Spin-Orbit Coupling in Transition Metal Dichalcogenide Heterobilayer Flat Bands

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The valence flat bands in transition metal dichalcogenide (TMD) heterobilayers are shown to exhibit strong intralayer spin-orbit coupling. This is reflected in a simple tight-binding model with spin-dependent complex hoppings based on the continuum model. A perpendicular electric field causes interlayer hybridization, such that the effective model is equivalent to the Kane-Mele model of topological insulators. The proposed model can be used as a starting point to understand interactions and the experimentally observed topological phases.

Heterostructures with Moiré patterns, emerging from the lattice mismatch or misalignment of two-dimensional materials, have proven to be a fertile ground for novel electronic states.[1–3] Amongst possible materials, ‘flat bands’ are expected to occur in bilayers of transition metal dichalcogenide (TMD) monolayer semiconductors. TMD heterobilayers, when the two layers are of a different material, have been proposed as an ideal realization of the triangular lattice Hubbard model with real short-range hoppings.[4]

Indeed, experimental results in WS₂/WSe₂ and MoTe₂/WSe₂ bilayers include the observation of a Mott insulating state at half-filling of the flat bands[5, 6] and generalized Wigner crystals at other fillings[7–10]. Recently, it came as quite a surprise that upon tuning a perpendicular electric field, both the quantum spin Hall effect (QSH) and quantum anomalous Hall (QAH) effect were observed in bilayer MoTe₂/WSe₂.[11] After all, the original heterobilayer model[4] did not include any spin-orbit coupling, in contrast to the homobilayer model that predicted topological bandstructures.[12]

There is a growing literature aiming to understand and characterize TMD heterobilayers,[13–27] where the observed topological transitions are proposed to originate in lattice relaxation[28] or interlayer coupling.[29, 30] However, as of yet there is no simple tight-binding model describing topological phases.

In this Letter, I reinterpret the continuum model of Ref. [4], using symmetry arguments related to the backfolding of the monolayer momenta to the Moiré mini-Brillouin zone. This directly yields strong spin-orbit coupling in the form of a complex spin-dependent hopping. The corresponding tight-binding model is equivalent to the next-nearest neighbor hopping in the Kane-Mele (KM) model. By tuning the band offset through a perpendicular electric field, the second layer comes into play and we find a full realization of the KM model with a corresponding topological transition and band inversion to a topological insulator phase. The tight-binding model derived in this Letter can serve as a starting point for studying the interplay between topology and interaction effects in TMD heterobilayers.

Continuum model — Let me first recap the essence of the continuum model of flat bands in TMD heterobilayers.[4] Monolayer transition metal dichalcogenide (MX₂ with M=W, Mo and X = S, Se, Te) have a hexagonal lattice with C₃ rotational symmetry. They are semiconductors with a relatively large direct bandgap at the K and K’ point. The valence band states have a large Ising spin splitting, such that states at K are spin↑ and the states at K’ are spin↓. Spin (↑ or ↓) and valley (K or K') are therefore inextricably coupled.

Whenever two different TMD monolayers are combined in a heterobilayer with twist angle θ, a Moiré pattern is created with lengthscale a_M = 1/√(1/π₁ + 1/π₂ − 2sinθ/a₁a₂) where a₁, a₂ are the lattice constants of the monolayers. Because different TMD monolayers have different band gaps, the top of the valence band of one layer lies in the gap of the other (a type-I or type-II band alignment). Consequently, the valence band in a heterobilayer exclusively consists of states localized in a single layer.

These heterobilayer valence band states can be described, per spin/valley, by imposing a Moiré potential V(r) on the monolayer valence band states,

\[ H = -\frac{\hbar^2 Q^2}{2m^*} + V(r) \]

\[ V(r) = \sum_{g_j} V_{g_j} \exp[i\mathbf{g_j} \cdot \mathbf{r}] \]

where \( g_j = \frac{4\pi}{\sqrt{3}} \left(-\sin\frac{2\pi(j-1)}{n}, \cos\frac{2\pi(j-1)}{n}, 0\right) \) are the six reciprocal Moiré vectors and \( Q \) is the momentum relative to the K/K' point of the monolayer. Because \( \Delta(r) \) must be real and the C₃ symmetry, we have \( V_1 = V_3 = V_5 \) and \( V_2 = V_4 = V_6 = V_4^* \)[4, 31, 32] which means one can parametrize the Moiré potential using only two parameters (V, ψ) such that \( V_1 = V e^{i\psi} \). For MoSe₂/WSe₂ bilayers, Ref. [4] calculated \((V, \psi) = (6.6 \text{ meV}, -94°)\). This results in a topmost valence flat band with almost perfect Gaussian Wannier orbitals centered on a Moiré triangular lattice.

