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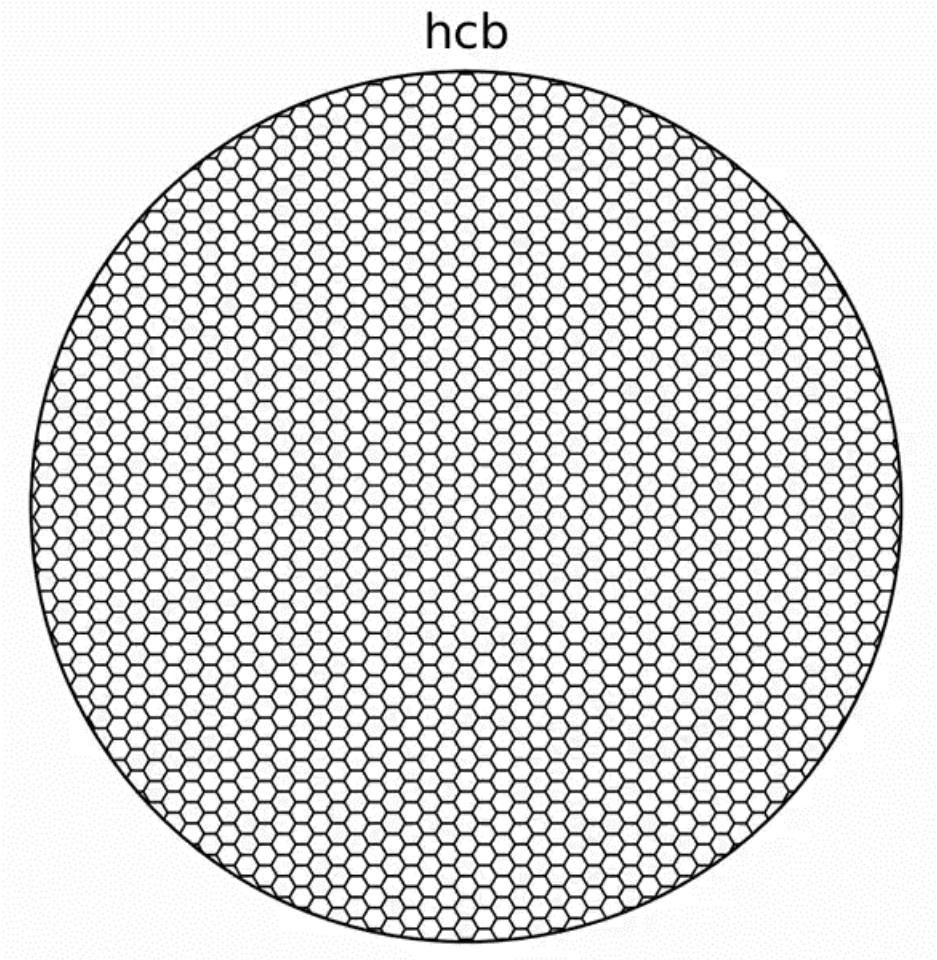
Chair of Theoretical Chemistry, TU Dresden

Relaxation effects in twisted transition metal dichalcogenide heterostructures

June 29, 2023 // Manchester

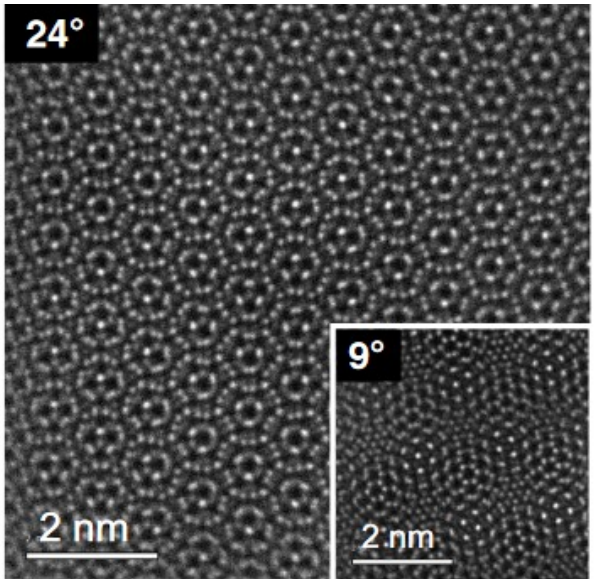
Why twisted: Moiré



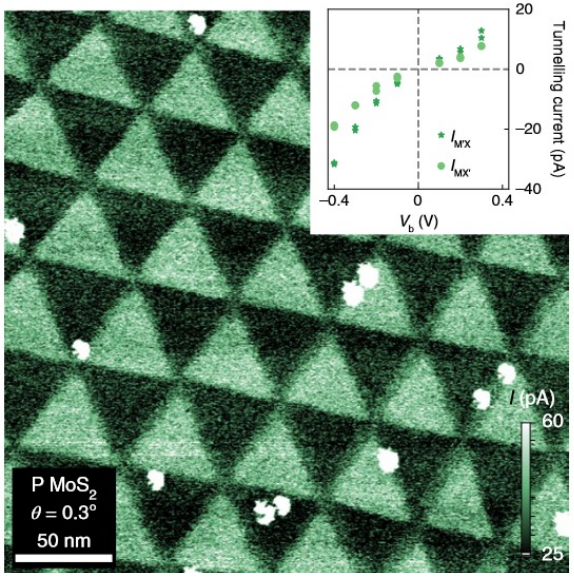
Florian Arnold. (July 5, 2022). Moiré clocks (Video File). Retrieved from https://www.youtube.com/watch?v=uxoKzBPFbrs&list=PL2LyfOO_UvEx5pfKmWIii_hmRBmZwJVG6&index=16.

Relaxation effects

Experimental observation: atomic reconstructions



Small θ



Twisted bilayer MoS_2[1]

Bilayer MoS_2 at $\theta = 0.3°$[2]

[1] *Nat. Comm.*, **2022**, *13*, 3898.
[2] *Nat. Nanotechnol.*, **2020**, *15*, 592.

