SAND2022-6215C

Sandia National Laboratories

Exceptional service in the national interest

Engineering of Nanoscale Heterogenous Transition Metal Dichalcogenide-Au Interfaces

Alex Boehm (amboehm@sandia.gov)
Sandia National Laboratories, Albuquerque, NM 87185

In collaboration with:
T. Ohta,[1] K. Thuermer,[2] J.D Sugar,[2] J. J. Fonseca,[3] J. T. Robinson[3]
1 Sandia National Laboratories, Albuquerque, NM 87185
2 Sandia National Laboratories, Livermore, CA 94550
3 U.S. Naval Research Laboratory, Washington, DC 20375

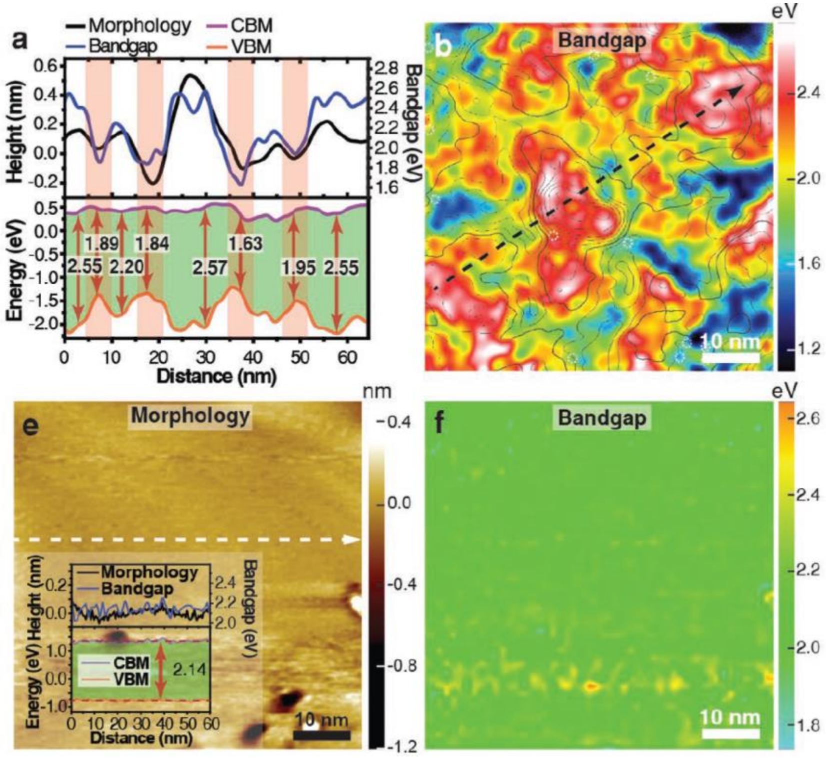
2022 MRS Spring Meeting: May 2022
Session NM01.20: Physical Properties of 2D Materials
Presentation Time: 9:00 AM to 9:15 AM
Session Location: Hawai'i Convention Center, Level 3, 311

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia LLC, a wholly owned subsidiary of Honeywell International Inc. for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

2D materials are susceptible to extrinsic factors

- 2D materials are attractive candidates for electronic and optoelectronic devices
- Reported sensitivity to extrinsic factors such as substrate interactions, mechanical strain, and charge transfer

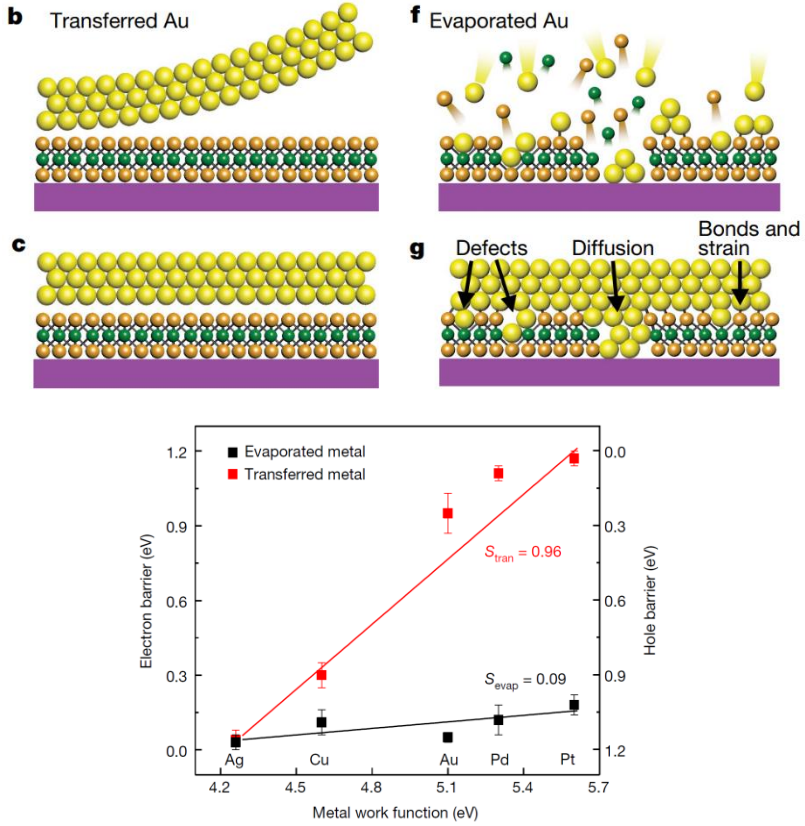


SiO2

HOPG

Shin, et al., Advanced Materials 28, 2016, 9378

- Interfaces with metallic contacts materials are crucial components of electronic and optoelectronic devices



