Exchange-bias controlled correlations in magnetically encapsulated twisted van der Waals dichalcogenides

D Soriano¹ and J L Lado²

¹ Radboud University, Institute for Molecules and Materials, NL-6525 AJ Nijmegen, The Netherlands
² Department of Applied Physics, Aalto University, 00076 Aalto, Espoo, Finland

E-mail: jose.lado@aalto.fi

Received 19 June 2020, revised 19 July 2020
Accepted for publication 28 July 2020
Published 25 August 2020

Abstract
Twisted van der Waals materials have become a paradigmatic platform to realize exotic correlated states of matter. Here, we show that a twisted dichalcogenide bilayer (WSe₂) encapsulated between a magnetic van der Waals material (CrBr₃) features flat bands with tunable valley and spin flavors. We demonstrate that, when electron–electron interactions are included, spin–ferromagnetic and valley–ferromagnetic states emerge in the flat bands, stemming from the interplay between correlations, intrinsic spin–orbit coupling (SOC) and exchange proximity effects. We show that the specific symmetry broken state is controlled by the relative alignment of the magnetization of the encapsulation, demonstrating the emergence of correlated states controlled by exchange bias. Our results put forward a new van der Waals heterostructure where symmetry broken states emerge from a genuine interplay between twist engineering, SOC and exchange proximity, providing a powerful starting point to explore exotic collective states of matter.

Keywords: twisted van der Waals materials, magnetic heterostructure, moiré material, exchange bias

(Some figures may appear in colour only in the online journal)

1. Introduction

Two-dimensional materials have risen as a paradigmatic platform to engineer emergent states of matter. This flexibility stems from the capability of stacking different two-dimensional materials on top of each other, allowing to combine electronic orders. Among the different two-dimensional materials, transition metal dichalcogenides [1–6] attracted much attention due to their strong spin–orbit coupling (SOC) effects. This has allowed exploiting different early proposals in spintronics such as spin-charge conversion [7], valley control [8–11], and optically generated spin currents [12, 13], demonstrating the possibilities for magnetic control in two-dimensional materials. In this line, the recent discovery, and isolation of two-dimensional magnetic materials [14–20] has allowed pushing this idea even further, providing a purely van der Waals platform for spintronic physics.

Moiré structures are known to emerge in generic twisted two-dimensional materials, including graphene [21, 22], transition metal dichalcogenides (TMDC) [23–29], boron nitride [30], MoO₃[31] and ferromagnets [32]. In particular, twisted transition metal dichalcogenide systems provide unique opportunities for spintronics, due to the strong SOC [8, 33–38]. Correlated states in twisted dichalcogenides materials have been also demonstrated, stemming from the emergence of quasi-flat bands [23, 24, 39–45, 45–53]. Importantly, the strong spin–orbit effects, together with the emergent valley and layer degrees of freedom, bring forward a whole new set of possibilities [3, 37, 38, 54] for engineering strongly correlated states of matter in twisted dichalcogenides. In particular,
the spin-valley locking [55, 56] provides a way of controlling the internal quantum numbers of a symmetry broken state. In this fashion, combining exchange fields with twisted correlated materials provides a unique knob to tailor the microscopic spin-sector of a many-body state.

Here we show that a correlated state controlled by exchange bias can be engineered in a van der Waals heterostructure made of magnetic two-dimensional materials and a twisted dichalcogenide bilayer. In particular, we demonstrate that a magnetic exchange bias can control the symmetry broken state of twisted dichalcogenide system, creating a transition from valley ferromagnetism to spin ferromagnetism. Our results put forward exchange bias as a powerful knob to control symmetry broken states in magnetically encapsulated twisted dichalcogenide materials, exemplifying the versatility of combining spintronics and twistronics. The manuscript is organized as follows. In section 2 we show the emergence of nearly flat bands controlled by SOC and exchange coupling in the magnetically encapsulated multilayer. In section 3 we show the selective switching between interaction-induced valley ferromagnetism and spin ferromagnetism by controlling the magnetization of the encapsulation. In section 4 we estimate the strength of the effective exchange fields by means of first-principles calculations for a specific CrBr$_2$/WSe$_2$ based structure. Finally, in section 3 we summarize our conclusions.

