Response to Report 1: Topological multiferroic order in twisted transition metal dichalcogenide bilayers

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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. In our revised version we have addressed all the comments and modified our manuscript accordingly. 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 1

The authors study topological multiferroic order in twisted transition dichalcogenides bilayers (TMDs) in the presence of spin-orbit interactions and applied electrical field. Starting from a model Hamiltonian that captures nearest-neighbor hopping between the AB/BA sites forming an effective honeycomb lattice, the authors analyze spin-/charge excitations (SDW/CDW) that emerge due to on-site and nearest-neighbor interactions within a self-consistent mean-field theory. They demonstrate that spin-orbit interactions render this model a topological multiferroic with non-vanishing Chern Number whose magneto-electrical coupling can further be tuned by the application of electrical fields. The topological character manifests in the existence of interface modes between either ferrimagnetic (FM), ferroelectric (FE) or multiferroic (FEM) domains. These findings are argued to exist in super-moire potentials arising from twisted TMDs encapsulated in substrates, e.g. hBN, with slightly different lattice constant. The authors demonstrate that the additional moiré potential leads to the formation of different topological regions of the multiferroic and hence naturally to the existence of topological interface modes.

The realization of topological states in twisted TMDs with strong intrinsic spin-orbit coupling represents an uprising field of research beyond the physics of twisted multilayer graphene and aligns with recent theoretical and experimental efforts. The discussion of topological multiferroic order is novel and adds to the toolbox of correlated topological phases that can be realized via moiré engineering. The connection to the super-moiré potential as possible candidate to host topological interface modes displays an interesting avenue, in particular because such effects have been observed experimentally in twisted trilayer graphene (TTG) to have major impact on correlated quantum states. The manuscript is soundly written and easy to follow for the reader. I therefore recommend publication as long as the following (minor) points are addressed properly:

Some technical aspects could be described in more detail to provide further clarification or to eventually allow for easier reproduction of the results, especially since there is no Appendix/Supplementary Material. Some aspects I came across are the following:

1. Concerning the TMD model Hamiltonian: The nearest-neighbor honeycomb model has been widely studied in the literature within, e.g. mean-field approximation, functional renormalization group studies, quantum Monte Carlo etc. Most of these studies focus to half-filling/filling in the vicinity to the van-Hove singularity. Could the authors clarify how quarter-filling in their manuscript is related to the position of the vHS? This would make it easier to compare with existing results.

As these materials show nearly flat bands, fillings from 0 to 1 fall in a flat band featuring a nearly divergent density of states. This applies both to the half-filled case mentioned by the referee and quarter filled case addressed in our manuscript. Even if the chemical potential does not fall in a van Hove singularity, the system is in the strongly interacting limit due to the reduced bandwidth of all the bands. The studies mentioned by the Referee often focus in half filled cases as these limits are the paradigmatic ones showing Mott insulating states. Furthermore, from the computational point of view, some many-body methods like quantum Monte Carlo may have severe sign problems away from half filling, and therefore for those methodologies the half filled case is more straightforward to tackle.

From a physical point of view, the quarter filling in the staggered honeycomb lattice offers a natural platform in which electronic interactions can lead simultaneously to the emergence of a charge order and a magnetization. Compared to the well studied half-filling case, in which electronic interactions lead to an antiferromagnetic insulator, the quarter-filling case allows the stabilization of charge order, thus leading to a net electric polarization in the staggered honeycomb lattice. The onsite Coulomb interaction U leads to a magnetic Stoner instability, in the half-filling case this leads to an equal magnetization in each of the sites since there are 2 electrons for 2 sites. For the realistic case in which first neighbor coulomb interactions V are smaller than onsite, $U > V$, a sublattice imbalance will not be promoted, since this is energetically unfavorable, i. e., an electron is already occupying

each site. In the quarter-filling case only 1 electron is available for the 2 sites, the onsite interaction will again promote a Stoner instability leading to magnetic order. However, in this case, for $U > V$, V will be able to promote a sublattice imbalance, since the sites are not fully occupied by an electron. Therefore, the quarter filling case allows for the simultaneous emergence of magnetic and electric orders.

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

2. The authors write: *The interacting model is solved using a self-consistent mean-field procedure including all the Wick contractions, including magnetic symmetry breaking, hopping renormalization, and charge order.* I would prefer to avoid the statement *all Wick contractions* as e.g. superconducting order is not accounted for as another possible Wick contraction or do the authors also account for this kind of instability, even though it may be irrelevant for purely repulsive interactions? Furthermore, what are the technical details of the mean-field analysis? How many moiré unit cells were taken into account (or what momentum resolution was used to sample the Brillouin zone) for the self-consistent procedure? Were the calculations performed in real- or momentum-space? Did the initial guess for the self-consistent procedure already include multiferroic order or was the initial state chosen randomly?

As the referee points out in the question, the Wick contractions that we have included correspond to those that allow magnetic symmetry breaking, hopping renormalization, and charge order. Anomalous terms related to the superconducting order are not included since we are dealing exclusively with repulsive interactions.

The self-consistent calculations were carried out in the unit cell of the staggered honeycomb lattice in momentum space with a well converged 10×10 k-mesh. An initial guess with finite ferroic orders was used in the calculations.

Action: We rephrase the sentence regarding the Wick contractions specifying that we do not include superconducting terms but just normal terms. We have also included a more detailed description of the calculations.

3. Concerning the results of the mean-field analysis: It is interesting that for $U/t < 4$ and up to $V/t < 2$ no charge-density wave is present, but the latter only emerges for $U/t > 4$ in the multiferroic phase. Is there an intuitive picture to understand why the formation of CDW order is diminished at $U/t = 0$?

