Response to Report 2: Topological multiferroic order in twisted transition metal dichalcogenide bilayers

Mikael Haavisto,[1] J. L. Lado,[1] and Adolfo O. Fumega[1]

[1] *Department of Applied Physics, Aalto University, 02150 Espoo, Finland*

We thank the referee for his/her consideration for the publication of our work in SciPost Physics and his/her positive feedback on our manuscript. Please, find below the response to each of the issues addressed. The actions denote the corresponding modifications that we have implemented in the new version of the manuscript.

REPORT OF REFEREE 2

The manuscript analyzes different broken symmetry states in a narrow band in twisted MoS2. The results are interesting, and deserve publication in some form. In the following, some topics whose consideration will enhance the clarity and interest of the study are highlighted:

1. The manuscript addresses the physics of one among many, flat bands near the valence band edge of twisted MoS_2. The existence of other bands, and the fact that the study concerns only the valence band needs to be mentioned.

We thank the Referee for mentioning this point, we have added a note about it in our manuscript. The flat bands near the Fermi level or valence band edge will be the easiest to access experimentally via electronic gating, and hence this has been our subject of study.

Action: We have included a note for this point in the new version of the manuscript.

2. The manuscript uses a tight binding parametrization supposedly based on calculations of the Wannier functions for the band under consideration. Refs. 41, 46, and 47 do not carry out a full calculation of the Wannier functions associated to this band. These references only mention that the symmetries of the band are well described by effective s orbitals centered at the edges of the unit cell. The parametrization of the band may need more than nearest neighbor hopping elements. In addition, these references do not mention the vertical location of the Wannier functions. The value of the electric moment in the ferroelectric phase discussed in the manuscript depends on this unknown vertical shift. This point is also worth mentioning in the manuscript. Another unknown parameter is the extension of the Wannier functions, which is needed in order to estimate the strength of the interaction, see below

We thank the Referee for bringing those points, which we have now included in our manuscript. It is worth noting that in the nearly flat regime, the effective model is dominated by first neighbor hopping due to localization of the Wannier states in specific point of the moire unit cell of transition metal dichalcogenides. Second neighbor hopping can of course become sizable away from this flat band regime.

Regarding the vertical localization of the Wannier orbitals, the symmetry of the staggered honeycomb lattice guarantees that the emergence of a sublattice imbalance will produce a vertical electric polarization in the system. This relies on the vertical localization of the Wannier orbitals in each of the layers. In particular, in transition metal dichalcogenides, the Wannier states correspond to localized modes in each layer that appear due to local band bending due to the local stacking. This yields an upper bound for the vertical shift as the interlayer distance controlling the electric polarization of the system. Variations to this perfect localization will reduce the value of the ferroelectric polarization.

Action: We have addressed all these points in the new version of the manuscript. We thank the referee for bringing these issues.

3. The possibility of observing the phenomena described in the manuscript depends on the actual values of the magnetic moments and electric dipoles discussed in it. An order of magnitude estimate of these values will help the interested reader to understand the relevance of the results. Along the same lines, there is not discussion in the manuscript of the strength of the Rashba spin-orbit coupling.

The specific values for the electric polarization and magnetic moments will depend on a set of different parameters. For instance, we have already commented in the previous question that the vertical localization of the Wannier orbitals will determine the value of the electric polarization, that will depend at the same time on the values of the electronic interactions U and V as shown in Fig. 2d. However, we can establish upper bounds for the electric and magnetic moments. For the magnetic moment, strong onsite interactions fix a value of 0.5 μ_B per moiré unit cell. For the electric dipole, considering an interlayer distance of 4 Å and a perfect localization of the electron in one of the layers leads to an order of $\approx$1 Debye per moiré unit cell.

The value of the Rashba spin-orbit coupling will be determined by the transition metal and the ligand atoms. It is important to consider that the energy scales relevant in the twisted system are on the order of meV, both for the hopping and Rashba SOC. Therefore, values on that order of magnitude for the Rashba spin-orbit cou-

pling will be enough to obtain a non-trivial topological character as we can see from Fig. 4b.

Action: We have included these discussions in the new version of the manuscript.

4. Related to the previous point, the presence of nearby bands with different characteristics might influence the results, if the parameters U and V where comparable to the separation between bands.

We fully agree with the Referee on this point, and our results focus on the regime in which U, V are smaller than the separation between moire bands.

Action: We have highlighted this issue in the new version of the manuscript.

5. The manuscript studies only one filling, $\nu = 1$. It may happen that the case $\nu = 3$ is similar, because of an approximate electron-hole symmetry, although this is not discussed. I would be interesting an analysis of the other commensurate filling, $\nu = 2$.