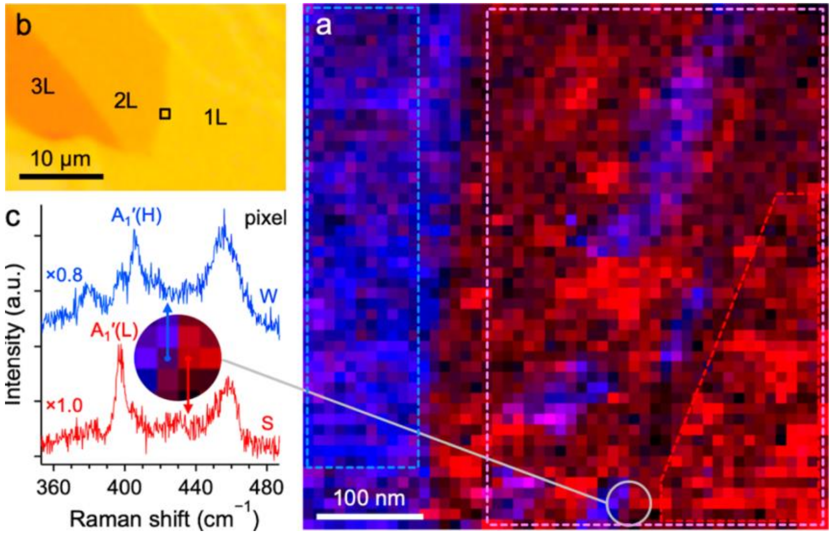
Liu, et al., Nature 557, 696 (2018)

Spatial nonuniformities in exfoliated MoS_2-Au interfaces

- One challenge toward realizing controlled TMD-metal interfaces are the spatial nonuniformities recently reported at these interfaces

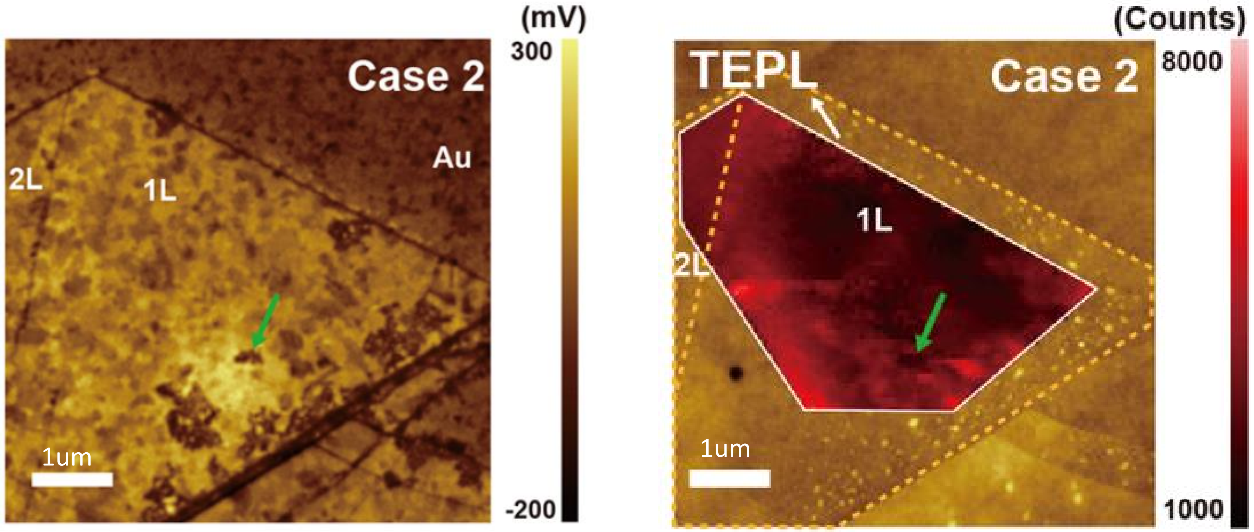
<u>MoS_2 exfoliated on Au</u>

Velickyì et al., J. Phys. Chem. Lett. 2020, 11, 6112



- Downshifted vibrational fingerprint revealed in Raman spectroscopy

Jo, et al., ACS Nano 2021, 15, 5618



- KPFM and TEPL identify regions with low contact potential and better electrical contact

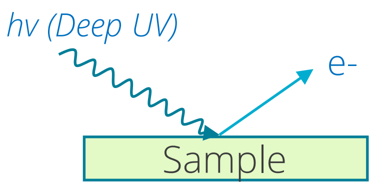
e^- doping of MoS_2

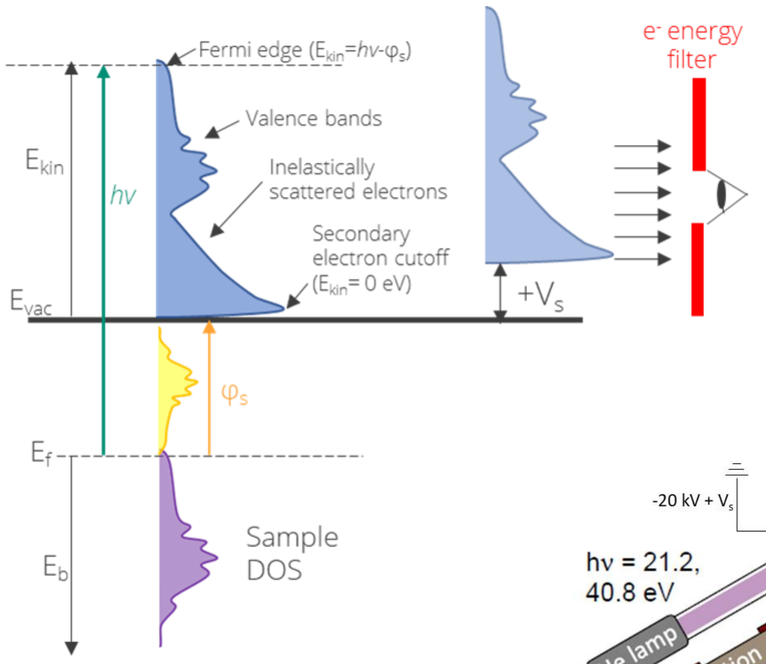
h^+ doping of MoS_2

Direct probe of electronic structure with high spatial resolution

Introduction: Photoemission electron microscopy and TMD-Au fabrication

- Photoemission electron microscopy (PEEM):
  A type of electron microscopy coupled with UV- or X-ray illumination to image nanoscale variations in photoelectron intensity





