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Engineering of Nanoscale Heterogenous Transition Metal Dichalcogenide-Au Interfaces

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In collaboration with:

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The Rio Grande Symposium on Advanced Materials: October 2022
Session Theory and modeling/Low Dimension 2D materials
Presentation Time: 3:45 PM to 4:00 PM
Session Location: Embassy Suites, Albuquerque, Piñon Room

SAND#####

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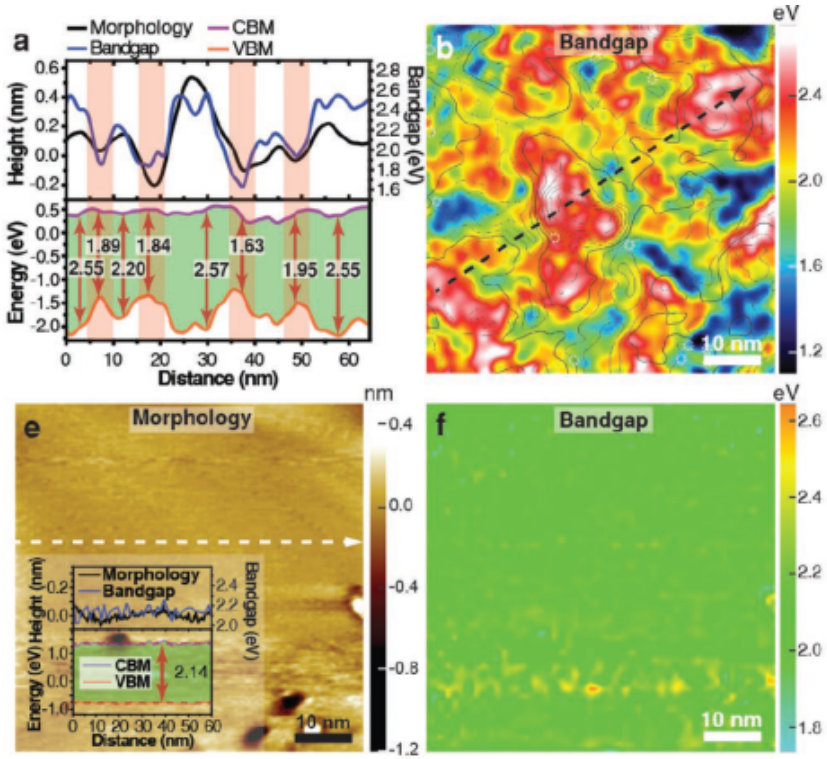
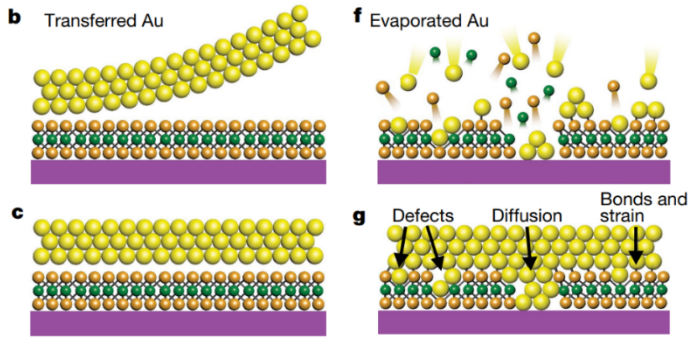
2D materials are susceptible to extrinsic factors

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



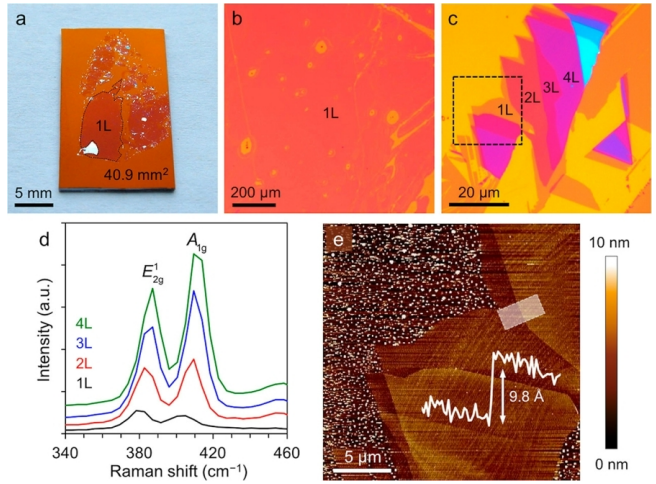
Shin, et al., Advanced Materials 28, 2016, 9378-9384

- Interfaces with metallic contacts materials are crucial components of electronic any optoelectronic device



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

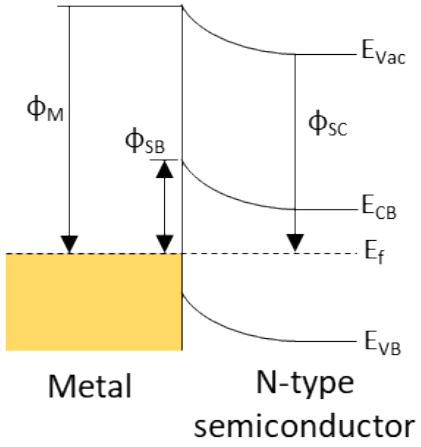
- Strong interactions with Au has enabled Au-assisted exfoliation to produce large area high-quality 2D TMDs



