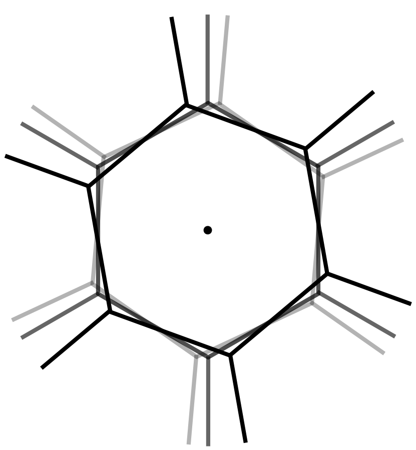
Moiré of moiré: modeling mechanical and electronic properties of twisted trilayer vdW heterostructures

Ziyan (Zoe) Zhu
Harvard University

Theory and Computation for 2D Materials Workshop,
UCLA IPAM, Los Angeles, CA
01/15/2020



Acknowledgement

- Stephen Carr, Shiang Fang, Efthimios Kaxiras (Harvard)

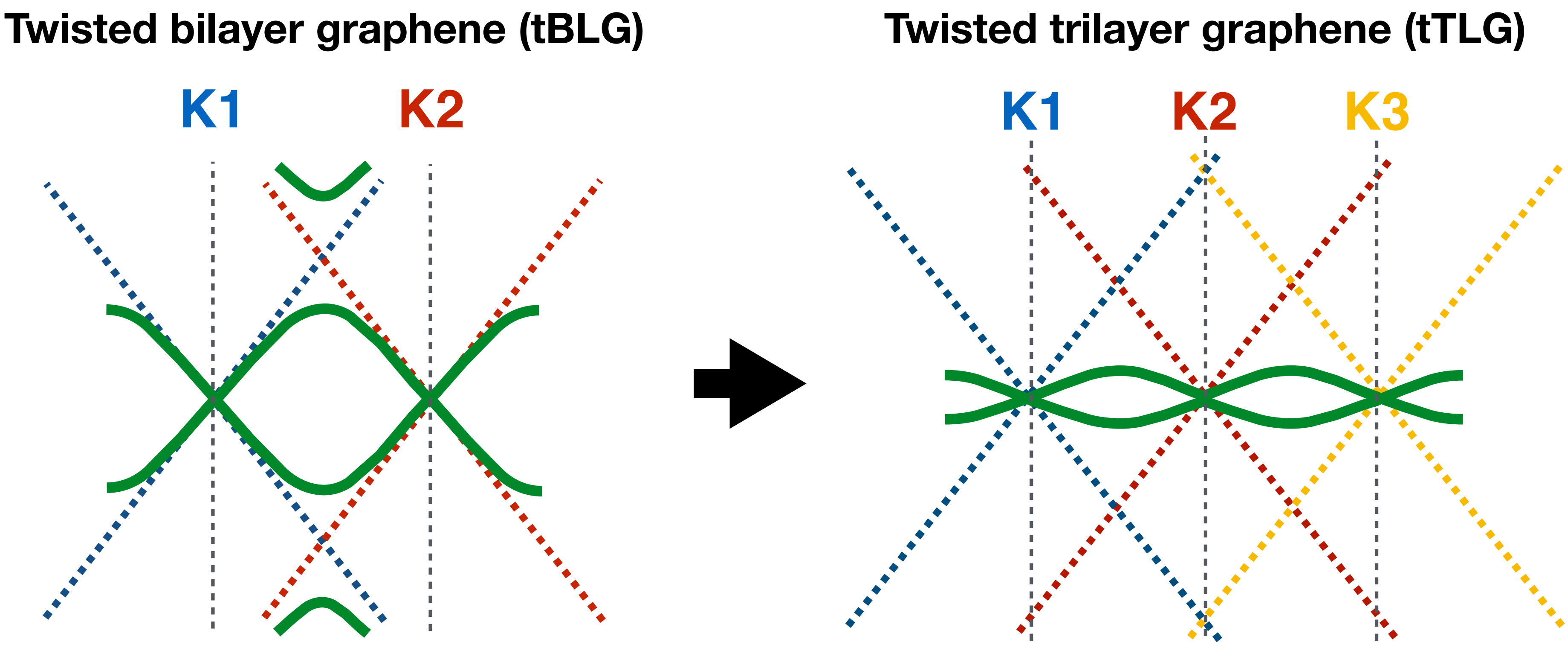
- Paul Cazeaux (U of Kansas), Mitchell Luskin (UMN), Daniel Massatt (UChicago)

- Xi Zhang, Kan-Ting Tsai, Ke Wang (UMN)

Outline

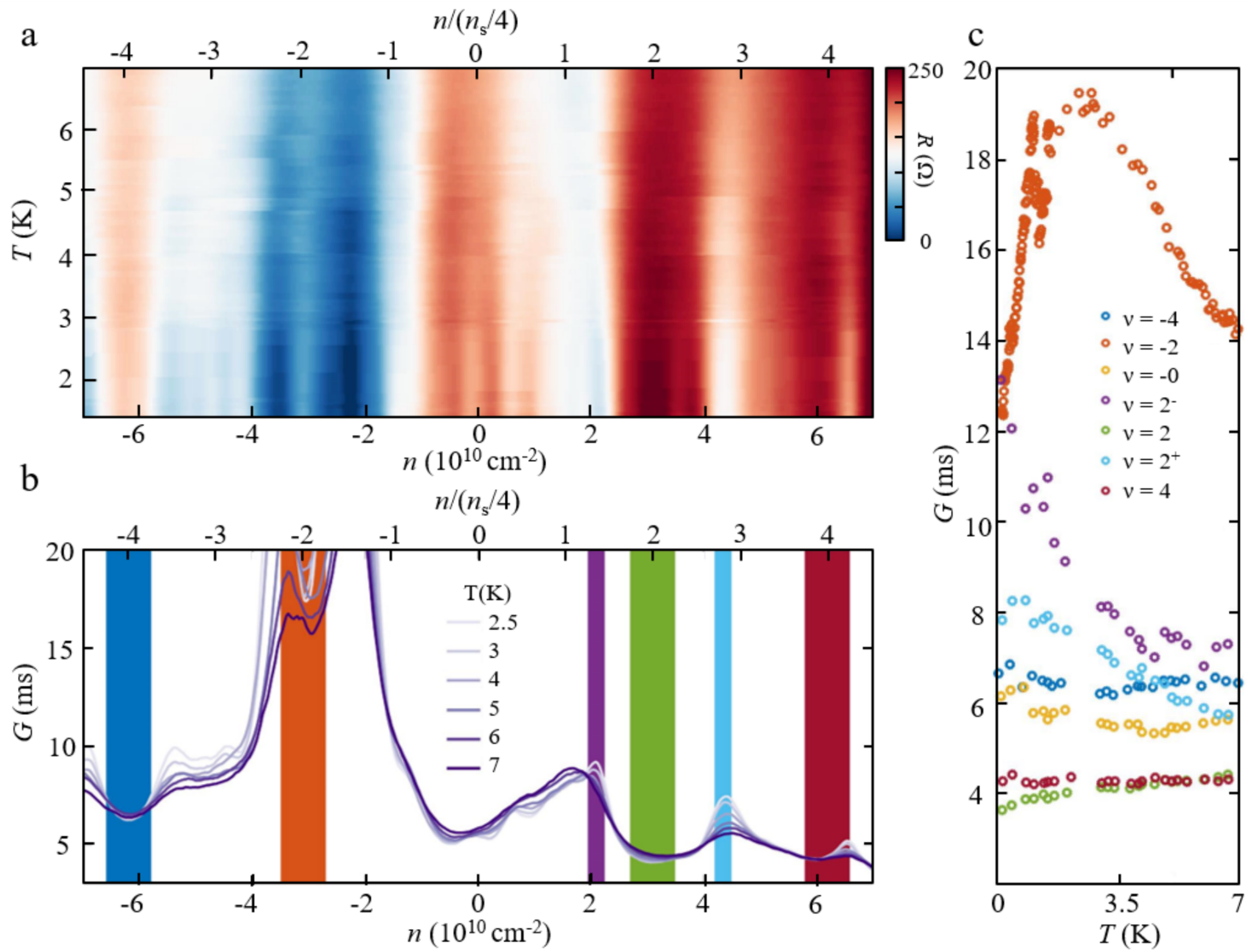
- Introduction
- Relaxation in general trilayers (brief review, focusing on $\theta_{12} = \theta_{23}$)
- Electronic structures in tTLG
- Conclusion & future directions

A new type of van der Waals heterostructures: twisted trilayer graphene (tTLG)



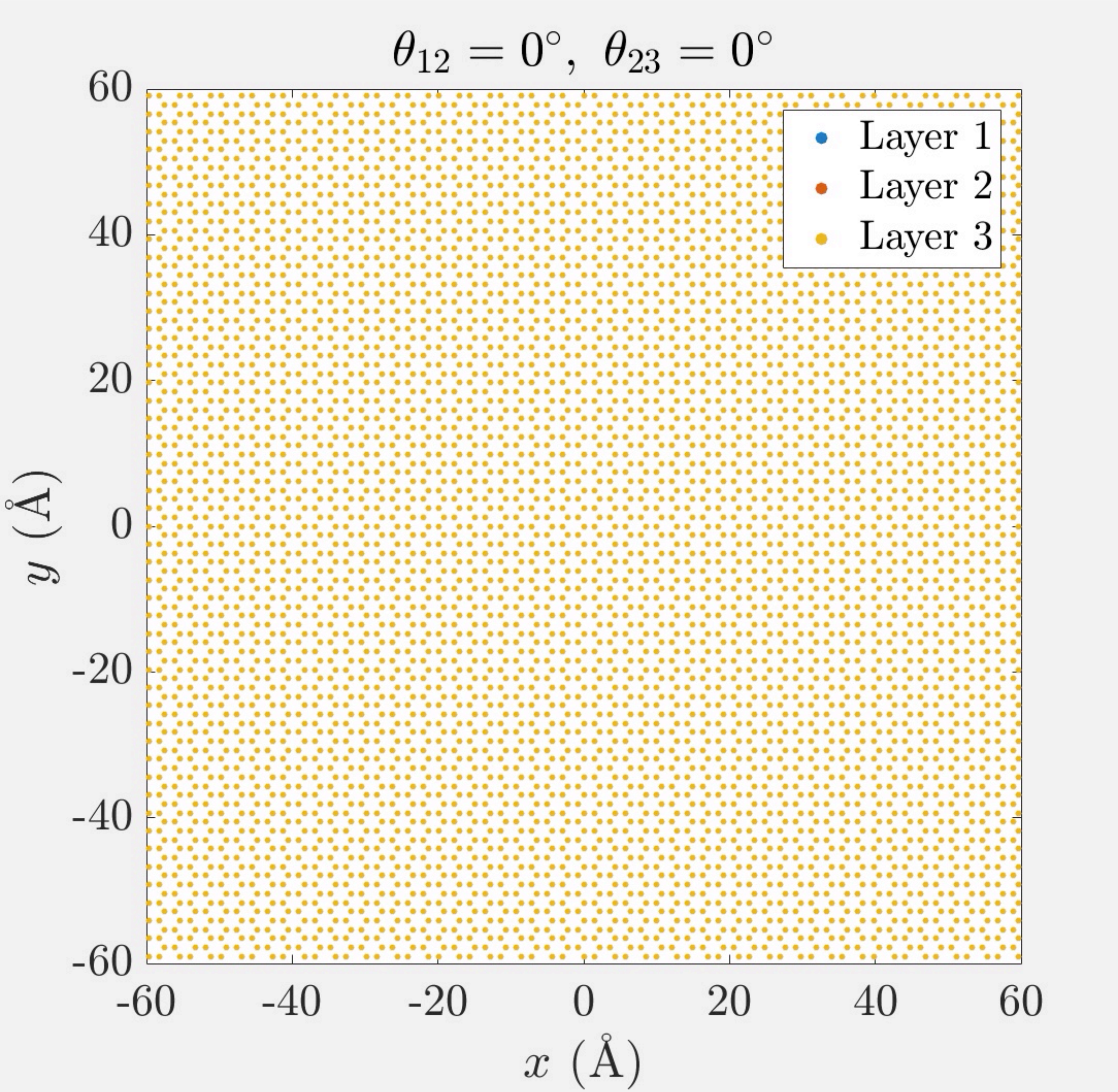
- New candidate for unconventional correlated states: flat bands, high density of states

Experimental Realization of Strongly Correlated States in tTLG



- Superconductivity and strongly correlated insulating states observed at half-filling of moiré of moiré superlattice

Experiment by K. Wang, K-T. Tsai, and X. Zhang: ArXiv:1912.03375



Modeling challenges

- Aperiodicity
- Large system size

Mechanical Relaxation in Twisted Trilayers

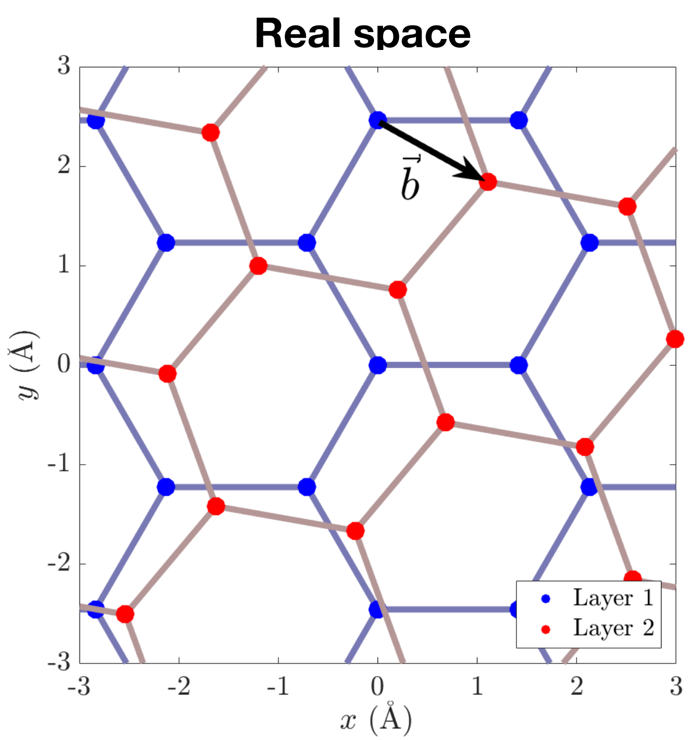
Ziyan Zhu, Paul Cazeaux, Mitchell Luskin, Efthimios Kaxiras, arXiv:1911.05324

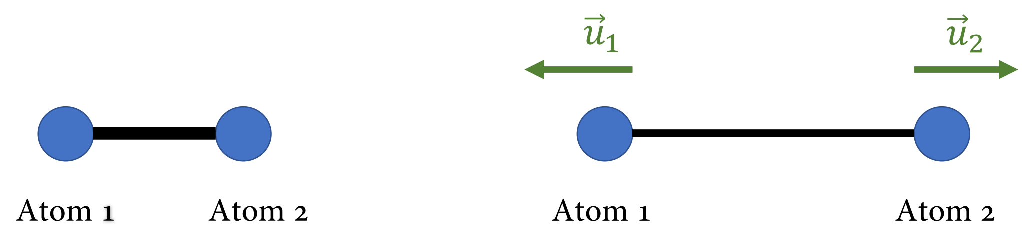
Goal: minimize the total energy as a function of the relaxation displacement vectors

$$E^{\mathrm{tot}}(\boldsymbol{u}) = E^{\mathrm{inter}}(\boldsymbol{u}) + E^{\mathrm{intra}}(\boldsymbol{u})$$

