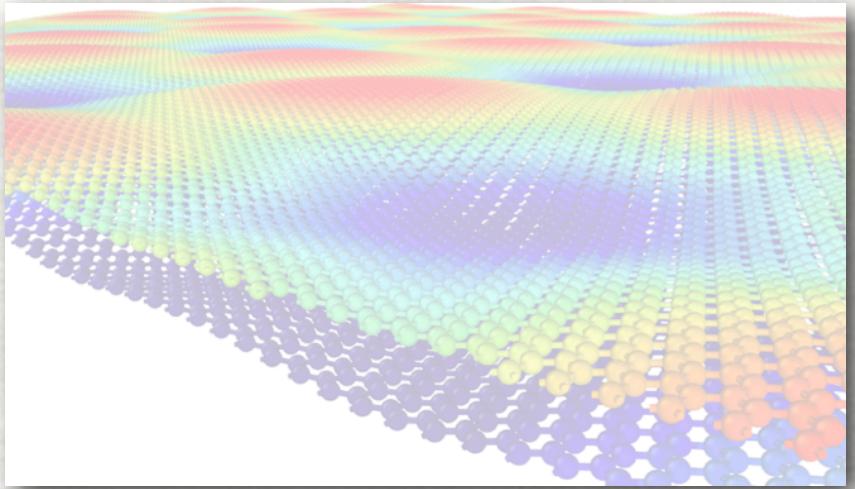
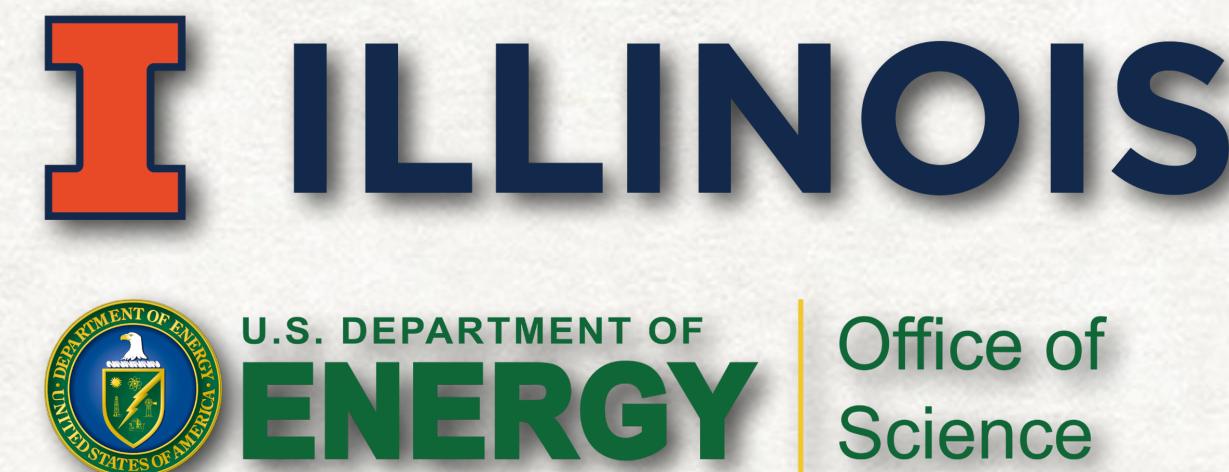


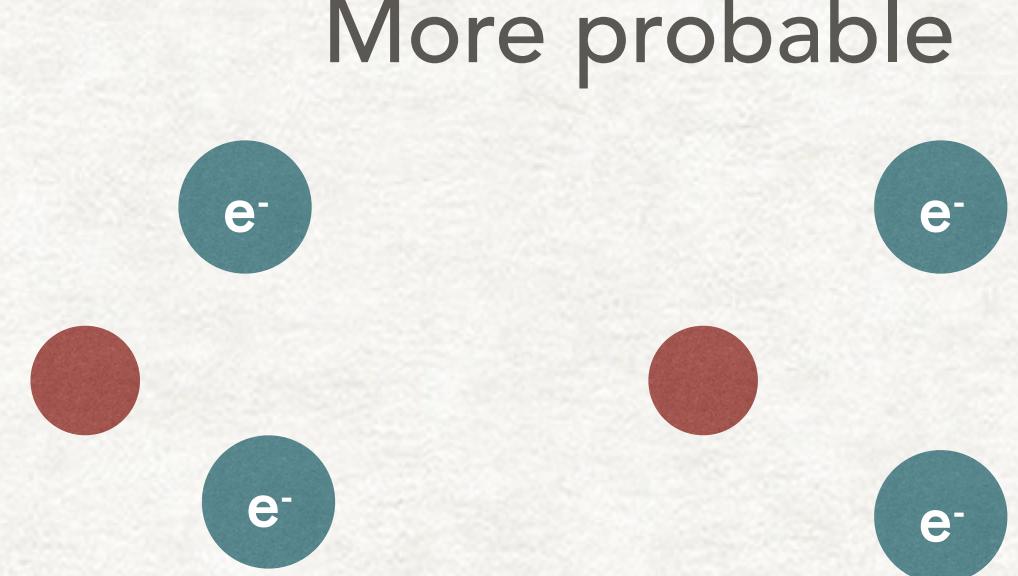
# First Principles Quantum Monte Carlo Results as a Data Source for Larger Scale Models



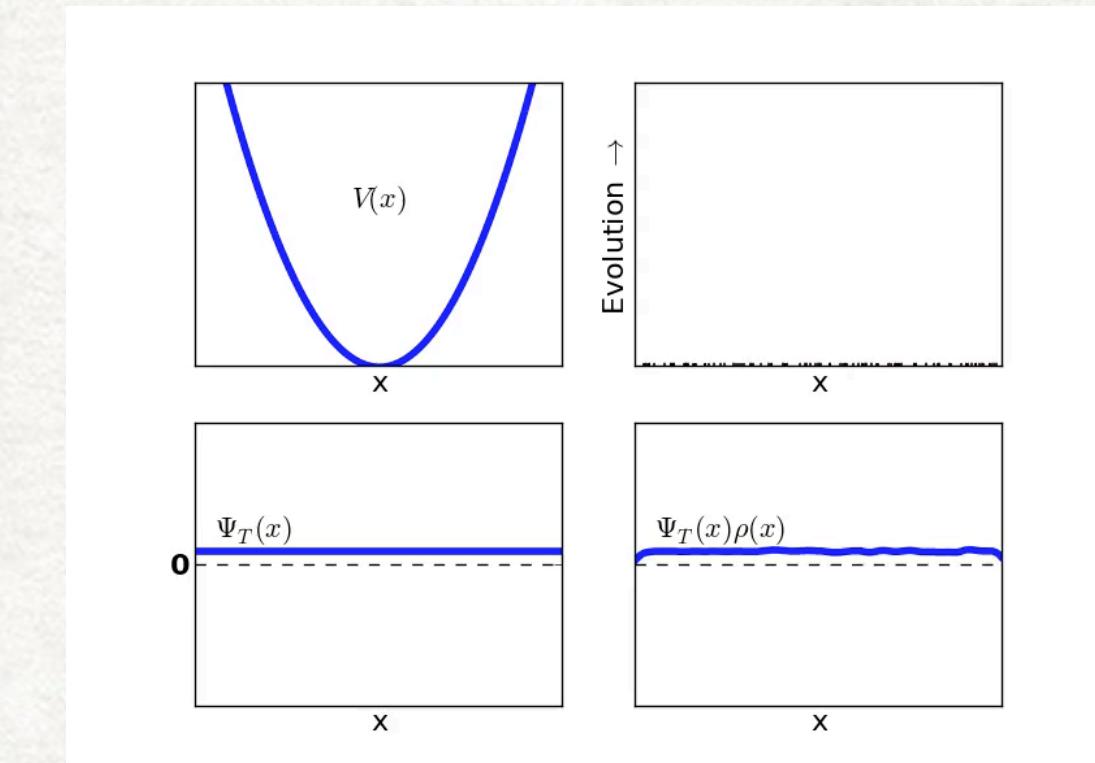
Lucas K. Wagner  
University of Illinois at Urbana-Champaign



