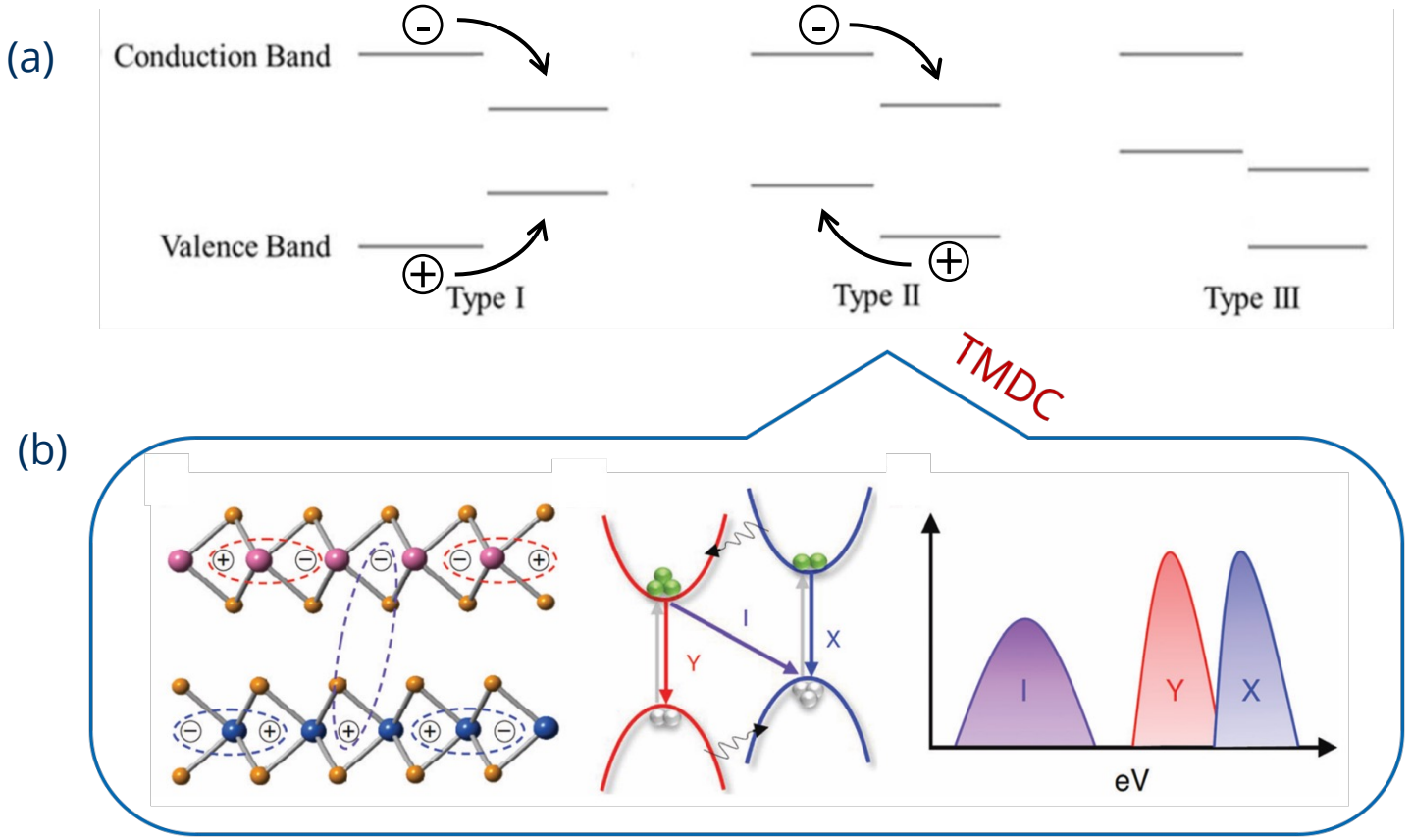
Wei Li, Gautam Jha, Thomas Brumme, Thomas Heine

Chair of Theoretical Chemistry, TU Dresden

Modulation of optical selection rules in twisted transition metal dichalcogenide heterobilayer

Mar 06, 2024 // Minneapolis

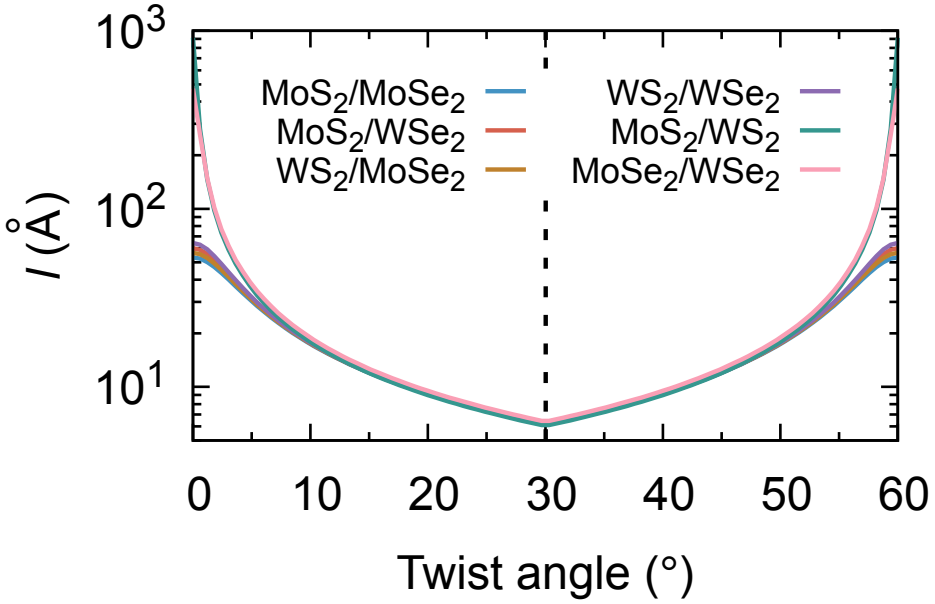
Why TMDC heterostructures?



