

Lithium Intercalation Dynamics in Twisted MoS₂

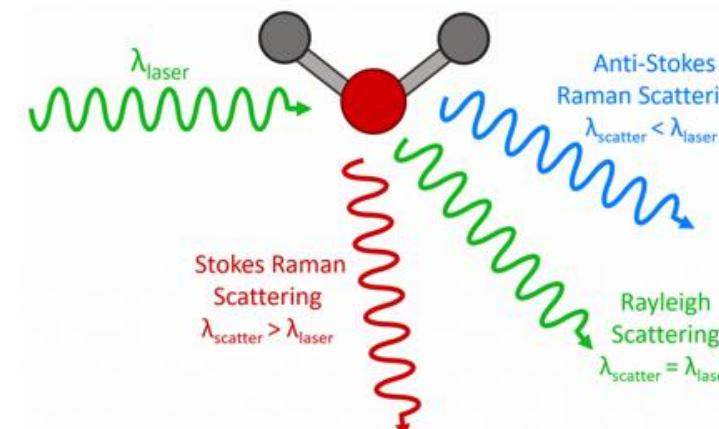
Ryan Cheng¹, Salman Kahn^{1 2}, Alex Zettl^{1 2}, Michael Crommie^{1 2}



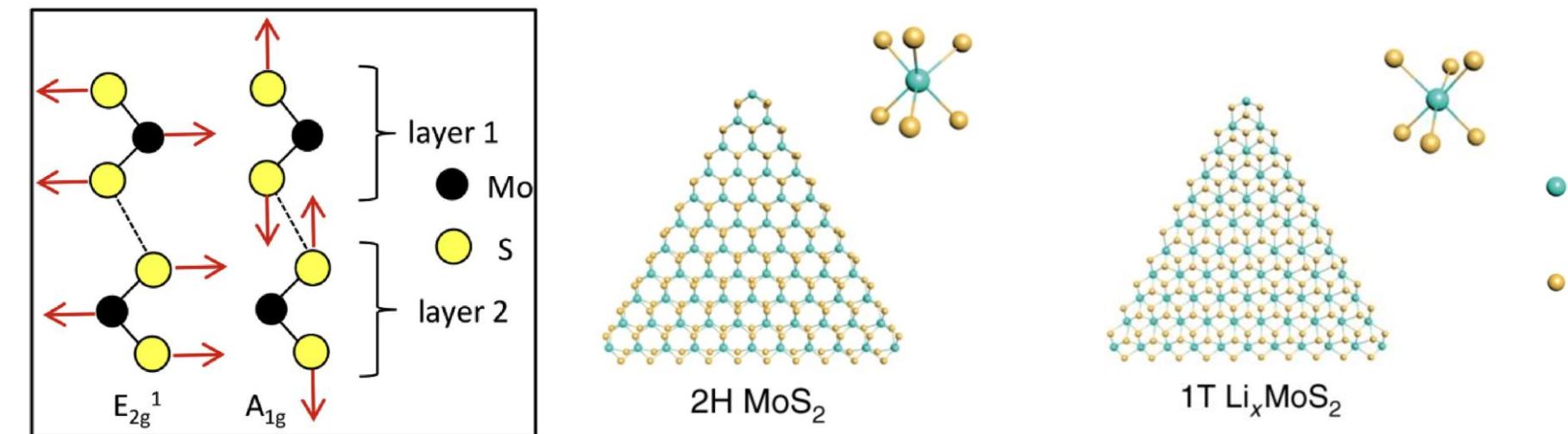
¹UC Berkeley, ²Berkeley Lab

Background & Theory

Raman Spectroscopy measures the differences in the wavelength of reflected photons due to inelastic scattering. This typically happens due to interactions with phonons within the material and gives us insight for the structure of the material.