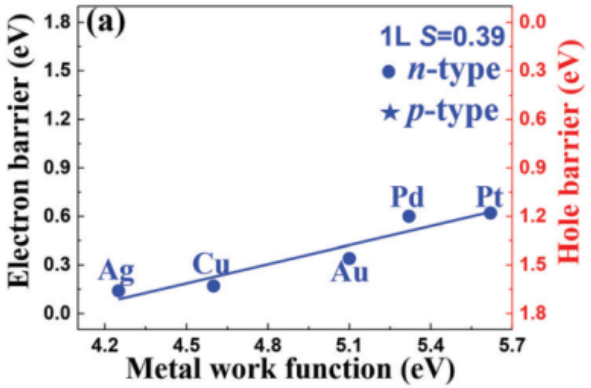
Velický, , et al., ACS Nano, 2018, 12, 10, 10463-10472

TMD sensitivity strongly impacts electronic performance

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

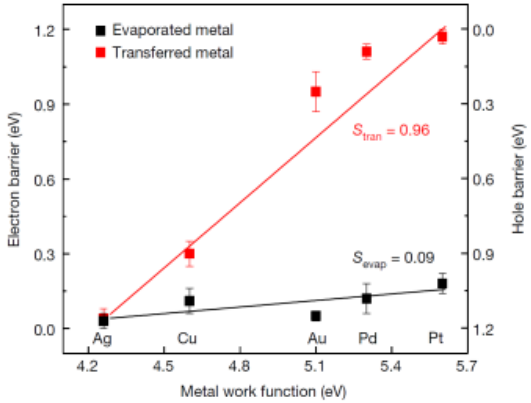


- Tuning SBH is difficult in TMD devices due to Fermi-level pining (FLP)



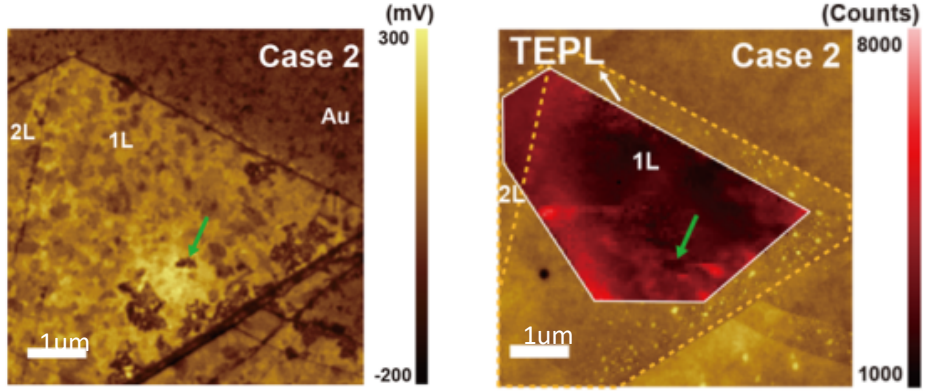
Wang, et al., Journal of Materials Chemistry C, 2020, 3113–3119.

- 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

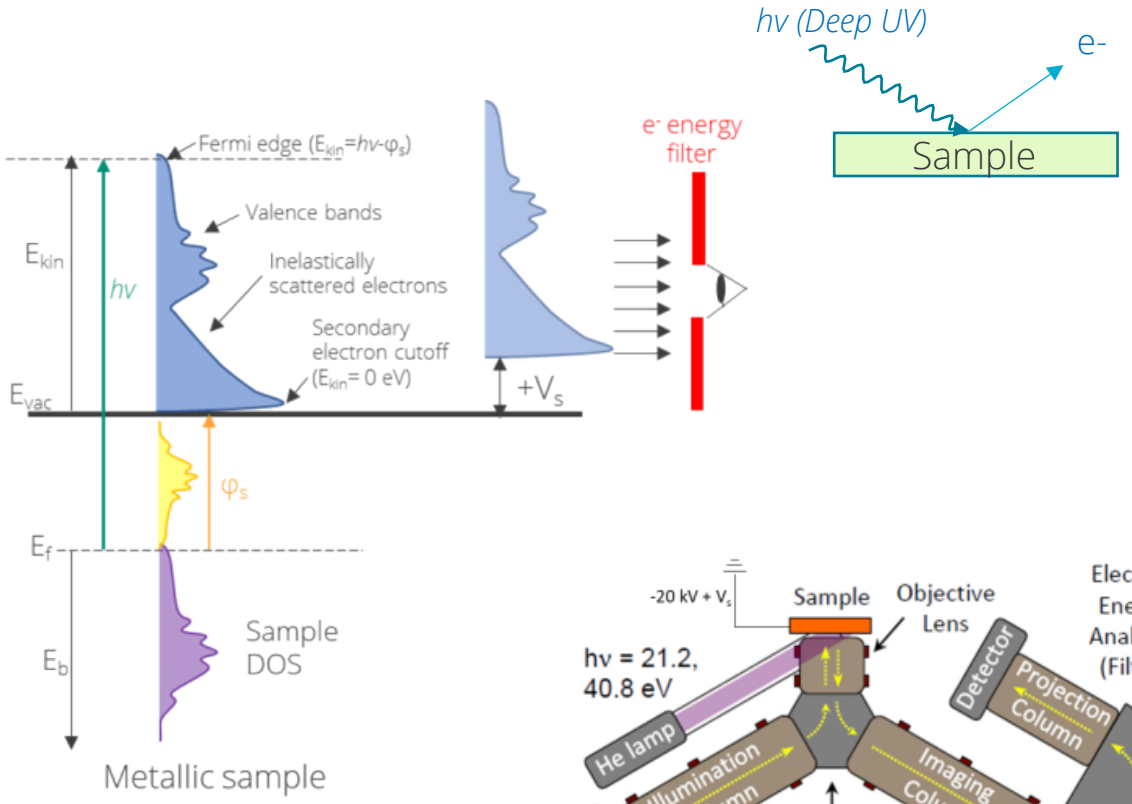
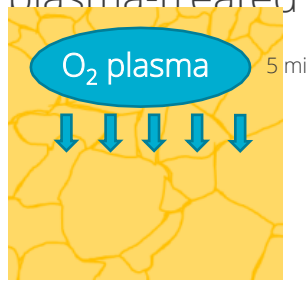