Momentum backfolding — The continuum model is expressed using the momenta of the monolayer, here denoted in capital letters \( K_p, K'_p \) with layer index \( l \). The Moiré potential folds the momenta back to which mini-Brillouin zone (BZ) whose high-symmetry points are \( \gamma = (0,0) \), \( \kappa = (\frac{4\pi}{3a_M}, 0) \), \( \kappa' = (\frac{2\pi}{3a_M}, \frac{2\pi}{\sqrt{3}a_M}) \) and \( \bm{\mu} = \frac{1}{2} \mathbf{g_1} \). The question is which monolayer momenta correspond to which mini-BZ momenta.
In Ref. [4], the authors map the monolayer $K$ to the "point of the mini-BZ. Such backfolding only occurs when the Moiré length $a_M$ is a multiple of three times the monolayer lattice constant, as seen in Fig. 1c. In general, however, this is not true: the Moiré length in aligned commensurate cases can be expressed as $a_M = pa_1 = qa_2$ for coprime integers $p,q$. When both $p,q$ are not multiples of three, the $K$-points of the single layers map onto the $\kappa$-points of the mini-BZ. This is the case for aligned WS$_2$/WSe$_2$ with $a_M = 25\alpha_{WS2} = 26\alpha_{WS2}$ and aligned MoTe$_2$/WSe$_2$ with $a_M = 13\alpha_{MoTe2} = 14\alpha_{WSe2}$. Therefore the relevant momentum backfolding to describe the experiments is $K_1,K'_2 \to \kappa$ and $K_2,K'_1 \to \kappa'$, as shown in Fig. 1a. For general incommensurate and possibly twisted Moiré patterns we retain this mapping of momenta as shown in Fig. 1b. Note that in the continuum theory of TMD homobilayers the same mapping of momenta is used.[12]

With the correct momentum backfolding, the valence band structure of aligned AA-stacked WS$_2$/WSe$_2$ and AB-stacked MoTe$_2$/WSe$_2$ are shown in Fig. 2. For WS$_2$/WSe$_2$, I use $a_M = 7.98$ nm, $m^* = 0.36m_e$, and the Moiré potential $(V,\psi) = (7.7 \text{ meV}, -106^\circ)$ is calculated using density functional theory with Quantum ESPRESSO[33, 34] with a Coulomb cut-off[35] and the method of Ref. [4]. For MoTe$_2$/WSe$_2$, I use the parameters $a_M = 4.55$ nm, $m^* = 0.65m_e$, and $(V,\psi) = (7 \text{ meV}, -14^\circ)$ based on Ref. [30]. The resulting flat bands have a very clear spin-orbit splitting at $\kappa$ and $\kappa'$. The general structure of spin/valley-split bands except along the line $\gamma - \mu$ was also observed in large-scale DFT calculations of WS$_2$/WSe$_2$[22] and MoTe$_2$/WSe$_2$[29].

Tight-binding model – The tight-binding model for the valence flat bands can be derived using symmetry arguments. I will focus only on nearest neighbor hopping $t_1$ since it is much larger than the longer-ranged hoppings $t_2,t_3$. The $C_3$ symmetry of the heterobilayer implies that $t_1$ can be complex, $t_1 = |t_1|e^{i\phi}$, as shown in Fig. 3a. Within the continuum model of Eq. (1), the band-structure is six-fold symmetric around the top of the flat band. Given that the top of the flat band is positioned at $\kappa$ or $\kappa'$, it directly follows that the complex phase equals $\phi = \pm \frac{2\pi}{3}$, respectively. The resulting tight-binding model is thus a triangular lattice with spin-orbit coupled hopping

