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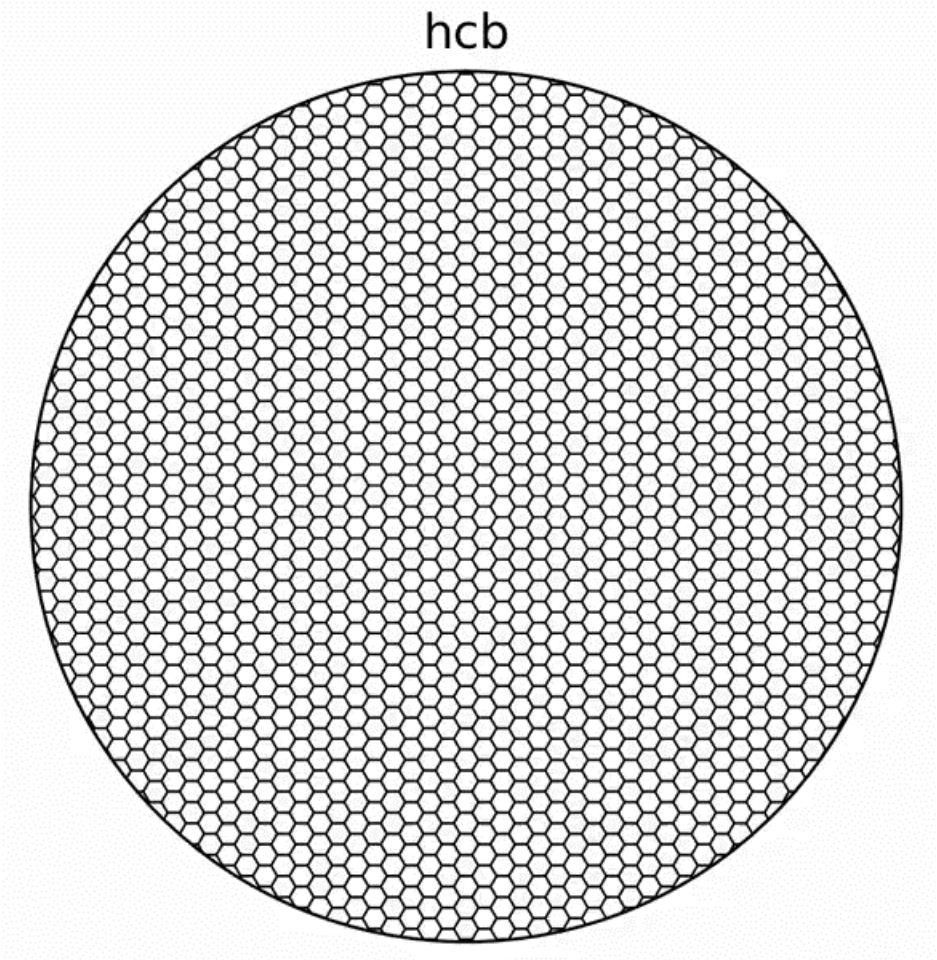
Relaxation effects in twisted transition metal dichalcogenide heterostructures

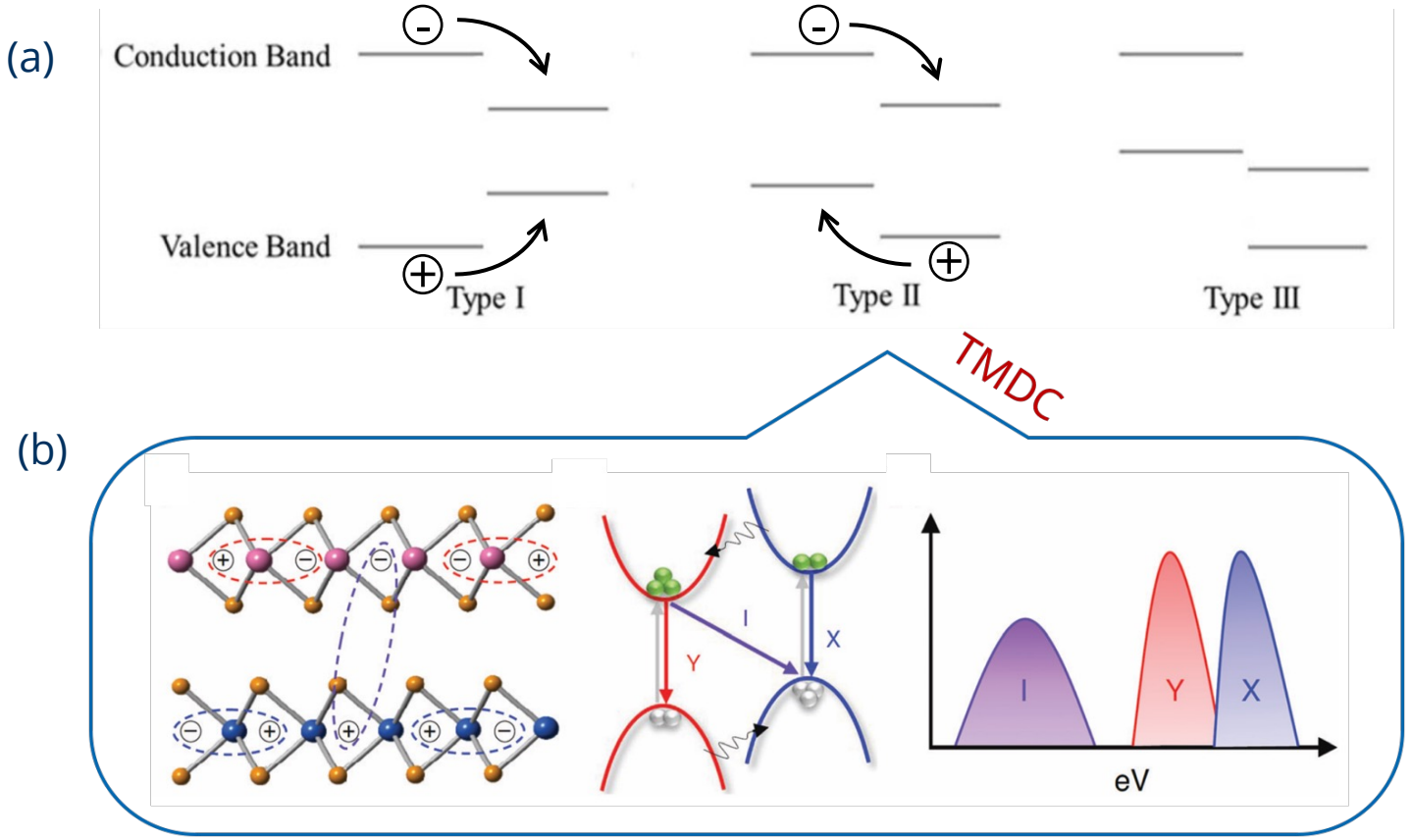
Sep 25, 2023 // Prague

Why twisted: Moiré



Florian Arnold. (July 5, 2022). Moiré clocks (Video File). Retrieved from https://www.youtube.com/watch?v=uxoKzBPFbrs&list=PL2LyfOO_UvEx5pfKmWIii_hmRBmZwJVG6&index=16.

