

Correlated electronic states in moiré heterostructures: insights from quantum chemistry

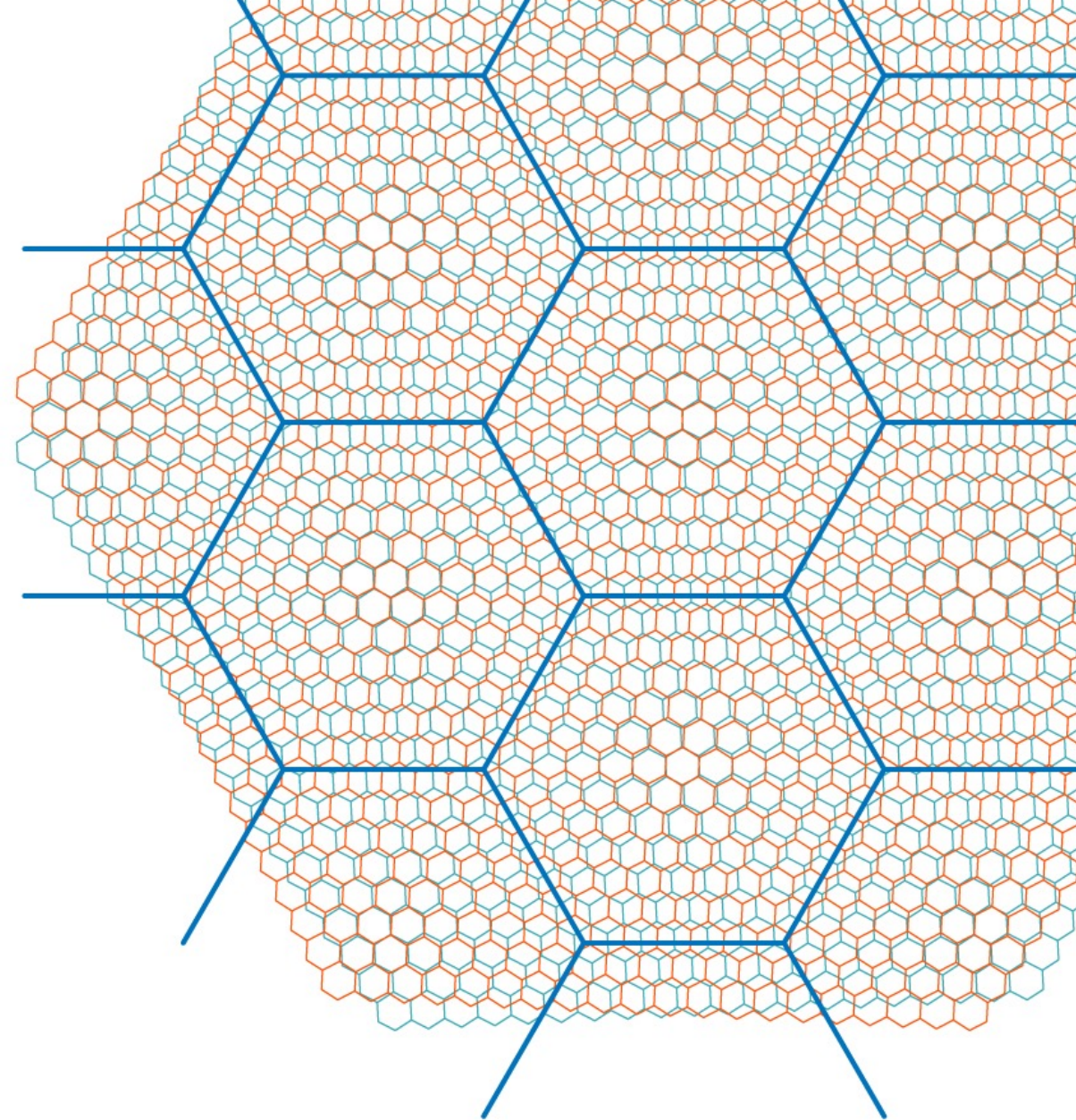
Shu Fay Ung

Thesis defense | 08/14/26

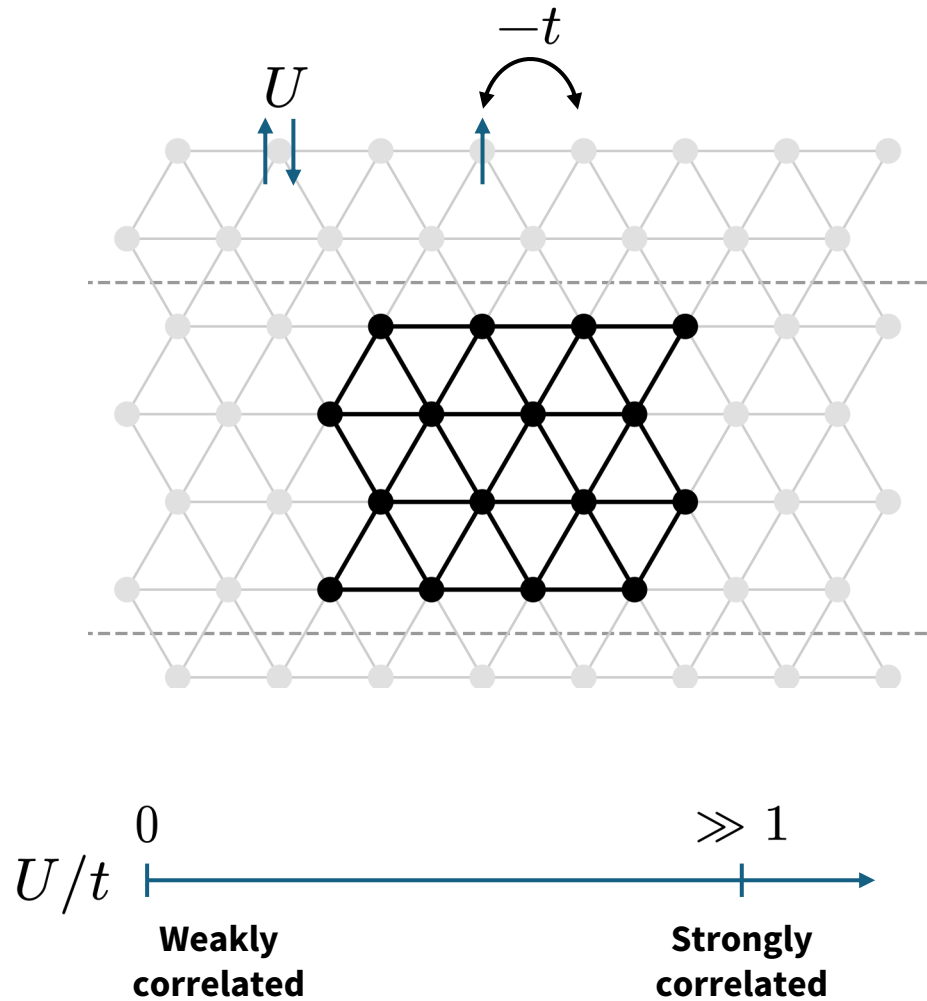
Advisor: Prof. David Reichman

Committee:

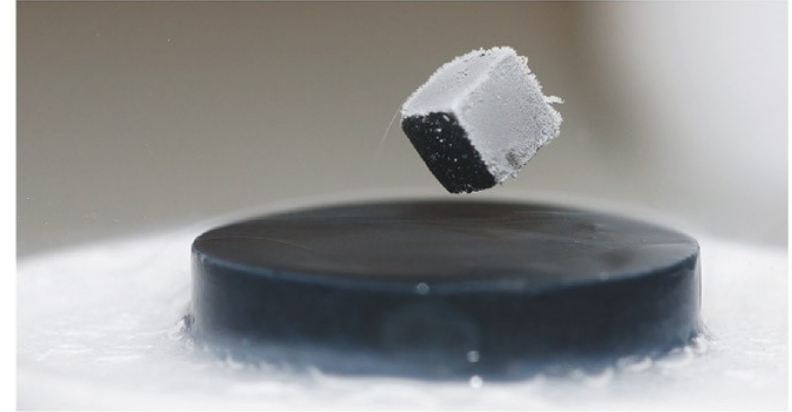
Prof. Timothy Berkelbach, Prof. Andrew Millis,
Prof. Diptarka Hait, Prof. Milan Delor



Strongly correlated systems

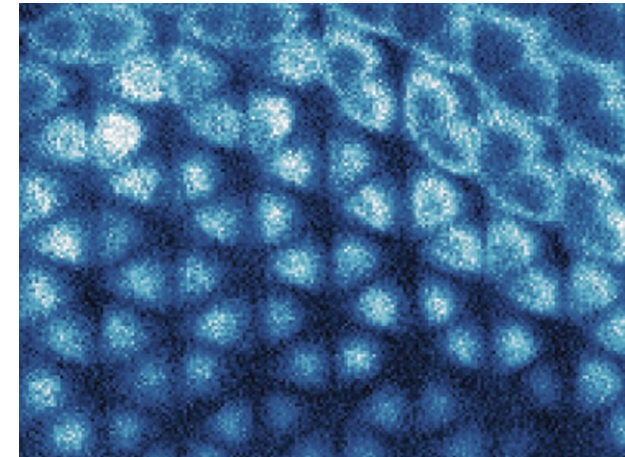


Superconductivity



Oak Ridge National Laboratory

Electron crystallization

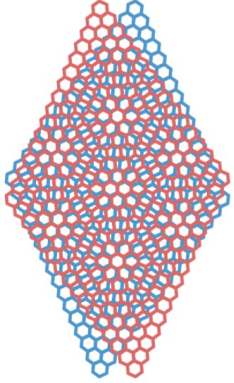


Nature 597, 650–654 (2021)

Moiré systems

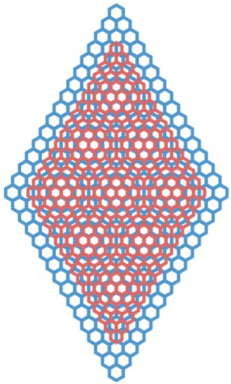
a Rotational misalignment

For example, twisted bilayer graphene



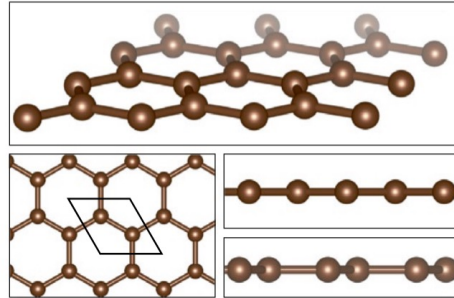
d Lattice mismatch

For example, aligned WSe_2/WS_2

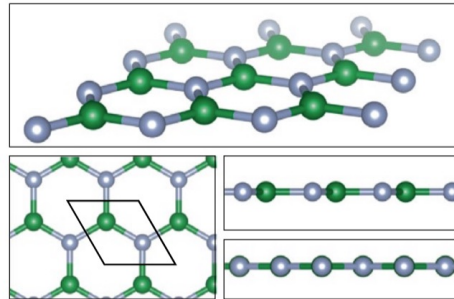


2D materials

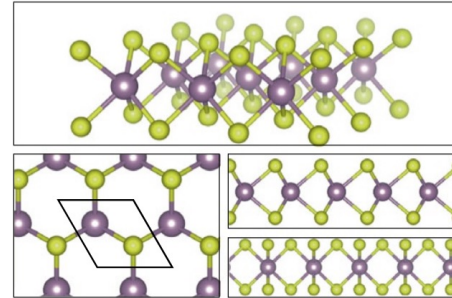
Graphene



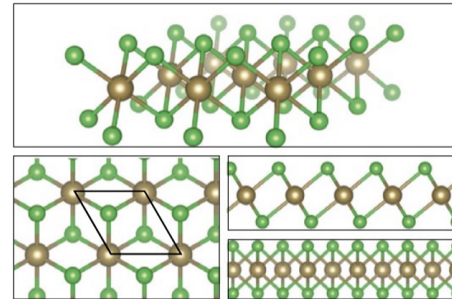
Hexagonal boron nitride



MoS_2



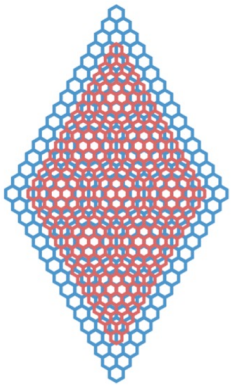
TaSe_2



**Transition-metal
dichalcogenides (TMDs)**

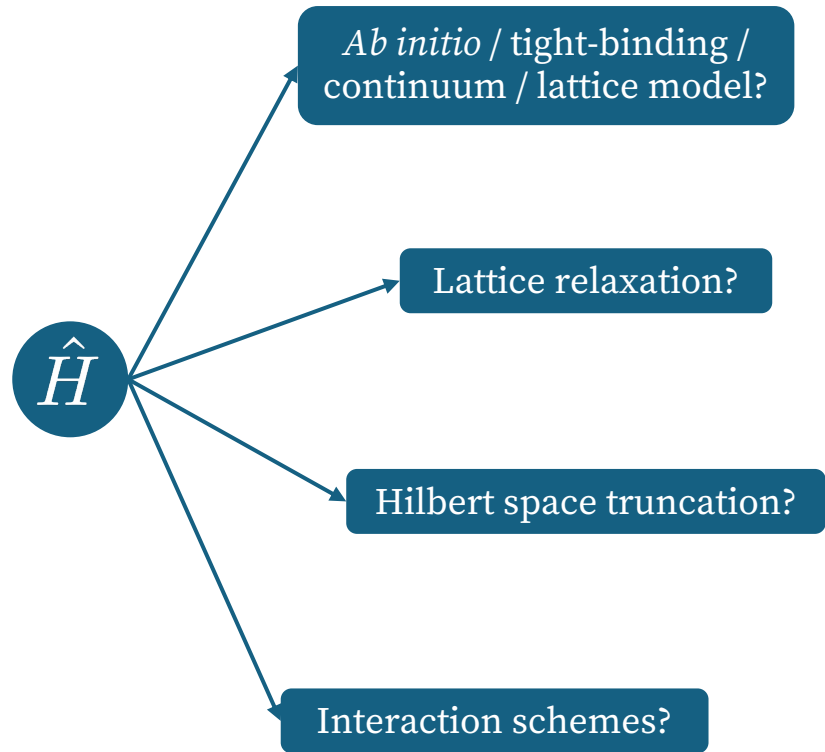
Nat. Rev. Mater. 5, 748-763 (2020)

Nat. Rev. Mater. 9, 460-480 (2024)