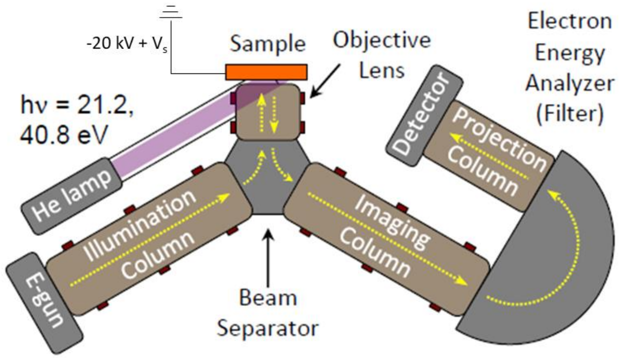
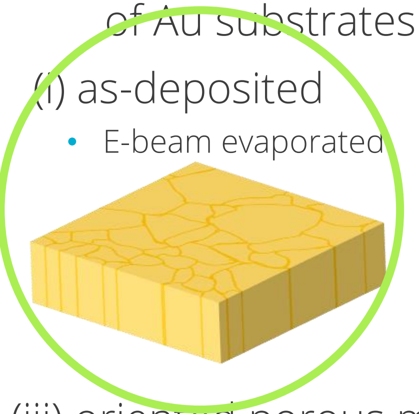


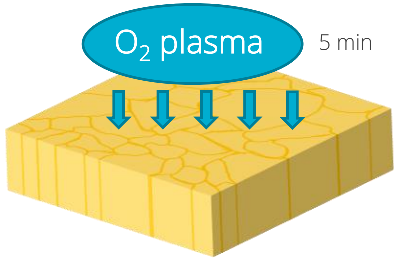
Figure courtesy: M. Berg

- We study TMD-Au interfaces fabricated through mechanical exfoliation with three different types of Au substrates

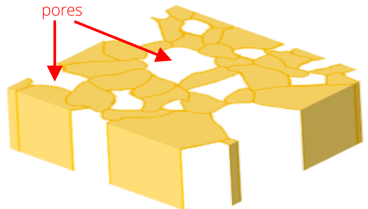
(i) as-deposited
- E-beam evaporated



(ii) plasma-treated

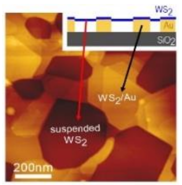


(iii) oriented porous metallic network (OPEN) Au

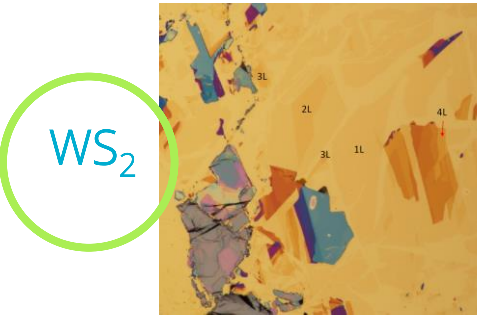


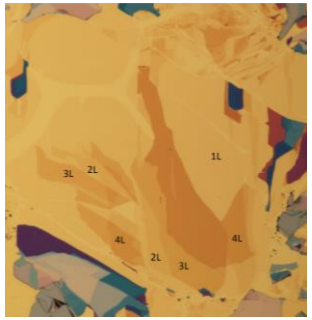
Fonseca et al., Nat. Commun., 2020, 11, 5

- Pseudo-epitaxial interface
- Resulting from the re-flow & recrystallization of Au by annealing



