MIP: PARADIM at Cornell University, DMR-2039380

In-House Research - 2025

Understanding Misfits—Theory and Measurement unravel 50+ Years of Mystery

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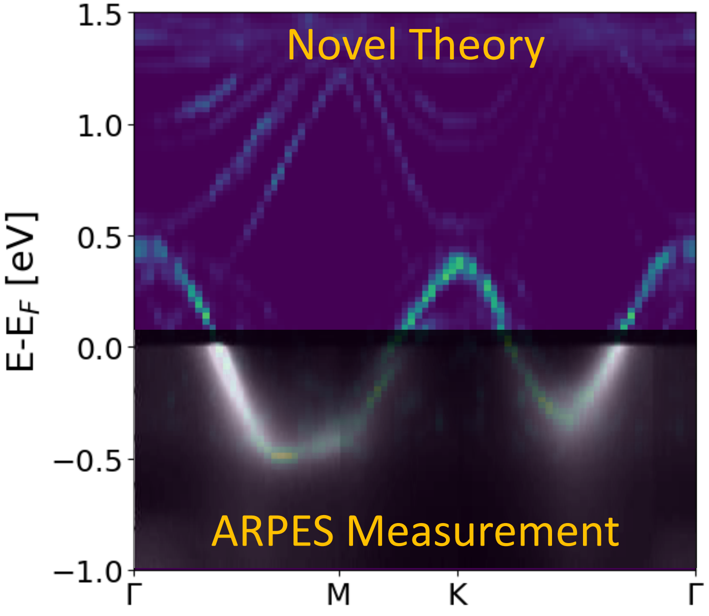
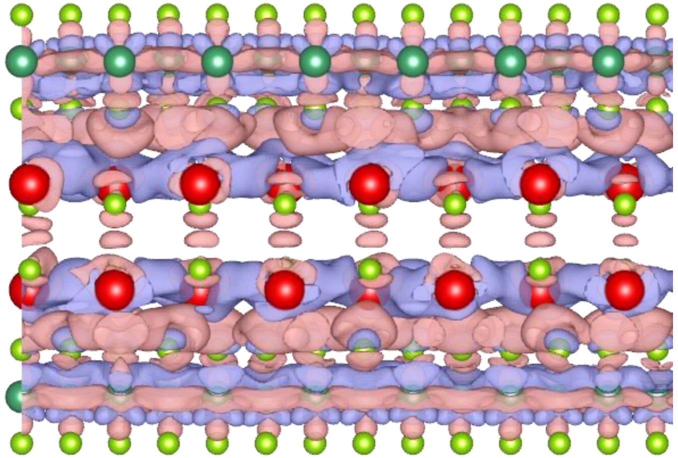
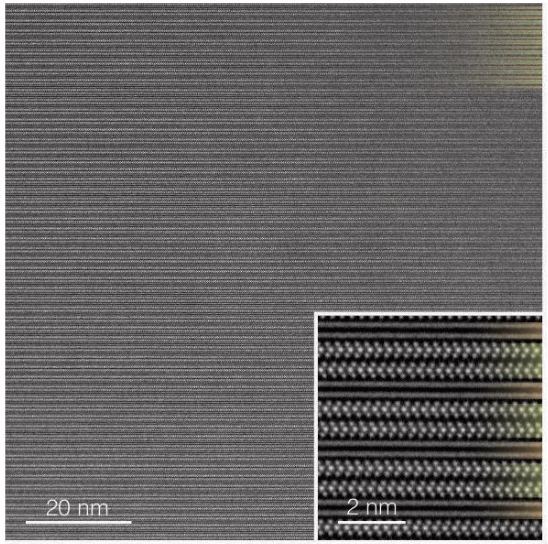
Misfit chalcogenides offer the opportunity of combining magnetism and superconductivity. The structure of the misfits consists of transition metal dichalcogenides (TMDs) stacked with alternating layers of a rare-earth rock salt. The TMDs can host many interesting electronic phases which can be heavily doped by the neighboring rock salt layers. Despite studies since the 1970, the extend of the charge transfer remained poorly understood.

Here, PARADIM's In-House Research Team applied their newly developed *ab initio* theoretical framework for mismatched interfaces (MINT) that enables accurate calculations of the electronic and vibrational properties of mismatched layered quantum materials. Applied to the misfit rock-salt/TMD heterostructures, the team shows that the large effective doping in the TMD layers arises from electron rearrangement within each layer, and not from net interlayer charge transfer, as previously assumed. ARPES measurements demonstrate mutual consistency of theory and experiment on PARADIM-grown high-quality bulk single crystals. The generality of our new method and insight into the nature of effective doping guide both theory and experimental efforts to create layered quantum materials with optimal properties for novel electronics and beyond.

D. Niedzielski, *et al.* *Phys. Rev. Lett.* **135**, 206202 (2025).
MINT Code and Tutorial DOI: 10.34863/seqm-4p70







(Top left) Overlay of measured ARPES data with new theoretical predictions, showing excellent agreement. **(Top right)** Electron density change upon interaction between LaSe and $NbSe_2$ layers showing the formation of interlayer covalent bonds and intralayer electron re-arrangement rather than the previously assumed net-interlayer charge transfer. Regions colored red (blue) indicate gain (loss) of electrons.
(Bottom right) Scanning transmission electron microscopy reveals consistent layered structure across length scales. Orange and green stripes indicate LaSe and $NbSe_2$ layers.