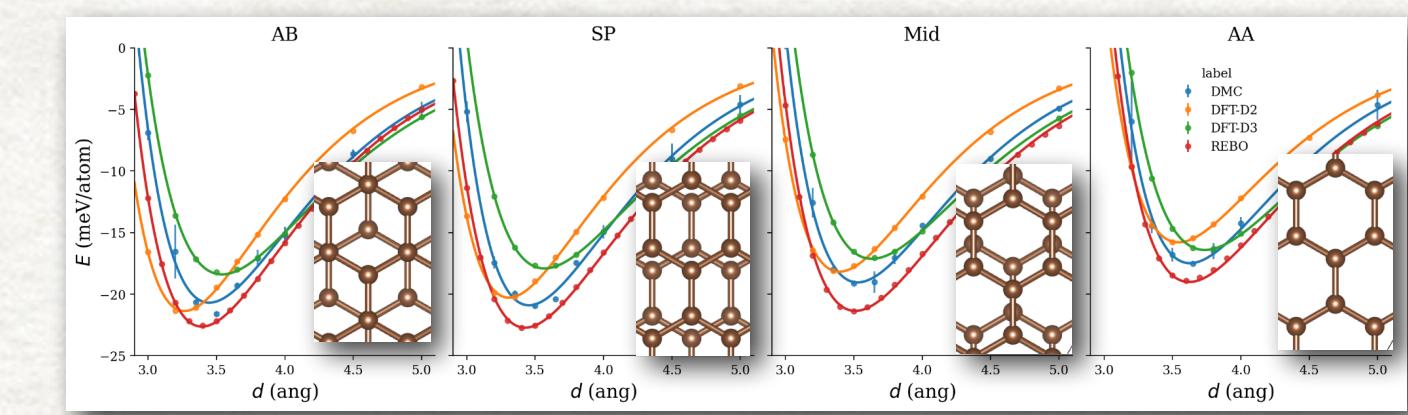
Twisted bilayer  
graphene exhibits a  
rich phase diagram  
(micron scale)



Electron correlation  
induced interaction  
between the layers  
(atomic scale)



Quantum Monte Carlo  
methods describe the  
correlation very  
accurately, but is very  
expensive  
(sub-atomic scale)



Quantum Monte Carlo  
data on which we fit  
micron-scale models.

