Large scale tight-binding methods: hBN encapsulated multilayer graphene

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Twistronics workshop 2023 - Jan 11-13, Seoul, Korea

Acknowledgments

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Theory graphene/hBN (U. Antwerp)

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Graphene, hBN samples, supermoire experiments

group of K. Novoselov (Manchester)

group of K. Wantanabe, T. Taniguchi (Tsukuba)

Funding:

Outline

1) Large scale tight-binding simulations with Pybinding and Quantum-Kite

- Computational methodology: spectral expansions
- Details of the codes and functionalities

2) Supermoiré in hBN-graphene-hBN

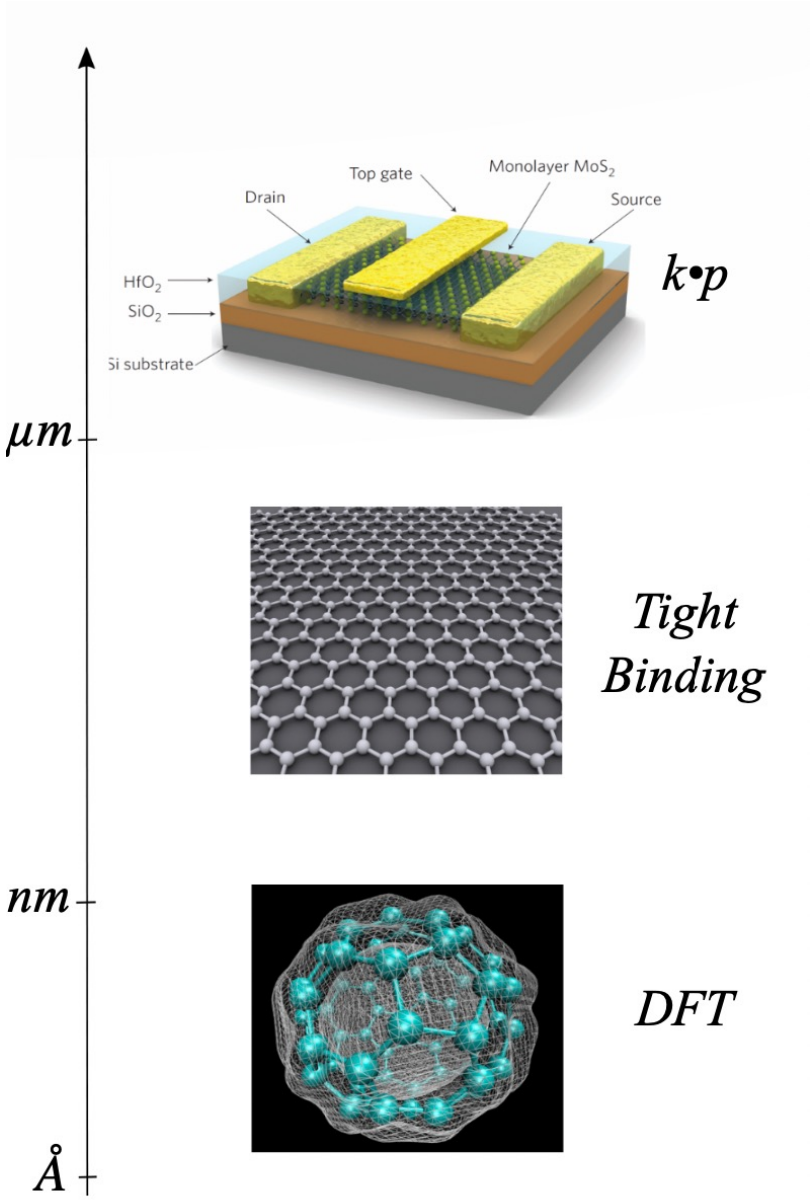
- Tuning the sub-band structure with a double moiré potential

3) Flat bands in hBN encapsulated multilayer graphene

- Tuning the band flatness in Bernal bilayer graphene by gating

Quantum Kite and Pybinding software

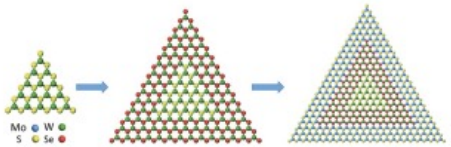
Modelling length scales and methods



$$\hat{H}_{\mathrm{TB}} = \sum_{(i,j,\alpha_i,\beta_j)} t_{\alpha i,\beta j} \, |\alpha_i, i\rangle\langle\beta_j, j|$$

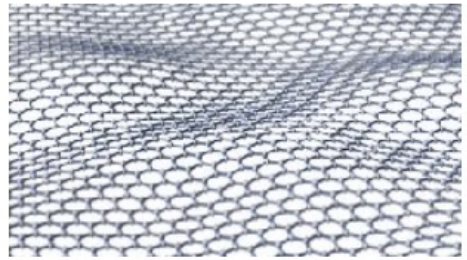
Tight-binding challenges

Scaling challenge



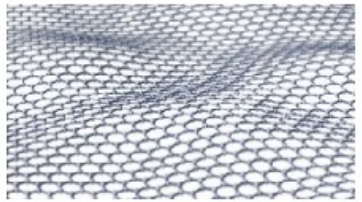
"**Nano**": *L* = 1-100 nm

length scales

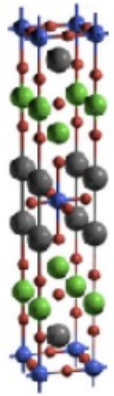


real-size 2D flakes *L* = 1-100 μm

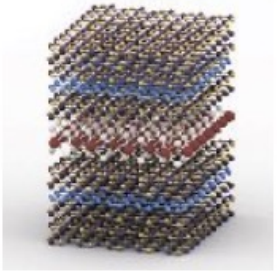
Additional challenge



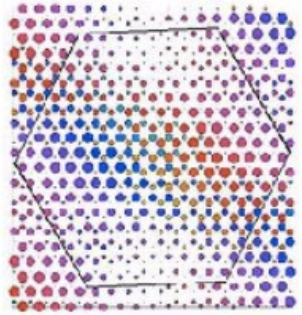
honeycomb layers



complex structure

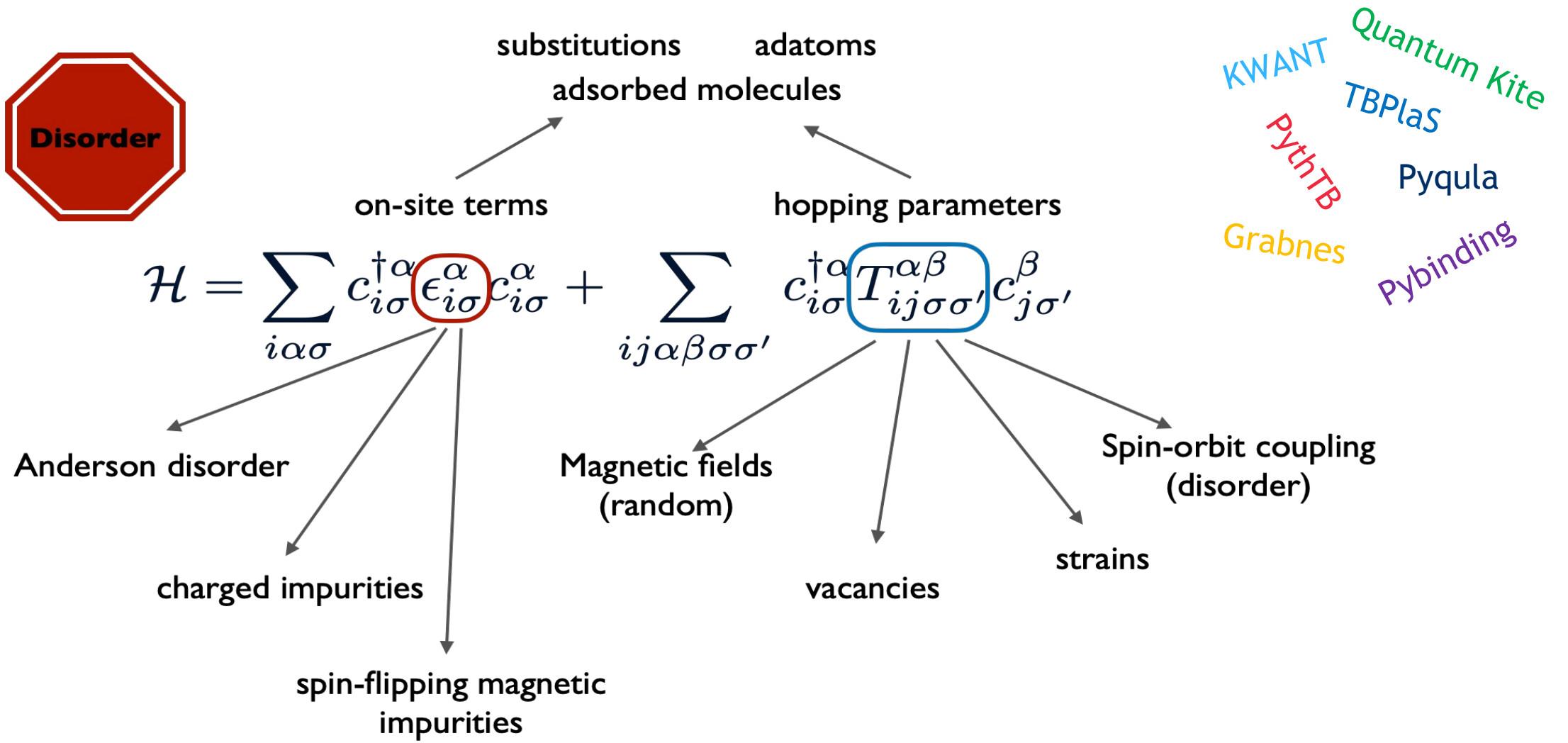


hybrid materials



large unit cell

Real space computational tools (Pybinding + KITE)



https://docs.pybinding.site
https://quantum-kite.com

Spectral methods

target function

$$f(x) = \sum_n \langle \phi_n | f \rangle \phi_n(x)$$

- real variable (e.g., energy)
- x is defined on a finite interval

$\{\phi_n(x)\}$

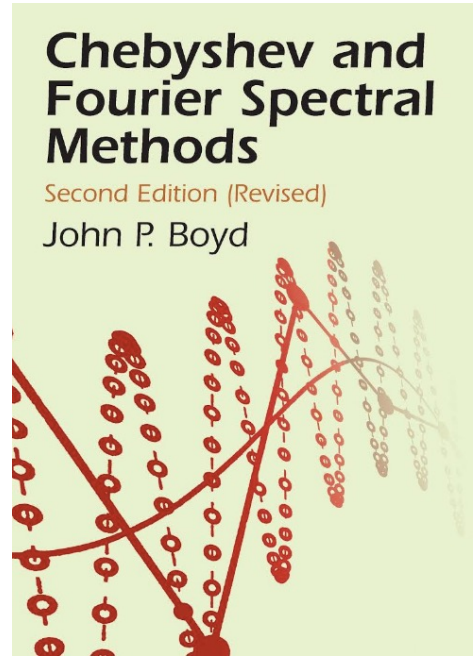
basis set ?

Boyd's Moral principle:

(i) When in doubt, use Chebyshev polynomials

(ii) Unless you are sure another set of basis function is better, use Chebyshev polynomials

(iii) Unless you are really, really sure another set of basis function is better, use Chebyshev polynomials



Spectral methods

Tight-binding → Bounded spectrum: [-1,1]

Operators in frequency space → Chebyshev spectral expansions

Recursive evaluation of moments

$$\mu_{n_1 \ldots n_p} = \langle P_{n_1}(\hat{H}) \cdot \ldots \cdot P_{n_p}(\hat{H}) \rangle$$

moments of N-body operators

$$\mathrm{LDoS}(\vec{x}, \epsilon)$$

$$\sigma_{ij}(\mu, T, \omega, \vec{q})$$

etc...

$$\#\ \text{operations} \propto O \equiv D \times N \times R$$

Hilbert space dimension *(max resolution)*

 # Chebyshev steps *(actual resolution)*

Accuracy

Highly stable, uniform resolution, general purpose

Chebyshev based methods

80s: TalEzer, Kosloff, …	• Chebyshev expansion of evolution operator
90s: Wang, Silver, Roder, …	• Kernel polynomial method (KPM) • Spectral densities • Zero-temperature dynamical correlations
2000 - now: Roche, Mayou, Weisse, Chapelier, Yuan, …	• Finite-temperature dynamical correlations • Electronic transport (wave packet propagation) • Full spectral methods

Ferreira et al., PRB 83 165402 (2011)	• KPM-based moments (“full spectral”) • XX conductivity graphene
Garcia, Covaci & Rappoport, PRL 114 116602 (2015)	• Kubo-Bastin approach (“full spectral”) • DC XX and XY conductivity at finite temperature
Ferreira & Mucciolo, PRL 115, 106601 (2015)	• Chebyshev Polynomial Green’s Function (CPGF) • Single-shot algorithm (3.6 billion atoms)

Green's function in frequency space (KPM)

$$G_{ij}(\omega + i\eta) = \langle i | \frac{1}{\omega + i\eta - \mathcal{H}} | j \rangle$$

Chebyshev expansion of the Green's function

$$G_{ij}(\omega) = \frac{-i}{\sqrt{1-\omega^2}} \left[\mu_0 + 2 \sum_{n=1}^{\infty} \mu_n e^{-in \arccos(\omega)} \right]$$

$$\mu_n = \langle i \mid T_n(\mathcal{H}) \mid j \rangle$$

$$T_n(x) = cos[n \ arcos(x)]$$

$$T_{n+1}(x) = 2xT_n(x) - T_{n-1}(x)$$

Recursive procedure:

$$\begin{aligned} \text{define: } |j_n\rangle &= T_n(H)\,|j\rangle \\ |j_0\rangle &= |j\rangle \\ |j_1\rangle &= H\,|j\rangle \\ |j_{n+1}\rangle &= 2H\,|j_n\rangle - |j_{n-1}\rangle \end{aligned}$$

Sparse matrix-vector

LDOS at site i:

$$\nu_i(\omega) = -\frac{1}{\pi} Im[G_{ii}(\omega + i\delta)]$$

DOS:

$$\nu(\omega) = -\frac{1}{\pi D} \sum_i Im[G_{ii}(\omega + i\delta)]$$

$$\mu_n = \frac{1}{D} Tr[T_n \mathcal{H}]$$

A. Weisse et al., Rev. Mod. Phys. 78, 275 (2006)

Chebyshev Polynomial Green's Function (CPGF)

- Truncate the expansion → Gibbs oscillations → Kernel polynomial method (KPM)
- ~~Jackson kernel ~ 1/M, Lorentz kernel ~ λ/M~~ Use CPGF instead!

$$\hat{\mathcal{G}}(\omega + i\eta) = (\omega + i\eta - \mathcal{H})^{-1} = \frac{1}{i}\int_0^\infty e^{i(\omega+i\eta)t} e^{-i\mathcal{H}t}\, dt$$