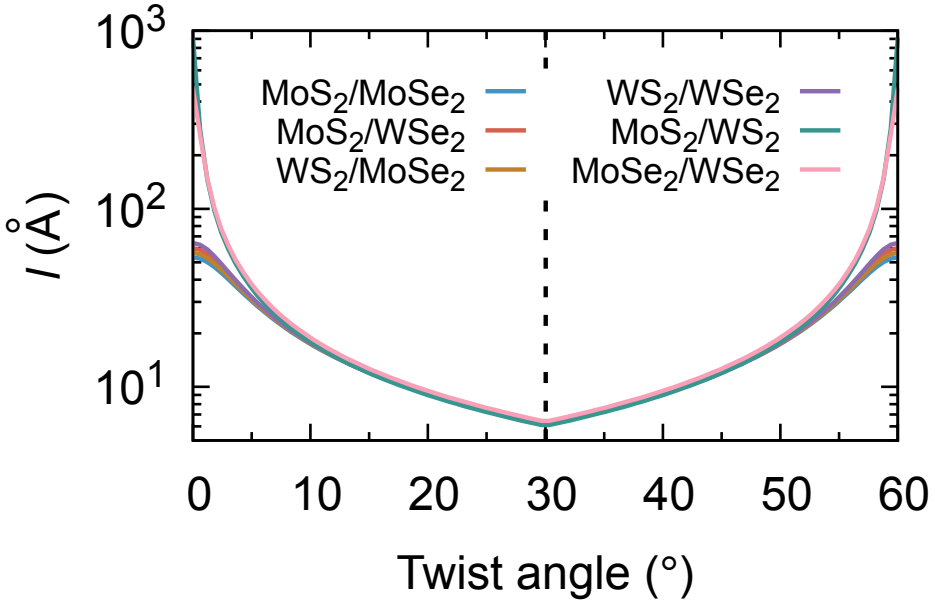
Why TMDC heterostructures?



(a) Different types of vdWH band structures.[1] (b) Excitonic behavior in Type-II alignment vdWH.[2]

[1] *Nat. Phys.* **2021**, *17*, 92.
[2] *J. Phys. D: Appl. Phys.* **2021**, *54*, 053001.

How: Multiscale approach



- Geometry optimization performed by Force-Field[1][2]
- Electronic properties calculated by DFTB[3]

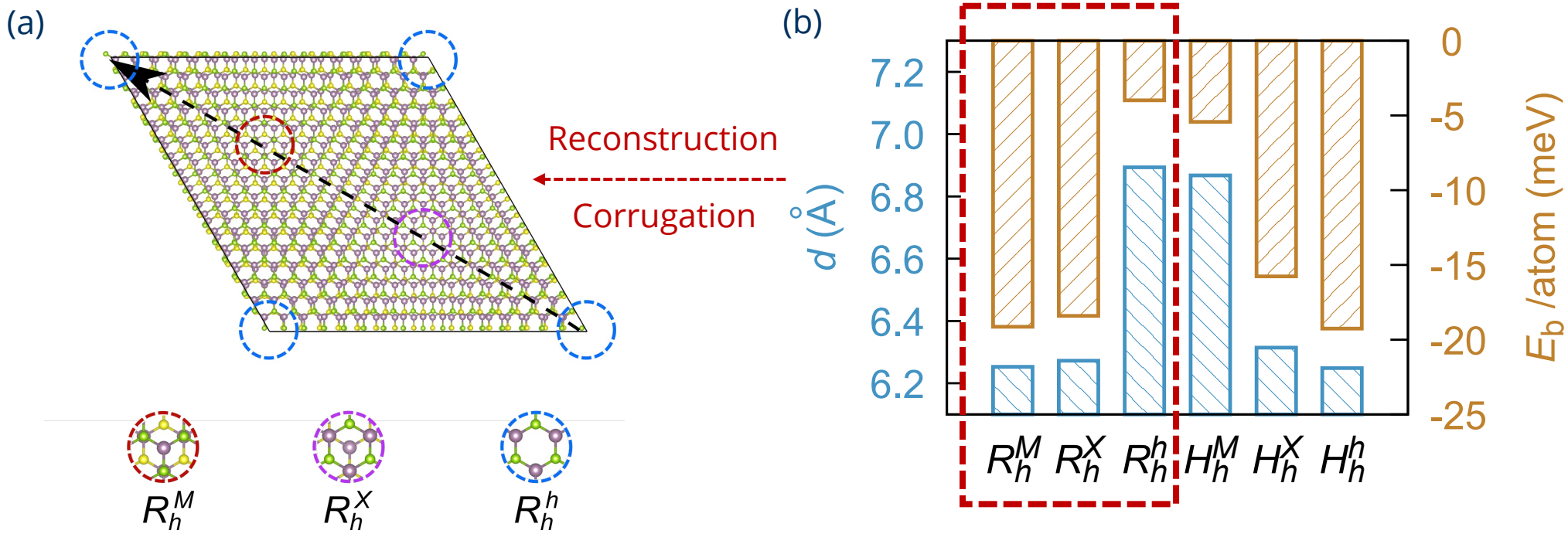
- Lattice size: 10^1 – 10^3 Å
- Number of atoms: 50 – 5×10^5

[1] *J. Appl. Phys.* **2013**, *114*, 064307.

[2] *J. Phys. Chem. C* **2019**, *123*, 9770.