Figure courtesy: M. Berg

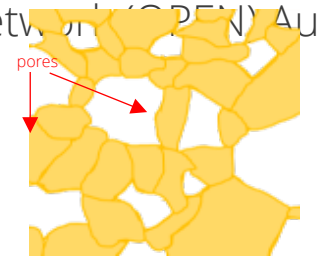
- We study TMD-Au interfaces fabricated through mechanical exfoliation with three different types of Au substrates

(i) as-deposited
- E-beam evaporated

(ii) plasma-treated

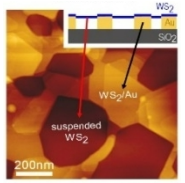


(iii) oriented porous metallic network (OPEN) Au



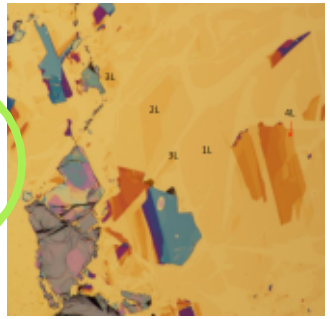
Fonseca et al., Nat. Commun., 2020, 11, 5

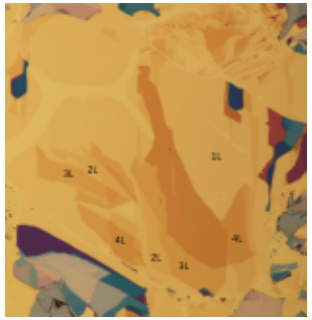
- Pseudo-epitaxial interface
- Resulting from the re-flow & recrystallization of Au by annealing



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

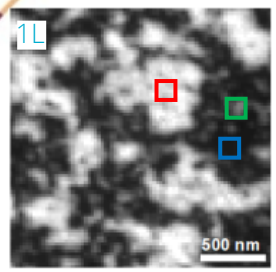
WS_2





WSe_2

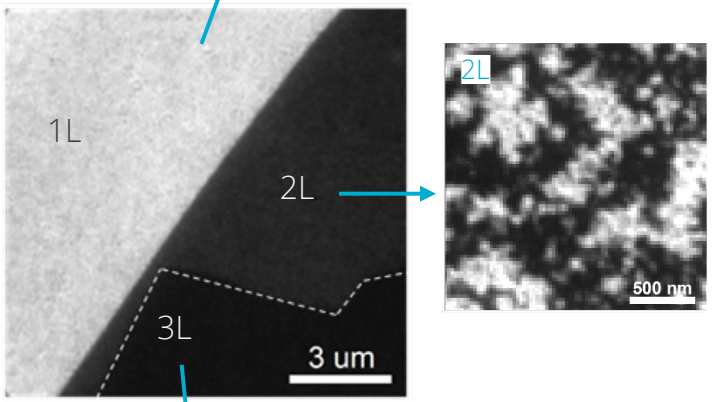
Micron-scale work function heterogeneity in as-deposited Au-WS_2 interfaces



Photoelectron intensity (arb. units)

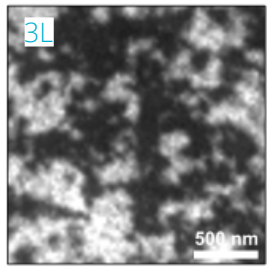
Low High

As-deposited Au

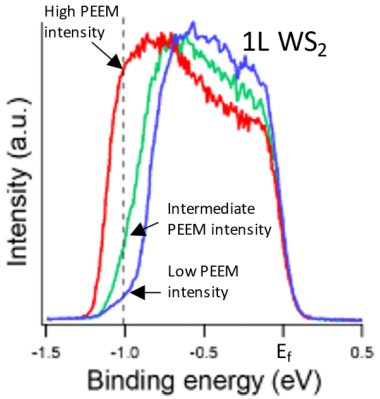


Photoelectron intensity (arb. units)

