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# Engineering of Nanoscale Heterogenous Transition Metal Dichalcogenide-Au Interfaces

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**AVS 68th International Symposium & Exhibition: November, 2022**
**2D Materials Technical Group**
**2D Materials: Electron Microscopy and Photoemission Spectroscopy**
**Session 2D+AS+EM-ThA**
**Room 303**

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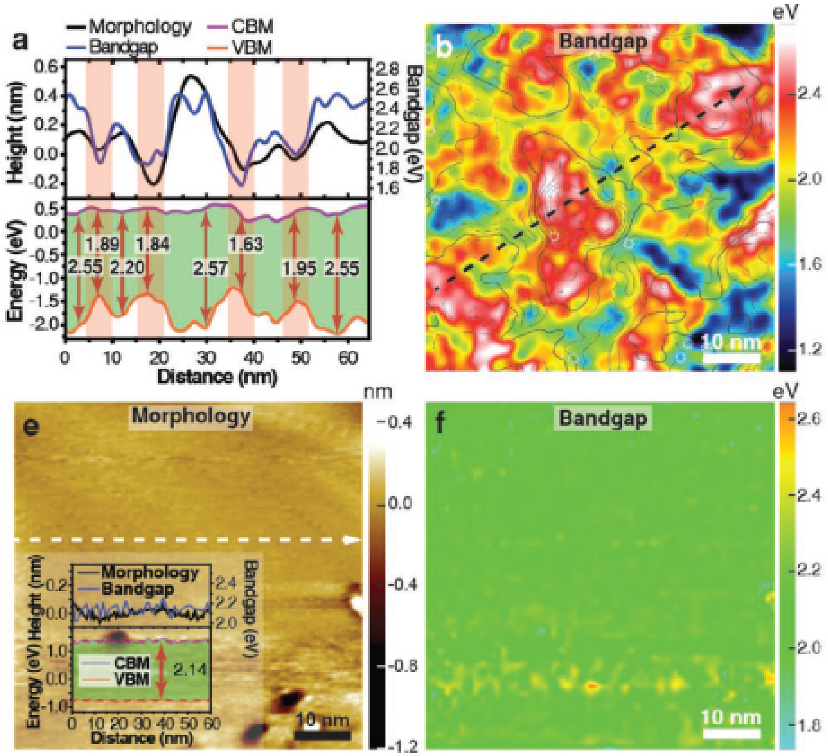
# 2D materials are susceptible to extrinsic factors

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

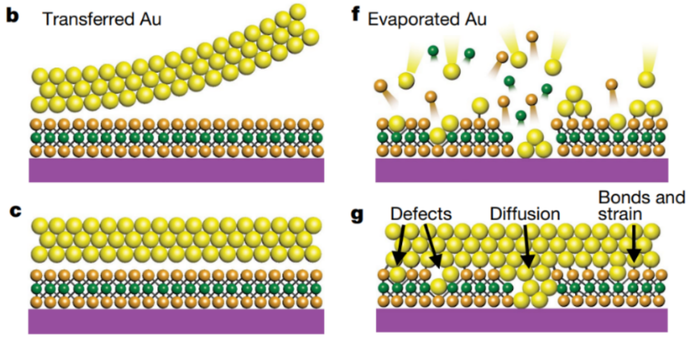



SiO2

HOPG

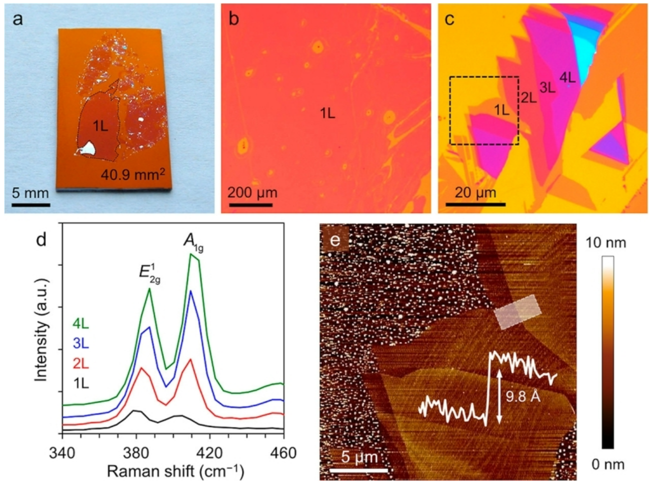
Shin, et al., Advanced Materials 28, 2016, 9378-9384

- Interfaces with metallic contacts materials are crucial components in electronic/optoelectronic devices




Liu, et al., Nature 557, 2018, 696-700

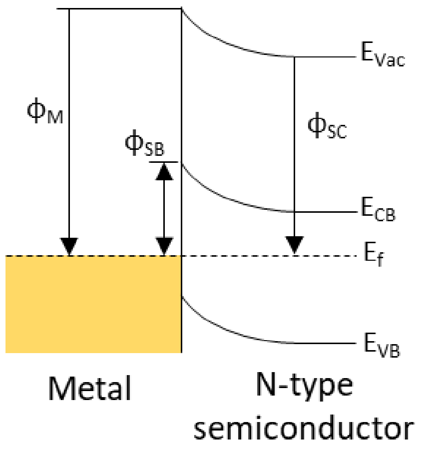
- Strong interactions with Au has enabled Au-assisted exfoliation to produce large area high-quality 2D TMDs