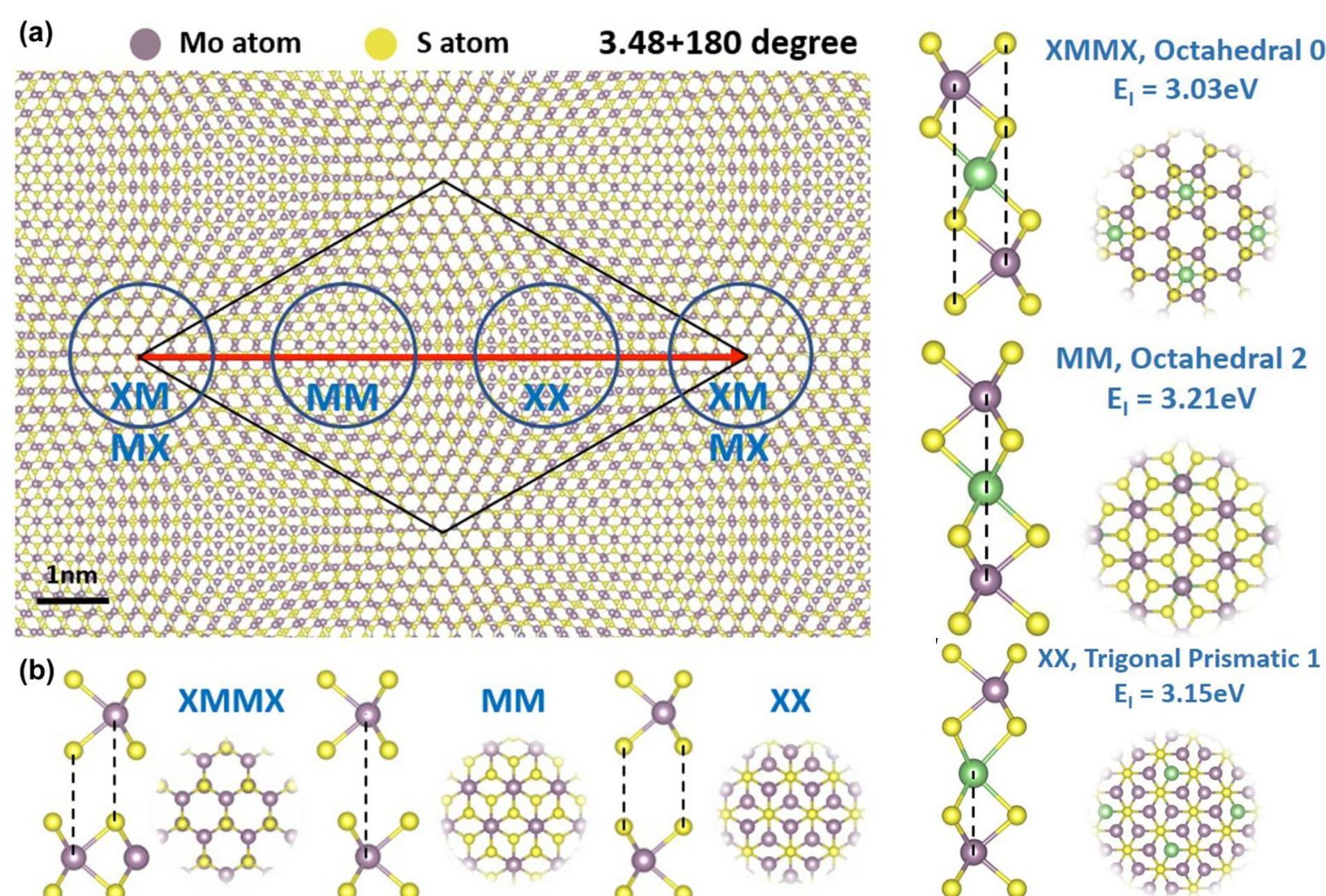


Lithium intercalation of MoS₂ for untwisted bilayers alters the crystal structure and results in a phase transition from the semiconducting 2H phase to the metallic 1T or 1T' phases.



Lithium intercalation within twisted MoS₂ is predicted to amplify interlayer interactions and enhance the moiré potential, leading to better flatband isolation.