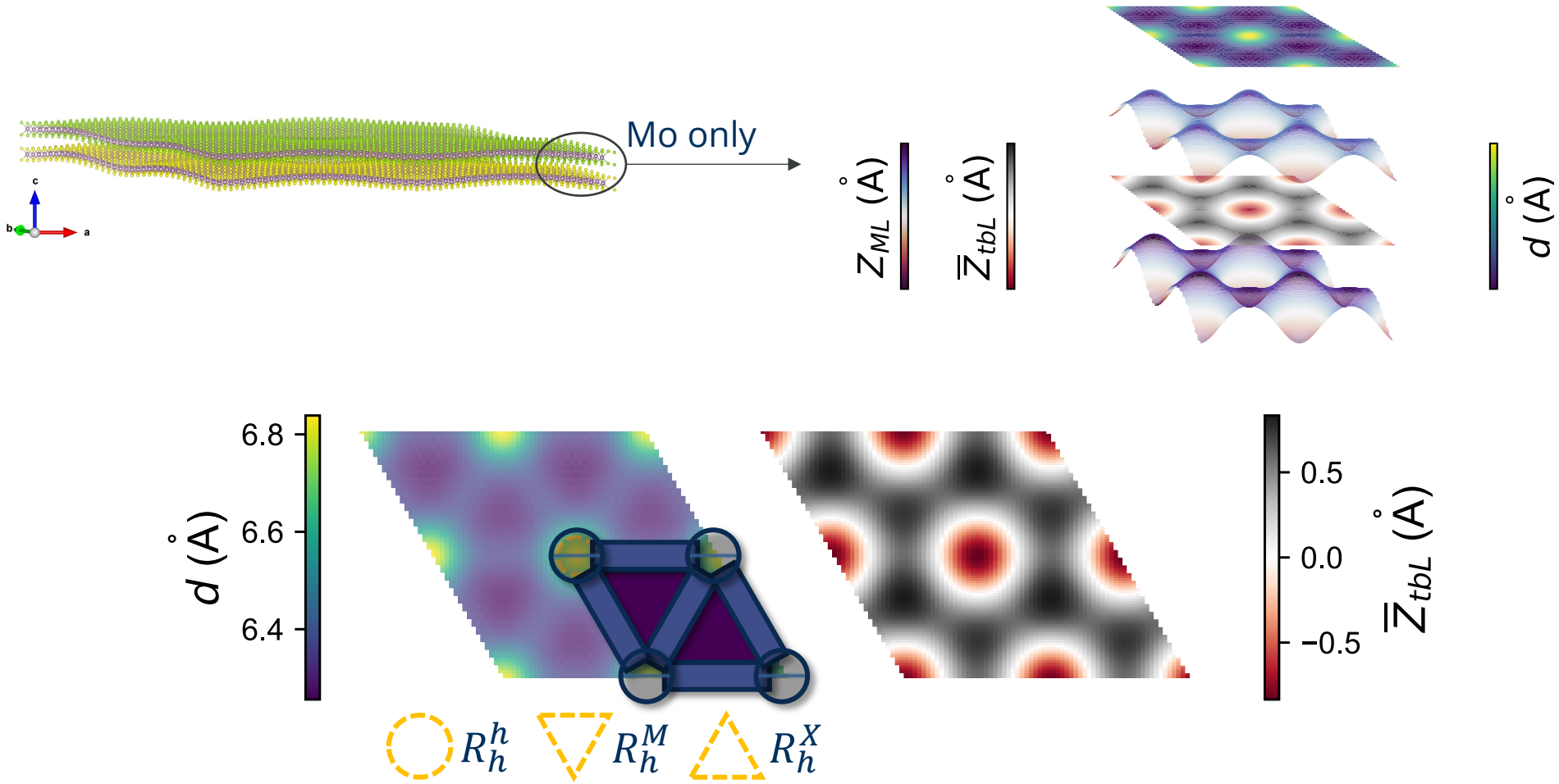
[3] *J. Chem. Theory Comput.* **2022**, *18*, 4472.

R-type stackings – Unrelaxed $MoS_2/MoSe_2$ at 0°

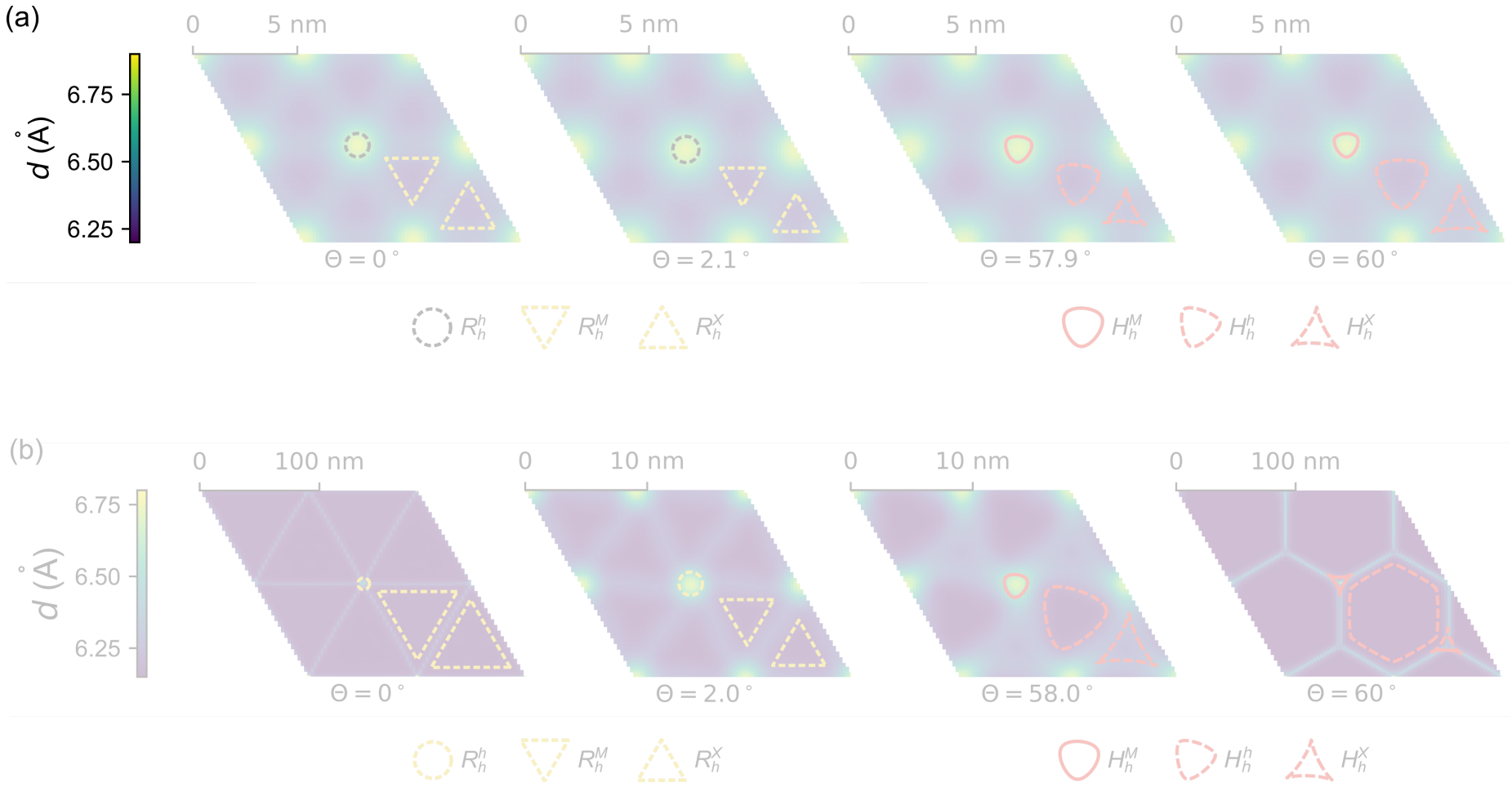


(a) 3 high-symmetry stackings and transition stacking regions. (b) Interlayer distance and binging energy of corresponding high-symmetry stackings.