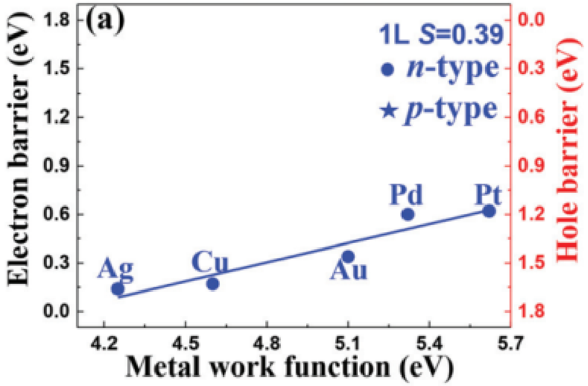

Velický, , et al., ACS Nano, 2018, 12, 10, 10463-10472

# TMD sensitivity strongly impacts electronic performance

- The Schottky barrier height (SBH) is a key parameter in device functionality

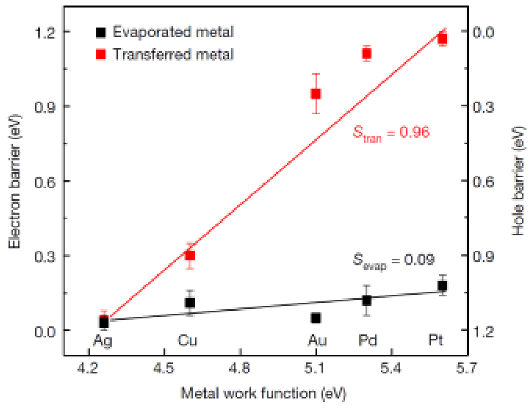



- Tuning SBH is difficult in TMD devices due to Fermi-level pining (FLP)



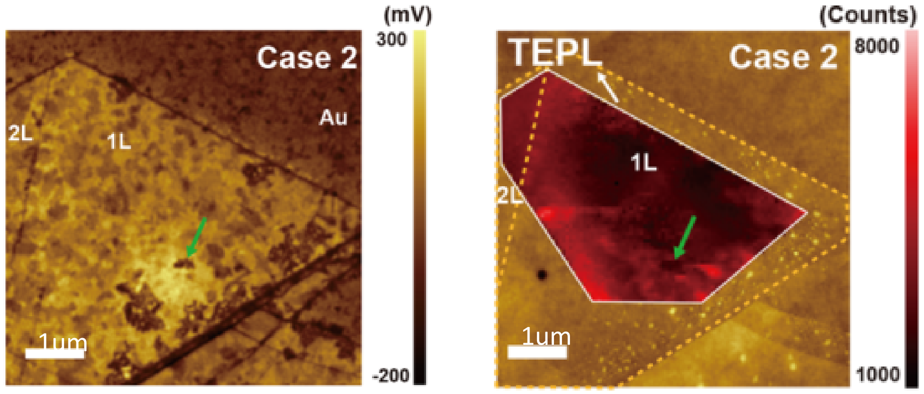

Wang, et al., Journal of Materials Chemistry C, 2020, 3113–3119.

- Multilayered TMDs can alleviate FLP




Liu, et al., Nature 557, 2018, 696-700

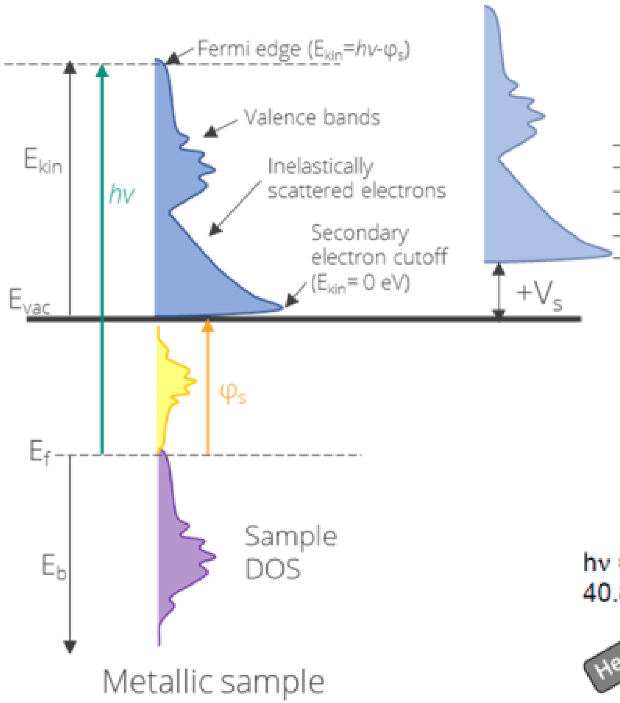
- Archetypal methods for extracting SBH from transport measurements average over the entire contact area

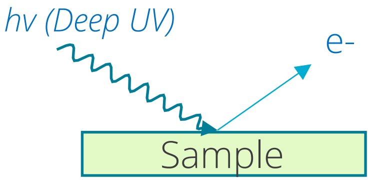



Jo, et al., ACS Nano, 2021, 15, 5618

# 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



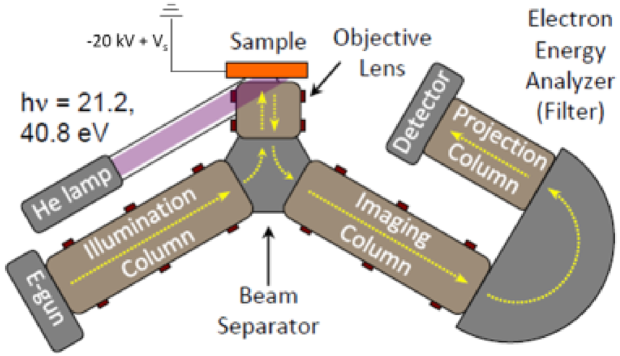
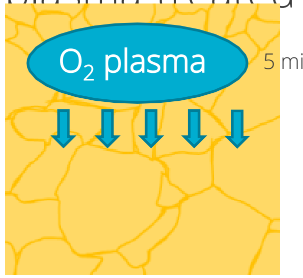