2. Controlling flat bands with exchange bias

We first address the emergence of flat bands in the magnetic van der Waals heterostructure, and in particular how spin-orbit and exchange effects the low energy flat bands. In the following, we consider an effective model for a transition metal dichalcogenide in a honeycomb lattice, that captures in an effective fashion the low energy valence and conduction bands. The physics that we explore is expected both in the valence and conduction bands of a twisted TMDC, provided the low energy states stem from the states located at the K-points of the original Brillouin zone. In this fashion, the physics of one electron per moiré unit cell (i.e. an electron in the conduction bands), will be equivalent to those of one hole per moiré unit cell (i.e. one missing electron in the valence band). For the sake of concreteness, in the following we will focus on the physics emerging in the conduction band of the model. We will consider a twisted van der Waals dichalcogenide, encapsulated between a ferromagnetic insulator, where we have integrated out the degrees of freedom of the ferromagnetic insulator. The total Hamiltonian is of the form

$$H = H_0 + H_{SOC} + H_J,$$

with $H_0$ the non-relativistic Hamiltonian, $H_{SOC}$ the SOC correction and $H_J = H_{AF}/H_{FE}$ the exchange proximity effect

$$H_0 = \sum_{\langle ij \rangle} \epsilon_{i,s} c_{i,s}^\dagger c_{j,s} + m \sum_{i,s} \partial^2_{\nu} \epsilon_{i,s} c_{i,s}^\dagger c_{i,s} + \sum_{i,j,s} t^{\pm}(\mathbf{r}_i, \mathbf{r}_j) c_{i,s}^\dagger c_{j,s},$$

where $t^{\pm}(\mathbf{r}_i, \mathbf{r}_j)$ parametrize the interlayer coupling [57–59], $\partial^2_{\nu} \epsilon_{i,s}$ is the sublattice Pauli matrix, $m$ denotes the onsite energy imbalance between the two sites of the honeycomb lattice, $\sigma^z_{i,s}$ is the spin Pauli matrix, $\nu_{ij} = \pm 1$ for clock-wise/anti-clockwise hopping [60], $\langle \rangle$ denotes sum over first neighbors in a layer.

Figure 1. Sketch of the van der Waals exchanged dichalcogenide multilayer (a) and sketch of the emergence of the moiré superlattice (b). Panel (c) shows a sketch of the typical band structure of TMDC when SOC effects are considered. Panel (d) shows the possible lifting of degeneracies that the magnetic encapsulation can create, which in the absence of exchange field accounts for a low energy band structure with four degrees of freedom. Panels (e) and (f) show the interaction-induced symmetry breaking of the low energy states. For antiferromagnetic alignment (e) spin symmetry broken state appears, in which one of the spin channels gets filled and the other one empty. In contrast, for ferromagnetic alignment (f) a valley symmetry broken state emerges, in which interactions create an spontaneous filling of one of the valleys, with the other one remaining empty.

$$H_{SOC} = \lambda_{SOC} \sum_{\langle ij \rangle, i',s'} \nu_{ij} \sigma^z_{i,s} c_{i,s}^\dagger c_{i',s'},$$

$$H_{AF} = J_{AF} \sum_{i,s,i',s'} \tau_{i,s}^z \sigma^z_{i,s} c_{i,s}^\dagger c_{i',s'},$$

$$H_{FE} = J_{FE} \sum_{i,s,i',s'} \sigma^z_{i,s} c_{i,s}^\dagger c_{i',s'},$$
Figure 2. Band structure of a twisted dichalcogenide with a twist angle of $1.8^\circ$ in the absence of exchange effects with zero SOC (a) and with finite SOC (b). Panels (c) and (d) show the bandstructure in the absence of SOC but with a finite exchange bias, having a ferromagnetic alignment between the layers in (c) and antiferromagnetic in (d). Panels (e) and (f) show the band structures with finite SOC and exchange fields, having a ferromagnetic alignment between the layers in (e) and antiferromagnetic in (f). In the cases (e,f), the low energy states are formed by two bands, yet with dramatically different orbital content as sketched in figure 1(d).

second neighbors in a layer, and $t_i = \pm 1$ for upper/lower layer. In particular, we take the following functional form for the interlayer coupling $t^{\perp}(\mathbf{r}_i, \mathbf{r}_j) = t^{\perp} \left( \frac{\mathbf{r}_i - \mathbf{r}_j}{r} \right) e^{-\gamma |(\mathbf{r}_i - \mathbf{r}_j)|}$, with $t^{\perp}$ the strength of the interlayer hopping, $d$ the distance between layers and $\gamma$ a parameter controlling the decay of the hopping between layers. We emphasize that our model is an effective model for the conduction band of the TMDC, and as a result $t_{\perp}, \gamma$ are effective parameters that are taken to reproduce flat bands obtained by first principles results [49]. The previous model applied for a single layer faithfully captures the electronic structure at the $K$ and $K'$ points [56, 61, 62], and therefore can be used as a starting point for flat bands stemming from the dichalcogenide valleys. We note that more sophisticated models for a dichalcogenide could be considered, yet for the sake of simplicity we here focus on the minimal one that captures the spin-valley physics at the valleys.