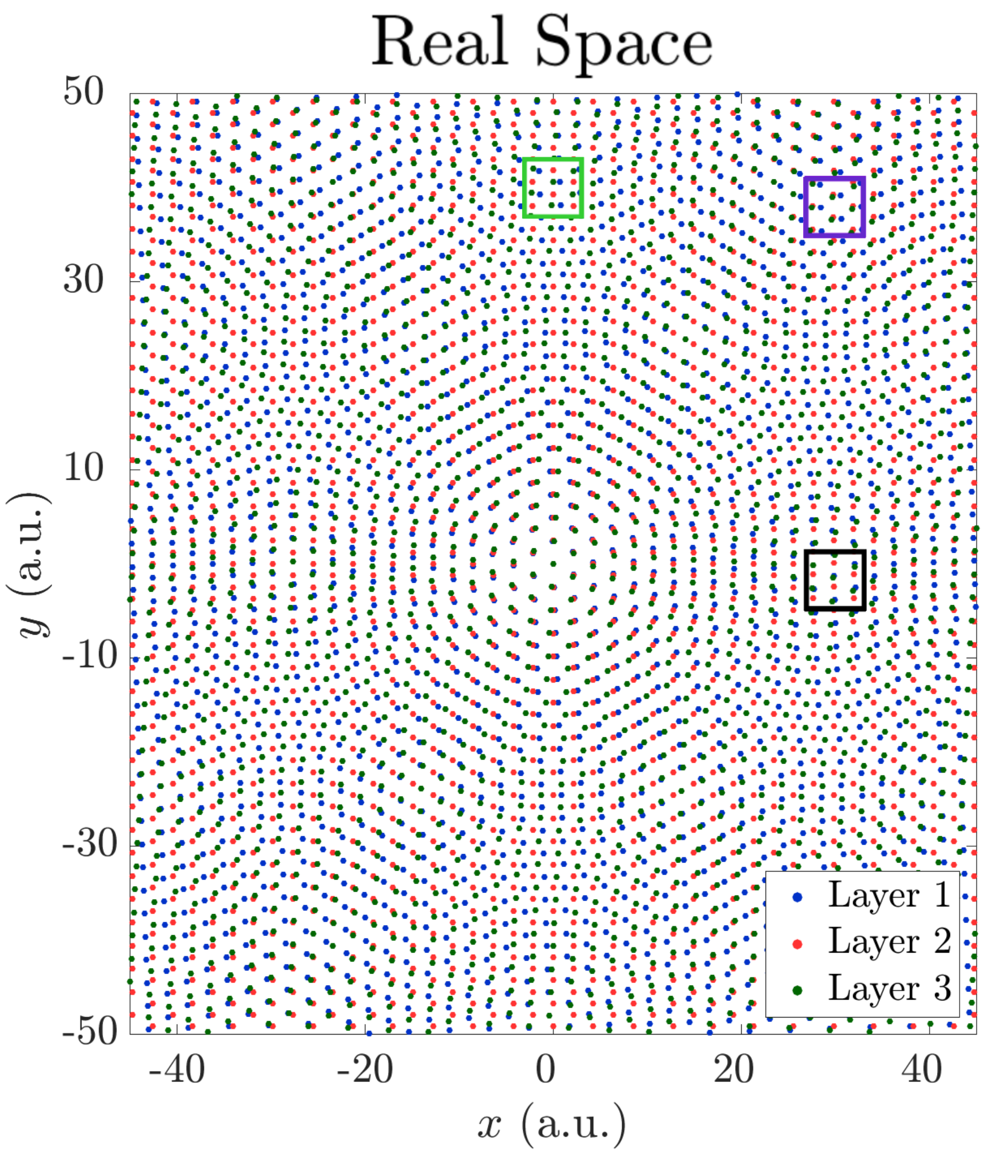
Layer misalignments
(equilibrium stacking: AB/BA)

Elastic energy

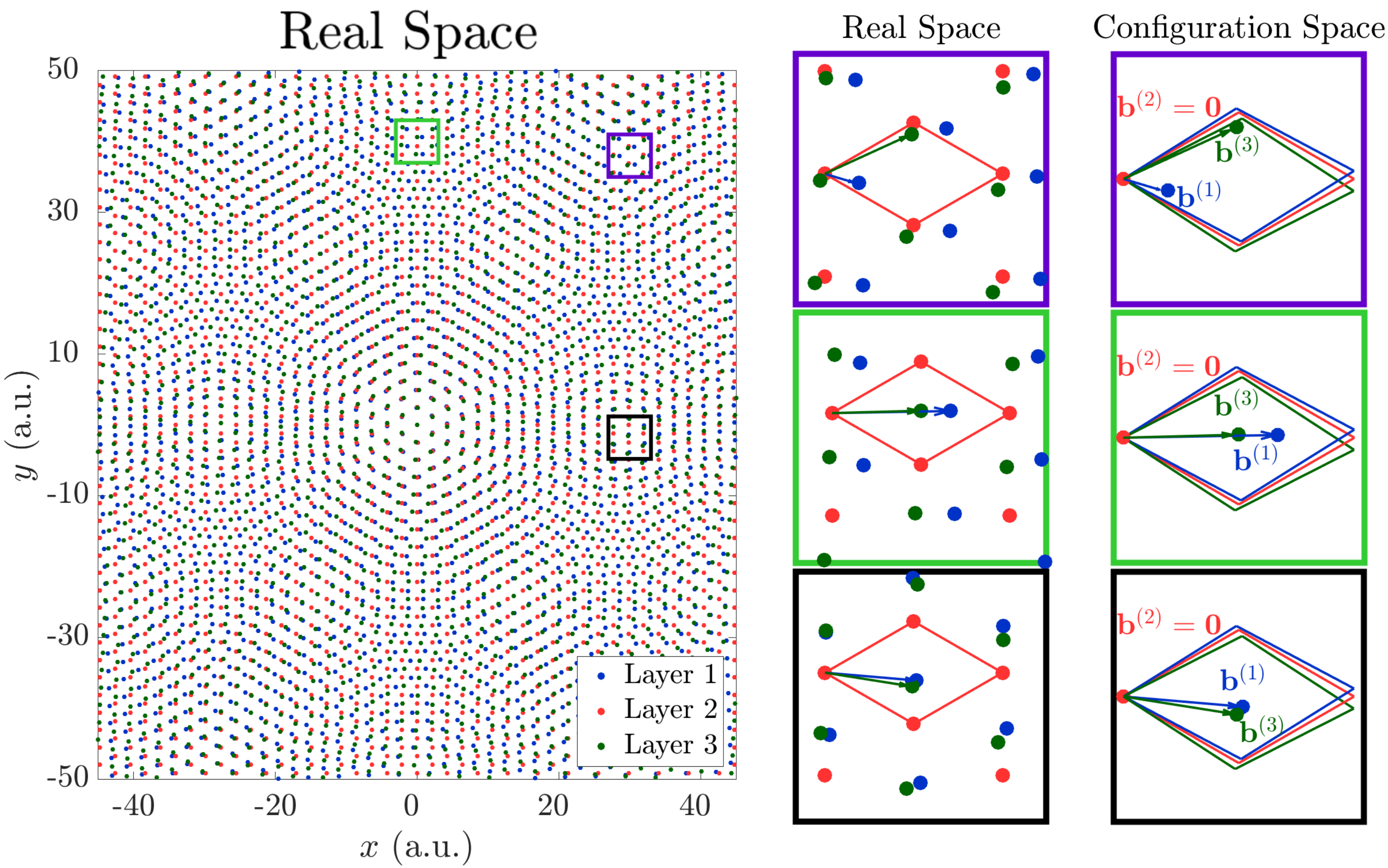




Configuration space: a description of aperiodic systems



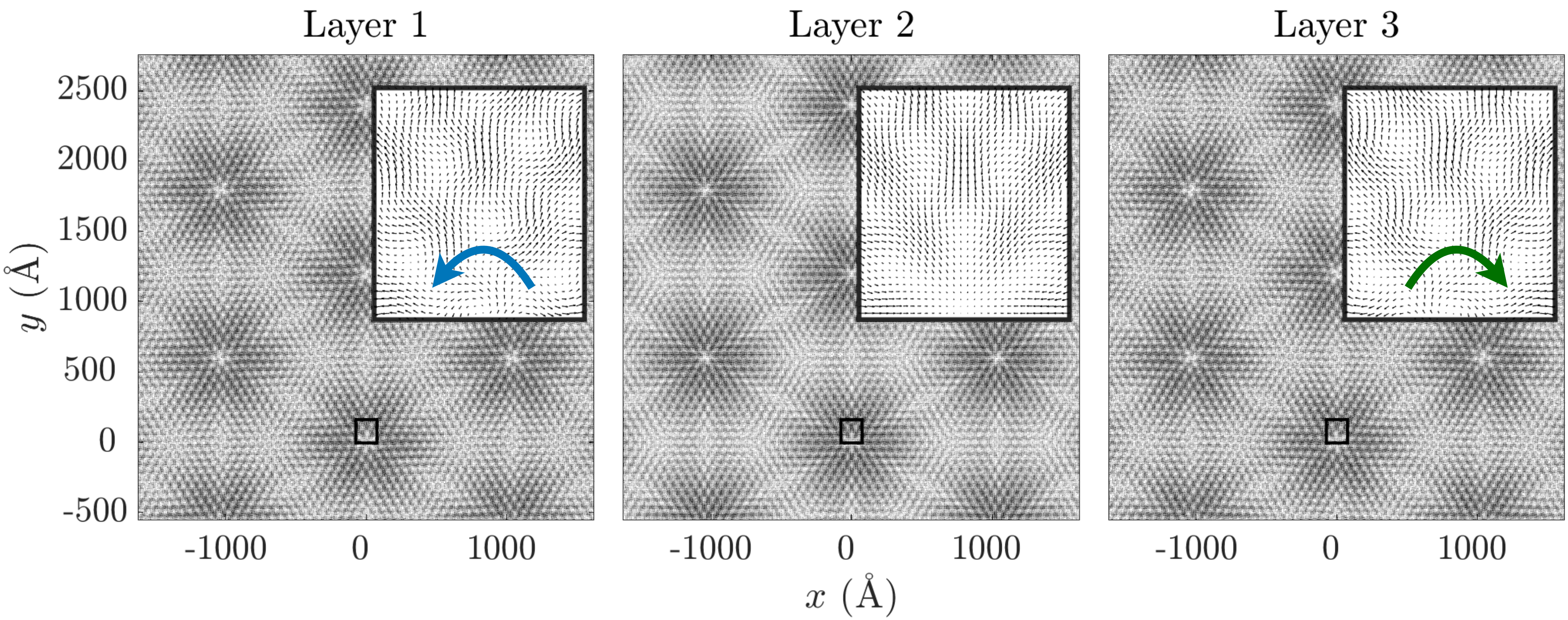
Configuration space:
a description of aperiodic systems



- Local environment of each atomic position

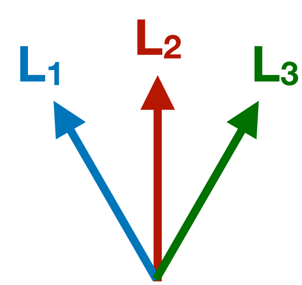
Real space relaxation pattern

WSe2 trilayer, $\theta_{12} = \theta_{23} = 3^\circ$



Zooming in on the AAA spots

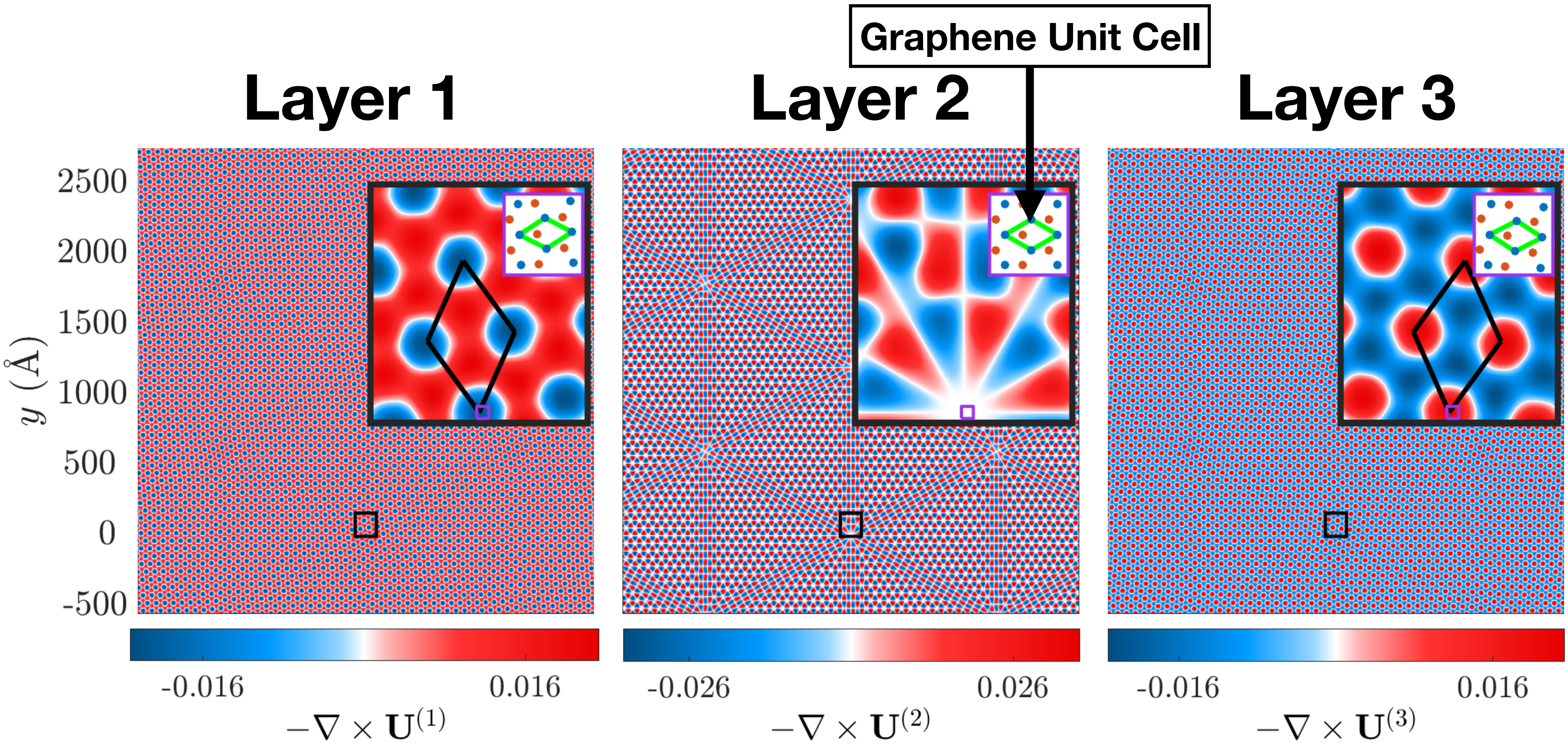
Rotation convention:



Ziyan Zhu, Paul Cazeaux, Mitchell Luskin, Efthimios Kaxiras, arXiv:1911.05324

Real space relaxation pattern

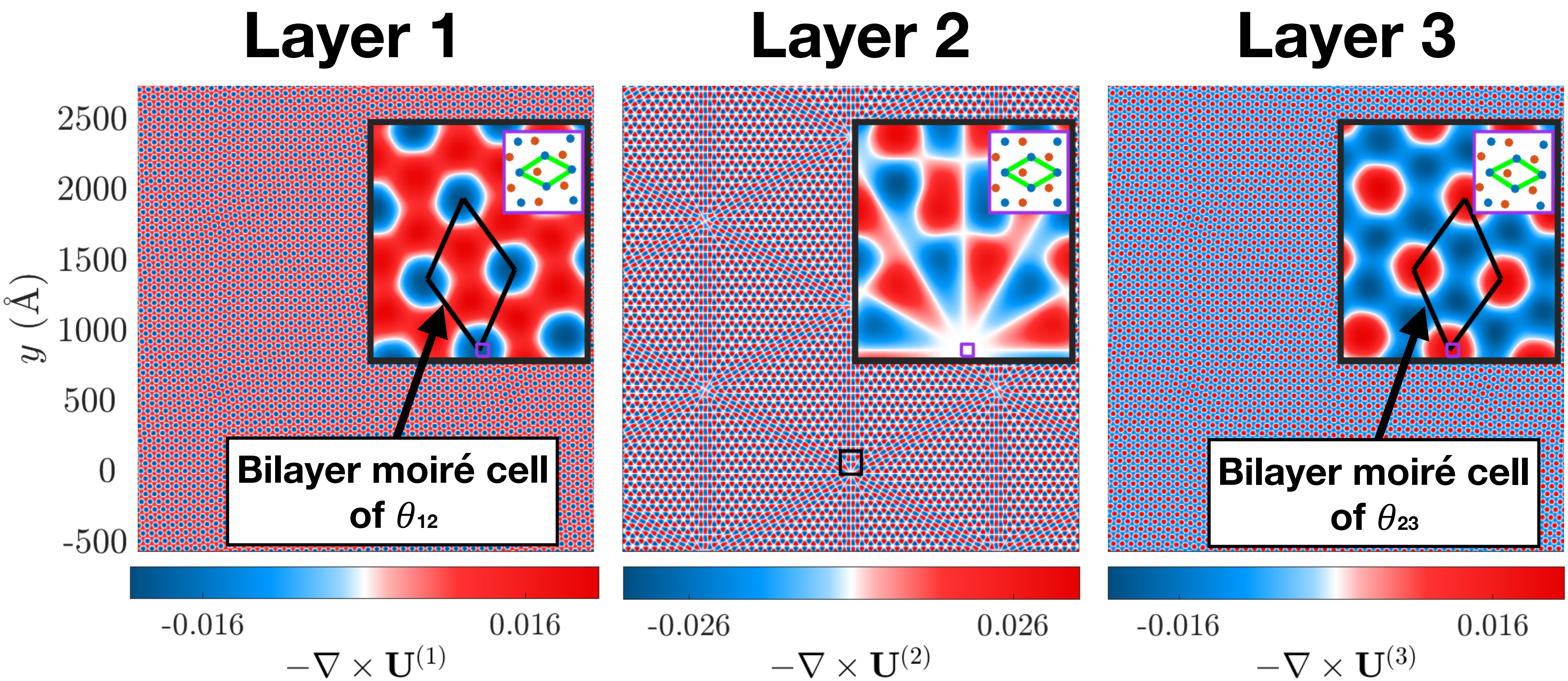
WSe2 trilayer, $\theta_{12} = \theta_{23} = 3^\circ$



Ziyan Zhu, Paul Cazeaux, Mitchell Luskin, Efthimios Kaxiras, arXiv:1911.05324

Real space relaxation pattern

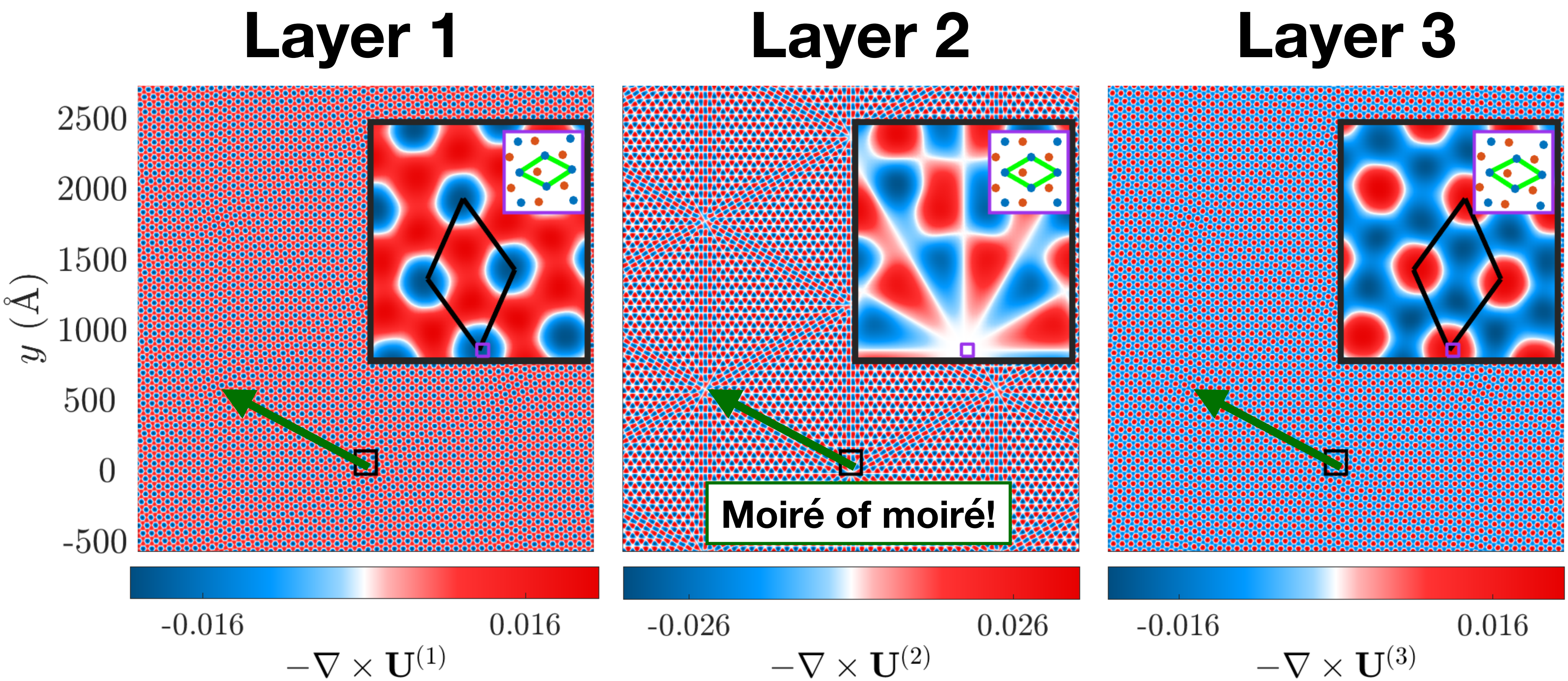
WSe2 trilayer, $\theta_{12} = \theta_{23} = 3^\circ$



Ziyan Zhu, Paul Cazeaux, Mitchell Luskin, Efthimios Kaxiras, arXiv:1911.05324

Real space relaxation pattern

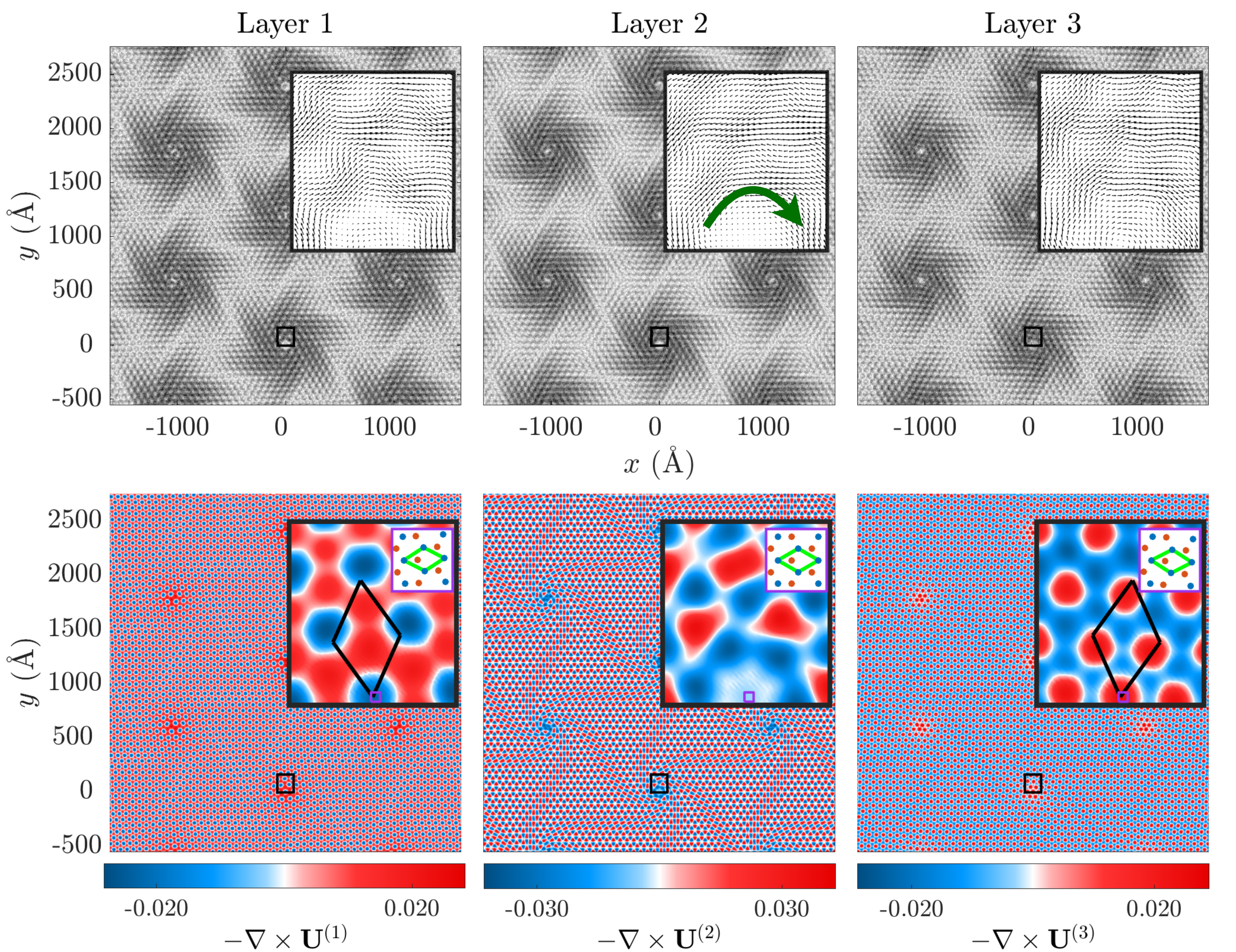
WSe2 trilayer, $\theta_{12} = \theta_{23} = 3^{\circ}$



Ziyan Zhu, Paul Cazeaux, Mitchell Luskin, Efthimios Kaxiras, arXiv:1911.05324

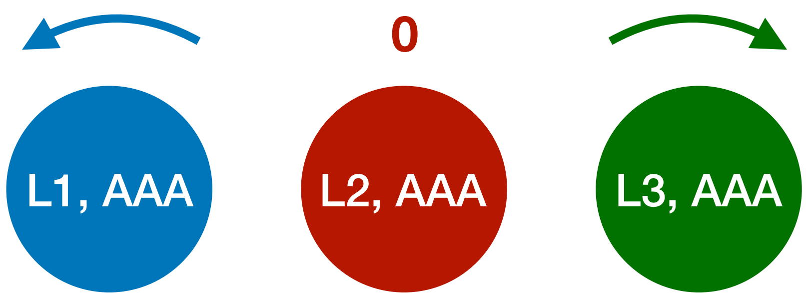
Not the whole story!

Spontaneous symmetry breaking at $\theta_{12} = \theta_{23}$

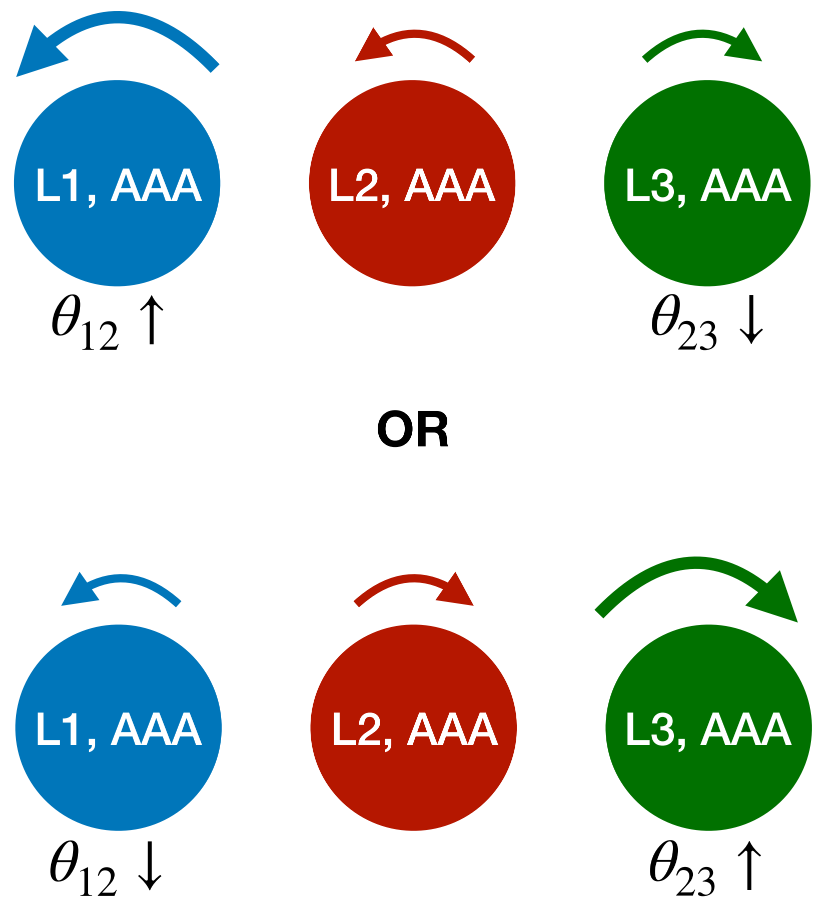


Why does this happen?

