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Electronic Band Alignment Measurement of Individual MoS_2 Flakes Supported on SiO_2 using PEEM with Deep Ultraviolet Illumination

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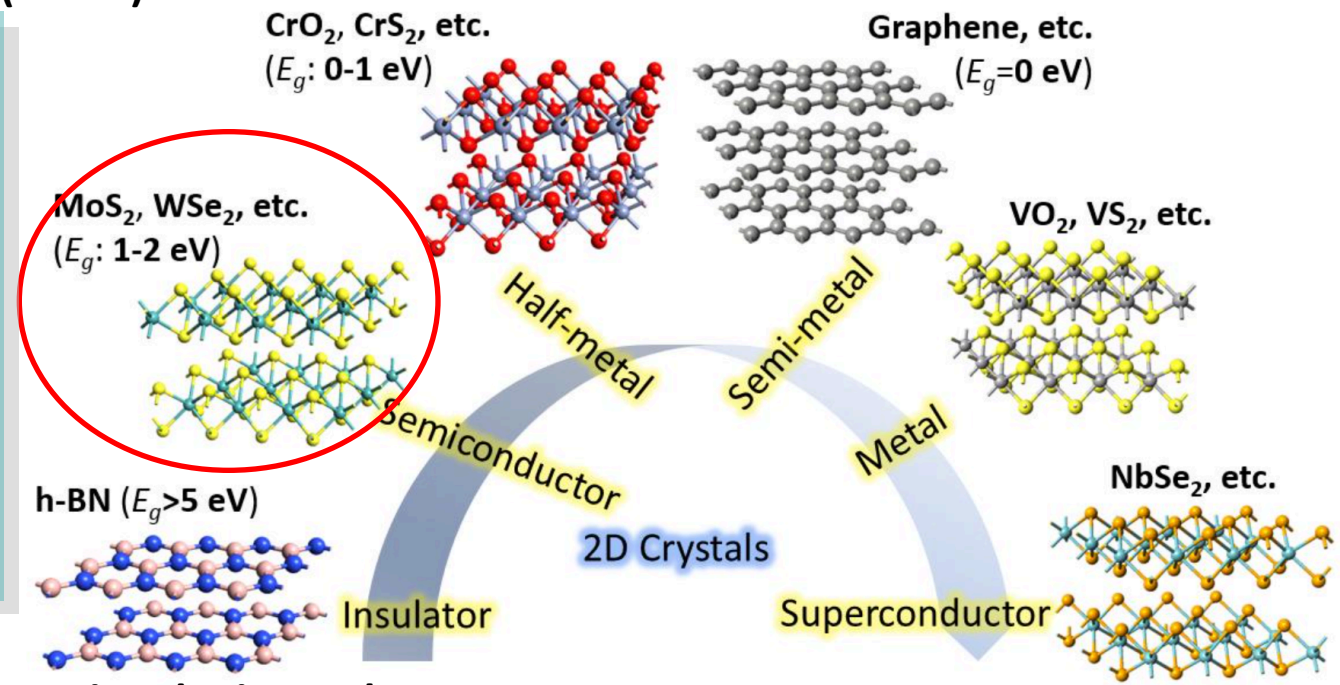
2D semiconductors are promising for semiconductor junctions in devices

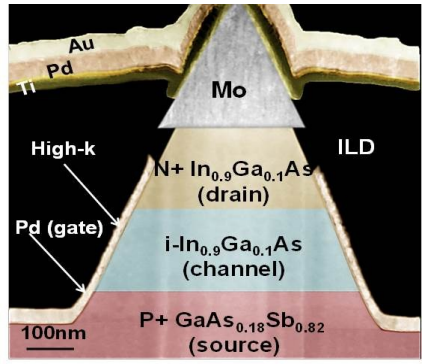
Transition Metal Dichalcogenides (TMDs)

- Tunable band gap
- Gate control of carriers
- Spin and valley degrees of freedom



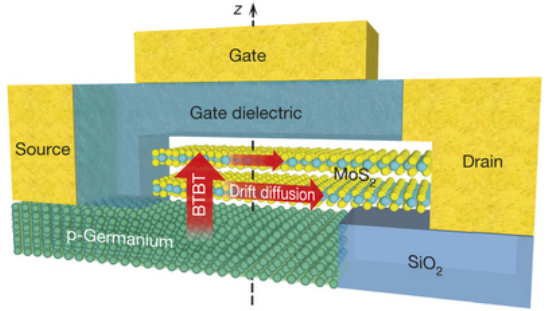
Next-generation devices: Ultra-thin and flexible

