Beyond the Unit Cell: Computing Electronic Properties in Twisted and Disordered Nanomaterials

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In Collaboration with:
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(Harvard) S. Fang, S. Shirodkar, E. Kaxiras.

BIRS 2016: Coupled Mathematical Models for Physical and Biological Nanoscale Systems and Their Applications(16w5069)

Sponsored by: ARO MURI Award No. W911NF-14-0247

Multi-scale approach

Increasing size

Scale:

Method:

In:

Out:

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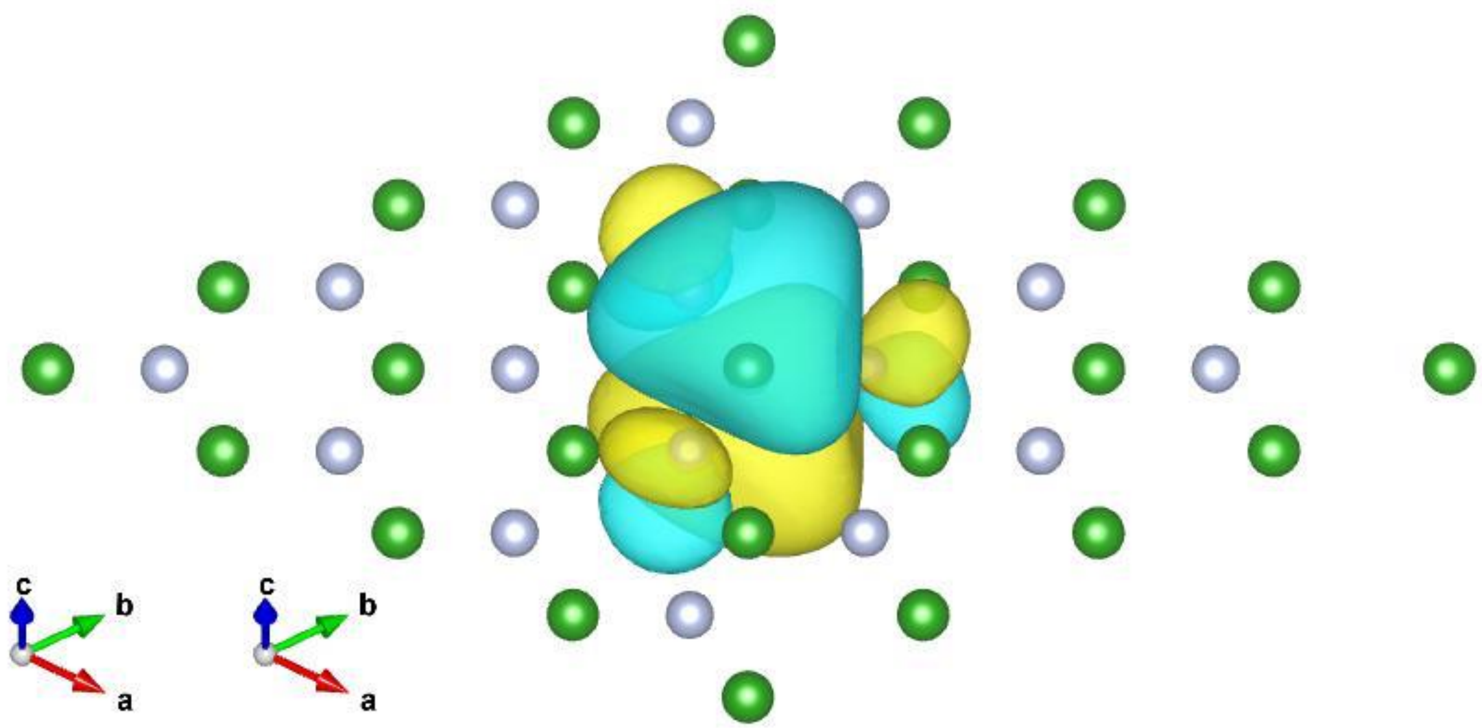
Multi-scale approach

Increasing size →

Scale:	<u>Quantum:</u>
Method:	DFT, Wannier
In:	*Ab-initio*
Out:	Tight-binding parameters



S. Fang, BIRS 2016

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Multi-scale approach

Increasing size →

Scale:	<u>Quantum:</u>	<u>Nano-scale:</u>
Method:	DFT, Wannier	TBM, Locality
In:	*Ab-initio*	Tight-binding
Out:	Tight-binding parameters	Physical Properties (DoS, σ, ε)

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Multi-scale approach

Increasing size →

Scale:	Quantum:	Nano-scale:	Macroscopic:
Method:	DFT, Wannier	TBM, Locality	PDEs, Continuum
In:	*Ab-initio*	Tight-binding	σ, ε
Out:	Tight-binding parameters	Physical Properties (DoS, σ, ε)	Engineered Devices

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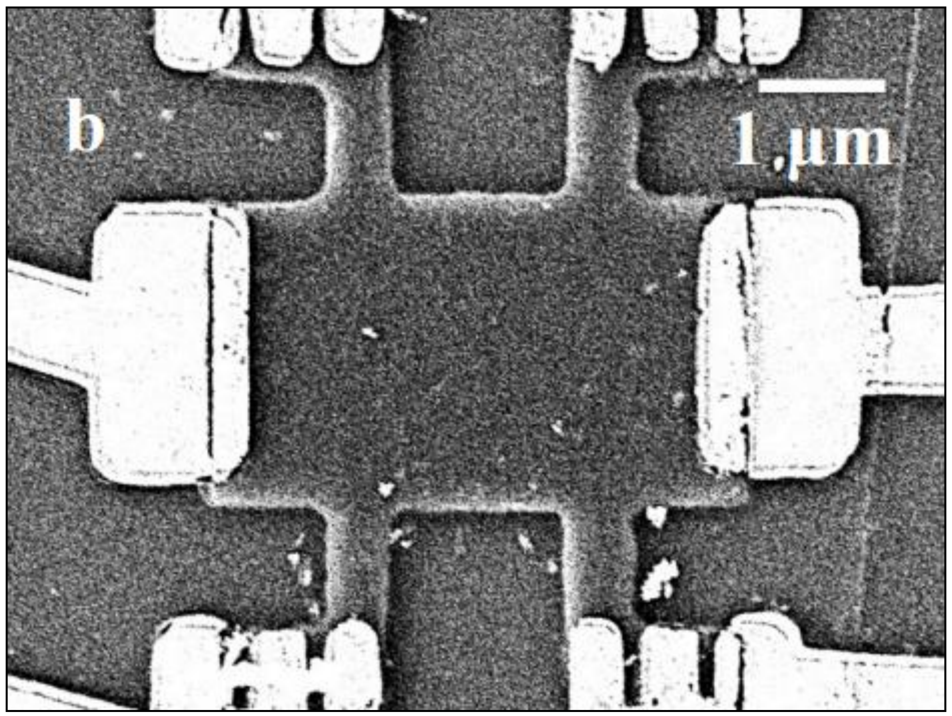
Simulating nanomaterial devices…?



S. Bhandari, G. H. Lee, A. Klales, K. Watanabe, T. Taniguchi, E. Heller, P. Kim, R. M. Westervelt.

Imaging Cyclotron Orbits of Electrons in Graphene.

Nano Lett., **2016**, *16* (3), pp 1690–1694

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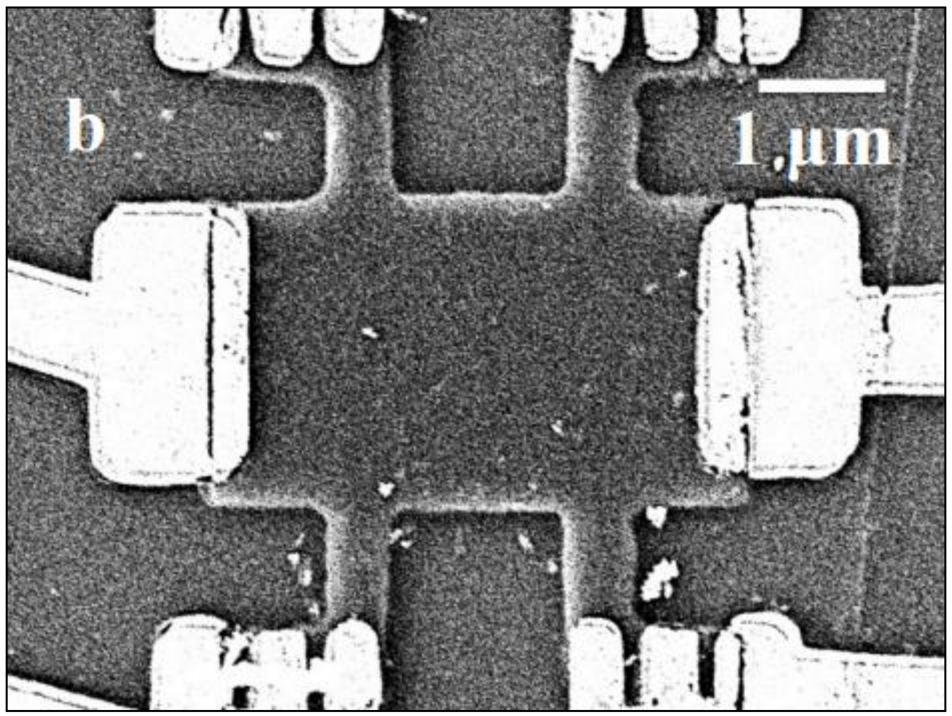