With the previous effective model, we now compute the electronic structure of the magnetically encapsulated dichalcogenide multilayer. In particular, it is interesting to look in detail at the degeneracy of the low energy manifold in different regimes. Let us start with the case $J_{AF} = J_{FE} = \lambda_{SOC} = 0$, the case in which both exchange and SOC effects are neglected. As shown in figure 2(a) the band structure for a structure with a twist angle $1.8^\circ$ shows nearly flat bands as expected from twisted dichalcogenide systems. It is important to note that such energy bands are four-fold degenerate, two times from valley, and two times due to spin. Such phenomenology is analogous to twisted graphene multilayers [21] or twisted boron nitride [30]. It is interesting to note that the flat band regime in twisted dichalcogenides appears for a range of small angles, in contrast with the fine tuning required for twisted graphene multilayers. Due to the existence of such degeneracy, it is expected that the addition of additional terms breaking that symmetry will create splittings between the different sectors. When we consider the effect of intrinsic SOC ($\lambda_{SOC} = 0.02t$), as shown in figure 2(b), a momentum-dependent spin splitting appears, yet the valley degeneracy remains along the shown k-path. As a result, the low energy manifold remains four-fold degenerate. It is interesting to note that the inclusion of SOC slightly increases the bandwidth of the low energy bands, yet still keeping an overall small bandwidth. Besides the splitting created in the low energy bands, additional splittings are created at higher energies.

Let us now focus on the cases hosting only exchange fields, and in the absence of SOC. The simplest case to consider is the ferromagnetic configuration ($J_{FE} = 0.02t$), in which the spin degeneracy is lifted figure 2(c). In this situation, a uniform shift between up and down channels take place, and the low energy bands become just two-fold degenerate due to valley degeneracy. A more interesting scenario happens for antiferromagnetic exchange bias ($J_{AF} = 0.05t$) (figure 2(d)) in which a small splitting appears in the bands, yet without fully lifting the degeneracy of the low energy manifold. It is especially interesting to note that the band structure with SOC (figure 2(b)) and antiferromagnetic exchange bias (figure 2(d)) both show a four-fold degeneracy of the low energy manifold, in contrast with the trivial lifting of degeneracy created by the ferromagnetic exchange field (figure 2(c)).

We now move on to the case when both exchange fields and SOC are non-zero. We start with the case of ferromagnetic exchange ($J_{FE} = 0.02t$) and non-zero SOC ($\lambda_{SOC} = 0.02t$), shown in figure 2(e). The low energy manifold is still two-fold degenerate stemming from the valley degree of freedom (both bands are superimposed in figure 2(e)), yet with a clear asymmetry between $-k$ and $+k$ due to the combination of SOC effect and exchange field. We emphasize that the red band of figure 2(e) consists actually on two bands, located one exactly on top of the other, that belong to different microscopic valleys. A more interesting scenario corresponds to the antiferromagnetic exchange case ($J_{AF} = 0.02t$) with non-zero SOC ($\lambda_{SOC} = 0.02t$) shown in figure 2(f). It is clearly observed that now the low energy manifold is two-fold degenerate, in contrast with figures 2(b) and (d) (see the splitting showed in the insets). The low energy degeneracy comes from a combination of time-reversal and mirror symmetries, and could be lifted with an electric field between the layers. It is worth to remark that in both cases, ferromagnetic and antiferromagnetic bias, the low energy manifold is two-fold degenerate, yet...
where $\mathcal{G}$ is the valley-spin Green’s function defined as

$$\mathcal{G}(\omega) = [\omega - H(\mathbf{k}) + i0^{+}]^{-1} \mathcal{V}_{c},$$

with $\mathcal{V}_{c}$ the valley operator [66, 68–71], $S_{c}$ the spin operator and $H(\mathbf{k})$ the Bloch Hamiltonian.

The valley-spin Chern number $\mathcal{C}$ can be computed as

$$\mathcal{C} = \int_{0}^{2\pi} d\phi \int d^{2}\mathbf{r} \Xi(\mathbf{r}, \omega),$$

and as result $\Xi(\mathbf{r}, \omega)$ is the energy resolved real-space valley-spin flux [65–68]. The valley operator $\mathcal{V}_{c}$ is defined in the tight binding basis as [66, 68–71]

$$\mathcal{V}_{c} = \lambda \sum_{\langle ij \rangle, s} \phi^{\dagger}_{i,j} c_{i,s} \phi_{j,s},$$

where $\nu_{ij} = \pm 1$ for clock-wise/anticlock-wise hopping. $\langle ij \rangle$ denotes sum over second neighbors in a layer, and $\phi^{\dagger}_{i,j}$ is the sublattice Pauli matrix. The previous operator takes the eigenvalues $\pm 1$ at the microscopic $K$ and $K'$, and as a result can be used to compute the valley flavor of an eigenstate. It is worth to note that this operator will be especially useful in the next section when computing valley symmetry breaking in the real tight binding model.