(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]

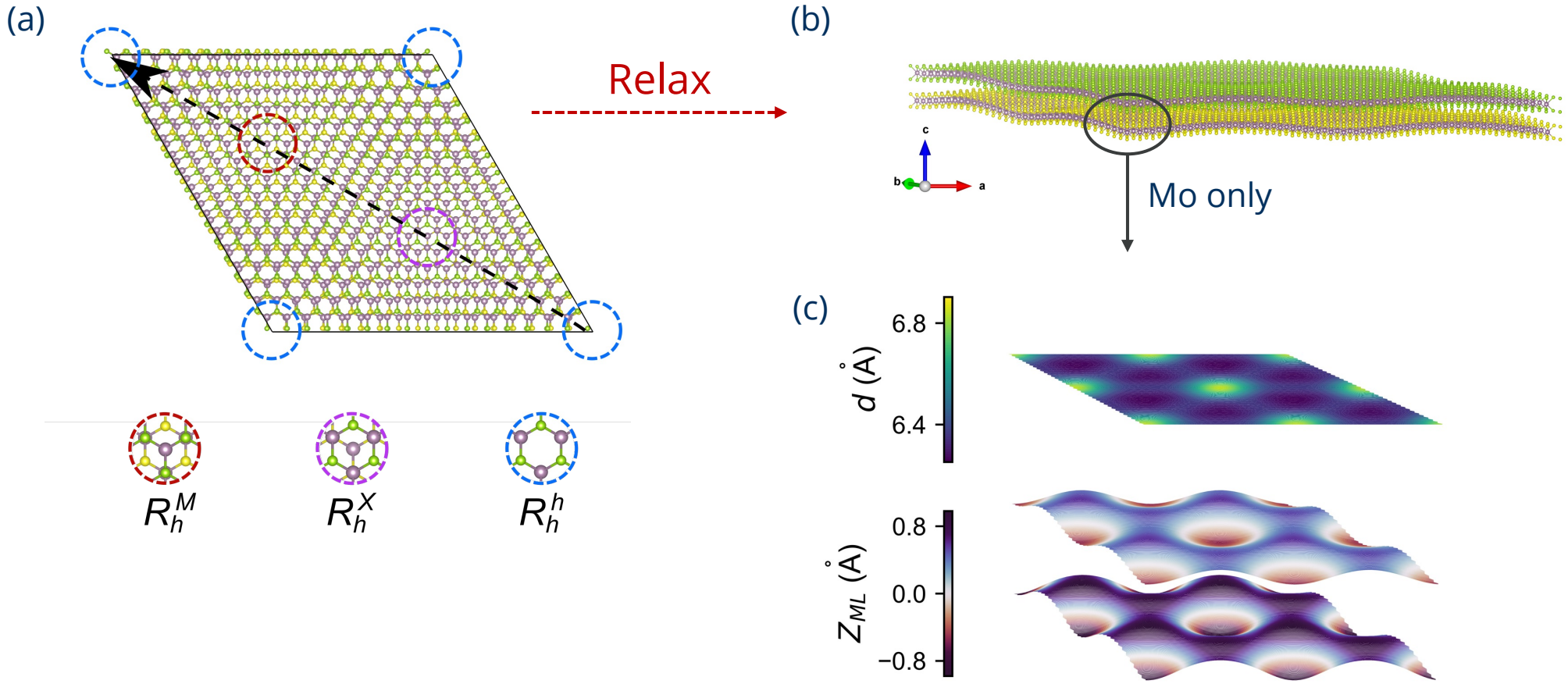
- Lattice size: 10^1 – 10^3 Å
- Number of atoms: 50 – 5×10^5

[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 – $MoS_2/MoSe_2$ at 0°



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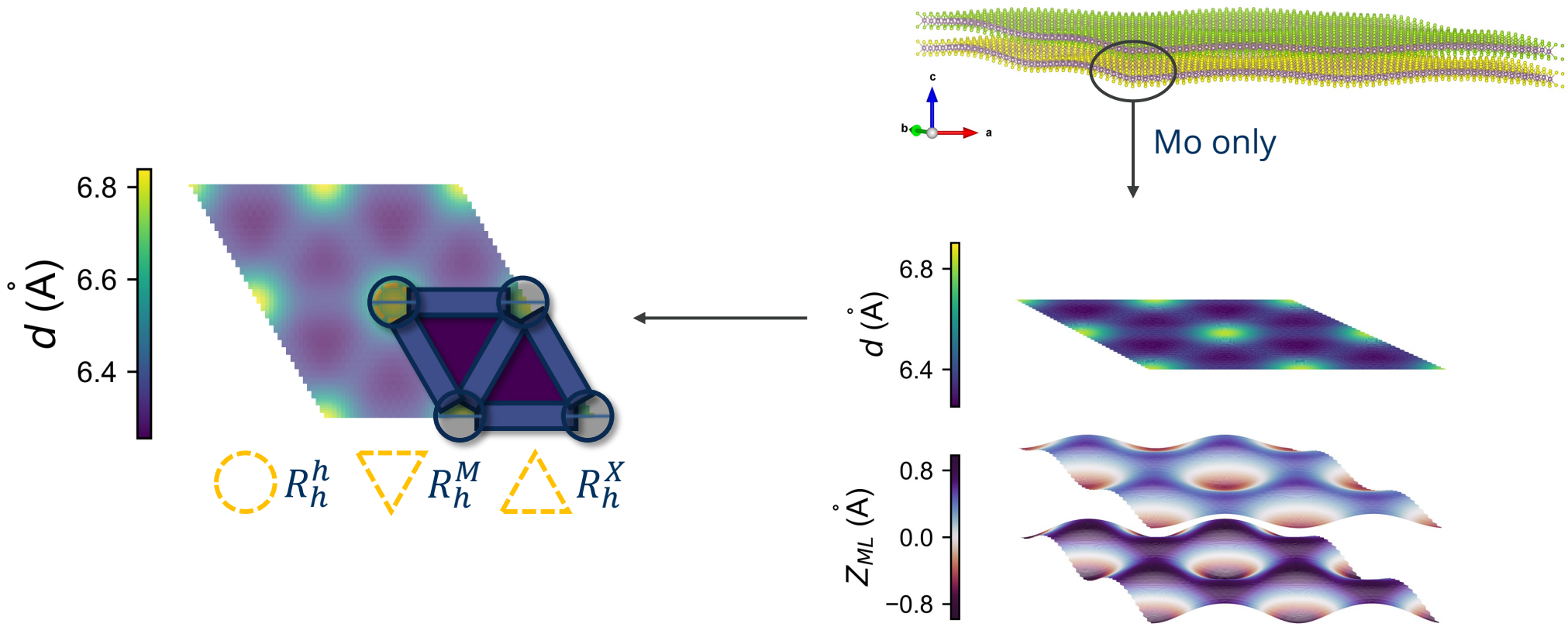
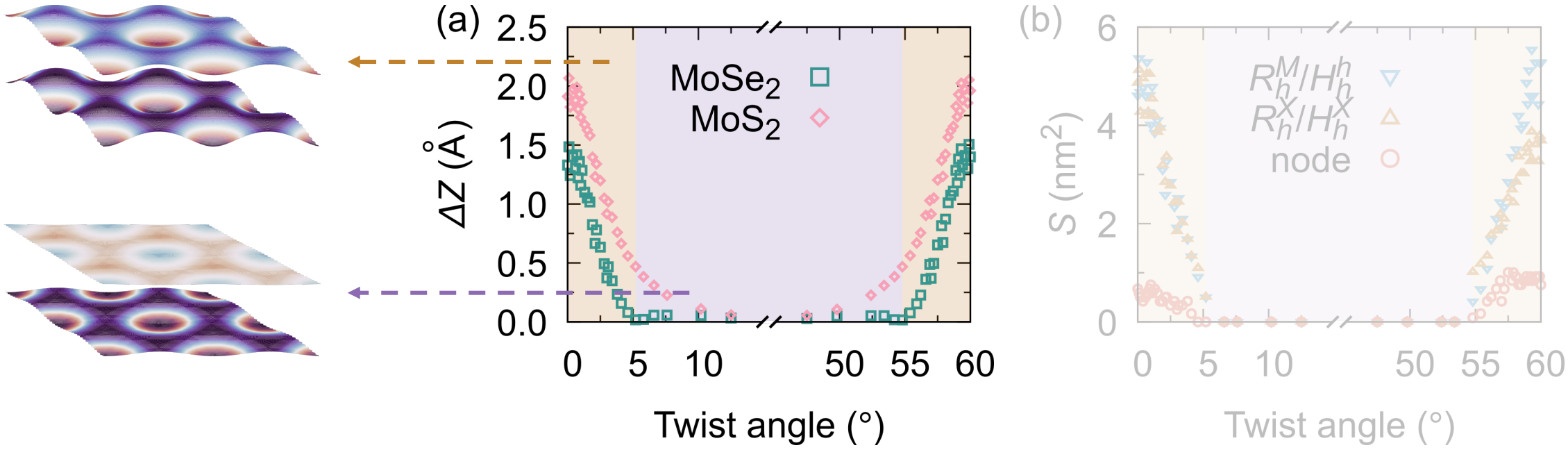
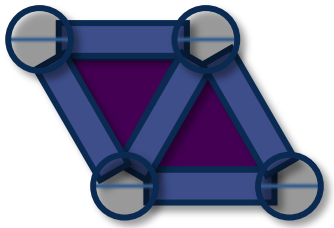


