Strain Engineering of Transition Metal Dichalcogenides: A Combined First-Principles and Experimental Study

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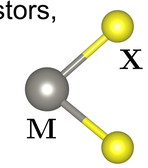
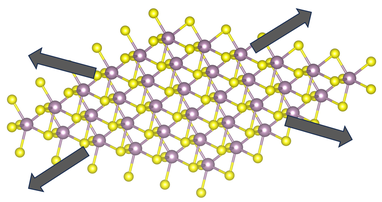
Background

- Transition metal dichalcogenides (TMDs) are a promising class of **2D materials** with use cases for novel transistors, optoelectronics, and sensors[1]
  - Monolayer group-VI TMDs follow an **MX_2** structure with M={Mo, W}, X={S, Se, Te} and are direct bandgap semiconductors

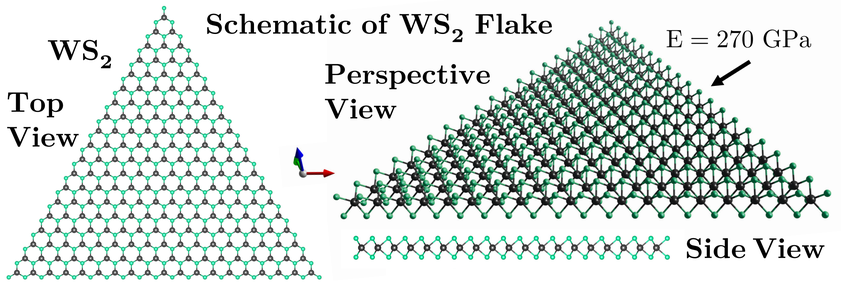


- TMDs are sensitive to **strain**, **alloying**, and **electric fields,** allowing precise, continuous modulation of optical and electronic properties like bandgap[1]



Straining 1H MoS_2

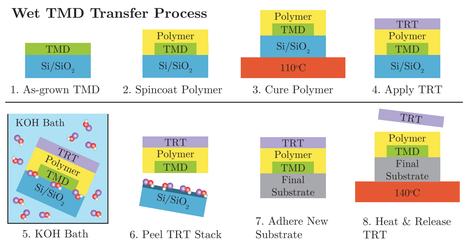
- Strain engineering is promising due to TMDs high **fracture resistance**[2]
- Prior studies report highly inconsistent strain tunability, often with slippage or fracture, motivating further study[2]



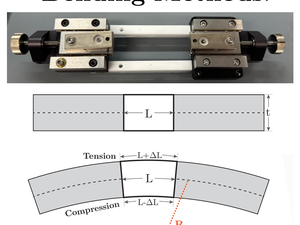
Methods

- Biaxial strains from -10% to +10% are simulated on monolayer MoS_2, $MoSe_2$, $MoTe_2$, WS_2, and WSe_2 using VASP-based density-functional-theory (DFT) with the PBE functional
- Monolayer WS_2 flakes are fabricated with CVD on a Si/SiO_2 substrate, transferred to a polymer substrate using a wet-transfer procedure for straining

Wet TMD Transfer Process

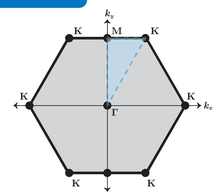


Uniaxial Stretching and Bending Methods:

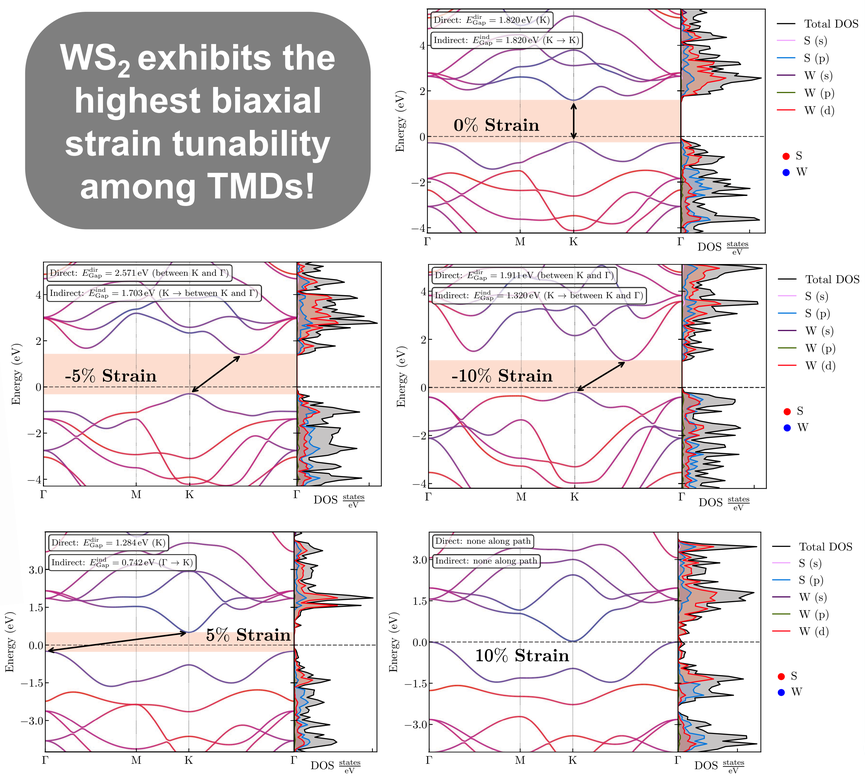


Computational Results

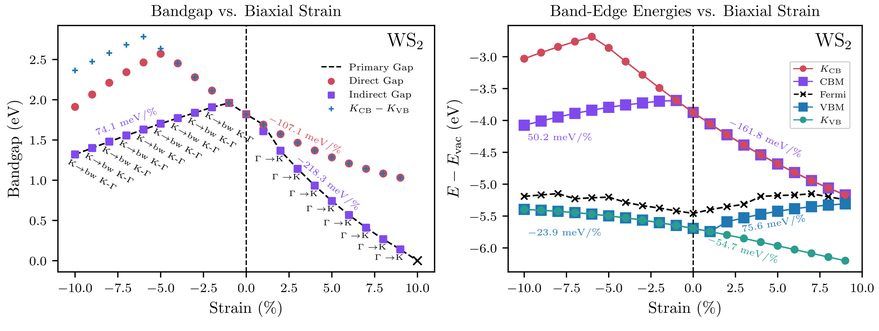
- Relaxed TMD unit-cells are attained using the PBE functional (far right); then, biaxial strain is applied and the atomic positions are relaxed
- Band structure calculations are performed over the irreducible Brillouin zone (right, in blue)
- Strained band structure plots for WS_2 are shown below:

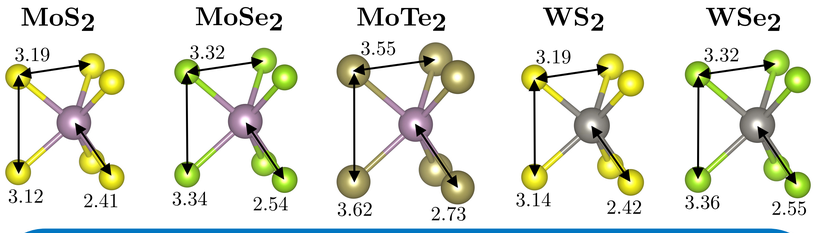


WS_2 exhibits the highest biaxial strain tunability among TMDs!



- There is a direct to indirect bandgap transition under small strains
- The bandgap narrows under both compressive and tensile strains, with highest tunability of **-218.3 meV/%** under large tensile strains
- The **CBM** shows significantly greater strain-tunability than the VBM

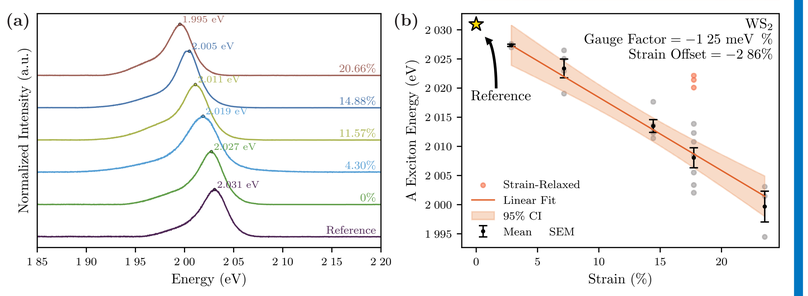




Experimental Results

- Small **uniaxial tensile strains** have a predicted **-53 meV/%** bandgap tunability (roughly half the computed biaxial result)[3]
- Photoluminescence spectroscopy is used to measure the optical bandgap of WS_2 flakes under strain





- Under uniaxial tensile stretching on PDMS, there is a maximum A-exciton shift of **-36 meV** with a low tunability rate of **-1.25 meV/%** (above) due to poor strain transfer (~10%)
- A higher **-48.3 meV** shift is achieved through bending on PET, with a maximal tunability rate of **−19.62 meV/%**
- Poor strain transfer efficiency causes low tunability rates, yet the maximum tunability exceeds results of many past studies![4]

Conclusion

- Sulfide-based TMDs demonstrate consistently larger strain tunability compared to their selenide and telluride counterparts
- WS_2 shows promise with the highest strain tunability, especially for devices operating under tensile strains with e^- carriers
- **Strain transfer limitations are the limiting factor in experimental strain engineering: substrate optimization is necessary to improve experimental strain tunability**

References: **[1]** Samadi *et al.* (2018) *Nanoscale Horizons*, 90–204. **[2]** Peng *et al.* (2020) *Light: Science & Applications*, 190. **[3]** Oliva *et al.* (2022) *ACS Applied Materials & Interfaces*, 19857–19868. **[4]** Rodríguez *et al.* (2024) *Journal of Physical Chemistry Letters*, 6355–6362.