Figure courtesy: M. Berg

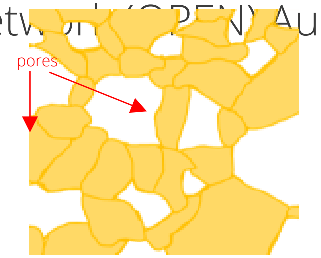
- We study TMD-Au interfaces fabricated through mechanical exfoliation with three different types of Au substrates

(i) as-deposited
- E-beam evaporated

(ii) plasma-treated

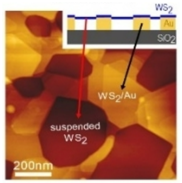



(iii) oriented porous metallic network (OPEN) Au



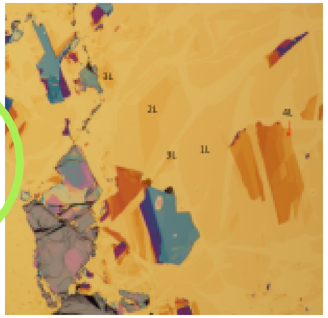

Fonseca et al., Nat. Commun., 2020, 11, 5

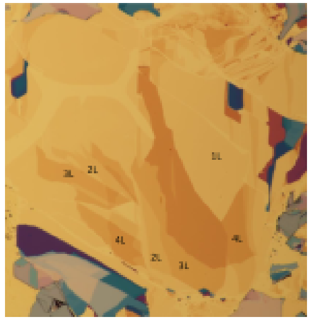
- Pseudo-epitaxial interface
- Resulting from the re-flow & recrystallization of Au by annealing




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

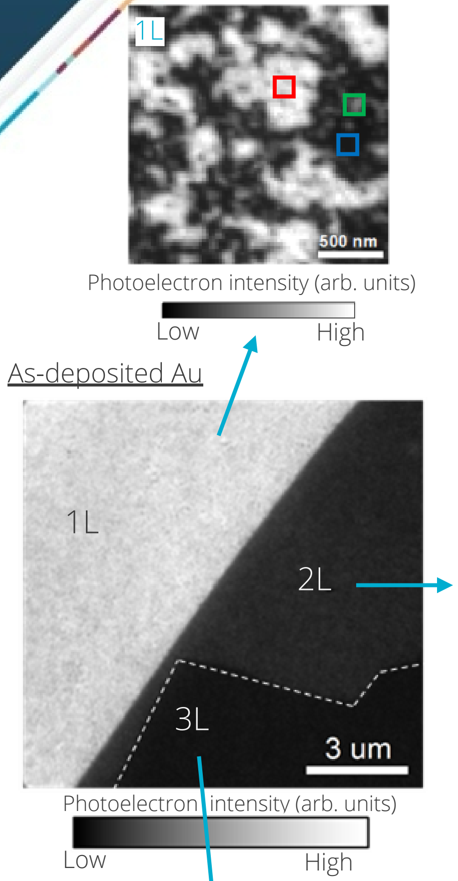
$WS_2$

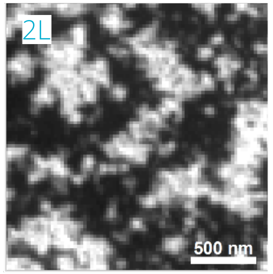


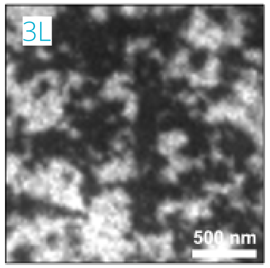


$WSe_2$

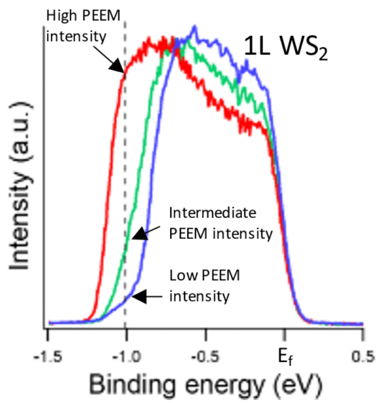
# Micron-scale work function heterogeneity in as-deposited Au-$WS_2$ interfaces



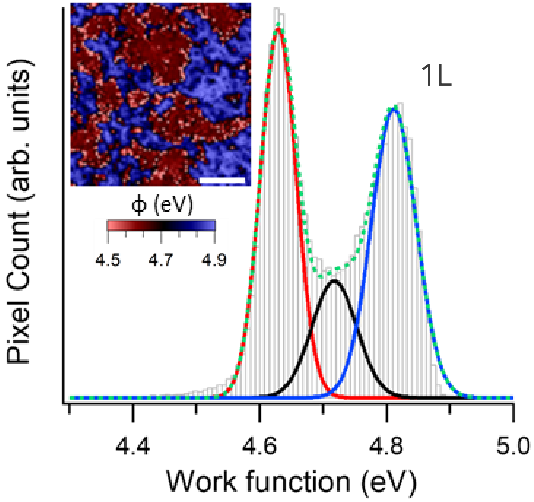



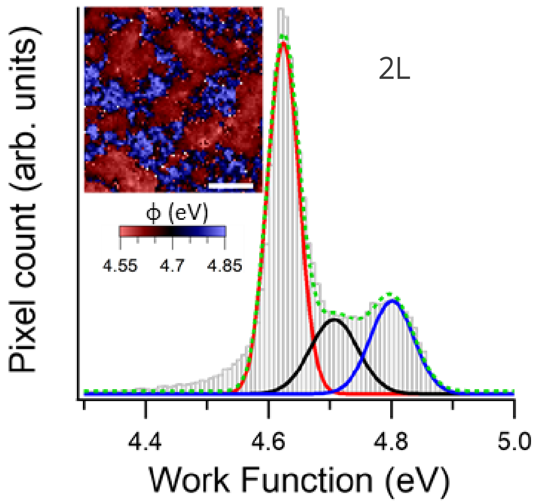




- Contrasting um sized domains in PEEM intensity
- Present in 1-3L $WS_2$ thickness