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<u>1 μm radius disk of graphene</u>

10.24×10^{6} atoms

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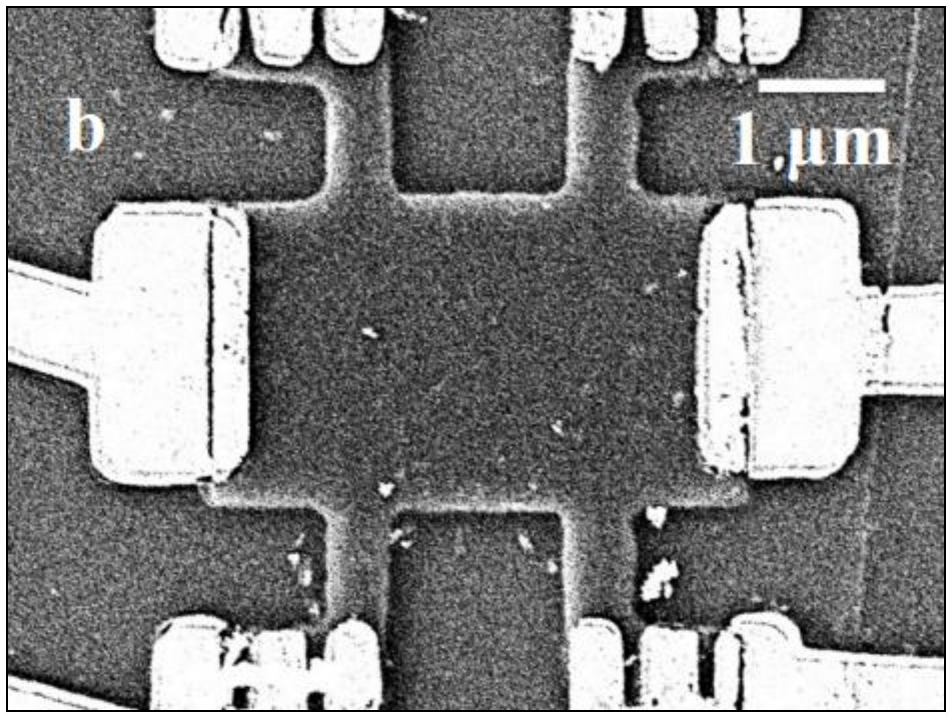

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<u>1 μm radius disk of graphene</u>

10.24×10^6 atoms

<u>Sparse TB-Hamiltonian:</u>

Max # of entries: 10^{14}

Expected # of entries: 1.64×10^9

Diagonalization not feasible…

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Kernel Polynomial Method

$$g_x(\epsilon) = \frac{1}{N} \sum_{s=1}^{N} \delta(\epsilon - \epsilon_s) |\psi_s(x)|^2$$

$$\sum_x g_x(\epsilon) = Tr[\delta(\epsilon - H)]$$

x = target orbital
v_x = projector onto x
a_i = Chebyshev coeff.
T_i = Chebyshev polynomial
T_i^{xx} = Cheby. proj. onto x

A. Weisse, G. Wellein, A. Alvermann, H. Fehske

The Kernel Polynomial Method

Rev. Mod. Phys. **78**, 275 (2006)

$$g_x(\epsilon) = v_x^\dagger \delta(\epsilon - H) v_x$$

$$\delta(\epsilon - H) \approx \sum_{i=0}^{p} a_i(\epsilon) T_i(H)$$

$$g_x(\epsilon) \approx \sum_{i=0}^{p} a_i(\epsilon) \left(v_x^\dagger T_i(H) v_x \right) = \sum_{i=0}^{p} a_i(\epsilon) T_i^{xx}(H)$$

$$g_0(\epsilon) \approx \sum_{i=0}^{p} a_i(\epsilon) T_i^{00}(H)$$

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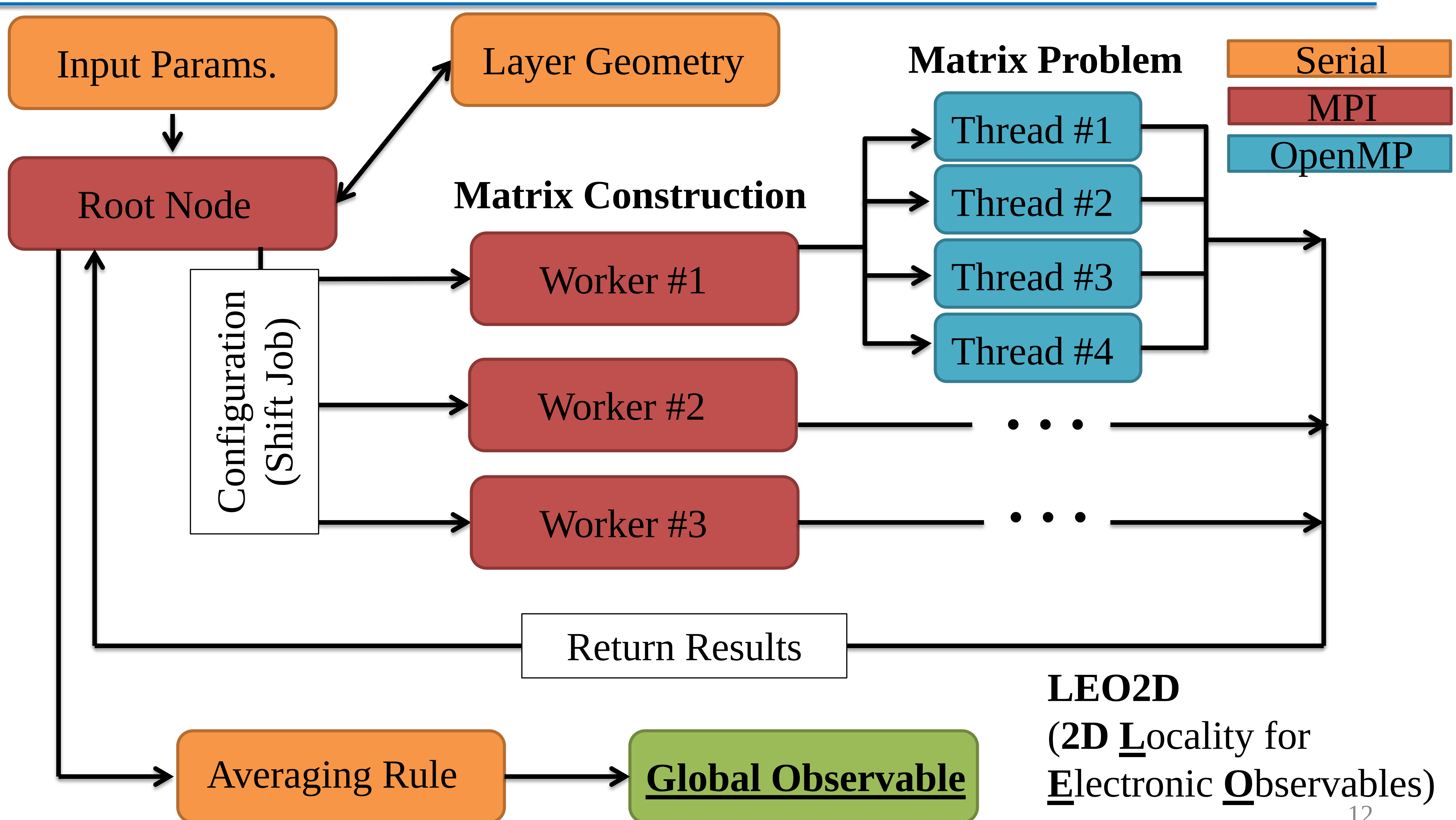
Parallelized Implementation



LEO2D
(**2D** **L**ocality for **E**lectronic **O**bservables)