# Multiscale Modeling

△ Accuracy ▾

## Quantum Monte Carlo

- Explicit correlation
- High accuracy
- Large computational cost

## Density Functional Theory

- Implicit correlation
- Lower accuracy
- Lower cost

## Standard approach

Maximum time/length scales ▷

## QMC-HAMM approach

### QMC-ML models

- Quantum lattice models (tight binding)
- Atomistic potentials

### DFT-ML

- Quantum lattice models (tight binding)
- Atomistic potentials

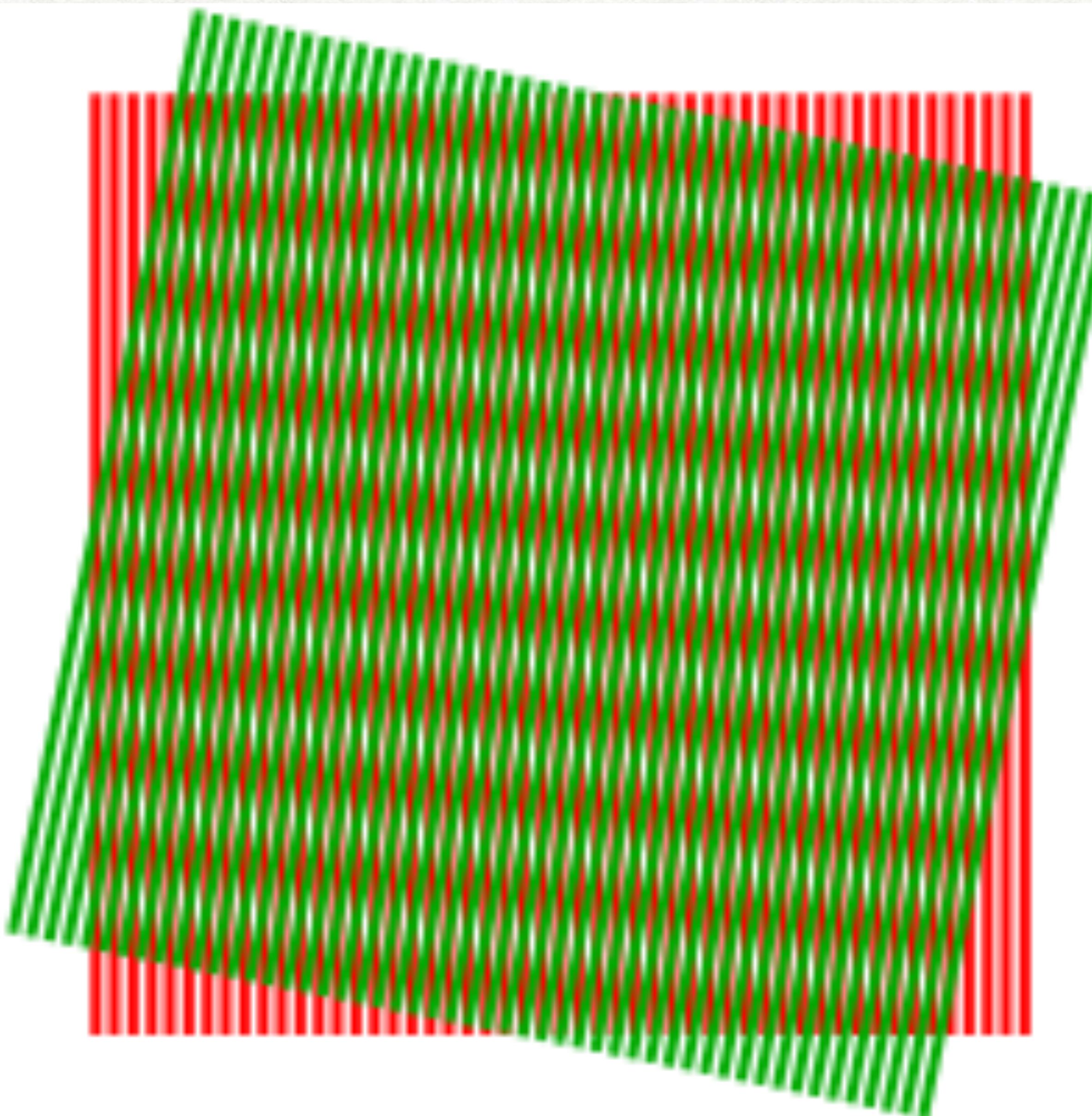
### QMC Mesoscale

- Phase diagrams
- Continuum models
- $k \cdot p$  models

### DFT mesoscale

- Phase diagrams
- Continuum models
- $k \cdot p$  models

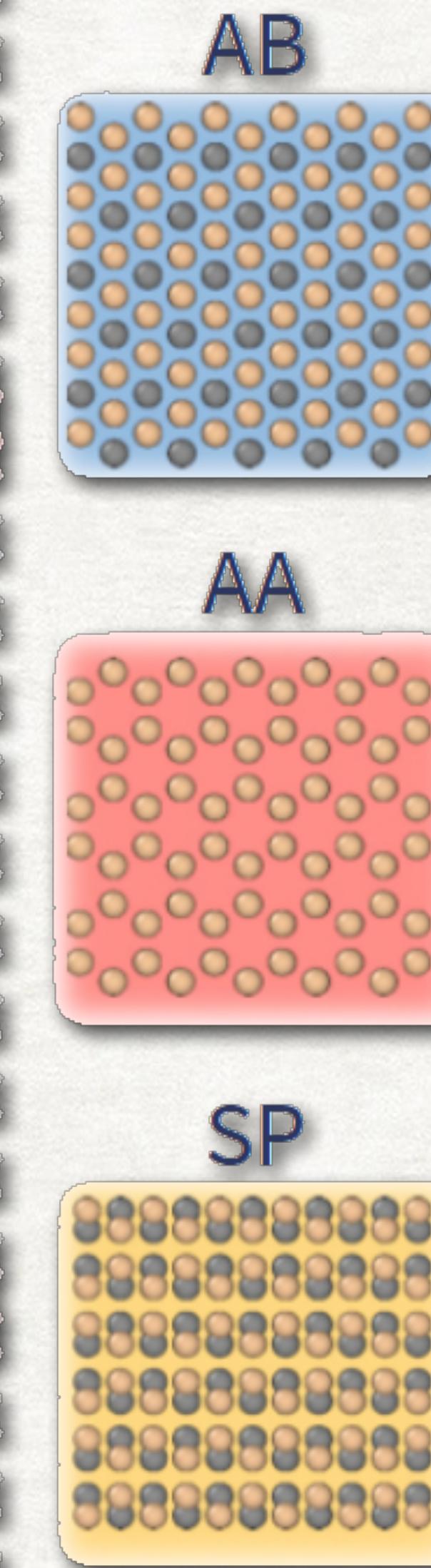
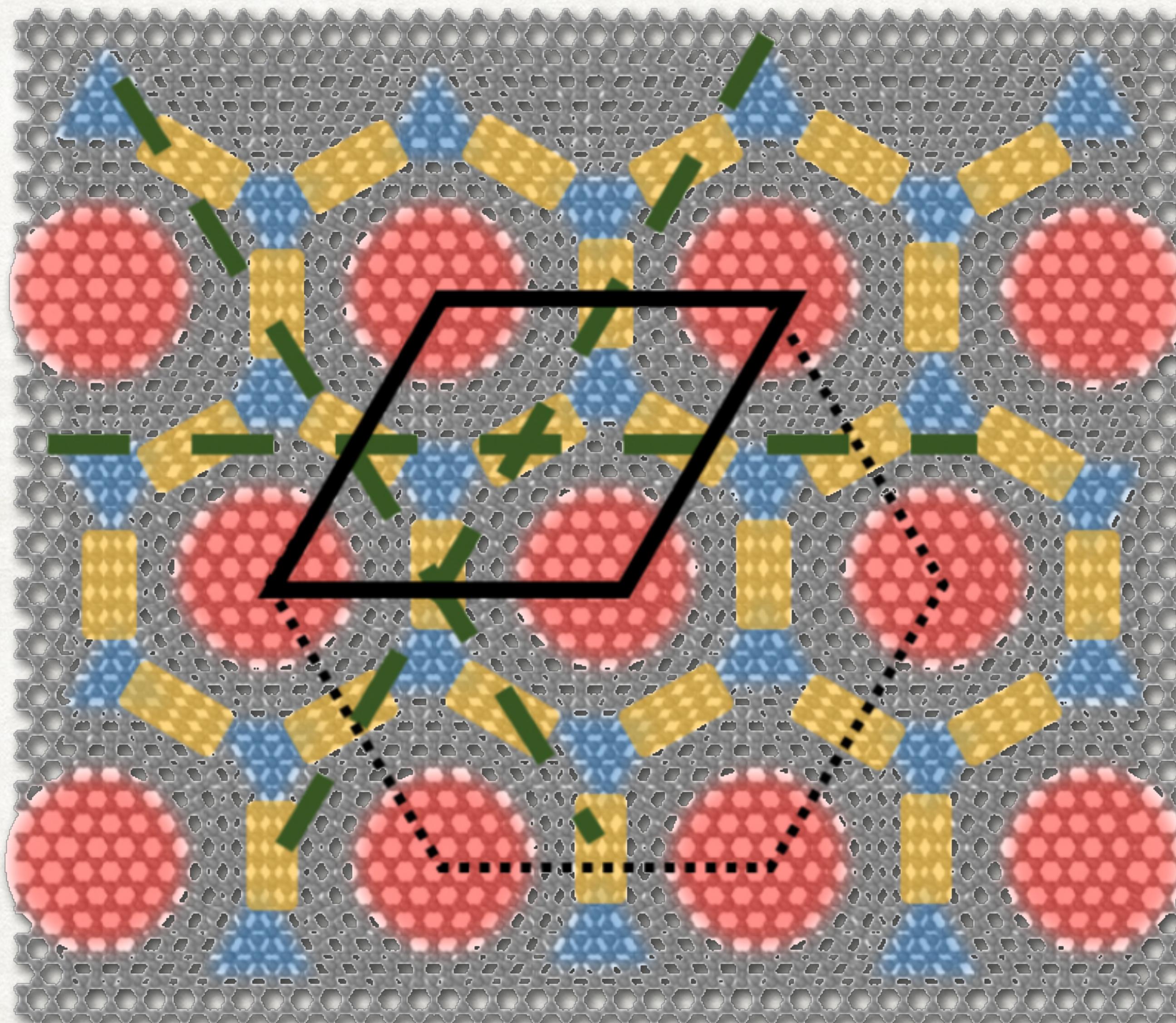
# Moire Patterns



Two regular lattices placed at an angle.

New longer length scale patterns emerge.

# Twisted Bilayer Graphene

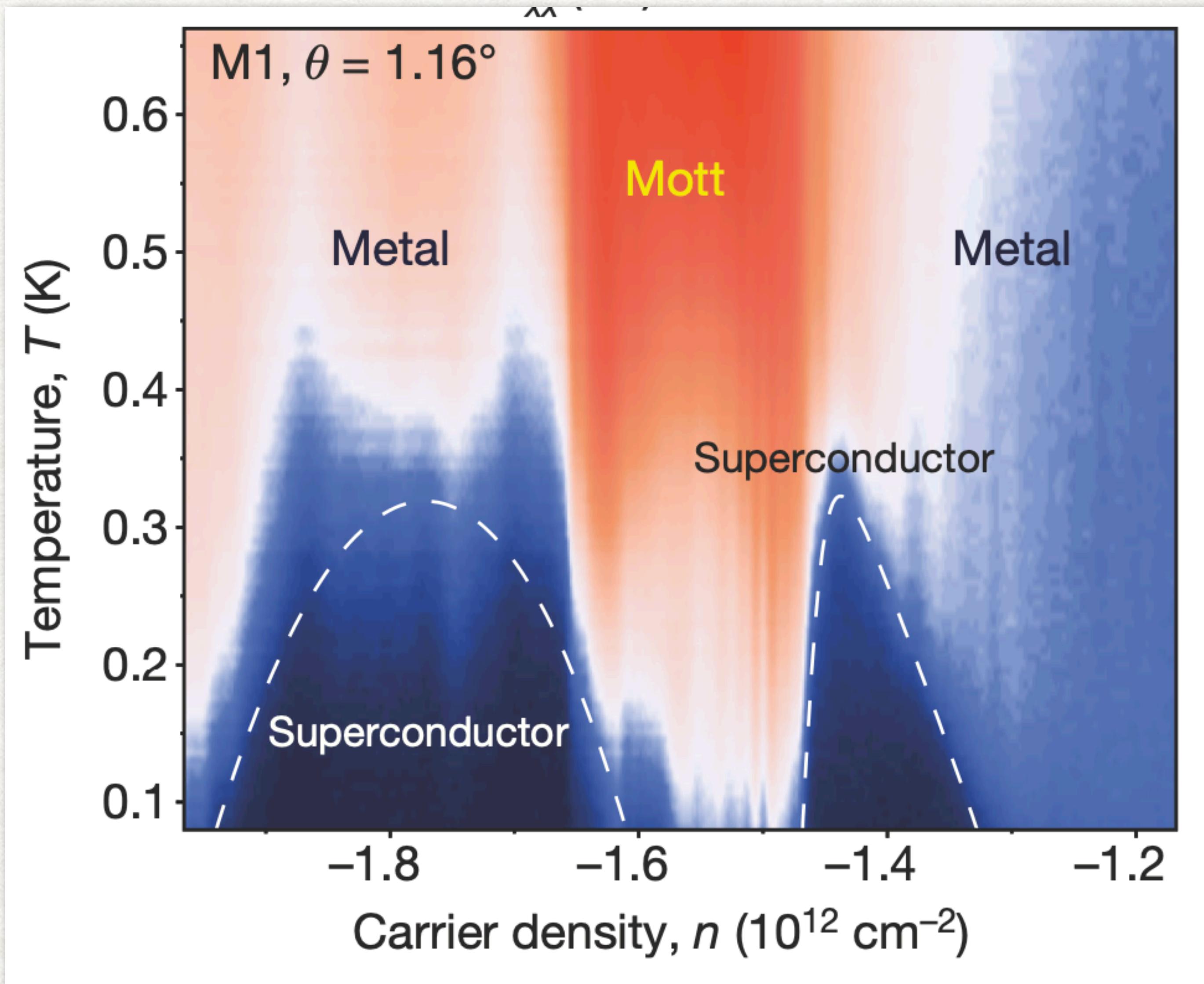


Two layers of graphene, one rotated above the other.

The twist creates Moire patterns.