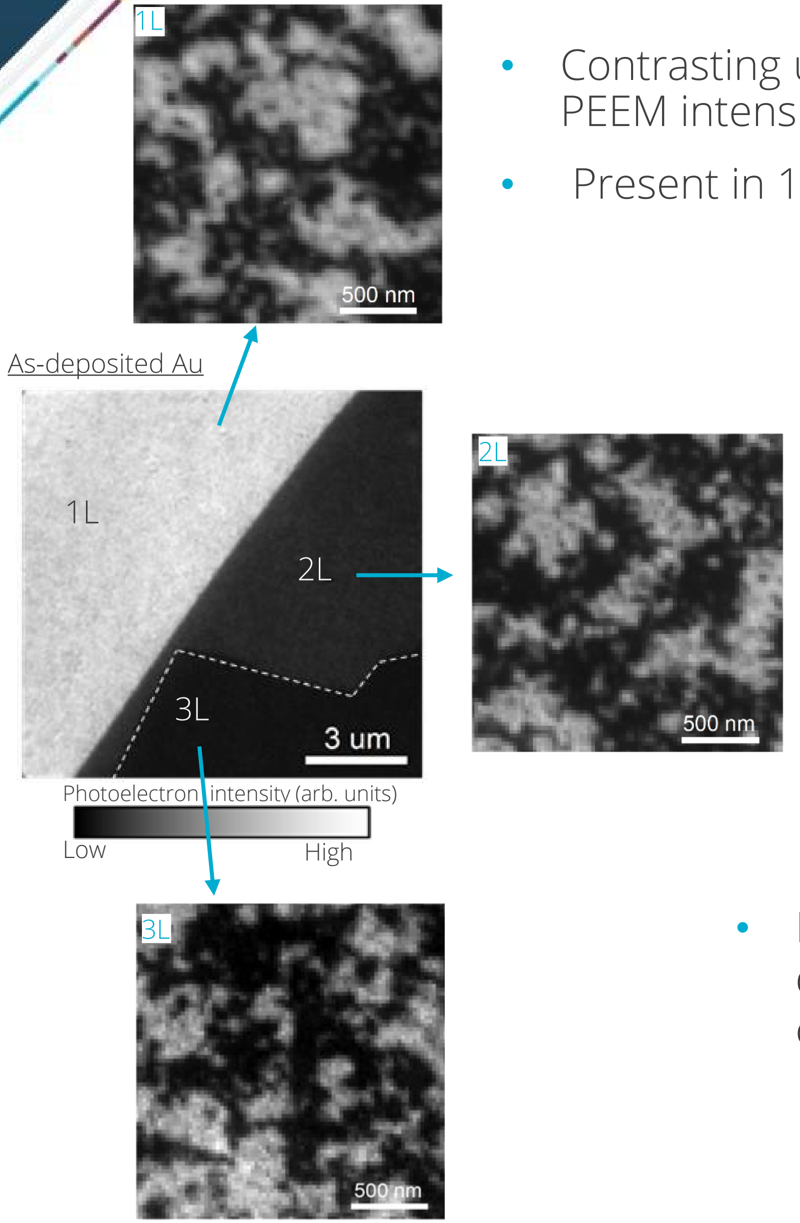
Thomas et al., ACS Nano. 2021, 15, 18060



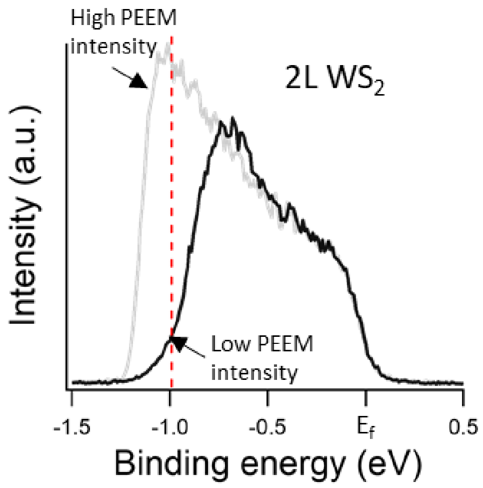


WSe_2

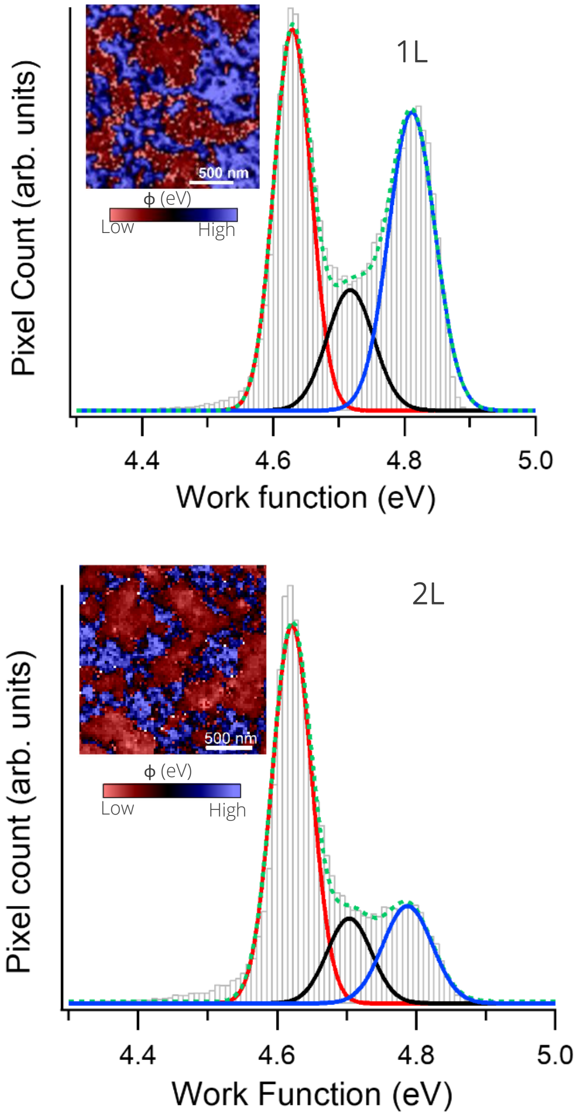
Micron-scale work function heterogeneity in as-deposited Au-WS_2 interfaces



- Contrasting um sized domains in PEEM intensity
- Present in 1-3L WS_2 thickness

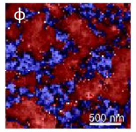


- PEEM contrasts arise from difference in secondary electron cutoff i.e. work function