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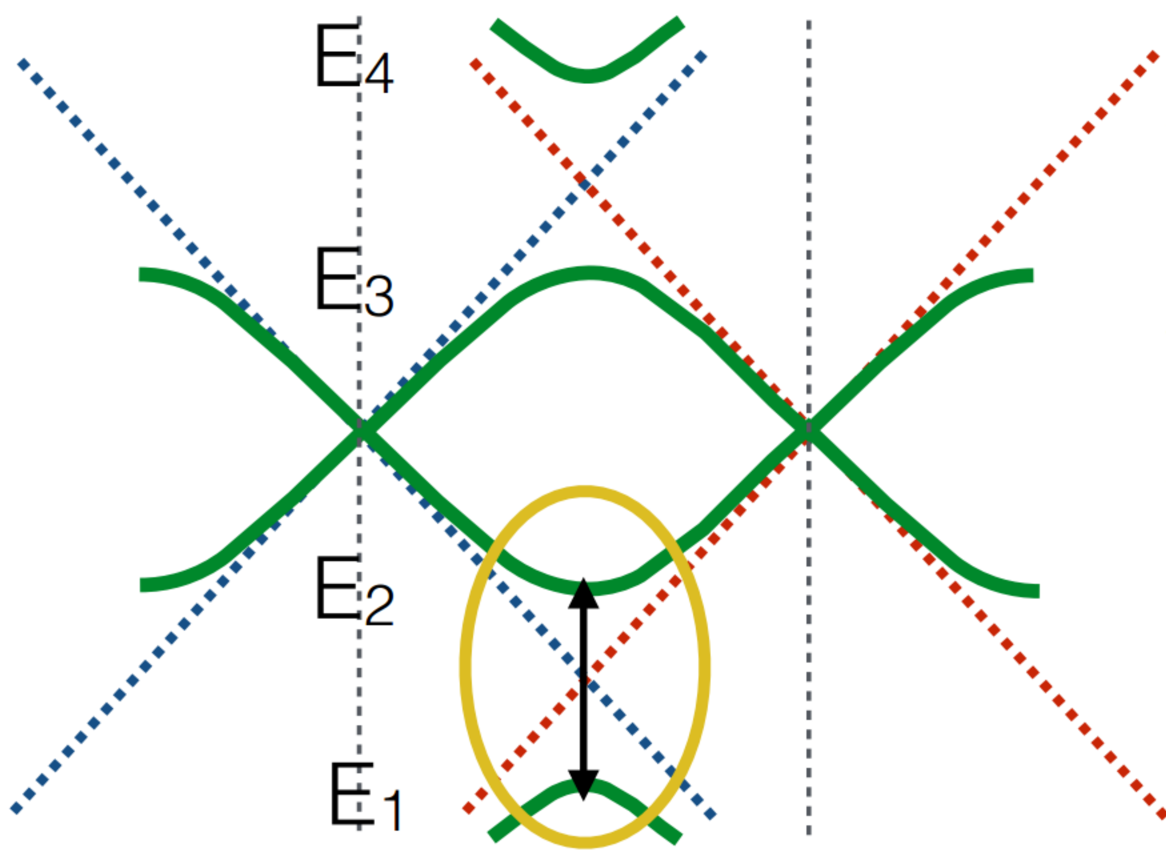
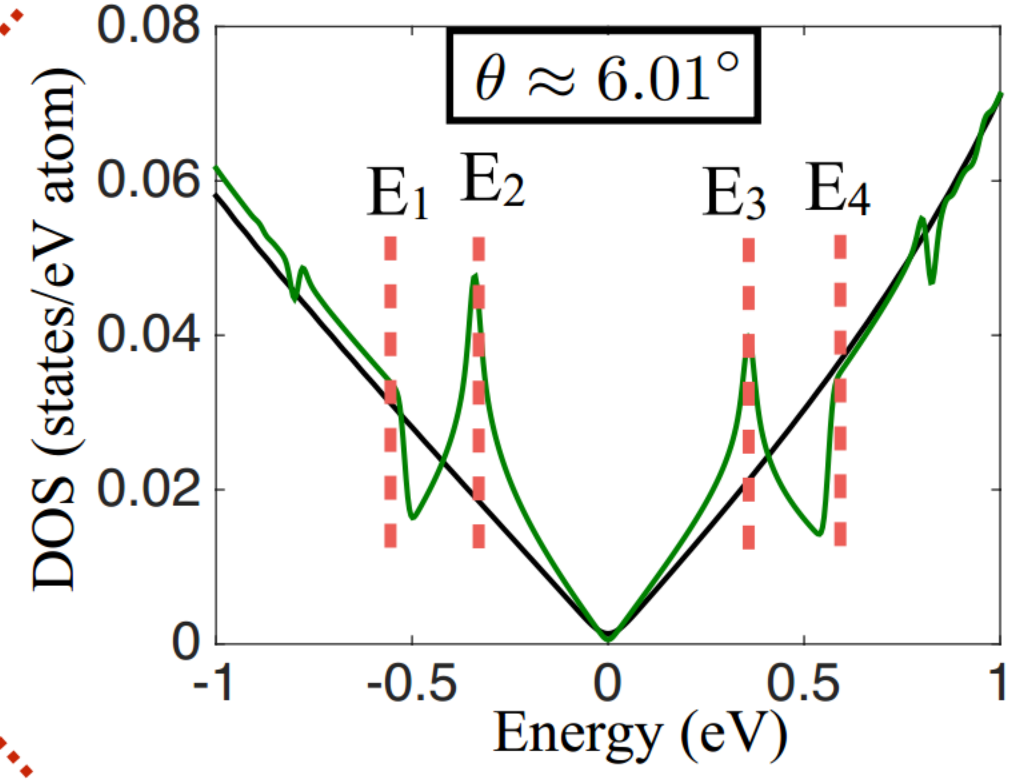
Twisted Bilayer Graphene (tBLG)

Two shifted Dirac cones



Van Hove singularity in density of states



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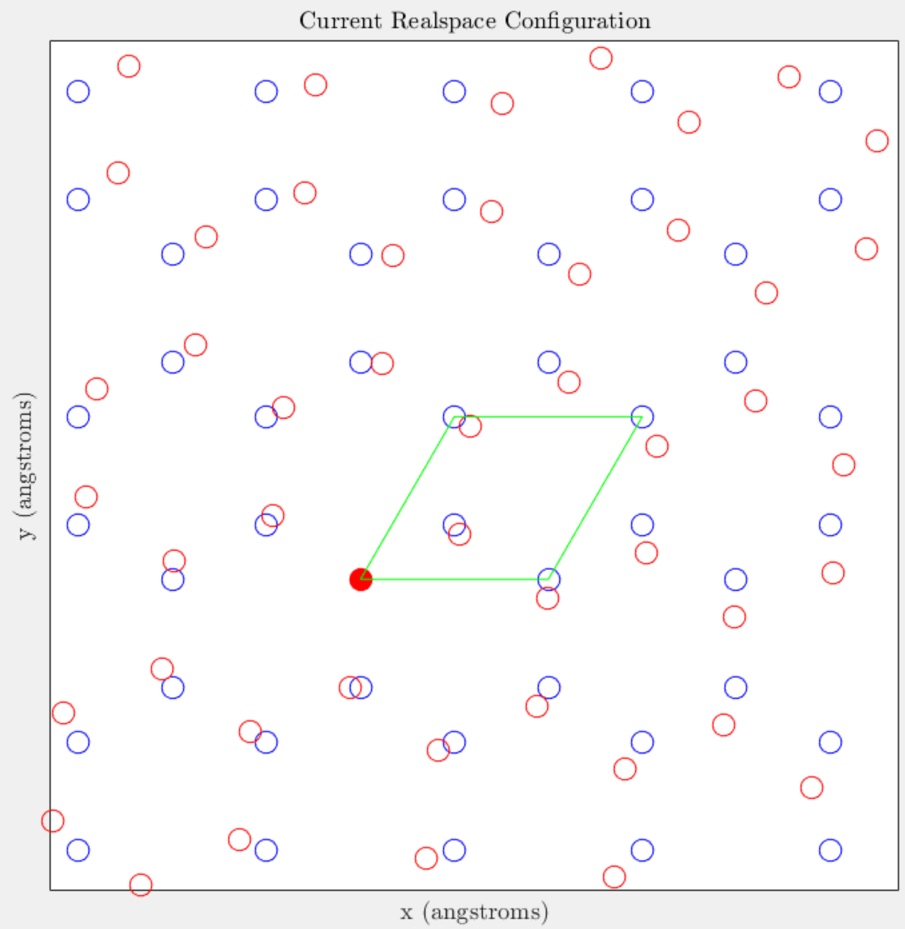