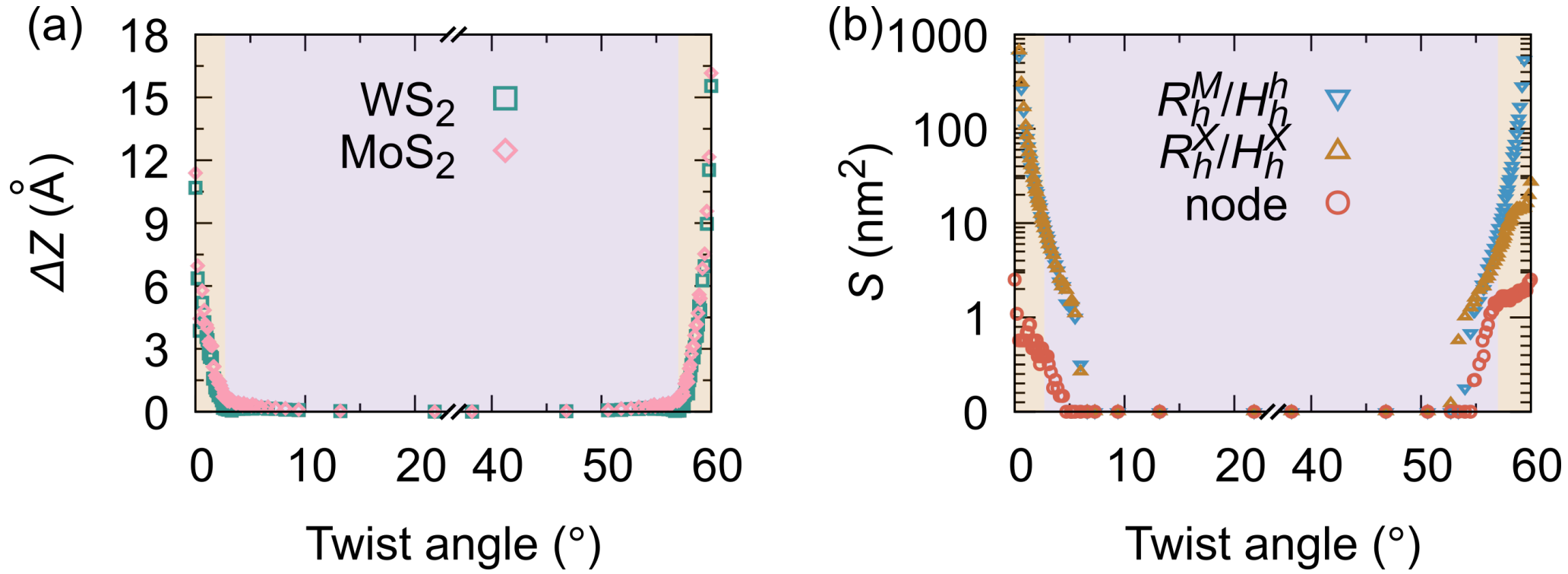
$MoS_2/MoSe_2$ at different twist angles