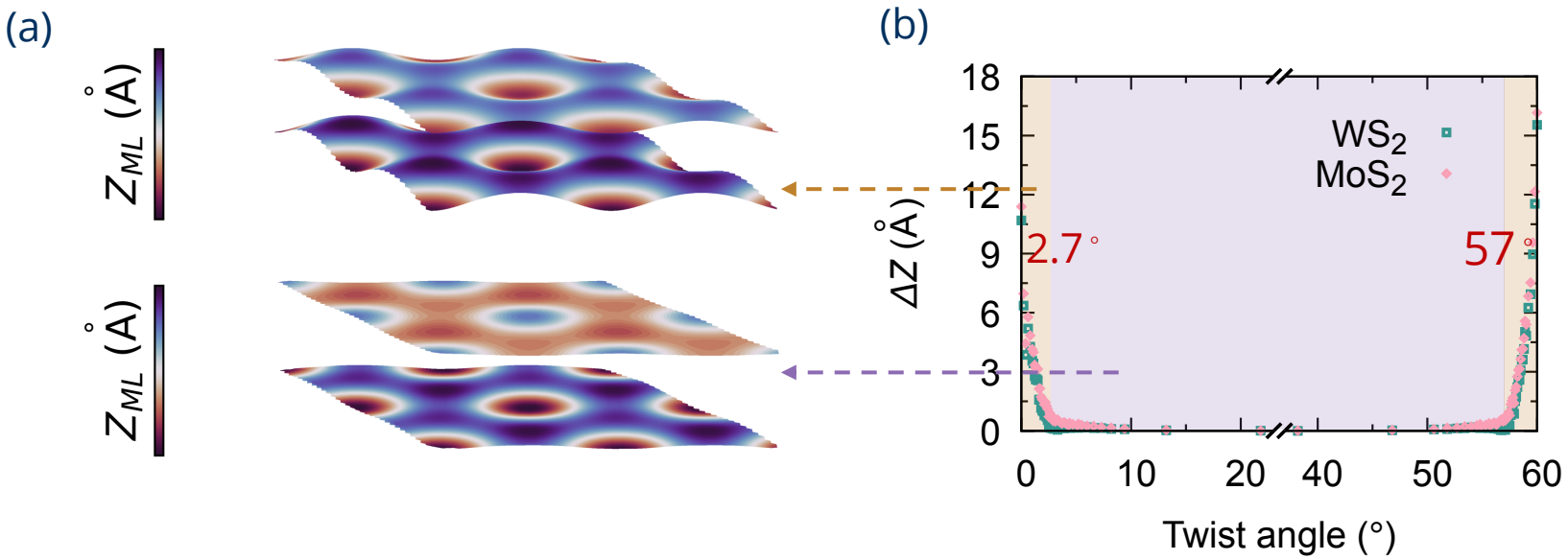
Relaxed $MoS_2/MoSe_2$ at 0°



$MoS_2/MoSe_2$ and MoS_2/WS_2 at small twist angles

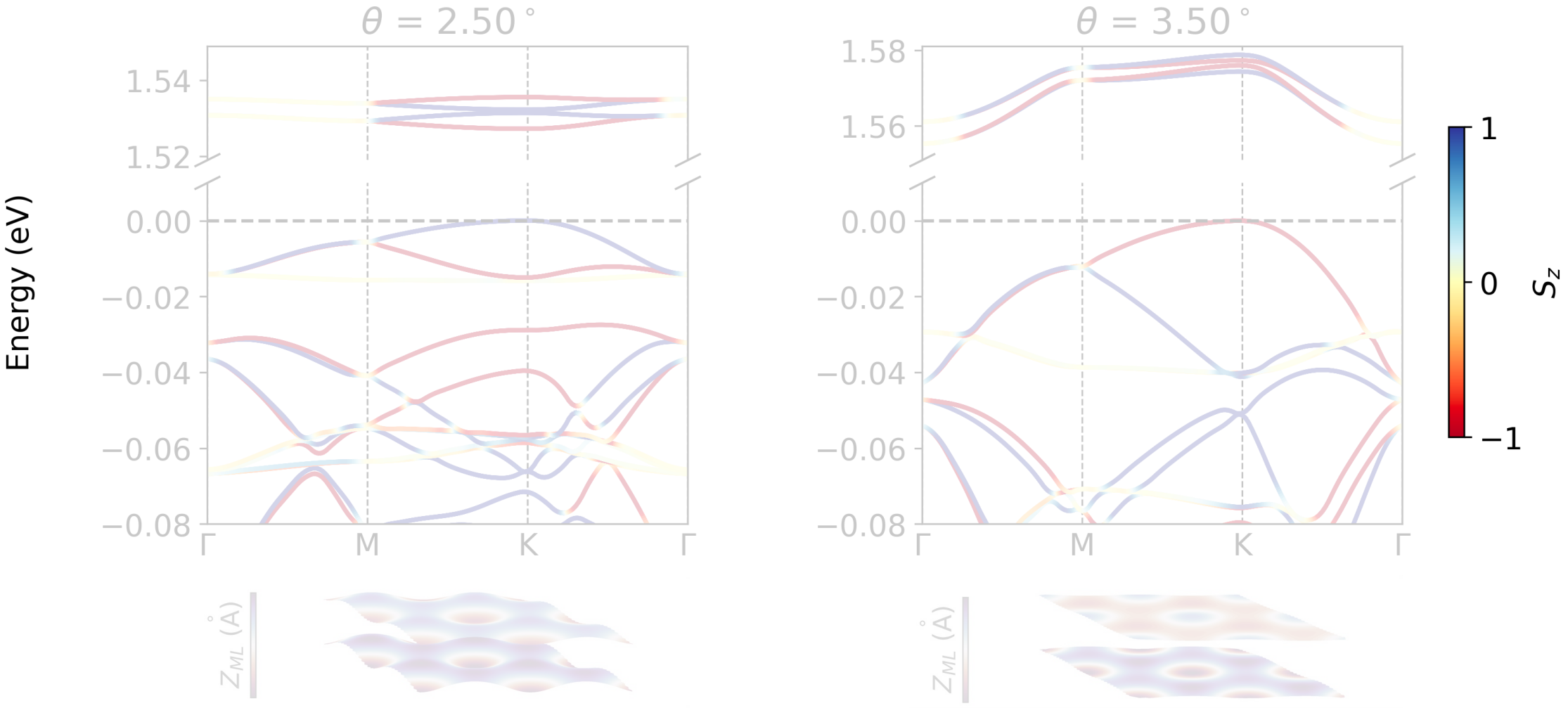


Small vs. large twist angles in MoS_2/WS_2

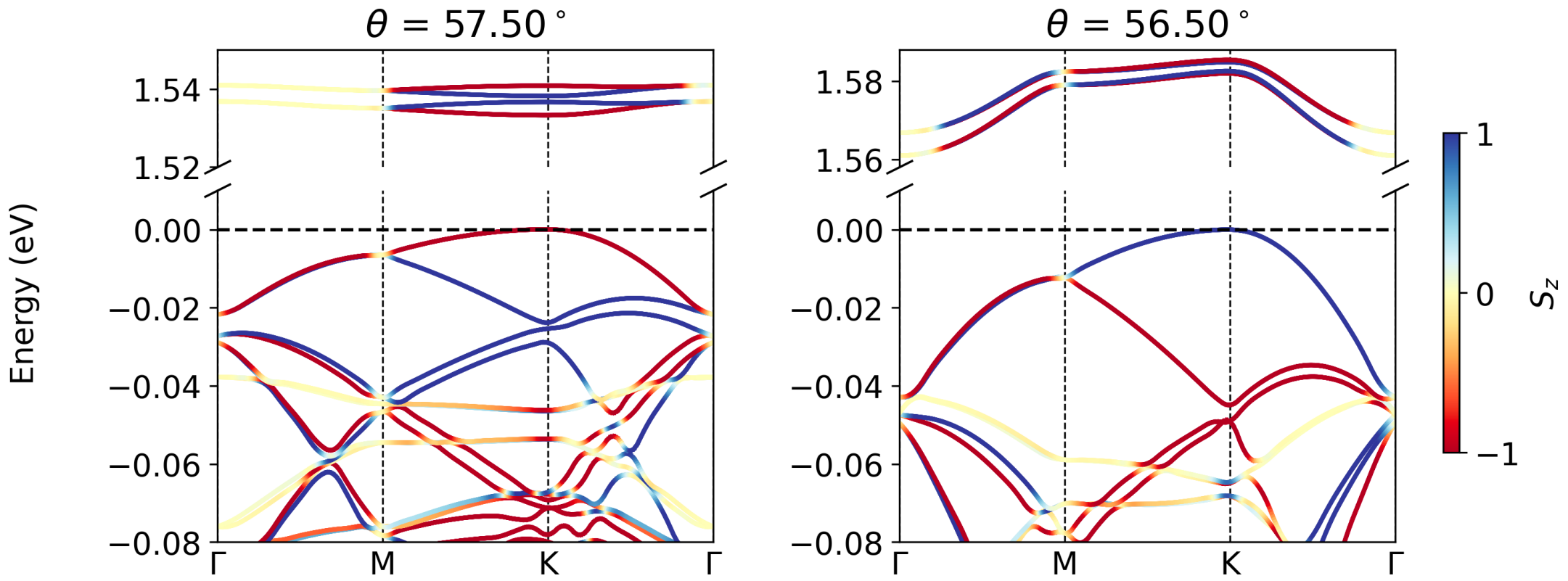


- (a) Out-of-plane corrugation of Mo. (b) Magnitude of corrugation of each layer.

MoS_2/WS_2 at 2.5° and 3.5°

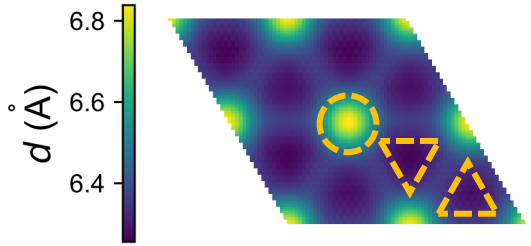


MoS_2/WS_2 at 57.5° and 56.5°

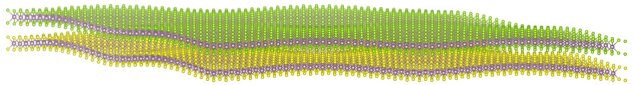


Summary

- Significant lattice reconstruction