Focusing on the case with antiferromagnetic configuration, we observe that real space valley fluxes at the Fermi energy $\omega = \epsilon_{F}$ emerge in the unit cell (figure 3(b)), similarly to other van der Waals multilayers [66–68]. The combination of triangular arrangement (figure 3(a)) and real space valley-spin flux (figure 3(b)) suggest that the low energy band structure can be captured with an effective triangular lattice with spin-valley fluxes $H = \sum_{\langle ij \rangle, s,s'} \epsilon^{\sigma}_{ij} \phi^{\dagger}_{i,s} d_{j,s'} \phi_{j,s'}$, where $d_{j,s}$ are the creation/annihilation operators for the Wannier states, $\phi_{i}$ are the phases associated to the real space flux of the supercell $\phi_{i}$ (figure 3(d)). This low energy Wannier model would allow to study interaction effects without having to perform a calculation for the whole moiré unit cell, and similar models have been explored in a variety of twisted two dimensional materials [48, 67, 72]. The form of the effective interactions in these low energy models can be non-trivial, due to the potentially extended nature of the Wannier states, $\mathcal{E}_{ij}$, which is available in the whole modeling of the moiré unit cell, and second neighbor interactions.

3. Interaction-induced symmetry breaking in exchanged-bias flat bands

We now focus on the effect of interactions in the full real space model. For that purpose, we consider a generic interacting Hamiltonian with interactions up to second neighbors of the form

$$H_{I} = H_{I}^{(0)} + H_{I}^{(1)} + H_{I}^{(2)},$$

where $H_{I}^{(0)}$ is the local Hubbard interaction with strength $U$.

$$H_{I}^{(0)} = U \sum_{i} c_{i,\uparrow}^{\dagger} c_{i,\downarrow} c_{i,\downarrow} c_{i,\uparrow},$$

Figure 3. Local density of states of the lowest flat band, showing the emergence of localized states in a triangular lattice (a). Focusing now in the heterostructure with antiferromagnetic alignment of figure 2(f), panel (b) shows the real space spin flux at $\omega = \epsilon_{F}$, showing the emergence of non-trivial spin topology. Panel (c) shows the sketch of the different parts of the superlattice, and panel (d) a low energy model for the nearly flat bands.

with dramatically different quantum numbers. This feature will be important when considering the effect of electronic interactions.

Before moving on to interaction effects, it is interesting to understand the nature of the low energy states (figures 3(a)–(d)). The states associated to the two low energy flat bands form an emergent triangular lattice, as shown in the local density of states of (figure 3(a)). This is of course already suggested by the band structures of figure 2, that features the typical dispersion of a triangular lattice. Interestingly, the location of this emergent triangular superlattice corresponds to regions of the twisted system showing AB stacking (figure 3(c)), namely a region where one of the atoms fall in the hollow site, in strike contrast with the conventional AA localization in twisted graphene bilayers [57, 63, 64]. This is easily rationalized by taking into account that, whereas for graphene AB and BA regions are equivalent, in a twisted dichalcogenide bilayer AB corresponds to Mo-Mo stacking, whereas BA corresponds to S-S stacking, and therefore are inequivalent. As a result, a triangular arrangement of the states in the AB regions becomes possible.

The triangular arrangement between the states, together with the low energy dispersion, suggests that this system can be described with a low energy effective model. To gain insight into this, it is interesting to look at the valley spin flux [65–68] in the moiré unit cell defined as

$$\Xi(\mathbf{r}, \omega) = \int \frac{d^{2}\mathbf{k}}{(2\pi)^{2}} \epsilon^{\sigma}_{r} \langle \phi(\partial_{x}, \mathcal{G}^{-1}) \phi(\partial_{y}, \mathcal{G}) \rangle |\mathbf{r}|,$$
trodes, the spin or valley degeneracy is lifted. In the low energy figure 4 above, depending on the configuration of the magnetic ele-
magnetic configuration (figures 4(a), (b)), and with ferro-
magnetic configuration (figures 4(c), (d)). In the case of an antiferromagnetic alignment interactions give rise to a valley polarized ground state (c), with a net valley moment per unit cell (d).