(a) Magnitude of corrugation of each layer. (b) Area of domains and node



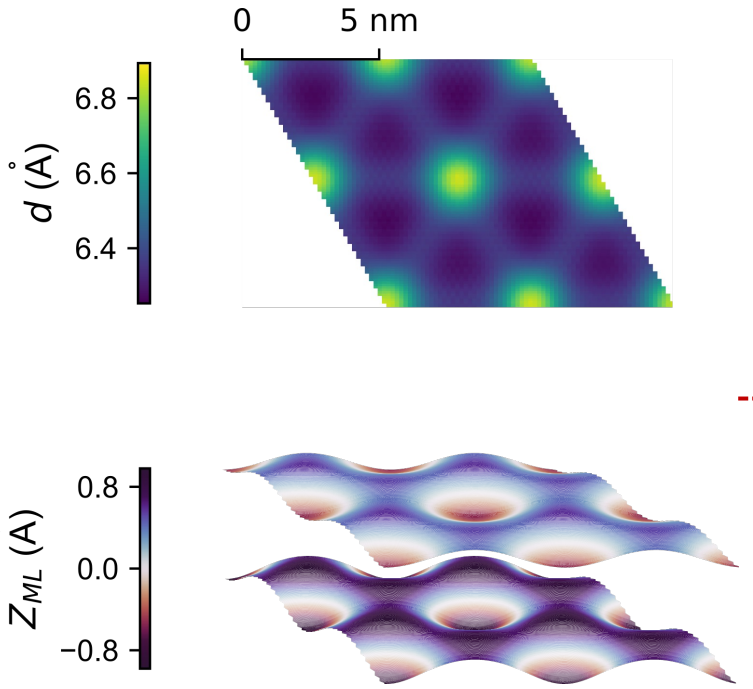
MoS_2/WS_2 at different twist angles



(a) Magnitude of corrugation of each layer. (b) Area of domains and node

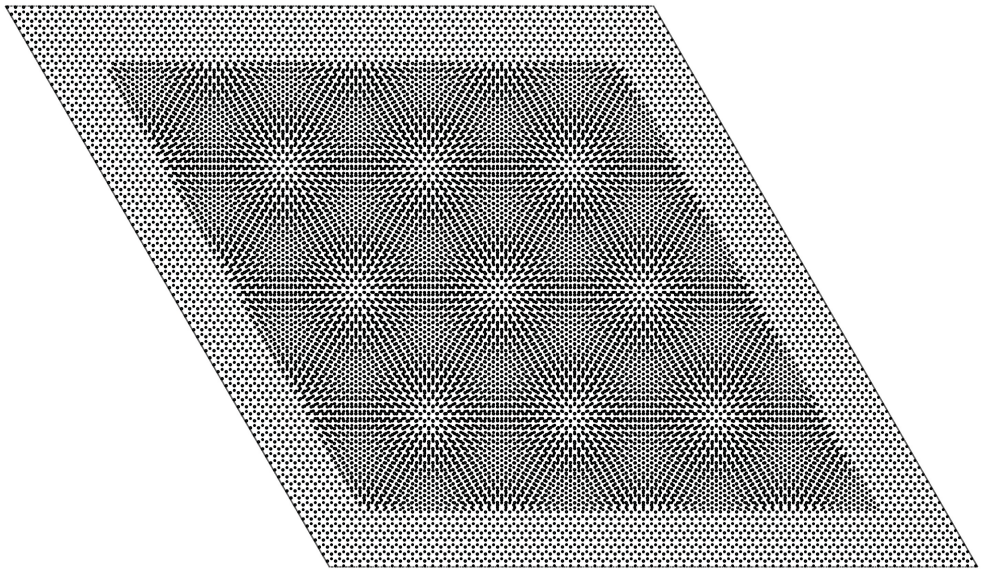
Theory vs experiment

(a)

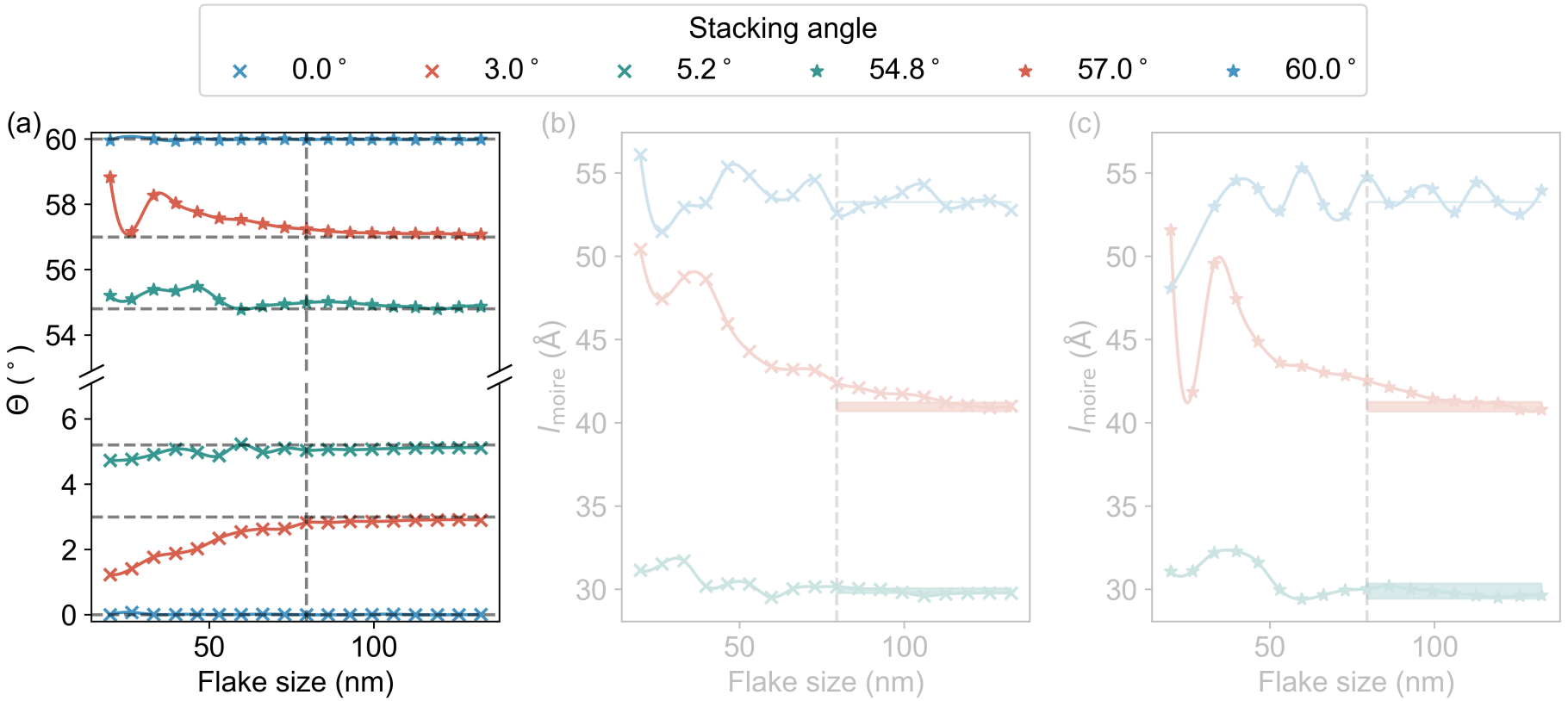


Infinite

(b)