  - Domain formation



  - Out-of-plane corrugation



  - Depending on the twist angle

- Corrugation-dependent spin orbital coupling effect

Acknowledgement

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- M.Sc. Gautam Jha
- Antje Völkel
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- All the members in 2EXCITING network

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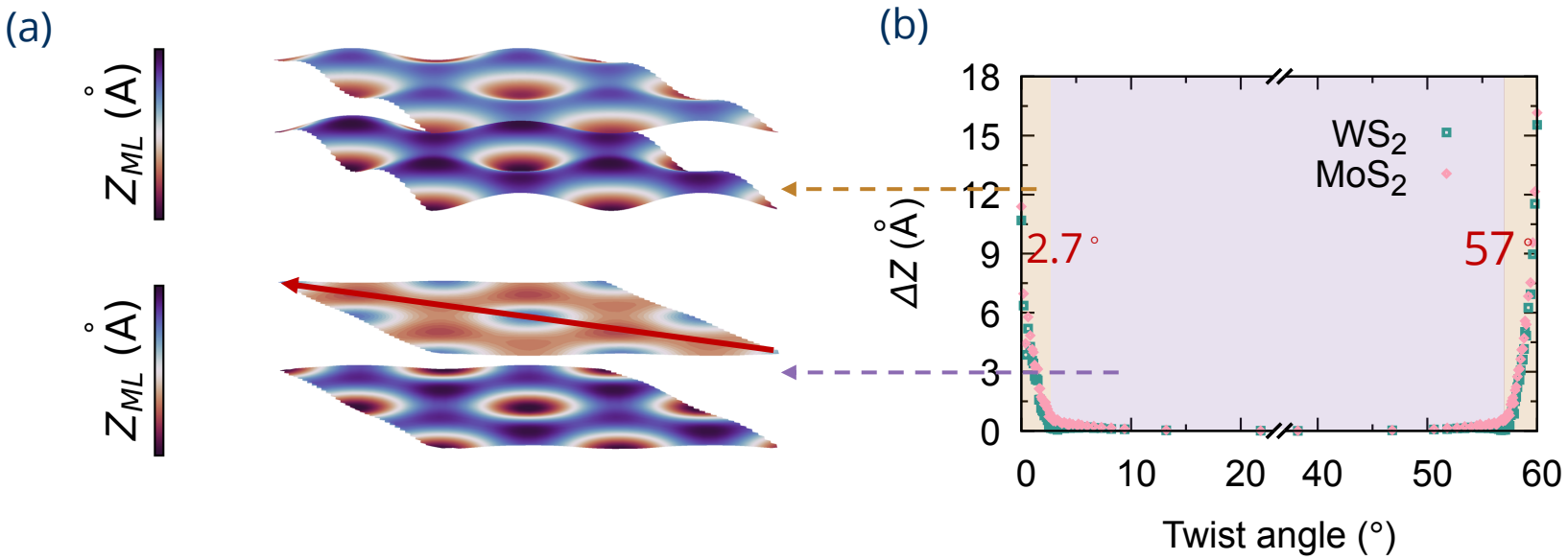
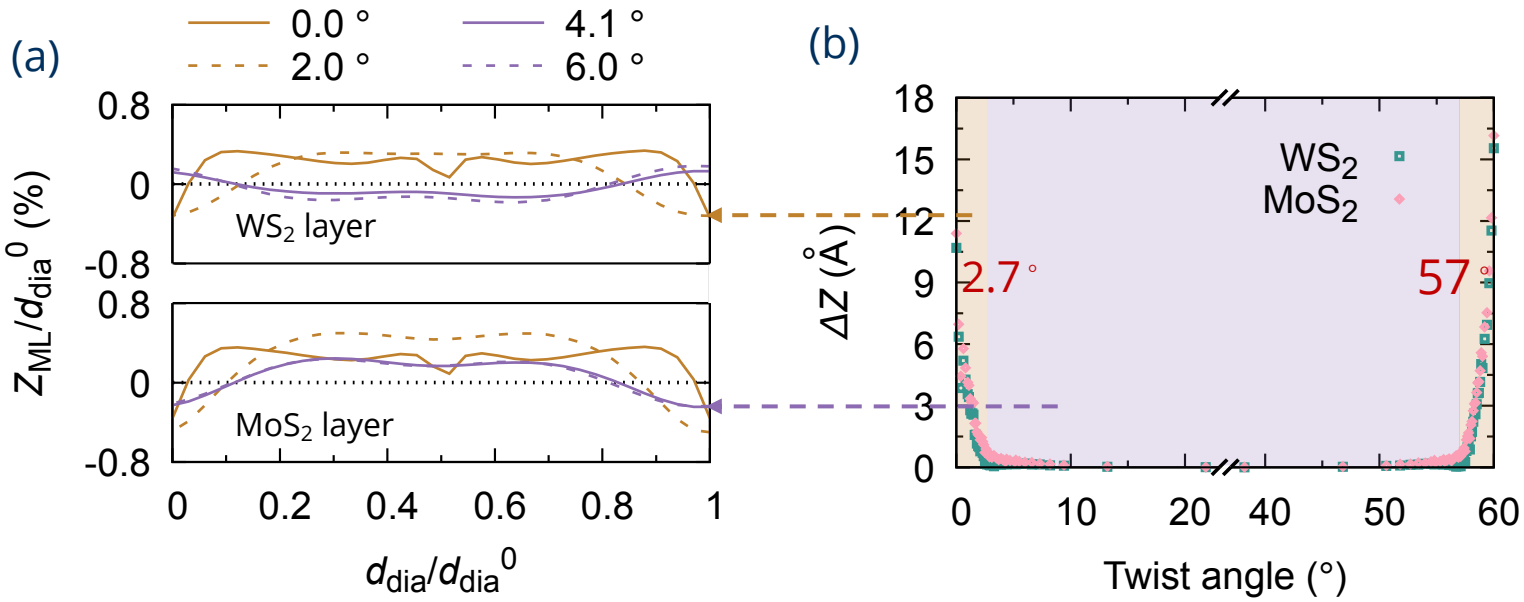
Small vs. large twist angles in MoS_2/WS_2