$H_I^{(1)}$ is the first neighbor interaction with strength $V_1$

$$H_I^{(1)} = V_1 \sum_{\langle ij \rangle} \left[ \left( \sum_i c_{i,s}^\dagger c_{i,s} \right) \left( \sum_j c_{j,s}^\dagger c_{j,s} \right) \right].$$

(11)

$H_I^{(2)}$ is the second neighbor interaction with strength $V_2$

$$H_I^{(2)} = V_2 \sum_{\langle \langle ij \rangle \rangle} \left[ \left( \sum_i c_{i,s}^\dagger c_{i,s} \right) \left( \sum_j c_{j,s}^\dagger c_{j,s} \right) \right].$$

(12)

We note that by construction, the interaction Hamiltonian is SU(2) symmetric. We decouple the previous Hamiltonian with a conventional non-collinear mean-field approximation, which we solve selfconsistently. In the following we take $U = 2t$, $V_1 = 1.5t$ and $V_2 = 0.5t$, and we will focus on the case of a single electron per moiir unit cell, that turns the low energy flat bands half filled. We note that the same results are expected for a single hole per unit cell and, as it will be shown later, this would be a physically achievable regime for the WSe$_2$/CrBr$_3$ twisted multilayer.

We now consider the two different scenarios for the mag-
netic encapsulation, the heterostructure with with antiferro-
magnetic configuration (figures 4(a) and (b)), and with ferro-
magnetic configuration (figures 4(c) and (d)). As elaborated above, depending on the configuration of the magnetic electrodes, the spin or valley degeneracy is lifted. In the low energy manifold, which is now just two fold degenerate, interactions can give rise to an additional symmetry breaking.

We focus first on the case with antiferromagnetic config-
uration, whose low energy manifold has two bands, one per spin channel (figure 2(f)). Due to this spin degeneracy, correlation effects could be able to lift the degeneracy and open up a gap, as we explicitly show below. We now take the interaction terms introduced in equations (10)–(12) and solve the selfconsistent mean field equations. We obtain that, upon introducing inter-
actions, symmetry broken state emerges, hosting a net spin magnetic moment per supercell and lifting the degeneracy of the low energy band structure [75]. The mean field band struc-
ture shown in figure 4(a) shows that an spontaneous exchange splitting appears in the band structure, lifting the degeneracy of the low energy manifold introduced in figure 2(f). The projection in real space of the spin operator $S_z(r)$ shows that, associated with the lifting of degeneracy, a non-zero magnetic moment emerges, localized in the regions with AB stacking as shown in figure 4(b).

We now move on to the case in which the magnetic leads have a ferromagnetic alignment. In this situation, the low energy manifold host two bands, one per valley (figure 2(e)). Given that these two energy band belong to the same spin channel, a magnetic instability cannot lift their degeneracy, in stark contrast with the antiferromagnetic encapsulation. Upon introducing electronic interactions of equations (10)–(12) and solving the selfconsistent problem, we obtain a mean field Hamiltonian whose band-structure has an spontaneous interaction induced valley splitting (figure 4(c)). We can compute the expectation value of the valley operator in the unit cell $\mathcal{V}(r)$, and we observe that a non-zero valley polarization emerges in the regions with AB stacking as shown in figure 4(d). The real space expectation value of the valley operator is computed by taking the valley operator of equation (8) and adding a localized real space envelope to each site.

The previous calculations demonstrate, using a micro-
scopic interacting model, that the symmetry breaking induced by interactions is controlled by the magnetic alignment of the magnetic substrate. Importantly, the genuine combination of spin–orbit and exchange effects leads to the realization of pure-valley or pure-spin electronic instabilities in the flat bands of twisted dichalcogenides. Finally, it is worth to men-
tion that the previous discussion has focused in a low energy atomistic effective model. In the next section we show, using first principles methods, that a specific van der Waals material formed by WSe$_2$/CrBr$_3$ would be described with the effective model considered.

4. First principles calculations of the WSe$_2$/CrBr$_3$ exchange bias

We finally show via first principles calculations that prox-
imity effect between WSe$_2$ and CrBr$_3$ leads to an exchange splitting in the valence band of WSe$_2$. The emergence of such splitting can be understood from a second order pro-
cess in which an electron jumps from the semiconductor to the ferromagnet and back [76–82]. Since the emergence
of flat bands in twisted dichalcogenides have already been demonstrated by first-principles, here we will focus on the combined effect of exchange proximity effect and SOC. For this sake, we consider a minimal multilayer consisting on WSe$_2$ on top of CrBr$_3$, as shown in figure 5(a). In this situation, the exchange proximity effect created by CrBr$_3$ on WSe$_2$ will give rise to valley splitting in the low energy band-structure, as sketched in figure 5(b).

