uniquely determined by the condensate phase. In situ measurement of the in-plane electric dipole can therefore give direct information about the local condensate phase $\theta$. Note that only for pairing gap with both $s$- and $p$-wave components, the in-plane electric dipole is nonzero, while in other cases with $s$- or $p$-wave component alone, the in-plane electric dipole always vanishes and the overall electric dipole points perpendicular to the layer plane.

In the moiré supercell of AA stacking, the phase at high symmetry local AA0 is an ES, as $s$-wave component of interlayer tunneling vanishes there. Away from AA0, the strength of $s$-wave tunneling gradually increases, which tends to pin the phase of exciton condensate and therefore destroys counterflow superfluidity. So there is a crossover from ES phase to EI phase as the spatial deviation from AA0 stacking region increases. The moiré superlattice consists of patterns of ES and QSH phases separated by EI phases. Quantitative features of the ES to EI phase crossover can be explored using a minimal-field-theory model [38, 39], and will be an interesting subject for future study.

There is numerical evidence of gap closing at the NI and QAH phase boundary in figures 2(b) and 5, though due to symmetry change across the topological phase transition, the gap does not necessarily close in principle [40]. The gap-non-closing features at ES/QSH and QAH/QSH phase boundaries occur as first-order quantum phase transitions, like those reported in other systems [11].

The phase diagrams in figures 4 and 6 demonstrate the versatility of phases tunable experimentally by interlayer translation, together with the gate switchability of these phases, making this van der Waals heterobilayer advantageous over previous proposals of large-gap quantum spin Hall insulator systems.

6. Conclusion

Using WSe2/BAs heterobilayer as an example, we show that van der Waals heterobilayer of TMD/BAs can be a candidate system for realizing a QSH insulator with sizable band gap, which can be tuned between the topological and trivial phases by both interlayer bias and interlayer translation. This heterobilayer features a type II band alignment, with spin doubly degenerate conduction band and spin-valley locked valence band from BAs and TMD layer, respectively. When the bands get inverted by an interlayer bias, hybridization leads to a stacking-configuration dependent topological band inversion. However, in the absence of Coulomb interaction, the double spin degeneracy of BAs conduction band leaves an un-hybridized conduction band inside the hybridization gap, and hence the system is a spin-valley locked metal. Further taking into account the effect of Coulomb interaction, we find the un-hybridized conduction band can be shifted upwards in energy due to the interaction with the hybridization induced electric dipole associated with charge transfer between the two layers. A sizable global gap, of the order of hundred meV, appears in a broad parameter regime, tunable by both interlayer bias and stacking registry. Besides, the helical edge states in the QSH insulator phases can be switched on/off by the interlayer bias alone. Therefore, this heterobilayer provides a promising platform for engineering large gap QSH insulator and its moiré pattern in electrically tunable ways.

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Appendix A. Three-band model without interlayer hopping

The ground state wave function of the three-band model is $|\Psi\rangle = \prod_{r \neq k} (u_{rr} + v_{rr} d_{rr}^{\dagger} |b_k\rangle + v_{rr} d_{rr}^{\dagger} |\bar{b}_k\rangle) |0\rangle$, where $s = \uparrow (\downarrow)$, for $\tau = 1 (-1)$, and $s$ reverses the value of $s$. The mean-field energy reads
Here the summation over $\tau$ alone means the value of $s$ is simultaneously determined by the relation $s = \uparrow (\downarrow)$, for $\tau = 1 (-1)$, while the summation over both $\tau$ and $s$ indicates their values are independent.

Parameterize the mean-field wave function as $u_{r\k} = \cos \theta_{r\k}$, $v_{r\k} = \sin \theta_{r\k} \cos \phi_{r\k}$, $v_{r\k}' = \sin \theta_{r\k} \sin \phi_{r\k}$ and then the normalization condition $|u_{r\k}|^2 + |v_{r\k}|^2 + |v_{r\k}'|^2 = 1$ is readily satisfied. The above mean-field energy can be expressed as

$$\langle H \rangle = \sum_{r\k} \left( \varepsilon_{k\k'} - \varepsilon_{k\k'} \right) \left( |v_{r\k}|^2 + |v_{r\k}'|^2 \right)$$

$$- \frac{1}{2} \sum_{r\k,k'} U(k - k') |v_{r\k}|^2 |v_{r\k'}|^2$$

$$- \sum_{r\k,k'} V(k - k') u_{r\k} v_{r\k}^* u_{r\k'} v_{r\k'}^*$$

$$\frac{1}{2} \sum_{r\k} U(k - k') \sum_s |v_{r\k}|^2 \sum_s |v_{r\k}'|^2$$

$$+ \frac{2 \pi e^2 d}{\varepsilon} \left( \sum_{r\k} |v_{r\k}'|^2 \right)^2.$$

(A.1)

Table B1. The optimized layer distance $D$, the valence band splitting $\lambda_v$ and the conduction band splitting $\lambda_c$ at the $K'$ point for bilayer BAs with different stacking manners. The lattice constant is fixed as 3.39 Å.