Some regions are AB, some are AA, and some are in between (SP)

# Rich Phase Diagram From Twisting



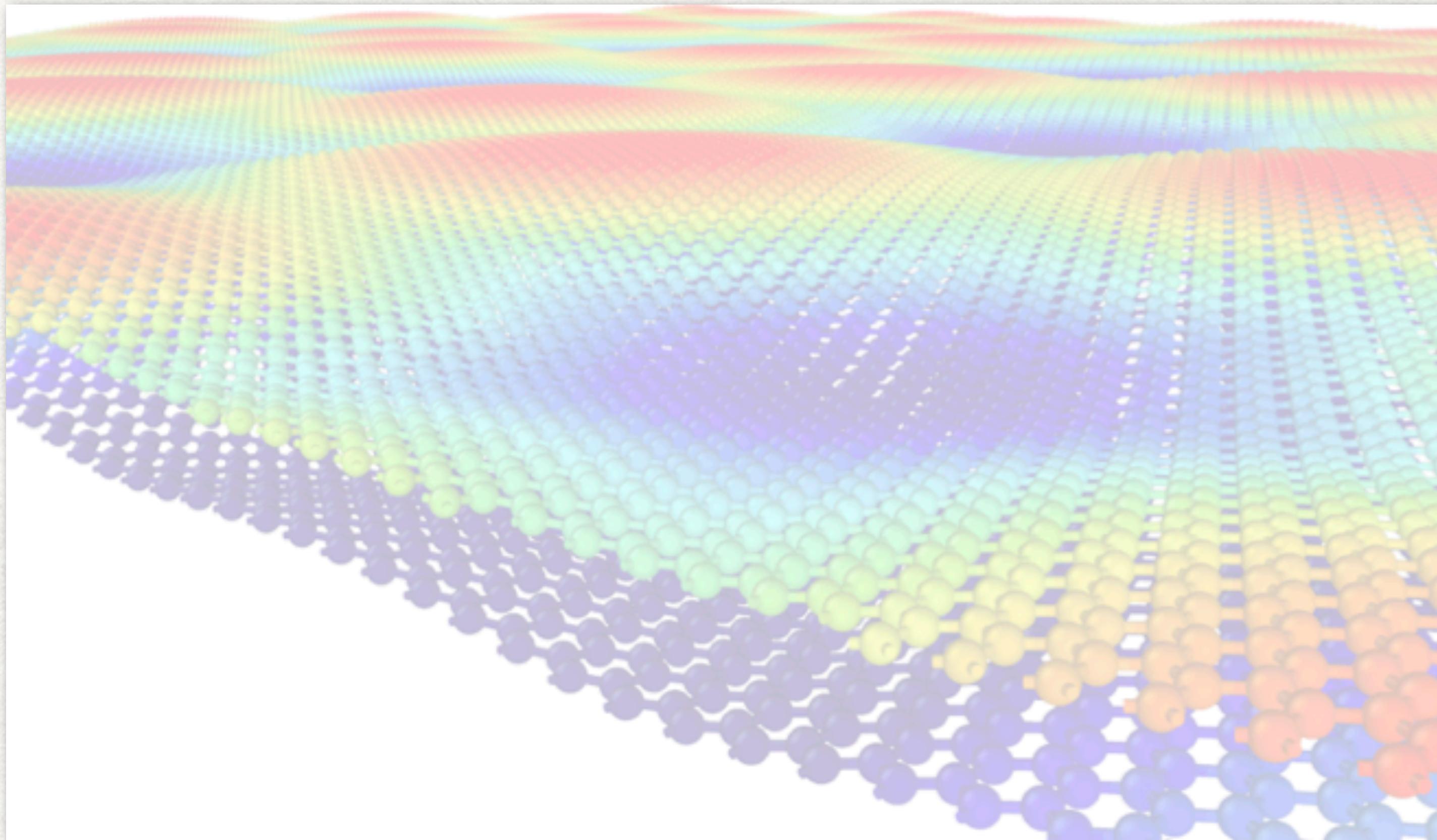
Sheets are angled at  $1.16^\circ$ .

Gating used to control electron density.

Induced superconductivity and correlated insulator phases.

Cao, Fatemi, Fang, Watanabe, Taniguchi, Kaxiras, Jarillo-Herrero. Nature 556: 43

# Focus of Today: Interlayer Interaction in Graphene Bilayers

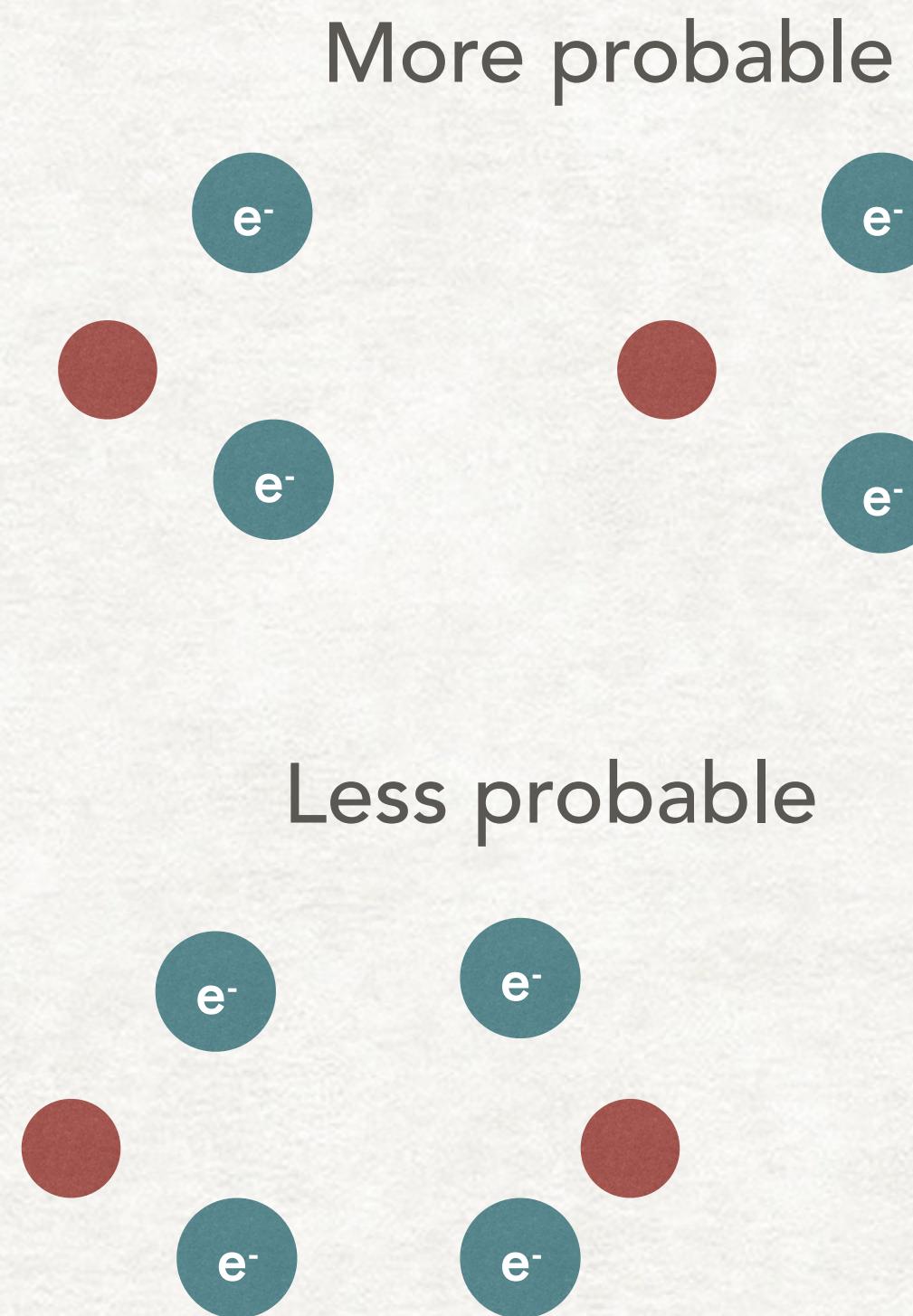


AA stacking: pushed apart  
AB stacking: pulled closer

-> corrugations

The corrugations then affect the electrons (another QMC-HAMM topic).

# Computing Van Der Waals (London Dispersion) Interactions



Two He atoms (4 electrons).

Probability density

$$\rho(r_1, r_2, r_3, r_4) = |\Psi(r_1, r_2, r_3, r_4)|^2$$

In the ground state, dipoles line up.

Attraction due to many-particle correlations!