GaAs-based tunneling FET*



Junction layers restricted by lattice matching

MoS_2 on p-Ge tunneling FET**



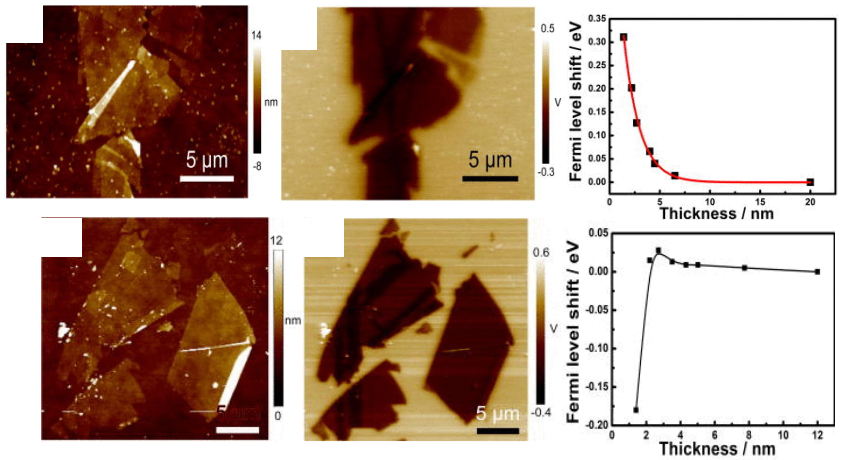
Van der Waals Materials- Lack of covalent bonding

* R. Bijesh *et al.*, *Proc. IEDM Tech. Dig.*, pp. 28.2.1–28.2.4 (2013).

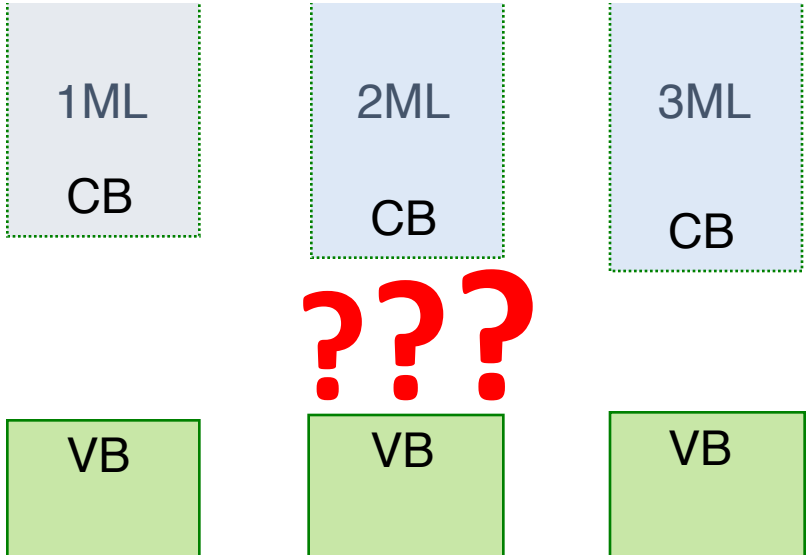
** D. Sarkar, X. Xie, W. Liu, W. Cao, J. Kang, Y. Gong, S. Kraemer, P. M. Ajayan, K. Banerjee, *Nature* **526**, 91 (2015).

The band alignment of MoS_2 layers is an open question

- Open questions
  - Layer-number dependence of band alignment
    - Work function, ϕ, evolution
    - N-type or p-type behavior
  - Interactions with the substrate



Kelvin-probe of unannealed vs. annealed MoS_2*



- Considerations
  - Layer-number dependence
  - Stacking sequence
  - Chemical modification
  - Interactions with the substrate

* Y. Li, C.-Y. Xu, L. Zhen, *Appl. Phys. Lett.* **102** 143110 (2013).