We fully agree with the Referee that the case with $\nu = 3$ would be similar to the one that we have studied and it would also lead to a multiferroic order in the case of electron-hole symmetry.

While addressing the case $\nu = 2$ would possible as the Referee suggests, such limit would not be as beneficial for the emergence of multiferroic order. The half filled case has been widely studied, and its phase diagram is now well known[1, 2]. In particular, at half filling U promotes an antiferromagnetic state, whereas V promotes charge order. However, those two phases do not show coexistence in the phase diagram, and therefore the emergence of multiferroic order is not possible. In stark contrast, the quarter filling in the staggered honeycomb lattice offers a natural platform in which electronic interactions can lead simultaneously to the emergence of charge order and a magnetism. We refer the Referee to the discussion with Referee 1.

Action: We have added in the new version of the manuscript a more detailed explanation justifying the choice of the quarter-filling.

6. The discussion of the effects of the substrate is sketchy. The perturbation induced by the substrate is only relevant when the lattices are aligned. There is no description of the type of potential induced by the substrate, and its strength is also not addressed.

We note that, in the presence of the moire, due to the spatial modulation of the stacking in some regions of the unit cell the lattices will be locally aligned. It is in those regions where the substrate has its maximal effect as the Referee notes. For computational reasons, the supermoiré potential is chosen to be commensurate with the supercell of the original moiré cell (staggered honeycomb unit cell) and it takes the following functional form:

$$E_{SM}(\mathbf{r}) = \sum_i \cos\left(\frac{\mathbf{b}_i \cdot \mathbf{r}}{n}\right), \tag{1}$$

where $\mathbf{b}_i$ are the reciprocal lattice vectors of the moiré supercell (the summation runs over the 3 $\mathbf{b}_i$ vectors related by the C_3 symmetry). The product $\mathbf{b}_i \cdot \mathbf{r}$ equals 2π when $\mathbf{r}$ takes the value of the lattice vectors of the original moiré unit cell, and n is an integer that commensurates the supermoiré length L_{SM} with the original moiré length L_M as $L_{SM} = nL_M$. The function in eq. (1) allows to generate a modulated potential as the one shown in Fig. S1 for $L_{SM} = 5L_M$. The modulation caused by the supermoiré potential in the original staggered honeycomb lattice can be seen in Figs. 6bc.

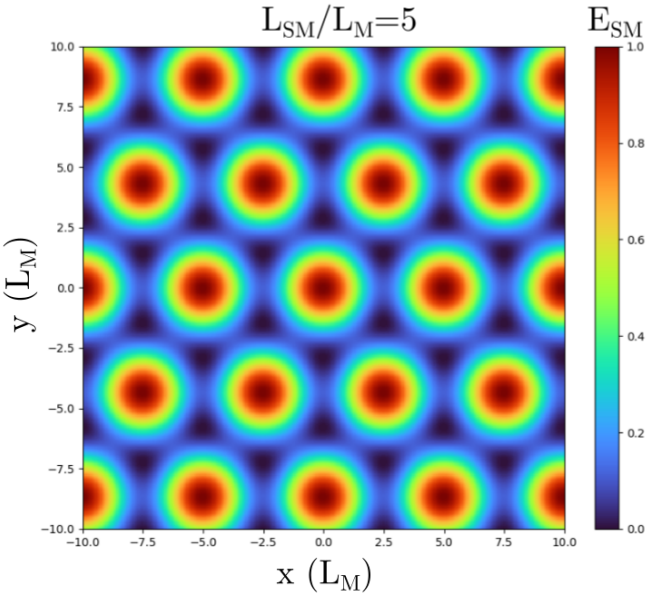

FIG. S1. Supermoiré potential generated with eq. (1) for $L_{SM} = 5L_M$. A periodicity of 5 times the original moiré supercell can be identified.

In our calculations the amplitude of the supermoiré potential E_{SM} is normalized to the range $E_{SM}/t = [0, 1.3]$.

Action: We have included the functional form of the supermoiré potential and the corresponding discussion.

We hope that, given our response to the different suggestions and the changes included in our manuscript, the Referee finds our manuscript suitable for publication in SciPost Physics.

[1] Igor F. Herbut, "Interactions and phase transitions on graphene's honeycomb lattice," Phys. Rev. Lett. **97**, 146401 (2006).

[2] Fakher F. Assaad and Igor F. Herbut, "Pinning the order: The nature of quantum criticality in the hubbard model on honeycomb lattice," Phys. Rev. X **3**, 031010 (2013).