$$H = t_1 \sum_{(ij)\sigma} e^{i\phi \sigma^z \nu_{(ij)}} c_{i\sigma}^\dagger c_{j\sigma}$$

where $\nu_{(ij)} = \pm 1$ depending on the direction of the bond,
where $\langle \langle ij \rangle \rangle$ represents the next-nearest neighbor on the honeycomb lattice where $\nu_{\langle\langle ij \rangle\rangle} = \pm 1$ depends on the direction, $\ell = \pm 1$ the layer index, and $\sigma^z = \pm 1$ the spin. This is exactly the Kane-Mele spin-orbit coupling term.\cite{37, 38}

The interlayer hopping is now given by nearest neighbor hopping on the honeycomb lattice. The $C_3$ symmetry once again constrains the possible complex phases of the interlayer hopping, as described in Ref. [29]. For the tight-binding model this yields

$$H_\perp = t_\perp \sum_{\langle\langle ij \rangle\rangle} e^{i\frac{2\pi}{3} \nu_{\langle\langle ij \rangle\rangle} \sigma_1} c_i^\dagger \sigma \ell c_j$$

where $\langle\langle ij \rangle\rangle$ now couples the nearest neighbors on the honeycomb lattice. The parameter $\nu_{\langle\langle ij \rangle\rangle} = 0.1$ or 2 depends on the direction and increases counterclockwise when going around the MoTe$_2$ lattice sites. Eqs. (3)-(4) form an effective tight-binding model constrained by symmetry and is visualized in Fig. 3b.

In the absence of an electric field the flat bands from the WSe$_2$ layer are at a much lower energy than the MoTe$_2$ states. A perpendicular electric field $V$ can shift the WSe$_2$ states upward,

$$H_V = (V + \Delta) \sum_i n_i, \ell = 2$$

where $\Delta < 0$ is the band offset. By increasing $V$ the bands from the two layers will overlap, at which point the interlayer coupling Eq. (4) becomes relevant. With $t_2 = 3.4$ meV extracted from the effective mass of the WSe$_2$ valence band and $t_\perp = 4$ meV, we calculate the bandstructure as a function of perpendicular field in Fig. 4. Above the critical field value $V_c$, a band inversion happens at the $\kappa'$ point for the states in the $K$-valley. For $V > V_c$, the top valence flat bands obtain a nonzero Chern number which is opposite for the two spin species.\cite{39} As such, the system has become a topological insulator fully described by the honeycomb lattice model of Kane-Mele.

The effective Kane-Mele model applies to all TMD heterobilayers provided the applied perpendicular electric field is sufficient to overcome the valence band offset.

**Interactions** – The presence of spin-orbit coupling has some implications for the possible interacting states. In particular, at half-filling of the topmost flat band the complex hopping phases lead to an effective spin model with Dzyaloshinskii-Moriya (DM) interactions.\cite{27} Its strength, characterized by the hopping phase $\phi = \frac{2\pi}{3}$, stabilizes an in-plane $120^\circ$ antiferromagnetic order. At the same time, Chern bands at half-filling are known to be susceptible to full spin polarization leading to a Quantum Anomalous Hall (QAH) effect.\cite{40, 41} The Mott-QAH transition observed in Ref. [11] can possibly by understood as a metamagnetic transition from in-plane Néel to Ising ferromagnetic order.

Note that within the current model, there is no band gap between the topologically nontrivial bands. However,
interaction-driven renormalization of the bands can open up the gap at full filling of the top valence flat bands.\cite{30}

**Conclusion** – I showed that by correctly mapping the monolayer momenta onto the mini-BZ, the effective model of TMD heterobilayers obtains a strong spin-orbit coupling as given by Eq. (2). The interlayer coupling can be described by a Kane-Mele model following Eqs. (3)-(5), yielding a topological transition as a function of perpendicular electric field.

Having an effective tight-binding model is an important step towards a full understanding of possible strongly correlated phases. This Letter shows that spin-orbit coupling cannot be ignored in further studies of TMD heterobilayers.

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