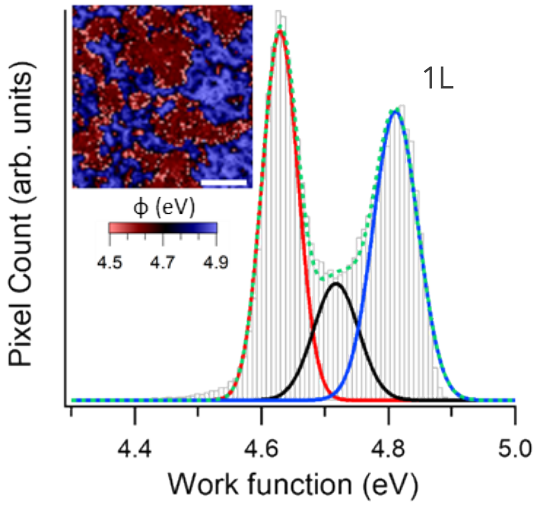
Low High

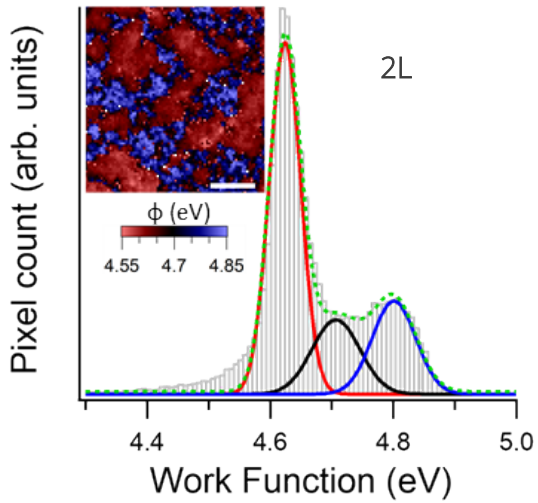


- Contrasting um sized domains in PEEM intensity
- Present in 1-3L WS_2 thickness



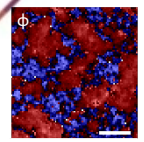
- PEEM contrasts arise from difference in secondary electron cutoff <u>i.e. work function</u>



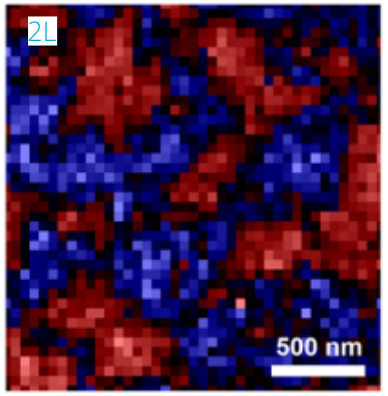


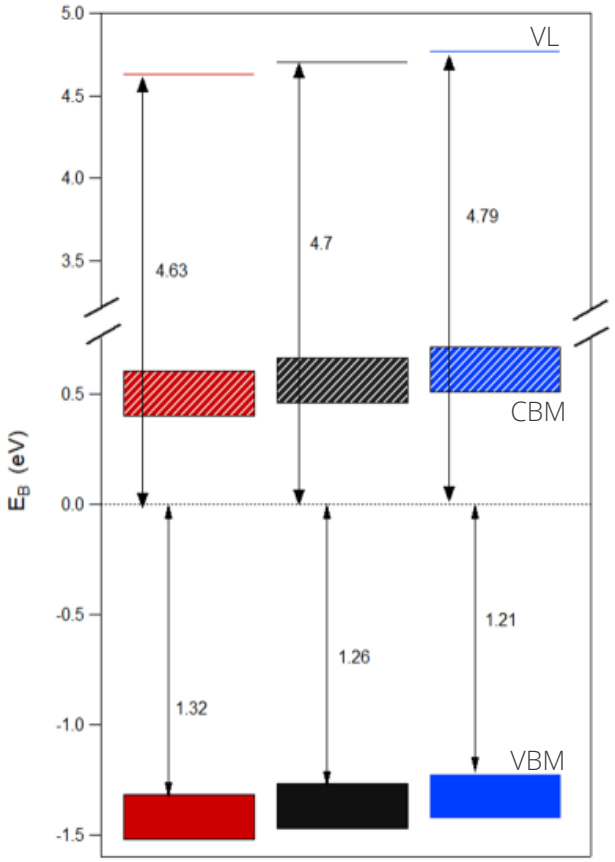
- Large work function variation (~200 meV)
- Indicates that the carrier density varies in the WS_2 flakes

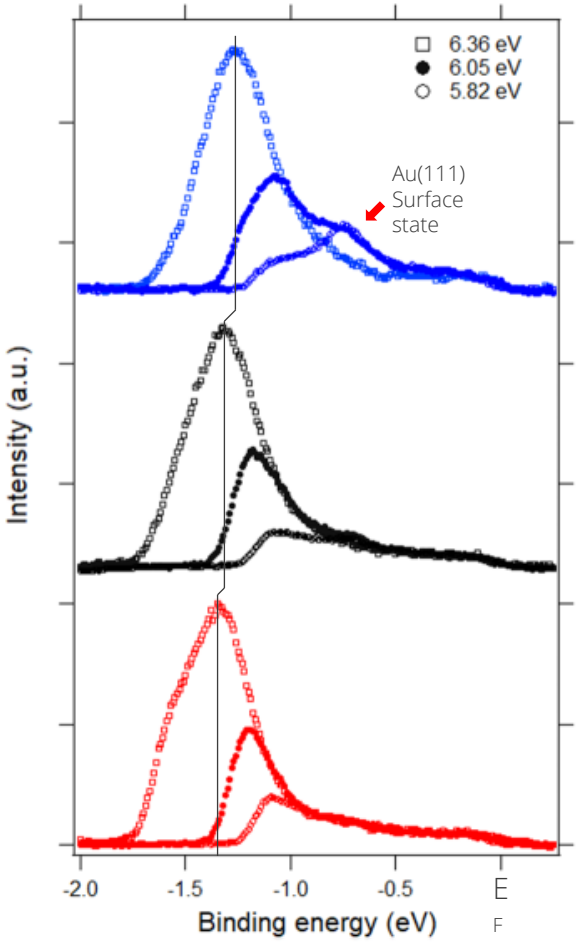
Micron-scale VBM heterogeneity in as-deposited Au-WS_2 interfaces