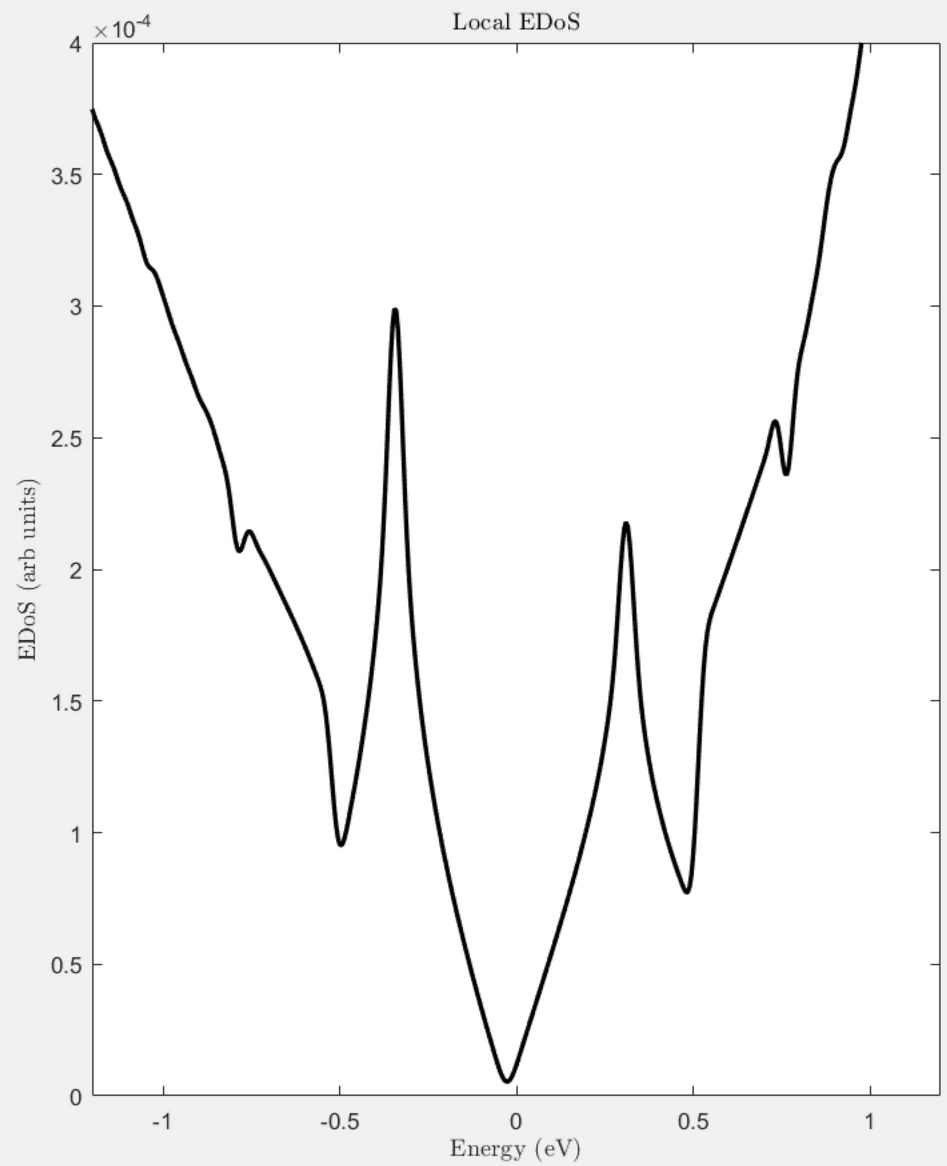

A new "Brillouin Zone" for disordered systems

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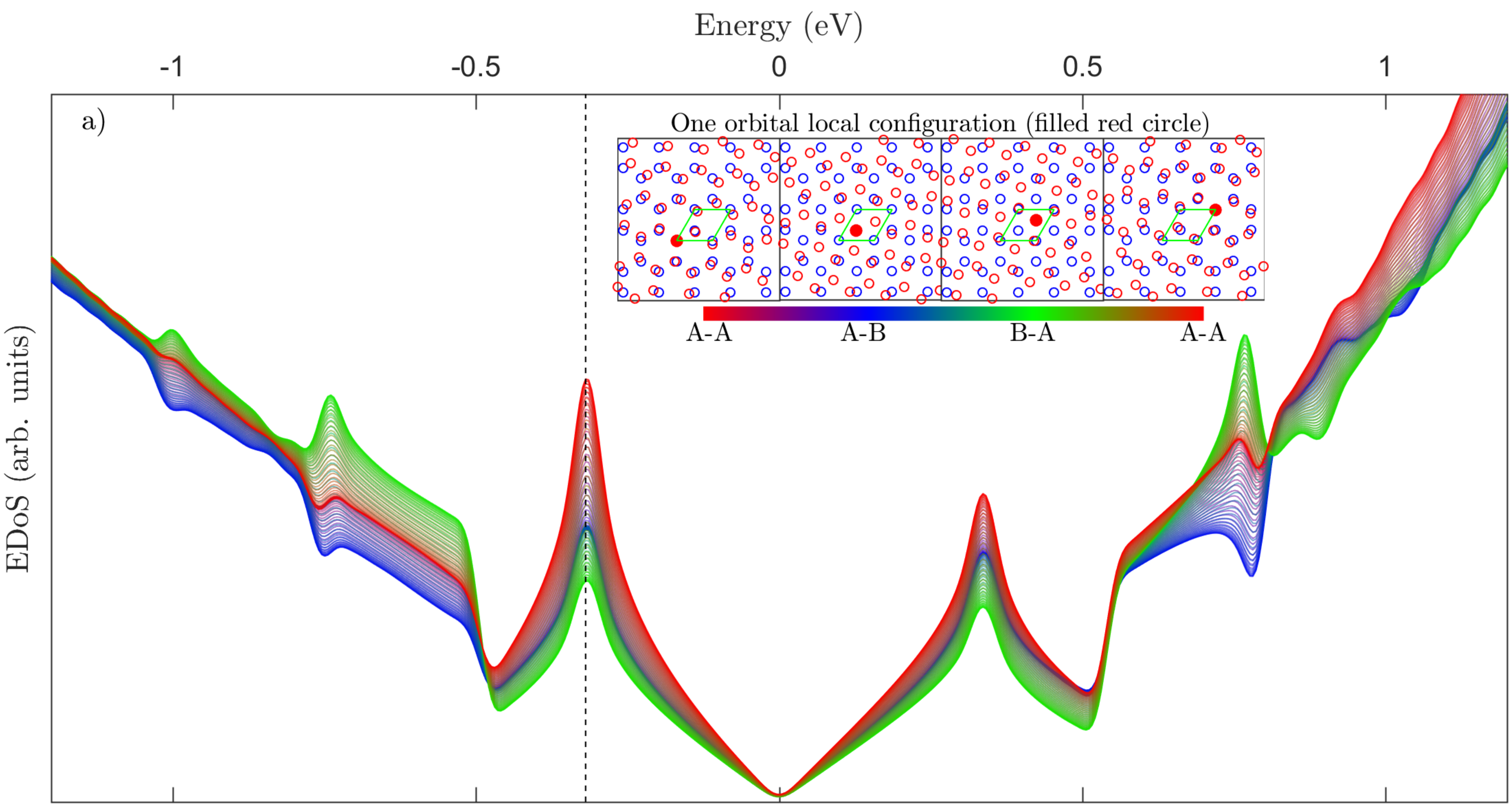

5.73° Twist angle
Simulation with 100 nm diameter (591,344 atoms)

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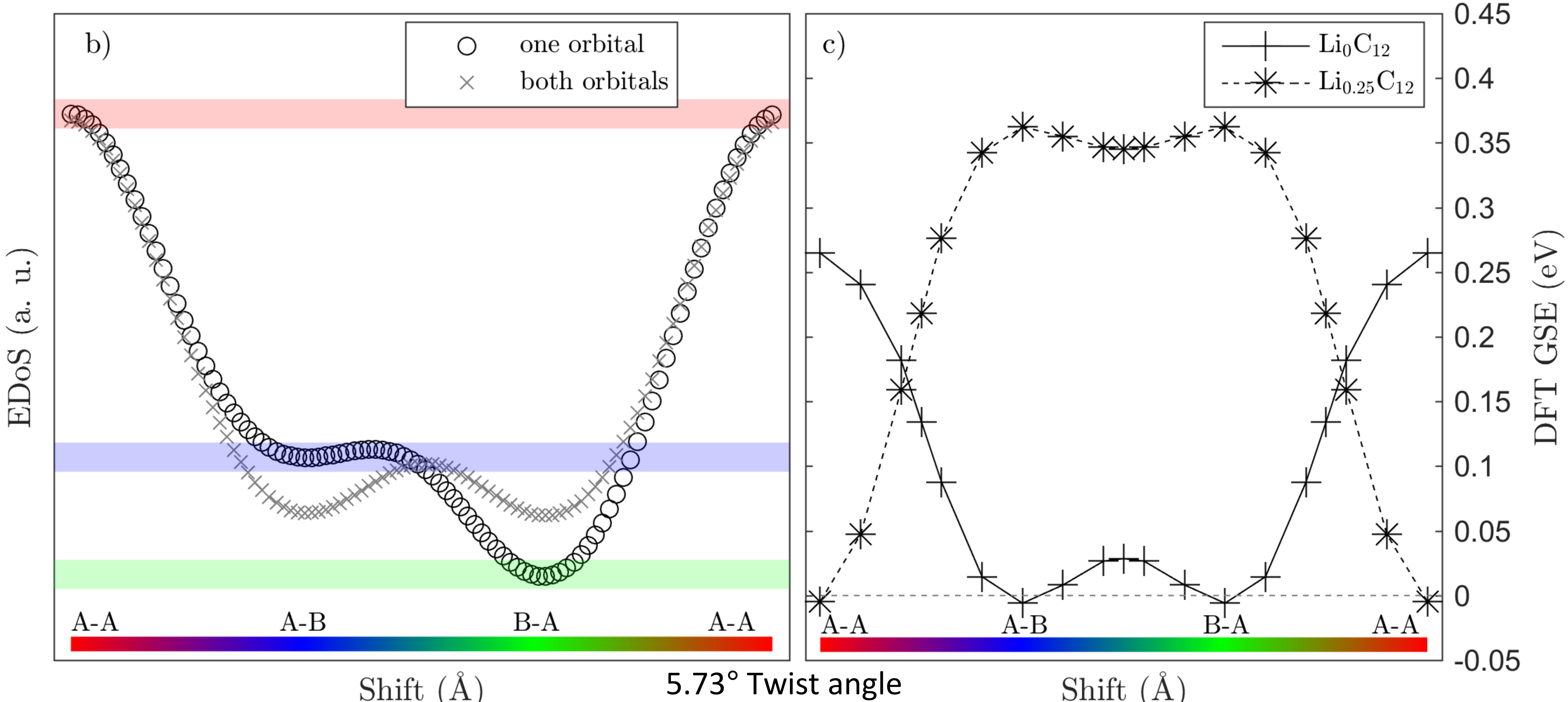