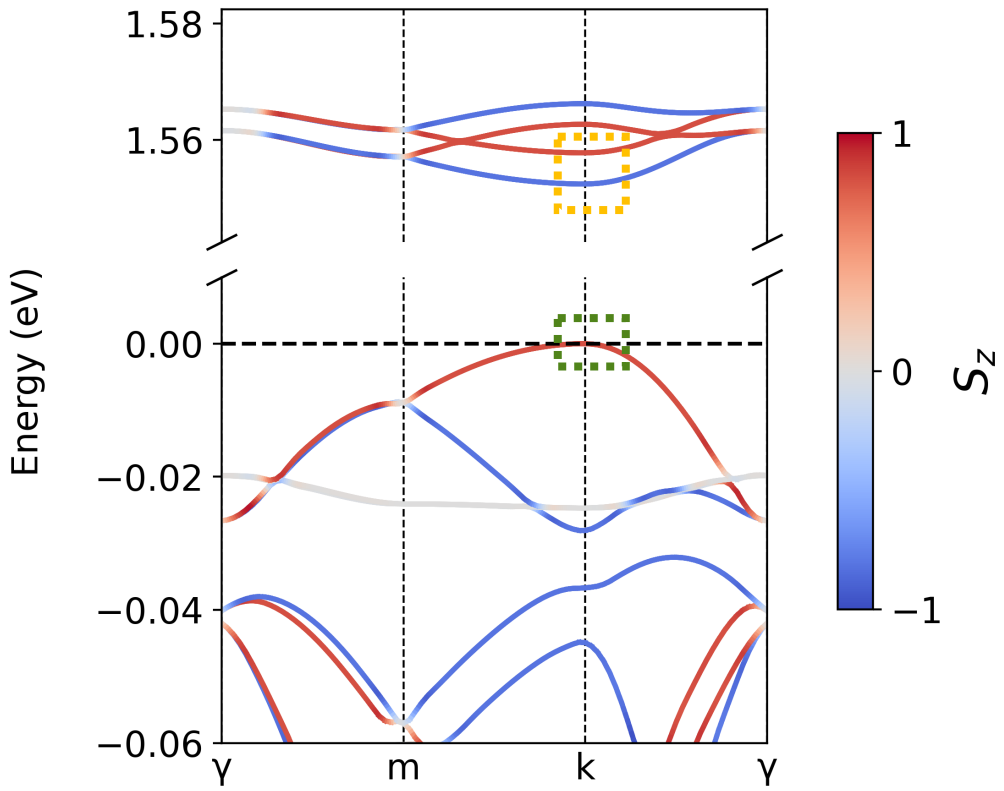
$MoS_2/MoSe_2$ flake systems

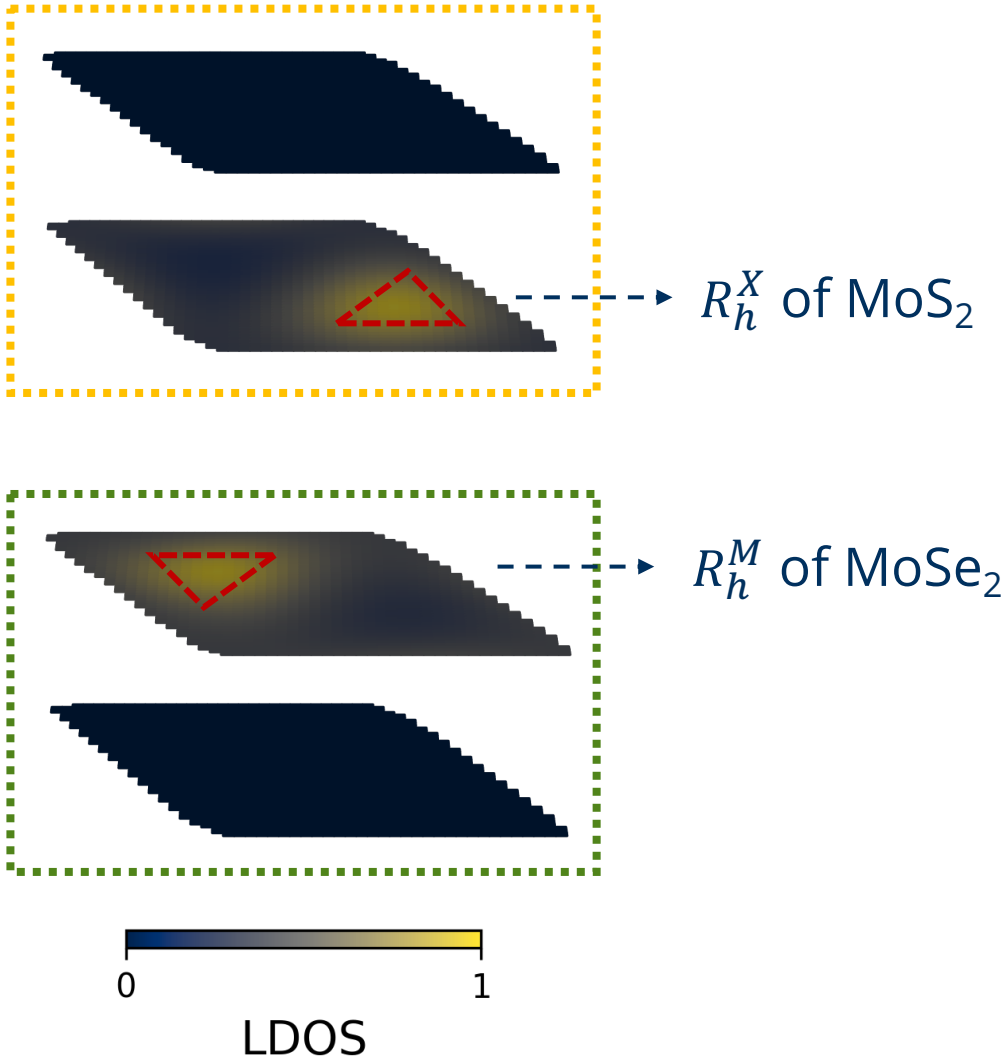


The effects of flake size on (a) twist angles and (b,c) moiré superlattice constant upon relaxation