# Variational Monte Carlo

$$\frac{\langle \Psi | \mathcal{O} | \Psi \rangle}{\langle \Psi | \Psi \rangle} = \frac{\int \langle \Psi | R' \rangle \langle R' | \mathcal{O} | R \rangle \langle R | \Psi \rangle dR dR'}{\int |\langle R | \Psi \rangle|^2 dR}$$

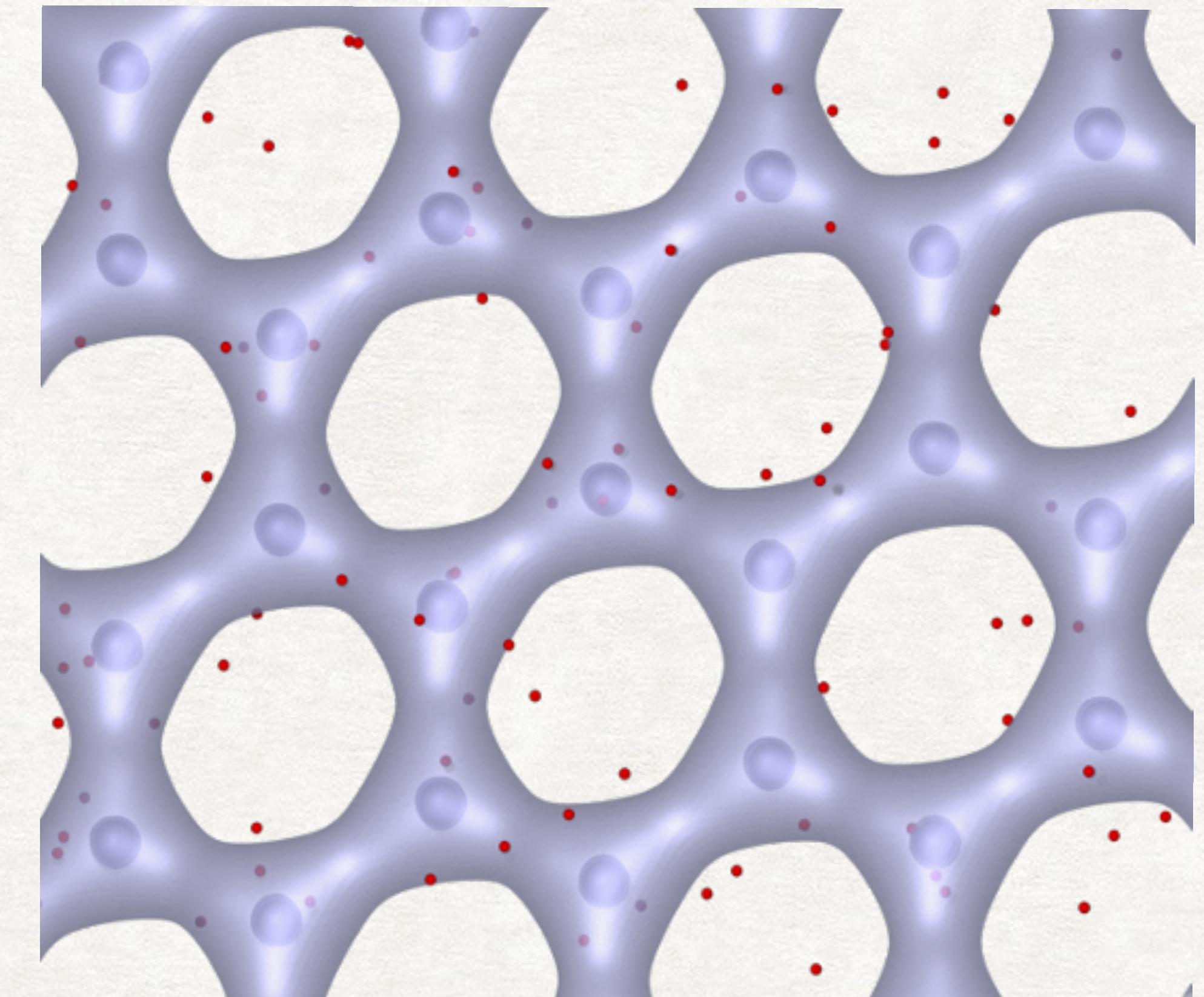
If  $\mathcal{O}$  is semi-local (doesn't need to be!):

$$\int \frac{\langle R | \mathcal{O} | \Psi \rangle}{\langle R | \Psi \rangle} \frac{|\langle R | \Psi \rangle|^2}{\int |\langle R | \Psi \rangle|^2 dR} dR$$

$\mathcal{O}_L(R)$                      $\rho(R)$

$$\langle \mathcal{O}_L(R) \rangle_{R \sim \rho}$$

A method for evaluating many-body integrals



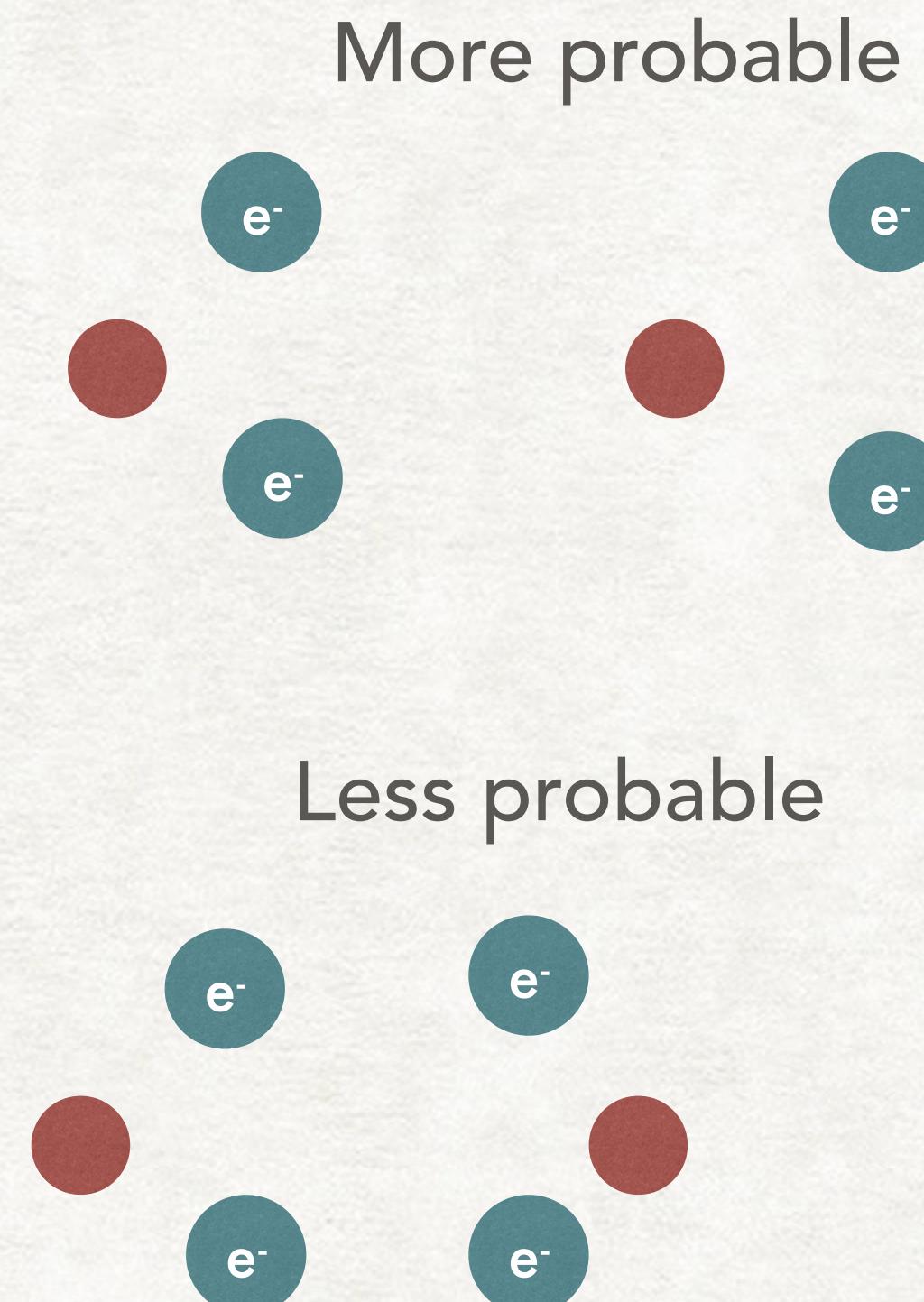
Sample **electronic coordinates**,  
average local energy