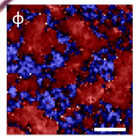

- PEEM contrasts arise from difference in secondary electron cutoff i.e. work function



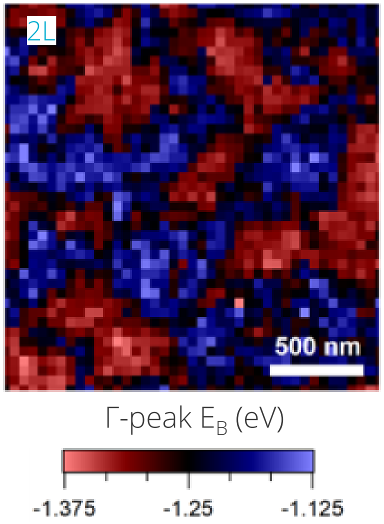



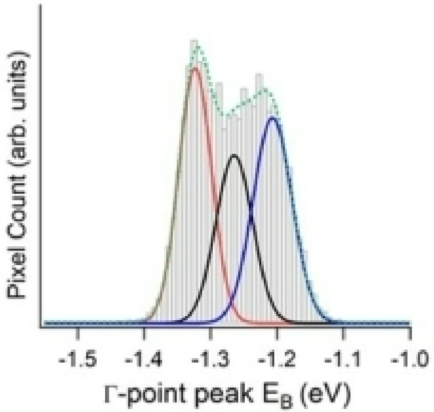

- Large work function variation (~200 meV)
- Indicates that the carrier density varies in the $WS_2$ flakes

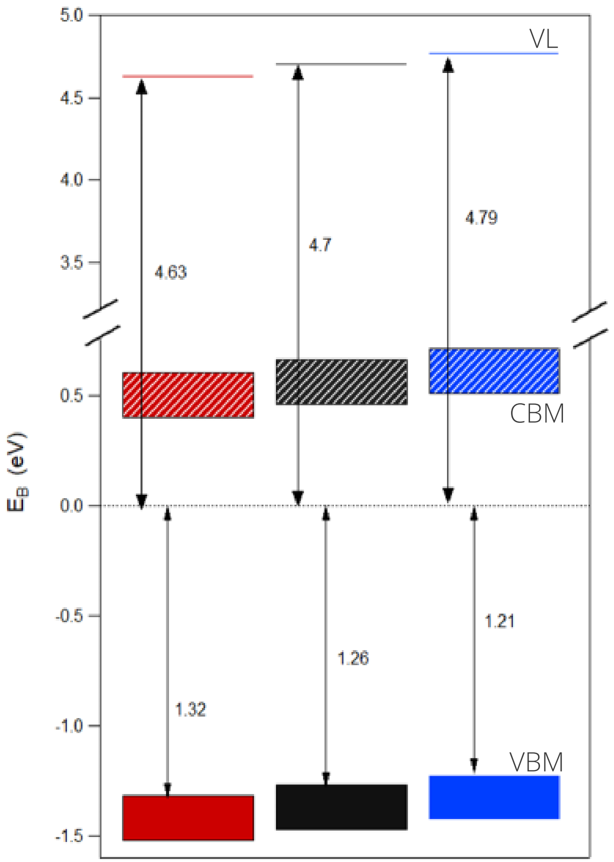
# Micron-scale VBM heterogeneity in as-deposited Au-$WS_2$ interfaces

