Electronic band alignment of few-layer transition metal dichalcogenides on SiO_2

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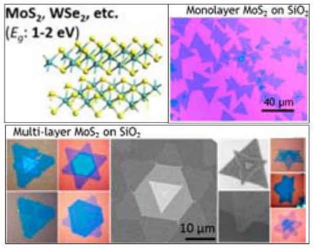
Introduction



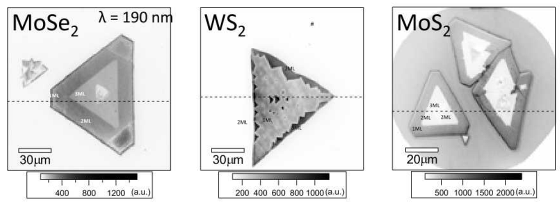
2D layers of transition metal dichalcogenides (TMDs) are a proposed alternative to conventional semiconductors for constructing junctions in next-generation devices. MoS_2, WS_2, $MoSe_2$, model TMDs, show favorable properties for electronic and optoelectronic applications including high on-off ratios, high photoluminescence quantum efficiency, high photogain and photoresponse. The open experimental questions we address in this study, relevant for device performance and considerations, include:

- Evolution of work function, ϕ, with layer number
- Doping of TMD on SiO_2
- The band alignment among MoS_2, WS_2, & $MoSe_2$

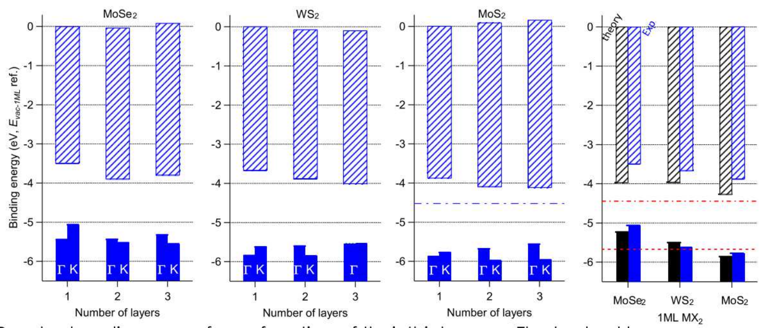
So far, environmental effects (chemical modification due to exposure, substrate interactions) have prevented a clear, systematic understanding of these properties via experiment.

Band alignment of MoS_2, WS_2, & $MoSe_2$: a complete picture

The ionization energy displays a progressive decrease from MoS_2, to WS_2, to $MoSe_2$, in agreement with reported density functional theory calculations. We deduce that a heterojunction comprising any of the three TMD monolayers would form a staggered (type-II) band alignment in the absence of the interlayer coupling.



PEEM intensity images of $MoSe_2$, WS_2, and MoS_2 acquired at 0.1 eV below E_{vac} of the 1 ML area. Higher photoemission intensity is shown as darker gray.

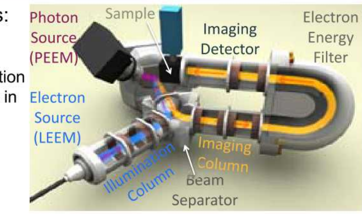


Band edge diagrams of as a function of their thicknesses. The hashed boxes represent the conduction bands, and the solid boxes represent the valence bands. The right figure compares experimentally determined band diagrams to those of DFT modeling.

Local Photoemission Measurement using Spectroscopic PEEM

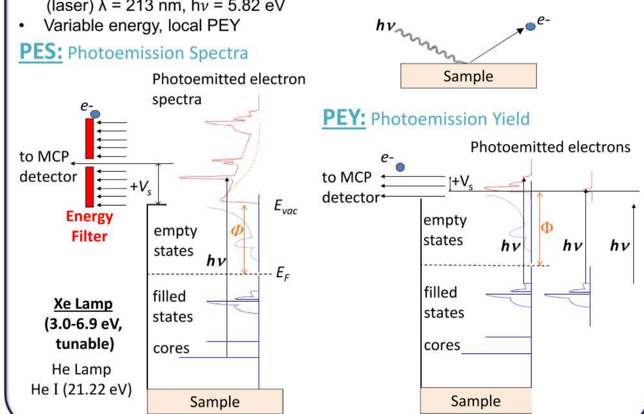
Spectroscopic PEEM features:

- Spatial resolution: 10 - 15 nm
- Spectral resolution: 200 meV
- Electron/Photon/Thermal excitation
- Real-time imaging of processes in situ

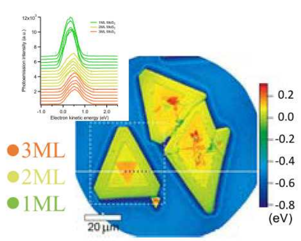


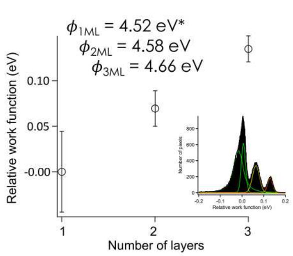
Two modes of photoemission measurement:

- Fixed energy, local PES (laser) λ = 213 nm, $h\nu$ = 5.82 eV
- Variable energy, local PEY



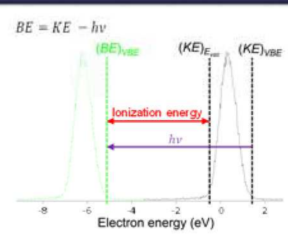
PES & PEY measurements of layer-dependent work function & ionization energy for MoS_2

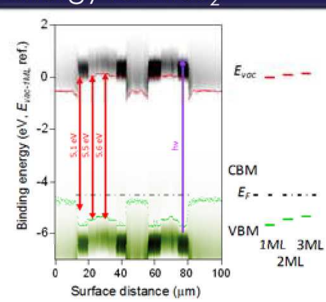




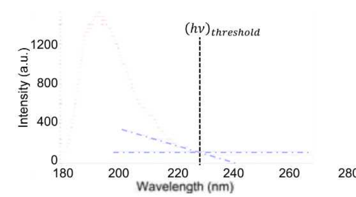
* J.-M. Yun, et al., *J. Mater. Chem. C* **1**, 3777 (2013).

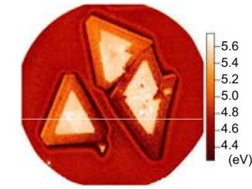
Relative vacuum level (E_{vac}) and valence band edge increase with each additional layer, establishing a layer-dependent surface potential and work function increase with increasing layer number.





We obtain the binding energy of the valence band edge relative to the vacuum level and construct an experimental band diagram that shows the local ionization energy measured with PES.





Despite agreement in PES and PEY results for I_{3ML}, there are disparities for I_{1ML} and I_{2ML} (see red bidirectional arrows in the experimental band diagram). Considering the mechanisms that differ between the two modes of photoemission measurement, we rely on our PES results for a more accurate determination of ionization energy.

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