- (a) Out-of-plane corrugation of Mo. (b) Magnitude of corrugation of each layer.

Small vs. large twist angles in MoS_2/WS_2

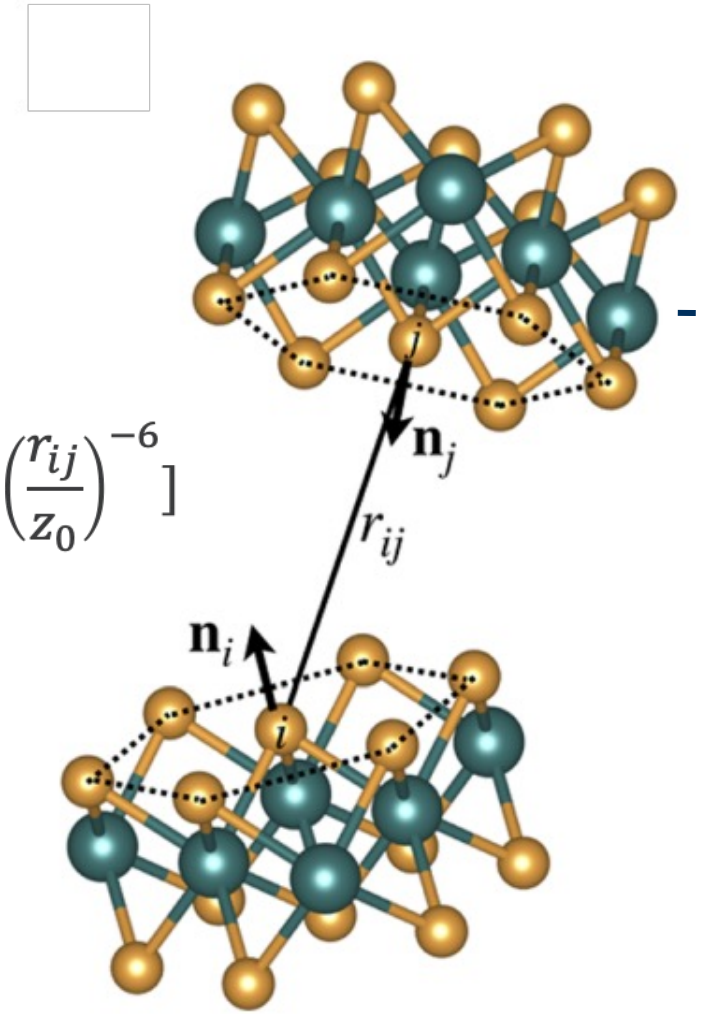


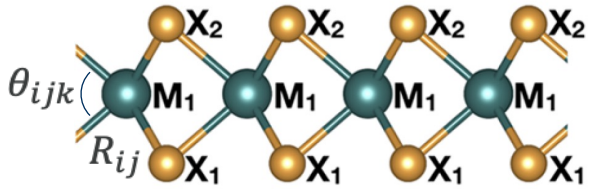
- (a) Out-of-plane corrugation of Mo along diagonal direction, (b) magnitude of corrugation of in each layer

Backup: Force-field method

$$\frac{1}{2}\sum_i \sum_{j\neq i} [e^{-\lambda(r_{ij}-z_0)} V_\rho - A\left(\frac{r_{ij}}{z_0}\right)^{-6}]$$

Interlayer





$$\sum_i \sum_{j>i} \phi_2(R_{ij}) + \sum_i \sum_{j\neq i} \sum_{k>j} \phi_3(R_{ij}, R_{ik}, \theta_{ijk})$$

