

Resubmission Letter

Dear Editor,

We thank the referee for reviewing our manuscript, and for recommending it for publication with minor revisions. The pertinent comments and suggestions have contributed to improve the quality of our paper. In what follows we address the points raised and list the changes made in the revised version of our article, which we believe is now ready for publication.

Color convention: (Blue) Questions of the referees - (Black) Our responses (Red) Changes in the manuscript.

Sincerely yours,

T. P. Cysne, on behalf of all authors.

REFEREE 1

1. *“The authors report a theoretical investigation of orbital currents in a Kekulé deformed graphene lattice, occurring when the couplings between carbon atoms are either enhanced or decreased (Figure 1a). How this particular situation is achieved in practice is not discussed in sufficient detail (doesn't the envisaged situation require a very careful fine-tuning?), and the model calculation has the risk of being "just a model" - not relevant to reality. (Later, the paper uses a value for the Kekulé parameter extracted from experiment, but the experiment is itself somewhat unclear.)”*

Response:

We thank the referee for addressing this issue and for encouraging us to contextualize our work within a more realistic scenario. The experimental reference [1] quoted in our manuscript reports a chiral symmetry-breaking gap opening induced by a type-O Kekulé ordering in graphene. In fact, the experiment utilizes two graphene layers intercalated with Li atoms. Nevertheless, the experimental gaped dispersion relation and the O-shaped pattern observed in the scanning tunneling microscope (STM) images near the band edges clearly establish the occurrence of Kekulé-O distortion. There are also theoretical proposals for experimental realization of the model used in our article in graphene monolayers with the Kekulé-O distortion. Realistic density functional theory (DFT) calculations show that the low-energy electronic spectra of the graphene/Bi₂Te₃ structure is well described by the Hamiltonian given by Eq. (4) of our manuscript [2]. **In the revised version of our article, we have included a few words about possible experimental realizations of type O Kekulé ordering in graphene, quoting Refs. [1, 2].** However, it is important to clarify that the experimental realization is not the specific point of our manuscript. It is focused on highlighting a situation where the valley currents are meaningless but there still is a transverse orbital current in the system that cannot be attributed to valleys.

2. *“But my main point is philosophical. The band structure shown in Fig. 1 c does not have two valleys because the energy minimum is shifted to the Gamma-point - hence it does not make any sense to speak about "valley currents", which are specific to the K and K' points in the undistorted lattice. Thus the authors are considering a situation where, by construction, the valley currents do not give any meaning. This does not remove the reason for doing the calculation, but it removes the foundations of all critical remarks made on using the valley currents as a vehicle of calculation. It also removes the hopes of resolving of some of the difficulties related to the interpretation/observation of valley currents.”*

Response: The main objective of this work is exactly to draw the community's attention to the fact that orbital angular momentum transport is not restricted to the valley contribution (valleytronics). This is exactly why we discuss the valleys that are present if the kekulé distortion goes to zero. The approach employed in our article is more comprehensive and includes the valleys' contributions when they are present. The main message of the article is that there is an transverse orbital current even in the absense of valleys.

3. *“In my view the authors are presenting a calculation (maybe a model calculation) in a situation where valley currents cannot be defined - and this should be clear in the submitted manuscript.”*

Response: This is correct and we have reiterated it in the revised version of our manuscript, following the referee suggestion.

REQUESTED CHANGES

1. “*remove "cristal" - it is crystal in English.*”

Response: We have corrected this typo.

2. “*Where does Eq.(11) come from - it is crucial for many subsequent developments. Either give a derivation, or a complete sequence of references.*”

Response: We have included references for Eq. (11).

3. “*I seem to recognize the expressions for the Berry curvature (e.g., Eq.(20)). Is the expression new, or a reincarnation of well-known results?*”

Response: No. This is not the Berry curvature Eq. (20) is just the result of Eq. (19), this is a new result which is the orbital Berry curvature for the electronic structure of the Kekulé- O model, and we have emphasized it in the revised version of our article. x

ADDITIONAL CHANGE

4. We had fixed a factor 2 in the plot of figure 3.

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- [1] C. Bao, H. Zhang, T. Zhang, X. Wu, L. Luo, S. Zhou, Q. Li, Y. Hou, W. Yao, L. Liu, et al., Phys. Rev. Lett. **126**, 206804 (2021), URL <https://link.aps.org/doi/10.1103/PhysRevLett.126.206804>.
- [2] Z. Lin, W. Qin, J. Zeng, W. Chen, P. Cui, J.-H. Cho, Z. Qiao, and Z. Zhang, Nano Letters **17**, 4013–4018 (2017), ISSN 1530-6992, URL <http://dx.doi.org/10.1021/acs.nanolett.6b05354>.