Symmetric

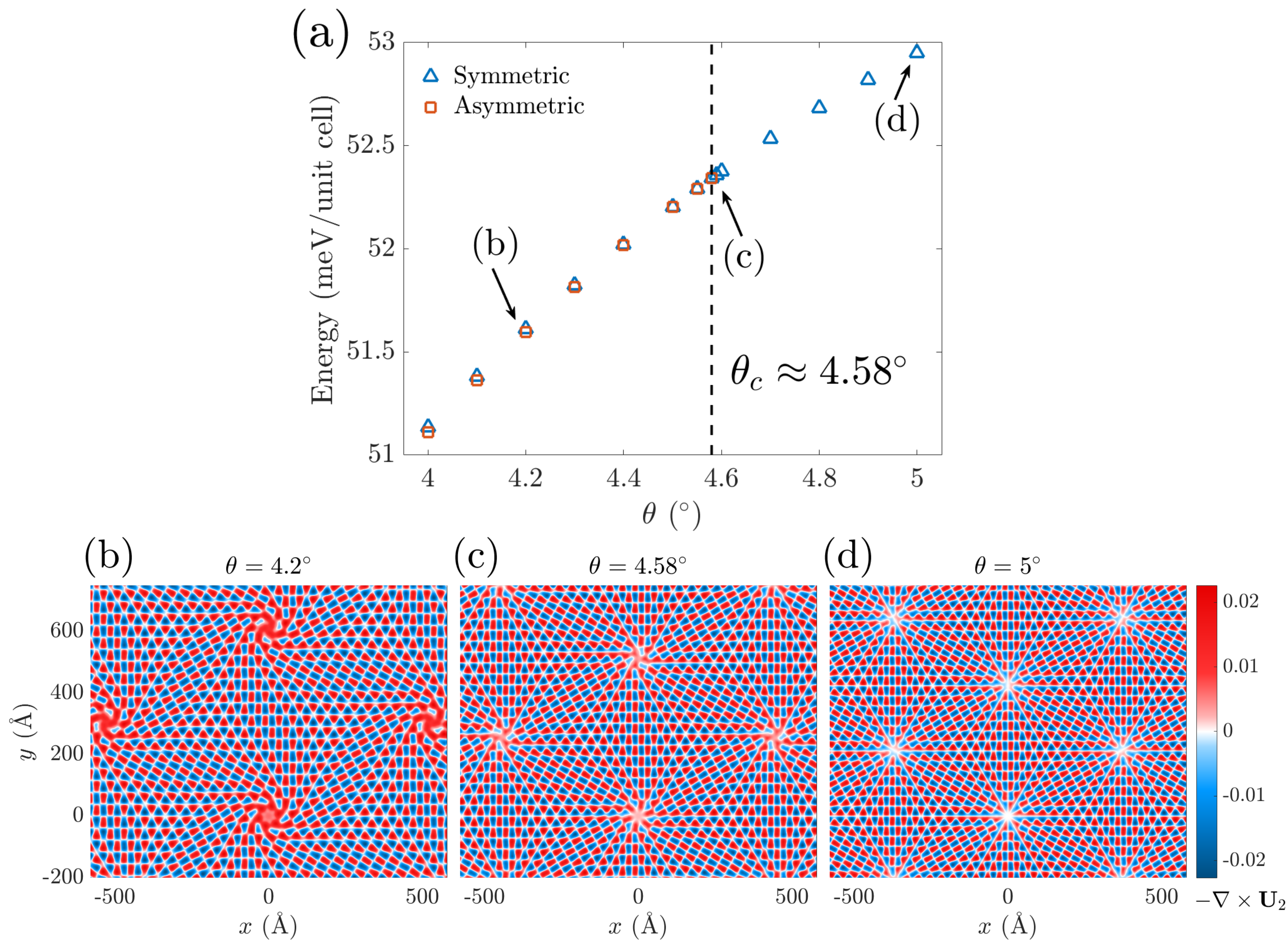


Asymmetric



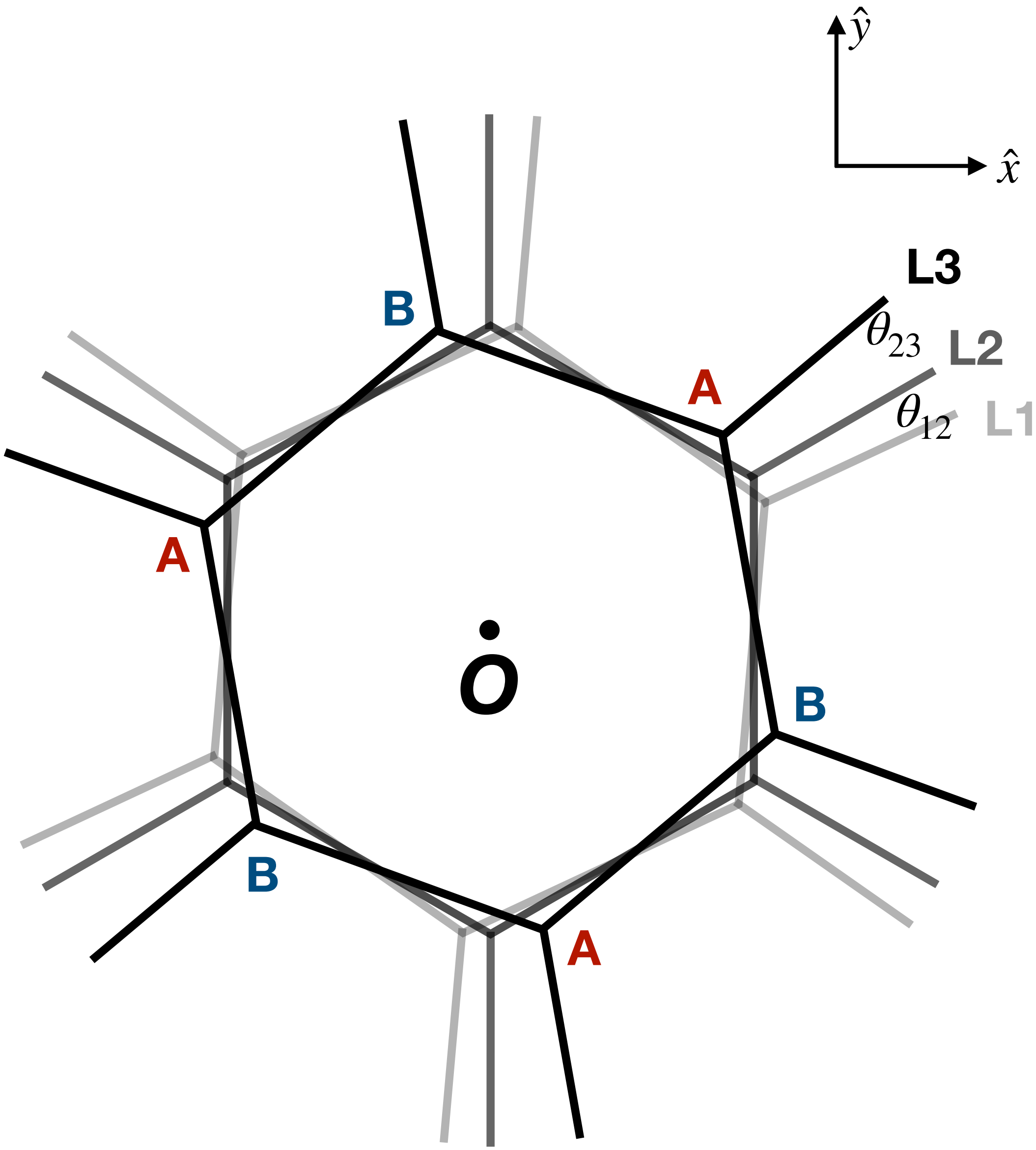
Physically: competition between intralayer (elastic) energy and interlayer (misfit) energy

Bifurcation of the solution for $\theta_{12} = \theta_{23}$

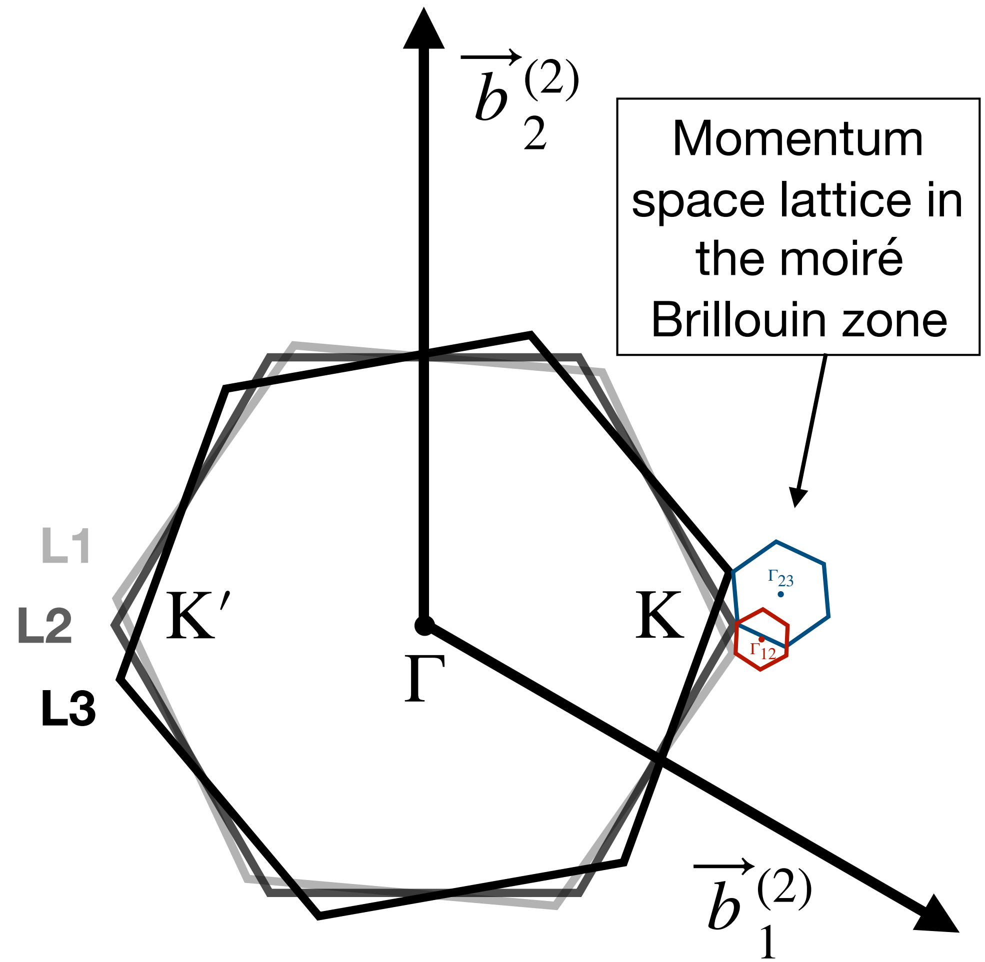


Electronic Structures in Twisted Trilayer Graphene

Set up: monolayer in real and reciprocal space



Real space



Reciprocal space

Real space model (tight-binding)

$$H = \boxed{\sum_{l=1}^{3} H^l} + \boxed{\sum_{l=1,2} \left(V^{l,l+1} + V^{l+1,l}\right)}$$

Intralayer:

$$H^\ell = -t \sum_{\boldsymbol{R}^{(\ell)}} c^\dagger_{\ell,A}(\boldsymbol{R}^{(\ell)})[c_{\ell,B}(\boldsymbol{R}^{(\ell)}) + c_{\ell,B}(\boldsymbol{R}^{(\ell)} - \boldsymbol{a}_1^{(\ell)}) + c_{\ell,B}(\boldsymbol{R}^{(\ell)} - \boldsymbol{a}_2^{(\ell)})] + h.c.$$

Interlayer:

$$V^{ij} = \sum_{\boldsymbol{R}^{(i)},\alpha,\boldsymbol{R}^{(j)},\beta} c^\dagger_{i,\alpha}(\boldsymbol{R}^{(i)}) t^{ij}_{\alpha\beta}(\boldsymbol{R}^{(i)}, \boldsymbol{R}^{(j)}) c_{j,\beta}(\boldsymbol{R}^{(j)})$$

Plane-wave expansion:

$$c^\dagger_{\ell,\boldsymbol{k}^{(\ell)},\alpha} = \frac{1}{\sqrt{A_{\mathrm{BZ},\ell}}} \sum_{\boldsymbol{R}^{(\ell)}} e^{-i\boldsymbol{k}^{(\ell)}\cdot(\boldsymbol{R}^{(\ell)}+\boldsymbol{\tau}_\alpha^{(\ell)})} c^\dagger_{\ell,\alpha}(\boldsymbol{R}^{(\ell)})$$

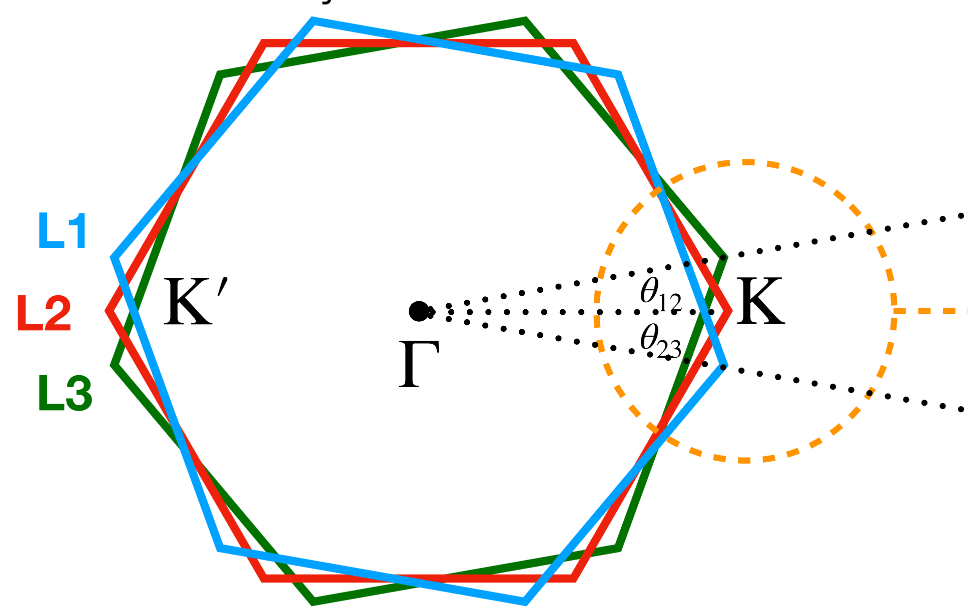
$$c_{\ell,\boldsymbol{k}^{(\ell)},\alpha} = \frac{1}{\sqrt{A_{\mathrm{BZ},\ell}}} \sum_{\boldsymbol{R}^{(\ell)}} e^{i\boldsymbol{k}^{(\ell)}\cdot(\boldsymbol{R}^{(\ell)}+\boldsymbol{\tau}_\alpha^{(\ell)})} c_{\ell,\alpha}(\boldsymbol{R}^{(\ell)})$$

Momentum space model

$$H_{\mathrm{K}} = \begin{array}{c} \begin{array}{ccc} \text{L1} & \text{L2} & \text{L3} \end{array} \\ \begin{bmatrix} H_D^1(\boldsymbol{k}) & T^{12}(\boldsymbol{r}) & 0 \\ T^{12\dagger}(\boldsymbol{r}) & H_D^2(\boldsymbol{k}) & T^{23}(\boldsymbol{r}) \\ 0 & T^{23\dagger}(\boldsymbol{r}) & H_D^3(\boldsymbol{k}) \end{bmatrix} \end{array} \begin{array}{c} \text{L1} \\ \text{L2} \\ \text{L3} \end{array}$$