bond stretching + angle bending

Intralayer

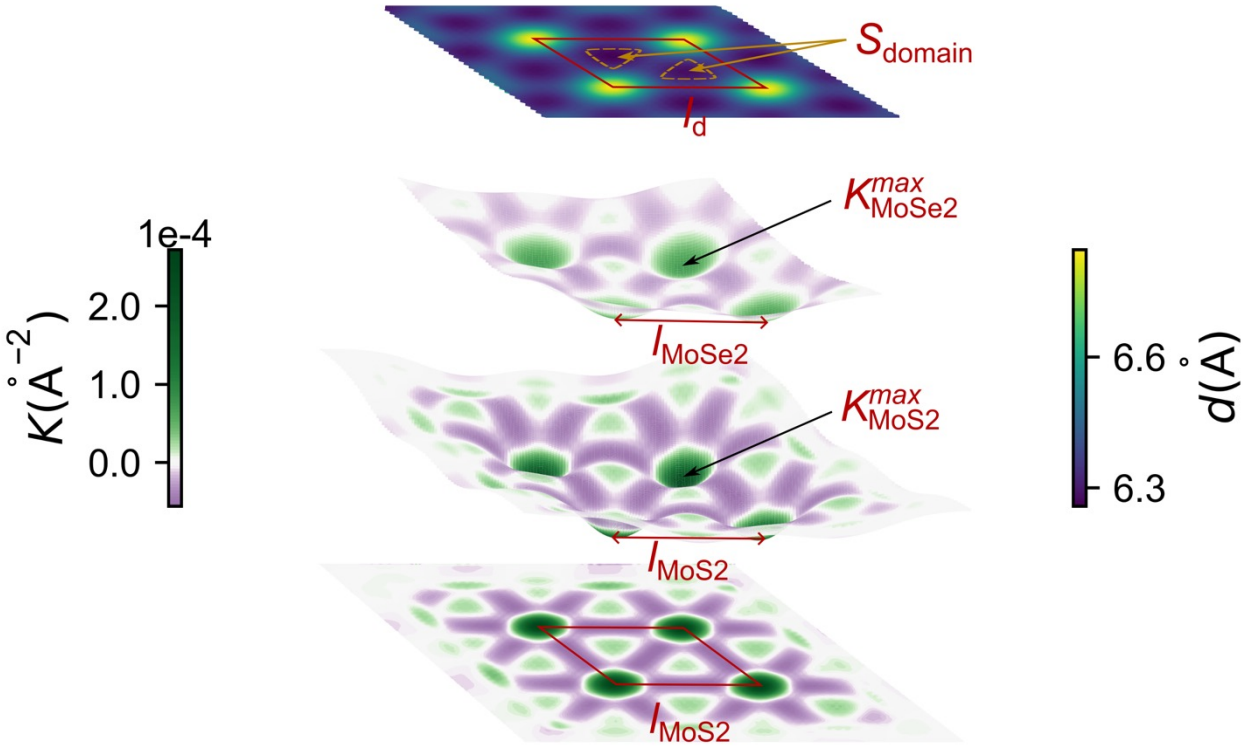
Backup: DFTB

$$E_{\mathrm{DFT}}[\rho] = T_{\mathrm{S}}[\rho] + \int \mathrm{d}\boldsymbol{r}\, v_{ext}(\boldsymbol{r})\rho(\boldsymbol{r}) + E_{\mathrm{H}}[\rho] + E_{\mathrm{xc}}[\rho]$$

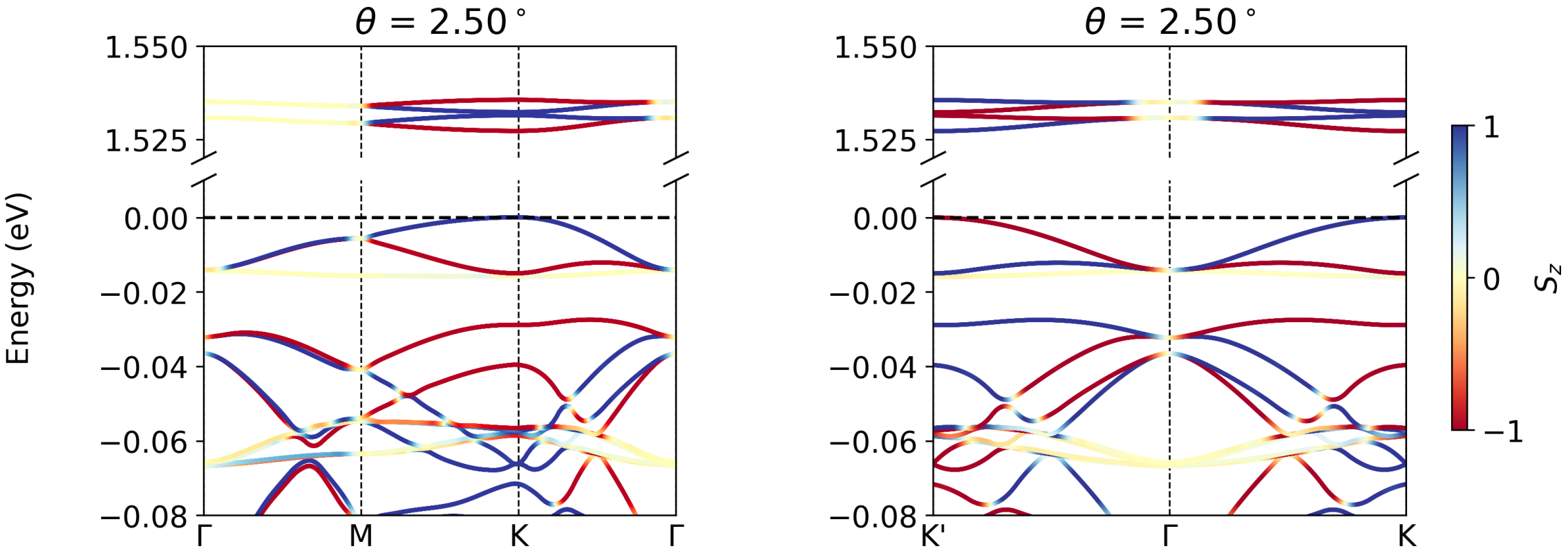
⬇ expand at ρ_0

$$E_{\mathrm{DFTB}}[\rho_0 + \delta\rho] = E^0[\rho_0] + E^1[\rho_0, \delta\rho] + E^2[\rho_0, (\delta\rho)^2] + E^3[\rho_0, (\delta\rho)^3]$$