Local photoemission measurements with PEEM (PES) can provide band information

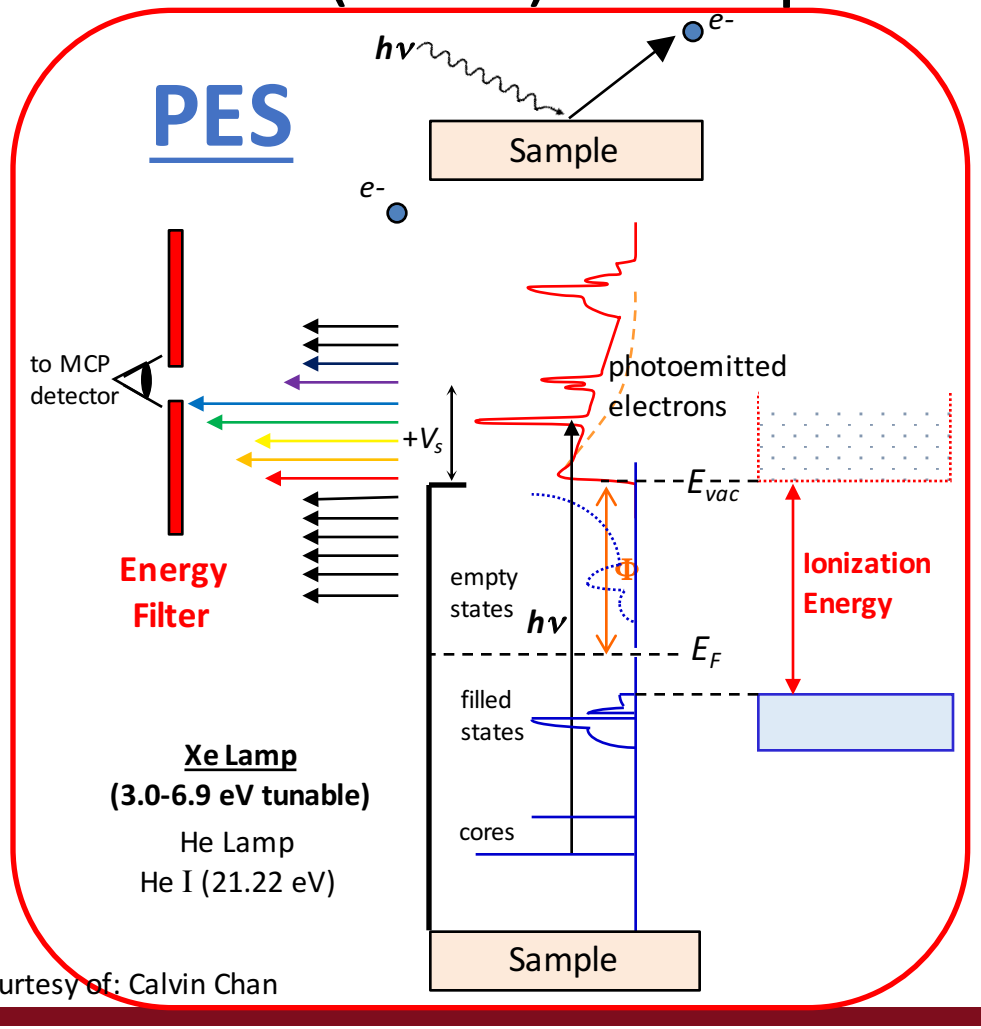
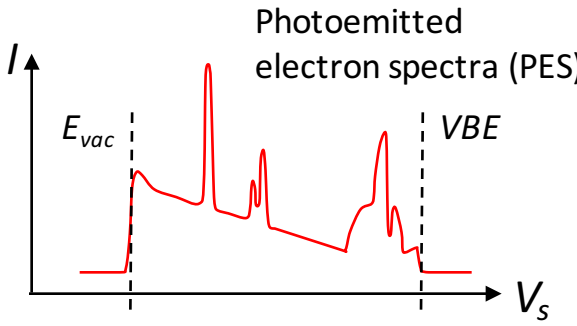
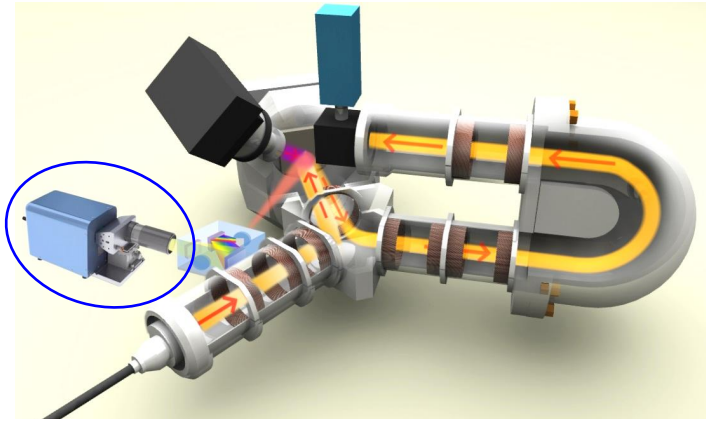
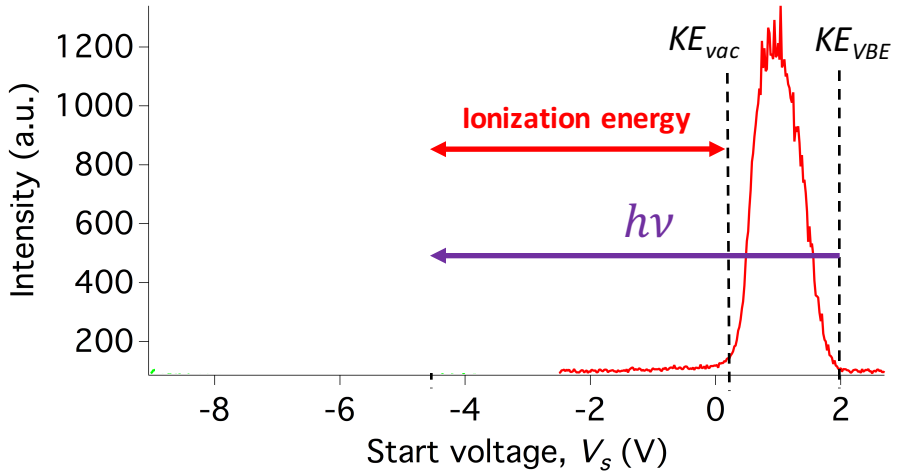