Having a small twist angle between monolayers generates a moiré pattern with characteristic length scale much greater than the lattice constant, leading to various correlated phenomena such as flat bands and enhanced electron-phonon coupling.

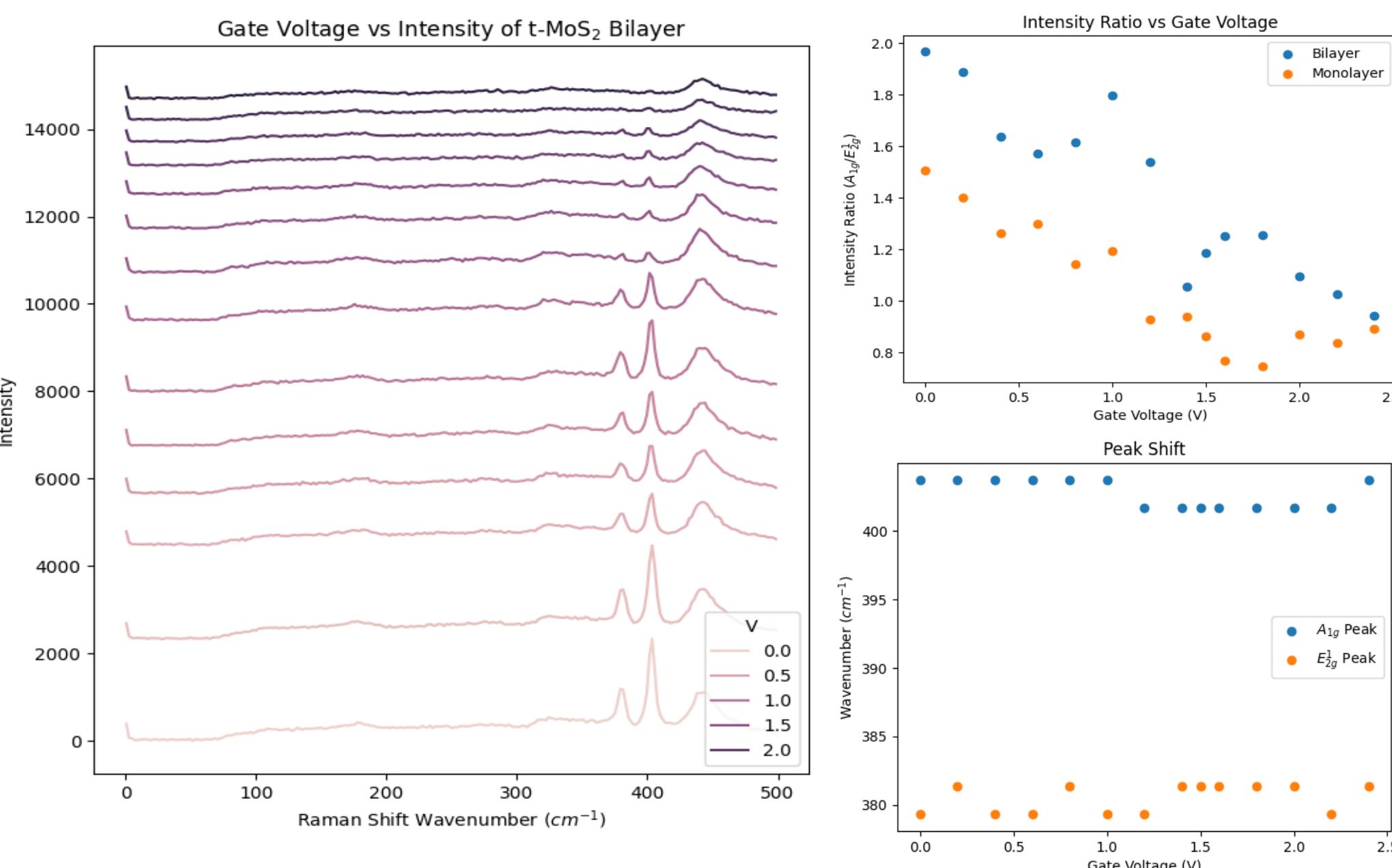


Raman Spectra

Overall decrease of A1g and E2g peaks are indicative of a continuous shift towards the 1T/1T' phase.

