Influence of atomic relaxations on the moiré flat band wavefunctions in antiparallel twisted bilayer WS_2

L. Molino[1], L. Aggarwal[1], I. Maity[2,3], R. Plumadore[1], J. Lichner[2,3] and A. Luican-Mayer[1]

[1]*Department of Physics, University of Ottawa, Ottawa, Ontario K1N 6N5, Canada*
[2]*Department of Materials, Imperial College London, London, SW7 2BP, UK*
[3]*Thomas Young Centre for Theory and Simulation of Materials, London, SW7 2BP, UK*

lmoli076@uOttawa.ca

Twisting bilayers of transition metal dichalcogenides (TMDs) gives rise to a periodic moiré potential resulting in flat electronic bands with localized wavefunctions and enhanced correlation effects. In this work, scanning tunneling microscopy is used to image bilayer WS2 marginally twisted off of antiparallel alignment. Room temperature scanning tunneling spectroscopy reveals the presence of localized electronic states in the vicinity of the valence band onset. However, the experimentally observed electronic structure was found not to agree with first principles density-functional theory calculations, in particular differing on the real-space location of the valence band onset wavefunctions, which are predicted to correspond to a flat band. Agreement with theory is recovered when the calculations are carried out on bilayers in which the atomic displacements from the unrelaxed positions have been reduced, reflecting the influence of the substrate and finite temperature. This demonstrates the delicate interplay of atomic relaxations and the electronic structure of twisted bilayer materials.

*AL-M, LM, RP, LA acknowledge funding from NSERC Discovery RGPIN-2022-05215, NRC Quantum Sensing Challenge, Ontario Early Researcher Award ER-16-218, NSERC AllianceQuantum Consortium ALLRP/ 578466-2022. This project received funding from the European Union's Horizon 2020 research and innovation program under the Marie Sklodowska-Curie Grant agreement No. 101028468. This work used the ARCHER2 UK National Supercomputing Service (https://www.archer2.ac.uk) via our membership of the UK's HECMaterials Chemistry Consortium, which is funded by EPSRC (EP/R029431).

.

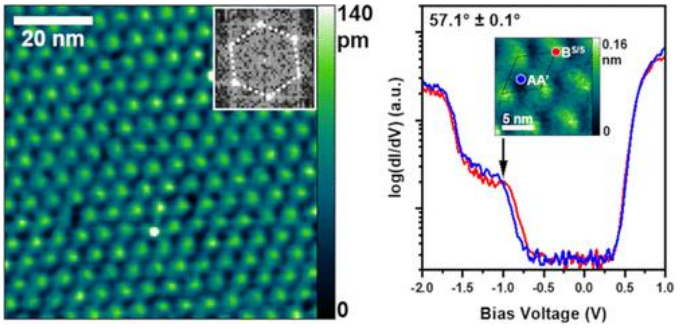

Fig.1. Scanning tunneling miscopy topography image of moiré pattern, scanning tunneling spectroscopy at high symmetry sites