Unrelaxed $MoS_2/MoSe_2$ at 0°

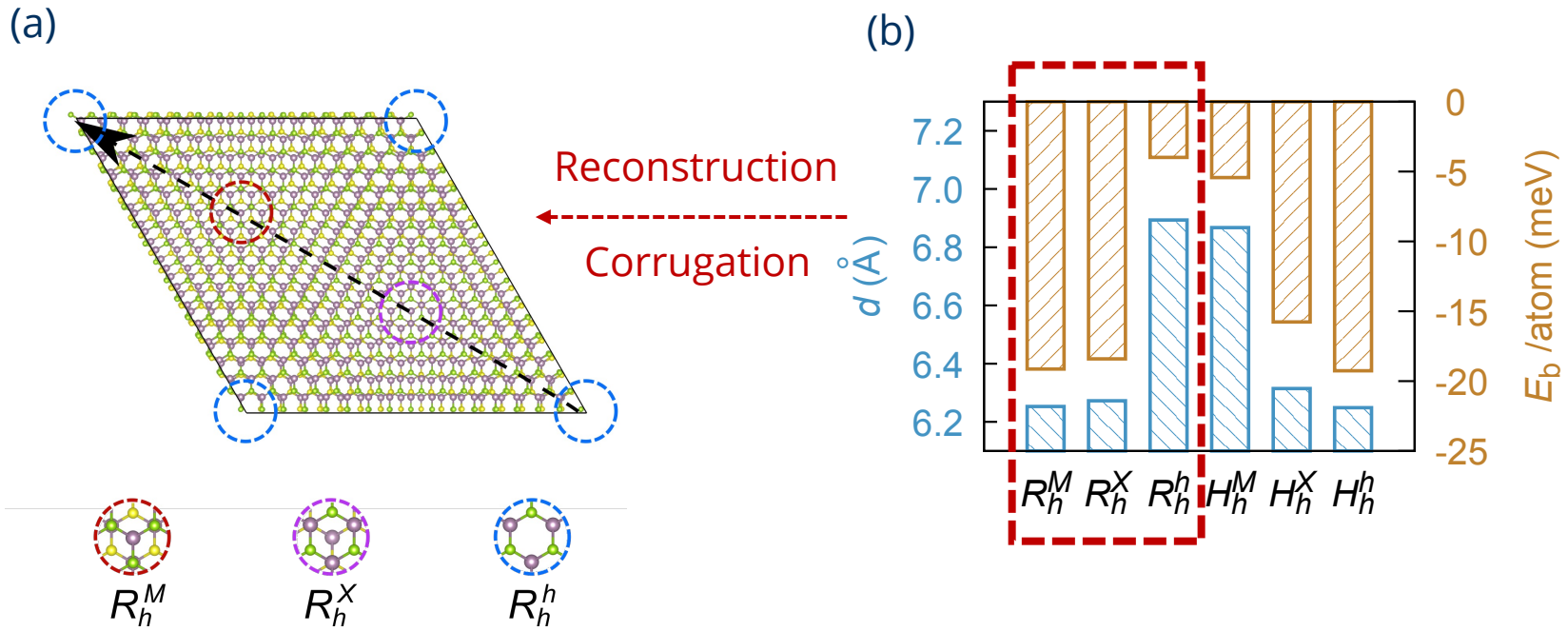
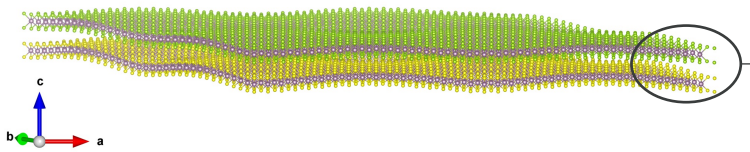


Fig 1. (a) 3 high-symmetry stackings and transition stacking regions. (b) Interlayer distance and binding energy of corresponding high-symmetry stackings.

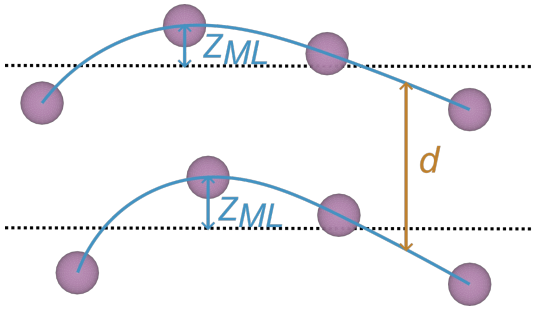
Relaxed $MoS_2/MoSe_2$ at 0°

(a)

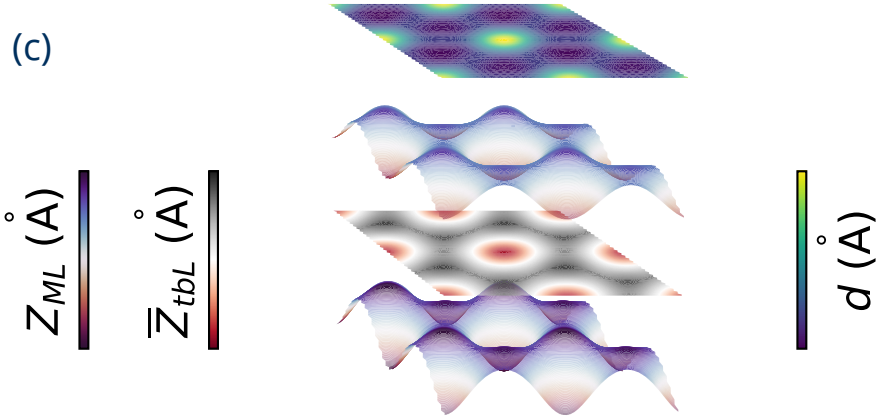


Mo only

(b)