No new peaks indicative of spatially dependent interlayer coupling have been observed, however.

The intensity ratio between the A1g and E2g peaks decrease over gate voltage, lithium tends to intercalate within layers rather than (A1g peak intensity goes down)

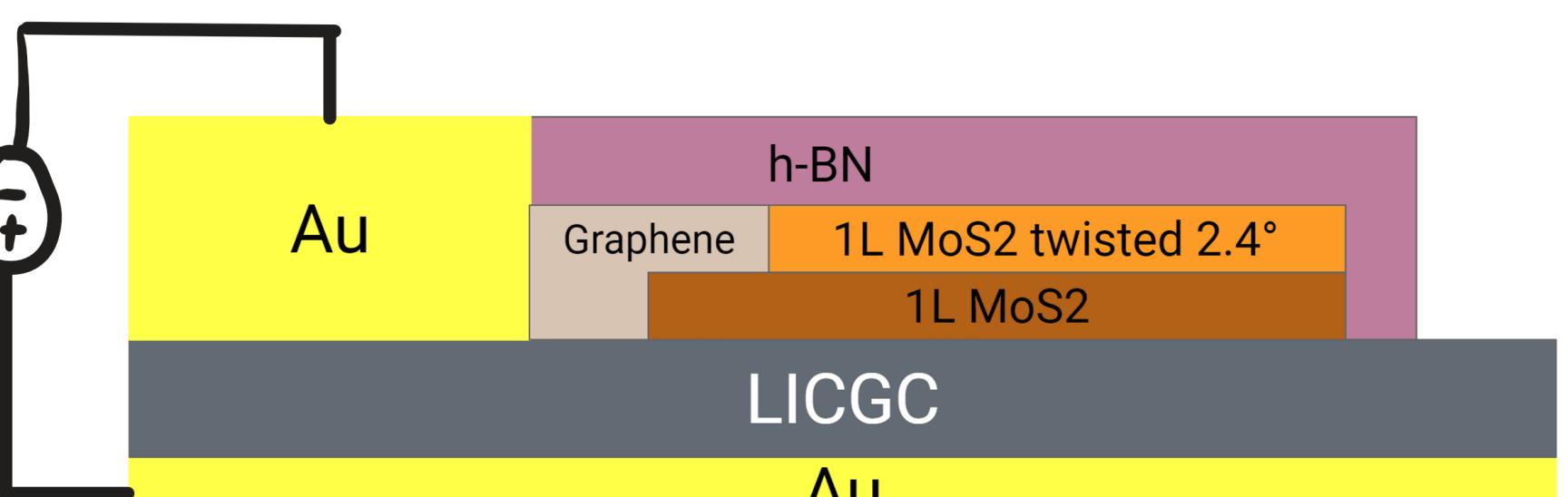


Device Fabrication

Gold contacts evaporated onto Lithium-Ion Conducting Glass Ceramic (LICGC)

MoS₂, h-BN, and graphene monolayers mechanically exfoliated and transferred via PDMS to be in contact between LICGC and gold contact.

Wire bonds are placed on gold contacts.