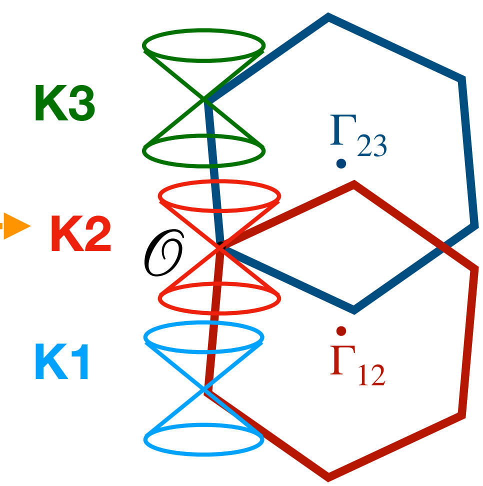
Diagonal blocks: intralayer hopping: rotated Dirac Hamiltonian

Monolayer Brillouin zone



$\theta_{12} = \theta_{23}$

Supercell Brillouin zone



$$H_D^1(\boldsymbol{q}^{(1)}) = v_F \begin{bmatrix} 0 & e^{i\theta_{12}} q_+^{(1)} \\ e^{-i\theta_{12}} q_-^{(1)} & 0 \end{bmatrix}$$

$$H_D^2(\boldsymbol{q}^{(2)}) = v_F \begin{bmatrix} 0 & q_+^{(2)} \\ q_-^{(2)} & 0 \end{bmatrix}$$

$$H_D^3(\boldsymbol{q}^{(3)}) = v_F \begin{bmatrix} 0 & e^{-i\theta_{23}} q_+^{(3)} \\ e^{i\theta_{23}} q_-^{(3)} & 0 \end{bmatrix}$$

Momentum space model

$$
\begin{array}{cccc}
\text{L1} & \text{L2} & \text{L3} & \\
\multicolumn{3}{c}{H_{\mathrm{K}} = \begin{bmatrix} H_D^1(\boldsymbol{k}) & T^{12}(\boldsymbol{r}) & 0 \\ T^{12\dagger}(\boldsymbol{r}) & H_D^2(\boldsymbol{k}) & T^{23}(\boldsymbol{r}) \\ 0 & T^{23\dagger}(\boldsymbol{r}) & H_D^3(\boldsymbol{k}) \end{bmatrix}} & \begin{array}{c} \text{L1} \\ \text{L2} \\ \text{L3} \end{array}
\end{array}
$$

Layer indices (adjacent layers)

Off-diagonal blocks: interlayer hopping

$$
T^{ij}_{\alpha\beta}(\boldsymbol{k}^{(i)}, \boldsymbol{k}^{(j)}) = \sqrt{A_{\mathrm{BZ},i} A_{\mathrm{BZ},j}} \sum_{\boldsymbol{G}^{(i)}, \boldsymbol{G}^{(j)}} e^{-i\boldsymbol{G}^{(i)} \cdot \boldsymbol{\tau}_{\alpha}^{(i)}} t^{ij}_{\alpha\beta}(\boldsymbol{k}^{(i)} + \boldsymbol{G}^{(i)}) e^{-i\boldsymbol{G}^{(j)} \cdot \boldsymbol{\tau}_{\beta}^{(i)}} \boxed{\delta(\boldsymbol{k}^{(i)} + \boldsymbol{G}^{(i)}, \boldsymbol{k}^{(j)} + \boldsymbol{G}^{(j)})}
$$

Scattering selection rule

Sublattices

Momentum space model

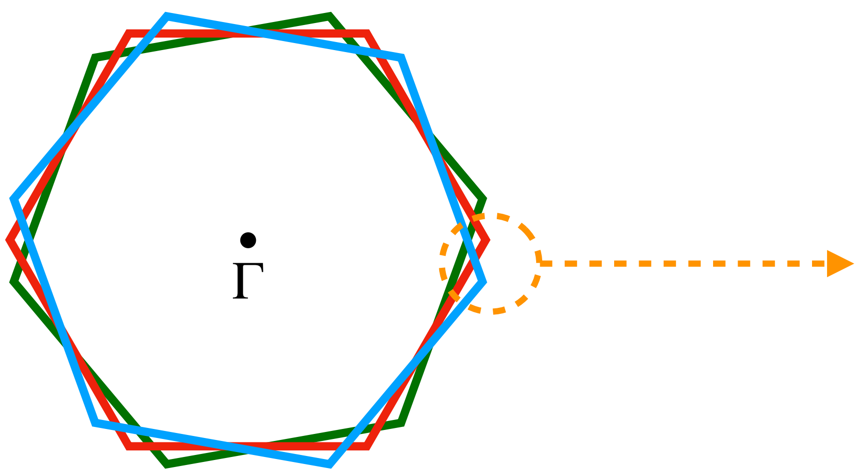
$$
\begin{array}{cc}
 & \begin{array}{ccc} \text{L1} & \text{L2} & \text{L3} \end{array} \\
H_{\mathrm{K}} = & \begin{bmatrix} H_D^1(\boldsymbol{k}) & T^{12}(\boldsymbol{r}) & 0 \\ T^{12\dagger}(\boldsymbol{r}) & H_D^2(\boldsymbol{k}) & T^{23}(\boldsymbol{r}) \\ 0 & T^{23\dagger}(\boldsymbol{r}) & H_D^3(\boldsymbol{k}) \end{bmatrix} \begin{array}{c} \text{L1} \\ \text{L2} \\ \text{L3} \end{array}
\end{array}
$$

Off-diagonal blocks: interlayer hopping (low-energy limit, first shell)

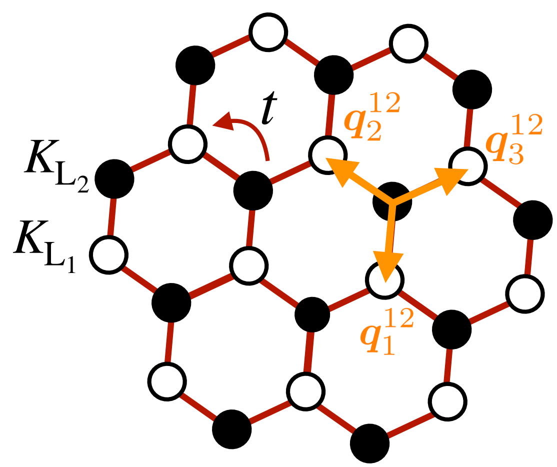
$$\boldsymbol{q}^{(i)} = \boldsymbol{k}^{(i)} - K_{\mathrm{L_i}}$$

$$T^{ij}_{\alpha\beta}(\boldsymbol{q}^{(i)}, \boldsymbol{q}^{(j)}) = \sum_{n=1}^{3} T^{\boldsymbol{q}_n^{ij}}_{\alpha\beta} \boxed{\delta_{\boldsymbol{q}^{(i)} - \boldsymbol{q}^{(j)}, \boldsymbol{q}_n^{ij}}}$$

k space NN coupling



Monolayer Brillouin zone



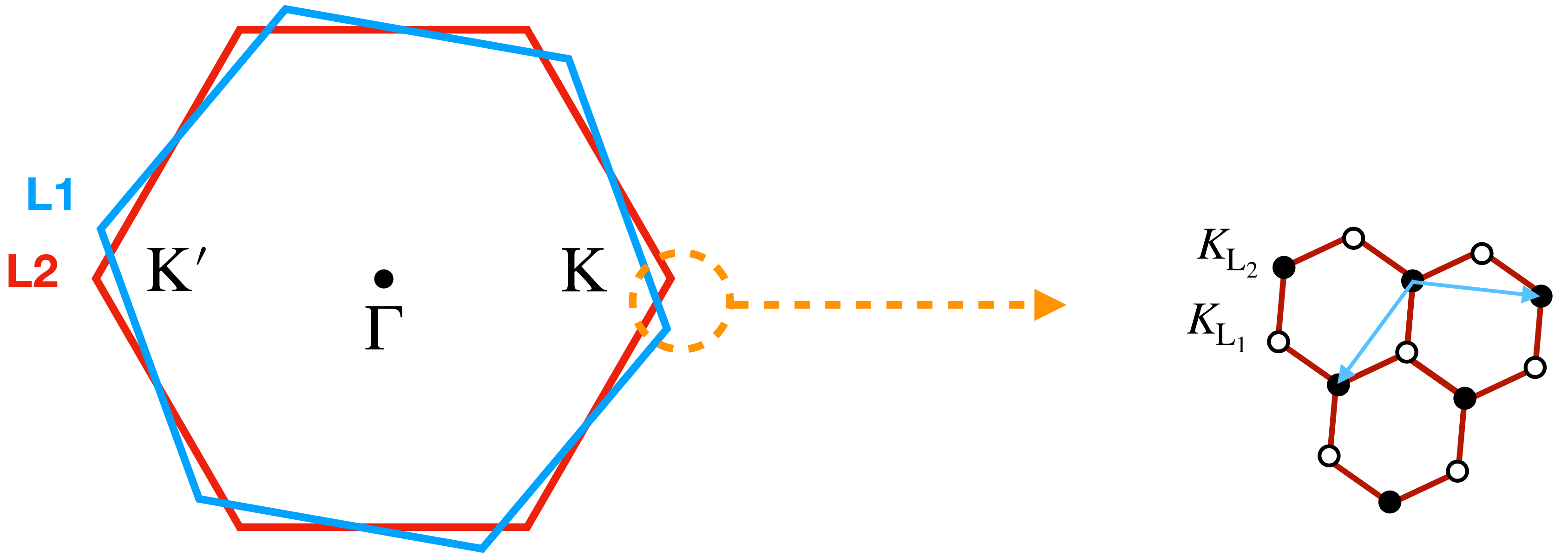
L1+L2 supercell Brillouin zone

k degree of freedom in bilayers vs. trilayers

$$\delta(\boldsymbol{k}^{(i)} + \boldsymbol{G}^{(i)}, \boldsymbol{k}^{(j)} + \boldsymbol{G}^{(j)})$$

Monolayer Brillouin zone

L1+L2 supercell Brillouin zone



○ Layer 1 momentum

● Layer 2 momentum

→ L1+L2 SC reciprocal lattice vector

Bilayer scattering selection rule

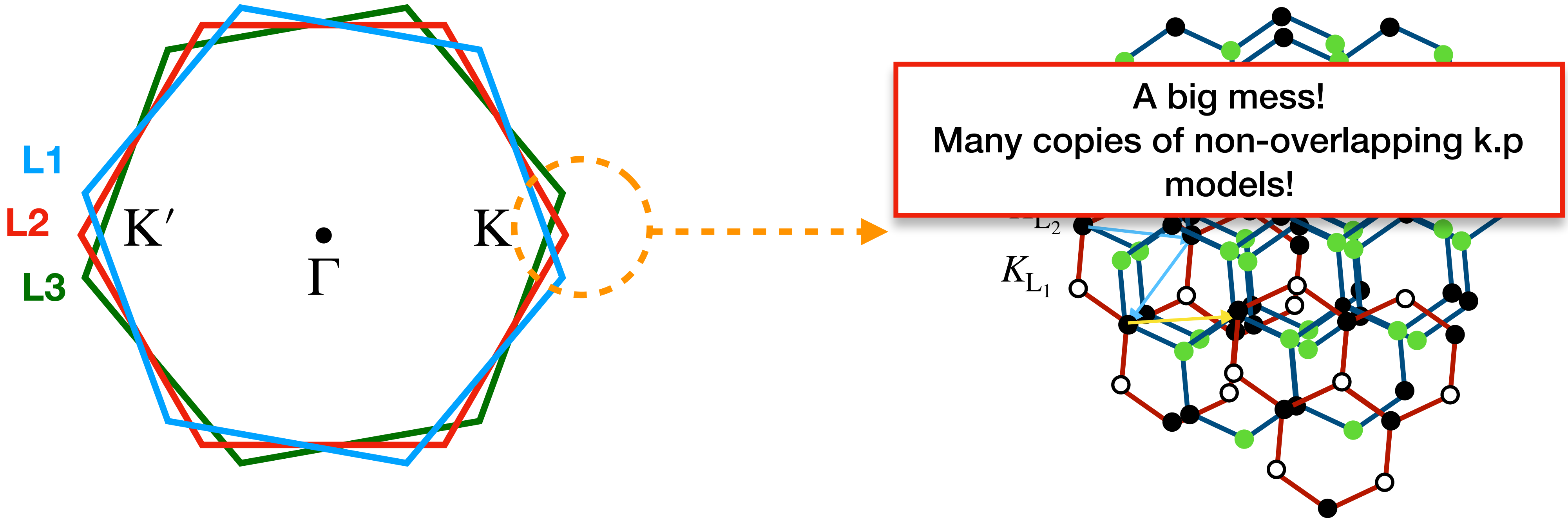
$$\boldsymbol{G}^{(1)} + \boldsymbol{q}^{(1)} + K_{\mathrm{L}_1} = \boldsymbol{G}^{(2)} + \boldsymbol{q}^{(2)} + K_{\mathrm{L}_2}$$

k degree of freedom in bilayers vs. trilayers

$$\delta(\boldsymbol{k}^{(i)} + \boldsymbol{G}^{(i)}, \boldsymbol{k}^{(j)} + \boldsymbol{G}^{(j)})$$

Monolayer Brillouin zone

L1+L2+L3 supercell Brillouin zone



- ○ Layer 1 momentum
- ● Layer 2 momentum
- ● Layer 3 momentum
- → L1+L2 SC reciprocal lattice vector
- → L2+L3 SC reciprocal lattice vector