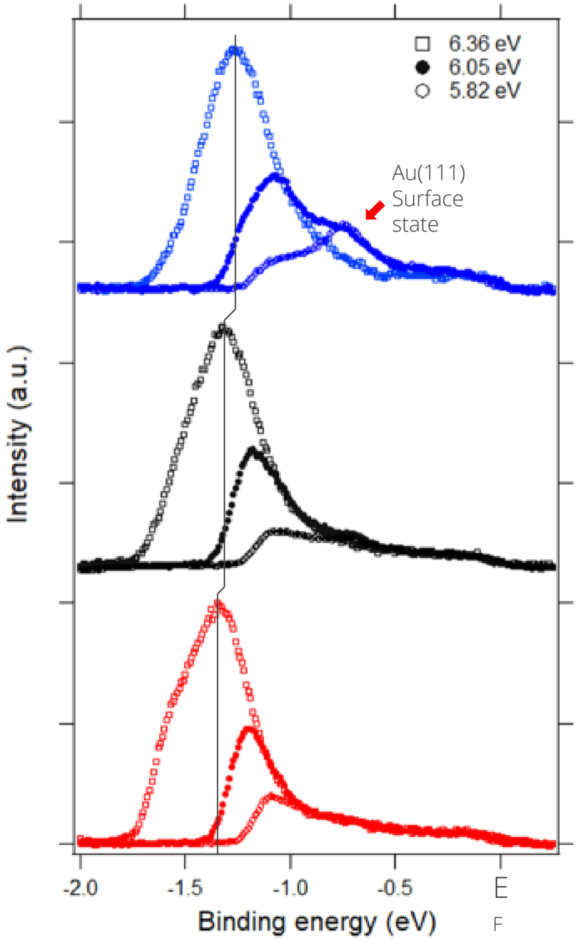



## 2L-$WS_2$ on as-deposited Au



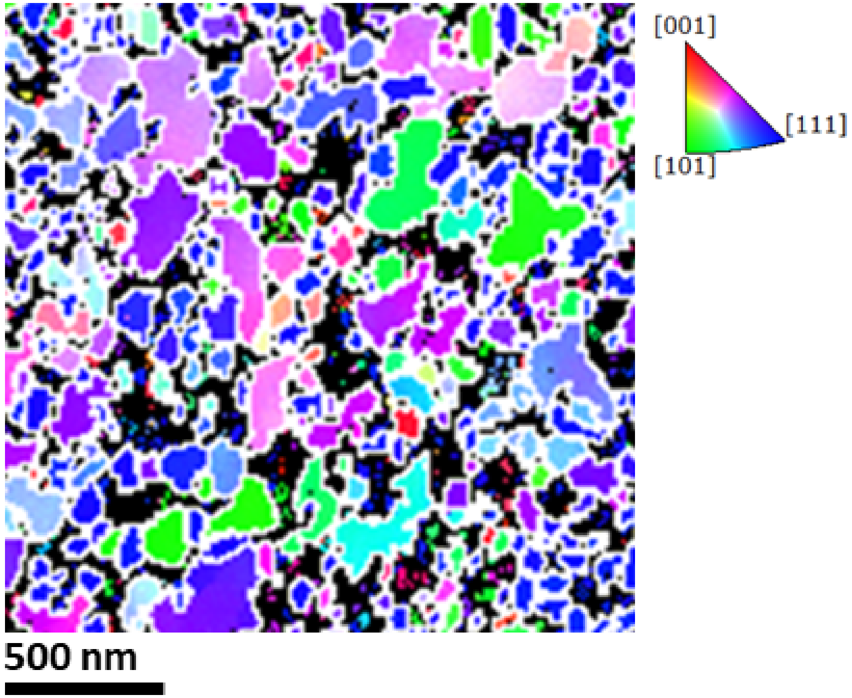

- Same micron-sized domains from photoelectron intensity and ϕ maps
- Higher work function regions show corresponding upshift in VBM = rigid shift
- Three distinct regions

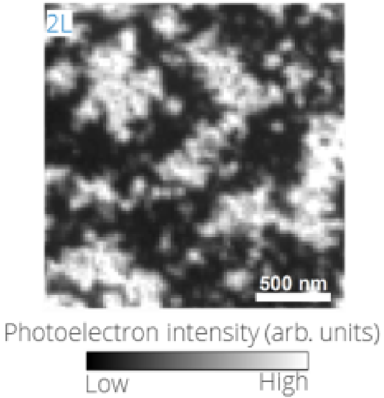
More n-type    More p-type

- Schottky barrier height is expected to vary across the metal contact
- Low BE peak observed near fermi level only in high work function areas
- Au (111) surface state

# Three predominant crystallographic orientations of the Au grains elucidated via EBSD

$WS_2$ covered as-deposited Au



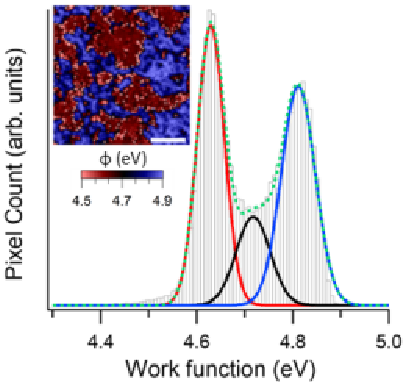

## EBSD: Electron Backscatter Diffraction

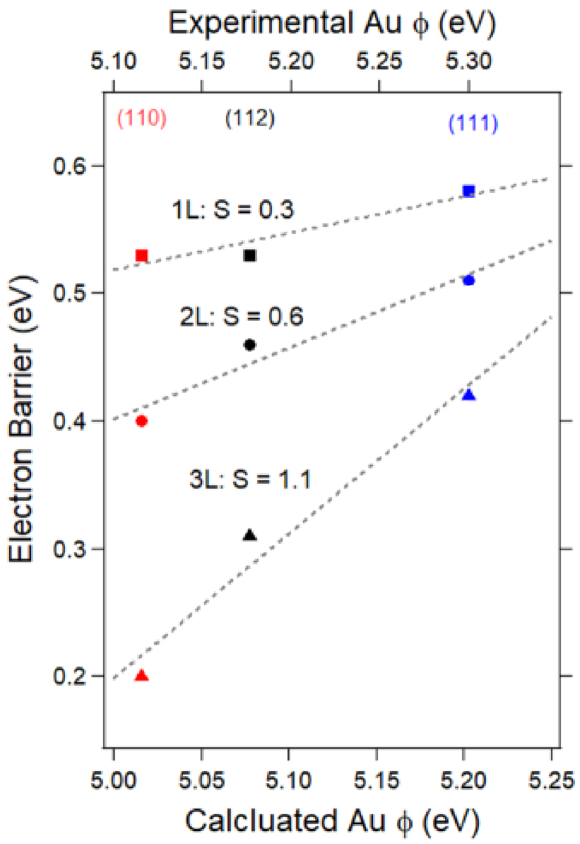
- Majority of Au grains have (111), (112), or (110) facets
- Consistent with absence of Au(111) surface state in some regions of PEEM
- Same length scale as heterogenous electronic structure domains

| Au | (110) | (112) | (111) | Δϕ (eV) |
|---|---|---|---|---|
| ϕ (eV) | 5.02 | 5.08 | 5.2 | 0.18 |