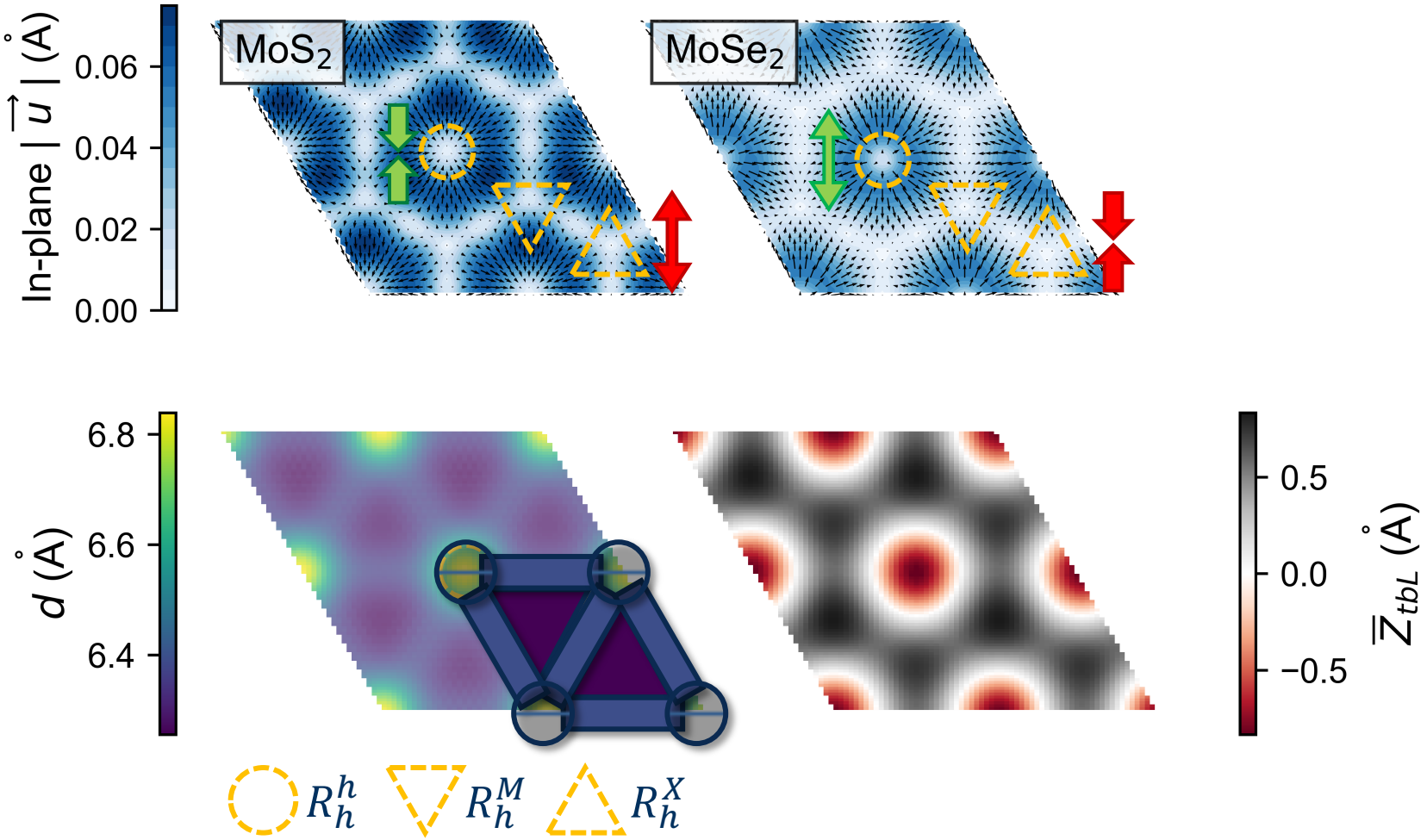


(c)

