The moiré potential in twisted transition metal dichalcogenide bilayers

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Moiré superlattices serve as a playground for emerging phenomena, such as localization of band states, superconductivity, and localization of excitons. These superlattices are large and are often modeled in the zero angle limit, which obscures the effect of finite twist angles. Here, by means of first-principles calculations we quantify the twist-angle dependence of the moiré potential in the MoS$_2$ homobilayer and identify the contributions from the constituent elements of the moiré potential. Furthermore, by considering the zero-angle limit configurations, we show that the moiré potential is rather homogeneous across the transition metal dichalcogenides (TMDs) and briefly discuss the separate effects of potential shifts and hybridization on the bilayer hybrid excitons. We find that the moiré potential in TMDs exhibits both an electrostatic component and a hybridization component, which are intertwined and have different relative strengths in different parts of the Brillouin zone. The electrostatic component of the moiré potential is a varying dipole field, which has a strong twist angle dependence. In some cases, the hybridization component can be interpreted as a tunneling rate but the interpretation is not generally applicable over the full Brillouin zone.

I. INTRODUCTION

Hexagonal bilayer transition metal dichalcogenides (TMDs) with a rotational misorientation between the monolayer sheets can exhibit an interference pattern in the atomic positions known as a moiré pattern. The latter can alter the electronic structure of the twisted monolayer sheets can exhibit an interference pattern in the atomic plane of the monolayer sheets, which we denote as the moiré unit cell (Fig. 1). We aim to provide a unifying perspective of the moiré potential in TMDs, in particular of the twist-angle dependence of the induced potential. Specifically, the moiré potential in MoSe$_2$/WSe$_2$ heterobilayers has been shown to be on the order of 150 to 300 meV and dominated by planar strain, and the band gap in MoS$_2$/MoTe$_2$ has been observed to be strongly twist-angle dependent. A comprehensive description that connects the aforementioned observations and establishes general trends that hold across different combinations of TMDs is, however, still needed.

Here, we therefore assess the moiré potential via first-principles calculations both on explicit moiré configurations and limiting configurations based on the primitive unit cell (Fig. 1). We aim to provide a unifying perspective of the moiré potential in TMDs, in particular of the origin of the potential and the similarity of the potential in different bilayer systems. Finally, we disentangle the electrostatic part of the moiré potential and determine the hybridization contribution at high symmetry points in the moiré superlattice.

II. METHODOLOGY

In order to construct the explicit moiré superlattices we have used the method outlined in Ref. 8. The commensurate hexagonal moiré superlattices are found at angles

$$\theta_k = \arccos \left( \frac{3k^2 + 3k + 0.5}{3k^2 + 3k + 1} \right),$$

where $k$ is a non-negative integer. In this work, we consider values up to $k = 7$, corresponding to an angle of 4.41°. The variations in the local atomic environment give rise to an electrostatic potential that varies in the plane of the monolayer sheets, which we denote as the intralayer moiré potential. By definition, the difference of the intralayer moiré potentials in the two adjacent layers is the interlayer moiré potential.

We consider two different measures of the electrostatic potential induced by twisting: (1) The (average) elect-
FIG. 1. (a) The explicit MoS$_2$ homobilayer moiré structure at a twist angle of $\theta = 2.9^\circ$ along with the limiting ($\theta \to 0^\circ$) local registry at three specific points. Blue (yellow) atoms indicate atomic species Mo (S). (b) Lateral relaxation for the MoS$_2$/MoS$_2$ homobilayer twisted 4.41$^\circ$. The arrows are amplified with a factor of 30.

trostatic core potential is a measure of the local electrostatic environment of the relevant electronic states, which in TMDs are primarily composed of $d$ orbitals of the transition metal.$^9$ (2) The second measure, which is closely related to the first is the induced dipole field.$^{10}$ It is strictly applicable only to the limiting configurations, since in explicit moiré superlattices, the field exhibits higher moments. The charge density difference upon bilayer formation is

$$\delta n(r) = n(r) - \sum_i n_i(r), \quad (2)$$

where $n(r)$ is the charge density of the bilayer structure and $n_i(r)$ denotes the charge densities of the constituent monolayers. The charge density difference $\delta n$ gives rise to a potential $\delta V$. In order to accurately determine the potential $\delta V$ and the induced vacuum level difference $D$

$$D = \delta V(-\infty) - \delta V(\infty), \quad (3)$$
dipole corrections are added when solving the Poisson equation for the electron density.