| $WS_2$ | 1L | 2L | 3L |
|---|---|---|---|
| Δϕ (eV) | 0.18 | 0.16 | 0.13 |

$$\Delta\phi_{Au} \approx \Delta\phi_{WS_2}$$



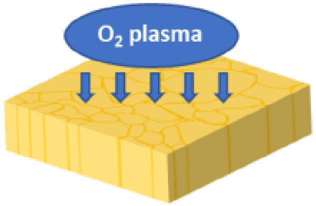

1L = strong FLP

↑# layers ↓FLP

3 SBH within a single junction

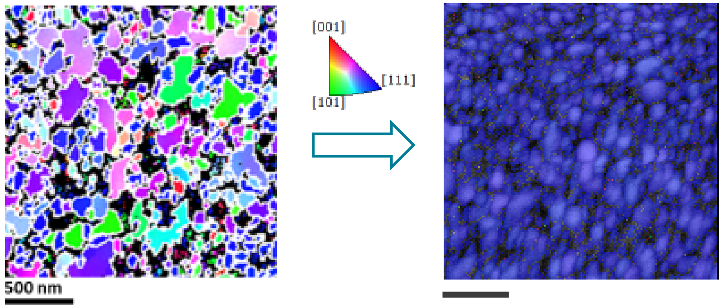
# TMD on OPEN and Plasma-treated Au yield more uniform electronic structures

Plasma-treated Au

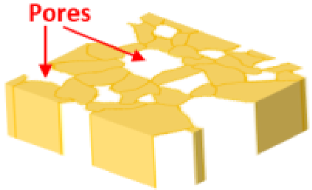


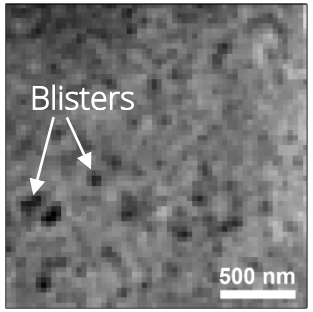

As-deposited Au

Plasma-treated Au



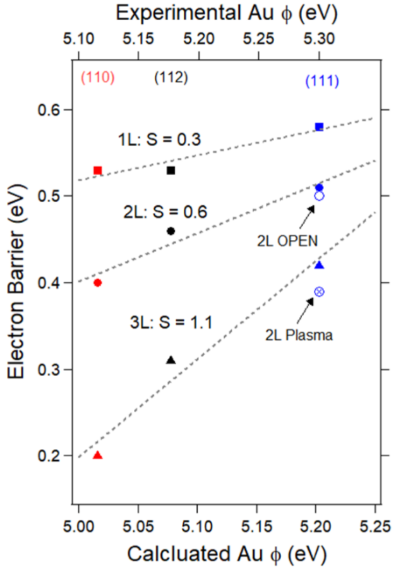

OPEN Au



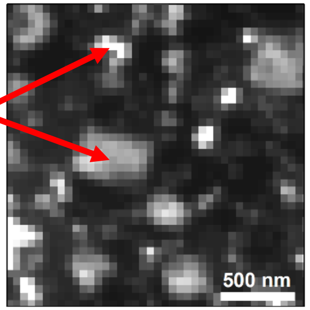




Photoelectron intensity (arb. units)

Low High

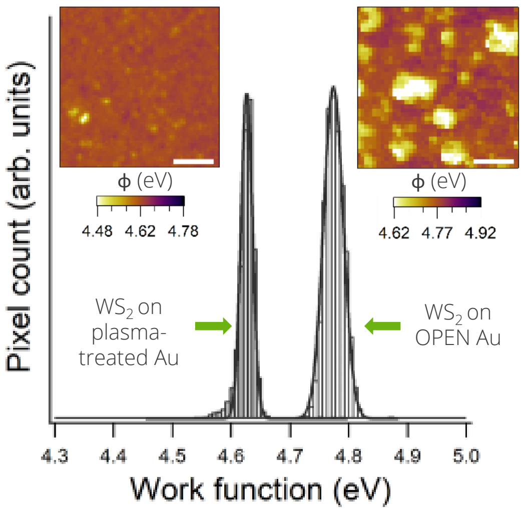



Suspended $WS_2$

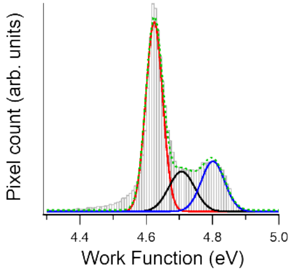



Photoelectron intensity (arb. units)

Low High

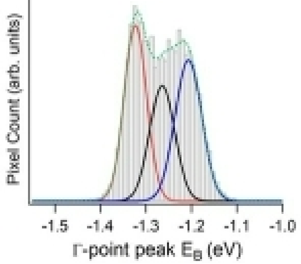


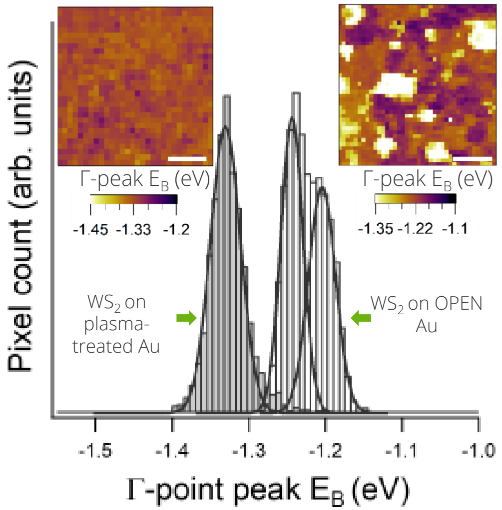

as-deposited Au




1 SBH for each $WS_2$–Au junction

as-deposited Au



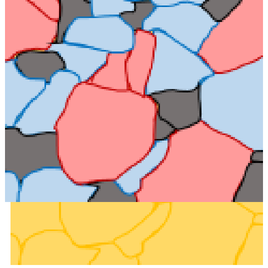



# Concluding remarks

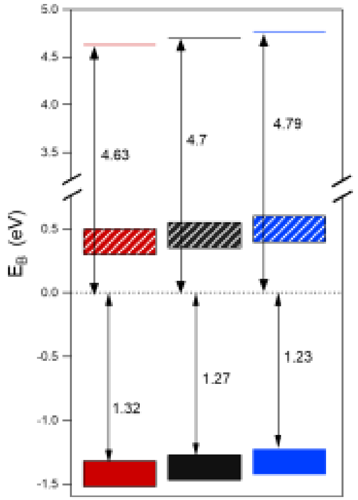
- We have quantified key variations in electronic structure of TMD-Au interfaces indicative of Schottky barrier height variation relevant for device application