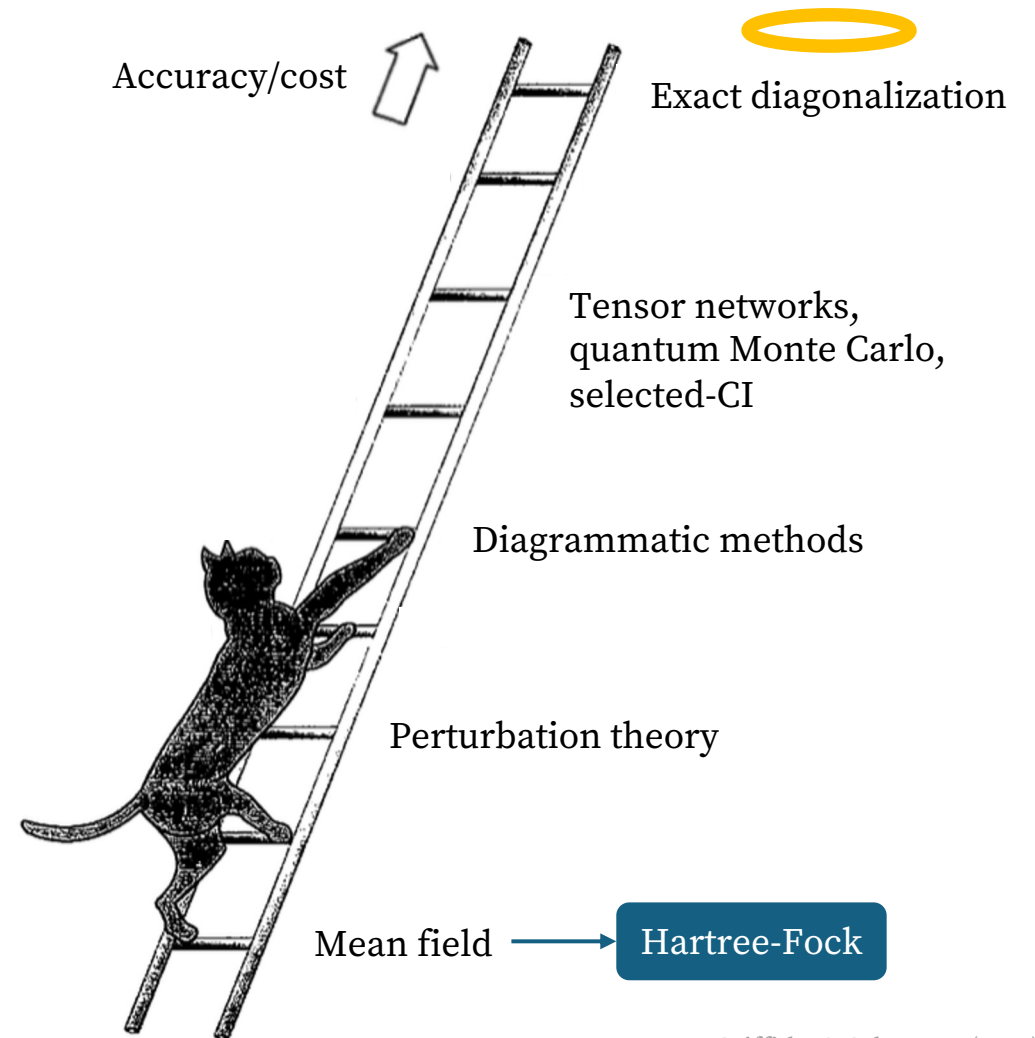


Theoretical efforts

Model construction

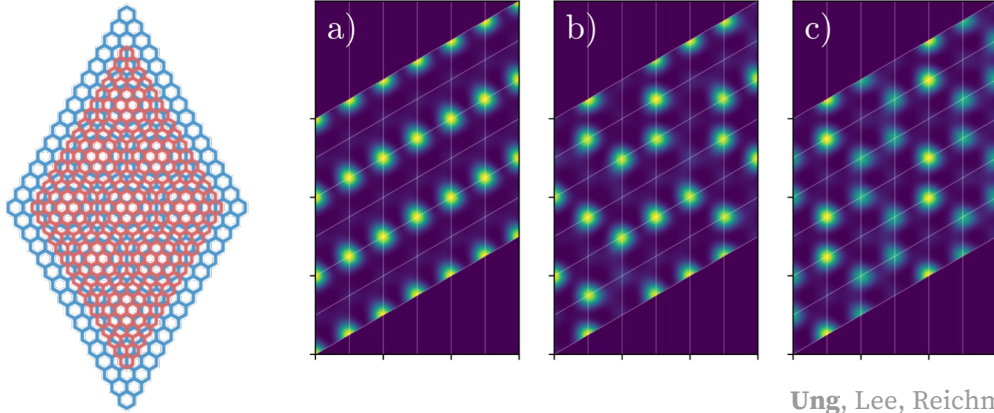


Model solution



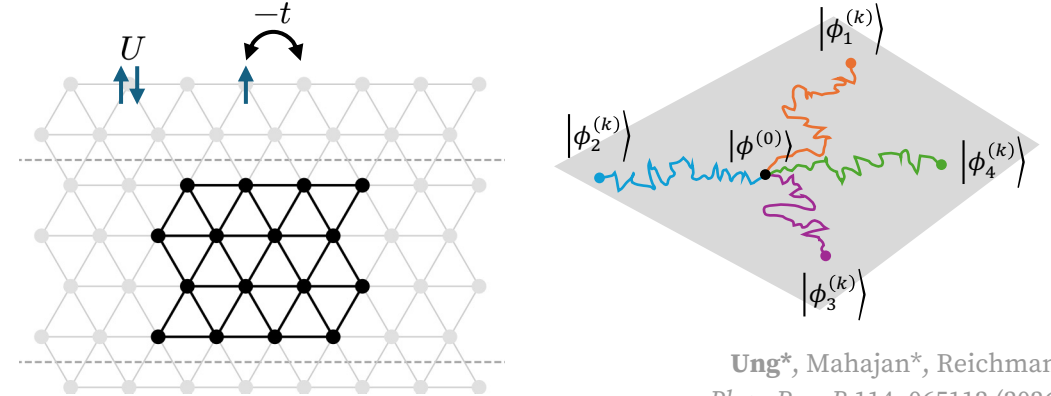
Thesis contributions

Generalized Wigner crystals in TMD heterobilayers



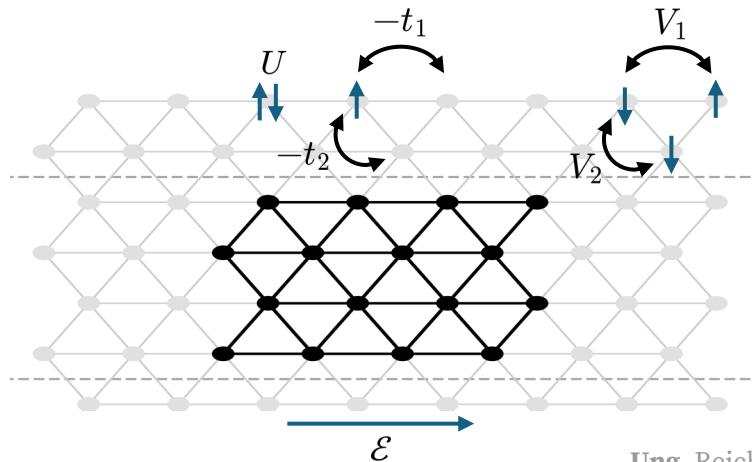
Ung, Lee, Reichman.
Phys. Rev. B 108, 245113 (2023)