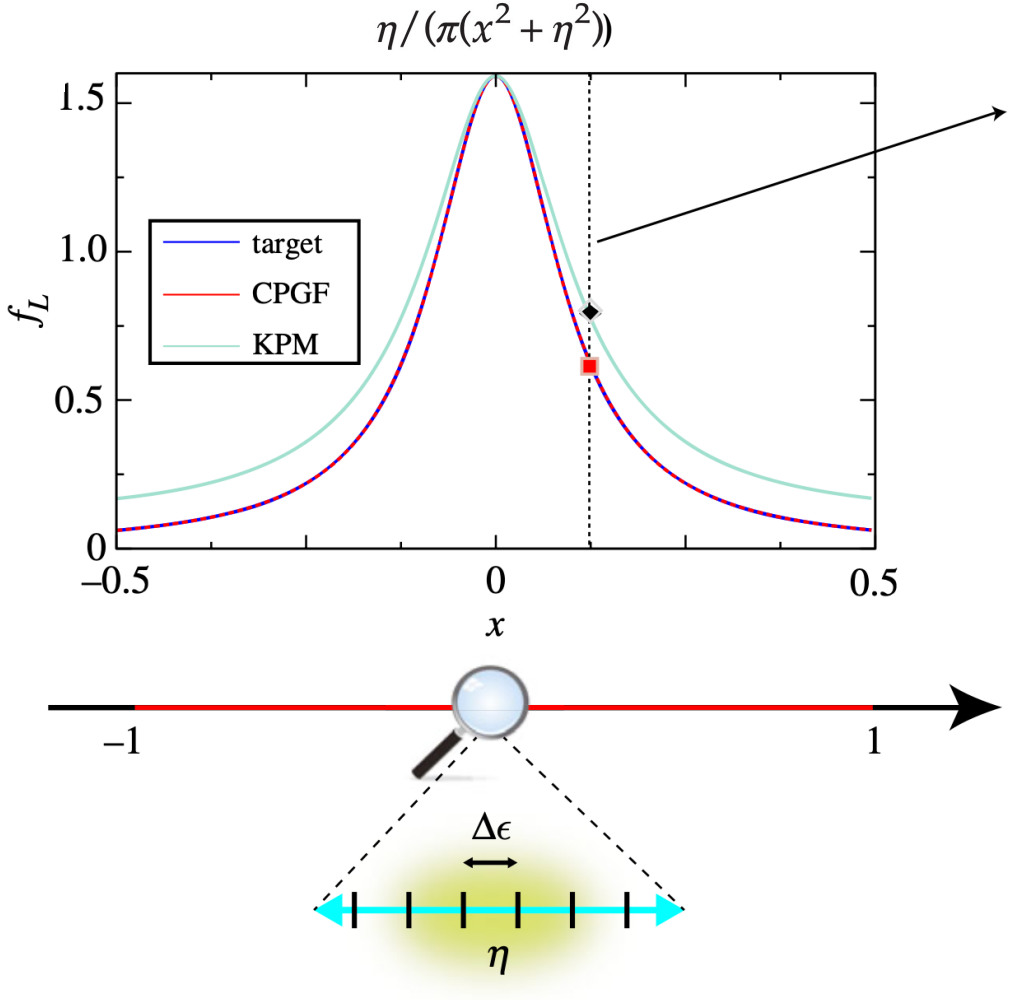
using $$e^{-ixt} = \sum_{n=0}^{\infty} \frac{2i^{-n}}{1+\delta_{0,n}} J_n(t) T_n(x)$$

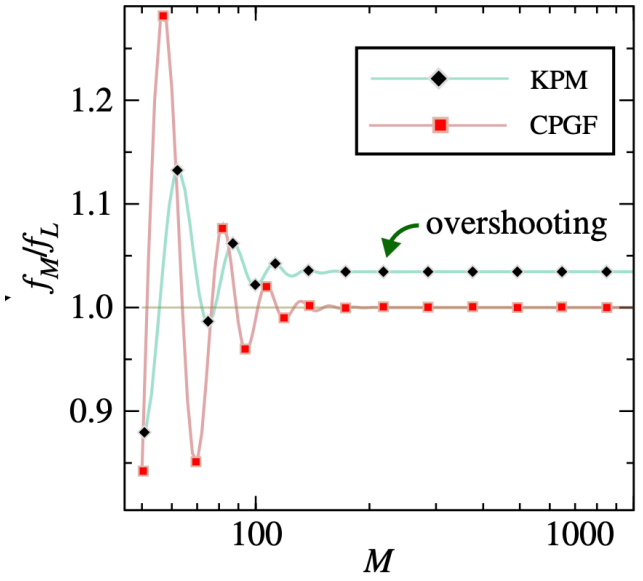
$$\int_0^\infty dt e^{-st} J_n(t) = \frac{(\sqrt{1+s^2} - s)^n}{\sqrt{1+s^2}}$$

$$\hat{\mathcal{G}}(\omega + i\eta) = \sum_{n=0}^{\infty} g_n(\omega + i\eta) T_n(\mathcal{H}) \qquad g_n(z) = \frac{2i^{-1}}{1+\delta_{0,n}} \frac{(z - i\sqrt{1-z^2})^n}{\sqrt{1-z^2}}$$

A Ferreira, E.R. Mucciolo, Phys Rev Lett 115, 106601 (2015)

CPGF compared with KPM (Lorentz kernel)





- KPM (Lorentz) does not converge to the correct function
- CPGF converges for all energies

- 60k x 60k graphene
- 0.4% vacancies
- DOS at Diract point
- KPM (Lorentz) for 1meV

S.M. João, M. Anđelkovic´, L. Covaci, T.G. Rappoport, J. Lopes, A. Ferreira, Royal Society open science 7 (2), 191809 (2020)

Two particle Green's function: optical conductivity

$$\Re\sigma_{xx}(\omega) = \frac{\pi}{\omega\Omega}\int_{-\infty}^{\infty} d\epsilon\,[f(\epsilon) - f(\epsilon+\omega)]\,\mathrm{Tr}\langle \hat{J}_x \hat{A}(\epsilon)\hat{J}_x \hat{A}(\epsilon+\hbar\omega)\rangle$$

$$\hat{J}_x = -\frac{ie}{\hbar}[\hat{x}, \hat{H}] \qquad \hat{A}(\epsilon) = -\frac{1}{\pi}\Im\hat{G}(\epsilon + i\eta)$$

- after performing the Chebyshev expansions (CPGF)

$$\Re\sigma_x x(\omega) = \frac{\pi}{\omega\Omega}\sum_{nm=0}^{N-1} \mu_{nm}\, A_{nm}(\mu, T, \omega)$$

$$\mu_{nm} = \mathrm{Tr}\langle \hat{J}_x\, T_n(\hat{\mathcal{H}})\, \hat{J}_x\, T_m(\hat{\mathcal{H}})\rangle$$

2D expansion!

$$A_{nm}(\omega, \mu, T) = \frac{\pi^2}{W^2}\int_{-\infty}^{\infty} [f(\epsilon) - f(\epsilon+\omega)] a_n(\epsilon) a_m(\epsilon+\omega)$$

T. P. Cysne, T. G. Rappoport, A. Ferreira, J. M. Viana Parente Lopes, N. M. R. Peres, Phys Rev B 94, 235405 (2016)

A. Weisse et al., Rev. Mod. Phys. 78, 275 (2006)

Stochastic evaluation of the trace

- the trace is performed by using statistical averages over R random vectors

$$|r\rangle \equiv (\xi_1^r, \ldots, \xi_N^r), \quad r = 1, \ldots, R$$

$$\langle\langle \xi_i^r \rangle\rangle = 0, \quad \left\langle\left\langle \xi_i^{r*} \xi_j^{r'} \right\rangle\right\rangle = \delta_{r,r'}\delta_{i,j}$$

$$\mathrm{Tr}\langle M\rangle \approx \frac{1}{R}\sum_{r=1}^{R}\langle r|M|r\rangle \qquad \text{with error} \qquad \mathcal{O}(1/\sqrt{RN})$$

- the 2D expansion can be approximated as:

$$\mu_{nm} \approx \frac{1}{R}\sum_{r=1}^{R}\langle r|\hat{J}_x\, T_n(\hat{\mathcal{H}})\; \hat{J}_x\, T_m(\hat{\mathcal{H}})|r\rangle = \frac{1}{R}\sum_{r=1}^{R}\langle \bar{r}_n|r_m\rangle$$

$\langle \bar{r}_n|$ $\qquad$ $|r_m\rangle$

- make use of large amounts of memory to speed-up the simulation
- make use of self-averaging effects: use large systems to reduce R

Quantum Kite

https://quantum-kite.com/

High-performance quantum transport package developed in collaboration with **J. Lopes** (U. Porto), **T. Rappoport** (U.F. Rio de Janeiro) and **A. Ferreira** (U. York).

Challenges

Accuracy: Green's functions are evaluated by means of **real-space polynomial expansions**, providing fine control over accuracy and energy resolution in pristine and disordered systems.

Scalability and speed: **fast and highly-scalable** state-of-the-art algorithms, capable of treating unprecedentedly large computational domains with multi billions of atomic orbitals in large RAM nodes. Highly efficient domain decomposition algorithms take advantage of CPU clusters.

Versatility: Generic equilibrium and out-of-equilibrium properties (e.g., local DOS and optical conductivity) of any system, from complex molecules to hetero-structures with large size unit cell. The **Pybinding interface** allows to easily define/import model parameters and select target functions/algorithms.

Quantum Kite functionalities

Linear optical conductivity

Non-linear optical conductivity

DC (spin) conductivity tensor (xx, xy)

Full spectral
Single shot (xx)

LDOS

ARPES

DOS

Wave packet propagation

KITE
quantum transport software

Large systems

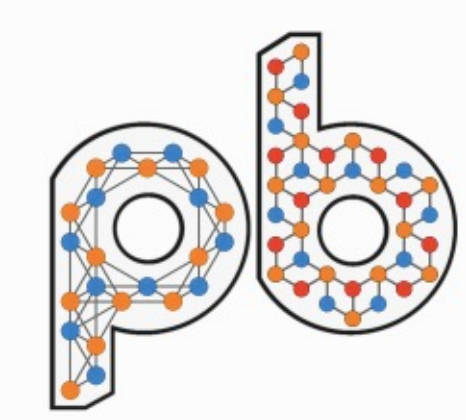
Generic multi-orbital + disorder

Magnetic field

2D or 3D

Quantum Kite workflow

Pybinding



http://docs.pybinding.site/

Pybinding initially developed by **Dean Moldovan** at the **University of Antwerp.**

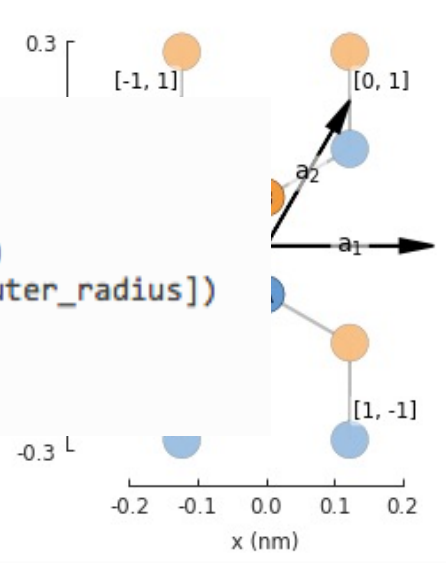
Features:

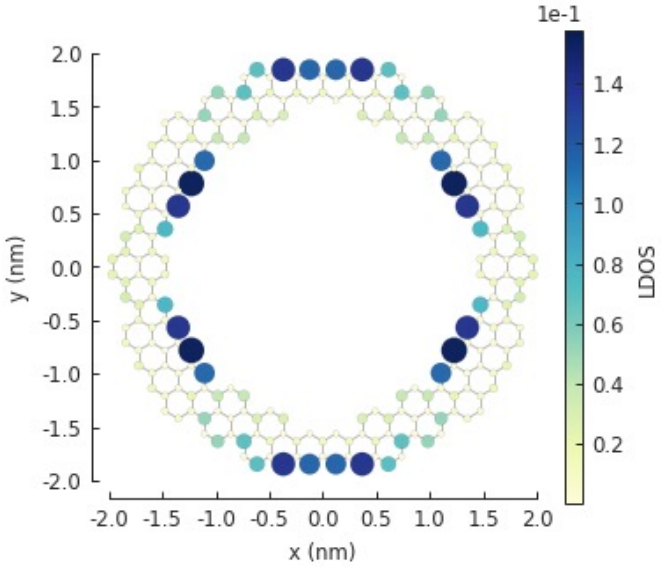
- straightforward definition of generic tight-binding model
- band structure calculation (exact diagonalization: lapack, arpack)
- periodic or finite size (nanoribbons, dots): DOS, LDOS (ED, KPM, CPGF)
- electric and magnetic fields
- defects and strains
- up to millions of orbitals (graphene, TMDs, input Wannier90, …)

```
    lat = pb.Lattice(a1=[a, 0],
```

```
def ring(inner_radius, outer_radius):
    def contains(x, y, z):
        r = np.sqrt(x**2 + y**2)
        return np.logical_and(inner_radius < r, r < outer_radius)
    return pb.FreeformShape(contains, width=[2*outer_radius, 2*outer_radius])

shape = ring(inner_radius=1.4, outer_radius=2)
shape.plot()
```

```
    return lat
```





Lots of tutorials are available!

Example KITE script

Making the lattice:

```
def graphene(onsite=(0, 0)):
    """Make a honeycomb lattice with nearest
neighbor hopping

    Parameters
    ----------
    onsite : tuple or list
        Onsite energy at different sublattices.
    """

    theta = np.pi / 3
    t = 2.8  # eV
    a1 = np.array([1 + np.cos(theta), np.sin(
theta)])
    a2 = np.array([0, 2 * np.sin(theta)])
    lat = pb.Lattice(
        a1=a1, a2=a2
    )
    lat.add_sublattices(
        # name, position, and onsite potential
        ('A', [0, 0], onsite[0]),
        ('B', [1, 0], onsite[1])
    )
    lat.add_hoppings(
        ([0, 0], 'A', 'B', - t),
        ([-1, 0], 'A', 'B', - t),
        ([-1, 1], 'A', 'B', - t)
    )

    return lat
```

Adding the disorder, onsite and structural:

Exporting the configuration:

```
# domain partition
n1 = n2 = 2
# number of unit cells along each direction.
l1 = l2 = 512
# boundaries - PBC
# is_complex - complex Hamiltonian
# precision - 0 float, 1 double, 2 long_double
configuration = kite.Configuration(
    divisions=[n1, n2], length=[l1, l2],
    boundaries=[True, True],
    is_complex=False, precision=1)
# request the calculation of DOS
calculation = kite.Calculation(configuration)
calculation.dos(num_points=4000,
                num_moments=512,
                num_random=5,
                num_disorder=1)
# configure the *.h5 file
kite.config_system(lattice,
                   configuration,
                   calculation,
                   filename='example.h5',
                   disorder=disorder)
```

```
disorder.add_disorder('A', 'Uniform', +1.5, 1.0
)
```

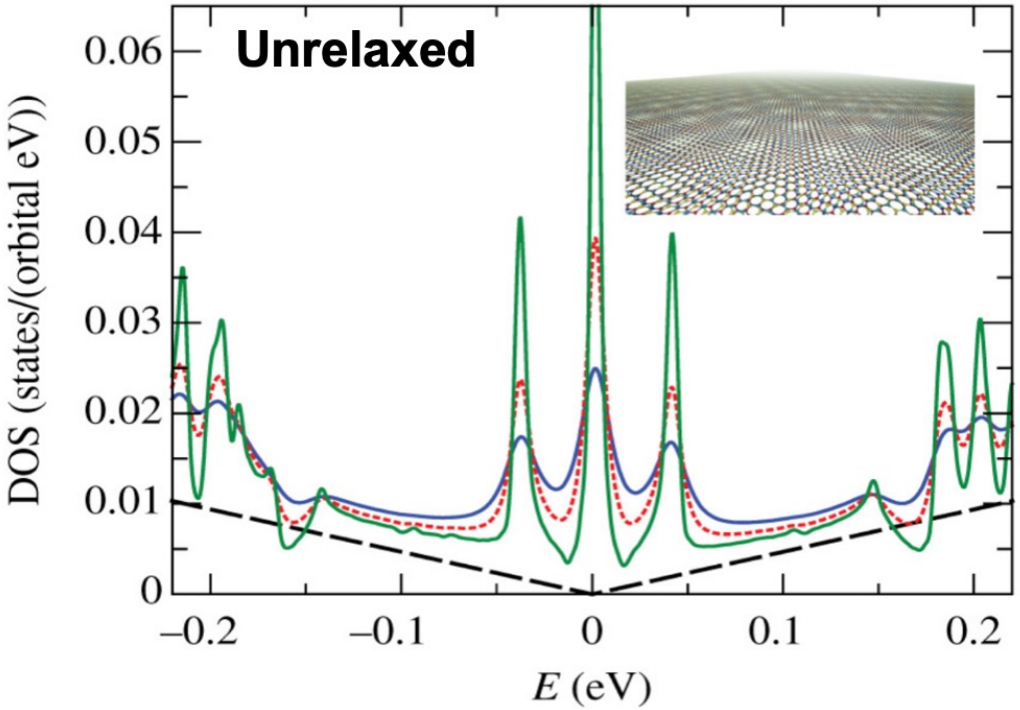
Adding the disorder, onsite and structural:

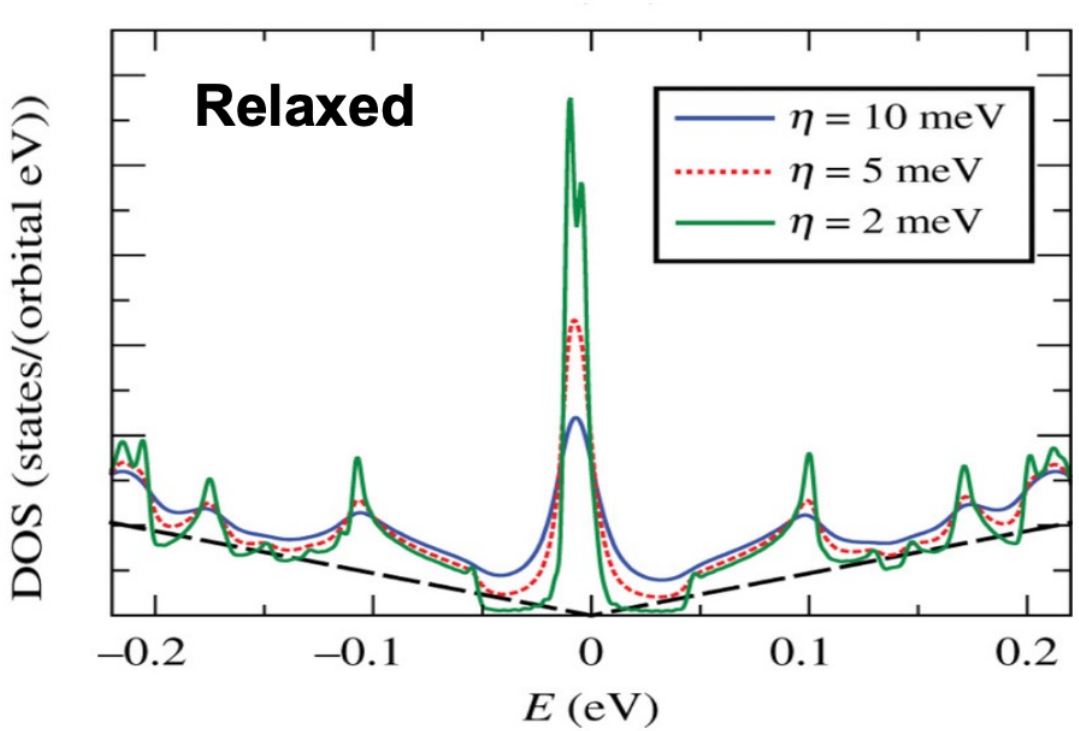
```
import kite

lattice = graphene((-0.5, 0.5))

# Add vacancy disorder:
vacancy_disorder = kite.StructuralDisorder(
lattice, concentration=0.1)
vacancy_disorder.add_vacancy('A')

# Add bond disorder
node0 = [[+0, +0], 'A']
node1 = [[+0, +0], 'B']
node2 = [[+1, +0], 'A']
node3 = [[+0, +1], 'B']

bond_disorder = kite.StructuralDisorder(lattice
, concentration=0.2)
bond_disorder.add_structural_disorder(
    # add bond disorder in the form
    # [from unit cell], 'sublattice_from', [
to_unit_cell], 'sublattice_to', value:
    (*node0, *node1, -0.6),
    (*node2, *node3, +1.2),
    # add onsite disorder
    ([+0, +0], 'A', -0.3)
)

# Adding onsite disorder distribution:
disorder = kite.Disorder(lattice)
disorder.add_disorder('B', 'Deterministic', -1.
0)
disorder.add_disorder('A', 'Uniform', +1.5, 1.0
)
```

KITE simulations

Twisted bilayer graphene

1.05 degs, N=12000, D=11908 x 640 x 512, Z=60





Numerical complexity depends on: num polyn (**N**) x system size (**D**) x coordination number (**Z**)

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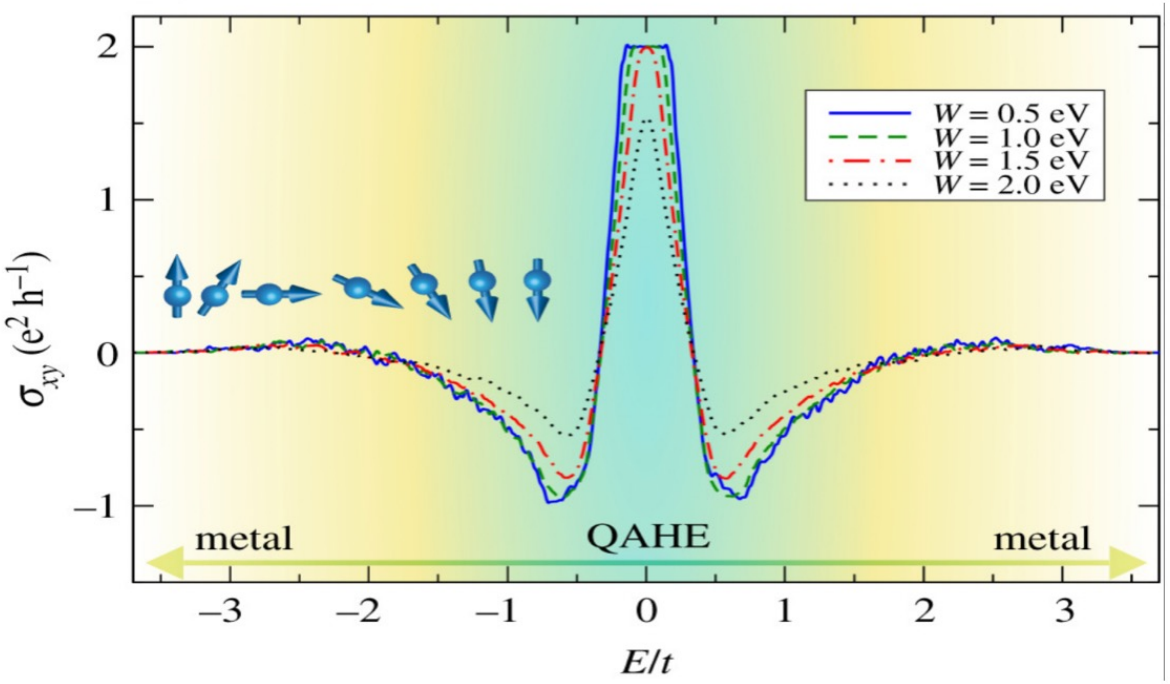
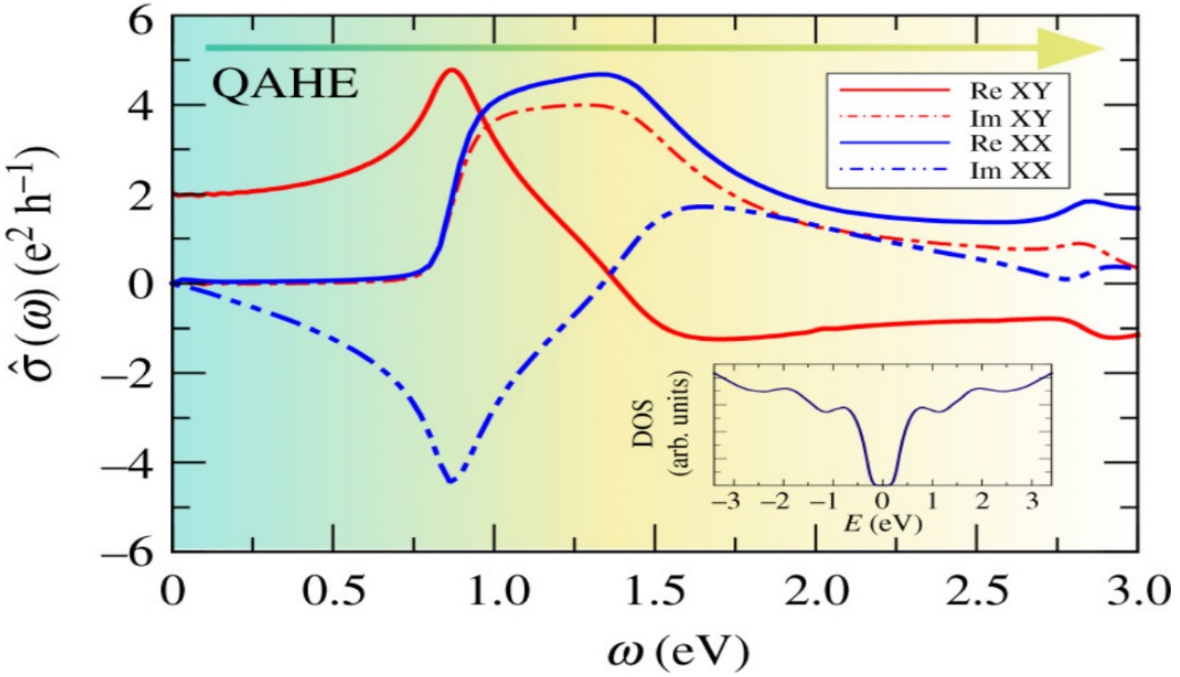
KITE simulations

Anomalous Quantum Hall + disorder

N=2048x2048, D= 4 x 8192 x 8192, Z=3





Numerical complexity depends on: num polyn (**N**) x system size (**D**) x coordination number (**Z**)

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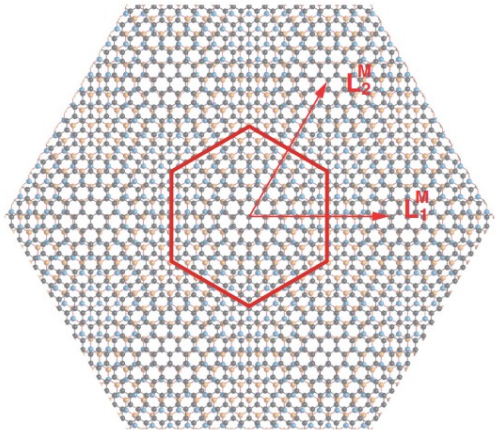
Comparison between Pybinding and KITE

	Pybinding	Quantum Kite
Hamiltonian storage	**sparse matrix storage**	**matrix free**
Parallelization (spectral methods)	**OpenMP***	**OpenMP**
Periodic, open BC	**x**	**x**
3D systems	**-**	**x**
Inhomogeneous strains	**x**	**-**
Magnetic field	**x**	**x**
LDOS, DOS, Green's func., spectral func. (KPM, CPGF)	**x**	**x**
Green's functions (KPM, CPGF)	**x**	**x**
Band structure calculation	**x**	**-**
Single shot XX conductivity (T=0)	**-**	**x**
Full spectral conductivity (XX, XY) (finite T)	**x**	**x**
Optical conductivity	**-**	**x**
Non-linear optical conductivity	**-**	**x**

Supermoiré in hBN encapsulated graphene

Moiré: aligned graphene on hBN

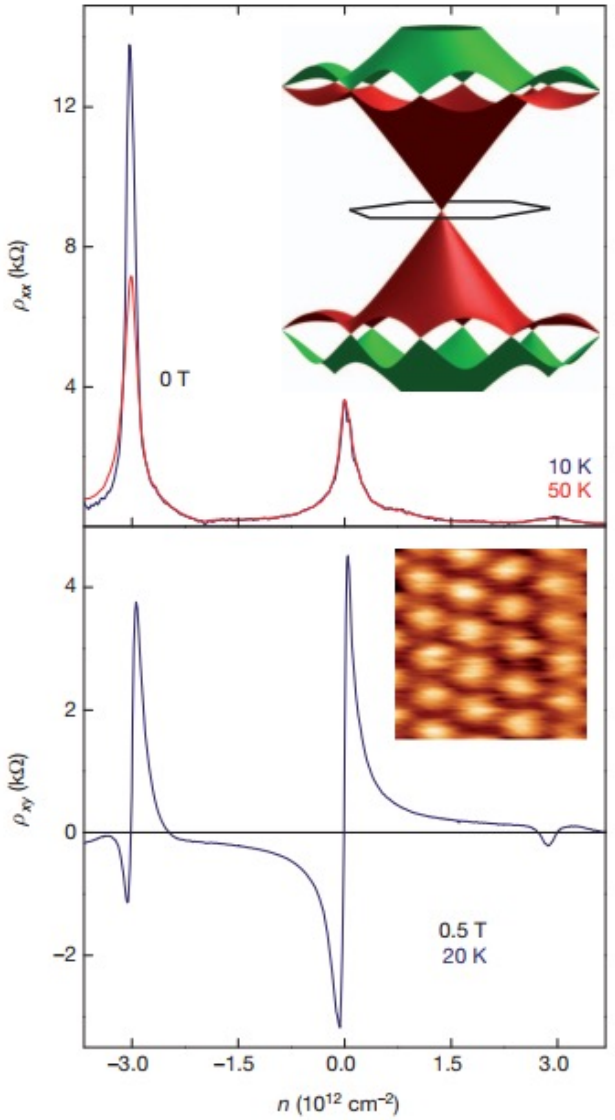
- hBN is an atomically flat 2D insulator → excellent for graphene devices
- Due to the small lattice mismatch between the two materials a moiré superlattice appears **(~13.9nm)**
- Moiré periodicity reconstructs the spectrum of graphene into minibands → secondary Dirac points emerge at $\pm E_D$



$$\mathbf{L}_i^{\mathrm{M}} = (\mathbf{1} - R^{-1}M^{-1})^{-1}\mathbf{a}_i \quad (i = 1,2)$$

$$M = (1+\varepsilon)\mathbf{1}$$

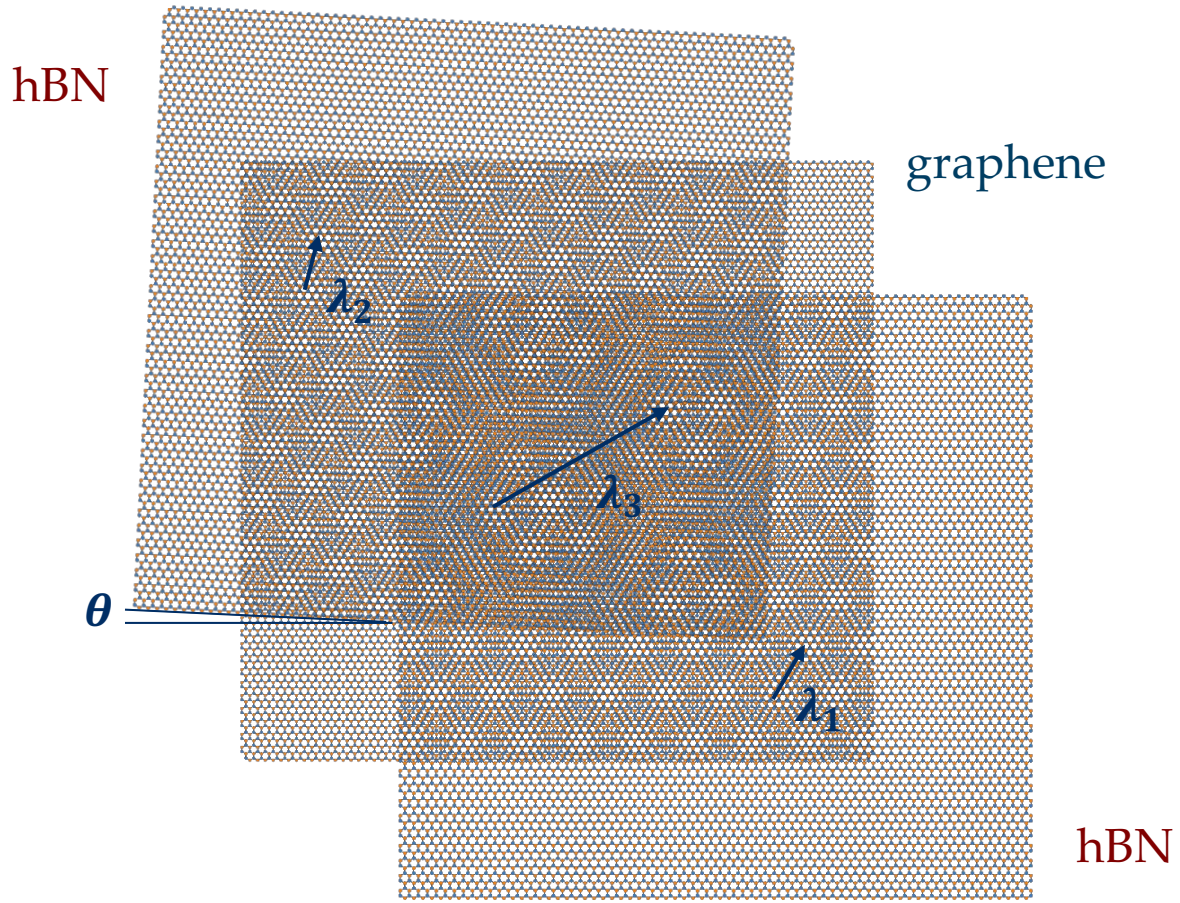
$$E_D = \pm 2\pi\hbar v_F/(\sqrt{3}\lambda)$$