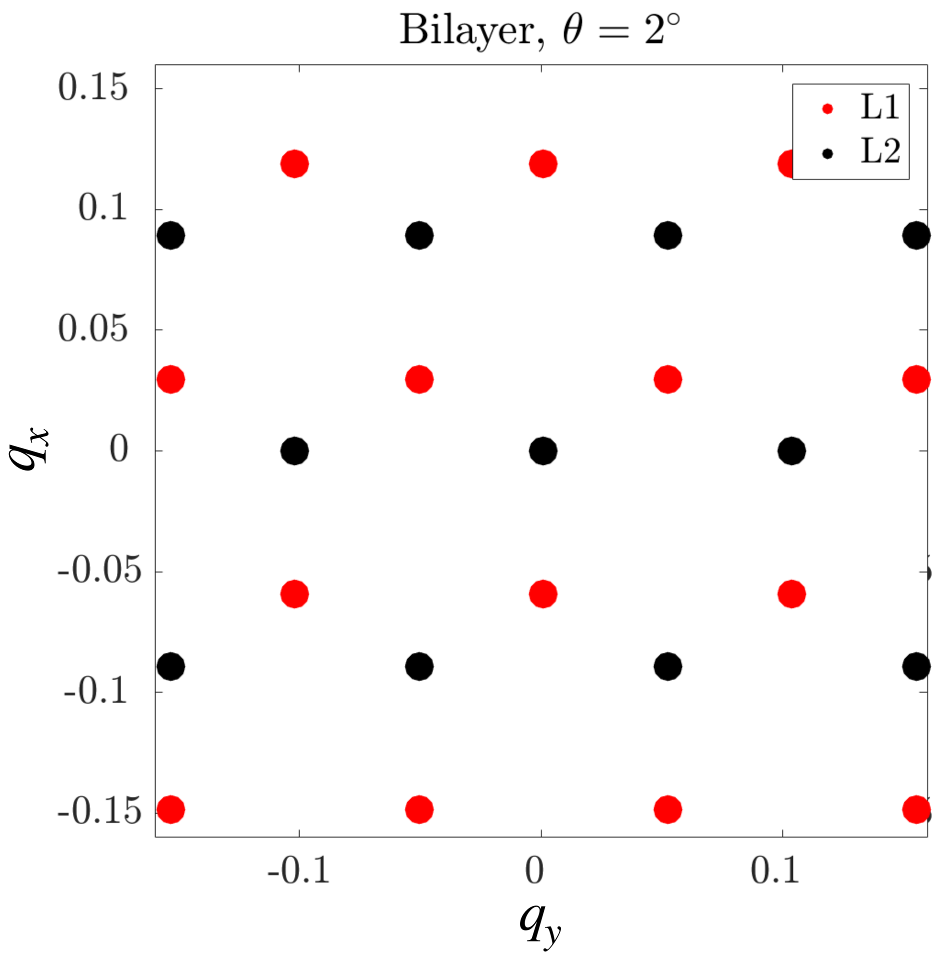
Trilayer scattering selection rules

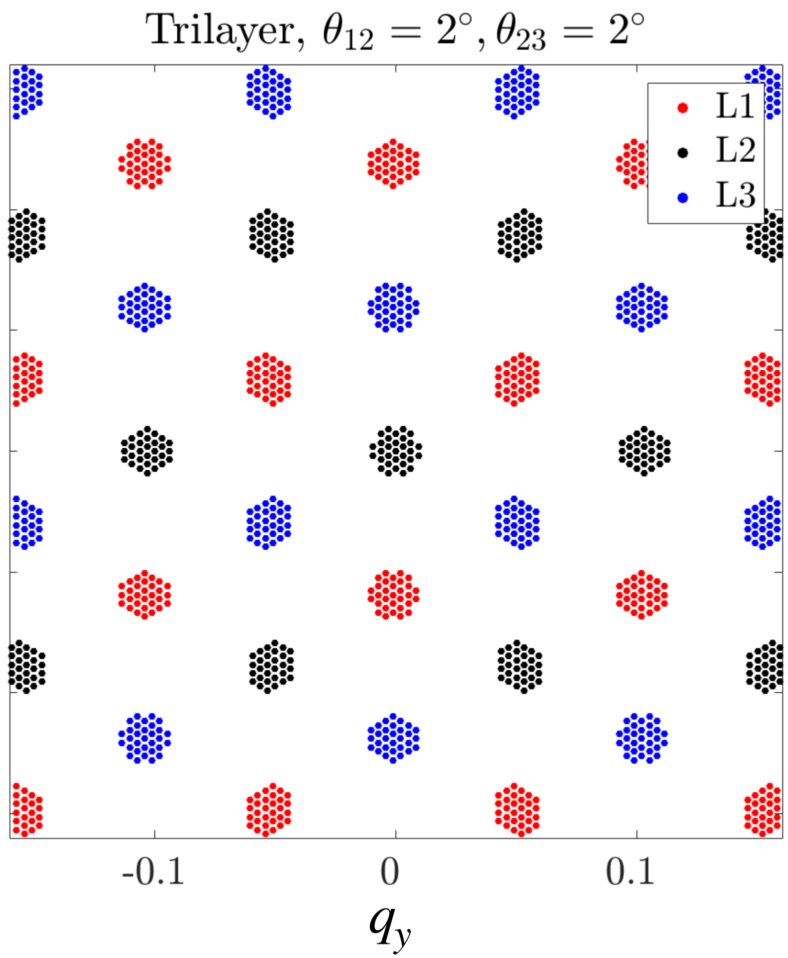
$$\boldsymbol{G}^{(1)} + \boldsymbol{q}^{(1)} + K_{L_1} = \boldsymbol{G}^{(2)} + \boldsymbol{q}^{(2)} + K_{L_2}$$

$$\boldsymbol{G}'^{(2)} + \boldsymbol{q}^{(2)} + K_{L_2} = \boldsymbol{G}^{(3)} + \boldsymbol{q}^{(3)} + K_{L_3}$$

k degree of freedom in bilayers vs. trilayers

Plotted quantities: scattering momenta in each layer
Origin: the Dirac point of Layer 2



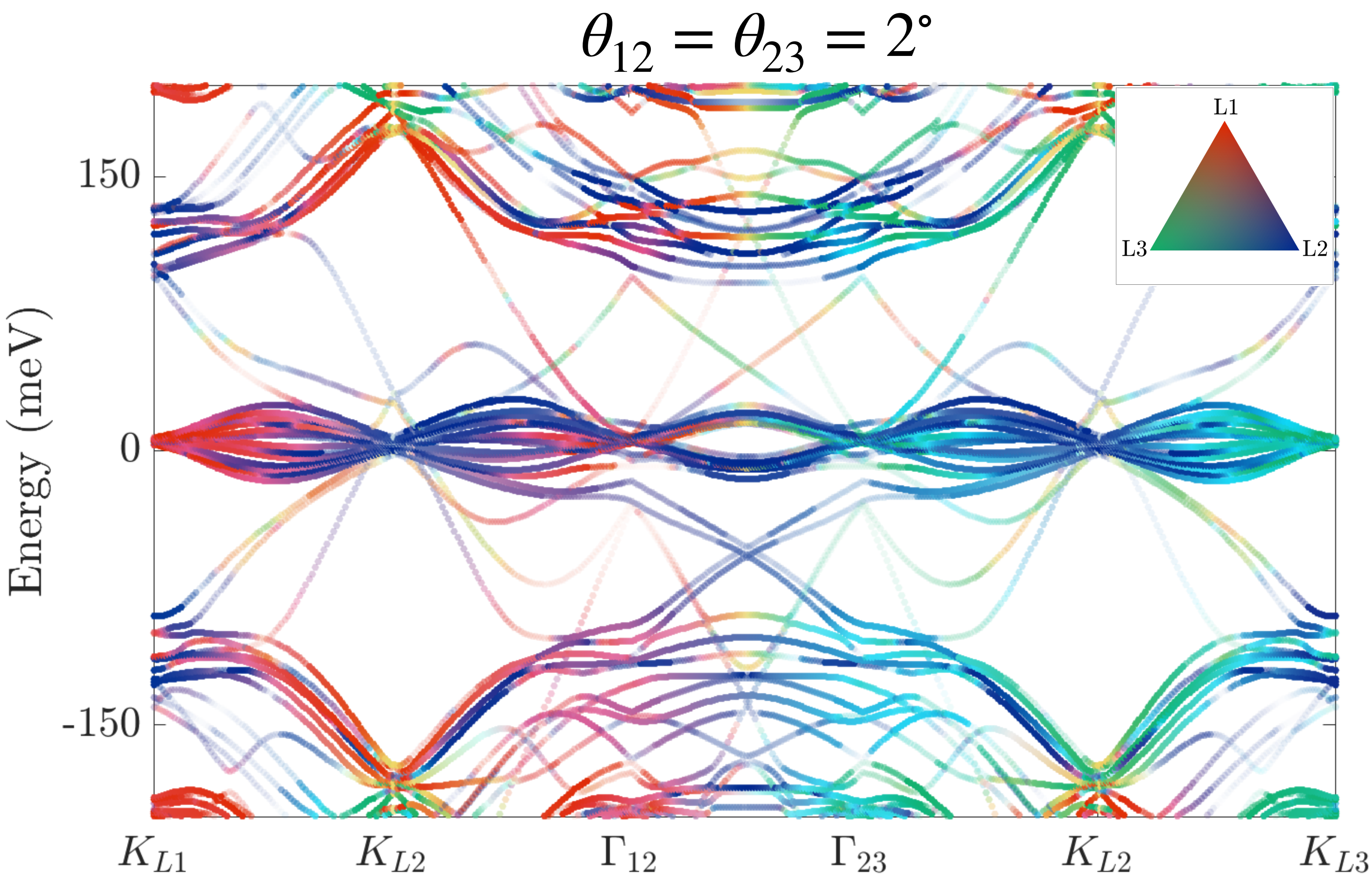


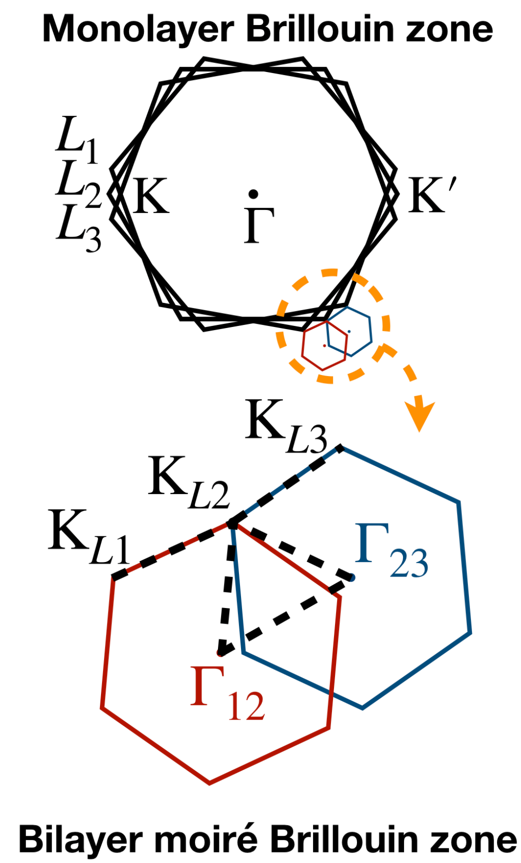
Scattering selection rules

$$\boldsymbol{G}^{(1)} + \boldsymbol{k}^{(1)} = \boldsymbol{G}^{(2)} + \boldsymbol{k}^{(2)}$$

$$\boldsymbol{k}^{(1)} + \boldsymbol{G}^{(1)} - \boldsymbol{G}^{(2)} = \boldsymbol{k}^{(3)} + \boldsymbol{G}^{(3)} - \boldsymbol{G}'^{(2)}$$

Electronic structure of tTLG

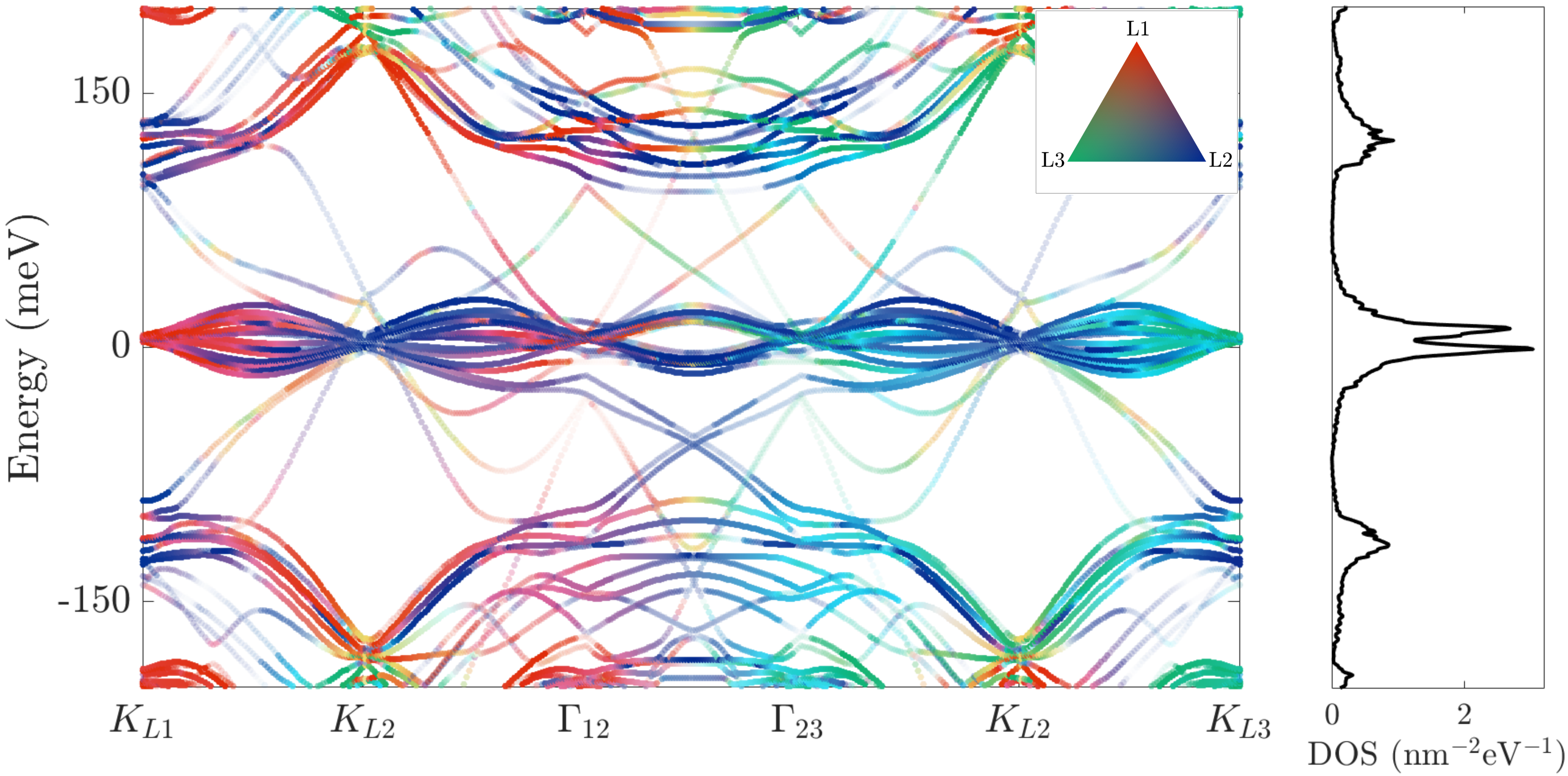




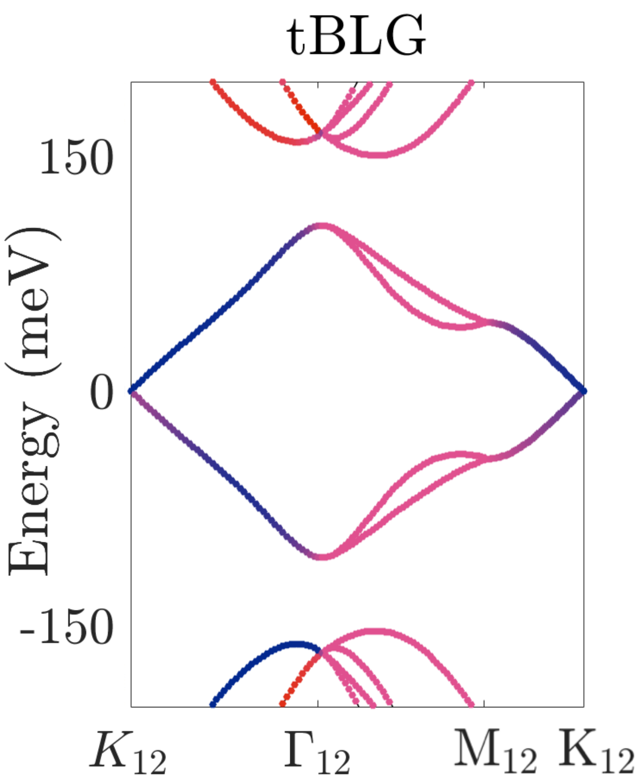
All connected bands
Hybridization between all three layers
Flat bands near the CNP