Measurement Procedure & Observations

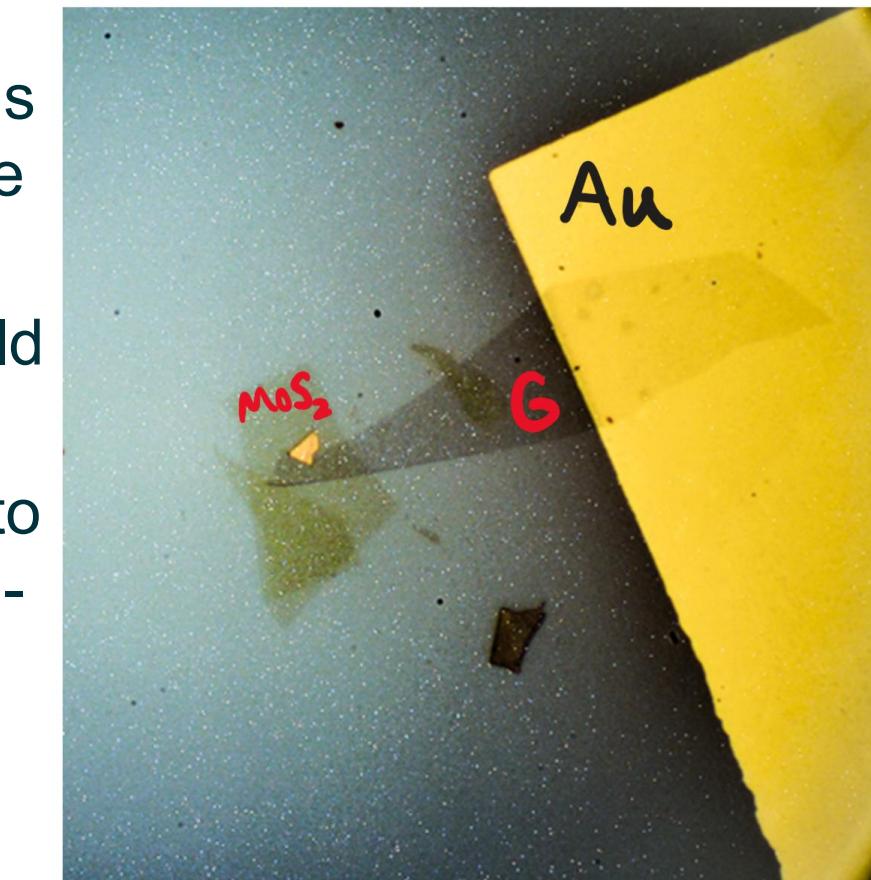
Measurements taken at room temperature with varying gate voltage in situ of an inVia confocal Raman microscope

Raman data taken at different xy positions around bilayer regions per gate voltage after 15 minutes of intercalation.

Graphene used as a contact between gold and bilayer to prevent contact erosion

Intercalation at high gate voltages leads to chemical reactions with atmosphere, h-BN is used to encapsulate the system and prevent this.

Reversing the bias seems to reverse intercalation, however the device does not seem to respond to further intercalation afterwards.



Conclusion & Future Directions

We confirmed and observed a continuous shift of the t-MoS₂ from the 2H phase towards the 1T/1T' phase.

We have not been able to observe new Raman peaks associated with interlayer coupling, however.

We will also take 4-terminal resistance and AFM measurements to confirm and corroborate observations.

References

- Lu, Z., Carr, S., Larson, D. et al. Lithium intercalation in MoS₂ bilayers and implications for moiré flat bands. *Phys. Rev. B* **102**, 125424 (2020). <https://doi.org/10.1103/PhysRevB.102.125424>
- Wan, J., Hao, Y., Shi, Y. et al. Ultra-thin solid electrolyte interphase evolution and wrinkling processes in molybdenum disulfide-based lithium-ion batteries. *Nat Commun* **10**, 3265 (2019). <https://doi.org/10.1038/s41467-019-11197-7>
- Zou, J., Li, F., Bissett, M. et al. Intercalation behaviour of Li and Na into 3-layer and multilayer MoS₂ flakes. *Electrochimica Acta* **331**, 135284 (2020). <https://doi.org/10.1016/j.electacta.2019.135284>
- Wu, Y., Wang, J., Li, Y. et al. Observation of an intermediate state during lithium intercalation of twisted bilayer MoS₂. *Nat Commun* **13**, 3008 (2022). <https://doi.org/10.1038/s41467-022-30516-z>