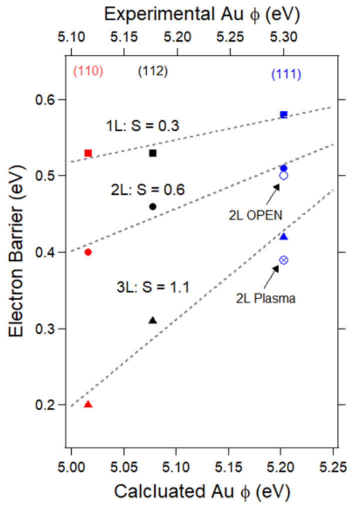
- Local electronic structure variations of $WS_2$ governed by the crystal orientation of Au grains
  - Metal microstructure plays an inherent role in contact formation with TMDs

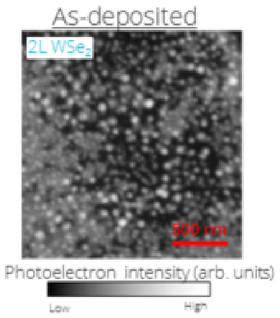
- Further, controlled processing of Au substrates can generate uniform TMD-Au interfaces



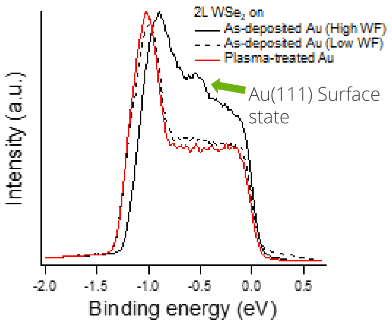
$WS_2$ on as-deposited Au

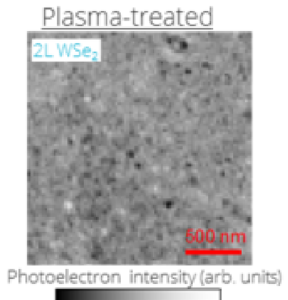



$WSe_2$



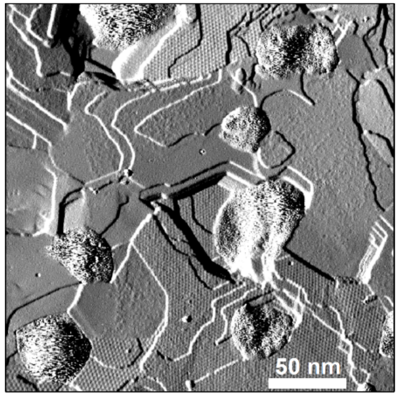

**Financial support:**

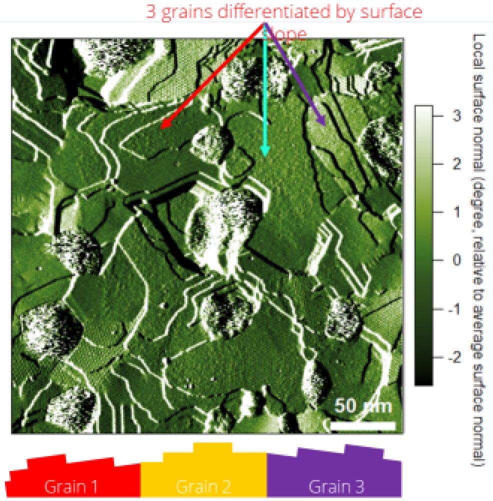
- Sandia LDRD program

**Special thanks:**

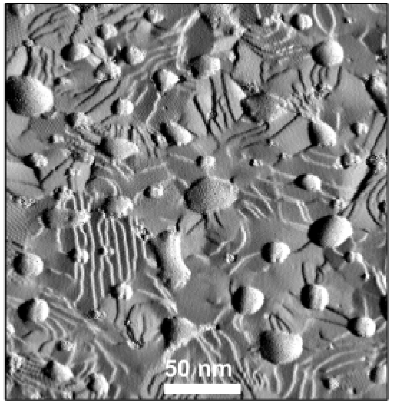
- N. Bartelt, F. Leonard, R. G. Copeland, Sandia National Laboratories