Benchmark study of the triangular-lattice Hubbard model with CPMC



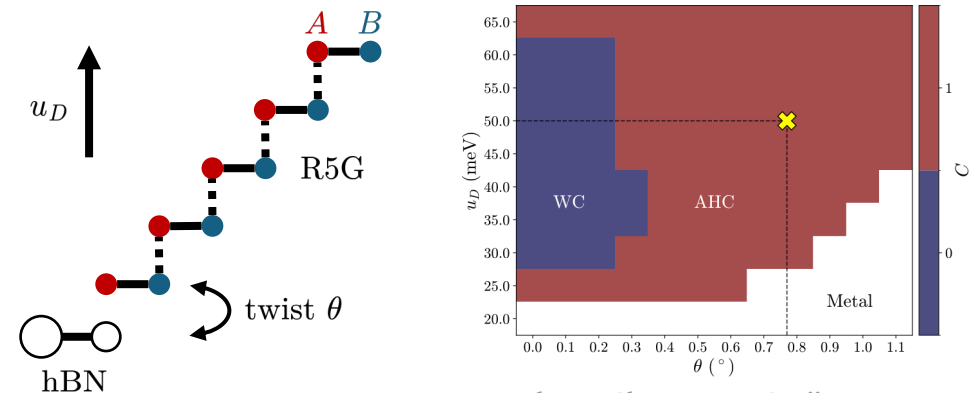
Ung*, Mahajan*, Reichman.
Phys. Rev. B 114, 065112 (2026)

Generalized Wigner crystals under strain



Ung, Reichman. *In prep.*

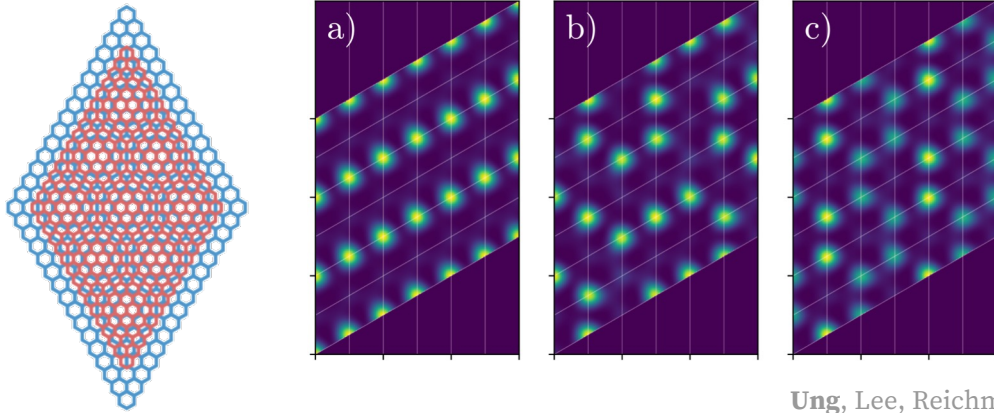
Electron correlation effects in rhombohedral pentalayer graphene/hBN