Figure courtesy of: Calvin Chan

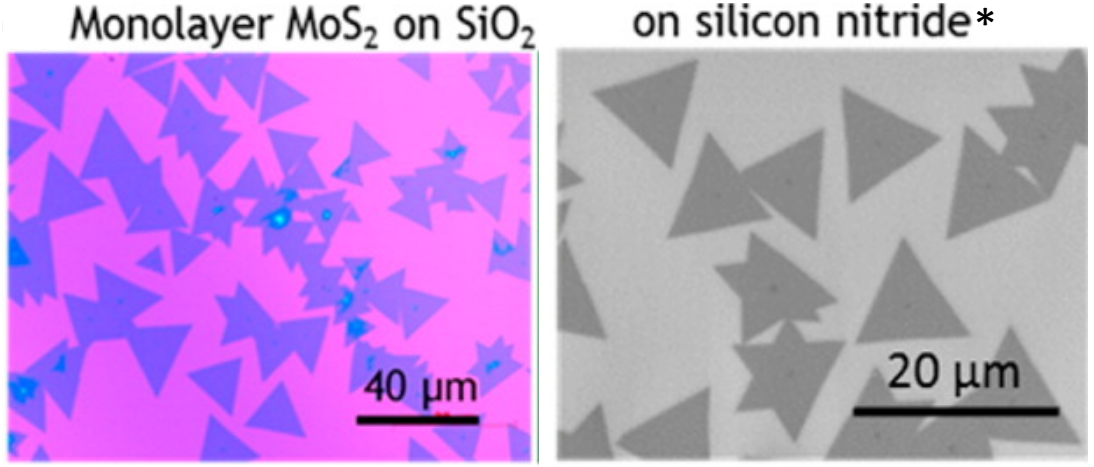


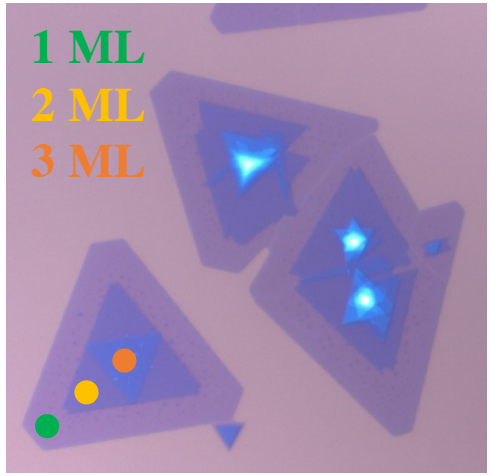


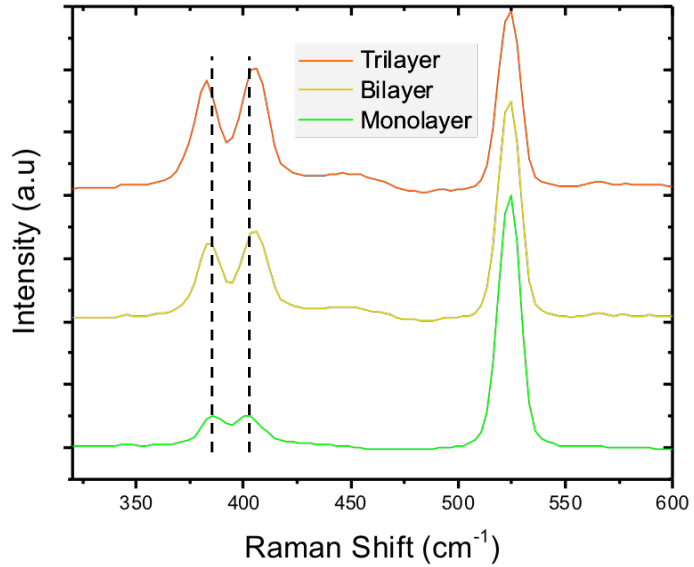
$$BE = KE - h\nu$$



We synthesized CVD TMD samples with mono-, bi- and tri-layer regions



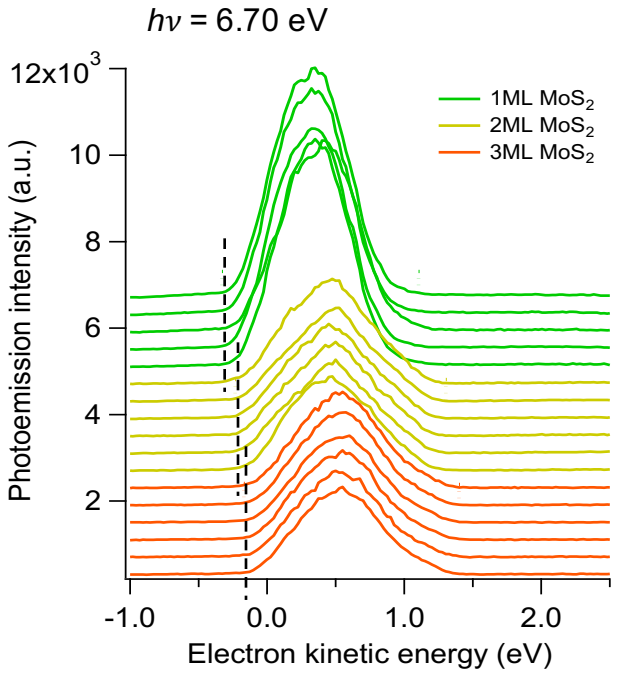


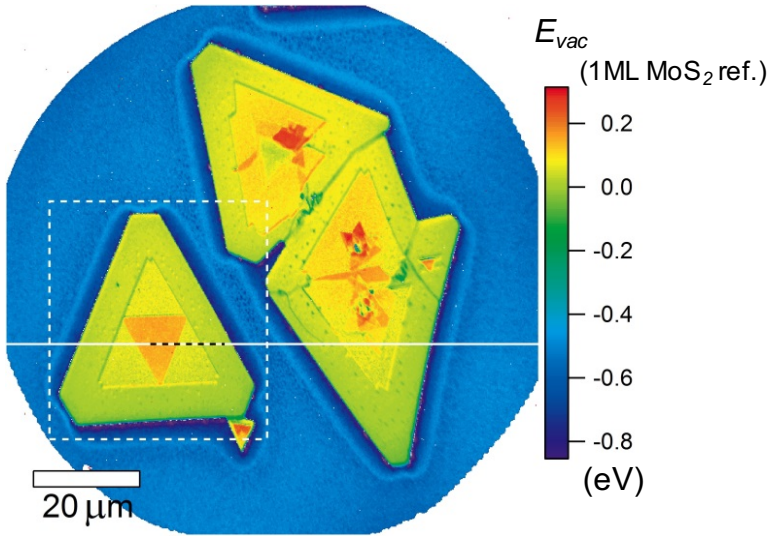


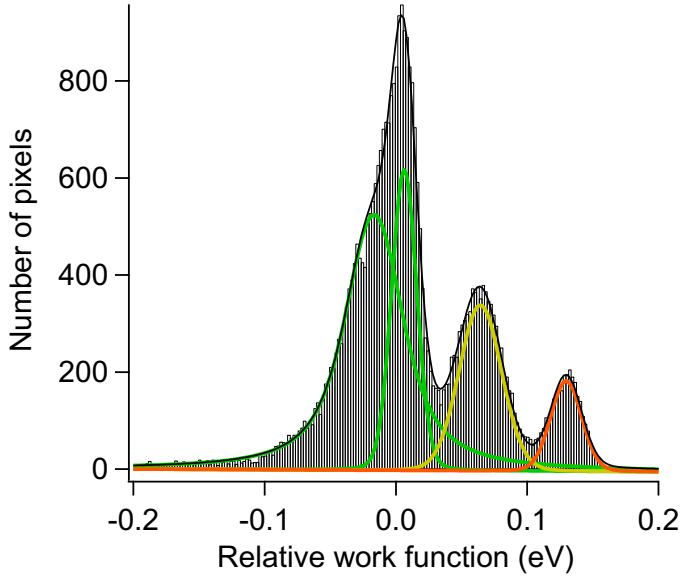
- As-grown samples
- CVD growth is compatible with a range of substrates
- P-type Si / 285 nm SiO_2 / MoS_2
- Thickness verified using the layer-number-dependent Raman shifts

* I. Bilgin, F. Liu, A. Vargas, A. Winchester, M. K. L. Man, M. Upmanyu, K. M. Dani, G. Gupta, S. Talapatra, A. D. Mohite, S. Kar, *ACS Nano* **9** (9), 8822 (2015).

We map layer-dependent vacuum level alignment with PES and determine local work function (ϕ)