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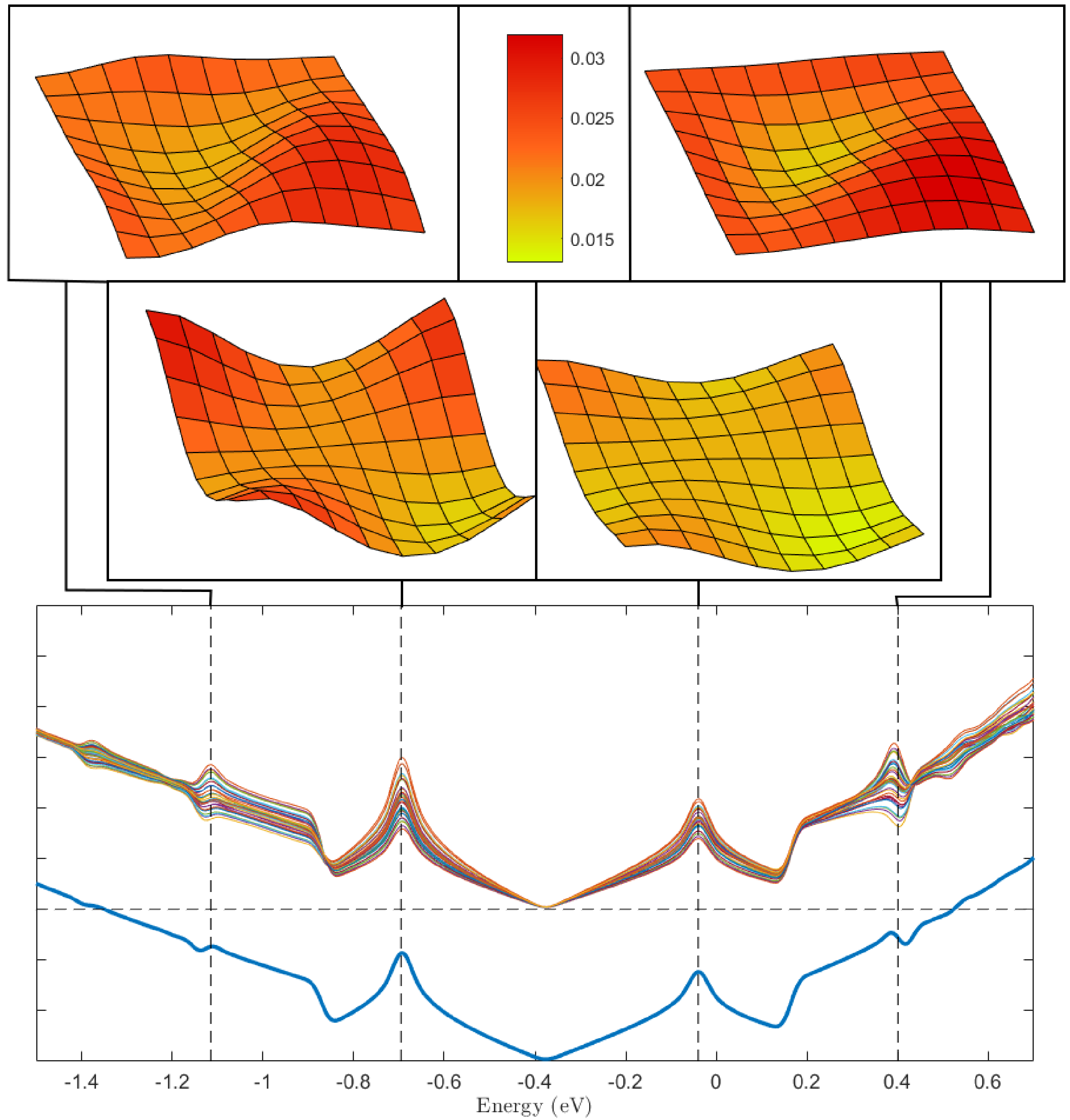

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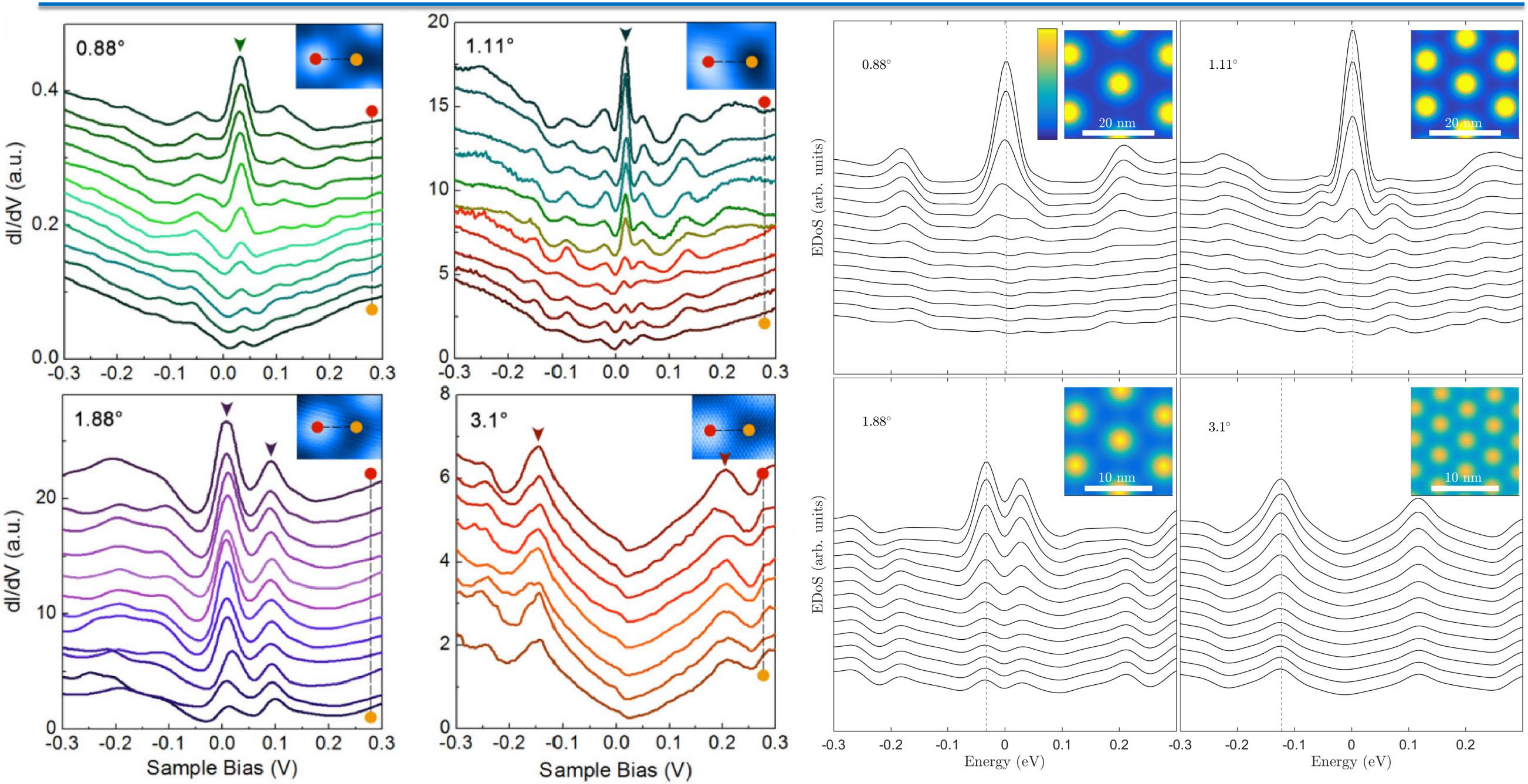
tBLG: Comparing to Exp.



Long-Jing Yin, Jia-Bin Qiao,Wei-Jie Zuo,Wen-Tian Li, and Lin He.
Experimental evidence for non-Abelian gauge potentials in twisted graphene bilayers.
Physical Review B. **2015**, 92, 081406