In order to estimate how $\delta V$ affects different single particle states we compute the following matrix elements for the monolayer states

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$$M^L_{nk} = \langle \psi^L_{nk} | \delta V | \psi^L_{nk} \rangle, \quad (4)$$

where $L$ is the monolayer index and $n$ is either $v$ (valence band) or $c$ (conduction band). In the spirit of perturbation theory, $M^L_{nk}$ is the first-order shift of the state $nk$ in monolayer $L$ due to the induced electrostatic potential. The (average) hybridization contribution to the level shift upon bilayer formation is then computed as

$$\Delta_{nk} = \frac{1}{2} \left[ \epsilon^\text{bilayer, +}_{nk} - \epsilon^\text{bilayer, -}_{nk} - (M^+_{nk} - M^-_{nk} + \Delta\epsilon^{\text{mono}}_{nk}) \right], \quad (5)$$

where $\epsilon^\text{bilayer, +}_{nk}$ is the energy of the bilayer state that shifts towards larger energies, $M^+_{nk}$ is the matrix element in the layer which experiences a positive potential, and $\Delta\epsilon^{\text{mono}}_{nk}$ is the energy level difference between the monolayers (which is zero for homobilayers). We will also make
use of the (average) interlayer tunneling rate

\[ T_{nk}^2 = \frac{1}{2} \left( \left( \varepsilon_{nk}^{\text{bilateral},+} - \varepsilon_{nk}^{\text{bilateral},-} \right)^2 \right) \]

\[ - (M_{nk}^+ - M_{nk}^- + \Delta_{nk}^{\text{mono}})^2 \],

(6)

which may be interpreted as the off-diagonal perturbation of a two-level system. These tunneling strengths may be, e.g., included in a tight-binding model for the exciton energies of twisted structures.\(^2\)

For the purpose of this paper, we consider a simpler case of untwisted structures to disentangle the effect of electrostatic effects and hybridization.\(^11\) To this end, we use the exciton density matrix formalism\(^12\)-\(^14\) and calculate the exciton energies for untwisted van-der-Waals heterostructures by first formulating a Hamiltonian in second quantization for the interaction free electronic energies

\[ H = \sum_{\alpha lk} \hat{E}_{\alpha lk}^\text{ex} a_{\alpha lk}^\dagger a_{\alpha lk} + \sum_{\alpha kl \neq l'} T_{\alpha l l'}^{\text{ex}} a_{\alpha l k}^\dagger a_{\alpha l l'}, \]

(7)

where \( l, l' \) are layer indices, \( \alpha = (\lambda, \xi) \) is a compound index with \( \xi \) denoting the valley and \( \lambda = (c, v) \) the conduction and the valence band respectively. Here \( a^\dagger (a) \) denotes the electronic creation (annihilation) operator. The possible excitonic transitions in this way are illustrated in Fig. 2. Here, \( k \) is the electronic wave vector and \( s \) is the stacking index. \( E_{\alpha lk}^\text{ex} \) are the electronic energies as well as the electrostatically induced alignment shifts. \( T_{\alpha l l'}^{\text{ex}} \) is the tunneling matrix element, which is modeled following a tight-binding approach.\(^15,16\) Consequently, the tunneling matrix element are directly proportional to the tunneling strength for those stackings, for which tunneling is allowed by \( C_3 \) symmetry. By following the method laid out in Refs. \(^2\) and \(^1\), the Hamiltonian is transformed into an exciton basis, thus taking into account the binding energies. The final hybrid exciton energies are calculated by first transforming into a hybrid basis \( \tilde{Y}_{\xi \eta}^L = \sum_{L} \tilde{C}_{\xi \eta}^{L}(Q)^{\dagger} X_{\xi \eta}^{L} \), which consists of a linear combination of the intra and interlayer exciton contributions \( \tilde{C}_{\xi \eta}^{L}(Q) \) to the final hybrid state. Here, \( L \) is a compound layer index, \( Q \) is the center-of-mass momentum, \( \eta \) the new band index and \( X^{\dagger}(X) \) are the exciton creation (annihilation) operators. The eigenvalue problem that arises in order to diagonalize this new hybrid exciton Hamiltonian yields the final exciton energies.