# The Slater-Jastrow Wave Function

$$\Psi(r_1, r_2, r_3, \dots) = \exp(U) \text{Det}^{\uparrow} \text{Det}^{\downarrow}$$

$$U = \sum_{ij} f(r_i, r_j, r_i - r_j)$$

Encodes the correlation



# Fixed Node Diffusion Monte Carlo

$$\lim_{\tau \rightarrow \infty} e^{-H\tau} |\Psi\rangle = |\Phi_0\rangle$$

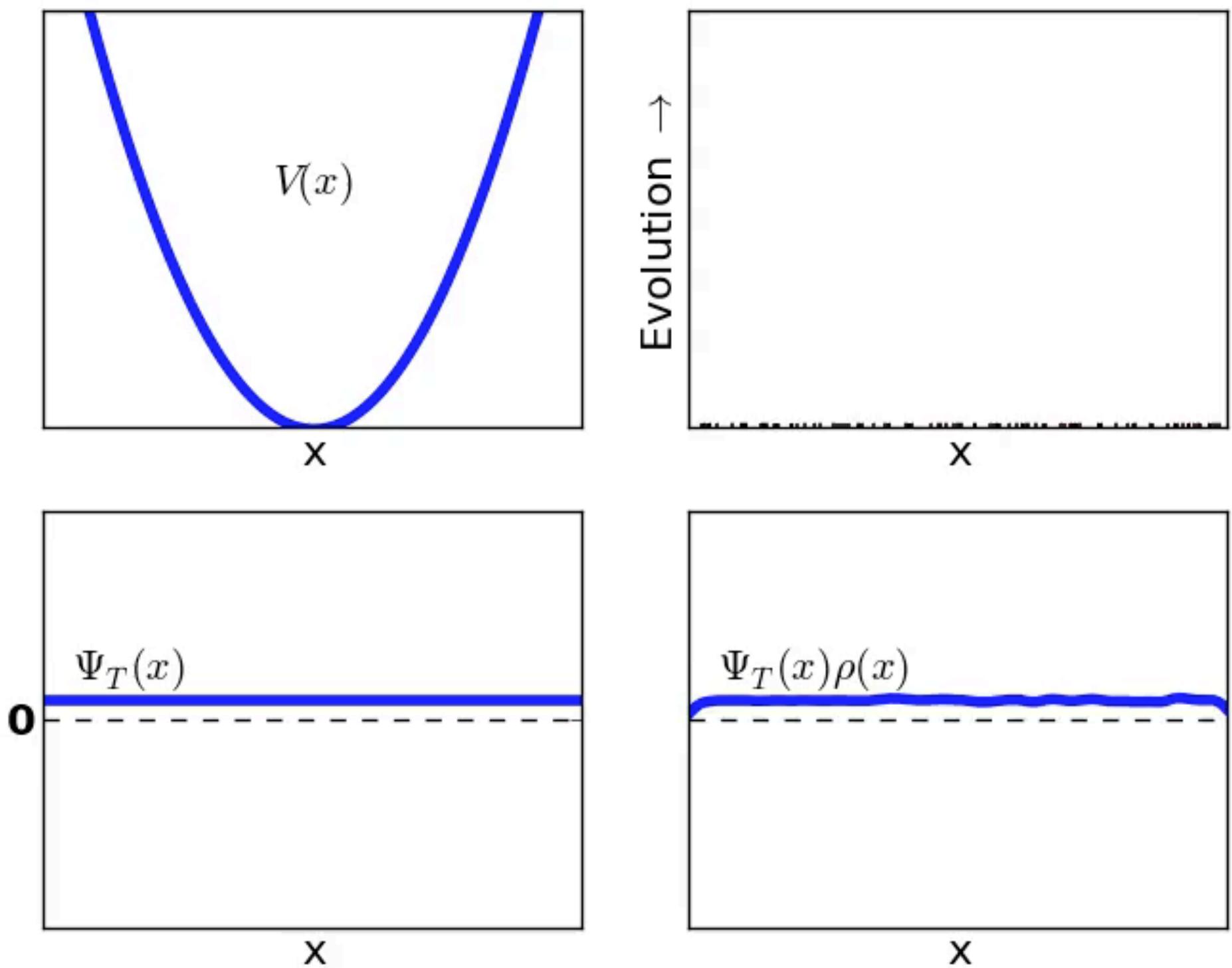
$$E_0 = \frac{\langle \Psi | e^{-H\tau} \mathbf{H} | \Psi \rangle}{\langle \Psi | e^{-H\tau} | \Psi \rangle} = \frac{\int \langle \Psi | R \rangle \langle R | e^{-H\tau} | R' \rangle \langle R' | \mathbf{H} | \Psi \rangle dR dR'}{\int \langle \Psi | R \rangle \langle R | e^{-H\tau} | R' \rangle \langle R' | \Psi \rangle dR dR'}$$

Sign problem: When  $\tau$  is large,  $\langle \Psi | R \rangle$  is uncorrelated with  $\langle R' | \Psi \rangle$  and the ratio goes to 0/0.

Fixed node: Set

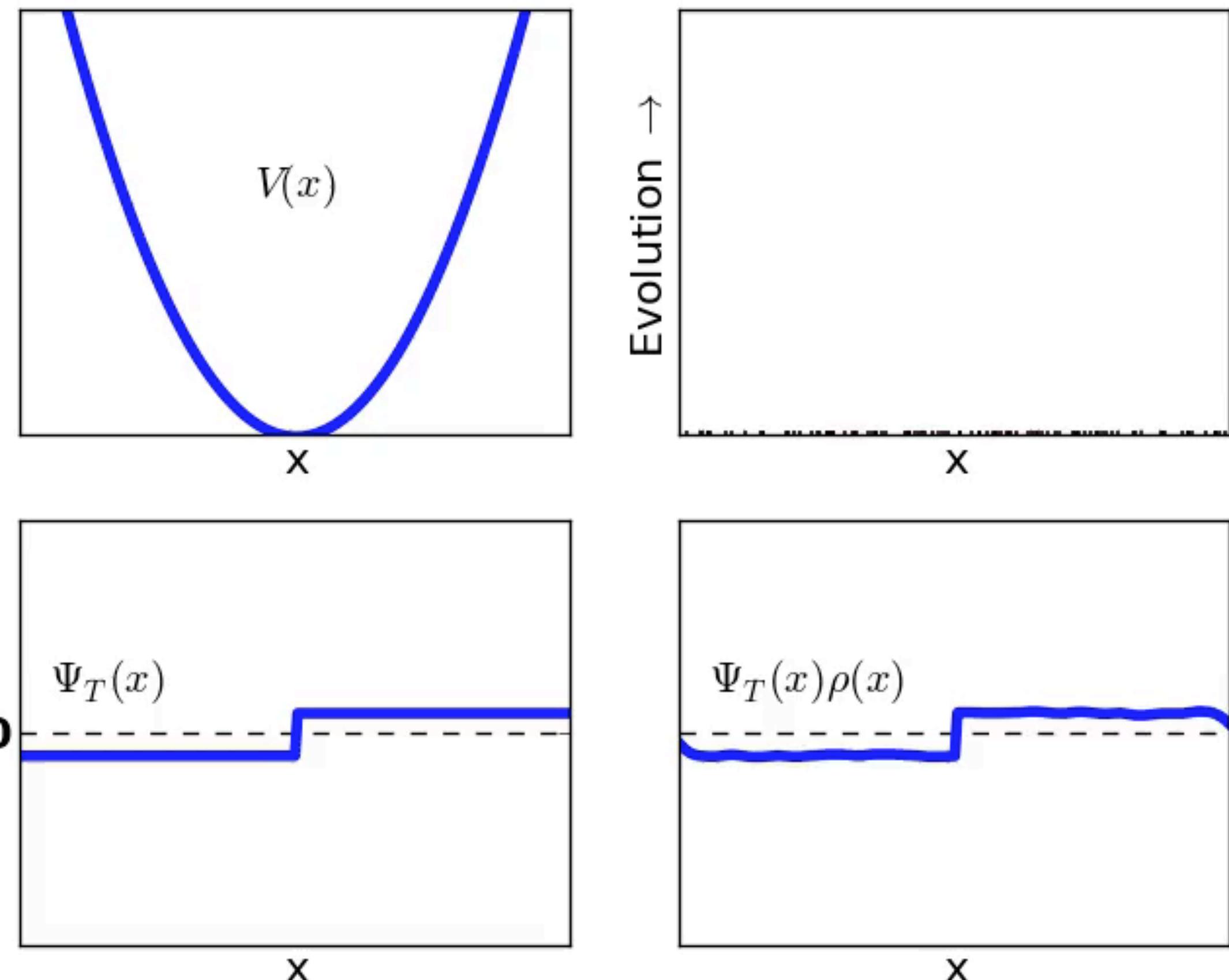
$$\langle R | e^{-H_{FN}\tau} | R' \rangle = \begin{cases} \langle R | e^{-H\tau} | R' \rangle & \text{if } \langle R | e^{-H_{FN}\tau} | R' \rangle \geq 0 \\ 0 & \text{otherwise} \end{cases}$$

Path integral!