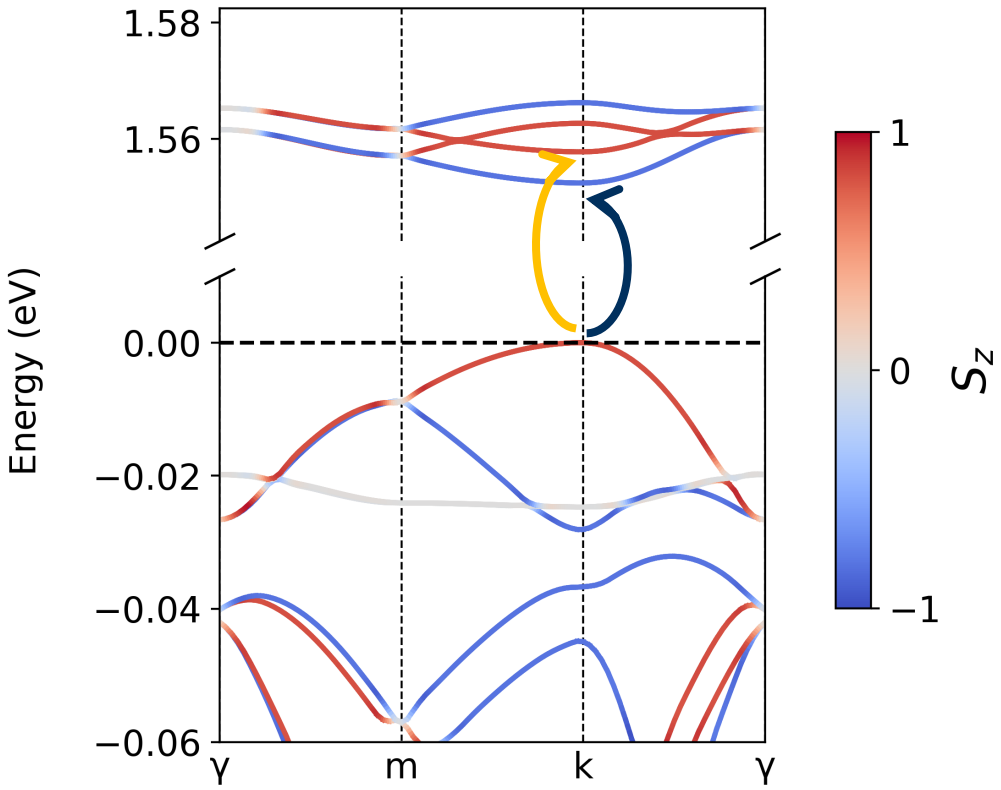
W Li, T Brumme, T Heine. Relaxation effects in transition metal dichalcogenide bilayer heterostructures. *npj 2D Mater. Appl.* (under revision)

MoS_2/WS_2 at 3° : Spin, angular momentum and g-factors



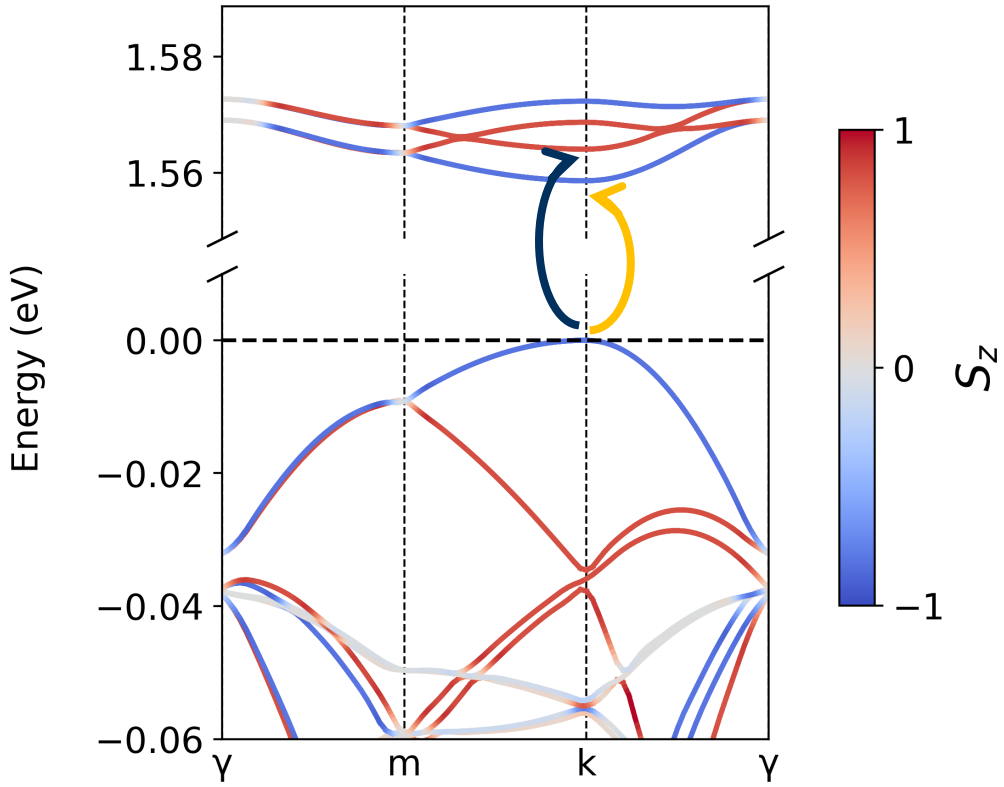


MoS_2/WS_2 at 3° : Spin, angular momentum and g-factors



R-type stacking		
g^{HS}	-1.2	2.78
transition	v → c	v → c+1
spin	↑↓	↑↑
ΔL	1.39	1.39

