Response to Report 3: 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. 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 3

This paper presents a detailed numerical study of the magnetism of low angle twisted bilayer TMD, using a Wannier Hamiltonian. It is shown how a topological multiferroic order can emerge in this system at quarter-filling. The competitive effects of on-site and first-neighbor interaction terms are studied in detail. As a result, a multiferroic order displaying simultaneously ferroelectric and ferrimagnetic orders can be obtained. Spin-orbit interactions induce a topologically non-trivial multiferroic order, and topological excitations can be control by and external electric fields. The author have also studied the impact of an underlying substrate in the moiré system by considering super-moiré cell, and thus show the emergence of topological excitations. This theoretical work is very rich and brings advances in a complex and new subject the physics of the twisted bilayer semiconductor. It is well presented and I think it deserves to be published in SciPost Physics.

To reinforce the relevance of this work it seems important to me to discuss the following points in more detail:

1. Wannier Hamiltonian model is widely used in the literature to study the electronic properties of the low-energy states of twisted bilayer. However, its relevance to the study of magnetism is not obvious. The low energy eigenstates, that are well simulated by the Wannier Hamiltonian (without interaction), are complex and localized on a large number of atomic orbitals (mainly d-obitals of metal). When the interaction terms are on, the local magnetic order (at the atomic scale) can be complex. Is it well simulated by Wannier states? It would be interesting for the authors to discuss the consequences of their results for the magnetization of the d-orbitals of metal.

Our study analyzes the quarter-filling regime for generic non-magnetic twisted transition metal dichalcogenide bilayers. In this case, the twist angle between layers introduces a renormalization of the energy bands that can be effectively described by localized Wannier orbitals [1–3]. Importantly, and addressing the issue raised by the referee, our interactions are directly projected in those nearly flat bands. Of course, such electronic interactions stem originally from electronic repulsion of the d-orbitals of the transition metal dichalcogenide and generically long-range electrostatic repulsion.

Action: We have clarified this point in the new version of the manuscript.

2. If I am not mistaken, the xy plane magnetization is not include in the calculations. The authors should justify that point, especially for a calculation that is not at half-filling.

We thank the Referee for bringing this point. This stems from the fact that in slightly doped monolayer semiconductors spin-orbit coupling favors an out-of plane magnetic ordering, thus driving the magnetization in the z-direction[4]. We also note that in the absence of such anisotropy or in regimes in which is not dominant, potentially non-collinear magnetic textures could emerge in the system. While this would be a very interesting issue to address in the future, we believe that this point is beyond the scope of our manuscript.

Action: We have included this discussion in the new version of the manuscript.

3. The authors focused on the quarter-filling case. It would be nice to justify this choice a little more. Is it also possible to discuss, at least qualitatively, the cases of other fillings?

We thank the Referee for bringing this point, which we also addressed in the response to Referee 2, which we adapt here. The 3/4 filling would yield an analogous phenomenology due to the underlying electron-hole symmetry of our model.

The half filled case in the honeycomb lattice has been widely studied, and its phase diagram is now well known[5, 6]. 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 and comparing it to other fillings as explained above.

4. The SciPost latex style should be used to produce the manuscript.

We thank the Referee for the suggestion, we have now use the SciPost latex style.

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.

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