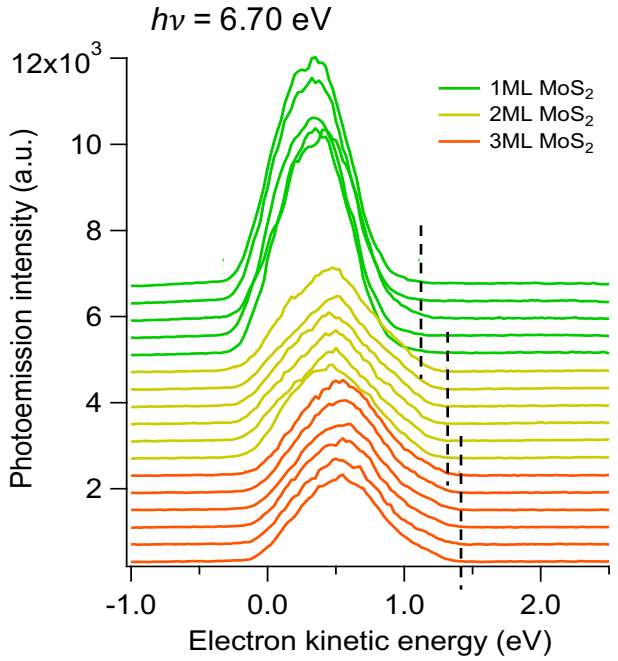
PES leading (left) edge → E_{vac}

- ~70 meV E_{vac} increase per layer

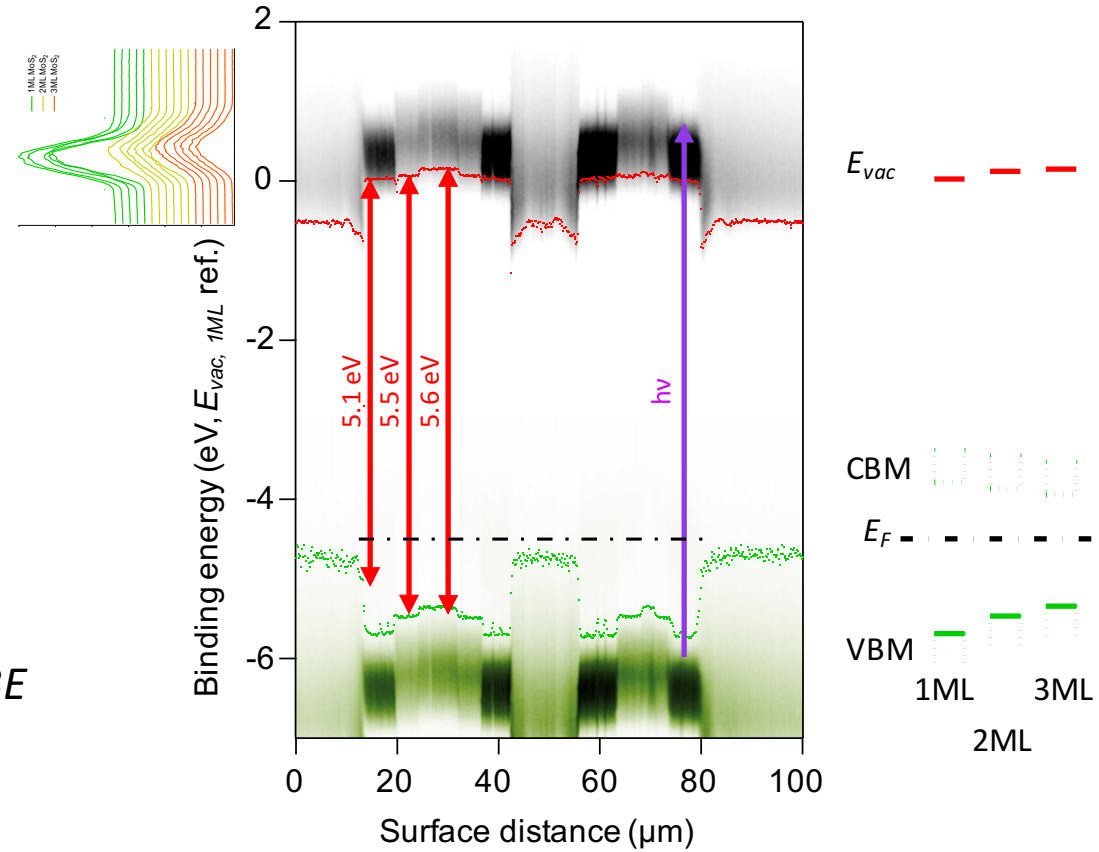
- E_{vac} histogram
  - ϕ_{1ML} = 4.52 eV*
  - ϕ_{2ML} = 4.58 eV
  - ϕ_{3ML} = 4.66 eV

* J.–M. Yun et al., *J. Mater. Chem. C* **1**, 3777 (2013)