2L-WS_2 on as-deposited Au





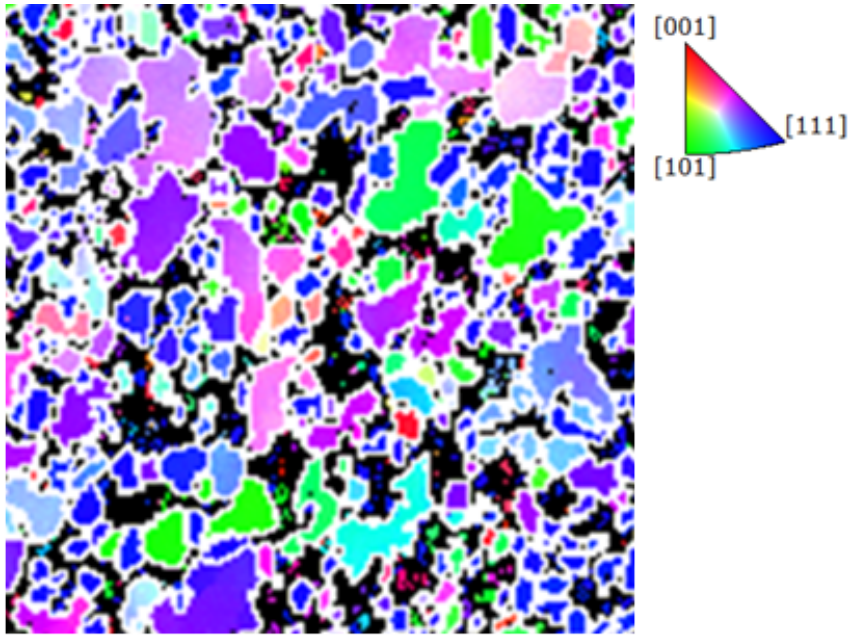


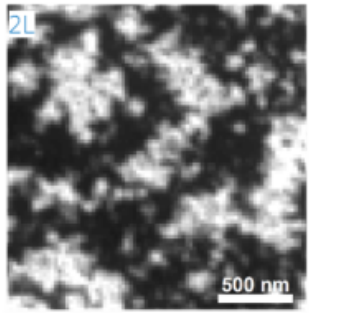
- Same micron-sized domains from photoelectron intensity and ϕ maps
- Higher work function regions show corresponding upshift in VBM = rigid shift
- Three distinct regions

  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





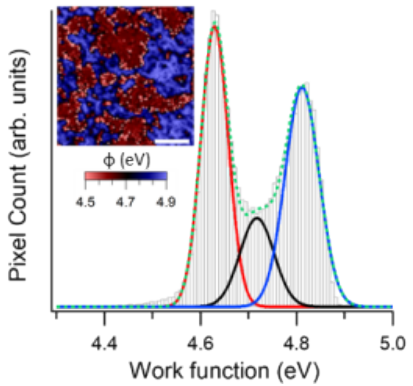
EBSD: Electron Backscatter Diffraction

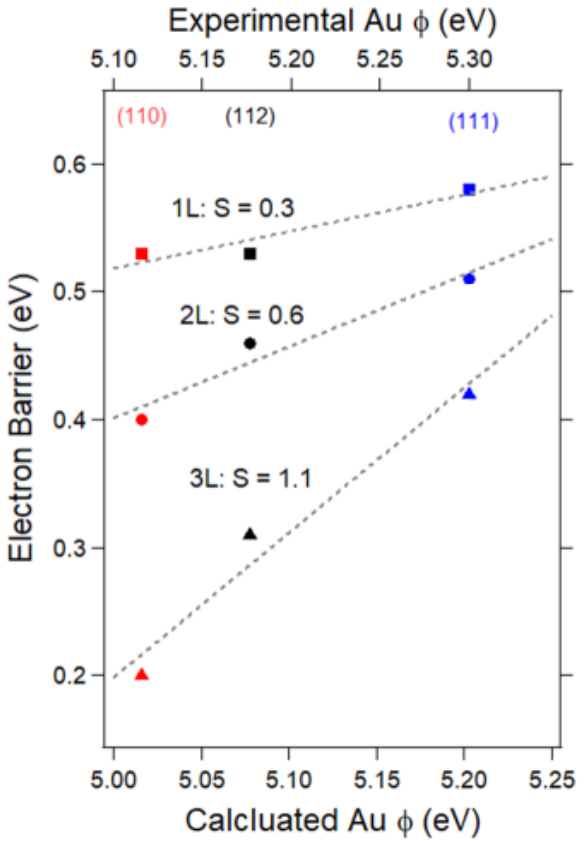
- Majority of Au grains have (111), (112), or (110) facets
- Consistent with absence of Au(111) surface state in some regions of PEEM
- Same length scale as heterogenous electronic structure domains