We performed our first principles calculations with the all-electron LAPW formalism as implemented in Elk [83]. Correlations in the Cr d-manifold where included with the DFT+U formalism in the Yukawa form [84], taking $U = 4$ eV. We first focus on the case without SOC the resulting band structure is shown in figure 5(c). The first important feature is that in the absence of SOC, the top of the valence band is located at $\Gamma$ instead of $K$, and as will be shown later this dramatically changes when SOC is included. Second, it is also worth to note that the conduction band states are formed by unoccupied Cr orbitals, instead of the WSe$_2$ states. This orbital arrangement as a function of energy is shown in figure 5(d). This indicates that for the current van der Waals heterostructure, correlated states in moiré flat bands should be searched in the hole-doped regime. Although it is well known that DFT underestimates the Mott gap in correlated magnets, our calculations performed with $U = 4$ eV for the Cr d-orbitals suggest that they might indeed be located inside the gap in real multilayer. Nevertheless, it must be noted that using a different ferromagnetic insulator such as EuS$_2$ may allow to explore the effect of exchange field in conduction electron flat bands.

We now move to consider the heterostructure computed with full non-collinear SOC, whose band structure is shown in figure 5(e). As anticipated before, in the presence of SOC the top of the valence band is located at $K$, in agreement with the low energy model used before. This feature highlights that first-principles calculations of flat bands in twisted WSe$_2$ can be strongly impacted by the presence of spin–orbit coupling, as it dramatically influences the nature of the states at the top of the valence band. In particular, we find that the top of the valence band is 60 meV below the top of the valence band at the $K/K'$ point. Second, it is observed that the combination of magnetism and SOC breaks the degeneracy between $k \rightarrow -k$, giving rise to different dispersion around the $K$ and $K'$ points (figure 5(e)). In the present case, we are particularly interested on its impact at the top of the valence band. Zooming the band structure on the top of the valence band as shown in figure 5(f), we clearly observe a splitting between the $K$ and $K'$ point, which is exactly associated to the combination of exchange proximity effect between CrBr$_3$ and WSe$_2$ and SOC [80, 85–87]. It is interesting to note that, even though the first principles methods fully capture the different orbital channels that contribute to the exchange splitting, tracing back the different contributions remains a non-trivial task [88, 89]. The exchange proximity effect is found on the order of 4 meV, similar to other van der Waals multilayers [80]. These first-principles results demonstrate both the validity of the low energy model used previously for the top of the valence band, and the possibility of breaking $K/K'$ via exchange proximity effect, the feature that we used to selectively lift the degeneracy of the flat bands.

We finally note that the previous first principles calculations do not account from additional effects appearing in the twisted system, such as atomic relaxations in the twisted WSe$_2$, rippling between the WSe$_2$ and CrBr$_3$, or atomic relaxations in CrBr$_3$ stemming from the moiré pattern between CrBr$_3$ and WSe$_2$. To capture those effects, first-principles calculations of a twisted CrBr$_3$/WSe$_2$/WSe$_2$/CrBr$_3$ multilayer should be performed, including non-collinear SOC and DFT+U. Nevertheless, given the large unit cells involved, this would represent a challenging system from the computational point of view.

5. Conclusions

We have shown that a van der Waals heterostructure consisting of twisted dichalcogenide bilayer (WSe$_2$) encapsulated between two-dimensional ferromagnets (CrBr$_3$) shows flat
bands and correlated states controlled by the magnetic encapsulation. By using an effective atomistic model that incorporates spin–orbit and exchange proximity effects, we showed that such moiré system shows flat bands whose internal orbital structure strongly depends on the magnetic encapsulation. Once we include electronic interactions, we demonstrated that depending on the magnetic arrangement, a spontaneous spin–orbit and exchange proximity effects, we showed that the proximity between CrBr$_3$ and WS$_{2}$ creates a spin splitting in the WSe$_2$, in agreement with the low energy model used. Our results put forward hybrid magnetic/dichalcogenide twisted multilayers as a versatile platform where spin-orbit and exchange effects drive controllable correlated states. Ultimately, our proposal brings together the fields of twistronics, spintronics and correlated physics, opening up new ways of engineering emergent moiré physics by means of exchange bias.

Acknowledgments

D S thanks financial support from EU through the MSCA project No. 796795 SOT-2DvdW. J. L. L acknowledges the computational resources provided by the Aalto Science-IT project. We thank Z Sun, T Wolf, G Blatter, O Zilberberg, F Guinea, B Amorim, M Sigrist, M Roesner and P Liljeroth for fruitful discussions. Part of this work was carried out on the Dutch national e-infrastructure with the support of SURF Cooperative.