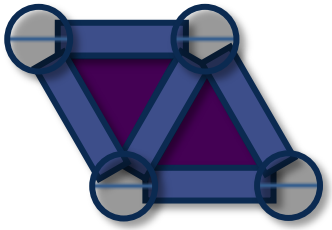


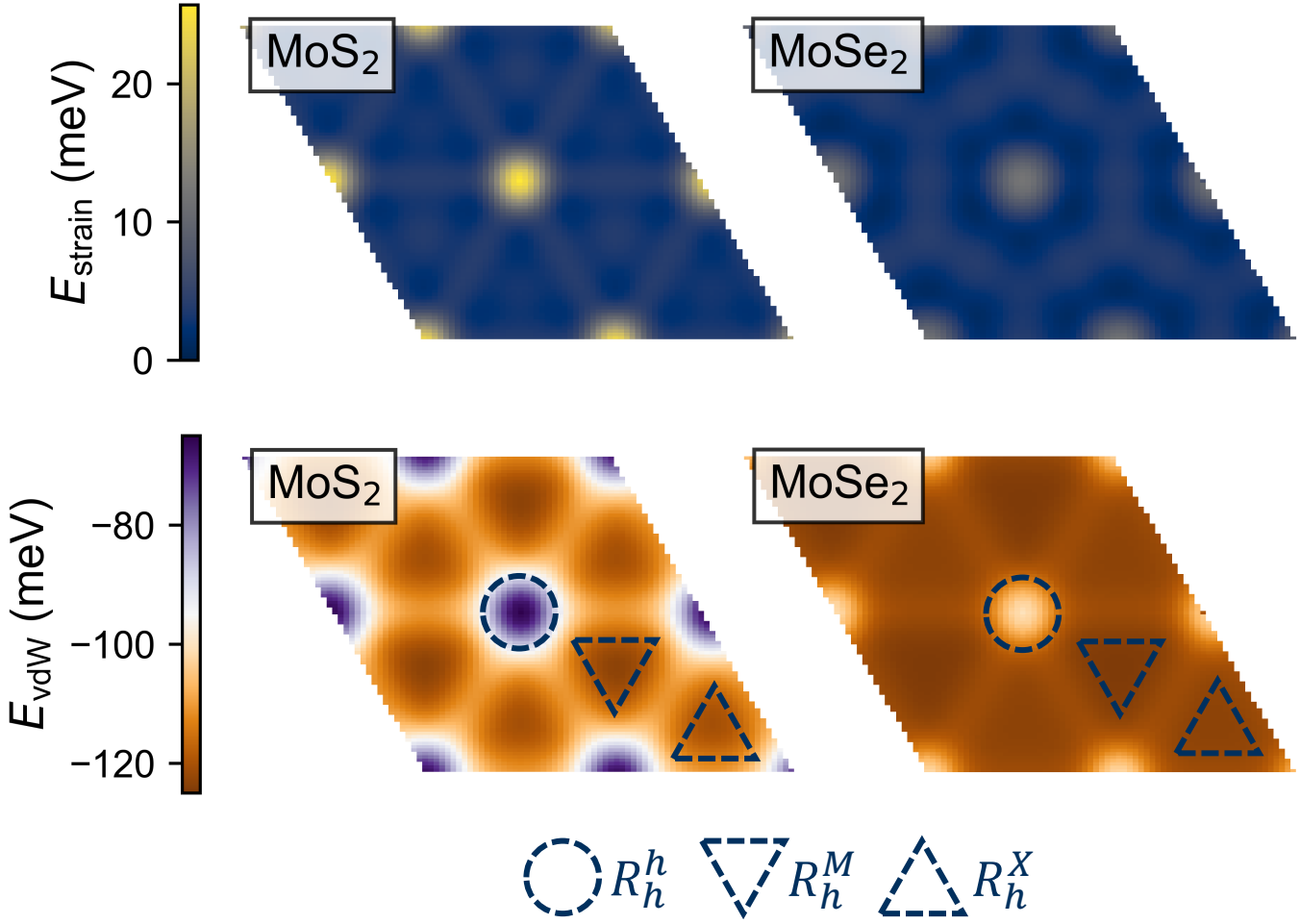
- Out-of-plane displacement Z_{ML}
- Average Out-of-plane displacement $\bar{Z}_{\mathrm{ML}}$
- Interlayer distance d