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

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





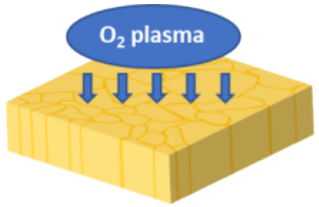
1L = strong FLP

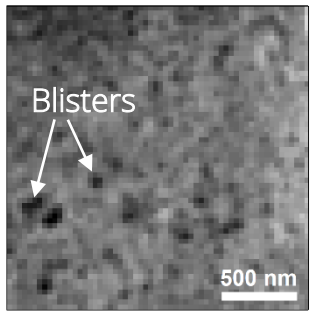
↑# layers ↓FLP

3 SBH within a single junction

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

Plasma-treated Au

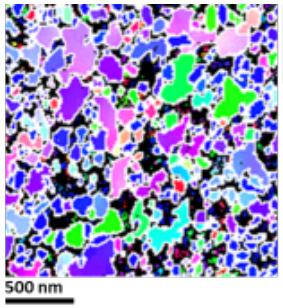


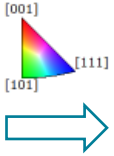


Photoelectron intensity (arb. units)

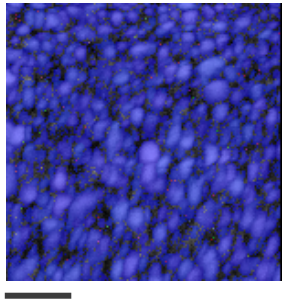
Low High

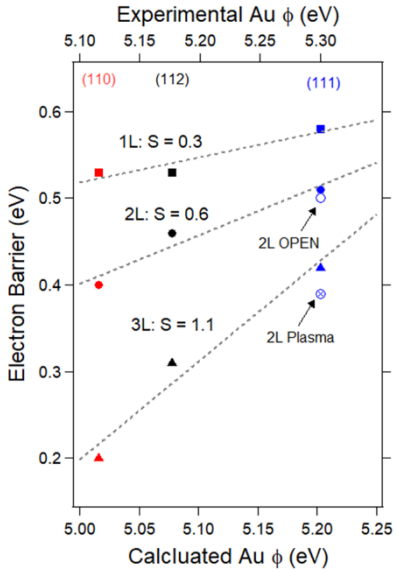
As-deposited Au





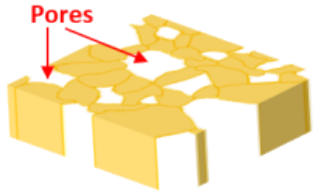
Plasma-treated Au



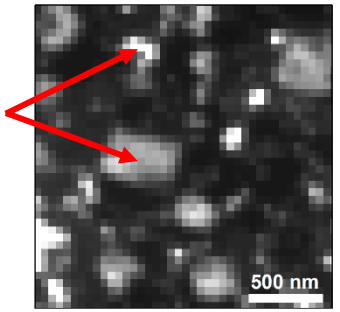


1 SBH for each WS_2–Au junction

OPEN Au

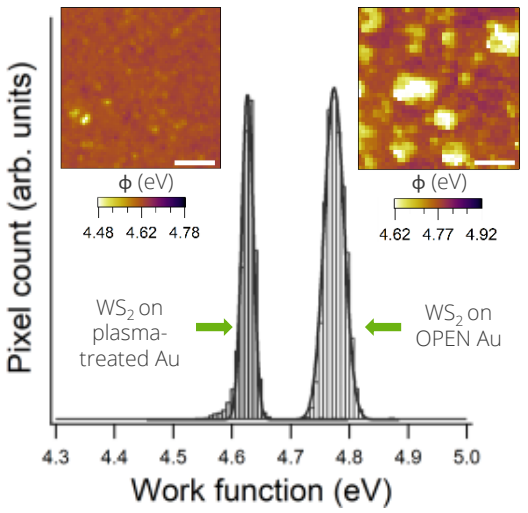


Suspended WS_2

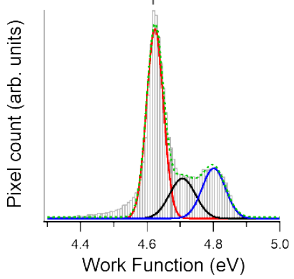


Photoelectron intensity (arb. units)

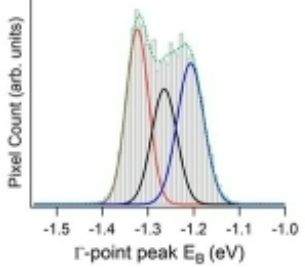
Low High

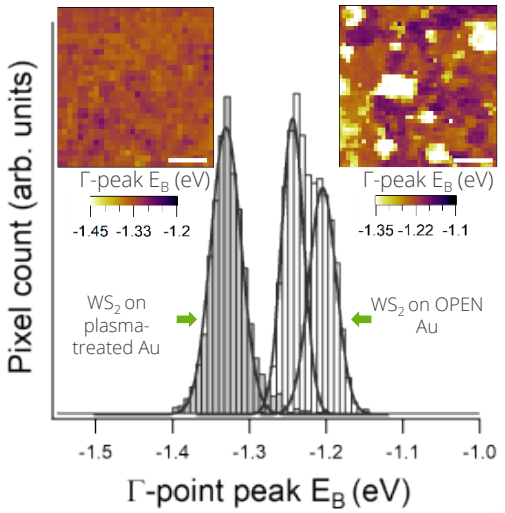


As-deposited Au



As-deposited Au



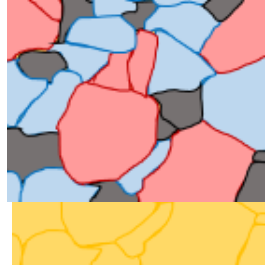


Concluding remarks

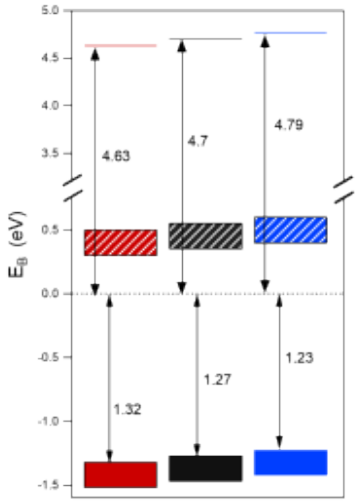
- We have quantified key variations in electronic structure of TMD-Au interfaces indicative of Schottky barrier height variation relevant for device application