ORCID iDs

D Soriano https://orcid.org/0000-0003-2358-526X
J L. Lado https://orcid.org/0000-0002-9916-1589

References

[1] Radisavljevic B, Radenovic A, Brivio J, Giacometti V and Kis A 2011 Nat. Nanotechnol. 6 147
[2] Splendiani A, Sun L, Zhang Y, Li T, Kim J, Chim C-Y, Galli G and Wang F 2010 Nano Lett. 10 1271
[3] Manzeli S, Ovchinnikov D, Pasquier O, Yazhev V and Kis A 2017 Nat. Rev. Mater. 2
[4] Mak K F, Lee C, Hong J, Shan J and Heinz T F 2010 Phys. Rev. Lett. 105 136805
[5] Dickinson R G and Pauling L 1923 J. Am. Chem. Soc. 45 1466
[6] Wilson J and Yoffe A 1969 Adv. Phys. 18 193
[7] Offidani M, Milletari M, Raimondi R and Ferreira A 2017 Phys. Rev. Lett. 119 196801
[8] Zeng H, Dai J, Yao W, Xiao D and Cui X 2012 Nano Lett. 12 1741
[9] Safeer C K et al 2019 Nano Lett. 19 1074
[10] Shao Q et al 2016 Nano Lett. 16 7514
[11] Mendes J B S, Aparecida-Ferreira A, Holanda J, Azevedo A and Rezende S M 2018 Appl. Phys. Lett. 112 242407
[12] Luo Y K, Xu J, Zhu T, Wu G, McCormick E J, Zhan W, Neupane M R and Kawakami R K 2017 Nano Lett. 17 3877
[13] Avsar A, Unuchek D, Liu J, Sanchez O L, Watanabe K, Taniguchi T, Özyilmaz B and Kis A 2017 ACS Nano 11 11678
[14] Huang B et al 2017 Nature 546 270
[15] Gibertini M, Koperski M, Morpurgo A F and Novoselov K S 2019 Nat. Nanotechnol. 14 408
[16] Samarth N 2017 Nature 546 216
[17] Gong C et al 2017 Nature 546 265
[18] Lin M-W et al 2016 J. Mater. Chem. C 4 315
[19] Zhao Du K, Zhi Wang X, Liu Y, Hu P, Utama M I B, Gan C K, Xiong Q and Kloc C 2015 ACS Nano 10 1738
[20] Kuo C-T et al 2016 Sci. Rep. 6
[21] Lopes dos Santos J M B, Peres N M R and Castro Neto A H 2012 Phys. Rev. B 86 155449
[22] Lopes dos Santos J M B, Peres N M R and Castro Neto A H 2007 Phys. Rev. Lett. 99 256802
[23] Wu F, Lovorn T, Tutuc E, Martin I and MacDonald A H 2019 Phys. Rev. Lett. 122 086402
[24] Naik M H and Jain M 2018 Phys. Rev. Lett. 121 266401
[25] Maity I, Naik M H, Maiti P K, Krishnamurthy H R and Jain M 2020 Phys. Rev. Res. 2 013335
[26] Liu C H et al 2015 Phys. Rev. B 91 165403
[27] Zhao Y, Yu W and Ouyang G 2017 J. Phys. D: Appl. Phys. 51 015111
[28] Jones A M et al 2013 Nat. Nanotechnol. 8 634
[29] Baugher B W H, Churchill H O H, Yang Y and Jarillo-Herrero P 2014 Nat. Nanotechnol. 9 262
[30] Zhao X-J, Yang Y, Zhang D-B and Wei S-H 2020 Phys. Rev. Lett. 124 086401
[31] Chen M, Lin X, Dinh T, Zheng Z, Shen J, Ma Q, Chen H, Jarillo-Herrero P and Dai S 2020 arXiv:2004.14588
[32] Hejazi K, Luo X-Z and Balents L 2020 Proc. Natl Acad. Sci. 117 10721
[33] Kormányos A, Zólyomi V, Drummond N D and Burkard G 2014 Phys. Rev. X 4 011034
[34] Mak K F, He K, Shan J and Heinz T F 2012 Nat. Nanotechnol. 7 494
[35] Cao T et al 2012 Nat. Commun. 3 887
[36] Sallen G et al 2012 Phys. Rev. B 86 081301
[37] Xu X, Yao W, Xiao D and Heinz T F 2014 Nat. Phys. 10 343
[38] Mak K F and Shan J 2016 Nat. Photon. 10 216
[39] Wang L et al 2019 arXiv:1910.12147
[40] Wu F, Lovorn T, Tutuc E and MacDonald A H 2018 Phys. Rev. Lett. 121 026402