DFT calculations were performed using the projector augmented wave method\(^17\) while atomic configurations were prepared and analyzed using \( \text{ASE} \).\(^18\) All structural relaxations were carried out using \( \text{VASP} \)\(^19,20\) and the \( \text{vdW-DF-cx} \) method.\(^21,22\) The monolayer lattice parameters were obtained using a plane wave cutoff of 340 eV and a \( k \)-point mesh with a density of 0.3 Å\(^{-1}\). The atomic positions for the limiting bilayer structures (Fig. 1) were relaxed using the same functional but with a cutoff energy of 500 eV and a \( 18 \times 18 \times 1 \) \( k \)-point mesh until the maximum force acting on any atom was less than 2.5 meV Å\(^{-1}\). The explicit moiré superlattices with up to 1,014 atoms were relaxed using a plane wave cutoff of 340 eV until the maximal force acting on any ion was less than 7.5 meV Å\(^{-1}\). The cell vector in the out-of-plane direction was 70 Å for these configurations to ensure decoupling across periodic boundary conditions.

The analysis of the electronic densities and potentials was carried out using \( \text{GPW} \)\(^23,24\) and the local density approximation (LDA).\(^25\) The electron densities of the limiting bilayer configurations were computed using a grid expansion with 0.15 Å spacing and a \( 18 \times 18 \times 1 \) \( k \)-point mesh. Here, a compensating dipole layer was included in the Poisson solver.

III. RESULTS

The TMDs with \( \text{MX}_2 \) (\( M=\text{Mo,W};X=S,\text{Se} \)) constitute a class of materials with similar behavior and here we therefore only carry out computations for large explicit moiré superlattices for the \( \text{MoS}_2/\text{MoS}_2 \) homobilayer. Computations and analysis of the limiting bilayer stackings have, however, been performed for all homobilayers and lattice constant matched heterobilayers.

A. Structural properties

First we report the computations of the interlayer distance, which is of fundamental importance since the wave function overlap of the constituent monolayer states is sensitively dependent on the interlayer distance. Therefore, both polarization and hybridization magnitude depend on the interlayer distance. In the zero-angle limit configurations, the interlayer distance for \( \text{MoS}_2/\text{MoS}_2 \)
varies between 6.86 Å in \( R_h^b \) and 6.10 Å in \( R_h^X \) and \( R_h^M \) in decent agreement with other theoretical estimations.\(^2\)\(^6\)

In moiré superlattices the interlayer distance varies with the lateral position. In order to quantify the interlayer distance for the explicit moiré structures, we therefore consider the distribution of distances (Fig. 3a), which we obtain by centering a cylinder of radius 3 Å on a transition metal atom in the upper layer and then taking the interlayer distance as the vertical distance between transition metal atoms found within the same cylinder.

At the largest considered twist angle (13\(^{o}\)) the interlayer distance is rather homogeneous with a value roughly equal to the mean of the interlayer distance in \( R_h^b \) and \( R_h^X \). This is not surprising since the interlayer interaction is very weak due to its van-der-Waals nature while the deformation of the monolayer sheets requires bending the much stronger covalent bonds. As the twist angle decreases the distribution widens. The extrema occur at \( R_h^b \) and \( R_h^M \) (\( R_h^X \)) and the local environment of the moiré superlattice at these specific points mimics the limiting configurations more and more as the twist angle decreases. The lower limit of the distribution shifts to smaller values (Fig. 3a) but even at the lowest twist angle considered, the smallest values are still slightly larger than the interlayer distance for the limiting \( R_h^b \) and \( R_h^M \) structures where the interlayer distance attains its minimum (which corresponds to the lower limit of the x-range in Fig. 3a).

One can also consider the distribution of the lateral displacements of the Mo atoms relative to the ideal moiré superlattice (\( \delta d_{Mo}^{xy} \)) that appear during relaxation (Fig. 3b-e). The largest twist angle shown is 7.34\(^{o}\) in which the maximal lateral displacement of the Mo atoms is 0.025 Å. For the smallest twist angle (4.41\(^{o}\)), the maximum relaxation distance of the Mo atoms is significantly larger at 0.065 Å. For the 4.41\(^{o}\) twist angle, the planar displacements forms three bands (Fig. 3e) corresponding to different distances of the displaced atom from the rotation center with a local AA stacking order. The direction of the lateral displacements for the Mo atoms is shown in Fig. 1b. The regions around the high symmetry points are almost vertically aligned and exhibit very little relaxation. The main relaxation occurs between the symmetry point \( R_h^b \) and \( R_h^X \) (and between \( R_h^M \) and \( R_h^b \)).