# Fixed Node DMC Properties

- 1) Energy is an upper bound to the exact ground state energy.
- 2) If the wave function nodes correspond to the ground state nodes, then will obtain the exact ground state energy.
- 3) If the wave function nodes correspond to an excited state, then under some conditions you obtain the exact excited state.



High accuracy for non-covalent interactions  
Dubecky, Mitas, Jurecka  
Chemical reviews 116, 5188

Ceperley and Alder. J. Chem. Phys. 81 5833 (1984)  
Foulkes, Hood, and Needs. PRB 60 4558 (1999)

Accuracy △

### Quantum Monte Carlo

- Explicit correlation
- High accuracy
- Large computational cost

### **QMC-HAMM approach**

#### QMC-ML models

- Quantum lattice models (tight binding)
- Atomistic potentials

#### QMC Mesoscale

- Phase diagrams
- Continuum models
- $k \cdot p$  models

### Density Functional Theory

- Implicit correlation
- Lower accuracy
- Lower cost

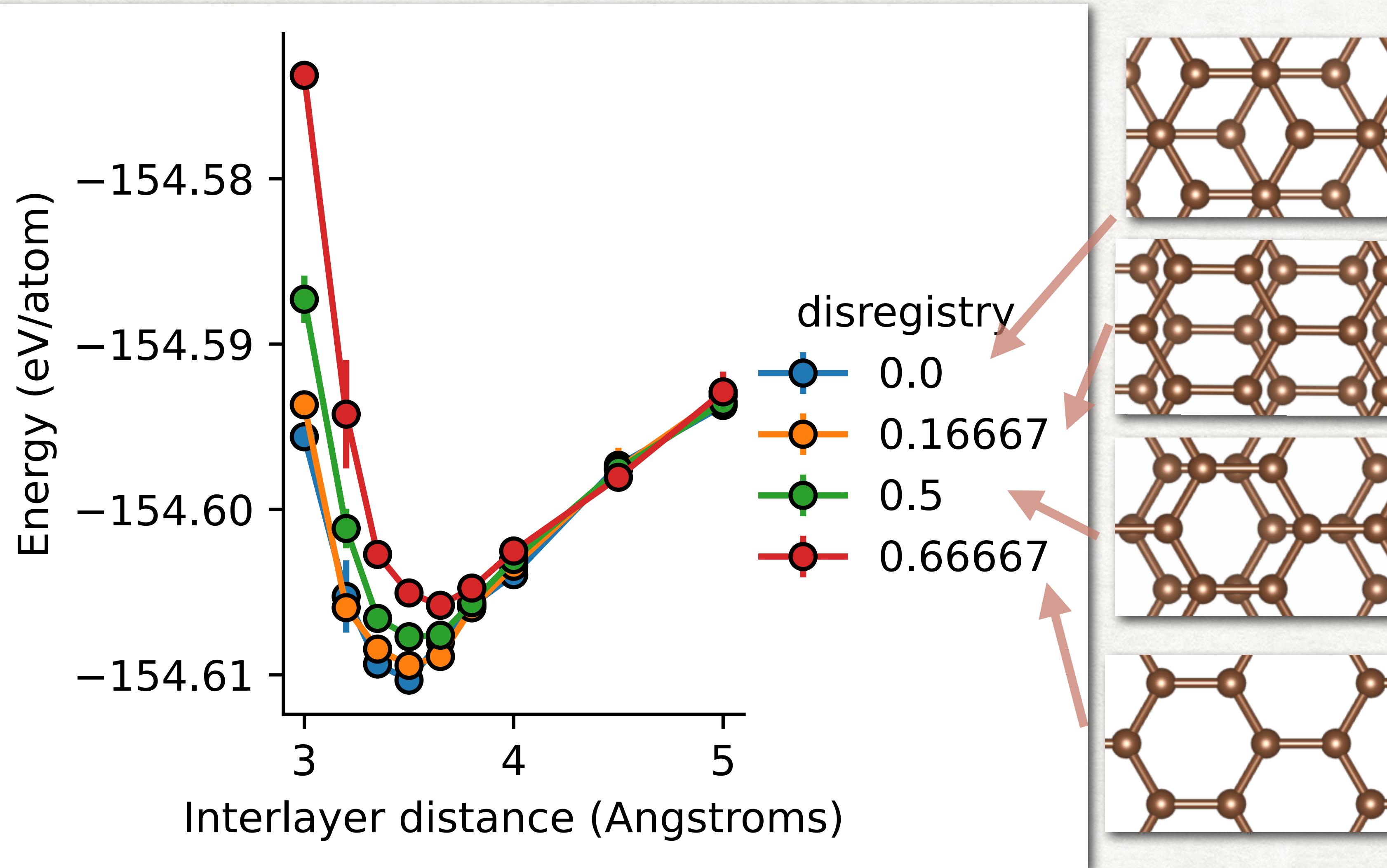
#### DFT-ML

- Quantum lattice models (tight binding)
- Atomistic potentials