We map layer-dependent valence band edge alignment at the *Γ*-point with PES

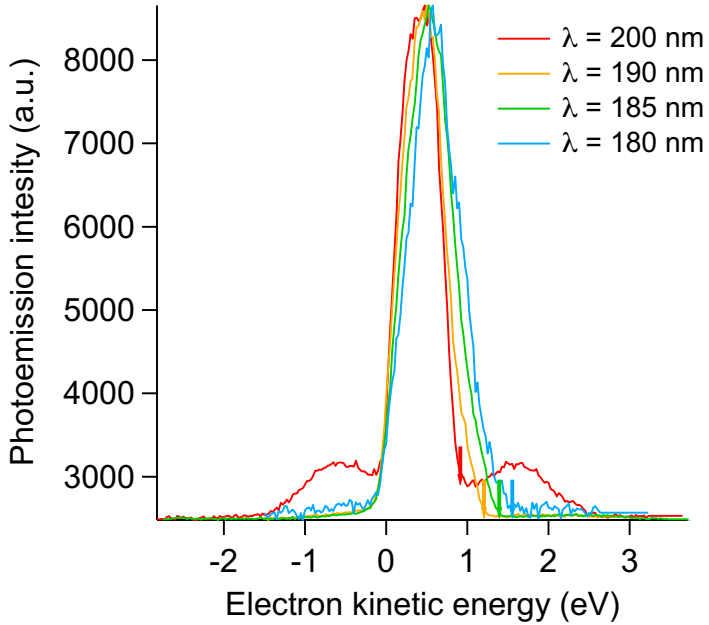


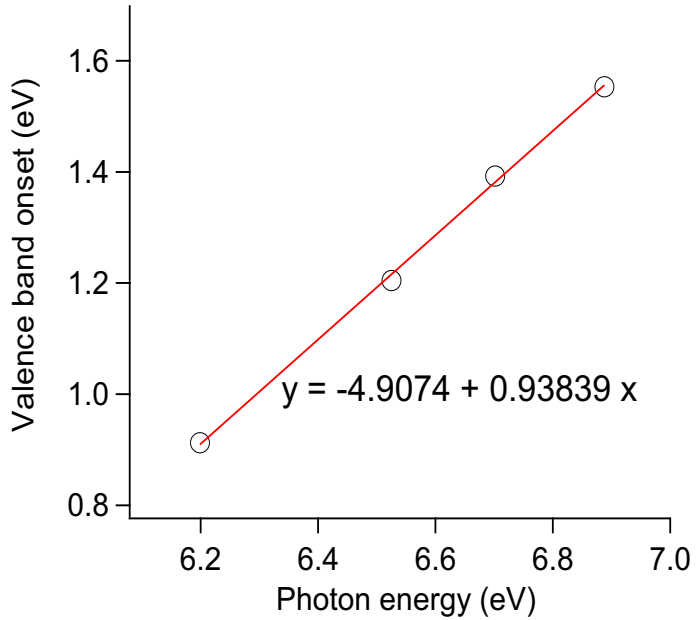
PES trailing (right) edge → *VBE*



- ~150 meV *VBE* increase per layer
- PES experimental band diagram

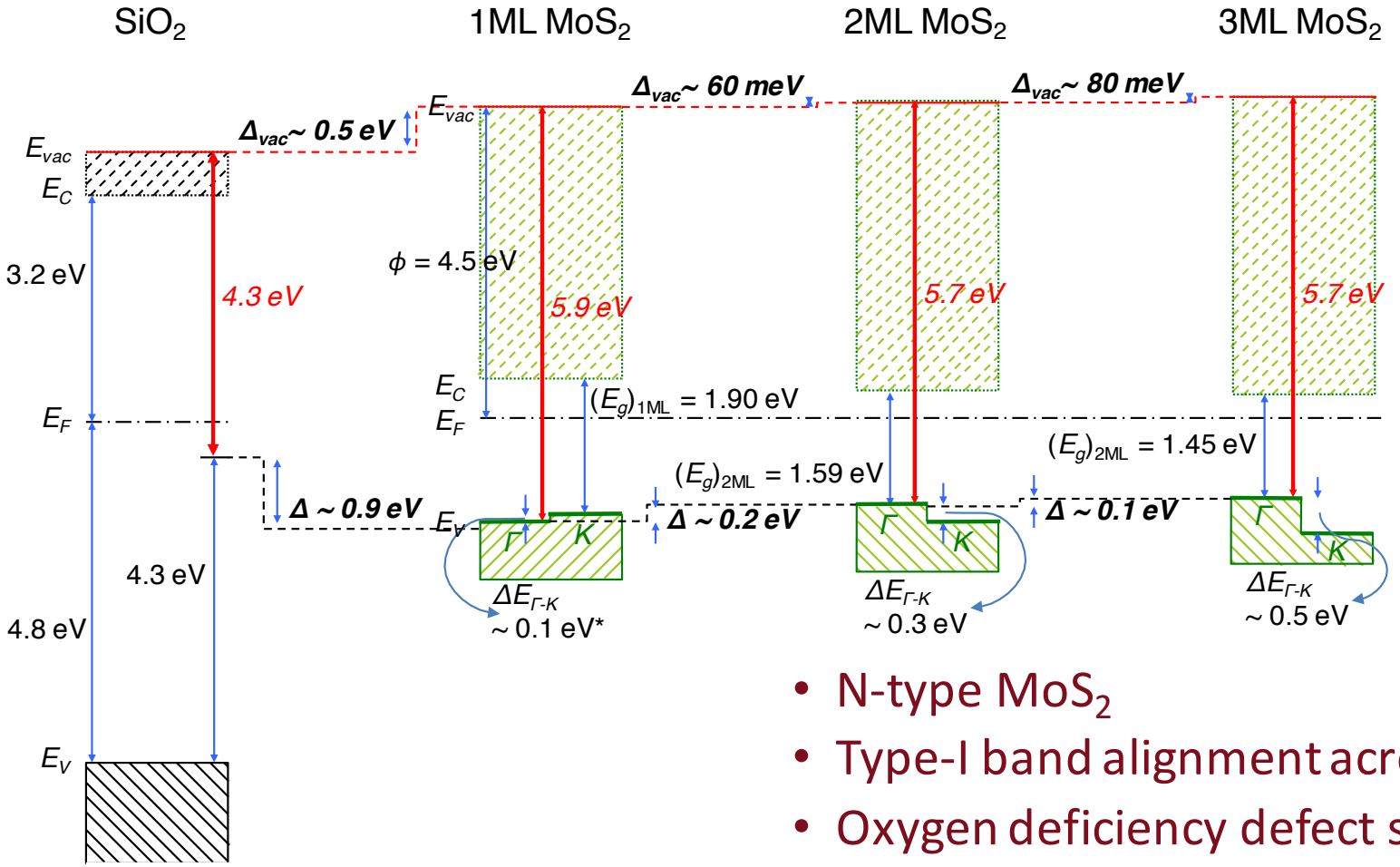
We use a photon-energy series to evaluate substrate contributions and sample charging





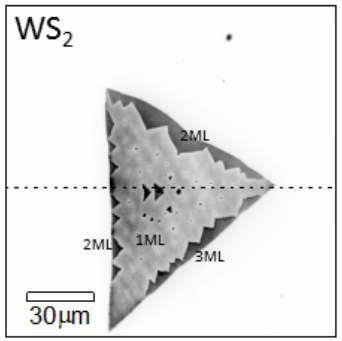
- Substrate contributions, $\lambda > 200$nm
- No obvious sign of charging