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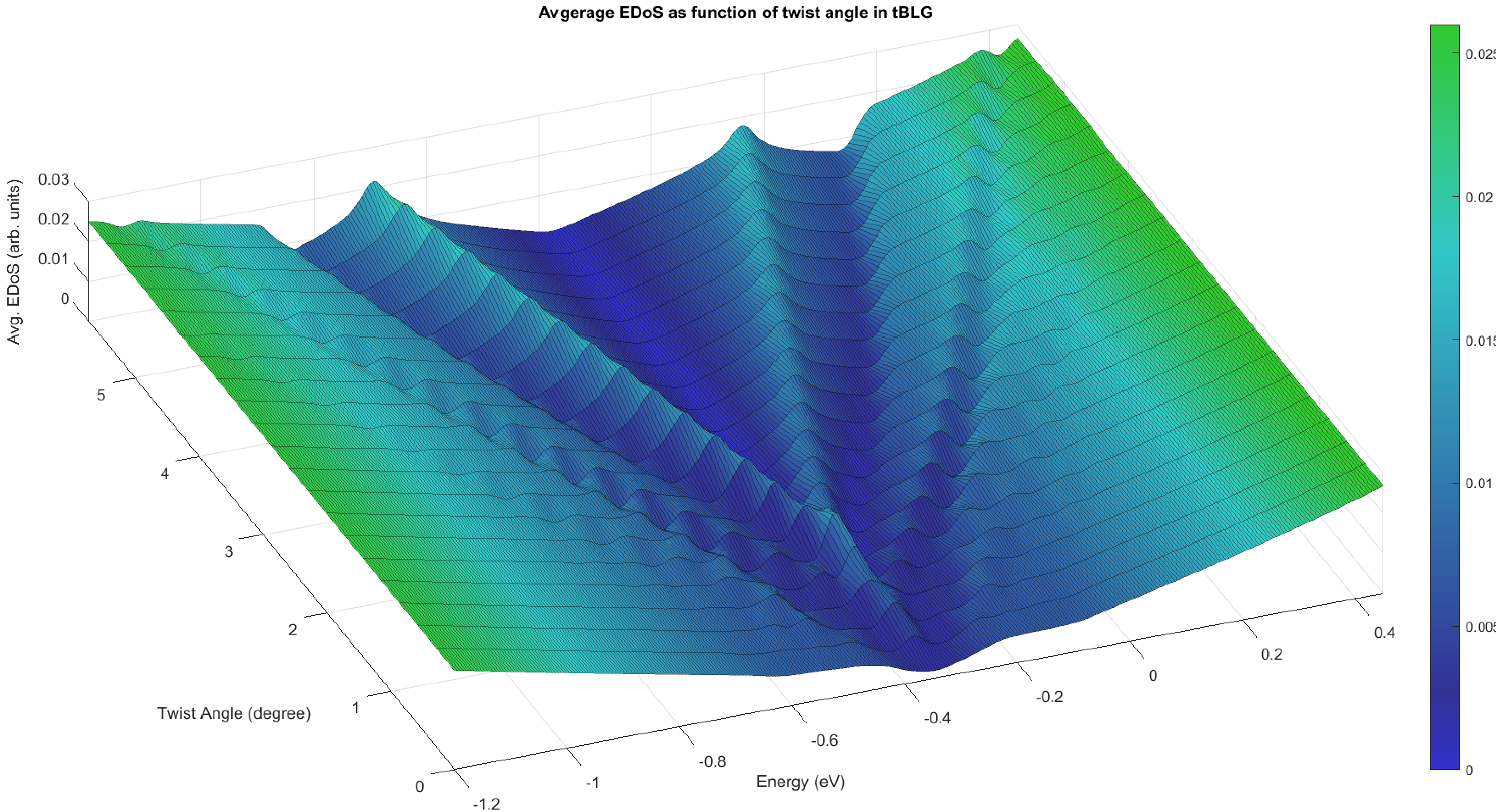
tBLG: Twist Degree of Freedom



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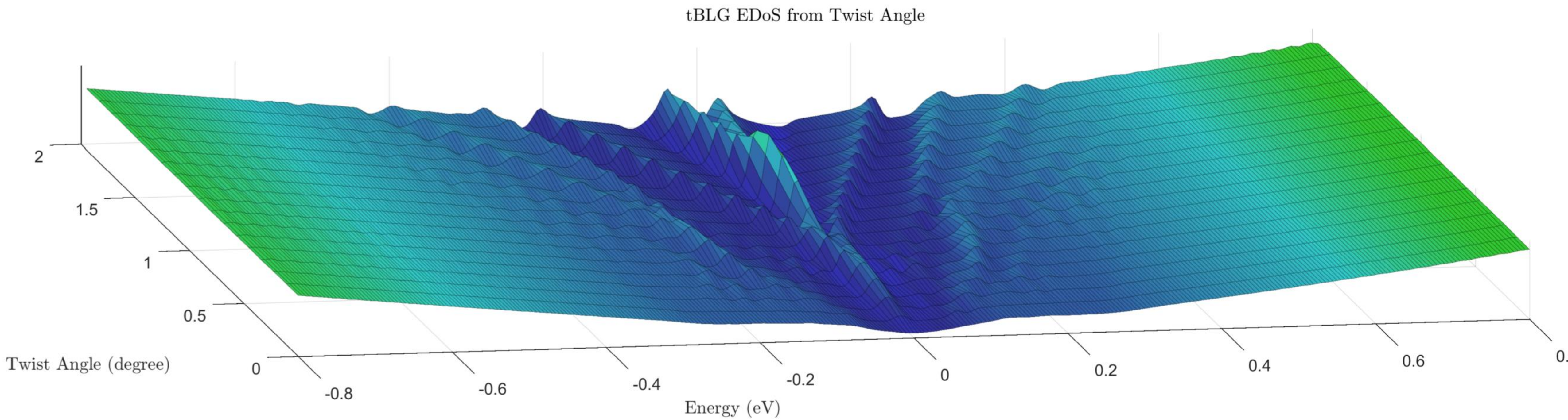
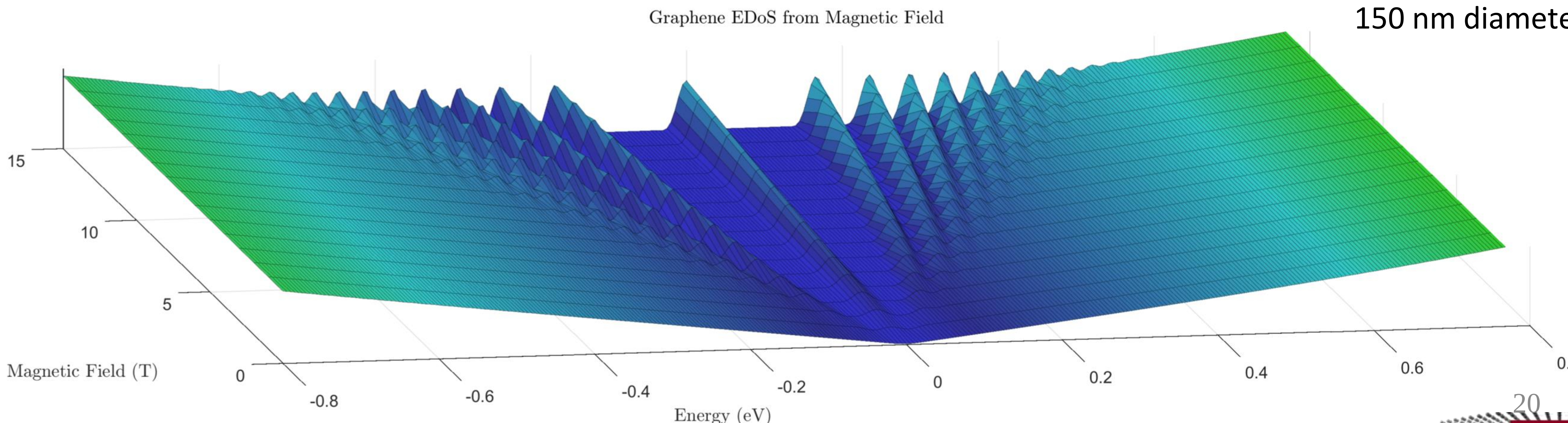