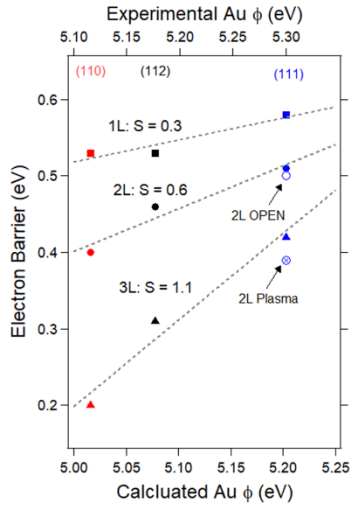
- Local electronic structure variations of WS_2 governed by the crystal orientation of Au grains
  - Metal microstructure plays an inherent role in contact formation with TMDs

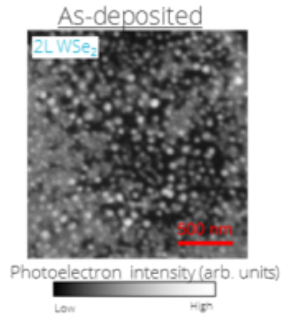
- Further, controlled processing of Au substrates can generate uniform TMD-Au interfaces



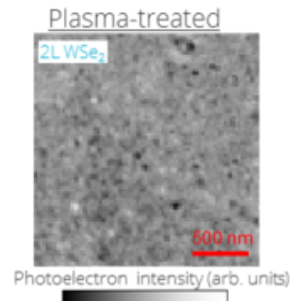
WS_2 on as-deposited Au

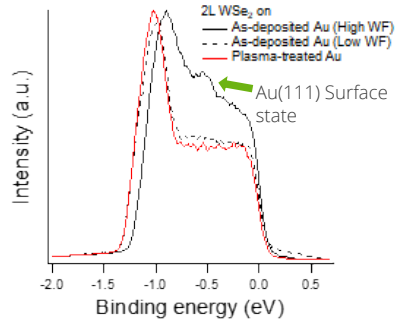






WSe_2





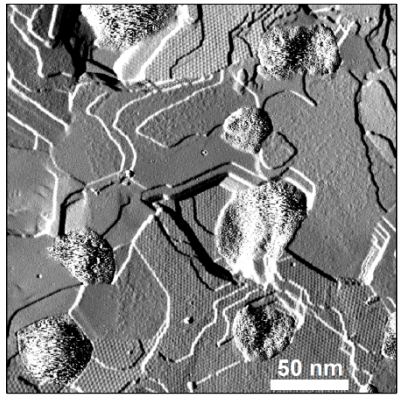
Financial support:

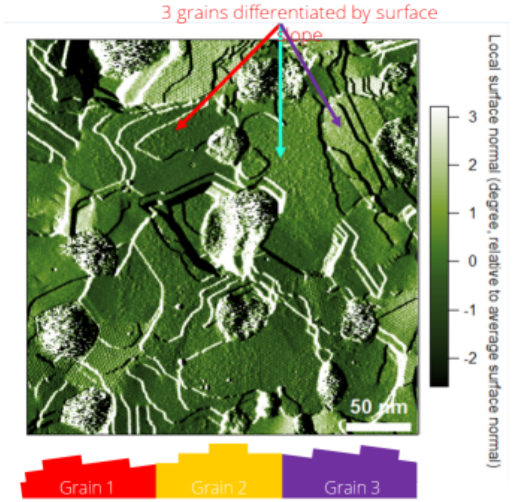
- **Sandia LDRD program**

Special thanks:

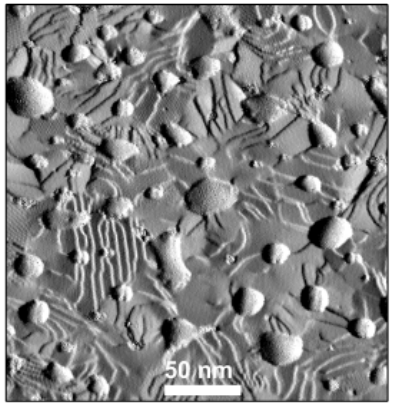
- **N. Bartelt, F. Leonard, R. G. Copeland, Sandia National Laboratories**