$WS_2$ covered as-deposited Au

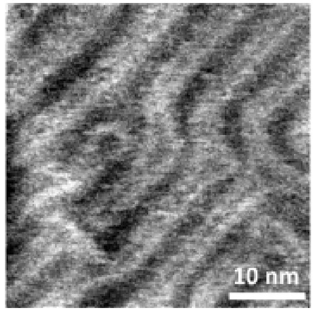


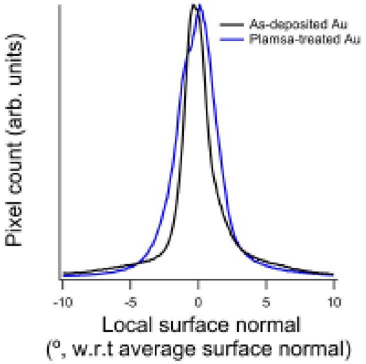



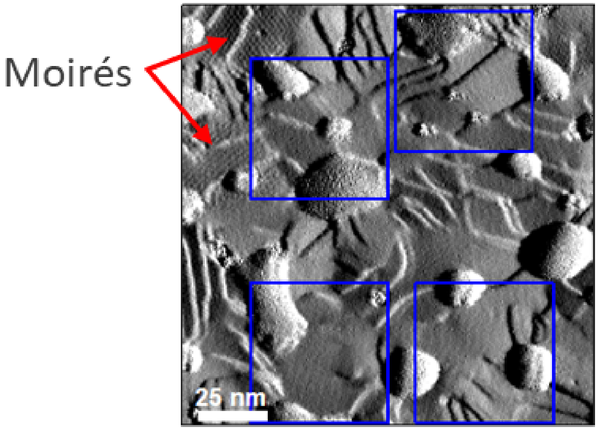

$WS_2$ covered plasma-treated Au




Exposed Au(111) in as-deposited

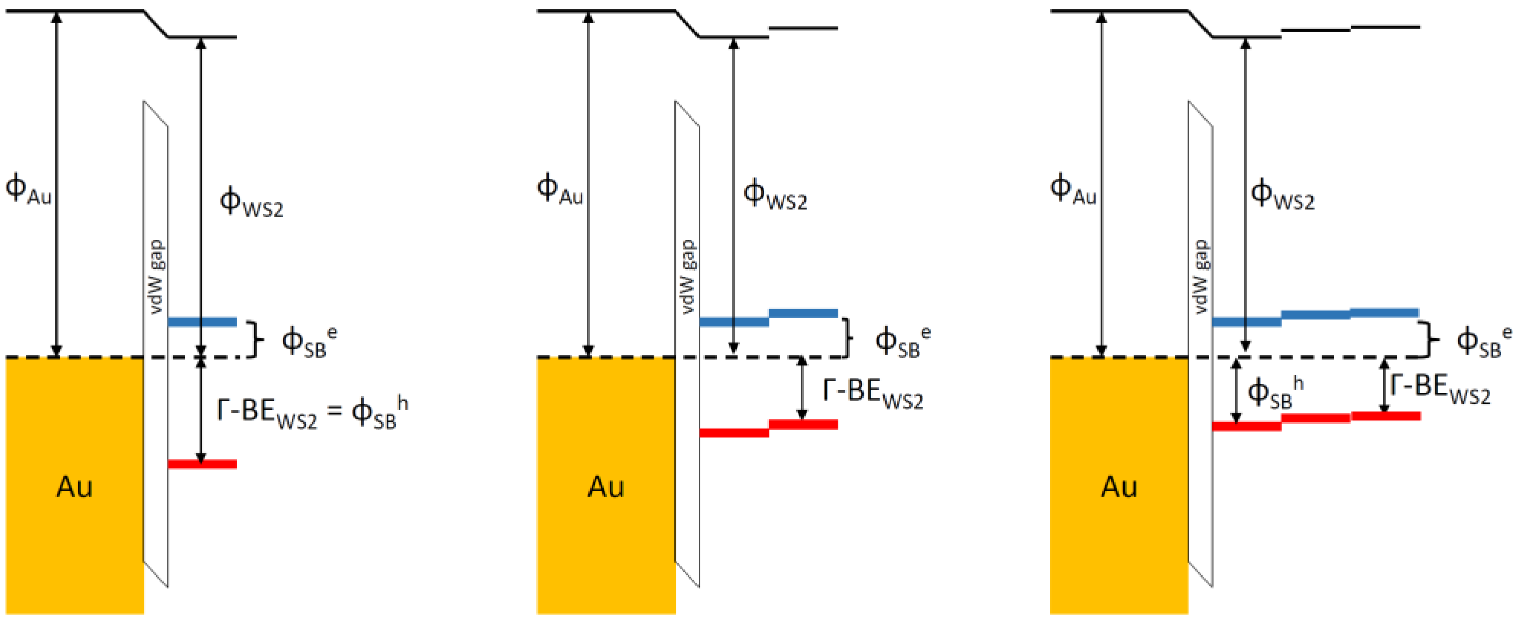


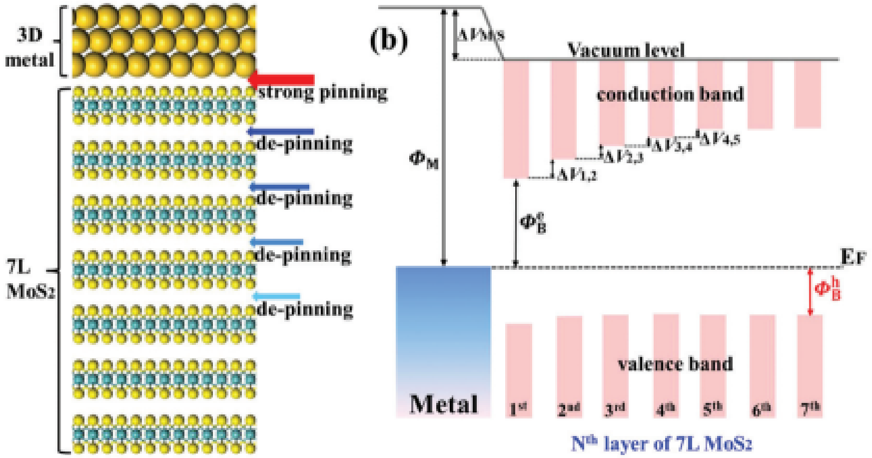




Herringbone reconstruction

## Model 3: Depinning effect




Wang, Q., Shao, Y., Gong, P., & Shi, X. (2020). Metal–2D multilayered semiconductor junctions: layer-number dependent Fermi-level pinning. *Journal of Materials Chemistry C*, *8*(9), 3113–3119. https://doi.org/10.1039/C9TC06331E