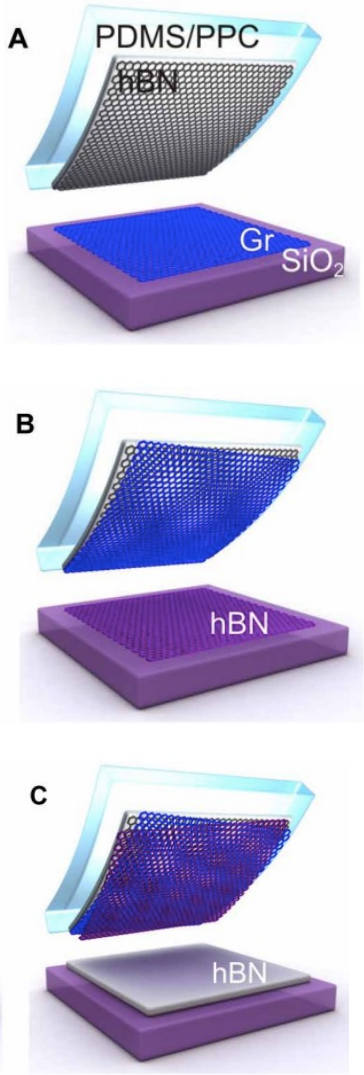
L. A. Ponomarenko *et al.*, Nature 497, 594 (2013)

Doubly aligned hBN - graphene - hBN

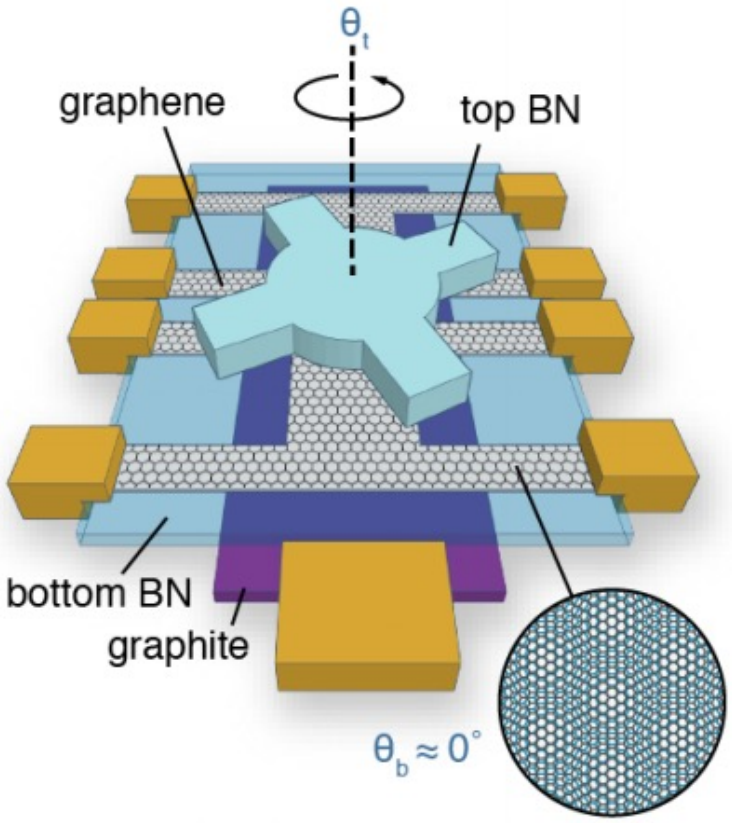
- *Question*: What happens when encapsulated graphene is almost aligned with both hBN layers



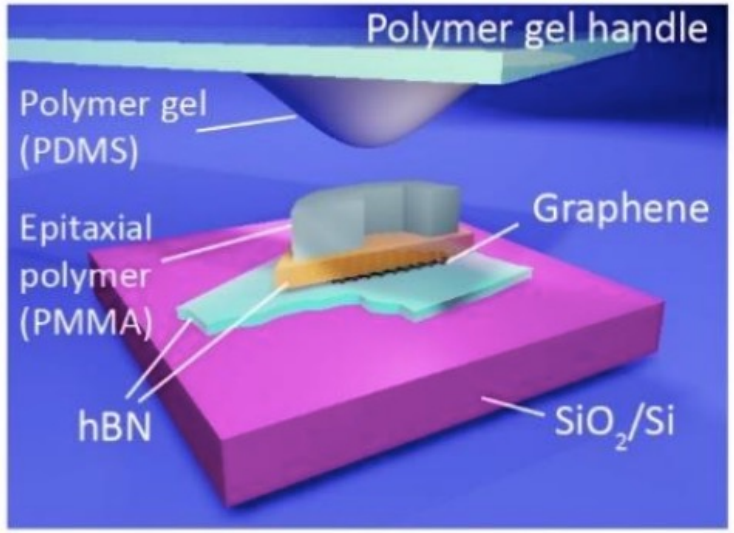
Experimental alignment procedure (in-situu)

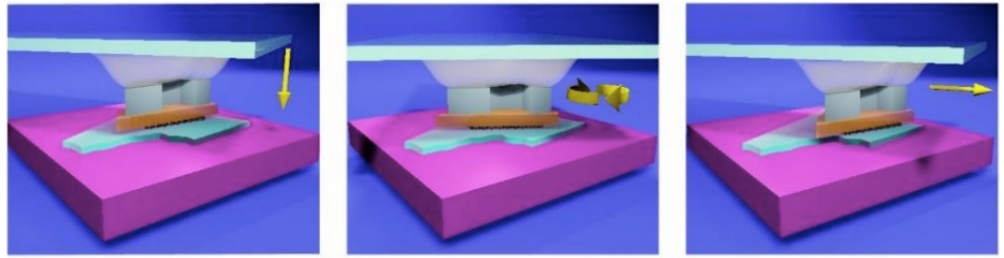


Z. Wang *et al.*, Sci. Adv. 5, eaay8897 (2019)



N. R. Finney *et al.*, Nat. Nanotech. 14, 1029 (2019)

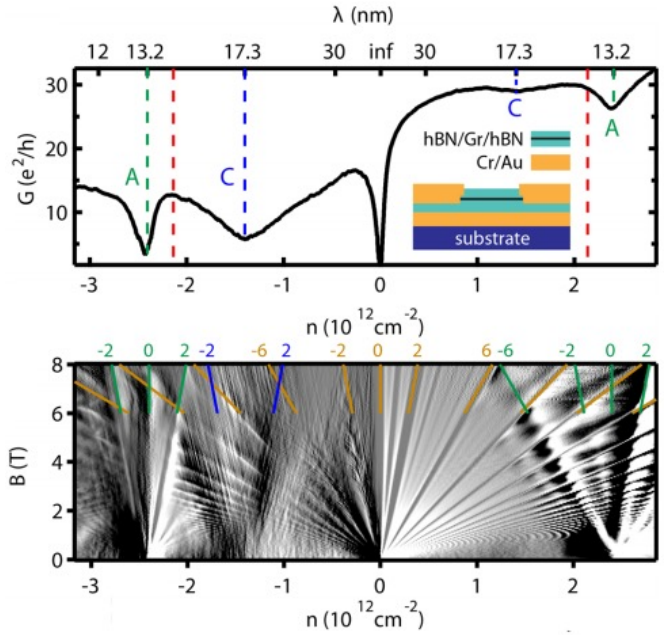


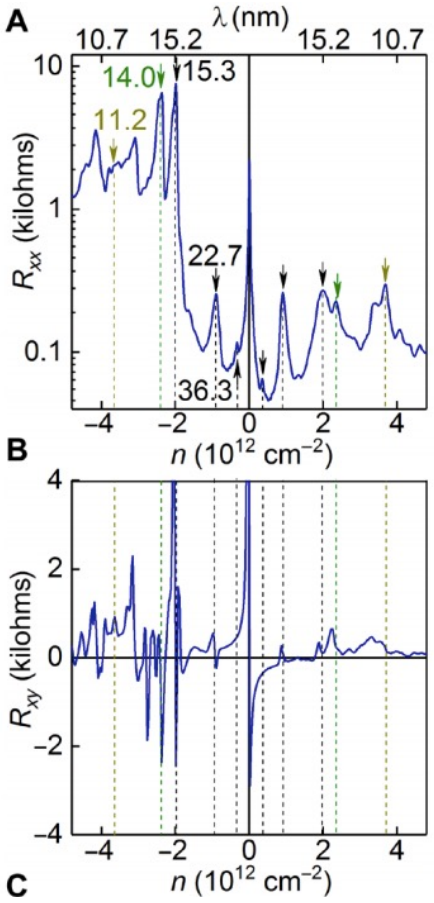


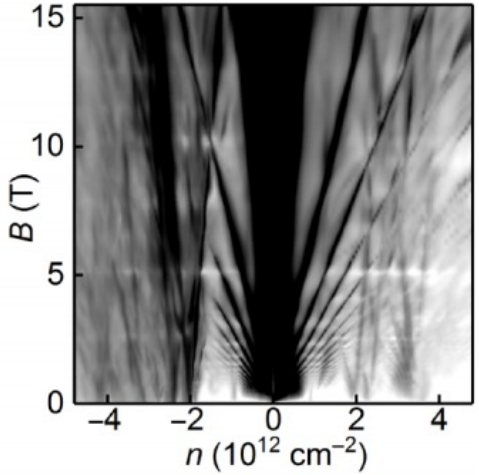
Y. Yang, …, A. Mischenko, Science Advances, 6, eabd3655 (2020)

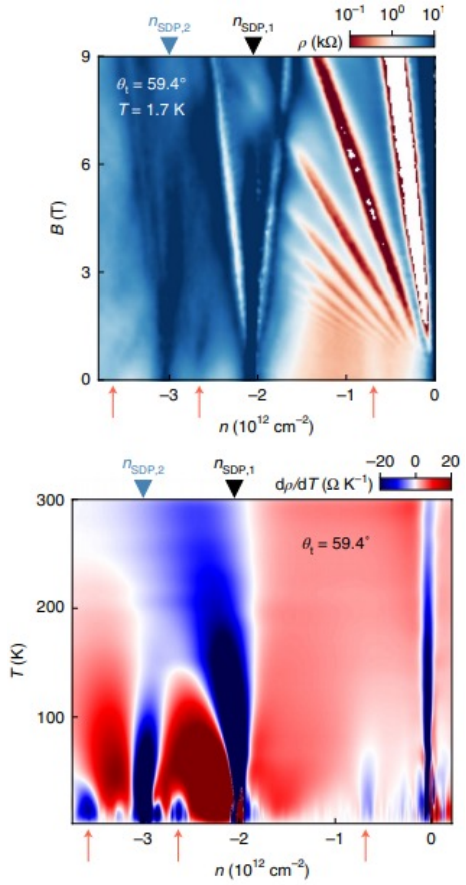
Moiré of moiré → Supermoiré

- Experiments on doubly aligned systems show SDPs lower energies → large periodicities









L. Wang *et al.*, Nano Lett. 19, 2371 (2019)

Z. Wang *et al.*, Sci. Adv. 5, eaay8897 (2019)

N. R. Finney *et al.*, Nat. Nanotech. 14, 1029 (2019)

Induced potential landscape

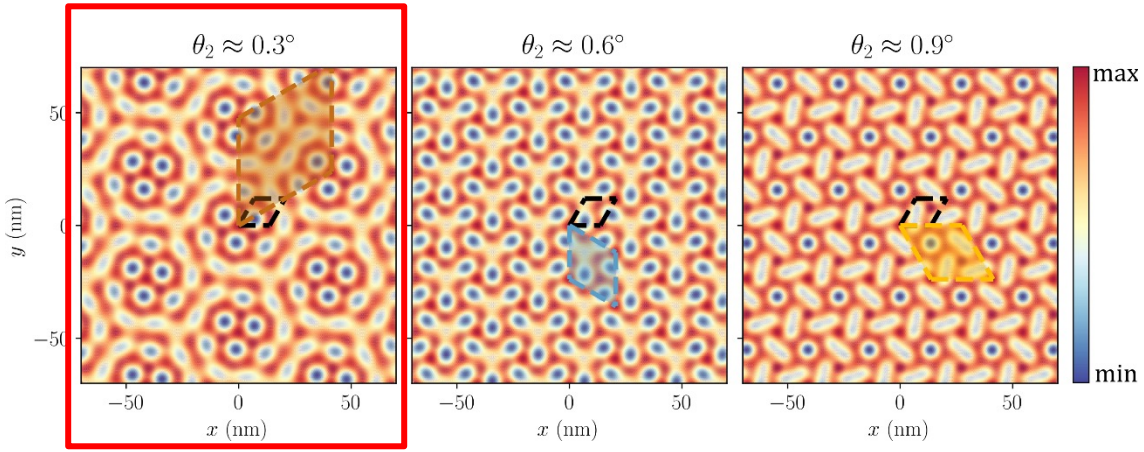
- Effective model for graphene on hBN [1]

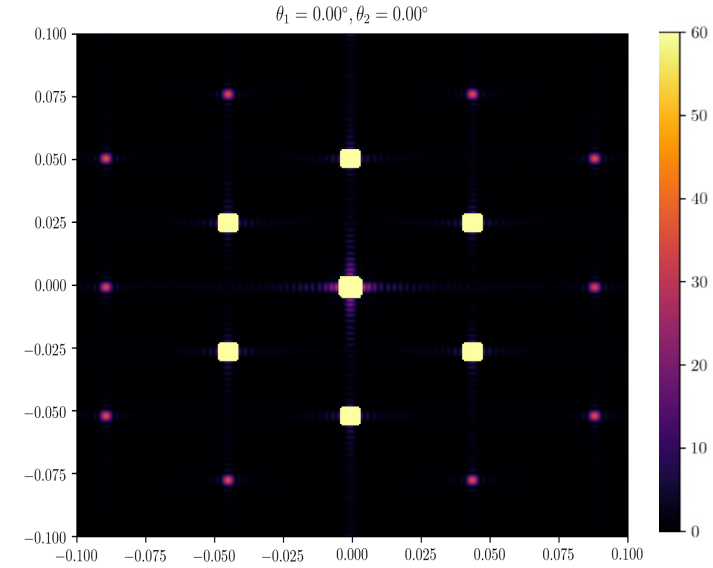
$$H = H_0 + \hbar v b U_1 f_+(\boldsymbol{r}) + i\xi\hbar v b U_2 \sigma_z f_-(\boldsymbol{r}) + i\xi\hbar v U_3 \left(\sigma_y \partial f_-/\partial x - \sigma_x \partial f_-/\partial y\right)$$

$$f_\pm(\boldsymbol{r}) = \sum_{m=0}^{5} (\pm 1)^m e^{i\boldsymbol{b}_m \boldsymbol{r}} = \pm f_\pm(-\boldsymbol{r})$$

$$\boldsymbol{b}_m = \frac{4\pi}{\sqrt{3}a} \hat{R}_{\pi m/3} \left[1 - (1+\delta)^{-1} \hat{R}_\theta\right] \begin{pmatrix} 0 \\ 1 \end{pmatrix}$$

- We assume that bottom hBN is aligned with graphene (θ_1 = 0) and the top hBN layer is rotated by angle θ_2





[1] J. R. Wallbank, Phys. Rev. B 87, 245408 (2013)

Geometrical consideration

- Reciprocal supermoiré vectors are given as the combination of reciprocal moiré vectors

$$\mathbf{b}^{\mathrm{SM}} = \mathbf{b}^{\mathrm{SM}}(i, j, k, l) = i\mathbf{b}_1^{M_1} + j\mathbf{b}_2^{M_1} - k\mathbf{b}_1^{M_2} - l\mathbf{b}_2^{M_2}$$