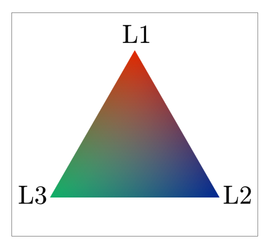
Electronic structure of tTLG

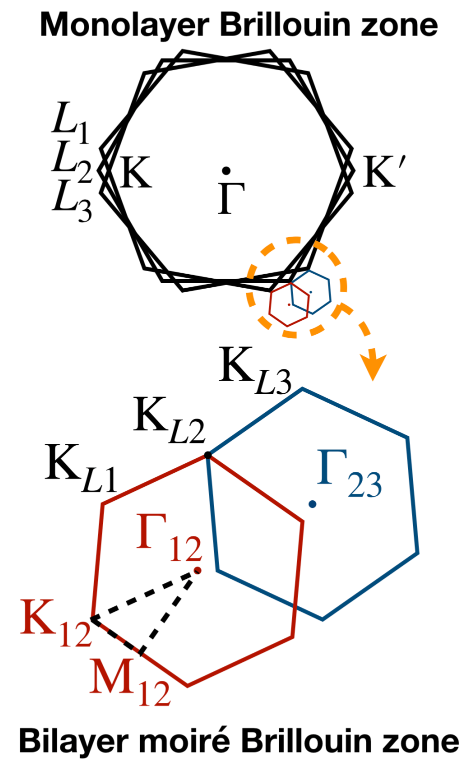
$\theta_{12} = \theta_{23} = 2^{\circ}$



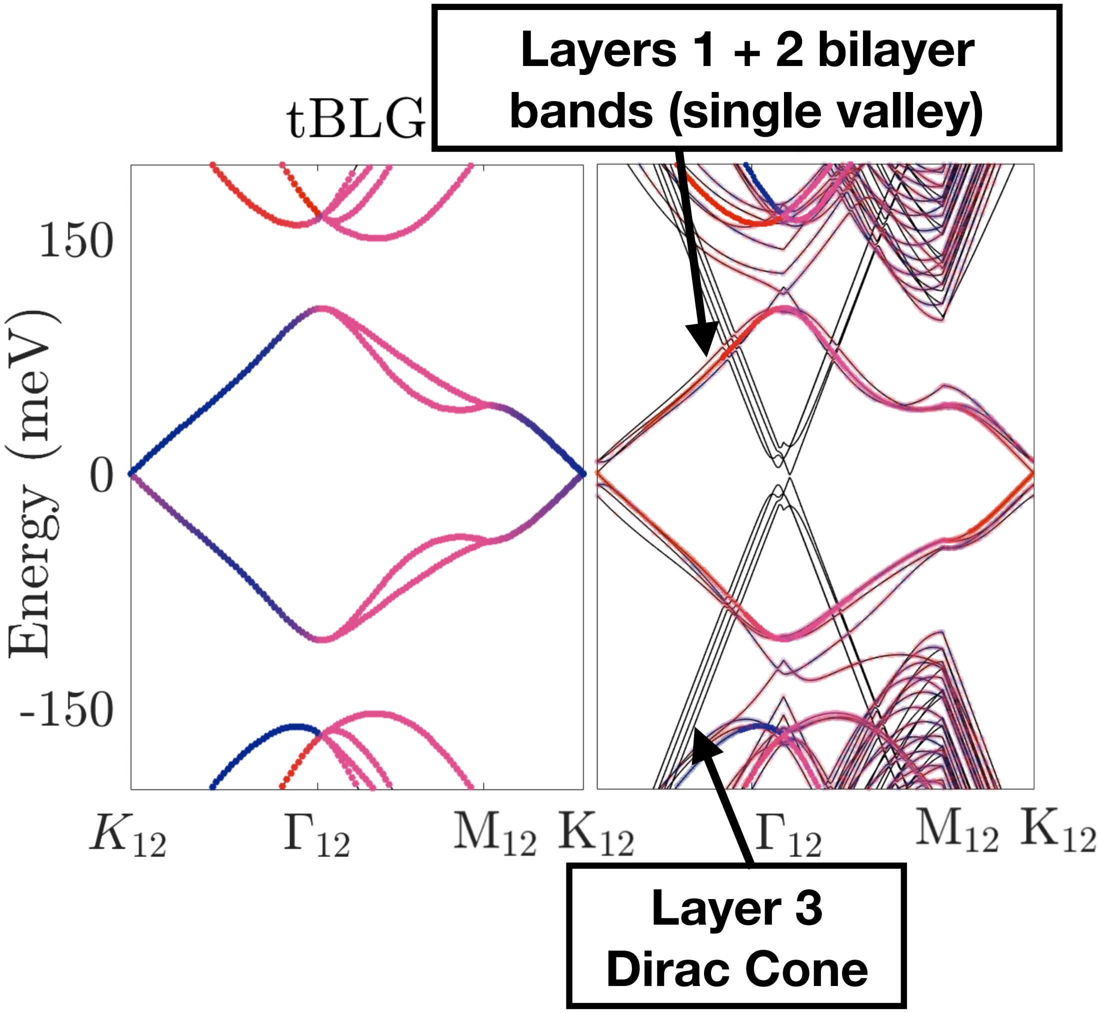
Comparison between tTLG and tBLG - band structure

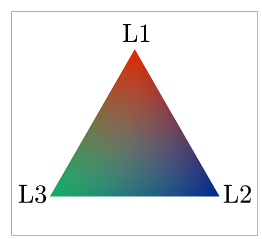


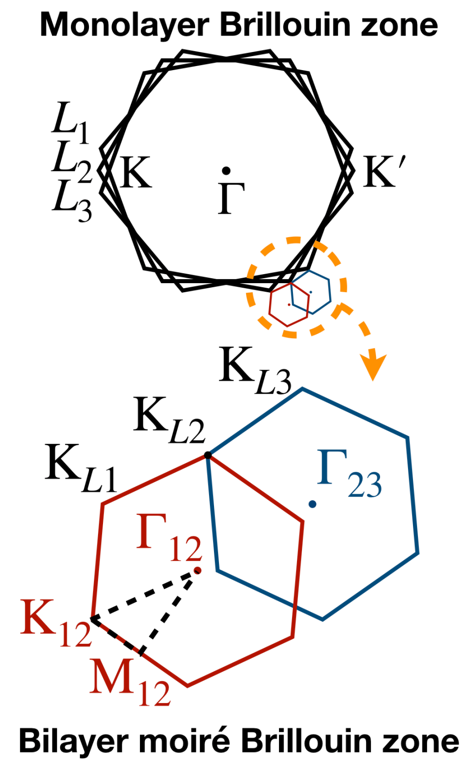




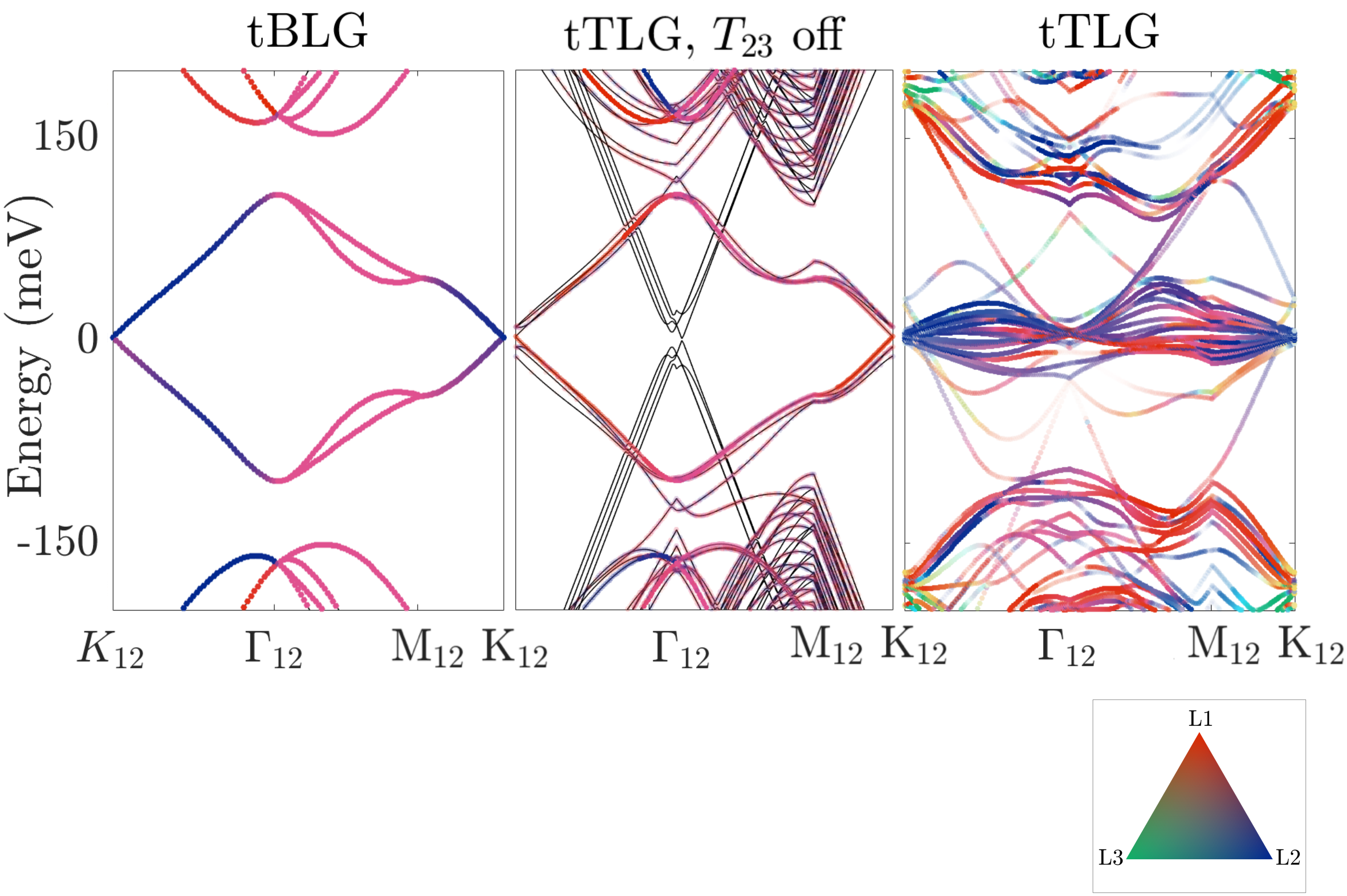
Comparison between tTLG and tBLG
- band structure