WS_2 covered as-deposited Au

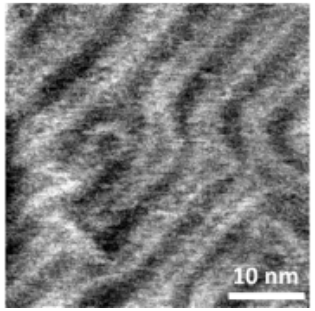


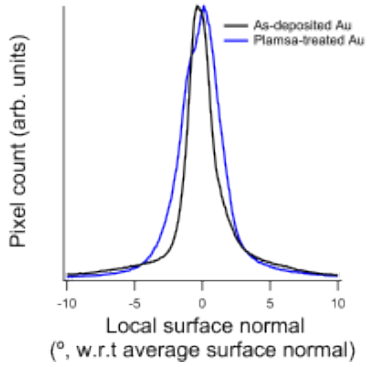


WS_2 covered plasma-treated Au

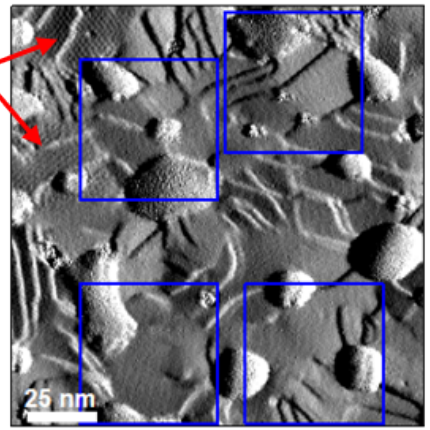


Exposed Au(111) in as-deposited



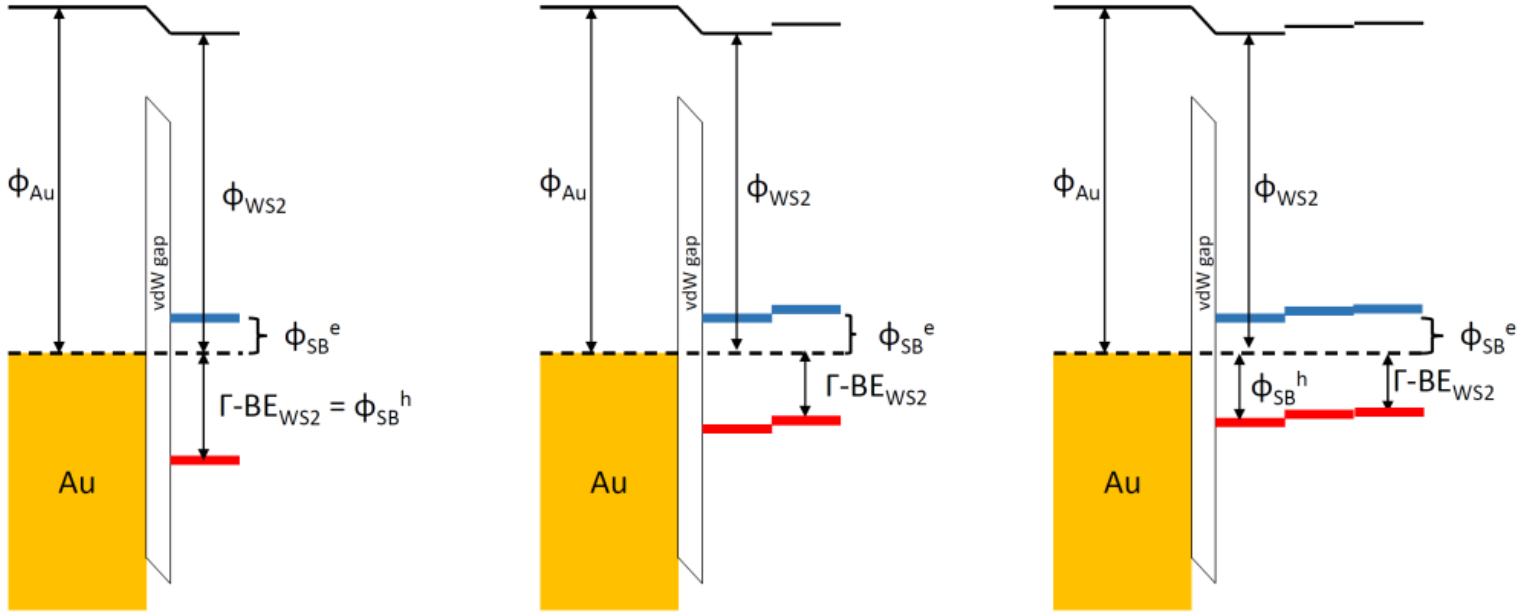


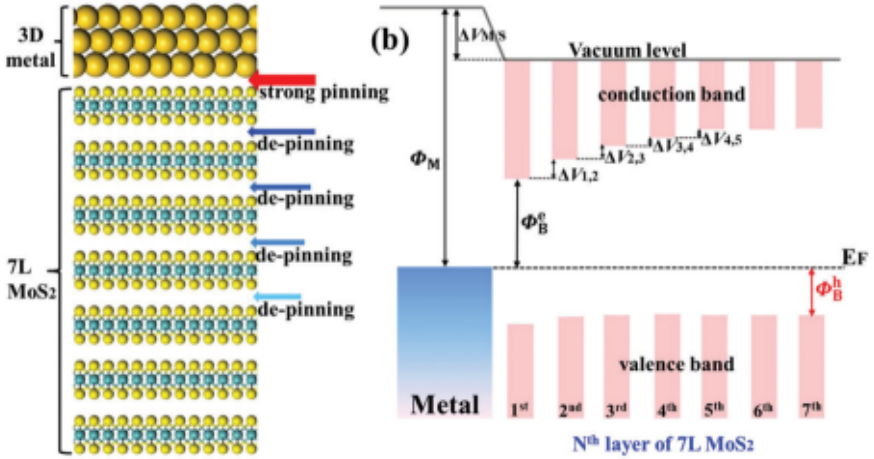
Moirés



Herringbone reconstruction

Model 3: Depinning effect





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