### **Standard approach**

Maximum time/length scales ▷

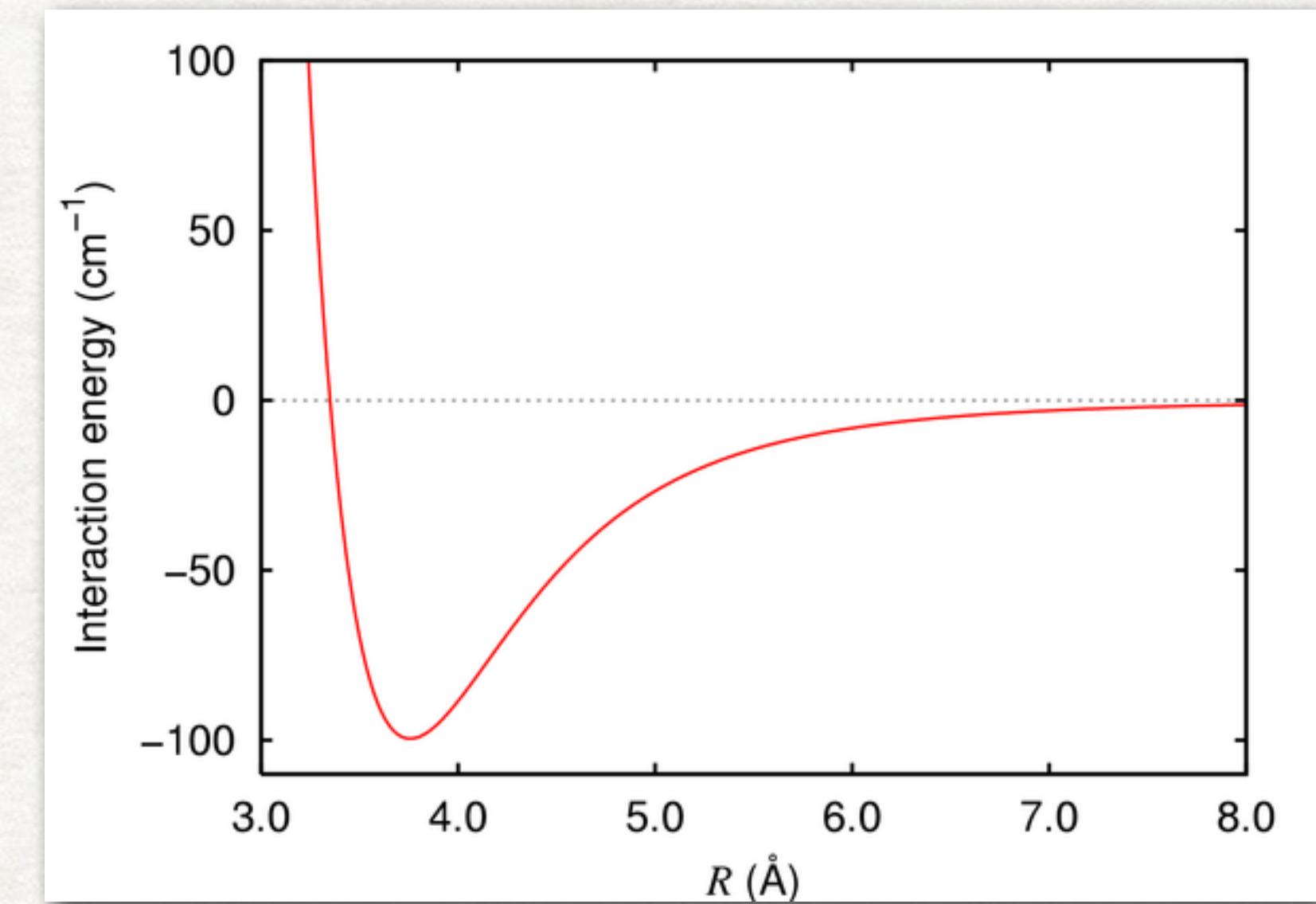
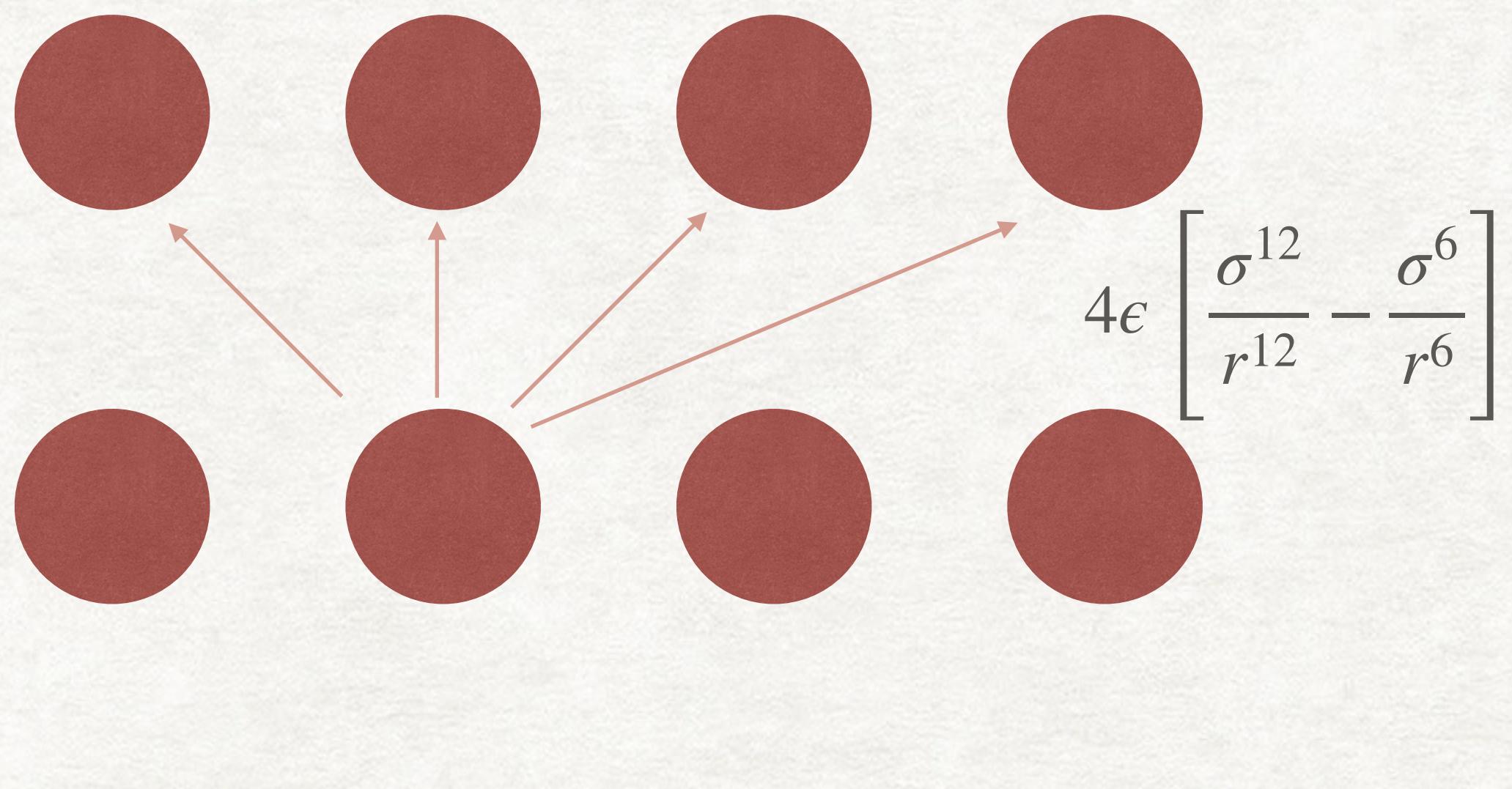
# QMC-HAMM Approach



Generate datasets and glue  
between different length scales.

Dataset for today: QMC  
calculations of AB, SP, AA, and  
'Mid' vs interactions.

# Using the QMC Data To Link to Larger Scales



Question: how well does the London formula (Lennard-Jones potential) applied atom-by-atom describe the QMC data?

If it does well, then we can compute the energy of a configuration of atoms in microseconds instead of weeks! This allows us to simulate much larger systems.

## Demo:

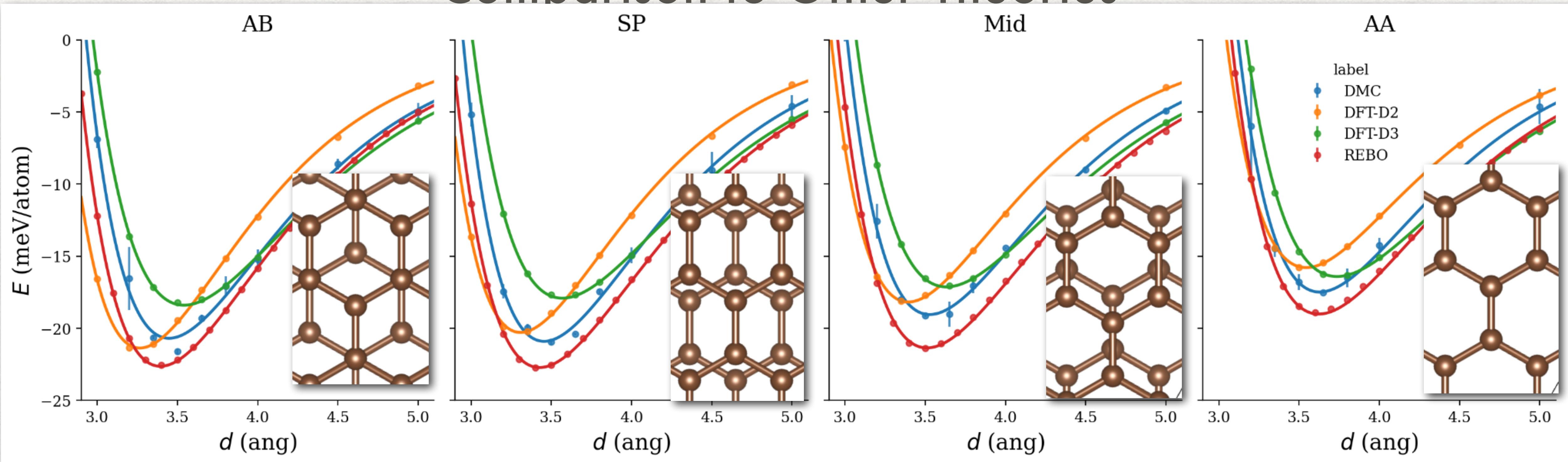
Exercise:

<https://colab.research.google.com/drive/1o9NpKKDAI1q28hMBRM9x8tfN3qsWalZq?usp=sharing>

Answers:

<https://colab.research.google.com/drive/1qENa9wtM4Dt8ML4CbFfilomA2Fpvjwze?usp=sharing>

# Comparison to Other Theories



Standard techniques do not quite replicate the QMC data either.

# More Complicated Functions Fit This