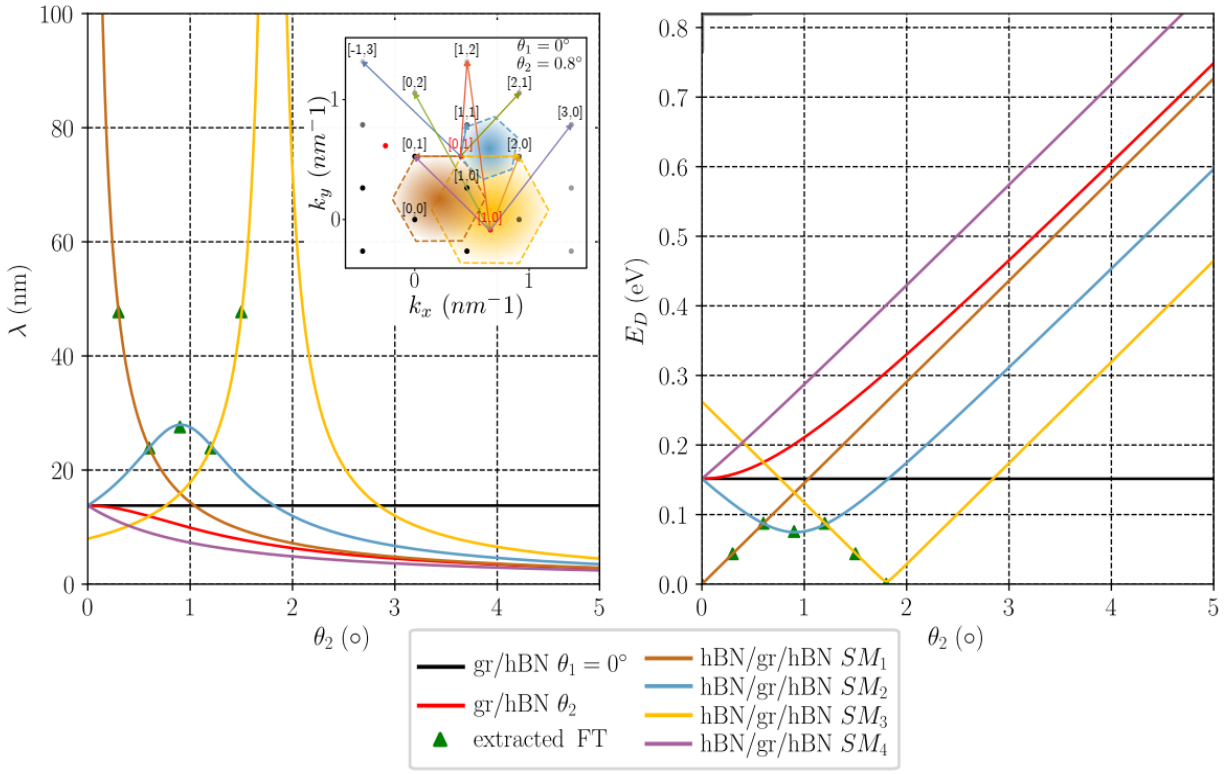
$$\lambda_1^{SM} = \frac{a(1+\delta)}{\sqrt{2-2\cos(\theta_2)}}, \qquad \mathbf{b}_1^{SM} = \mathbf{b}^{SM}(0,1,0,-1),$$

$$\lambda_2^{SM} = \frac{a(1+\delta)}{\sqrt{(2-\delta)(1-\cos(\theta_2)) + \delta^2 - \sqrt{3}\delta\sin(\theta_2)}}, \qquad \mathbf{b}_2^{SM} = \mathbf{b}^{SM}(0,1,-1,-1),$$

$$\lambda_3^{SM} = \frac{a(1+\delta)}{\sqrt{2+3\delta^2-2\cos(\theta_2) - 2\sqrt{3}\delta\sin(\theta_2)}}, \qquad \mathbf{b}_3^{SM} = \mathbf{b}^{SM}(0,1,-2,0),$$

$$\lambda_4^{SM} = \frac{a(1+\delta)}{\sqrt{(2+\delta)(1-\cos(\theta_2)) + \delta^2 + \sqrt{3}\delta\sin(\theta_2)}}, \qquad \mathbf{b}_4^{SM} = \mathbf{b}^{SM}(1,0,0,-1),$$

$$E_D = \pm 2\pi\hbar v_F/(\sqrt{3}\lambda)$$



Electronic properties: tight-binding model

- We use tight-binding technique to calculate electronic properties of the hBN/graphene/hBN trilayer

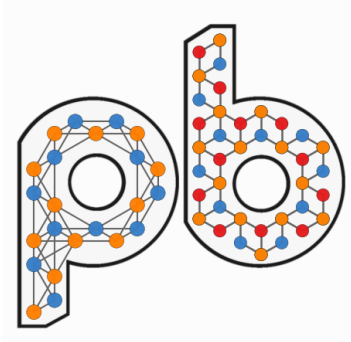
$$H = -\sum_{i,j} t(\mathbf{R}_i - \mathbf{R}_j)|\mathbf{R}_i\rangle\langle\mathbf{R}_j| + \sum_i V(\mathbf{R}_i)|\mathbf{R}_i\rangle\langle\mathbf{R}_i|$$

$$-t(\mathbf{R}) = V_{pp\pi}\left[1 - \left(\frac{\mathbf{R}\cdot\mathbf{e}_z}{R}\right)^2\right] + V_{pp\sigma}\left(\frac{\mathbf{R}\cdot\mathbf{e}_z}{R}\right)^2$$

$$V_{pp\pi} = V^0_{pp\pi}\exp\left(-\frac{R-a_0}{r_0}\right)$$

$$V_{pp\sigma} = V^0_{pp\sigma}\exp\left(-\frac{R-d_0}{r_0}\right)$$

Typical DOS calculation
System: ~5M atoms
Hamiltonian: ~200M non-zero elements



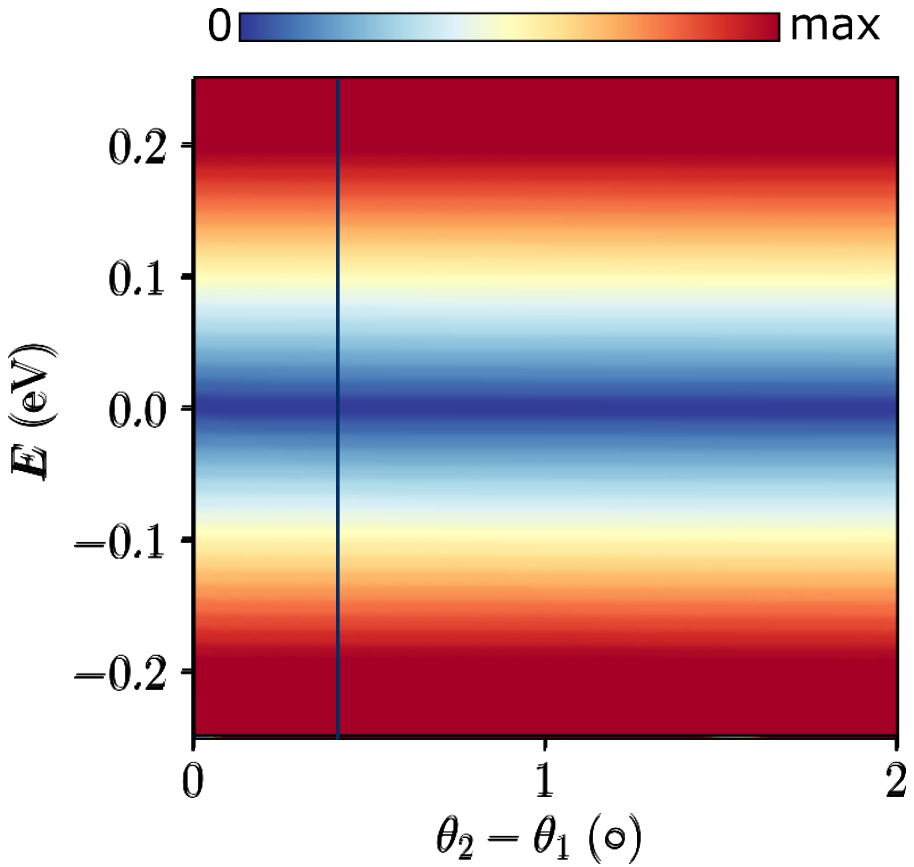
www.pybinding.site

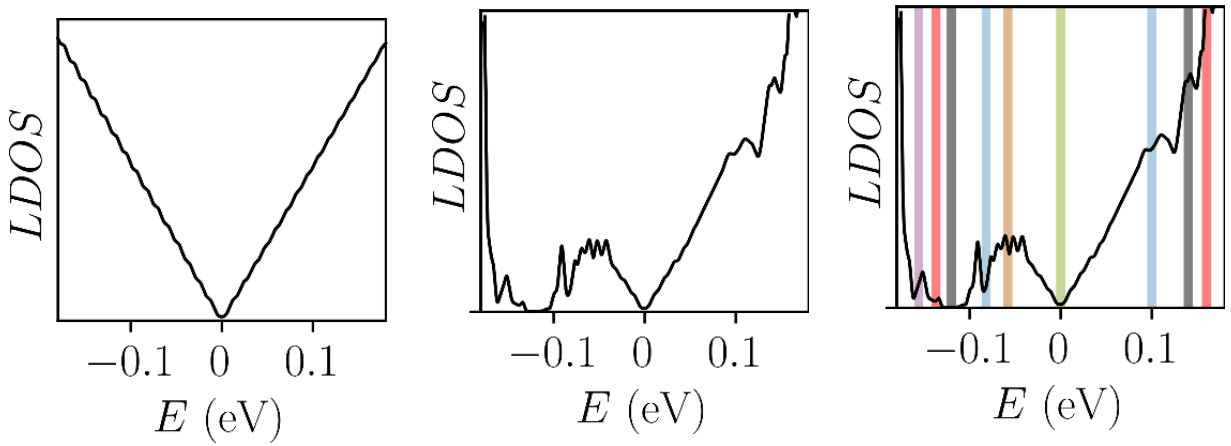
KITE
quantum transport software

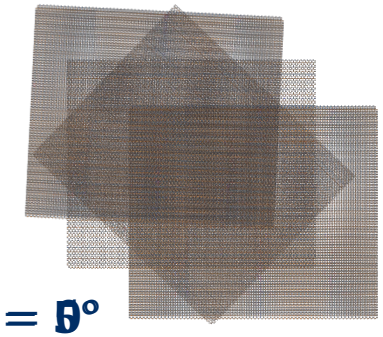
www.quantum-kite.com

LDOS calculation for a finite system

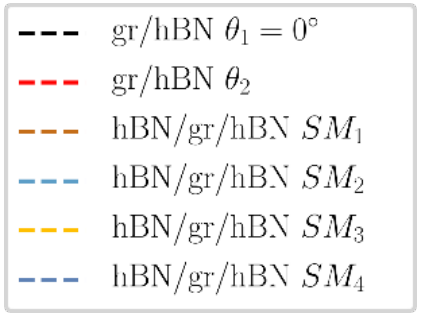
- We calculate LDOS for a point in the center of the unit cell in a disc ~150nm radius