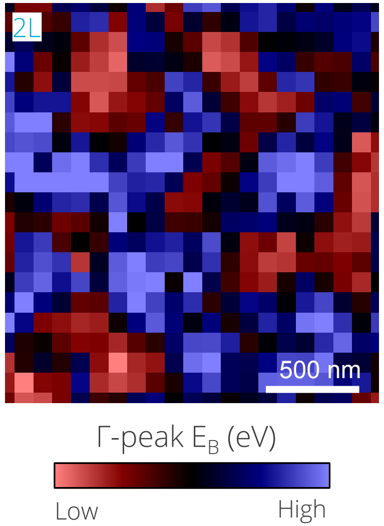


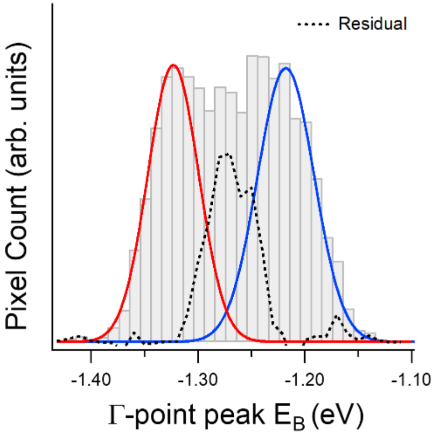
- Large work function variation (>200 meV)
- Indicates that the carrier density varies in the WS_2 flakes

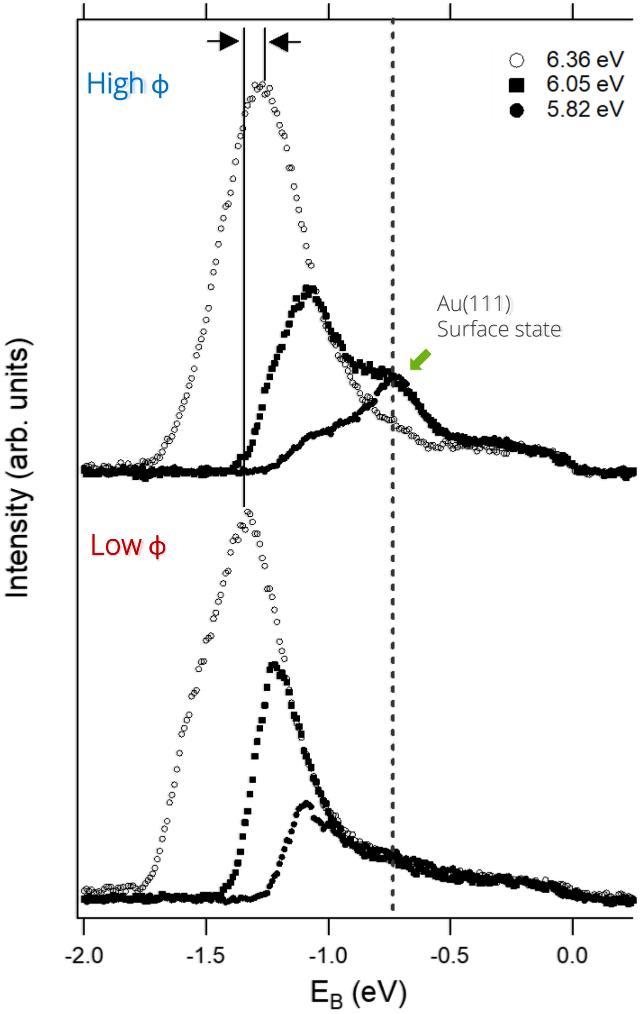
Micron-scale Γ-point peak position heterogeneity in as-deposited Au-WS_2 interfaces