The onsite interaction U plays also an important role in the stabilization of the CDW order. The relevance of U since stems from inducing a spin symmetry breaking, which in turn pins the chemical potential at the Dirac point at the majority spin channel. This pinning allows the first neighbor interaction V to drive a CDW state, as such a symmetry breaking allows opening up a gap at the Dirac point. Therefore, onsite interactions cooperate with V to drive CDW at quarter filling. At low values of U we have found in our analysis (but not shown, since $V > U$) that a CDW can also be formed for big values of V, i.e. $V > U$, but in this case the magnetic order does of course not emerge.

Action: we have added a comment in the new version of the manuscript addressing this point.

4. The authors write: *Since the origin of the interface modes is topological, small reconstructions at the interface will not impact the boundary modes.* For simple graphene, the edge termination (zig-zag/armchair) is crucial for the existence of edge states. Have the authors tried different edge terminations for the L/R domains and found invariant results?

This can be easily confirmed in Fig. 6d where the supermoiré potential induces regions with different topological invariant. In this case the boundaries have a different edge termination in each direction, but we can see that the circular boundary modes emerge without being affected by those different terminations. In the case of the device with two domains shown in Fig. 5, the Chern numbers would be unaffected by the termination. For topological states with a valley Chern number, if the interface is too sharp and there is strong intervalley scattering, a small gap could be opened driven by intervalley scattering.

Action: we have rephrased that sentence and add a discussion on the smoothness of the interface between different domains.

5. How is the momentum dependence of the interface spectral function $A(w, k_x)$ related to the sketch in Fig. 5 a) and what is the length of the domains in numerical simulations? From the sketch, I would intuitively expect that the system is periodic in y-direction and the interface separates the junction into the left/right domain along x. Then the momentum k_y would describe the bulk dependence. Maybe it would be helpful to add a small coordinate system in Fig. 5a) or to add a more detailed instruction why/how the spectral function $A(w, k_x)$ is computed.

The domains are semi-infinite domains in the y direction. The left one going from $y \to -\infty$ to $y = 0$ and the right domain from $y = 0$ to $y \to \infty$. The domain wall occurs at $y = 0$. In the x-direction the lattice is periodic, and hence this is the direction for which k_x describes the bulk dependence of the spectral function in the domain wall.

Action: We have clarified this issue in the new version of the manuscript and added a coordinate system to Fig. 5a accordingly to make the description of the analysis more intuitively.

6. Concerning the super-moiré potential in the last section: Can the authors give the functional form of the moiré potential and how it couples to the Wannier moiré orbitals? The potential looks to preserve the D_6h

symmetry of the original honeycomb lattice, though the LDOS in the multiferroic state only seems to preserve D_3h. Is that due to the inversion-breaking Rashba SOC term? Furthermore, how are the Chern Numbers calculated for different regions of the super-moiré? Was the set of parameters for the mean-field Hamiltonian (m, Δ_z, λ_R) the same as in the section before?

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 $L_{SM} = 5L_M$. We can see that the cosine functions of the supermoiré potential give rise to a 6 fold symmetry. The modulation caused by the supermoiré potential in the original staggered honeycomb lattice can be seen in Figs. 6bc.

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

The Chern numbers in the different regions are taken as the ones that would correspond in the uniform limit for the corresponding local values of the parameters. Therefore, they are not explicitly computed locally for the modulated system. This would be formally possibly using a Green's function formalism. Nonetheless, for big enough domains, as those considered in our manuscript, the local Chern number in the moire can be directly inferred from its value in the uniform case with the associated local Hamiltonian parameters.

Action: We have included the functional form of the supermoiré potential and the corresponding discussion, and added a note on how the Chern numbers are inferred.

I hope the authors appreciate the comments given above, which should not distract from the high quality of the manuscript.

We thank the Referee for their useful and positive feedback, and we hope that they find our manuscript suitable for publication in Scipost Physics.

Typos:

Action: We have corrected the typos pointed out by the referee and overall improved the readability of the paper.

We hope that, given our response to the different suggestions and the changes included in our manuscript, the

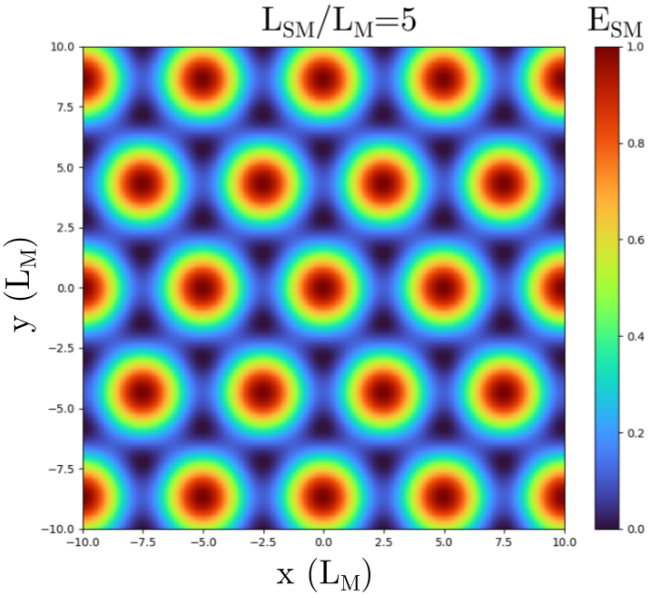

FIG. S1. Supermoiré potential generated with eq. (1) for $L_{SM} = 5L_M$. A periodicity of 5 times the original moiré supercell can be identified.

Referee finds our manuscript suitable for publication in Scipost Physics.