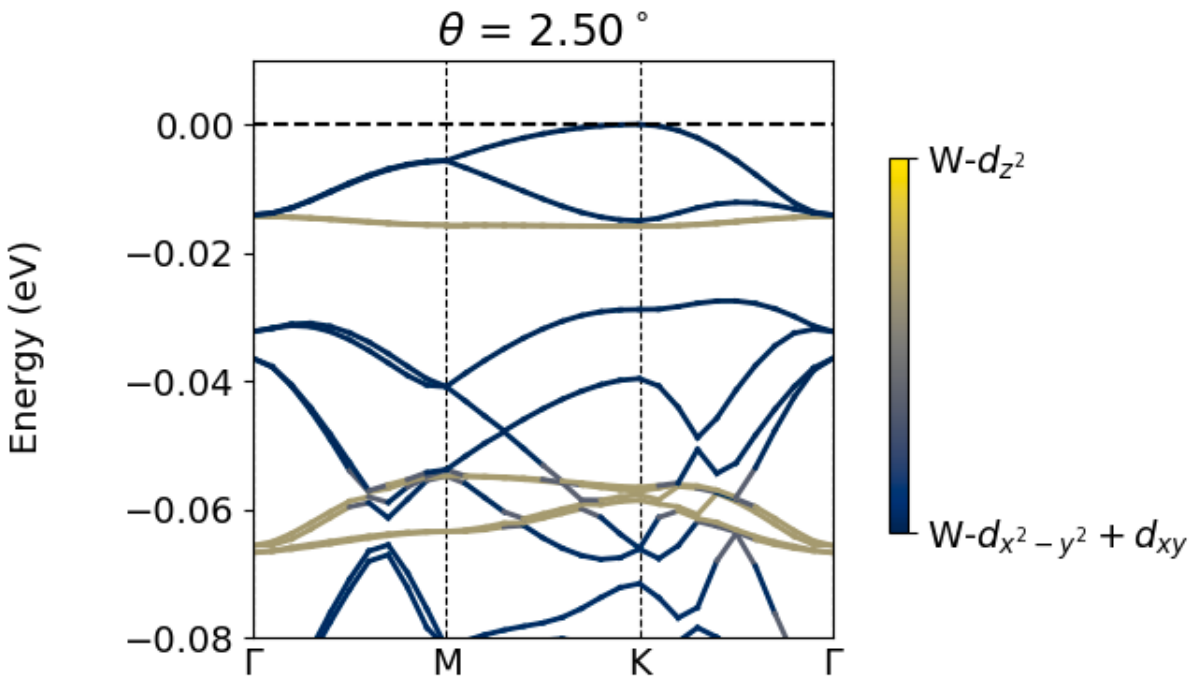
Backup: Flakes

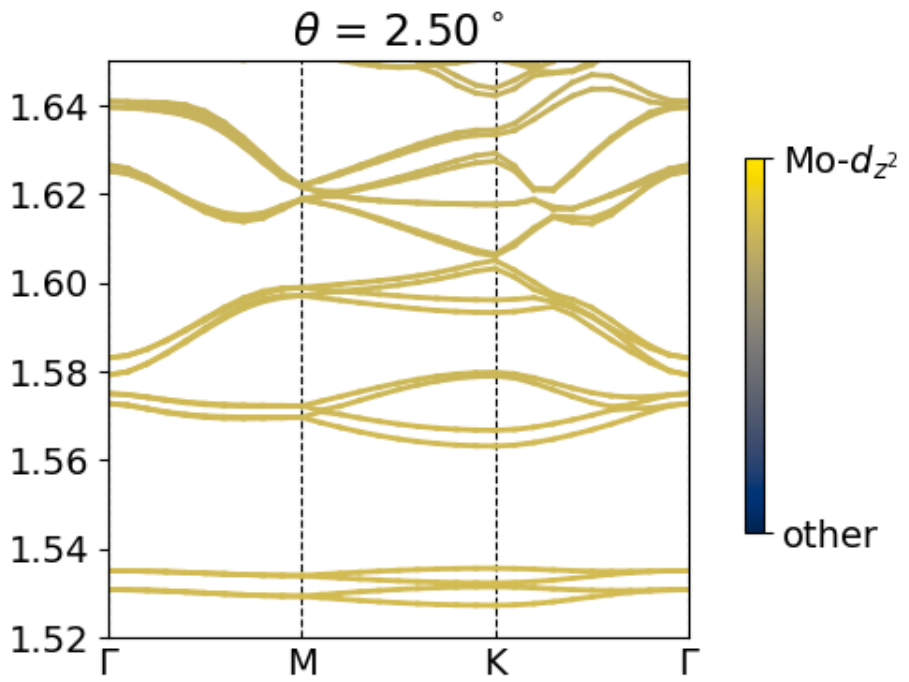


Backup: MoS_2/WS_2 at 2.5°

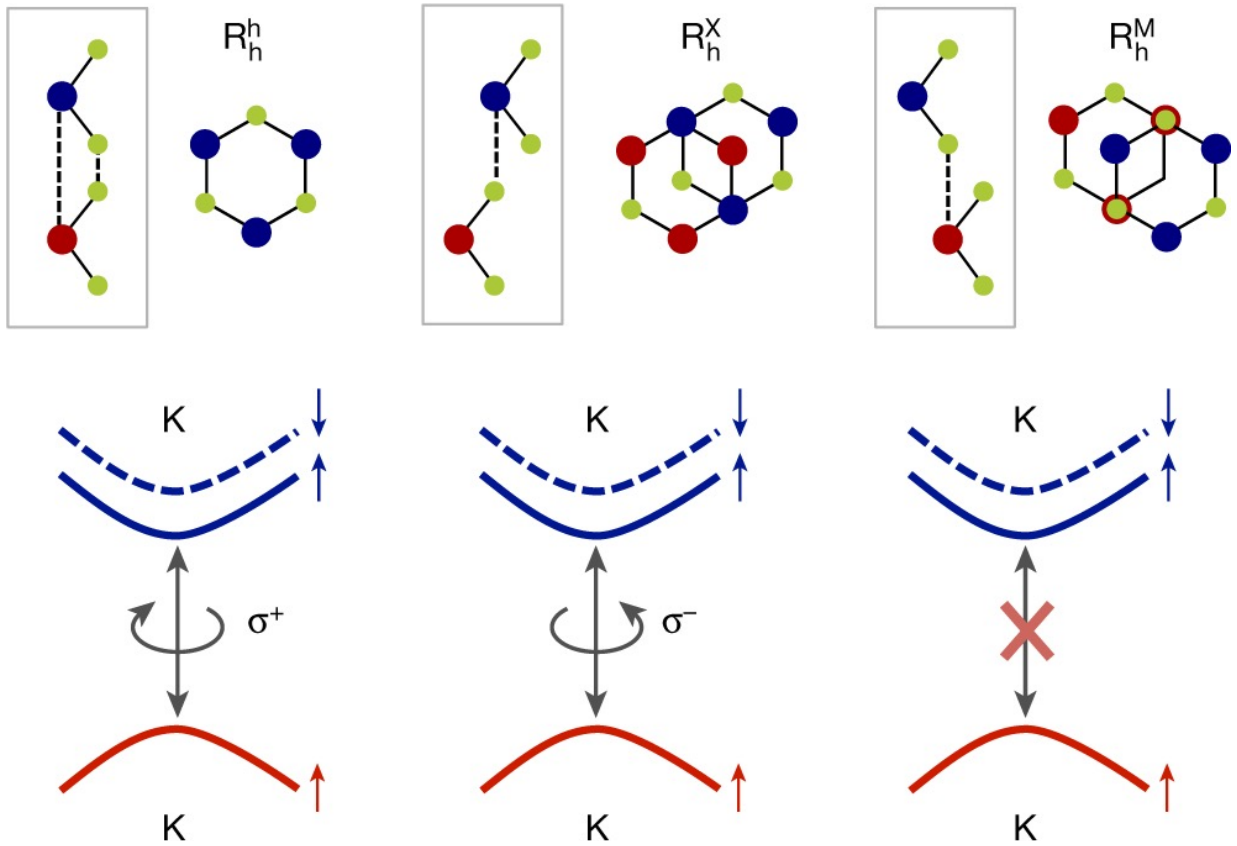


Backup: MoS_2/WS_2 at 2.5°





Backup: optical selection rules



[1] *Nature* **2019**, 567, 71.