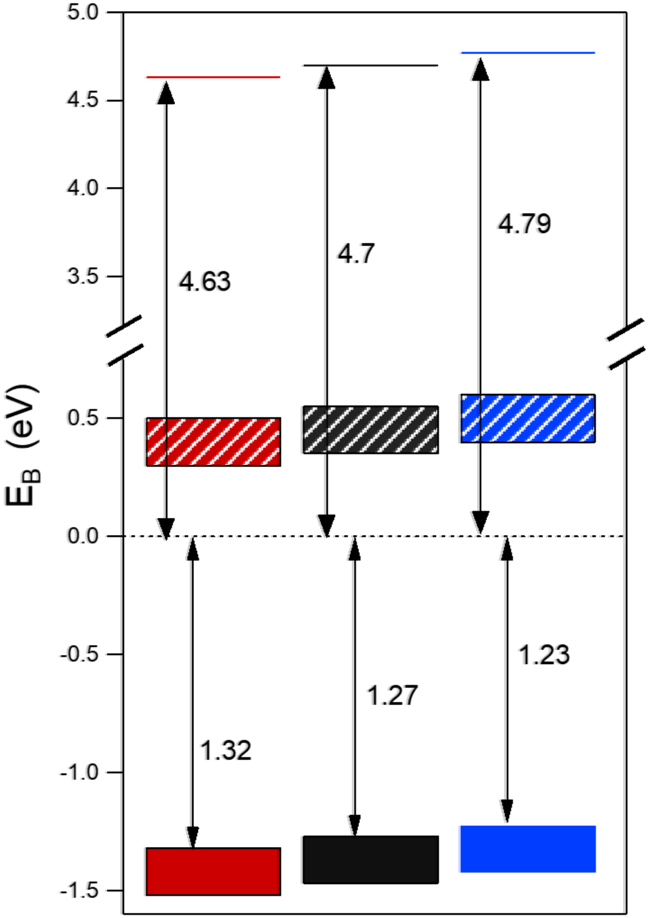


2L-WS_2 on as-deposited Au





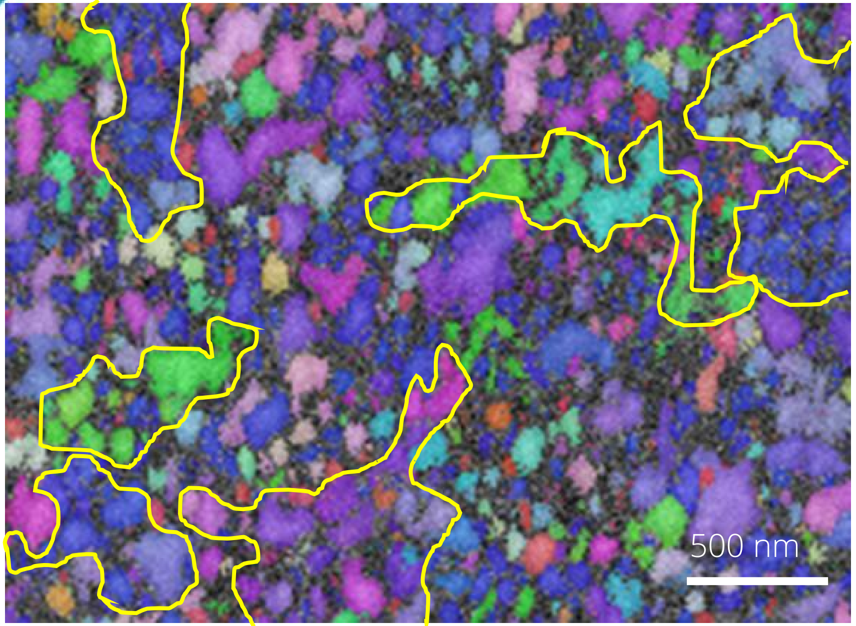


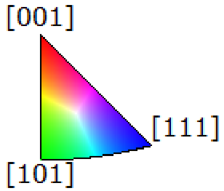


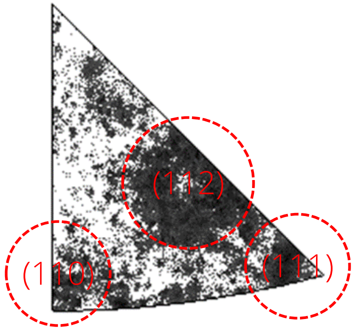
- Same micron-sized domains from photoelectron intensity and ϕ maps
- Higher work function regions show corresponding upshift in VBM = rigid shift
- Low BE peak observed near fermi level only in high work function areas
- Au (111) surface state
- More n-type
- More p-type
- Schottky barrier height is expected to vary across the metal contact

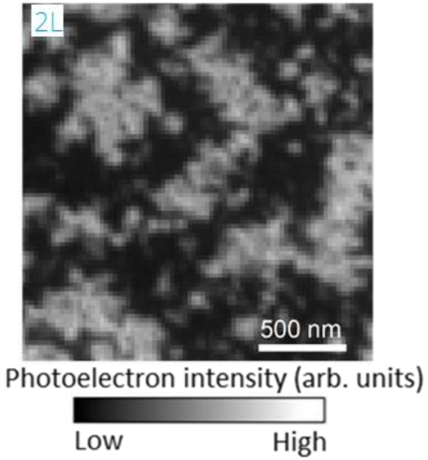
Three predominant crystallographic orientations of the Au grains elucidated via EBSD

WS_2 covered As-deposited Au







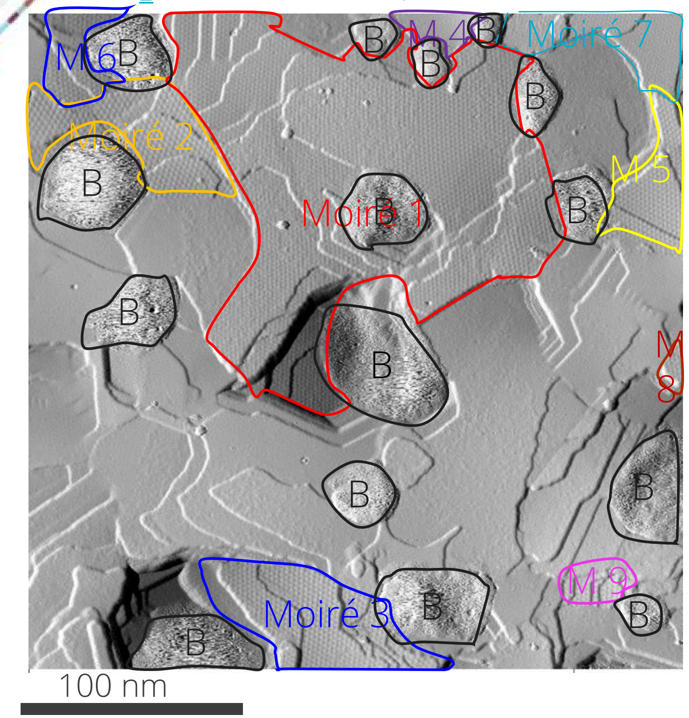


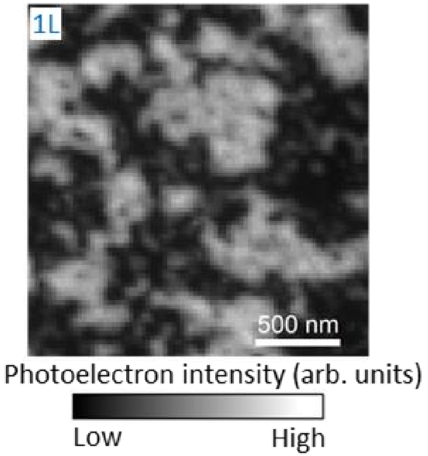
EBSD: Electron Backscatter Diffraction

- Majority of Au grains have (111), (112), or (110) facets
- Clusters with the same crystallographic orientation resemble the heterogenous domains of WS_2 probed using PEEM
- Consistent with absence of Au(111) surface state in some regions of PEEM