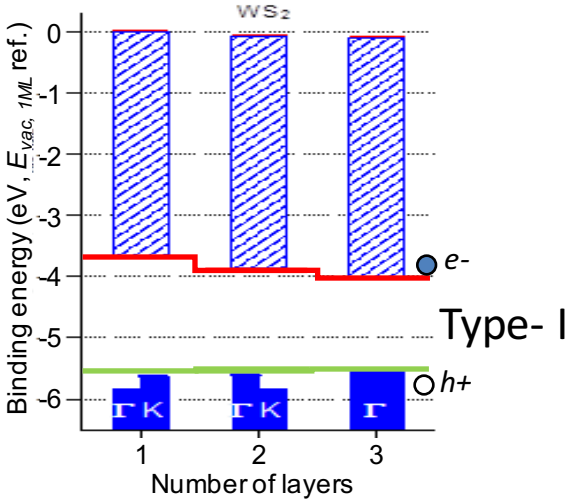
We compile all measured and known info about MoS_2 on SiO_2 into a surface band diagram



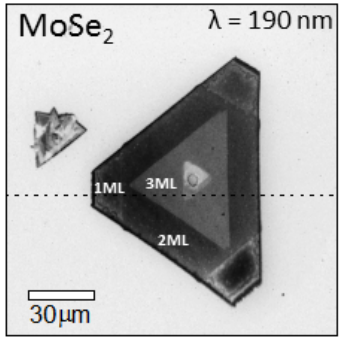
- N-type MoS_2
- Type-I band alignment across lateral junctions
- Oxygen deficiency defect state – charge dissipation

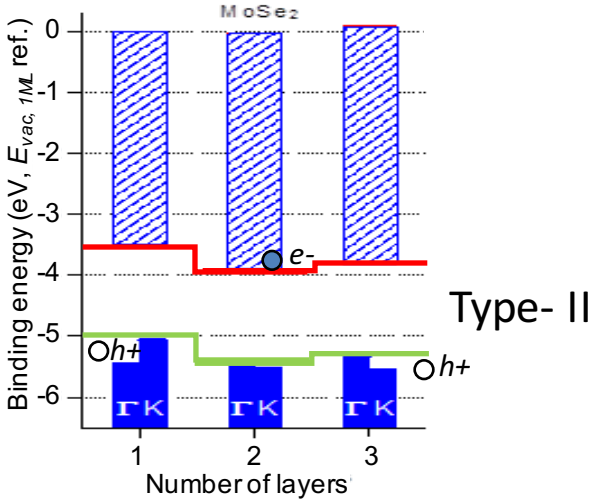
We venture into band alignment measurement of other TMDs





Type- I





Type- II

- WS_2: type-I alignment
- $MoSe_2$: type-II alignment
- Ionization energy decrease
  - MoS_2, to WS_2, to $MoSe_2$
- Type-II band alignment anticipated for heterojunctions comprised of different 1ML TMDs

Conclusions

- Clear layer-dependencies of TMDs
  - Work function increase w/increasing layer number (MoS_2)
  - Ionization energy decrease w/increasing layer number (MoS_2, WS_2, $MoSe_2$)
  - Type-I band alignment (MoS_2, WS_2)
  - Type-II band alignment ($MoSe_2$)
- Absolute surface band diagrams will inform heterojunction design
- DUV-PEEM: a new metrology for 2D crystals research

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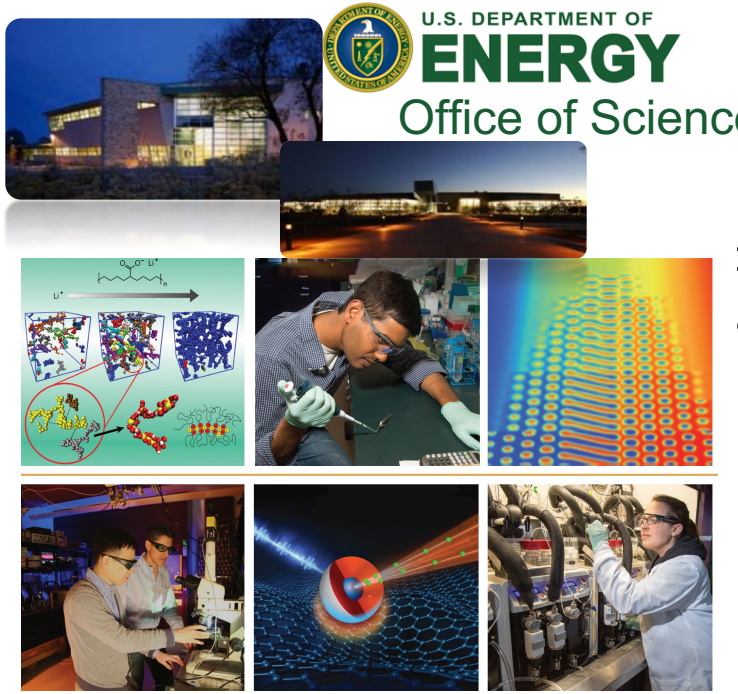

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