Ung, Zhang, Chen, Dong, Soejima, Lee. *In prep.*

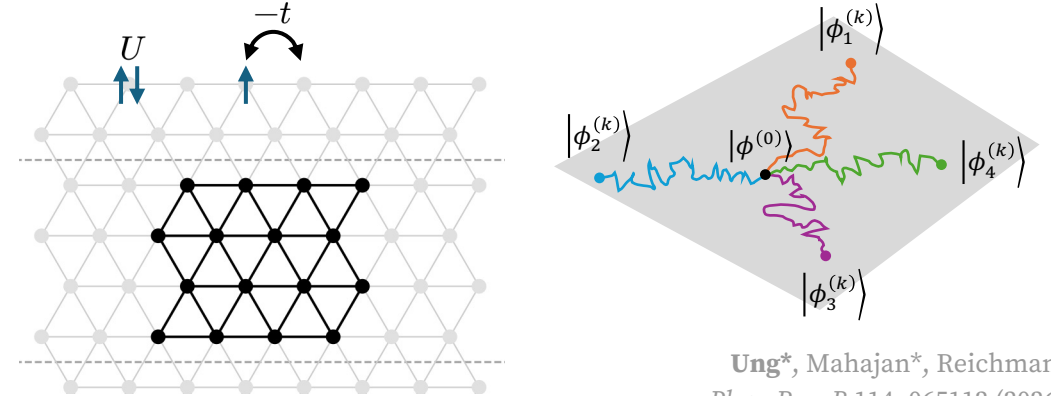
Talk outline

Generalized Wigner crystals in TMD heterobilayers



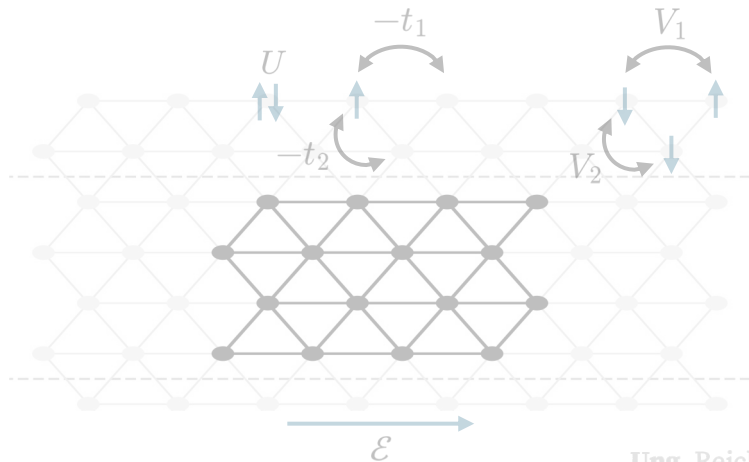
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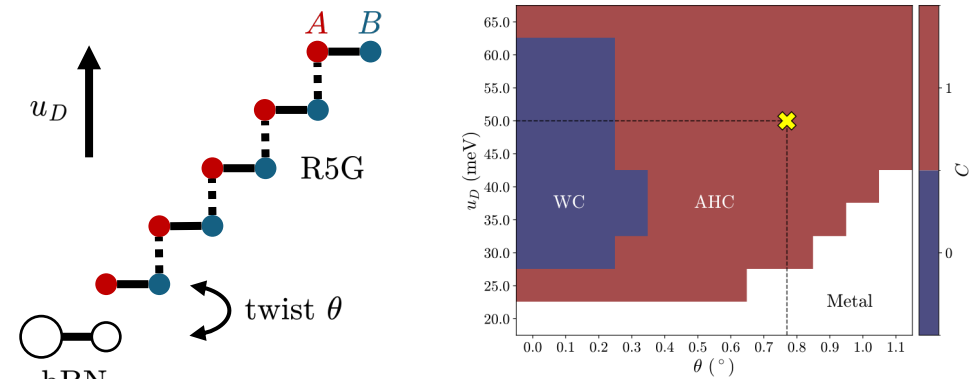
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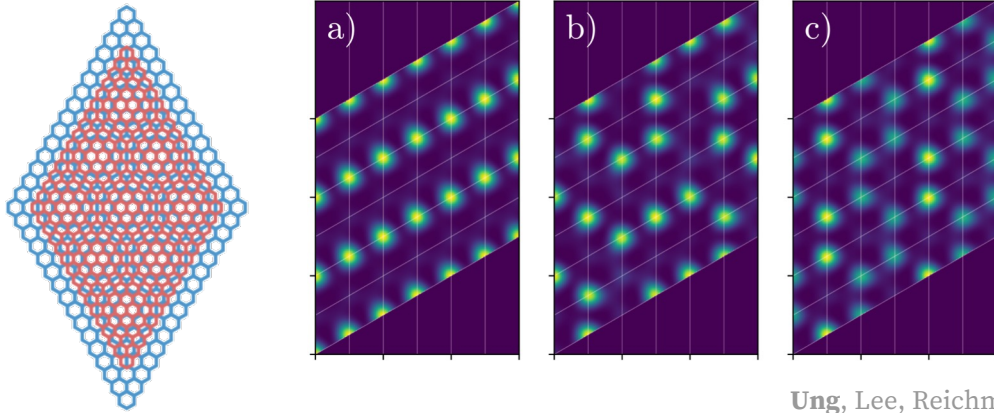
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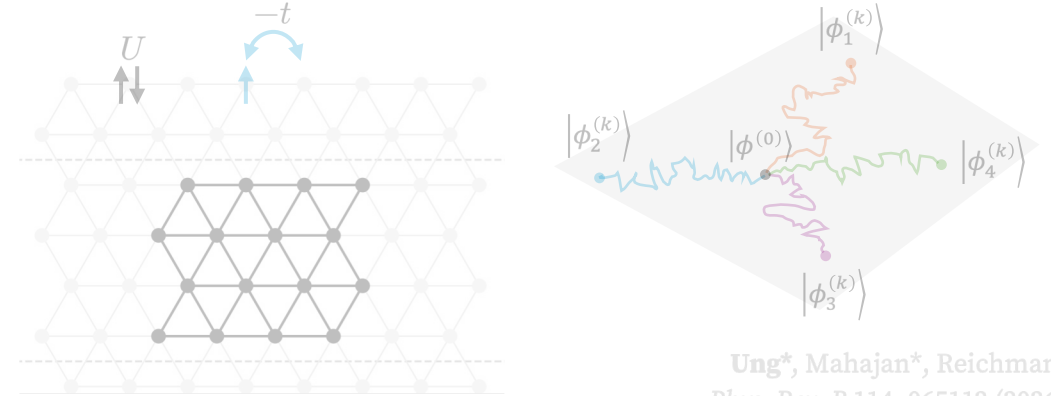
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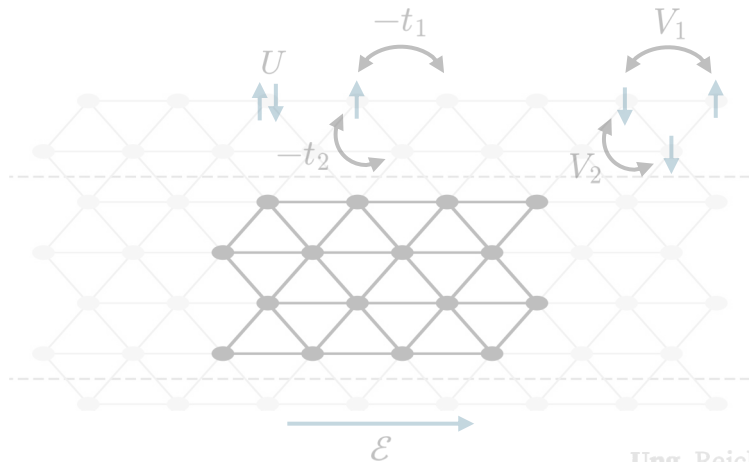
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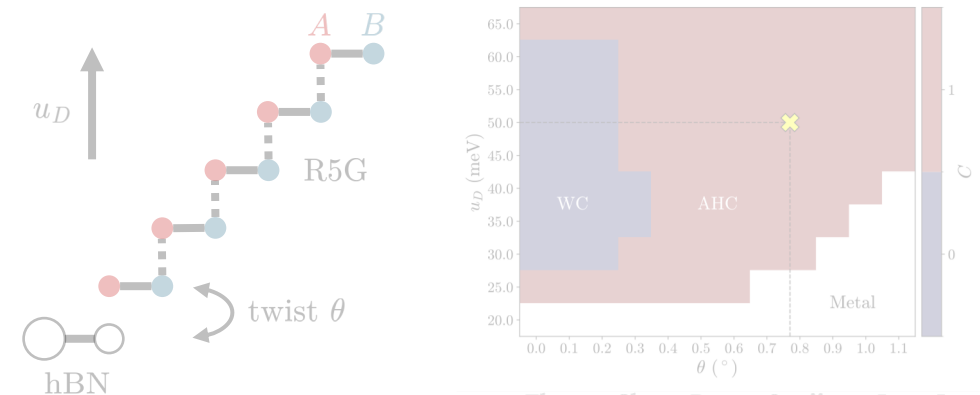
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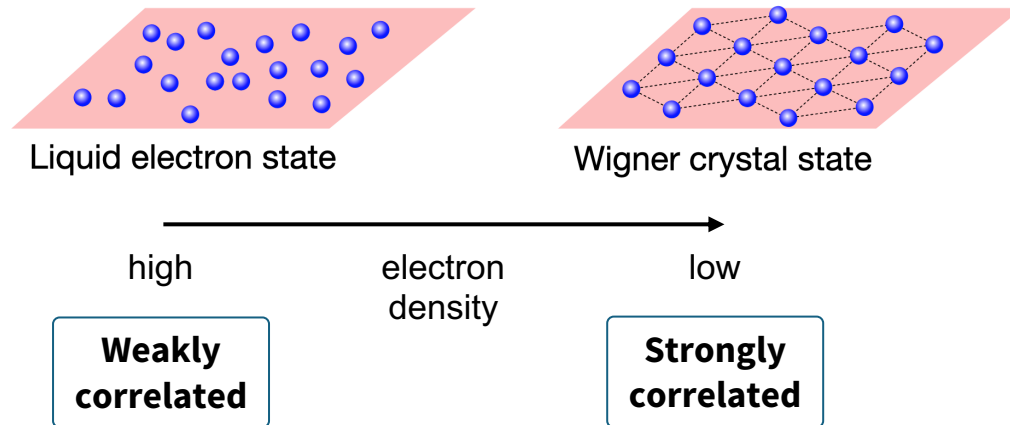
Generalized Wigner crystals (GWCs)

PHYSICAL REVIEW

On the Interaction of Electrons in Metals

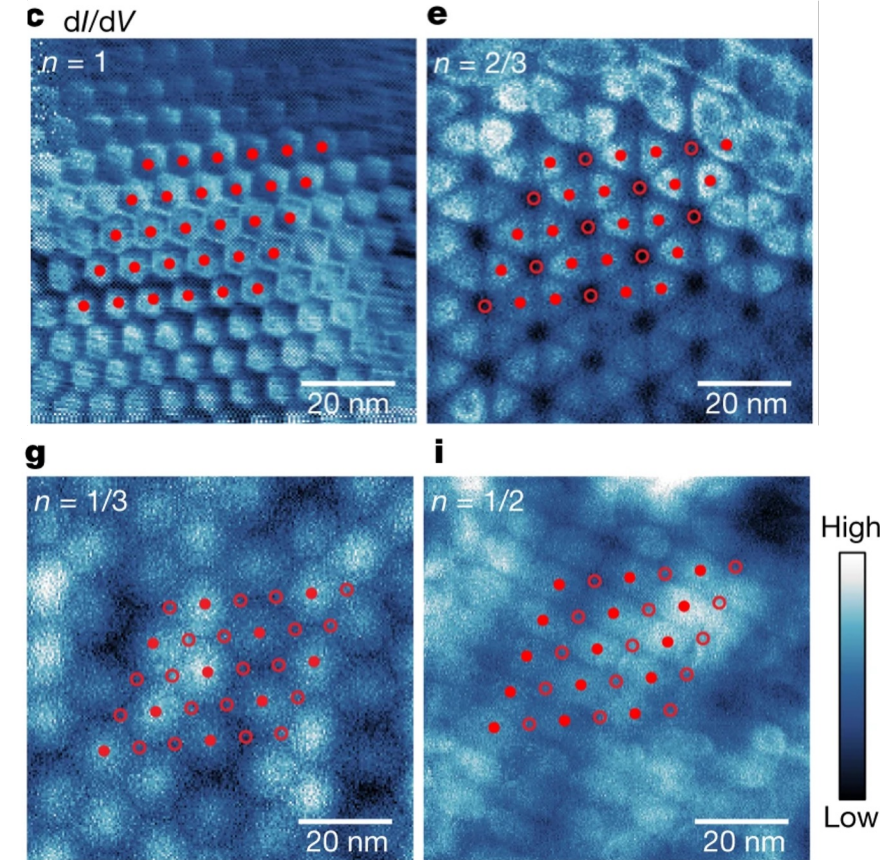
E. WIGNER, *Princeton University*

(Received October 15, 1934)