B. Moiré potential

Next we turn to the moiré potential. In Fig. 4a, a schematic illustration of the vacuum level difference (Eq. 3) as a function of displacement along the long diagonal of the unit cell is shown. The minimum occurs at \( R_h^X \) while the maximum occurs at \( R_h^M \). The origin of the spatially varying vacuum level difference is the stacking dependent density polarization (Fig. 4b-d), which gives rise to asymmetric (with respect to the plane halfway between the monolayers) potentials (Fig. 4e-g). The potentials shown in (Fig. 4e-g) would correspond to the interlayer moiré potential at these specific sites within the moiré superlattice and the magnitude of this potential is around 80 meV for the MoS\(_2\)/MoS\(_2\) homobilayer. Due to symmetry, this is also the full variation of the intralayer moiré potential, and taken together it places the full variation of the interlayer moiré potential at around 160 meV for this system.

We have also studied the twist angle dependence of the intralayer moiré potential by considering the maximal planar variation of the core potentials (Fig. 5). We find that it decays rapidly with increasing twist angle and already at \( \approx 5^{o}\) it has decayed to half of the zero limit value. Part of this decay can likely be explained by the increasing minimal interlayer distance with increasing twist angle and partly by the deviation from ideal stacking at \( R_h^b \) and \( R_h^M \). This behavior of the moiré potential is consistent with explicit calculations of the MoS\(_2\)/MoTe\(_2\) system,\(^7\) with measured energy barriers of exciton diffusion in twisted MoSe\(_2\)/WSe\(_2\)\(^2\)\(^7\) and in the localization of excitons in MoS\(_2\)/WS\(_2\).\(^2\)\(^8\)

The twist angle dependence of the moiré potential was computed without dipole corrections. In order to validate this approach we performed a test with an out-of-plane lattice vector of 100 Å and compared it with the 70 Å results. We found that the maximal planar variation of the Mo core potential for the 6.01\(^{o}\) structure was 26.1 meV in both cases confirming that the spurious electric field does not influence the results.

C. Hybridization and tunneling in MoS\(_2\)/MoS\(_2\)

The electrostatic moiré potential rigidly shifts the single particle electronic states according to Eq. (4). The bilayer states are, however, subject to interlayer hybridization as well, which for MoS\(_2\)/MoS\(_2\) homobilayer is quantified in Fig. 6 along the path between the \( \Gamma \) and \( K \) points. In all configurations, the hybridization contribution to the valence band shifts is small at the \( K \) point and large at \( \Gamma \). For the conduction band, the hybridization contribution is largest at \( \Lambda \) (halfway between \( \Gamma \) and \( K \)). At the \( \Gamma \) point the hybridization is much stronger in \( R_h^X \) and \( R_h^M \) than for \( R_h^b \). At \( K \), on the other hand, the valence band hybridization is slightly larger in \( R_h^b \) (15 meV) compared to \( R_h^X \), and \( R_h^M \) (5 meV) despite exhibiting a much larger interlayer distance. For the conduction band at \( K \) in \( R_h^X \), and \( R_h^M \) the hybridization contribution is negative with a value of \( -2 \) meV.

The tunneling rates defined in Eq. (6) largely follow the hybridization energy but become imaginary when the hybridization energy is negative, which implies that the solution becomes non-physical (Sect. IV). This occurs at the \( \Lambda \) point for the valence band and the \( K \) point for the conduction band in \( R_h^X \) and \( R_h^M \). The tunneling rate at \( \Lambda \) for the conduction band is 132 meV in \( R_h^b \) rising to 184 meV in \( R_h^X \) and \( R_h^M \). The tunneling rate for the \( K \) point is 2 meV (conduction band) and 15 meV (valence band) in \( R_h^b \). The latter value rises to 19 meV in \( R_h^X \) and
FIG. 3. (a) Distribution of interlayer distances in MoS$_2$/MoS$_2$ homobilayer as a function of twist angle. The lower and upper limits of the x-axis correspond to the layer spacing in $R^h_h$ and $R^M_h/R^X_h$, respectively. (b–e) The distribution of lateral relaxation for the four lowest twist angles.