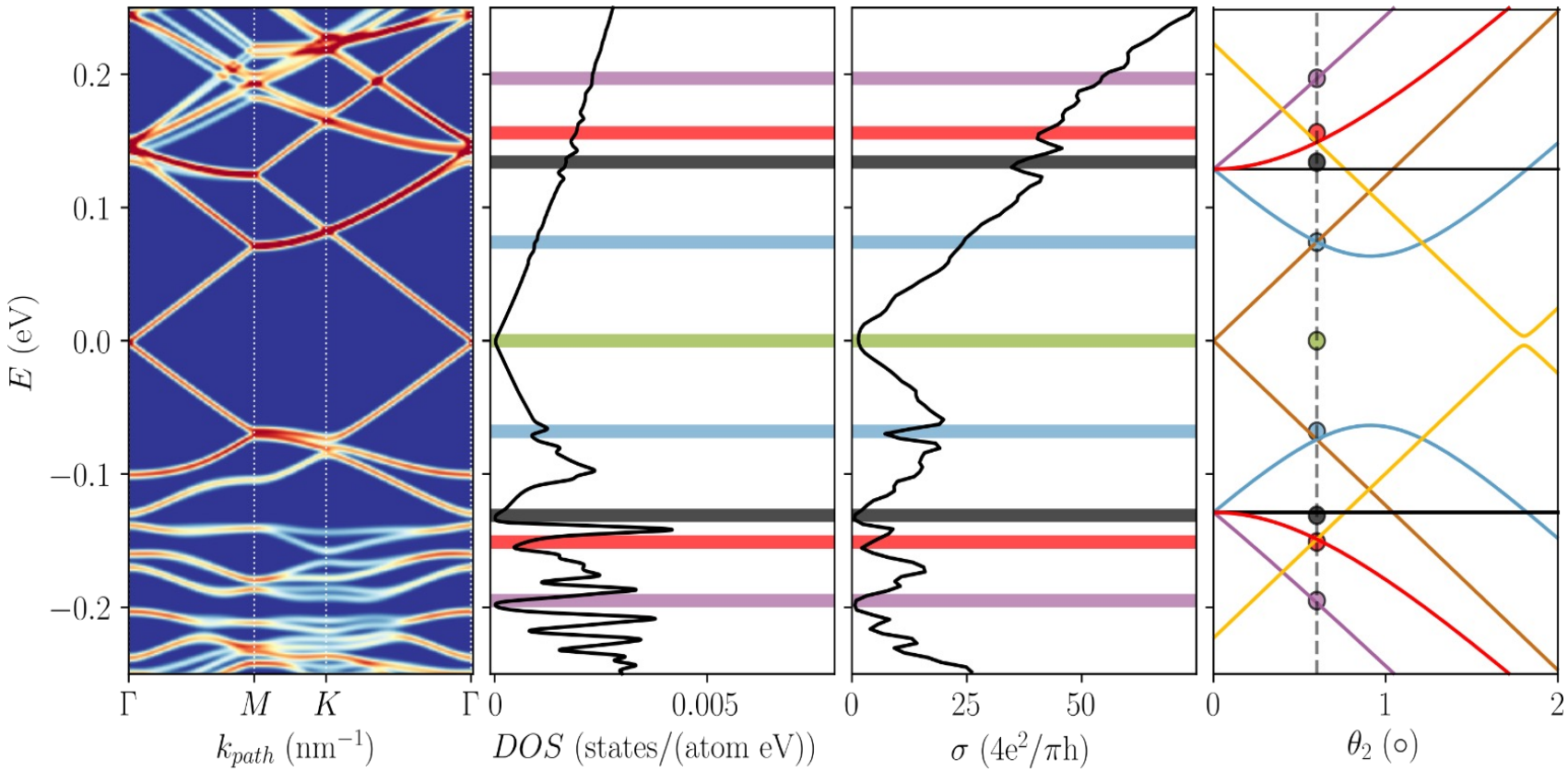




$\theta_1 = 0°$

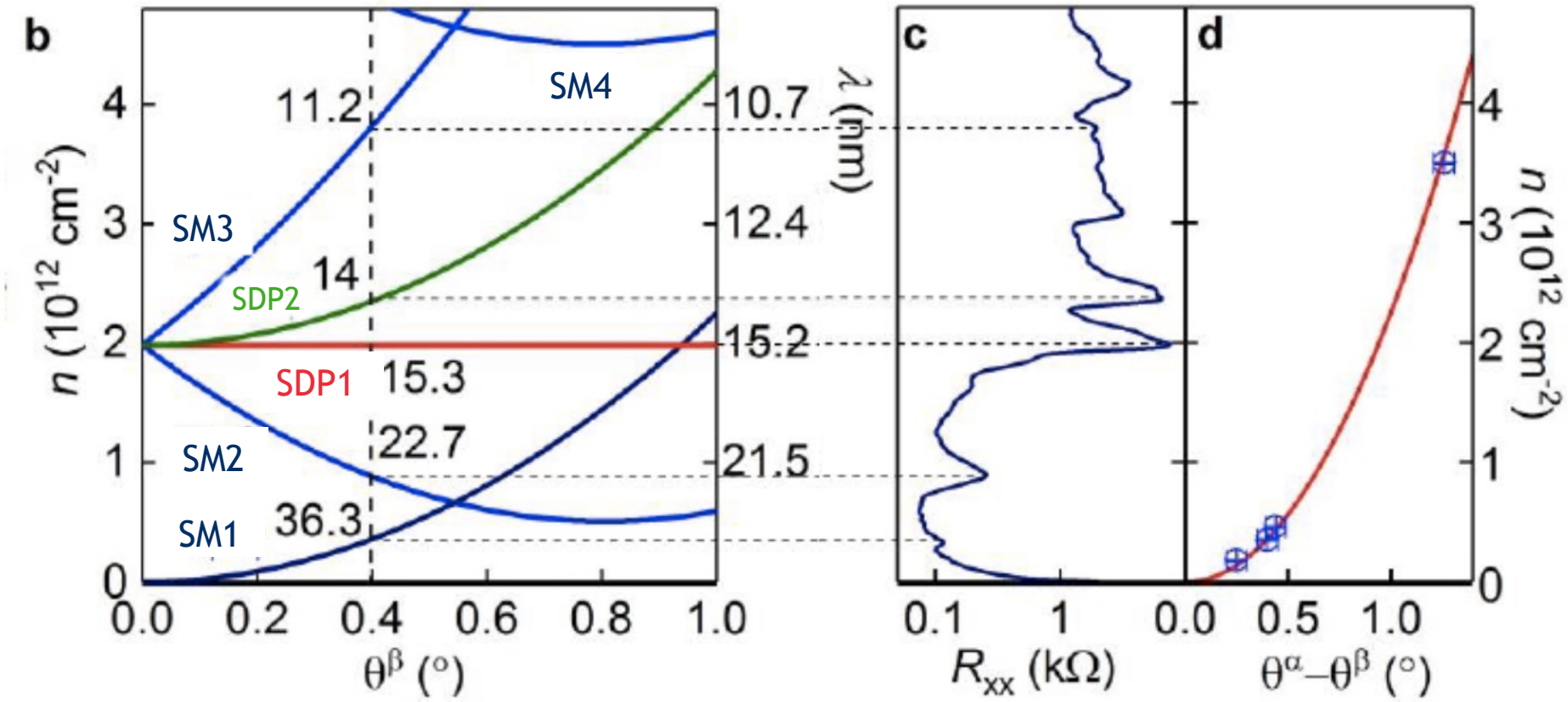


Spectral function, DOS, and conductivity at 0.6°



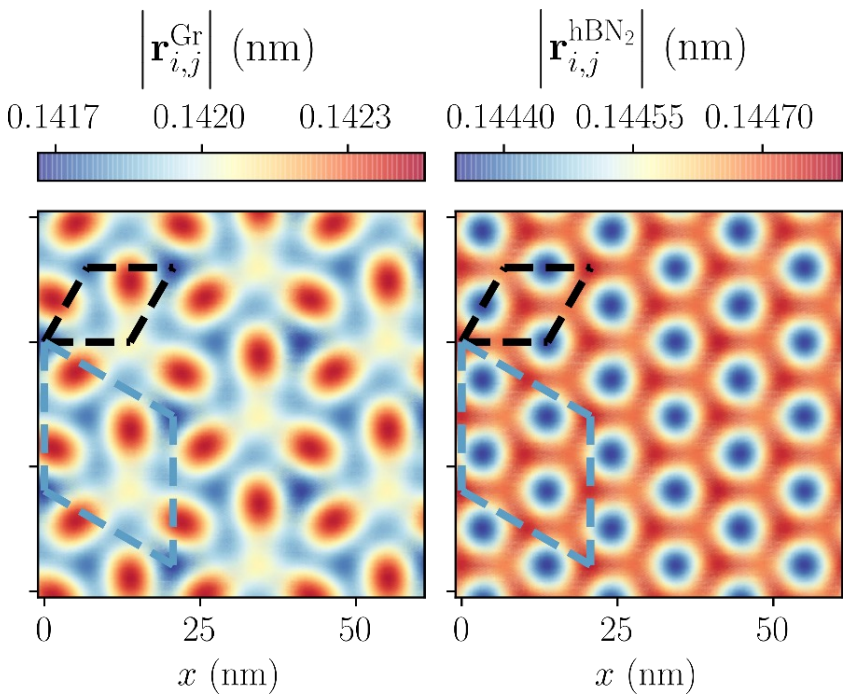
- Signatures of SM are found in global quantities, as well
- Strong reconstruction of the low-energy band structure

Comparison with experiment

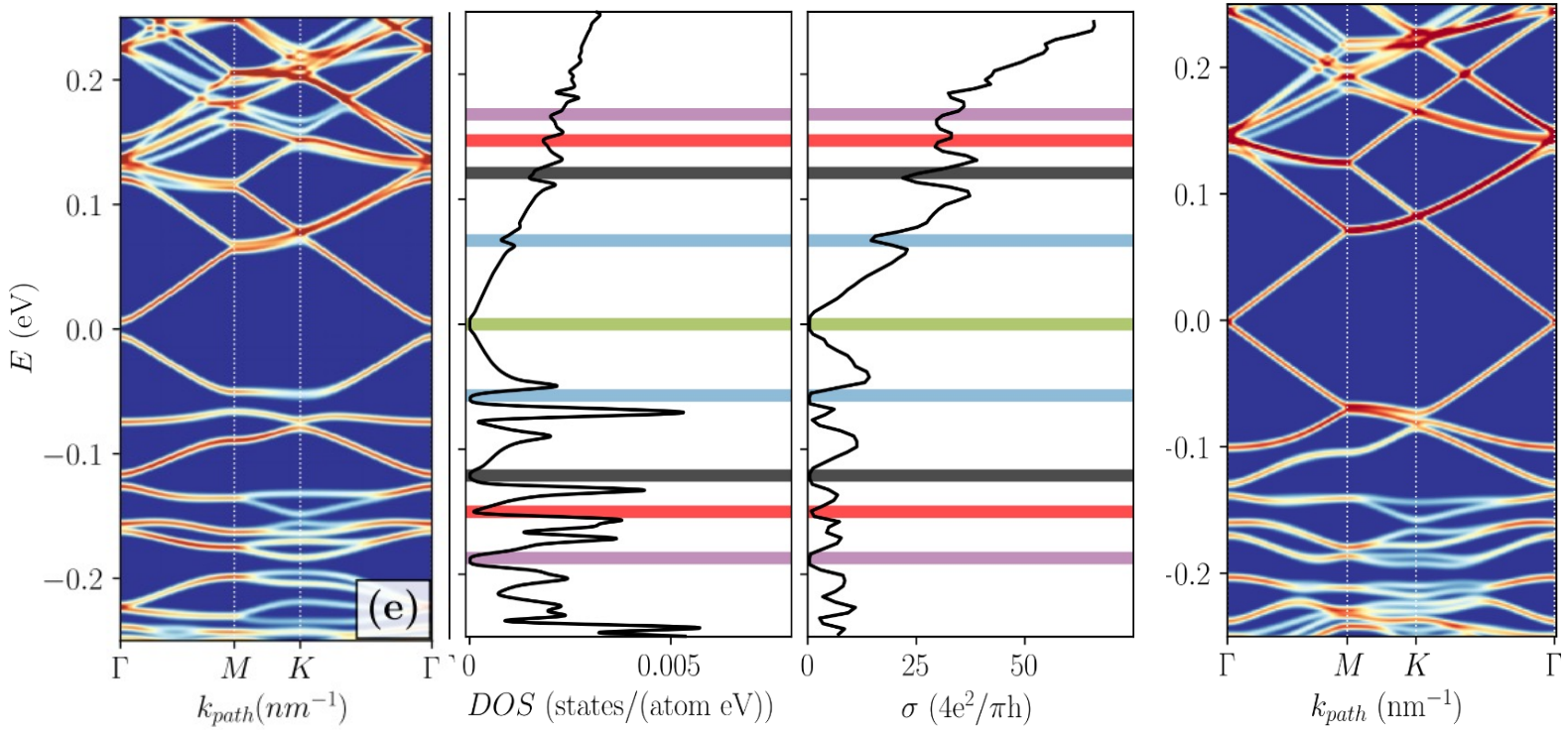


Z. Wang *et al.*, Sci. Adv. 5, eaay8897 (2019)

Effects of relaxation (MD simulation)



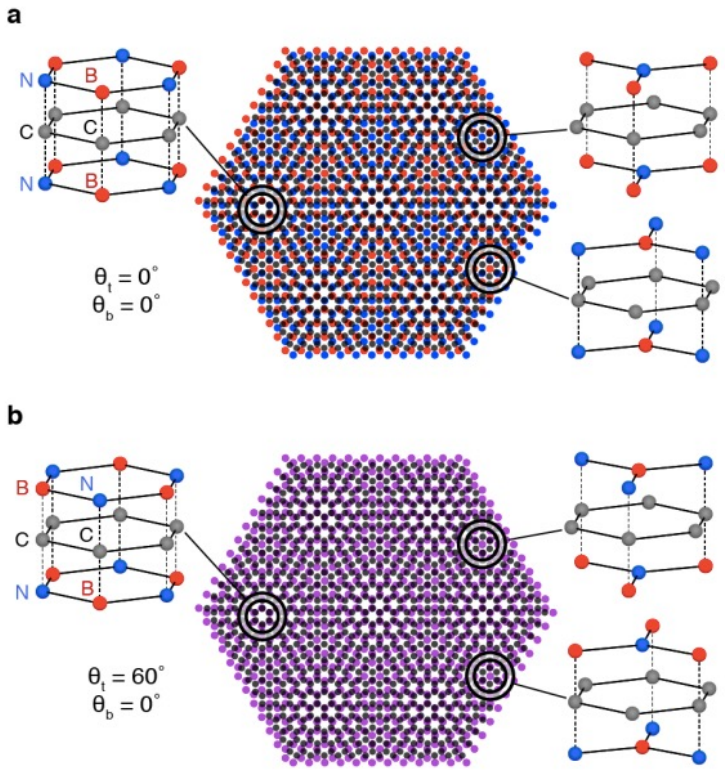
unrelaxed

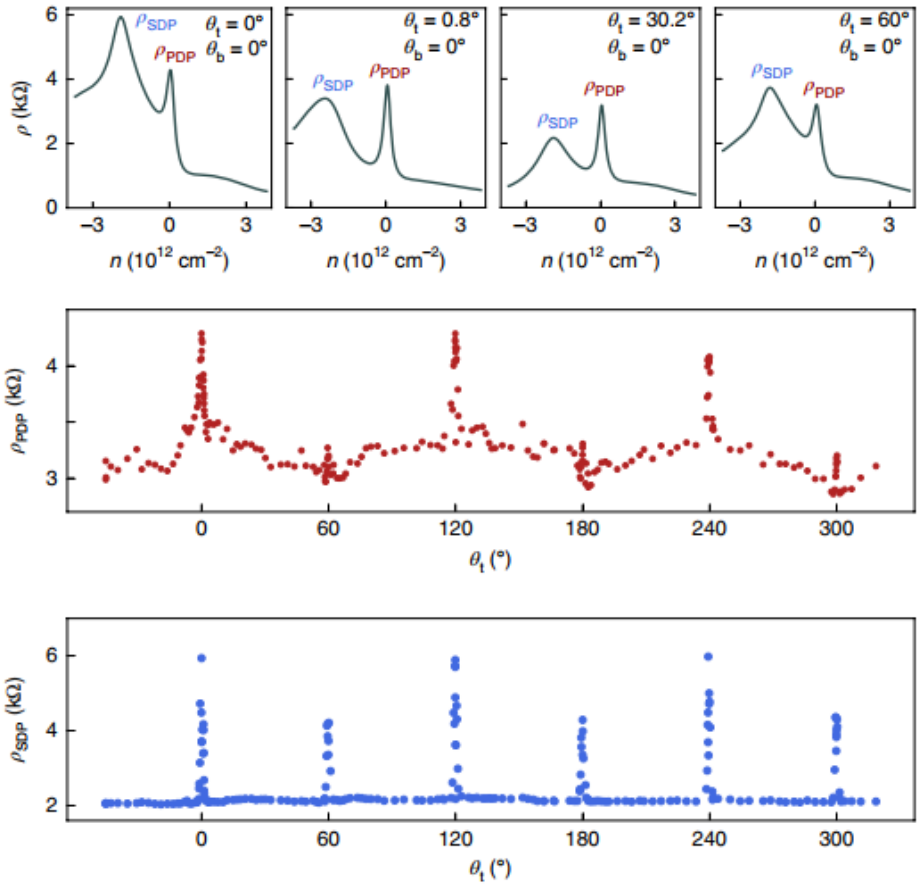


- Relaxation changes the interatomic registry, which results in the appearance of significant gaps at the primary and secondary DP
- Bottom hBN layer is kept rigid to mimic the effect of a thick substrate, while graphene and top hBN are freely relaxed