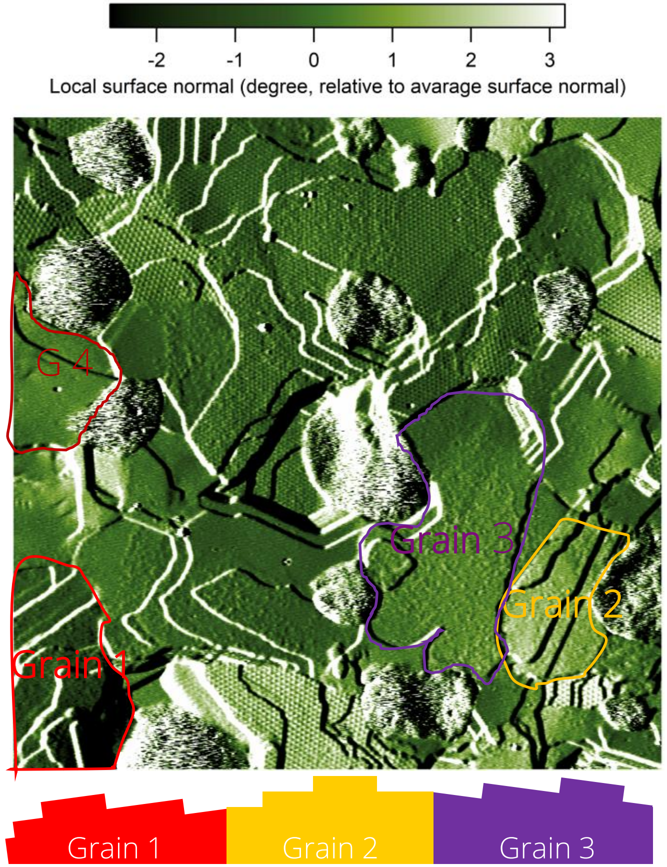
Moiré periodicities and absence of Moiré on some grains correlates with varying Au crystal orientations

WS_2 covered as-deposited Au





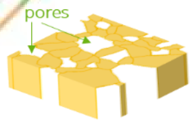
- Atomic terraces of Au and trapped blisters
  - Blisters/bubbles likely contain physisorbed molecules captured during exfoliation
- Hexagonal Moiré patterns indicate closely adhered WS_2
  - Arise from overlapping WS_2 and unreconstructed Au(111) lattices with azimuthal misalignment
  - Varying superlattice periodicities found on some Au terraces



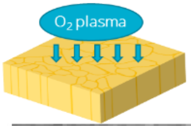
- Analysis of local surface normal can further dictate Au grains that do not show Moiré
  - (112) and (110) oriented grains
- Shapes of Au grains resemble the heterogenous domains of WS_2 probed using PEEM

Crystal facets of Au appear to govern the electronic properties of WS_2

TMD on OPEN and Plasma-treated Au yield uniform electronic structures

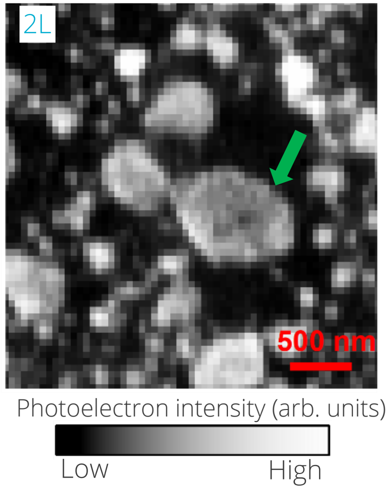


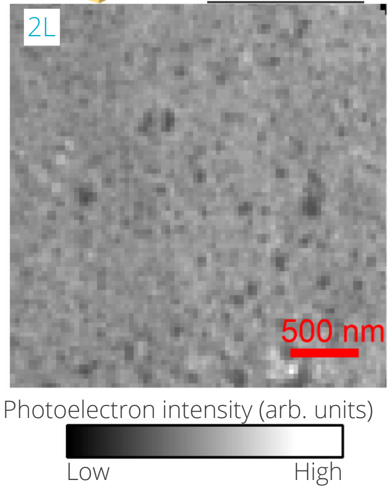
OPEN

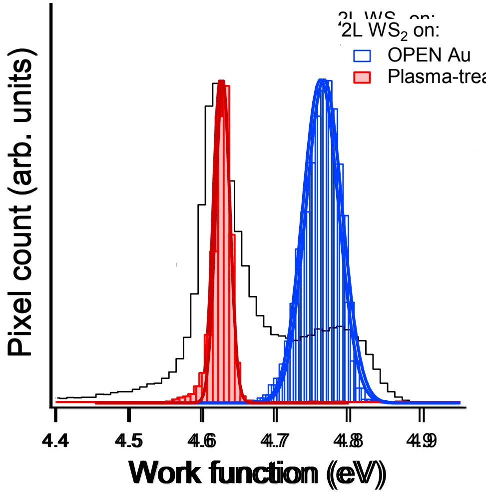


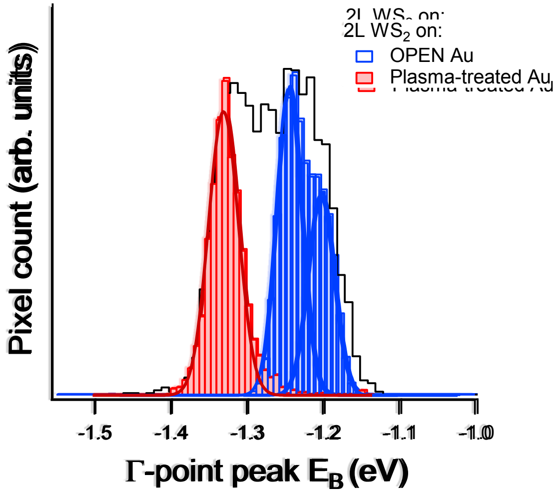
Plasma-treated

WS_2





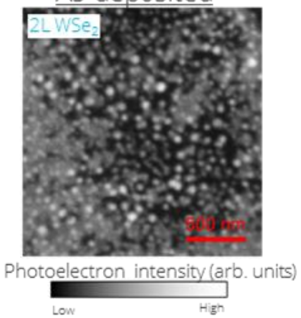




WS_2 exfoliation on:

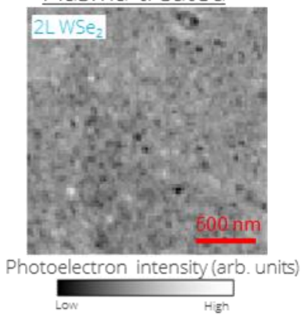
- OPEN Au = high work function/more p-type TMD
- Plasma-treated Au = low work function/more n-type TMD
  - These trends are consistent across 1-3L WS_2

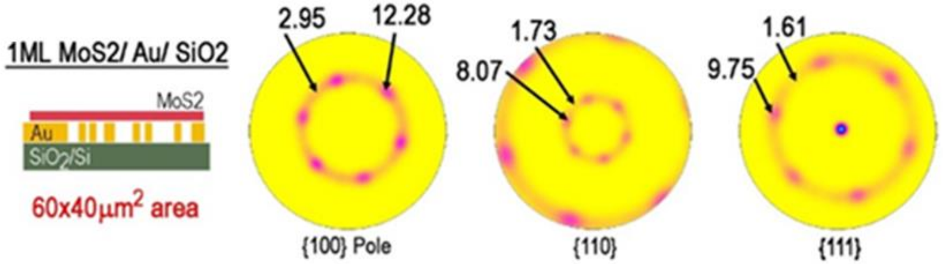
As-deposited



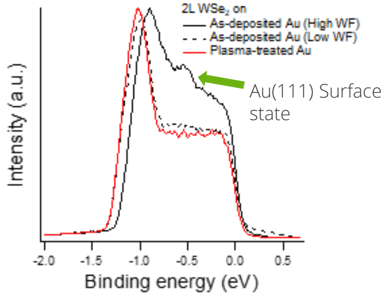
WSe_2

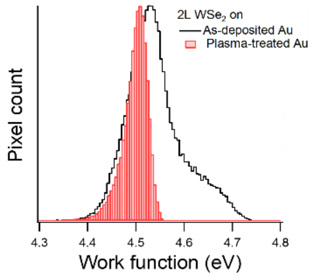
Plasma-treated





- Au grains in OPEN sample all (111)

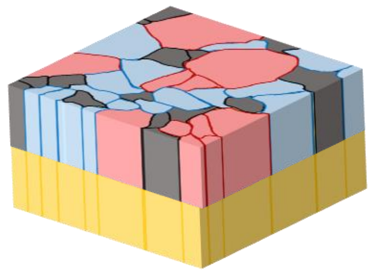


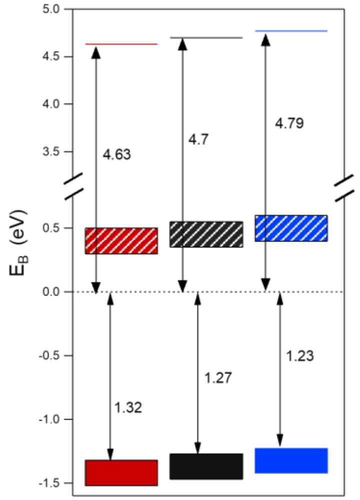


Fonseca et al., Nat. Commun., 2020, 11, 5

Concluding remarks

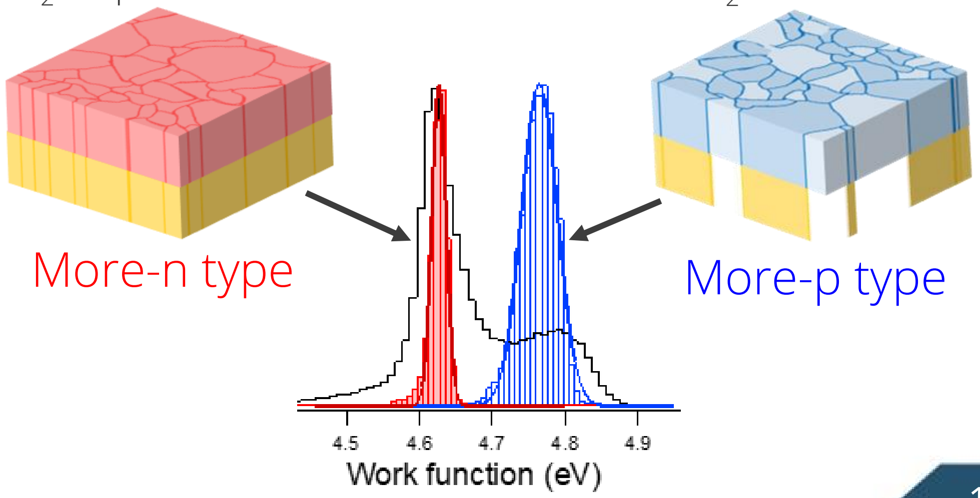
- We have quantified key variations in electronic structure of TMD-Au interfaces indicative of fluctuating doping levels and Schottky barrier height relevant for device application





- Local variations of the WS_2 electronic structures likely governed by the crystal orientation of Au grains
  - Appears microstructures of Au films plays an inherent role in the TMD-Au contacts

- Further, controlled processing of Au substrates can generate uniform TMD-Au interfaces and tune the electronic properties of the TMD



Financial support:

- **Sandia LDRD program**

Special thanks:

- **C.D. Spataru, N. Bartelt, F. Leonard, R. G. Copeland, Sandia National Laboratories**