$R^M_h$ while the former becomes imaginary. It was shown in Ref. 29 that the tunneling rate at $K$ vanished in $R^X_h$ and $R^M_h$ under the assumption that the orbital character of the monolayer band edges were completely composed of transition metal $d$ states. We attribute the appearance of a finite tunneling rate for the valence band at $R^X_h$ and $R^M_h$ at $K$ to orbital mixing with other states. Based on semi-local DFT calculations, it was found in Ref. 30 that there is a minor but non-zero contribution to the band edge states coming from the chalcogen species.

The tunneling rates at $\Lambda$ and $K$ for $R^h_h$ and $R^X_h$ are to a good approximation linear in the interlayer distance (Fig. 7). In $R^h_h$, the interlayer dependence is stronger for both the states at $\Lambda$ compared with $K$, whereas the dependence is very weak for the conduction band states. In $R^M_h$, the conduction band at $\Lambda$ and the valence band at $K$ exhibit stronger interlayer dependence than the other two states considered.

FIG. 4. (a) Schematic illustration of the vacuum level difference (Eq. 3) obtained by displacing the upper layer along the long diagonal of the primitive cell, keeping the interlayer distance fixed. (b–d) Electron density ($\delta n$ of Eq. 2) averaged in-plane for configurations $R^h_h$, $R^X_h$, and $R^M_h$ of MoS$_2$/MoS$_2$. (e–g) Corresponding potential $\delta V$ averaged in-plane.

FIG. 5. Maximal planar variation of the core potential for MoS$_2$/MoS$_2$ moiré superlattices. 4.41° is the smallest twist angle for which explicit computations have been performed. The data point (indicated by a square) at 0° is the zero degree limit computed as the Mo core potential difference at $R^h_h$ and the connecting line is a guide to the eye. The gray area indicates the range between the smallest and largest interlayer distance.
FIG. 6. Average hybridization contribution according to Eq. (5) of the valence band and conduction band for the limiting configurations of MoS$_2$/MoS$_2$ between the $\Gamma$ and $K$ points superimposed on the monolayer bands computed within the LDA.

FIG. 7. Tunneling rates as a function of interlayer distance for (a) $R^h_R$ and (b) $R^X_R$ of MoS$_2$ homobilayer.

D. Extension to other TMDs

In Fig. 8 the vacuum level difference $D$ as defined in Eq. (3) is shown for all Mo and W-based TMDs including the heterobilayers. For the homobilayers, the magnitude of the vacuum level difference is more or less uniform with minor variations between different TMDs despite rather large differences in the interlayer distance.

For the heterostructures, in contrast to the homobilayers, there is a vacuum level splitting present in $R^h_R$ due to the presence of different transition metals. However, the full variation of the interlayer moiré potential is still rather similar over both the homobilayers and heterobilayers with an average of 131 meV (standard deviation is 8 meV). This is likely due to the fact that the relevant orbital character of all the TMDs considered are similar.

E. Excitons in TMD bilayers

We have briefly investigated how the layer dependent polarization and hybridization influence the exciton spectrum in the different stackings originating from the AA stacked system. The present analysis has been performed by solving the Wannier equation in a basis of the constituent monolayers (see Eq. (7)) and the resulting excitons contains both intra and inter-layer components (Fig. 2).

The exciton spectra were investigated for the two prototype cases $i$) homobilayer WS$_2$, and $ii$) the MoSe$_2$/WSe$_2$ heterobilayer. In Fig. 9, the energy of the lowest lying exciton peak relative to the $K - K$ exciton in the $R^h_R$ stacked structure is shown for the WS$_2$ homobilayer, and in Fig. 10, the same data is shown for the MoSe$_2$/WSe$_2$ heterobilayer. The transparent bars include the polarization shift but not the hybridization energy. For the homobilayer, the lowest $K - K$ exciton is of intralayer nature and hence the stacking dependent polarization does not affect the position of the peak. In particular, there is a very weak hybridization of the $K - K$ excitons. For the case of excitons that involve the $K - \Lambda$ valleys, the energy is around 150 meV lower than the $R^h_R$ $K - K$ exciton with the largest contribution stemming from the hybridization of the conduction band at the $\Lambda$ point. To properly see the effect of hybridization, we consider the $\Gamma - K$ exciton in the $R^h_R$ stacking. Without accounting for hybridization, the exciton energy is about 200 meV higher in energy in comparison with the $R^h_R$ $K - K$ exciton, since the valence band maximum (VBM) at $\Gamma$ is lower in energy in comparison with at the $K$-point. The VBM at $\Gamma$ is pushed to higher energies by hybridization and the exciton energy becomes lower than the $R^h_R$ $K - K$ exciton. The $\Gamma - \Lambda$ indirect exciton is the one mostly affected by the hybridization since it is strong in both these valleys.