Rotations by 60°

- Rotations with 60° result in oscillating features of resistance at PDP and SDP

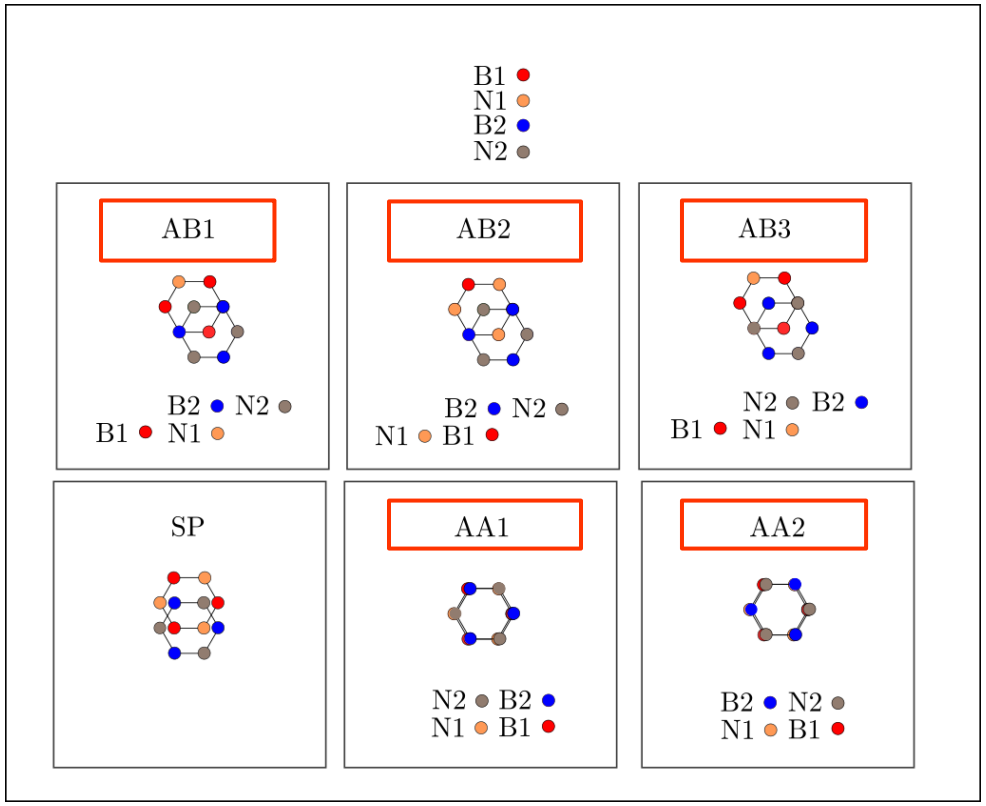


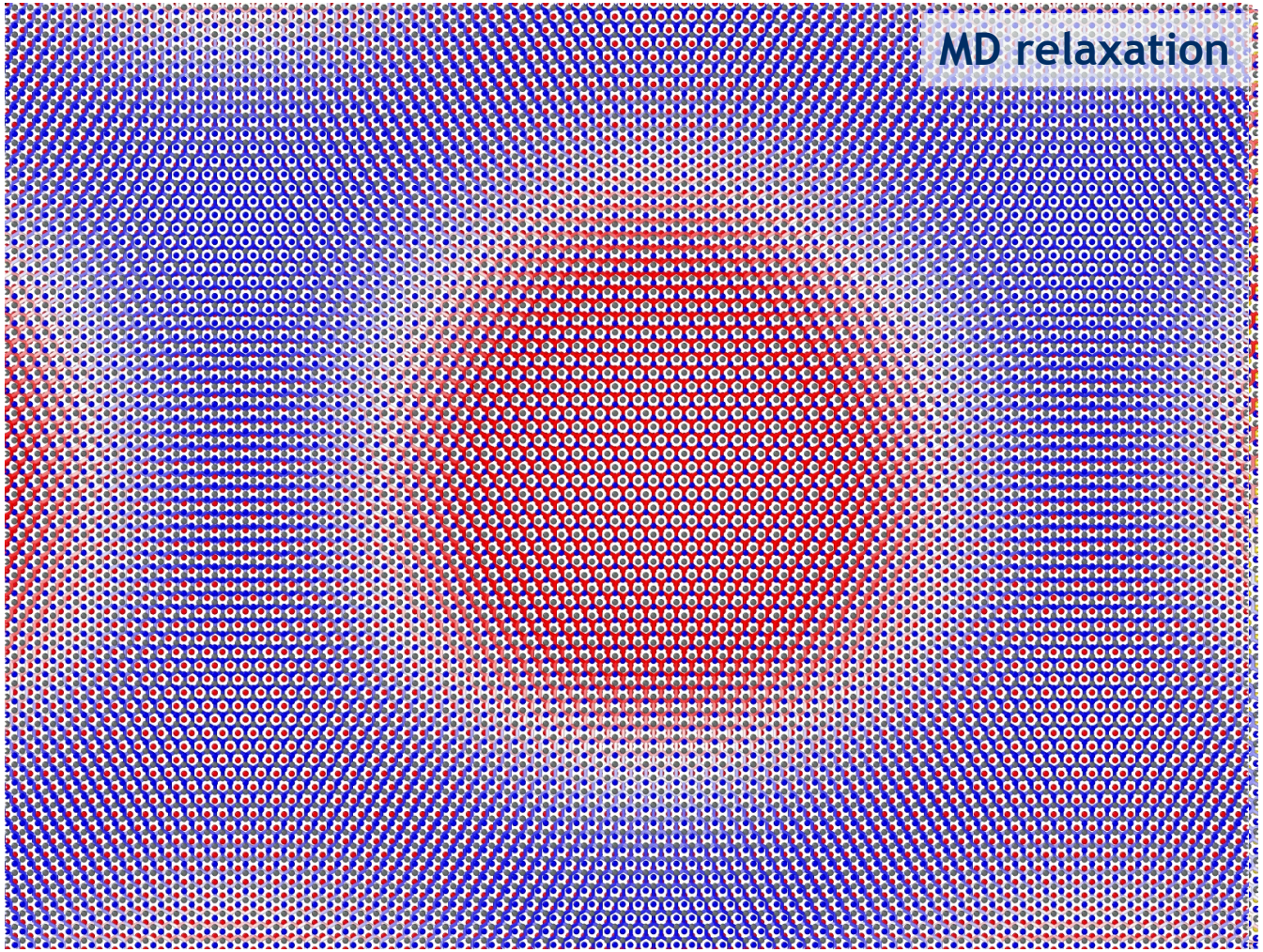


N. R. Finney *et al.*, Nat. Nanotech. 14, 1029 (2019)

Rotations by 60° (2 aligned moirés)

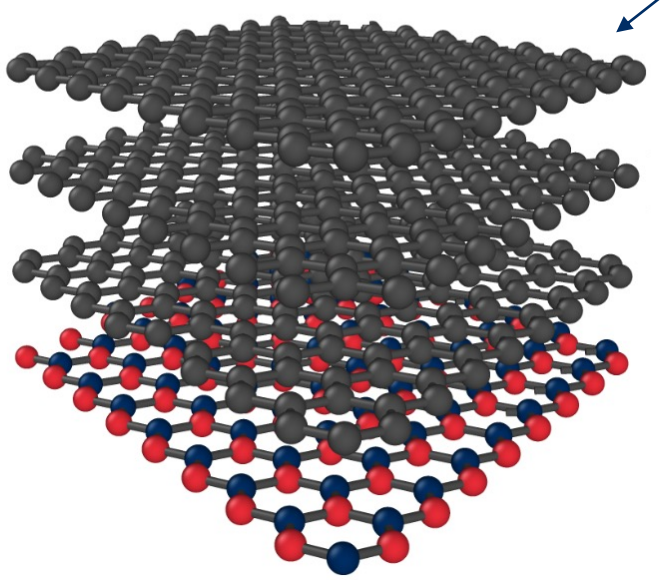
- Rotations with periods of 60° result in different stacking configurations





hBN encapsulated/aligned multilayer graphene

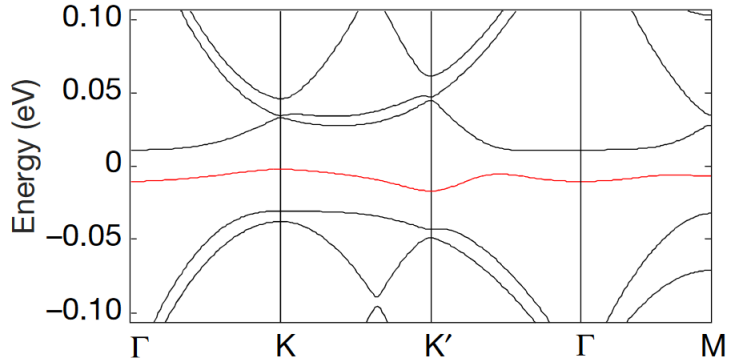
SC and correlated states in ABC trilayer graphene on hBN

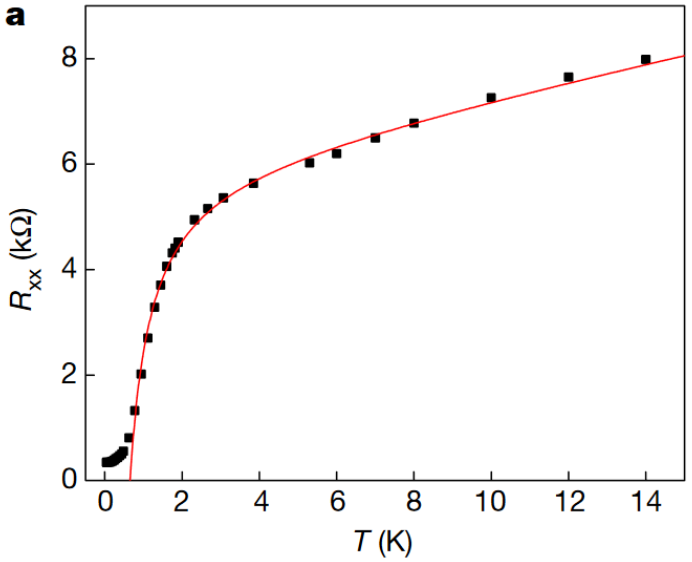


ABC-stacked trilayer graphene

+ applied vertical electric field

hBN





B. L. Chittari, G. Chen, Y. Zhang, F. Wang and J. Jung, *PRL* 122, 016401 (2019)

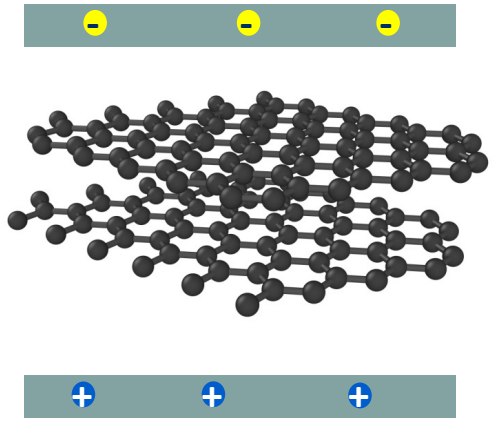
G.Chen et al, Nature Physics 15, 237 (2019)

G.Chen et al, Nature 572, 215 (2019)

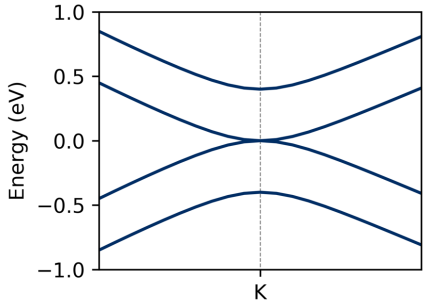
hBN supported (encapsulated) multilayer graphene

1

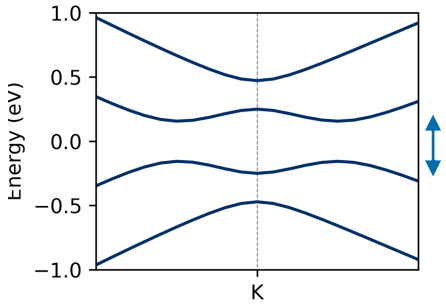
gating



normal



gated

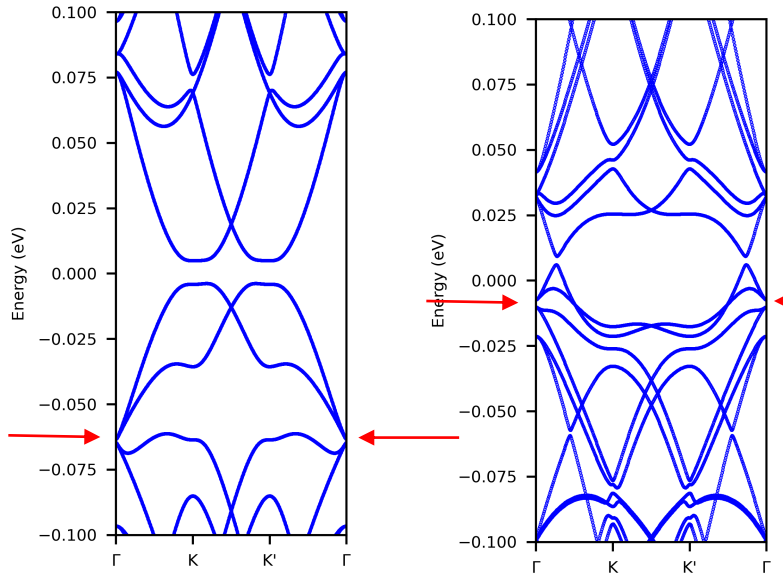


Bandgap

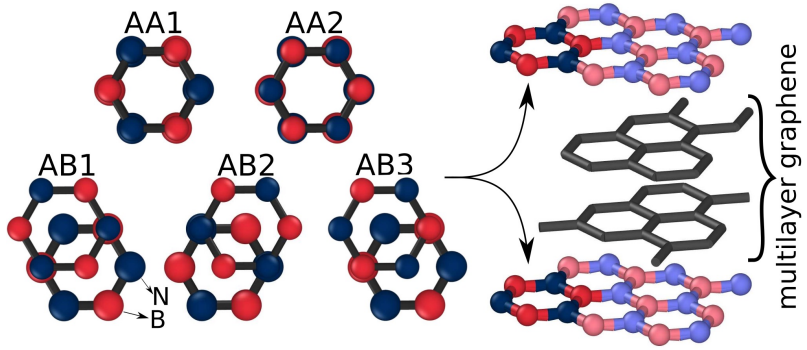
2

BLG/hBN

TLG/hBN



Can we engineer a gap at SDP through stacking manipulation?



NO TWISTING!

Induced effective potentials

$$(f_1(\mathbf{r}), f_2(\mathbf{r})) = \sum_m \left(1, i(-1)^{m-1}\right) \exp\left(i\mathbf{G}_m \cdot \mathbf{r}\right)$$

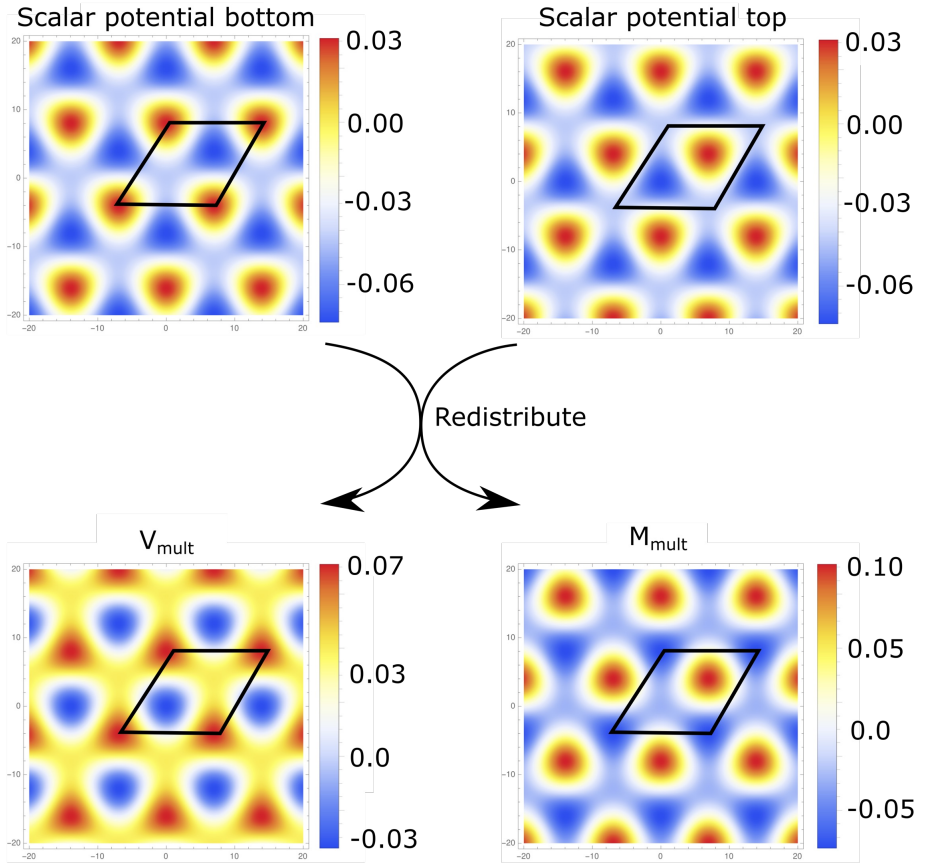
Single layer: $U_{eff}(\mathbf{r}) = vG(u_0 f_1 + \tilde{u}_0 f_2)\sigma_0 + \zeta vG(u_3 f_1 + \tilde{u}_3 f_2)\sigma_3$

$$+ \zeta v \left[\hat{\mathbf{z}} \times \nabla(u_1 f_2 + \tilde{u}_1 f_1)\right].\sigma,$$

Low energy sites in top and bottom layer

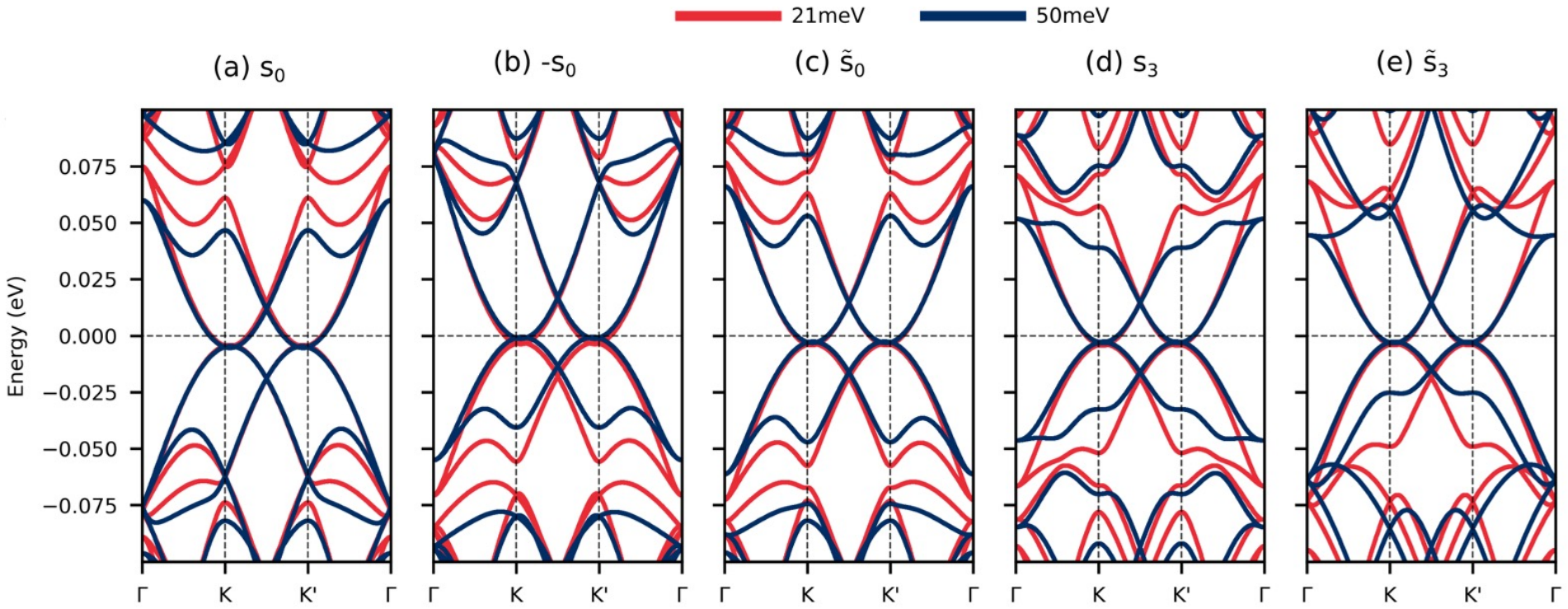
$$\tilde{\mathcal{H}} = \mathcal{H}_{mult} + \begin{bmatrix} V_t(\mathbf{r}) + M_t(\mathbf{r}) & 0 \\ 0 & V_b(\mathbf{r}) - M_b(\mathbf{r}) \end{bmatrix}$$

$$V_{mult}(\mathbf{r}) = V_0\sigma_0 + v_{mult}G(s_0 f_1 + \tilde{s}_0 f_2)\sigma_0$$
$$M_{mult}(\mathbf{r}) = M_0\sigma_3 + v_{mult}G(s_3 f_1 + \tilde{s}_3 f_2)\sigma_3$$