Lattice reconstruction and corrugation

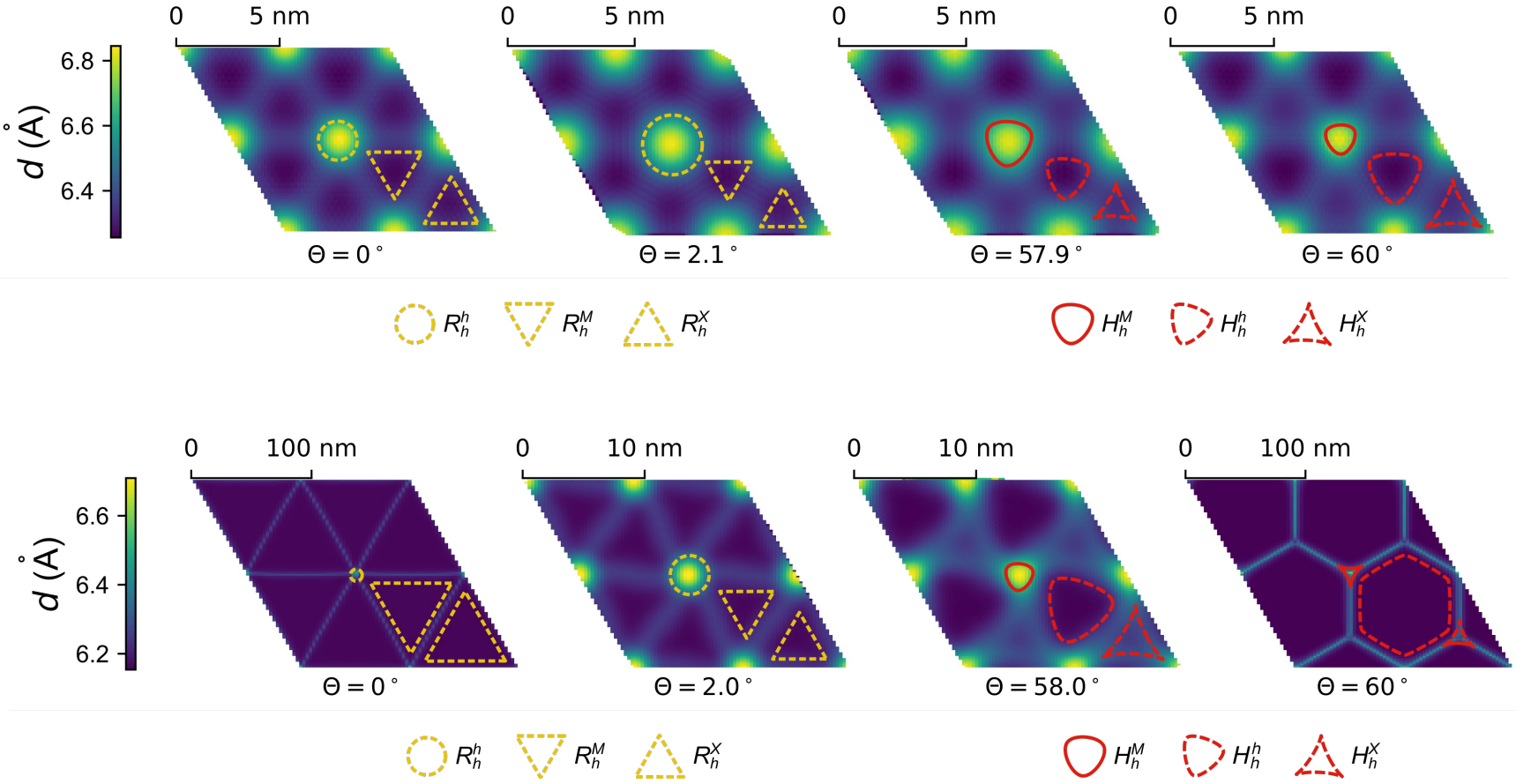


Strain energy and vdW energy

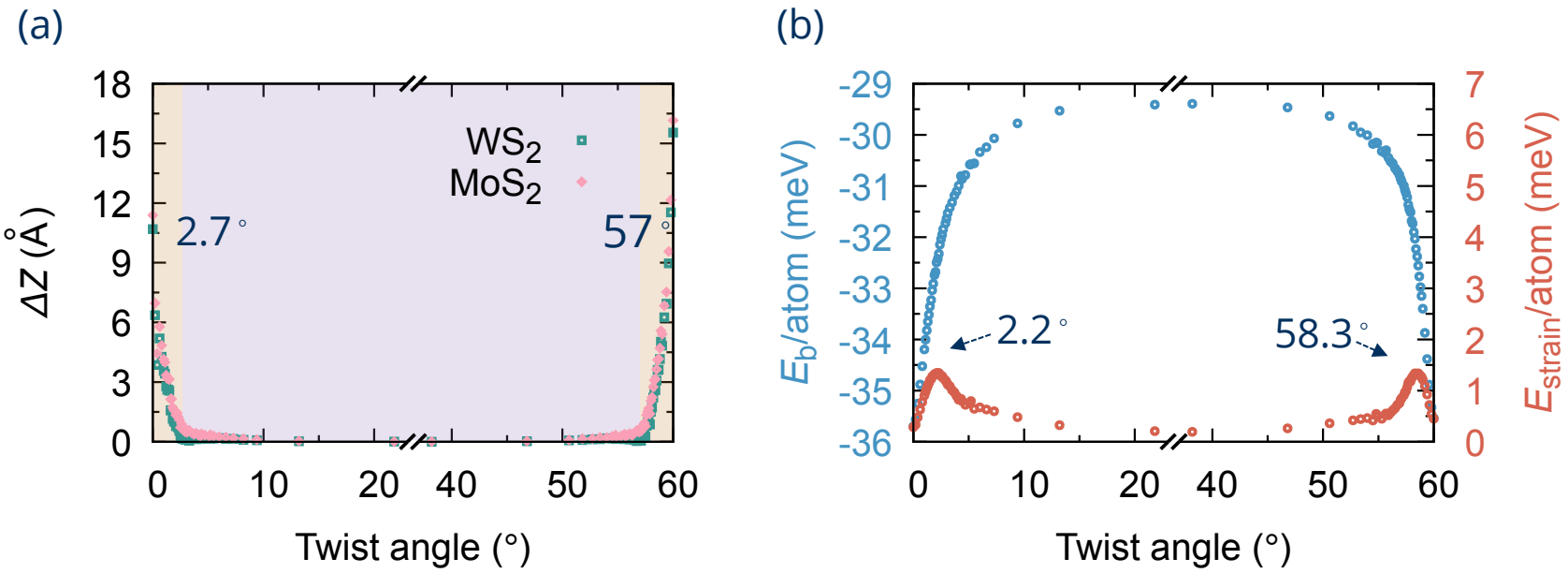




Twisted $MoS_2/MoSe_2$ and MoS_2/WS_2

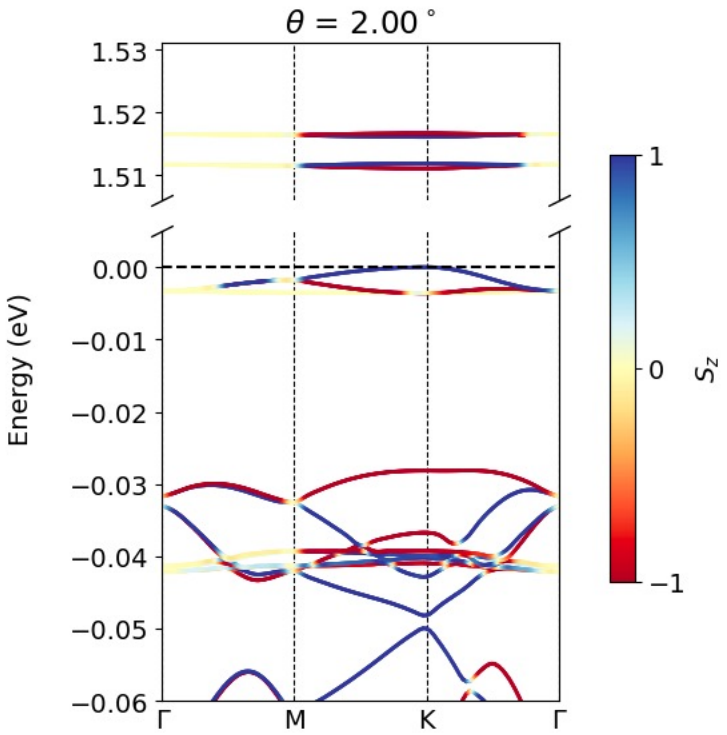


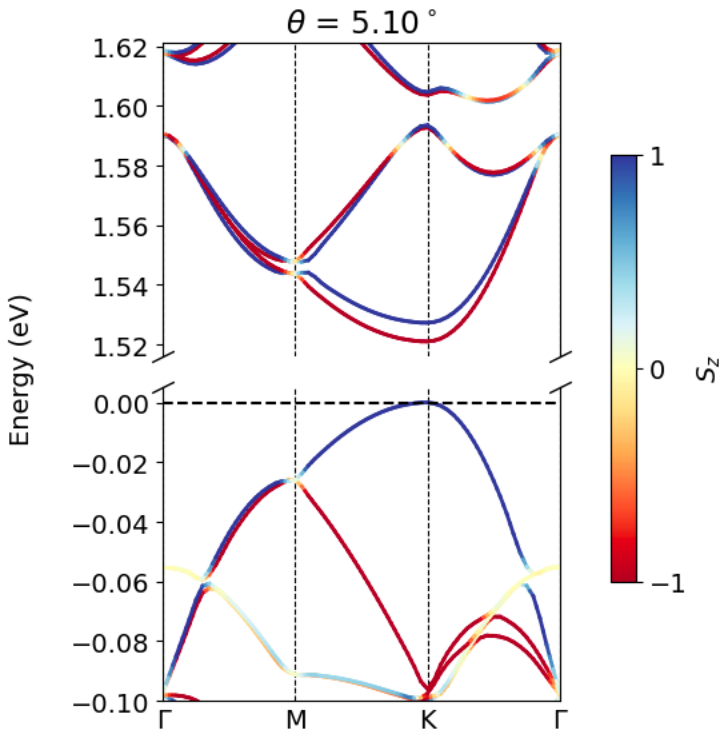
Small vs. large twist angles in MoS_2/WS_2



- Fig 1. (a) Out-of-plane corrugation of Mo along diagonal direction, (b) magnitude of corrugation of in each layer, (c) binding/strain energy at different twist angles.

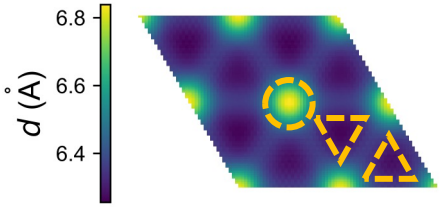
Spin-Orbit Coupling effect



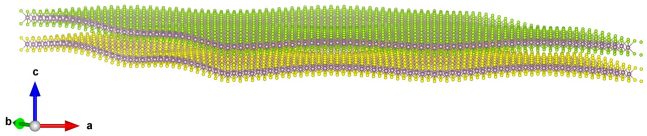


Summary

- Significant lattice reconstruction
  - Domain formation

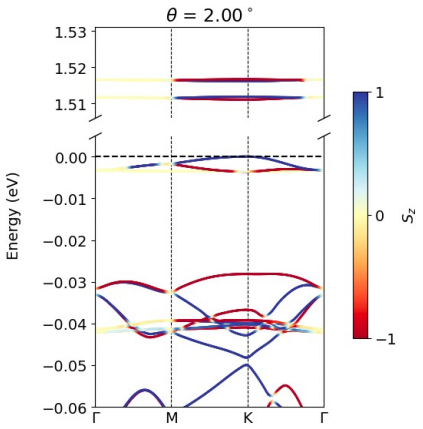


  - Out-of-plane corrugation



  - Depending on the twist angle

Inversion symmetry broken



- Strain energy cost and van der Waals energy gain