In the heterobilayer however, where the lowest exciton exhibits interlayer character, there is a pronounced effect of the polarization field on the lowest $K - K$ exciton and the exciton energy difference between the different stacking orders corresponds to the variation of the dipole field. The $K - \Lambda$ exciton is, without accounting for hybridization located around 150 meV-200 meV above the $R^h_R$ exciton. This energy drops significantly due to hybridization pushing the $\Lambda$ valley to lower energies. In contrast to the case of the homobilayer where the indirect $\Gamma - K$ exciton is lower in energy with respect to the $R^h_R$ $K - K$ exciton, the $\Gamma - K$ exciton in the heterobilayer has a much larger energy in comparison with the $R^h_R$ $K - K$ exciton.

IV. DISCUSSION

The main result of this study is that the moiré potential exhibits a strong twist angle dependence, as is evident from Fig. 5. The origin of this potential is an
The implications of a twist angle dependent potential are manifold. For example, it determines for which angles the electronic states at the band edges become localized. Furthermore, the twist angle dependence has implications for the temporal localization of charge carriers, since the potential is effectively an energy barrier. One of the charge carriers will be subject to an energy barrier induced by the dipole potential. For example, the transition from the lowest hole potential configuration to $R_h^i$ raises the hole energy by $D/2$. With increasing twist angle the energy barrier becomes smaller. Therefore, it is expected that the temporal localization of charge carriers is strongest at very small twist angles.

Exciton migration may proceed via resonant energy transfer but in the case that electron and hole migrate separately the current results are consistent with measurements on exciton diffusion. The interlayer exciton diffusion in MoSe₂/WSe₂ was investigated in Ref. 33 and it was found that the exciton diffusion was considerably larger at a 3.5° twist angle than a 1° twist angle.

The numerical values of the dipole field amplitude $D$ are quite similar for all the TMDs considered here (Fig. 8). The similarity of $D$ despite vastly different interlayer distances is interesting in itself and suggests that the dipole field is primarily a geometrical property, which, together with orbital similarity across this class of materials, results in a similar magnitude. In fact, the interlayer distance and the induced density difference $\delta n$ are not independent quantities. The density difference forms mainly between the layers, creating a bonding electron density that is slightly polarized, which in turn gives rise to an electric field that balances the bonding electron density and dispersion interaction.

The moiré potential affects inter and intralayer excitons differently in the bilayer structures (Fig. 9 and Fig. 10) and the energy for the $K - K$ exciton with interlayer character exhibits an energy variation over the stacking orders $R_h^i$, $R_h^X$, and $R_h^M$ that closely resembles the dipole field magnitude. The tunneling rates at the conduction band at $K$ and the valence band at $\Lambda$ are imaginary at $R_h^X$ and $R_h^M$ with similar values of around 10i – 20i meV. While the $\Lambda$ valence band is largely irrelevant due to its low energy, the conduction band at $K$ is rather important for the excitonic spectra. The tunneling rates are, as constructed, off-diagonal components to a perturbation of the two-level system consisting of the equivalent band from different monolayers. In this case,
FIG. 10. Energy of the lowest exciton peak relative to the lowest $K(\text{hole}) - K(\text{electron})$ exciton for the MoSe$_2$/WSe$_2$ heterobilayer. The shaded bars indicate the influence of the polarization shift and the solid bars indicate the influence of both the polarization shift and hybridization.

the splitting of the levels in the bilayer system is smaller than the difference in potential shift between the states. The tunneling matrix element represents the interaction that causes band edge shifts that go beyond the splitting that is caused by potential alignment. To describe this effect, here we employ a simple two-band model, see Eq. (6). The appearance of imaginary solutions can be attributed to the breakdown of this approximation and signify that the bilayer energy shifts are influenced by coupling to multiple states.

V. CONCLUSIONS

To summarize, we have shown that the moiré potential in TMDs exhibits both an electrostatic component and a hybridization component, which are intertwined and have different relative strengths in different parts of the Brillouin zone. The electrostatic component of the moiré potential is a varying dipole field, which has a strong twist angle dependence. In some cases, the hybridization component can be interpreted as a tunneling rate but the interpretation is not generally applicable over the full Brillouin zone.

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