Nature 595, 53–57 (2021)

Scanning tunneling microscopy (STM)



Nature 597, 650–654 (2021)

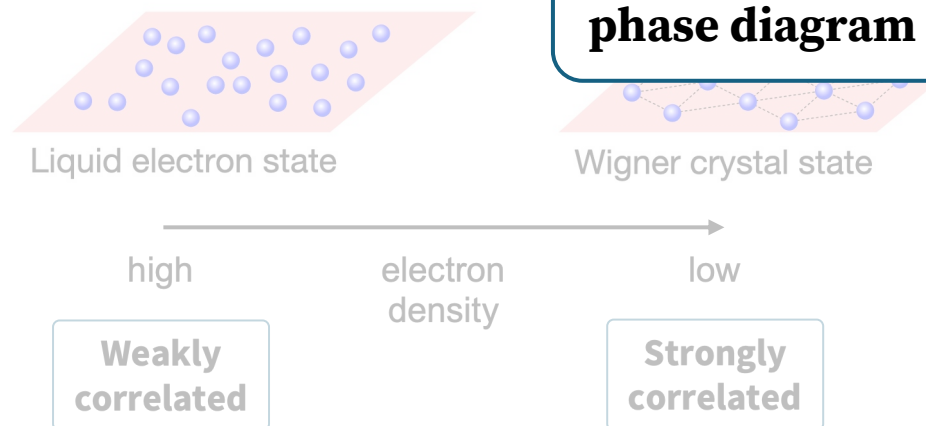
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PHYSICAL REVIEW

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E. WIGNER, *Princeton University*

(Received October 15, 1934)

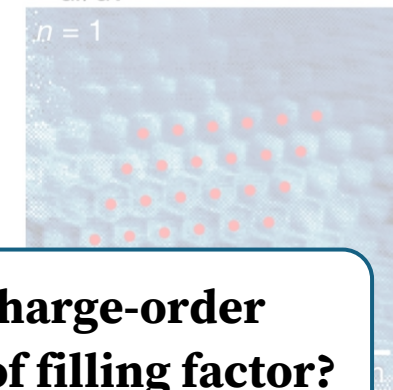


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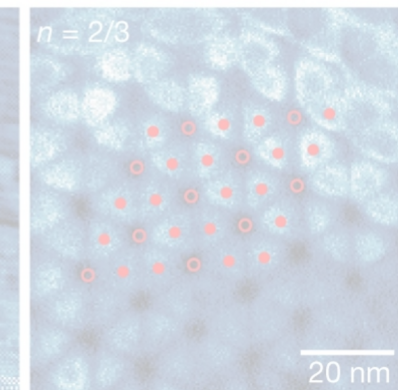
c dI/dV

$n = 1$

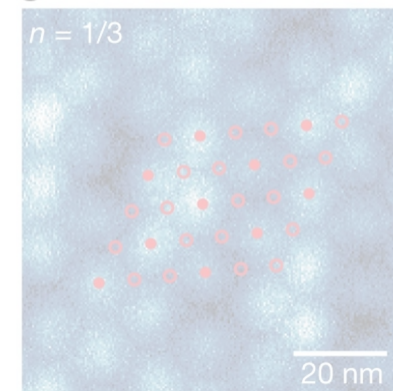


e

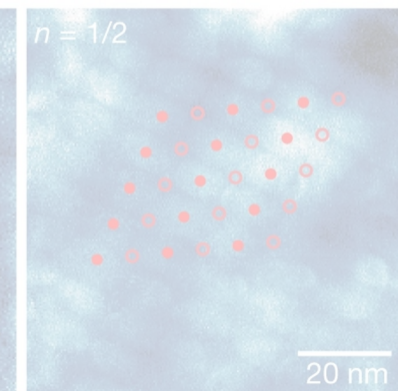
$n = 2/3$



g $n = 1/3$



i $n = 1/2$



High
Low

Nature 597, 650–654 (2021)

What is the ground state charge-order phase diagram as a function of filling factor?

Interacting continuum model

Continuum model

$$\hat{h}_i = -\frac{\nabla_i^2}{2m^*} + \Delta(\mathbf{r}_i)$$

m^* Effective mass

Phys. Rev. B 102, 201115(R) (2020)

Interacting continuum model

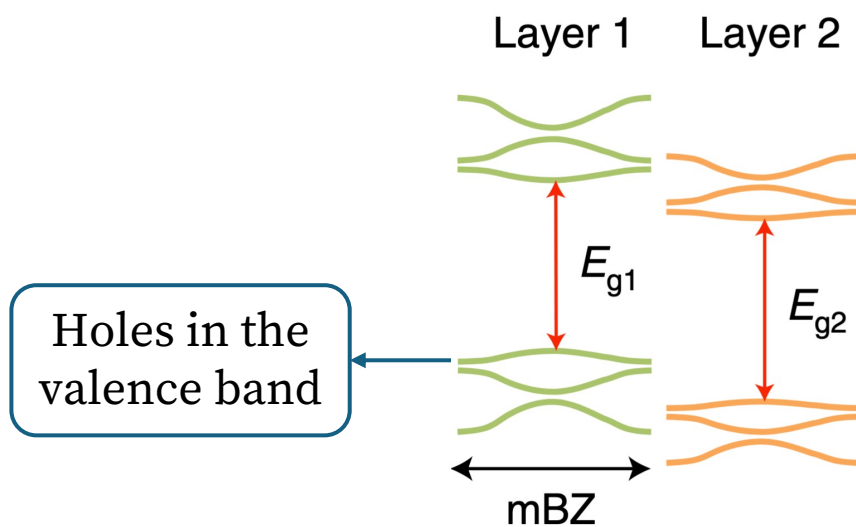
Continuum model $\hat{h}_i = -\frac{\nabla_i^2}{2m^*} + \Delta(\mathbf{r}_i)$

Moiré potential $\Delta(\mathbf{r}) = 2V_m \sum_{j=1}^3 \cos(\mathbf{g}_j \cdot \mathbf{r} + \phi)$