Simulations with
150 nm diameter



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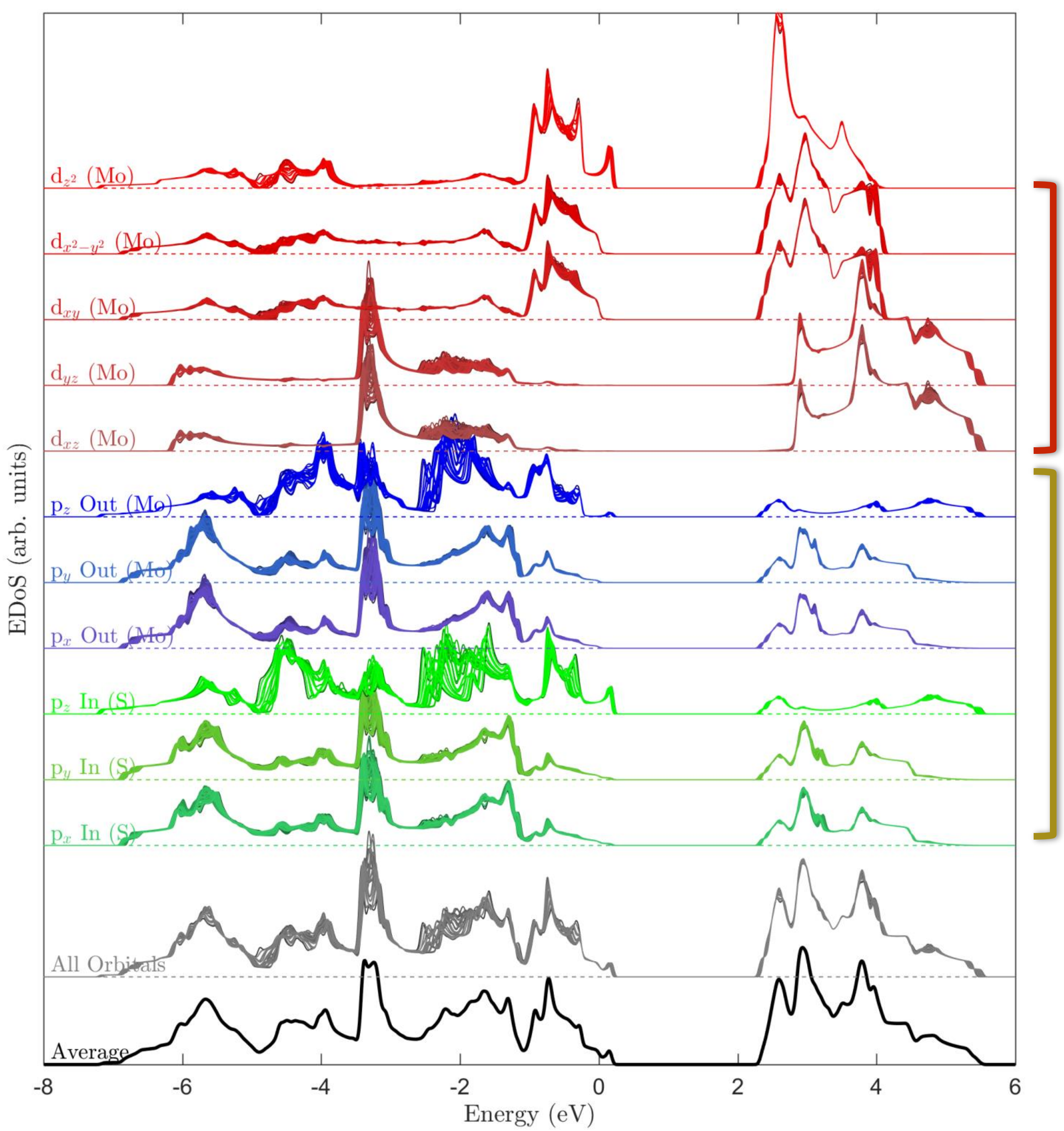
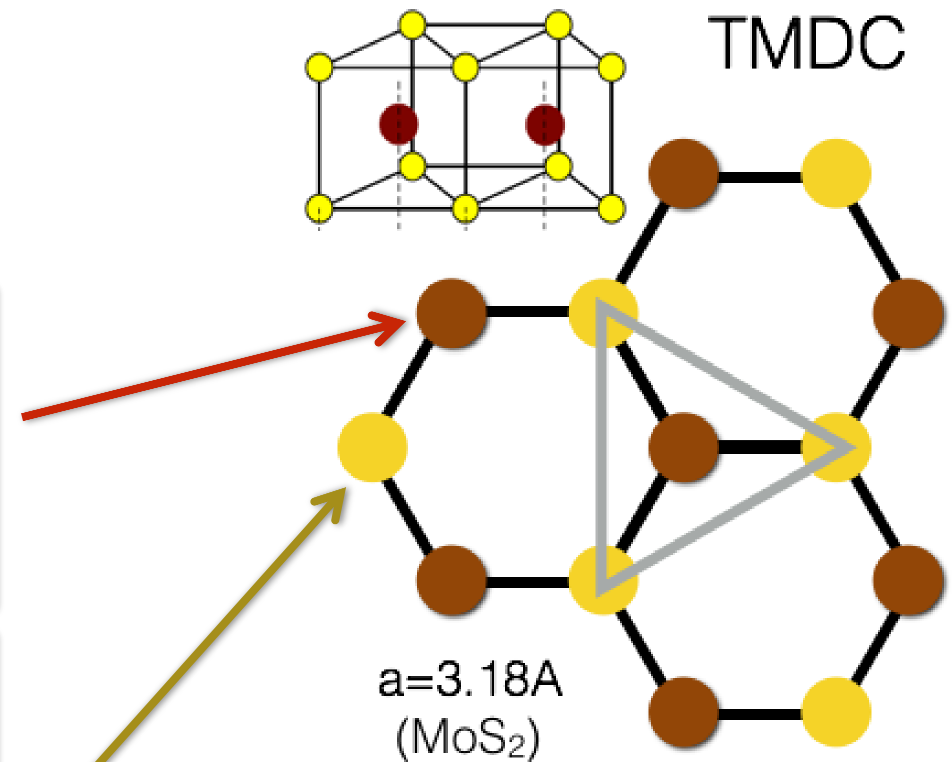
MoS_2 : Modeling TMDCs

Bilayer with 5.73° twist-angle





Semiconductor

- MX_2, M=Mo/W, X=S/Se
- Broken inversion symmetry
- Direct band gap 1-2 eV at K valleys
- Spin-orbit coupling

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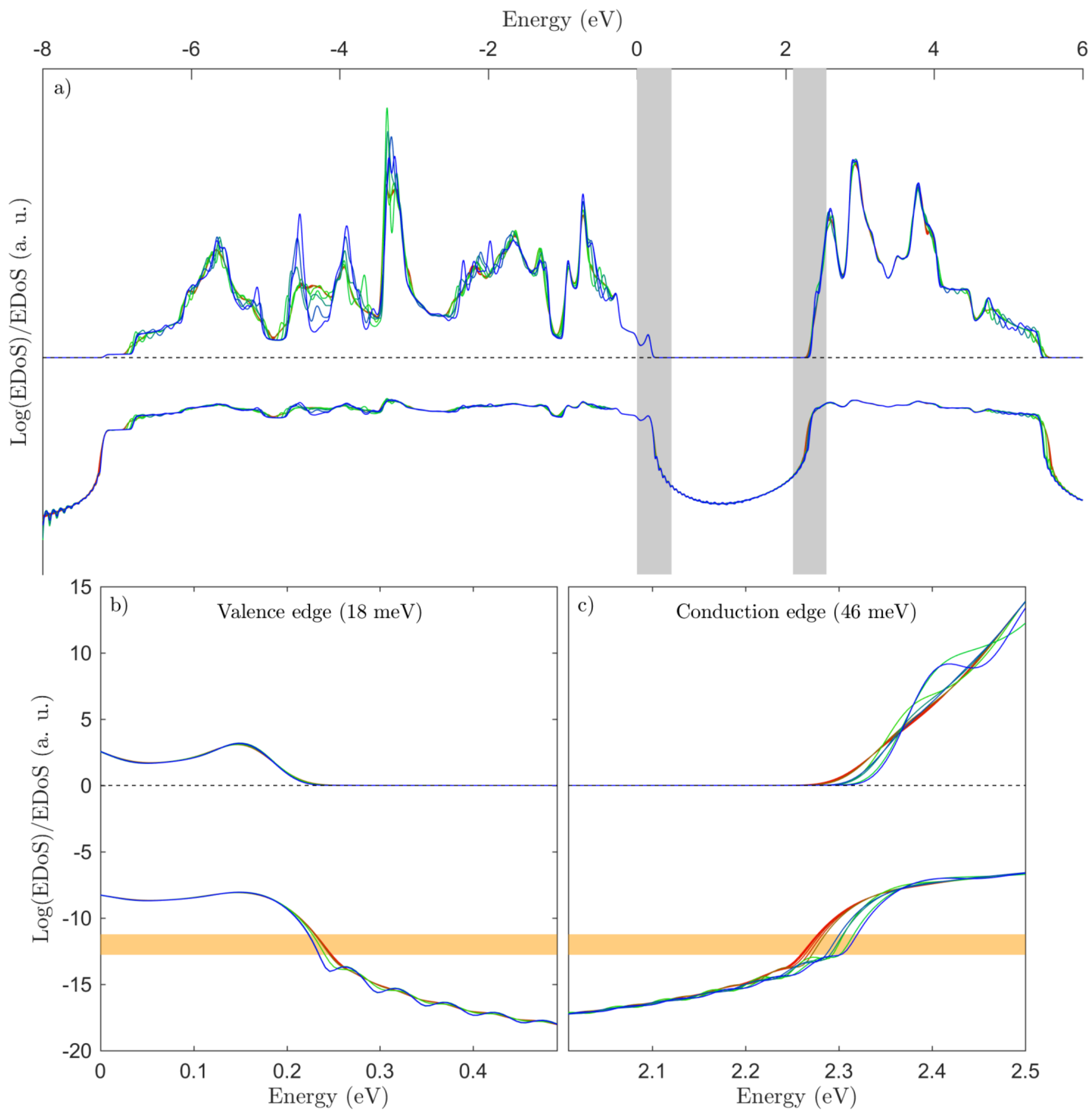
MoS_2 : Modeling TMDCs



Twist-angle dependent Electronic Density of States (EDoS)

[Logarithm of EDoS for band-gap study]

Band-edge EDoS

Band gap: 2.07 eV

Widens by: 64 meV

(3% increase)