Effect of potentials on bandstructure

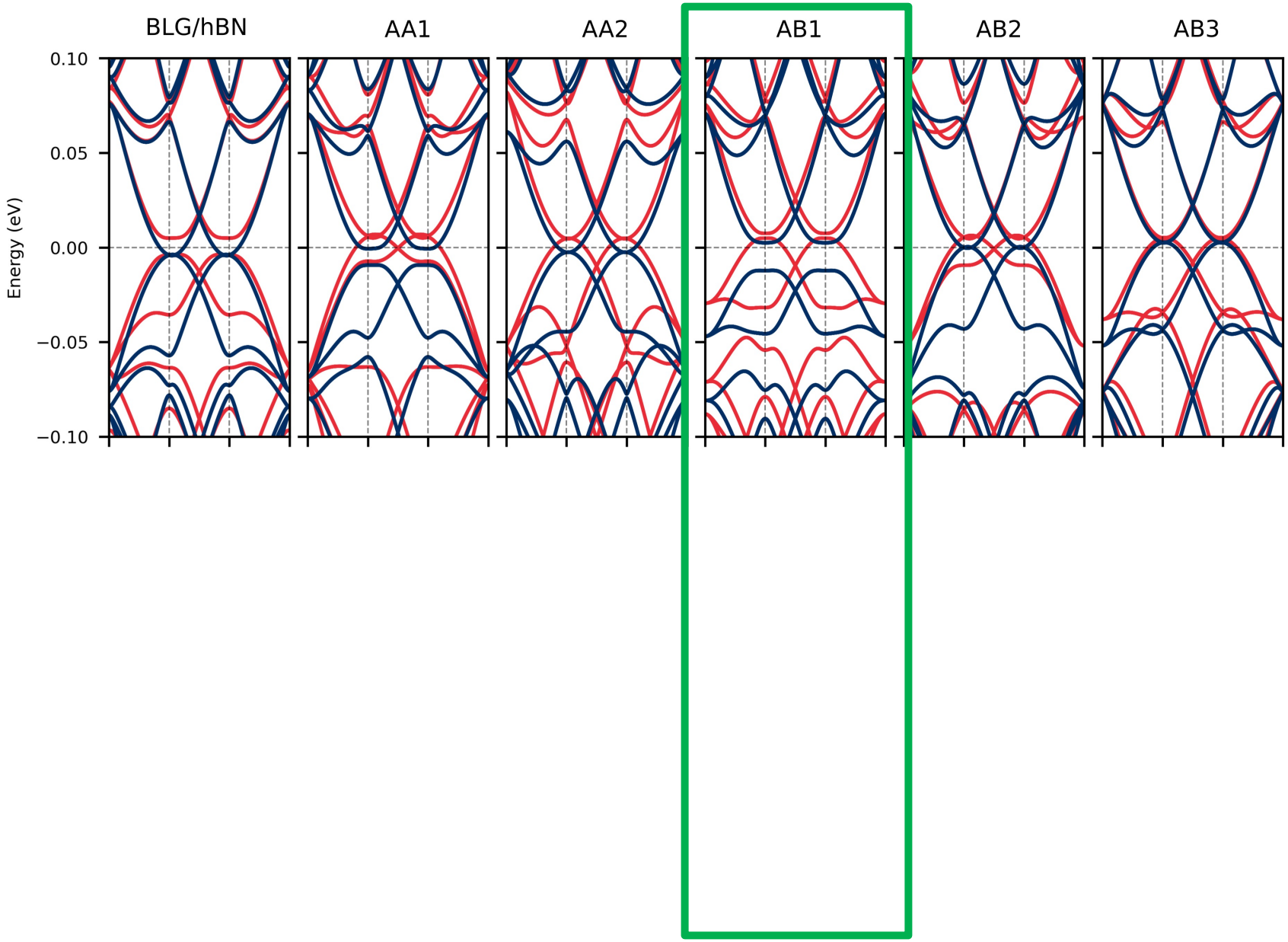
- tight-binding simulation with independent contributions of effective potentials



Full atomistic TB simulation (encapsulated AB bilayer)

No gating

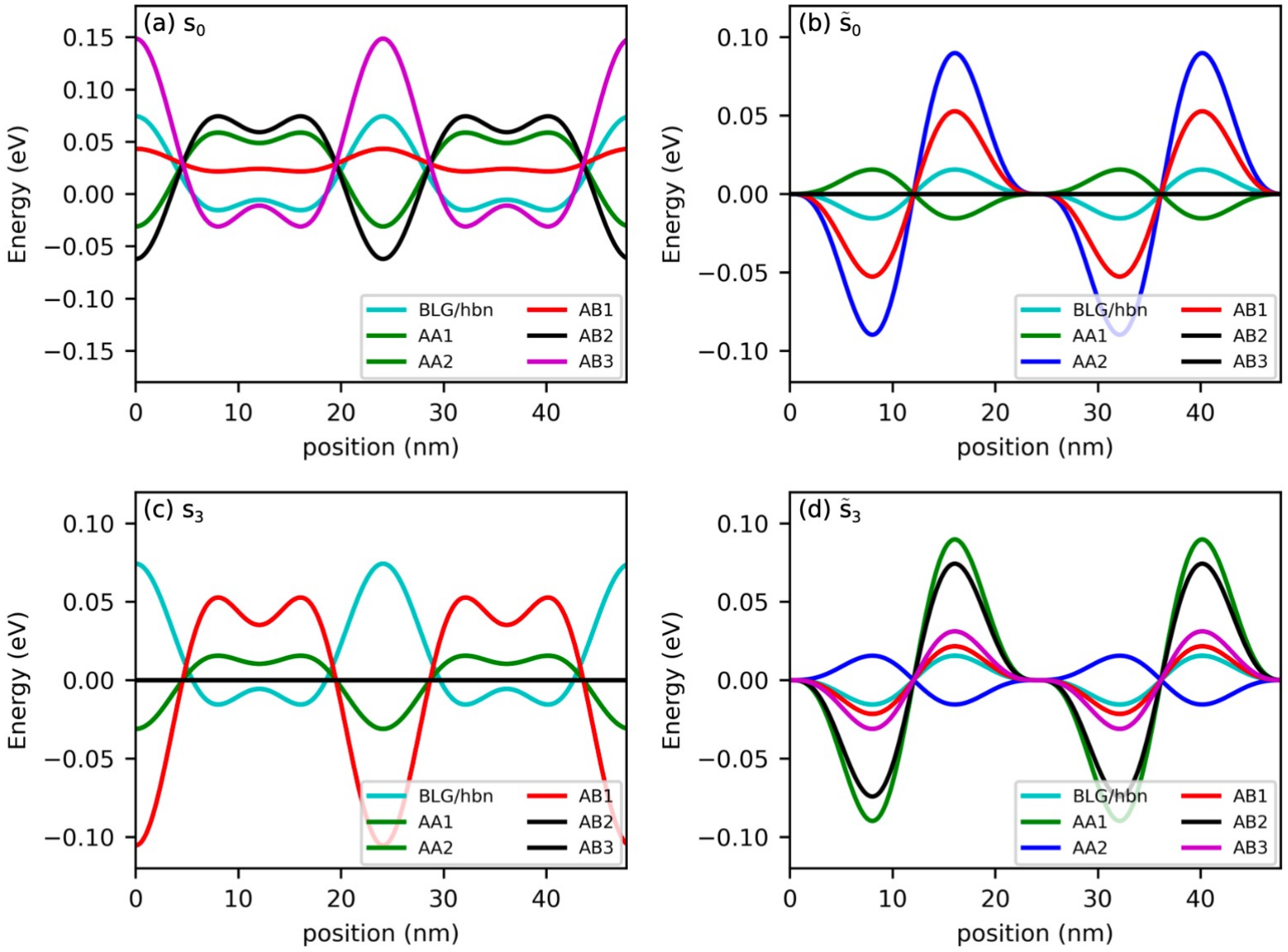
E=100mV/nm



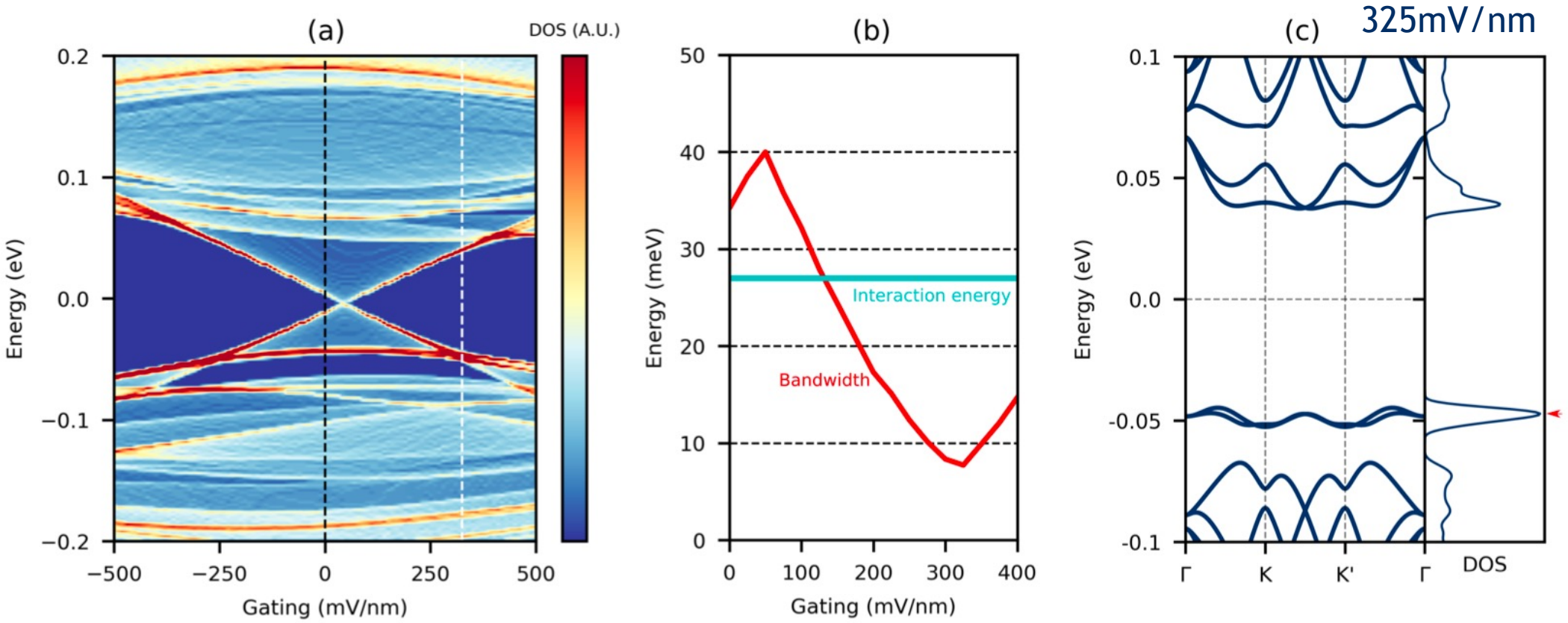
Relaxed

Unrelaxed

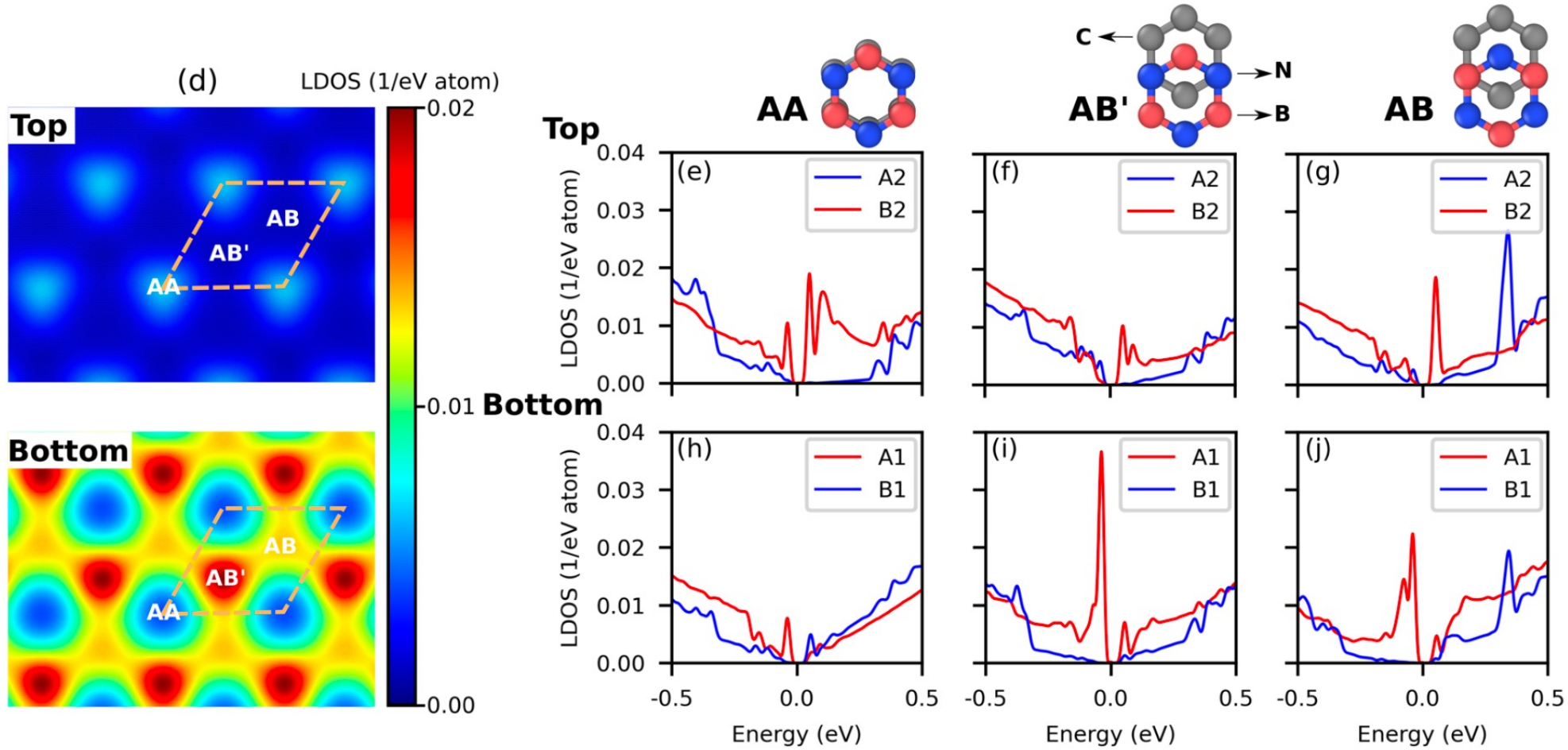
Effective potentials across the moiré unit cell



Electric field dependence of AB1 configuration



Localization of the wavefunction in the moiré



Conclusions

- Large scale tight-binding simulations are possible (KITE, Pybinding + other software)

- An additional aligned layer leads to a further reconstruction of the spectrum at all energies (esp. low energies) → supermoiré of arbitrary wavelength
- Further enhancement of scattering through relaxation (imprinting), leads to the appearance of significant gaps at all DPs
- hBN encapsulated + aligned multilayer graphene can provide further knobs for tuning DP gaps
- Further gating can lead to flat moire bands