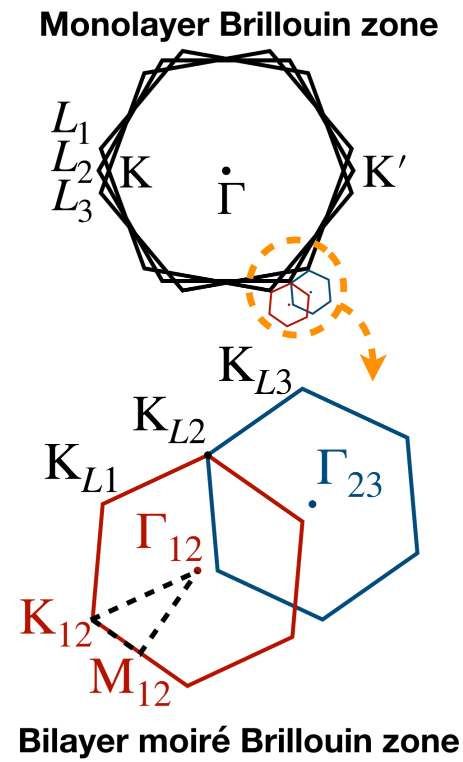




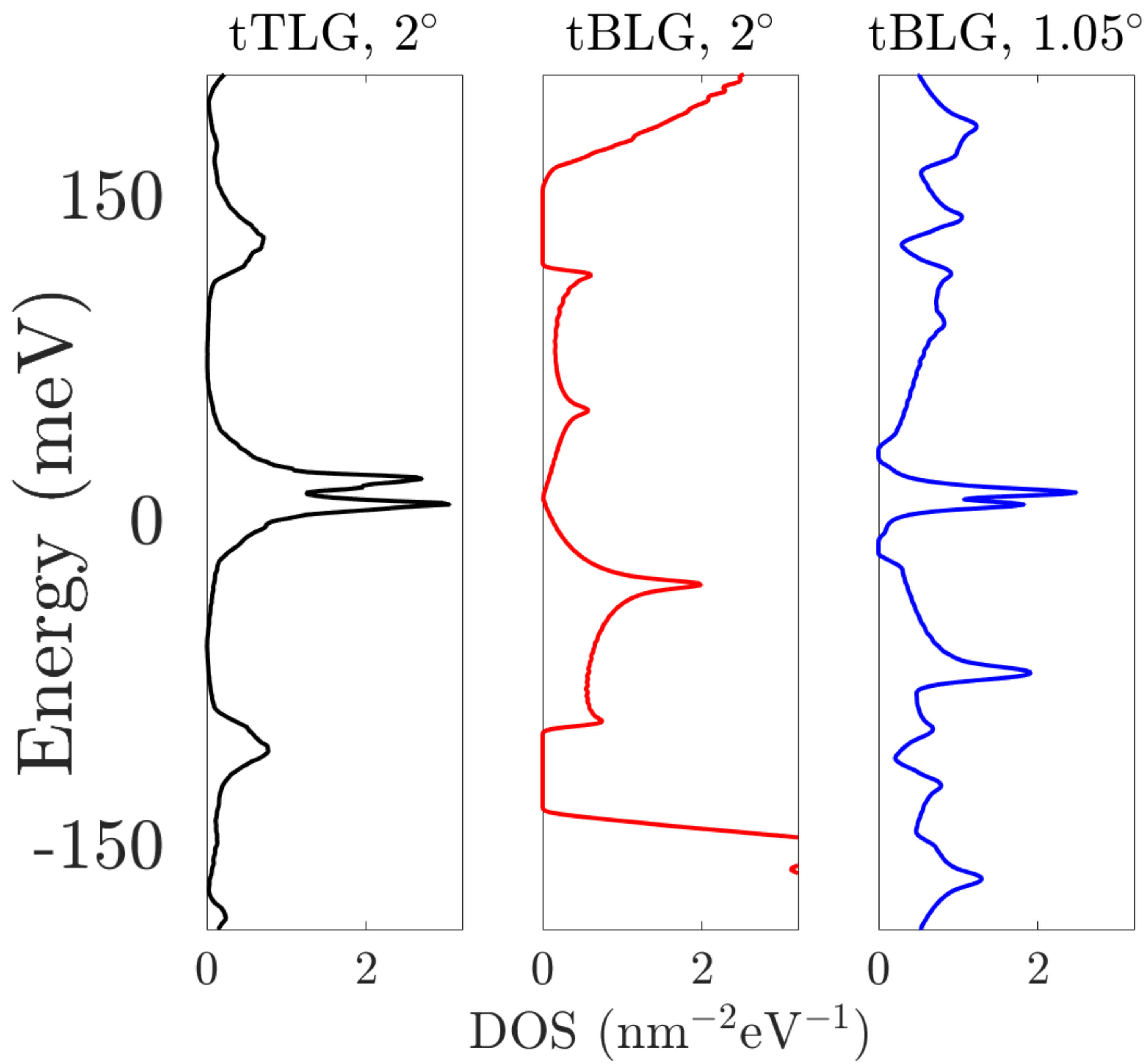


Comparison between tTLG and tBLG - band structure

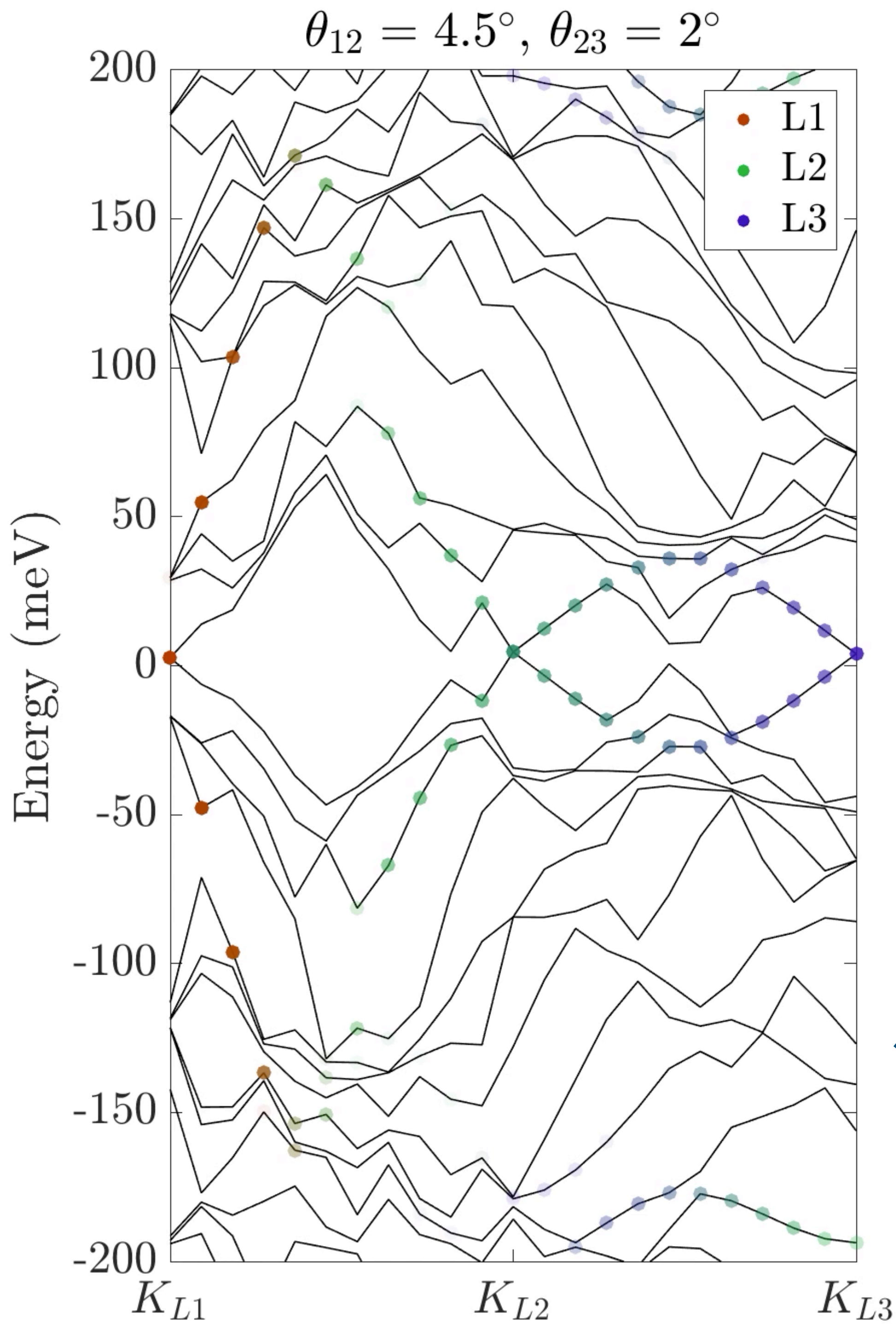


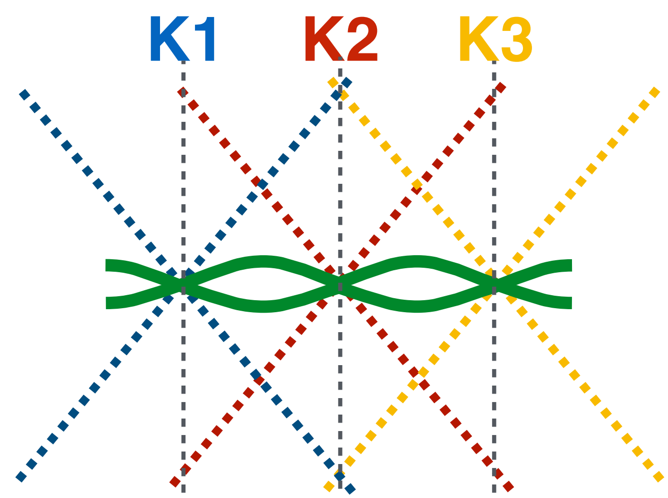


Comparison between tTLG and tBLG - DOS

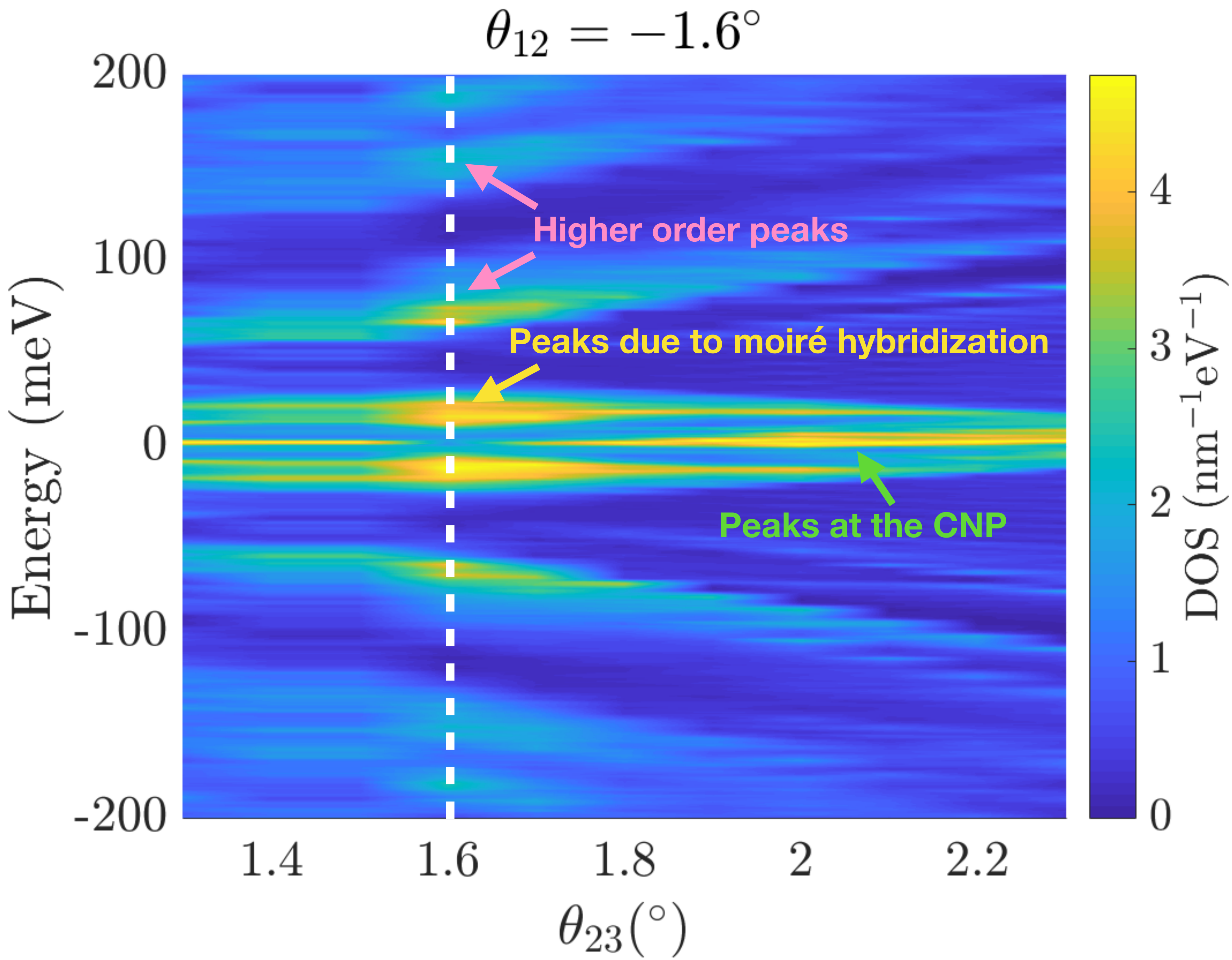


Band structure evolution at $\theta_{23} = 2°$



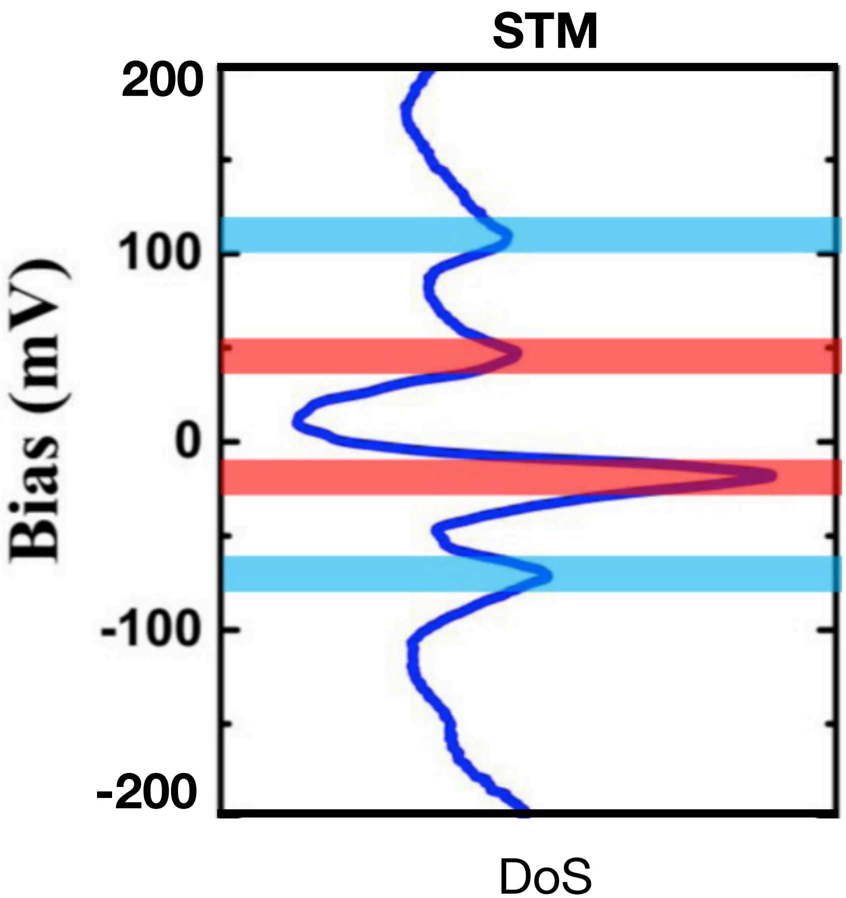


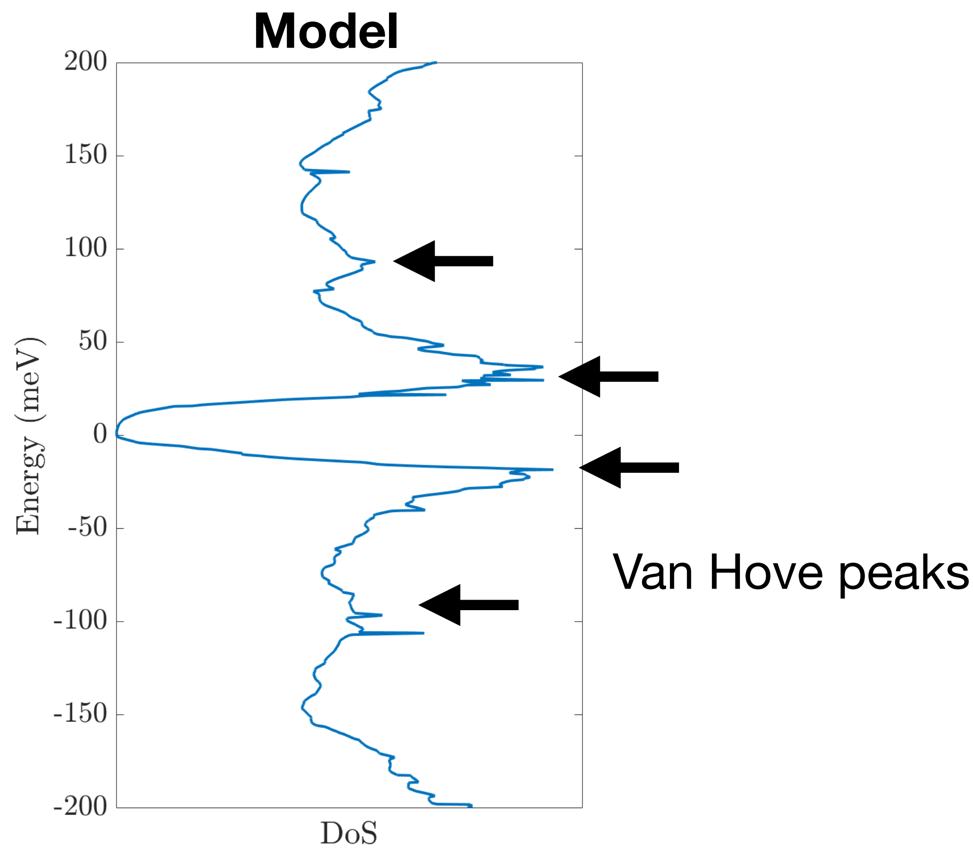
Density of states evolution



Agreement with the experiment

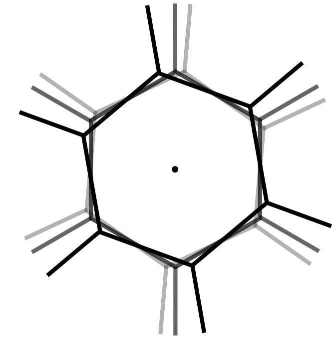
$\theta_{12} = 2.10^{\circ}, \theta_{23} = -2.81^{\circ}$





Ref: Zuo *et al*, Phys. Rev. B **97**, 035440 (2018)

Conclusion



- Relaxation in twisted trilayer vdW heterostructures
  - Moiré of moiré; capability of modeling aperiodic systems through configuration space
  - Spontaneous symmetry breaking at $\theta_{12} = \theta_{23}$
- Electronic structures of tTLG:
  - Flat bands and high DoS at equal twist angles
  - No singular DOS in tTLG, contrasting the tBLG results

Future directions

- Electronic states + relaxation
- Electron-phonon interaction