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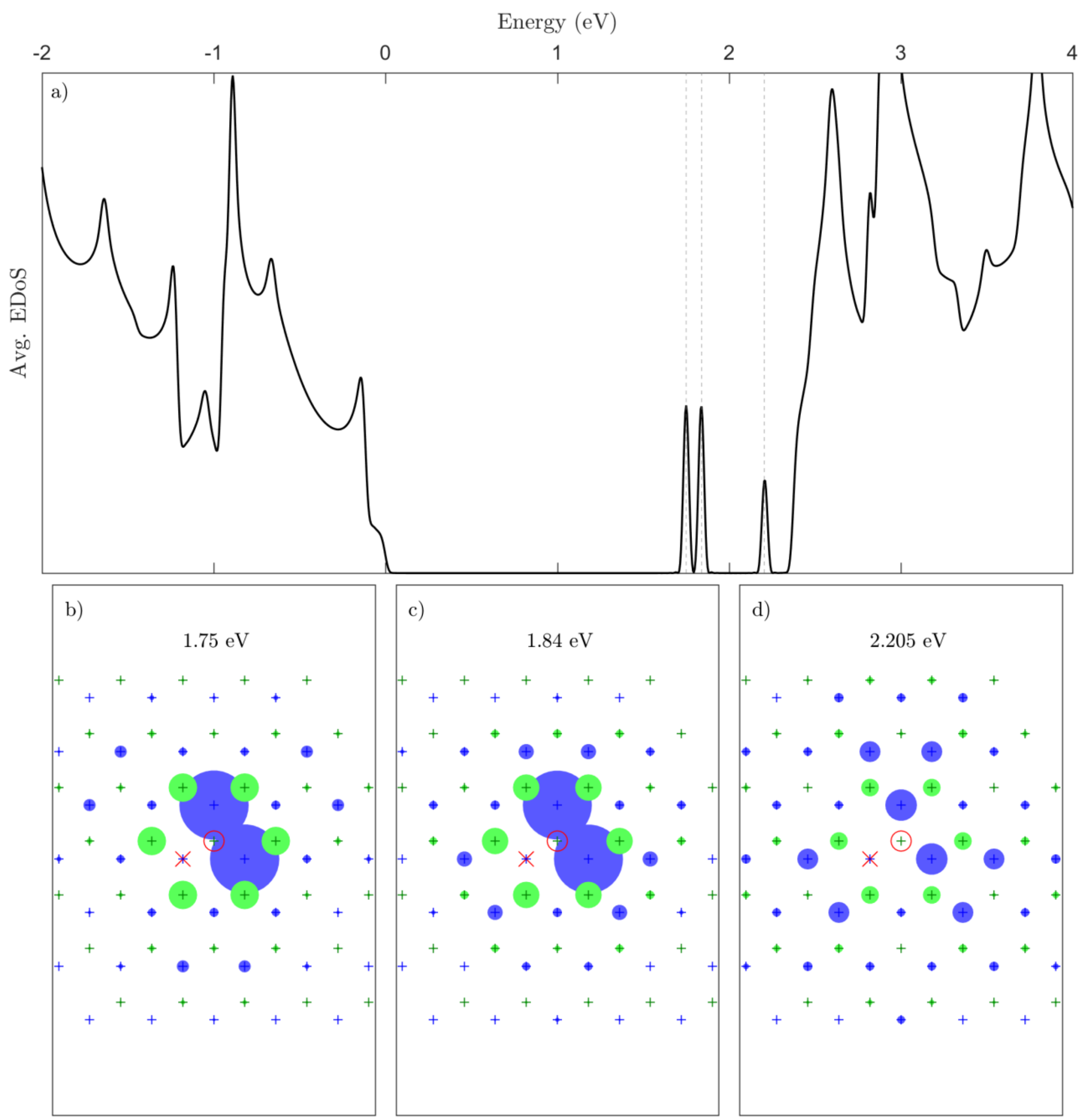
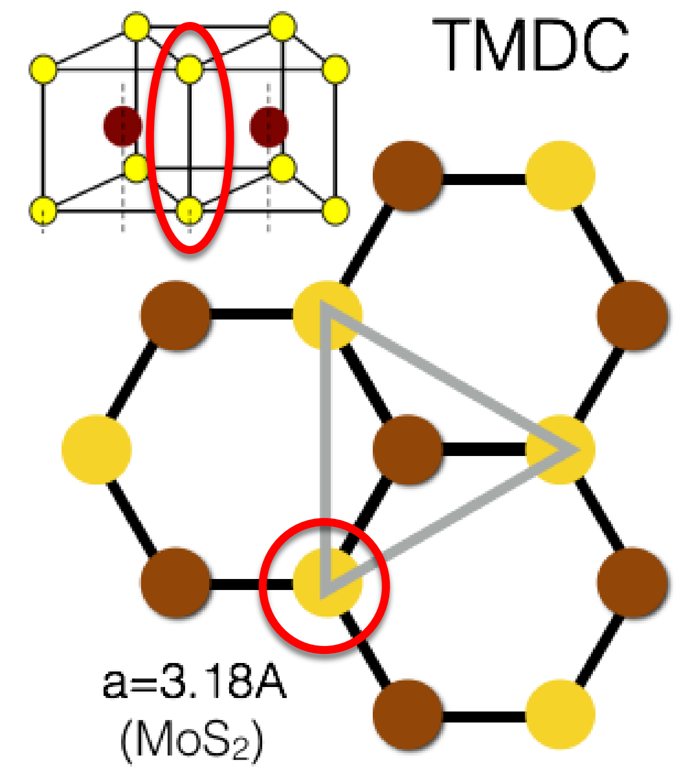
MoS_2 : Modeling TMDCs

Paired S Vacancy Defect in Monolayer





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Method Summary:

- A **multi-scale** approach to physics challenges and applications for engineered devices.

- **Locality** framework, backed up by mathematics and numerics.

- Large, finite **TBM** problems with Kernel Polynomial Method (**KPM**)

- Excellent **parallelization** efficiency

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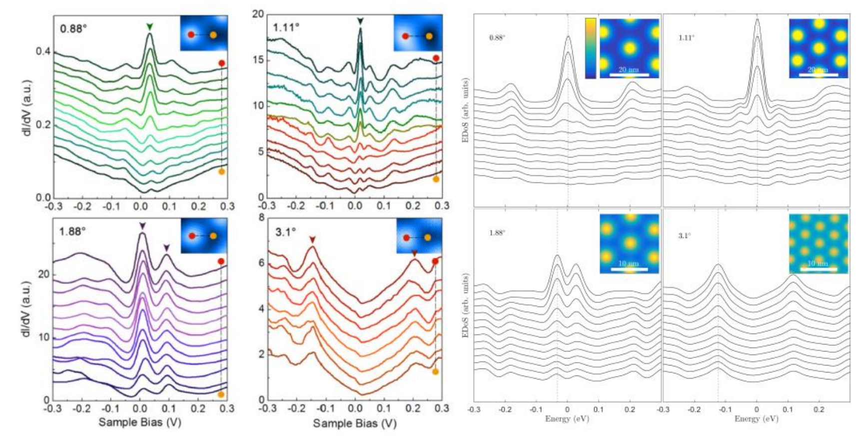
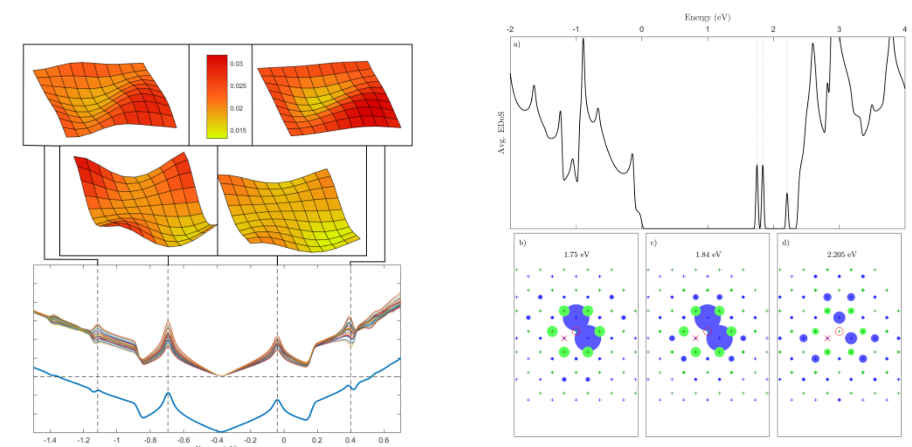
Results Summary:

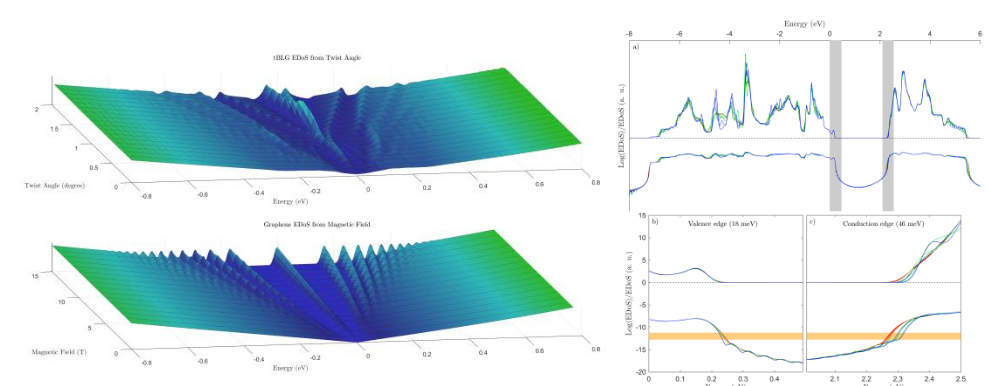
- Agreement with previous experiment and theory in tBLG and TMDC systems



- Simulations of real-space Local EDoS and defect wave functions



- Twist-angle as new “knob” for controlling localized states and doped semiconductors



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