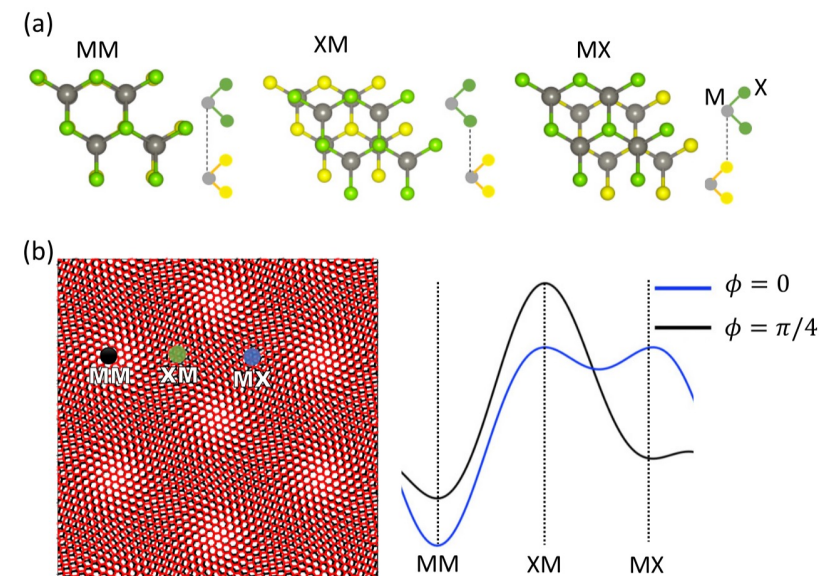
m^* Effective mass
 V_m Moiré potential strength
 ϕ Moiré potential shape

Interacting continuum model $\hat{H} = \sum_{i=1}^N \hat{h}_i + \frac{1}{2} \sum_{i \neq j}^N \frac{1}{\epsilon |\mathbf{r}_i - \mathbf{r}_j|}$

Phys. Rev. B 102, 201115(R) (2020)



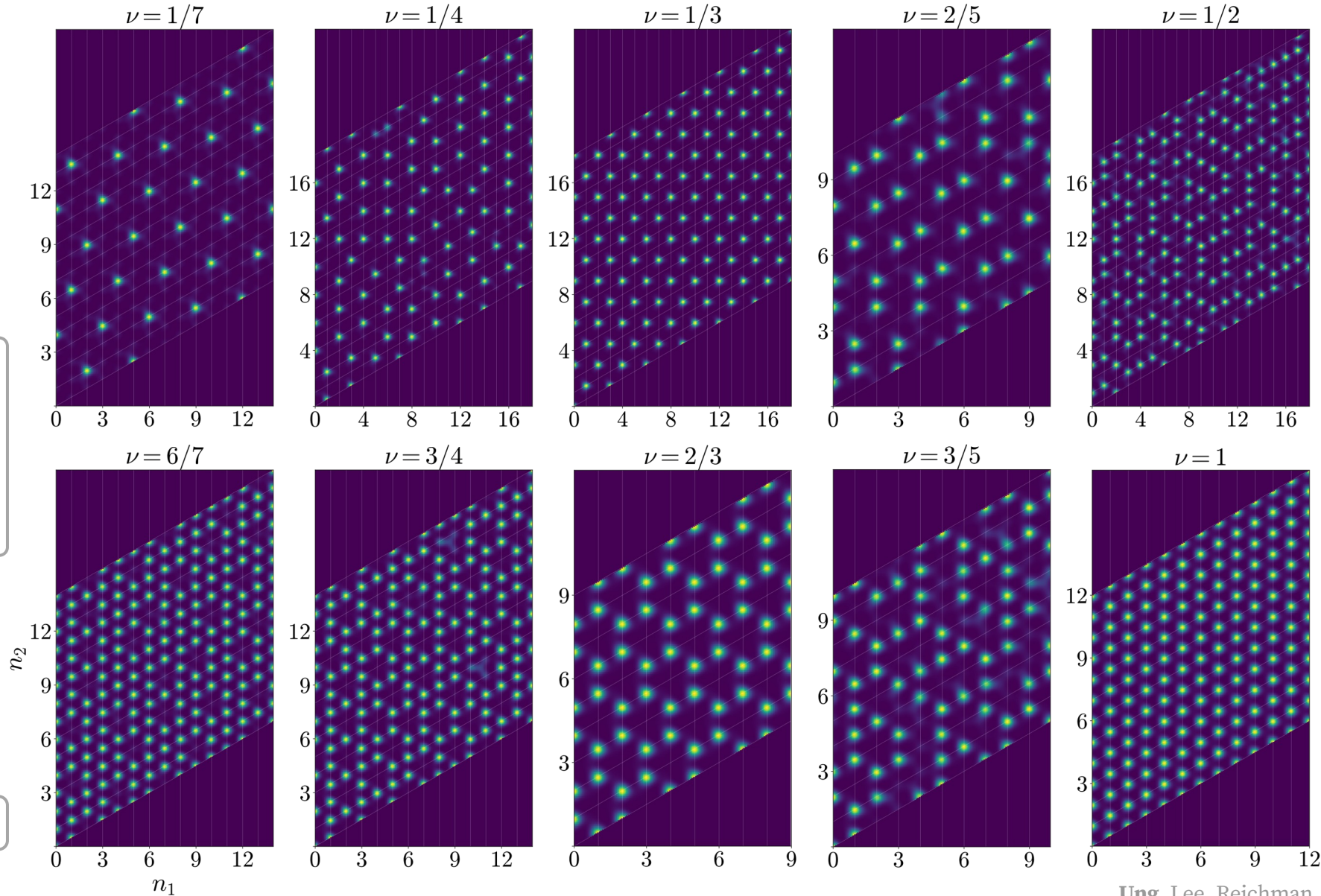
Nat. Nanotechnol. 17, 686-695 (2022)



Phys. Rev. B 102, 201115(R) (2020)