Acknowledgement

- Prof. Dr. Thomas Heine
- Dr. Thomas Brumme
- M.Sc. Gautam Jha
- M.Sc. Florian Arnold
- ThC group
- 2EXCITING network

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2exciting

Developing optoelectronics in two-dimensional semiconductors

MSCA ITN (GA 956813)

MARIE CURIE ACTIONS

European Commission | Horizon 2020 European Union funding for Research & Innovation

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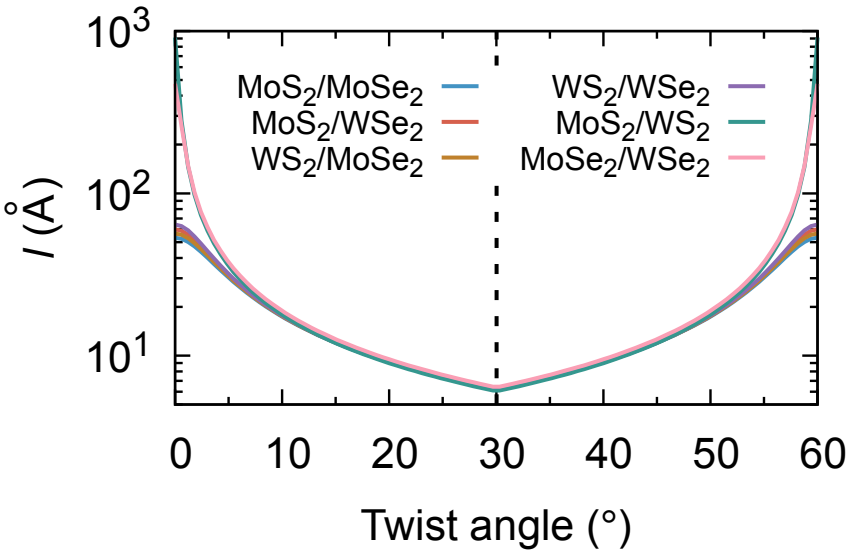
How: Multiscale approach



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

- 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.

Generating models

$$\begin{bmatrix} \tilde{a}_1^T \\ \tilde{a}_2^T \end{bmatrix} = M_a \begin{bmatrix} a_1^T \\ a_2^T \end{bmatrix}, \quad \begin{bmatrix} \tilde{b}_1^T \\ \tilde{b}_2^T \end{bmatrix} = RM_b \begin{bmatrix} b_1^T \\ b_2^T \end{bmatrix}$$