[41] Naik M H, Kundu S, Maity I and Jain M 2019 arXiv:1908.10399
[42] Wang J et al 2020 arXiv:2001.03812
[43] Jin C et al 2019 Nature 567 76
[44] Regan E C et al 2020 Nature 579 359
[45] Shimazaki Y, Schwartz I, Watanabe K, Taniguchi T, Kroner M and Imamoglu A 2020 Nature 580 472
[46] Tang Y et al 2020 Nature 579 353
[47] Zhang Y, Yuan N F Q and Fu L 2019 arXiv:1910.14061
[48] Slager K and Fu L 2020 arXiv:2003.13690
[49] Zhan Z, Zhang Y, Yu G, Silva-Guillen F G J A and Yuan S 2020 arXiv:2005.13868
[50] Fleischmann M, Gupta R, Sharma S and Shallock M 2019 arXiv:1901.04679
[51] Zhang Z, Wang Y, Watanabe K, Taniguchi T, Ueno K, Tutuc E and LeRoy B J 2019 arXiv:1910.13068
[52] Zhao Y, Liao C and Ouyang G 2018 J. Phys. D: Appl. Phys. 51 185101
[53] Zhang Y, Zhan Z, Guinea F, Silva-Guillen J A and Yuan S 2019 arXiv:1910.13068
[54] Hill H M, Rigosi A F, Kim K T, Flynn G W and Heinz T F 2016 Nano Lett. 16 4831
[55] Zhu Z Y, Cheng Y C and Schwendenschlögl U 2011 Phys. Rev. B 84 155402
Xiao D, Liu G-B, Feng W, Xu X and Yao W 2012 Phys. Rev. Lett. 108 196802
Suárez Morell E, Correa J D, Vargas P, Pacheco M and Barticevic Z 2010 Phys. Rev. B 82 121407
Sboychakov A O, Rakhmanov A L, Rozhkov A V and Nori F 2015 Phys. Rev. B 92 075402
González-Arraga L A, Lado J L, Guinea F and San-Jose P 2017 Phys. Rev. Lett. 119 107201
Suárez Morell E, Correa J D, Vargas P, Pacheco M and Barticevic Z 2010 Phys. Rev. B 82 121407
Bistritzer R and MacDonald A H 2011 Proc. Natl Acad. Sci. 108 12233
Wolf T M R, Lado J L, Blatter G and Zilberberg O 2019 Phys. Rev. Lett. 123 096802
Lopez-Bezanilla A and Lado J L 2020 arXiv:2005.02169
Colomés E and Franz M 2018 Phys. Rev. Lett. 120 086603
Ramires A and Lado J L 2018 Phys. Rev. Lett. 121 146801
Ramires A and Lado J L 2019 Phys. Rev. B 99 245118
Xu C and Balents L 2018 Phys. Rev. Lett. 121 087001
Koshino M, Yuan N F Q, Koretsune T, Ochi M, Kuroki K and Fu L 2018 Phys. Rev. X 8 031087
Kang J and Vafek O 2018 Phys. Rev. X 8 031088
Braz J a E H, Amorim B and Castro E V 2018 Phys. Rev. B 98 161406
Klein D R et al 2018 Science 360 1218
Cardoso C, Soriano D, García-Martínez N A and Fernández-Rossier J 2018 Phys. Rev. Lett. 121 067701
Zhou J, Qiao J, Duan C-G, Bournel A, Wang K L and Zhao W 2019 ACS Appl. Mater. Interfaces 11 17647
Mashahi S et al 2019 Nano Lett. 19 4659
Zollner K, Faria Junior P E and Fabian J 2019 Phys. Rev. B 100 085128
Zhong D et al 2017 Sci. Adv. 3 e1603113
Zhong D et al 2020 Nat. Nanotechnol. 15 187
Source Forge 2020 The Elk Code http://elk.sourceforge.net/
Bultmark F, Cricchio F, Gránás O and Nordström L 2009 Phys. Rev. B 80 035121
Catarina G, Peres N M R and Fernández-Rossier J 2020 2D Mater. 7 025011
Ciorciaro L, Kroner M, Watanabe K, Taniguchi T and Imamoglu A 2020 Phys. Rev. Lett. 124 197401
Lyons T P et al 2020 arXiv:2004.04073
Soriano D, Cardoso C and Fernández-Rossier J 2019 Solid State Commun. 299 113662
Sivadas N, Okamoto S, Xu X, Fennie C J and Xiao D 2018 Nano Lett. 18 7658