MoS_2/WS_2 at 57° : Spin, angular momentum and g-factors

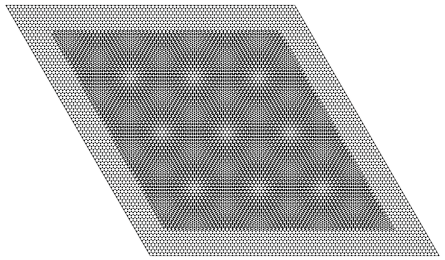


H-type stacking		
g^{HS}	-2.28	1.7
transition	v → c	v → c+1
spin	↑↓	↑↑
ΔL	-1.14	-1.14

Summary

- Significant lattice reconstruction
  - Domain formation
  - Out-of-plane corrugation

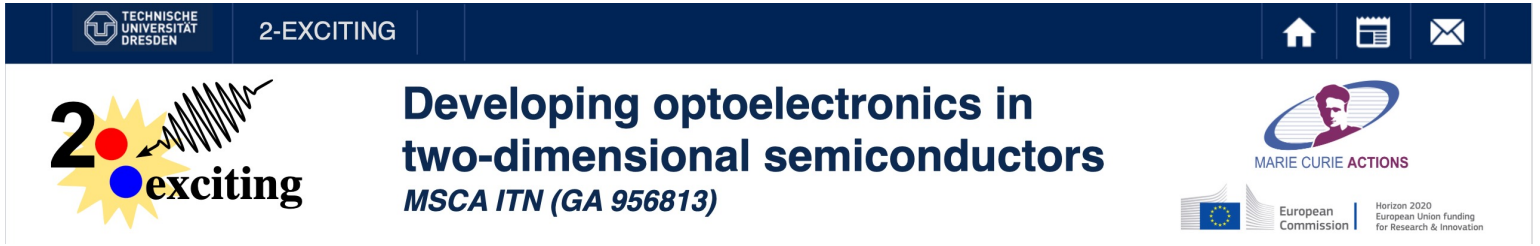
- Commensurate vs incommensurate model



- Spin orbital coupling effect and exciton g-factor

Acknowledgement

- Dr. Gautam Jha
- Dr. Thomas Brumme
- Prof. Dr. Thomas Heine



This project has received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 956813