⇓ Hexagonal symmetry

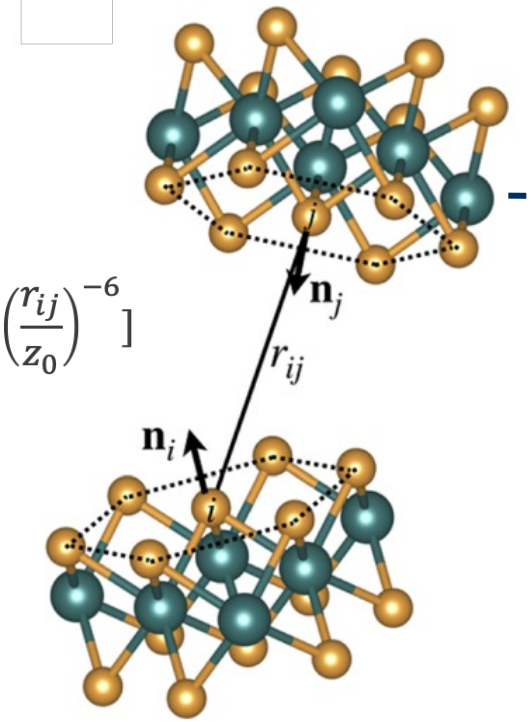
$$M_j = \begin{bmatrix} p_j & q_j \\ -q_j & p_j - q_j \end{bmatrix}$$

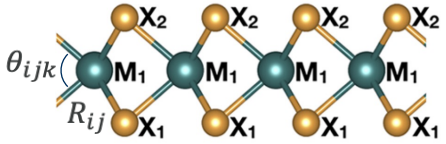
⇓ Coincidence

$$|\tilde{a}_i - \tilde{b}_i| < tolerance$$

Geometry optimization: Force-field method

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



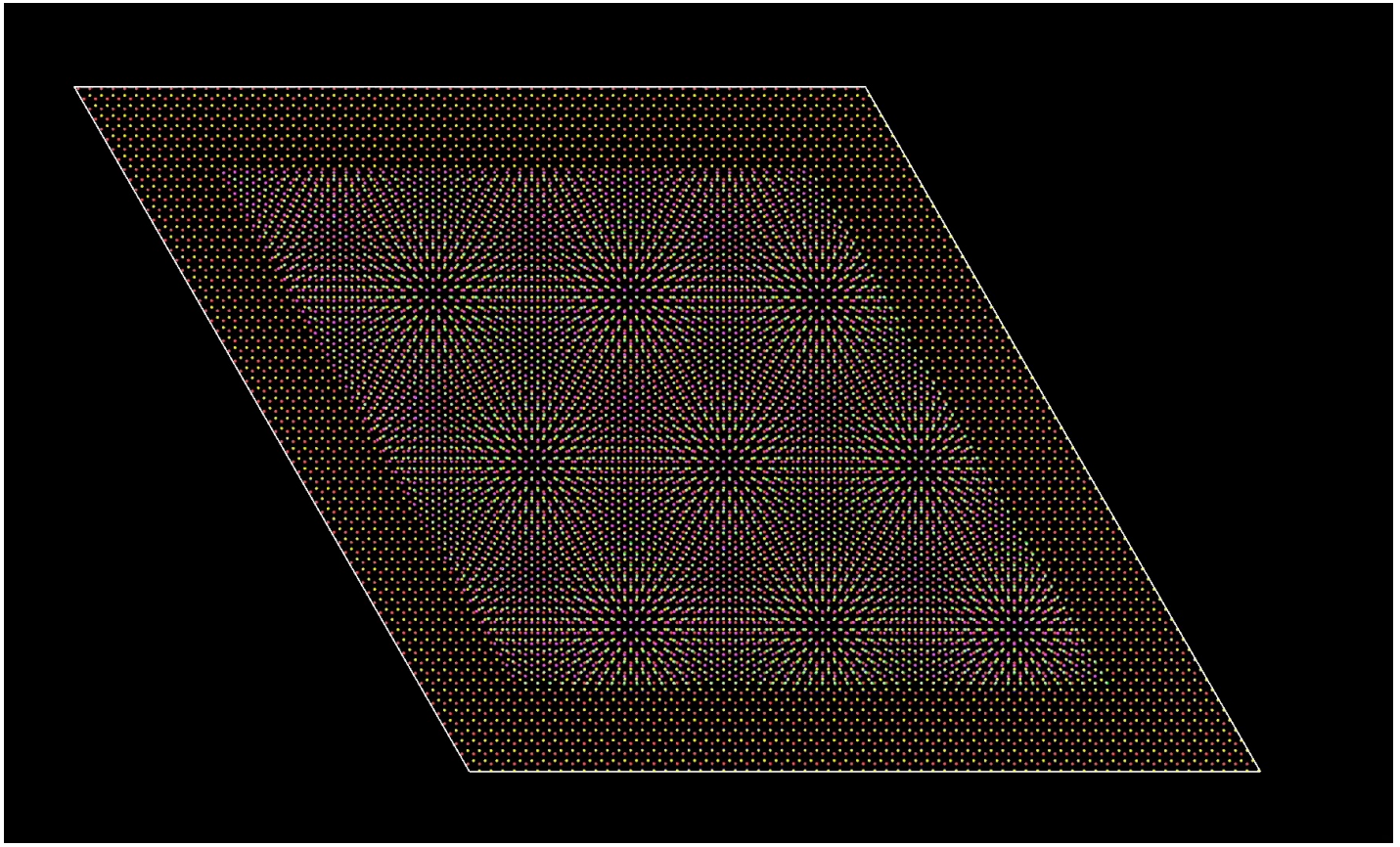


$$\sum_i \sum_{j>i} \phi_2(R_{ij}) + \sum_i \sum_{j\neq i} \sum_{k>j} \phi_3(R_{ij}, R_{ik}, \theta_{ijk})$$

Computational details

- Geometry optimisation:
  - Conjugate gradient method in LAMMPS
- Single point calculations (DFT):
  - GGA-PBE/TS in FHI-aims
- Single point calculations (DFTB):
  - SCC-DFTB theory level in DFTB+
  - QUASINANO2013 Slater-Koster parameters
  - Monkhorst Pack k-space