|           | AA0 | AA1 | AA2 | AB0 | AB1 | AB2 |
|-----------|-----|-----|-----|-----|-----|-----|
| $D$ (Å)   | 4.50| 4.00| 4.00| 4.42| 3.97| 4.29|
| $\lambda_v$ (meV) | 442 | 187 | 188 | 11  | 1   | 588 |
| $\lambda_c$ (meV)  | 340 | 230 | 230 | 1   | 688 | 1   |

It can be easily seen that minimization of the expressions inside the square brackets in lines (A.2)–(A.4) with respect to $\phi_k$ and $\phi_k'$ leads to the condition that these two phase angles are both constants independent of $k$. The general $3 \times 3$ mean-field Hamiltonian in the basis $(\hat{a}_{k\k'}, \hat{a}_{k\k'}, \hat{a}_{k\k'})$ is

$$\hat{H}_\tau = \sum_{r\k,k'} \left( \hat{a}_{k\k'}^\dagger \hat{a}_{k\k'} \right) a_{r\k} \left( \hat{a}_{k\k'}, \hat{a}_{k\k'}, \hat{a}_{k\k'} \right)^T,$$

$$h_{r\k} = \begin{bmatrix} \varepsilon_{k\k} - \xi_{k\k} + \xi_0 & -\chi_{r\k}(k) & -\Delta_{r\k}(k) \\ -\chi_{r\k}(k) & \varepsilon_{k\k} - \xi_{k\k} + \xi_0 & -\Delta_{r\k}(k) \\ -\Delta_{r\k}(k) & -\Delta_{r\k}(k) & \varepsilon_{k\k} + \xi_{k\k} - \xi_0 &$$

(A.6)

where $\xi_0 = \frac{2 \pi e^2 d}{\varepsilon} \sum_{r\k} |v_{r\k}'|^2$, $\xi_{k\k} = \sum_{k'} U(k - k') \left( |v_{r\k}'|^2 + |v_{r\k}|^2 \right)$, and $\xi_{k\k} = \sum_{k'} U(k - k') |v_{r\k}'|^2$. The pairing gaps are defined as $\Delta_{\tau\k}(k) = \sum_{k'} V(k - k') u_{r\k} v_{r\k}^* \chi_{r\k}(k) = \sum_{k'} U(k - k') v_{r\k}' v_{r\k}^*$. With the condition that $\phi_k$ and $\phi_k'$ are constants, namely,$v_{r\k}, v_{r\k}' = \text{constant}$, the general Hamiltonian can be diagonalized to give three quasiparticle band dispersions, one of which is decoupled from the other two. The two coupled bands have dispersion relations $E_{r\k \uparrow} = \frac{2 \pi e^2 d}{\varepsilon} \sum_{r\k} |v_{r\k}'|^2 \varepsilon_{k\k} + \xi_0 + \Delta_{r\k}(k) |v_{r\k}'|^2$, and the decoupled one is $E_{r\k \downarrow} = \varepsilon_{k\k} + \xi_0$. 

Figure B1. Orbital projected band structure of a AA-type WSe$_2$/BAs heterobilayer. Inset: crystal structure of the heterobilayer.
Appendix B. First-principles calculation results

The band structure of a AA-type WSe$_2$/BAs heterobilayer, as demonstrated in figure B1, is calculated with density functional theory implemented in QUANTUM ESPRESSO software [41, 42]. The full-relativistic pseudo-potential with local density approximation (LDA) for the exchange-correlation functional has been used to include the spin-orbital coupling. The lattice constant of the WSe$_2$/BAs heterobilayer is fixed as 3.30 Å, and its crystal structure is optimized with the Broyden–Fletcher–Goldfarb–Shanno (BFGS) quasi-Newton algorithm. The convergence thresholds on the total energy and force are set as $10^{-4}$ Ry and $10^{-3}$ Ry/Bohr, respectively. The kinetic energy cutoff for the plane-wave function is set as 80 Ry, and the convergence threshold for the electronic self-consistent calculation is set as $10^{-10}$ Ry. The $k$-points in Brillouin zone are sampled by the $15 \times 15 \times 1$ Monkhorst–Pack grid. With the correction of van der Waals interaction by the vdW-DF method, the layer distance between the W and B atoms is determined as 5.7 Å. The band structure in figure B1 shows that the WSe$_2$/BAs heterostructure has a type II band alignment, where the valence band comes from the WSe$_2$ layer with a large spin splitting and the conduction band comes from the BAs layer with negligible spin splitting. The direct band gap at $K$ point for this structure is about 0.568 eV. Similar band structures have also been obtained for the heterobilayer with other stacking registries.

The band structures of bilayer BAs with different stacking registries have also been calculated in order to estimate the interlayer coupling $t_{B\text{As}}$. Here, the Perdew–Burke–Ernzerhof exchange-correlation functional without spin-orbital coupling has been exploited. By fixing the lattice constant as 3.39 Å, the optimized layer distances and the values of band splitting at the valence band maximum and conduction band minimum are listed in table B1.

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