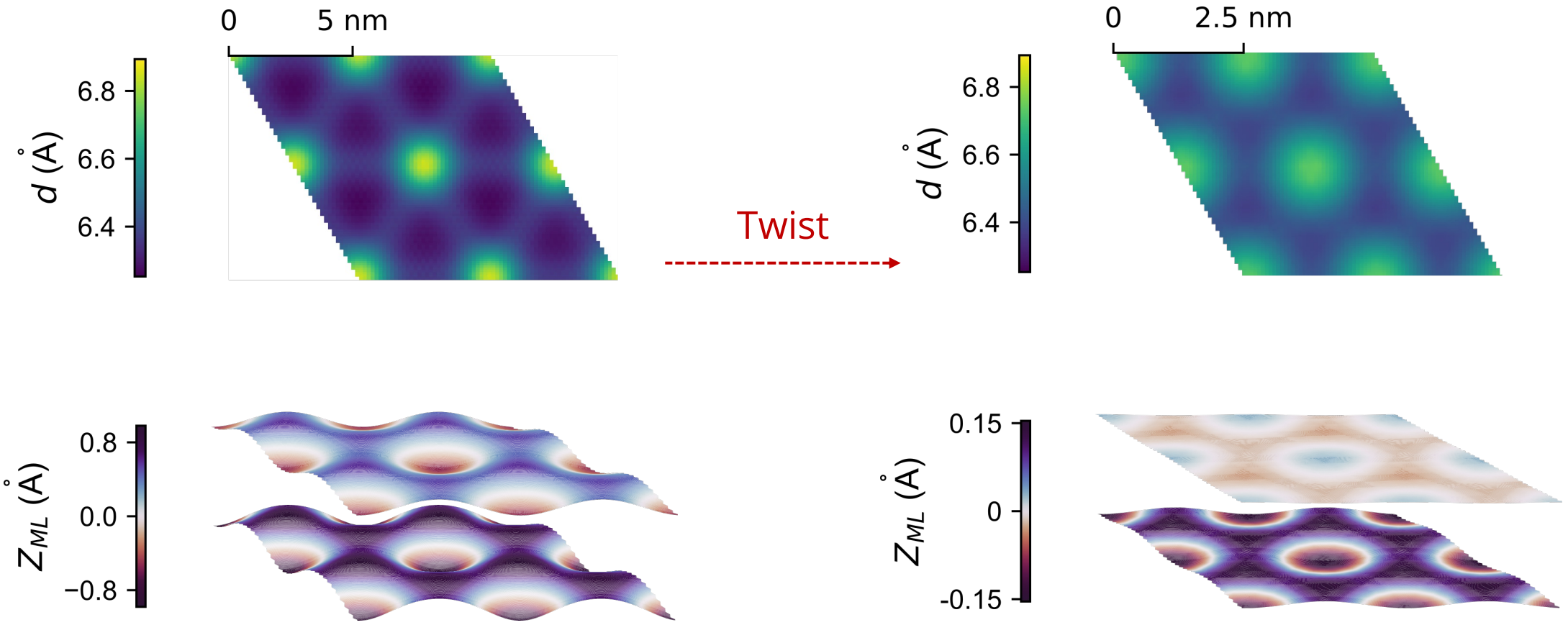
TECHNISCHE UNIVERSITÄT DRESDEN

HZDR HELMHOLTZ ZENTRUM DRESDEN ROSSENDORF

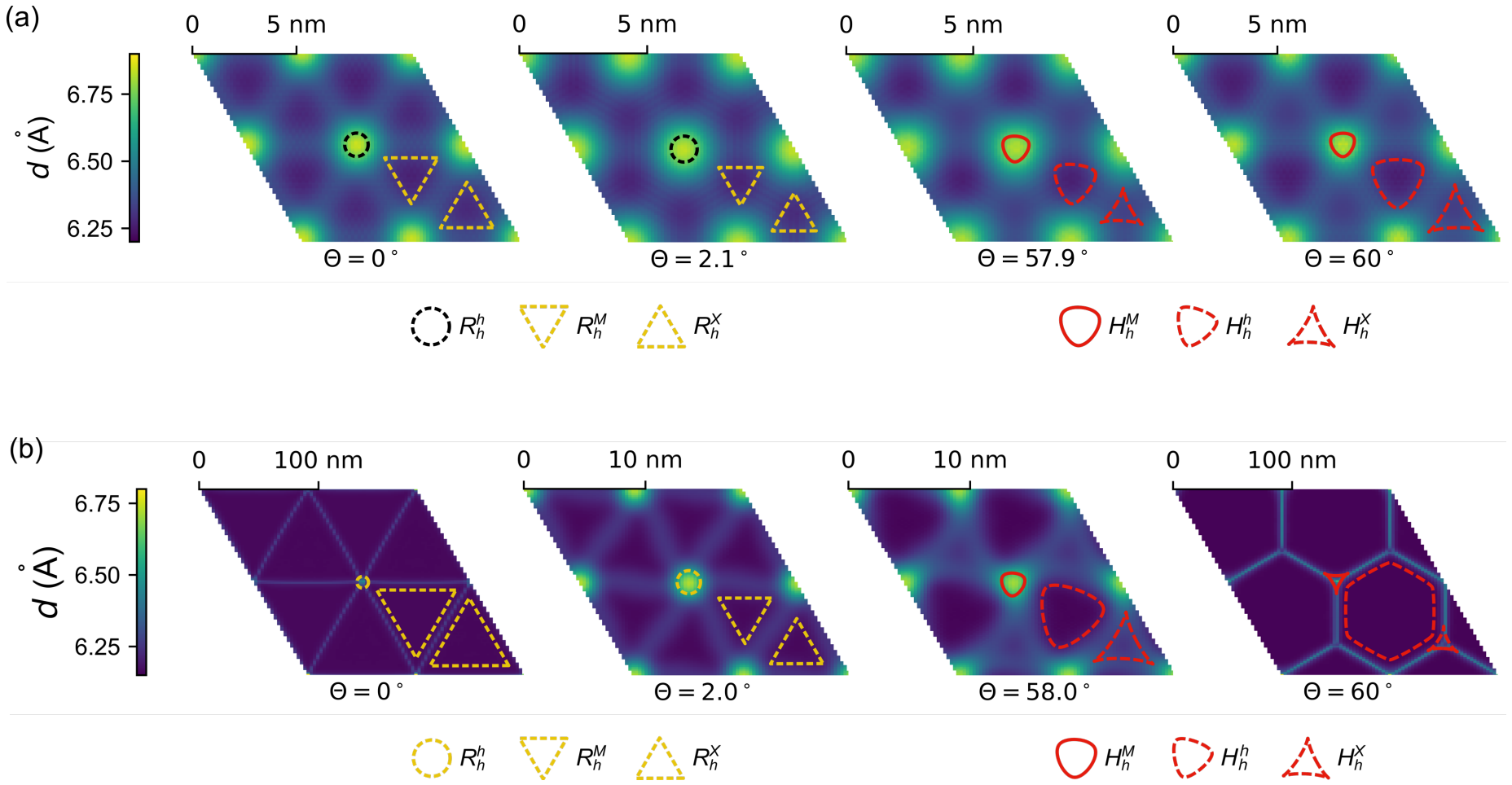
DRESDEN concept

THANK YOU

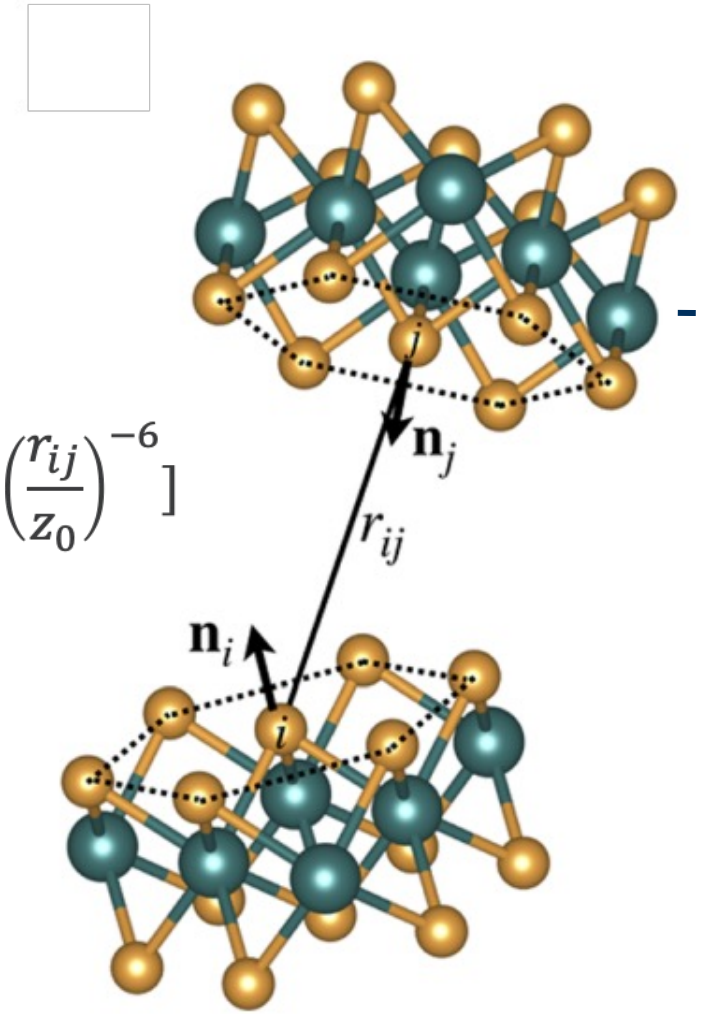
R-type stackings – Relaxed $MoS_2/MoSe_2$ at 0° and 6.6 °



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

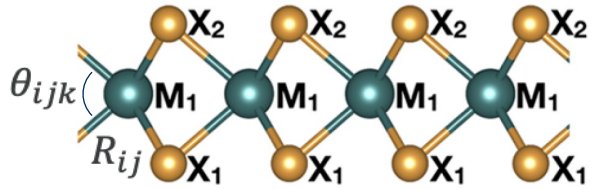


Backup: Force-field method



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

Interlayer



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

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: MoS_2/WS_2 at 2.5°