Boundary condition: flake



Unrelaxed $MoS_2/MoSe_2$ at 60°

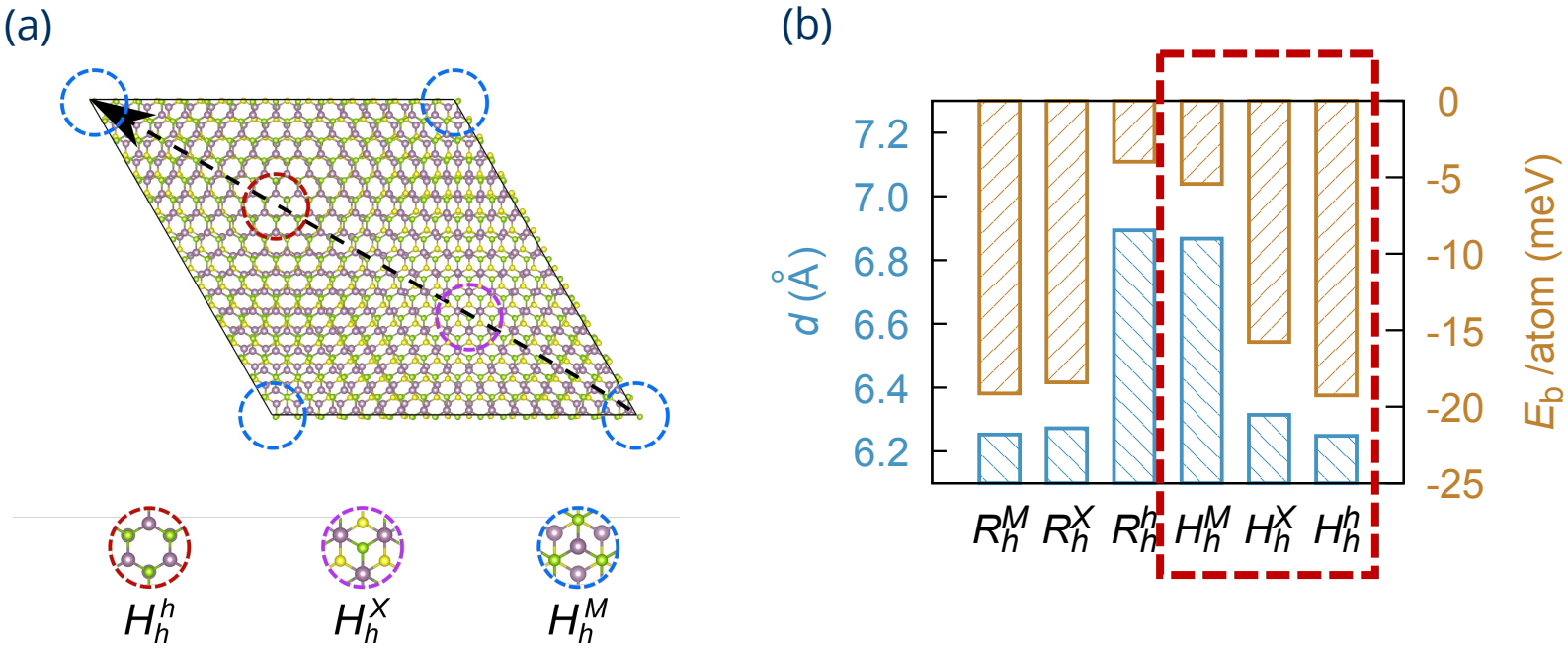
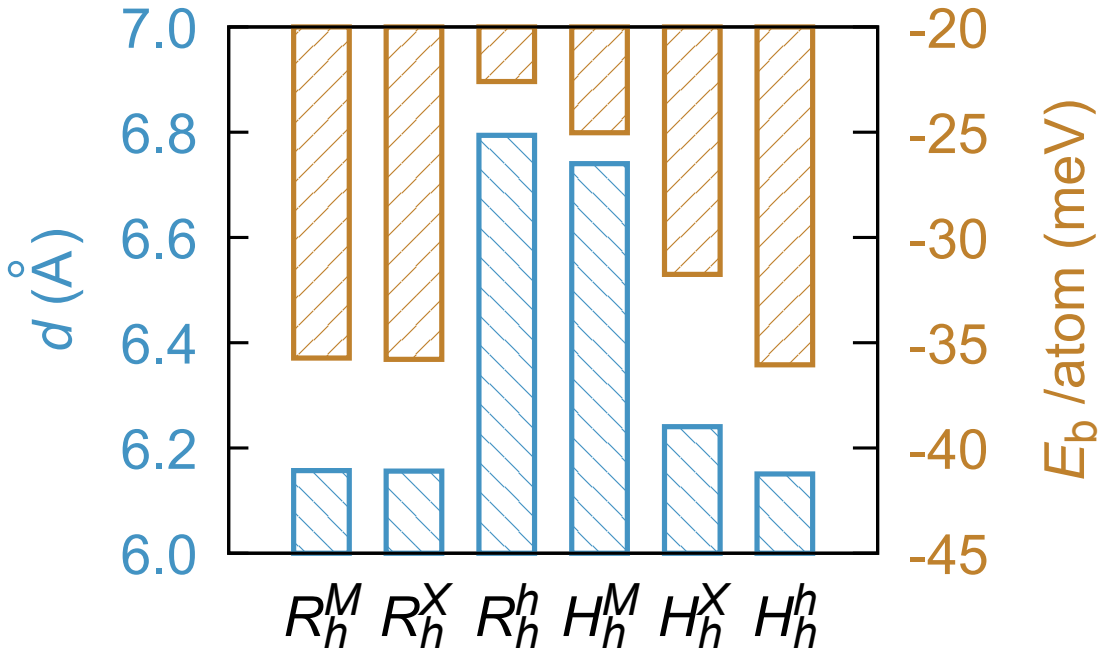


Fig 1. (a) 3 high-symmetry stackings and transition stacking regions. (b) Interlayer distance and binging energy of corresponding high-symmetry stackings.

Backup



- Fig 1. Interlayer distance and binding energy of high-symmetry stackings of MoS_2/WS_2