Hartree-Fock (HF) survey | charge densities

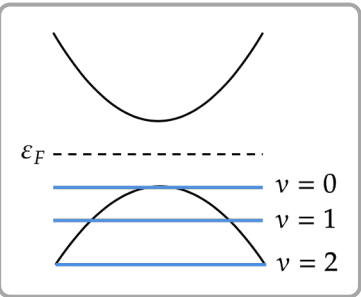
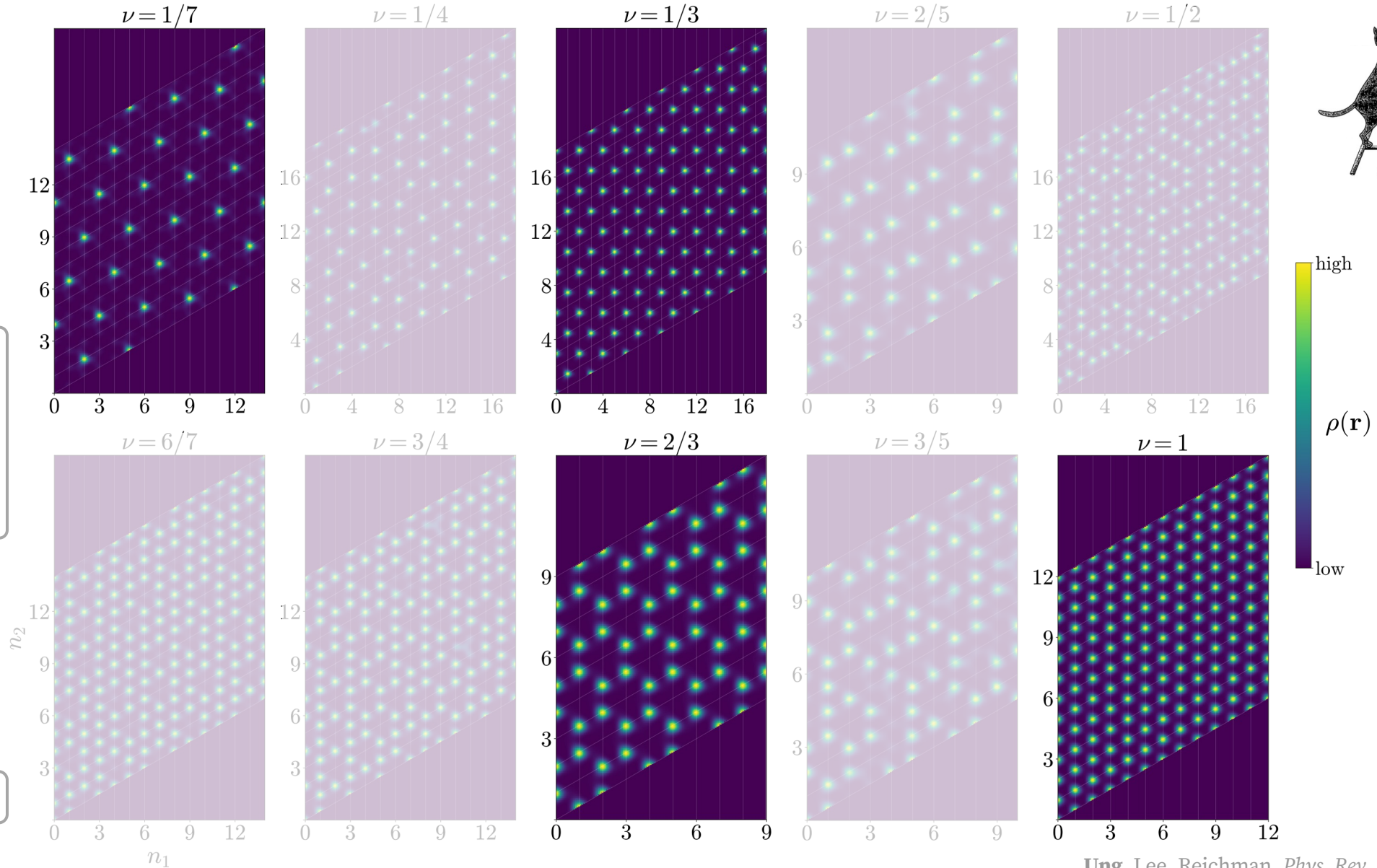
13



Moiré superlattice translational symmetry is generally broken.

Hartree-Fock (HF) survey | charge densities

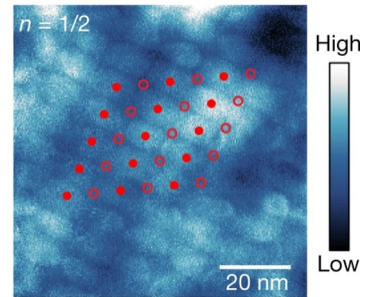
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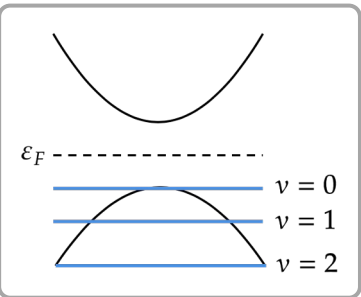
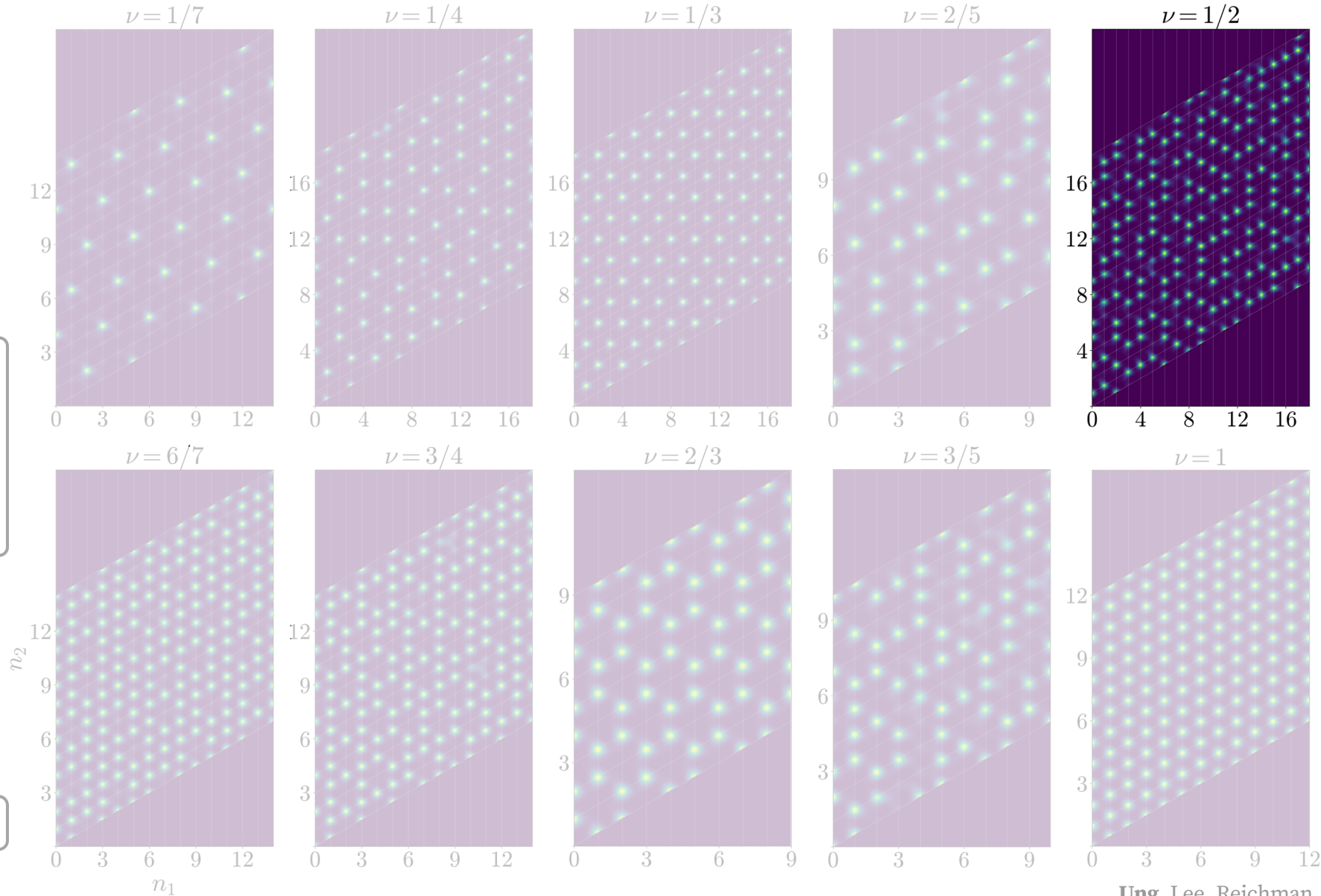
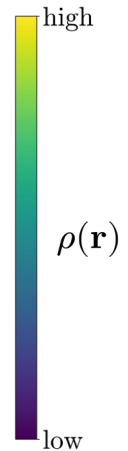
$$\mathbf{r} = n_1 \mathbf{L}_{m,1} + n_2 \mathbf{L}_{m,2}$$

Hartree-Fock (HF) survey | charge densities

Stripes

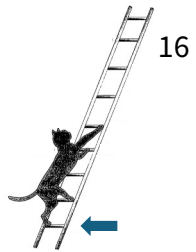


Nature 597, 650–654 (2021)

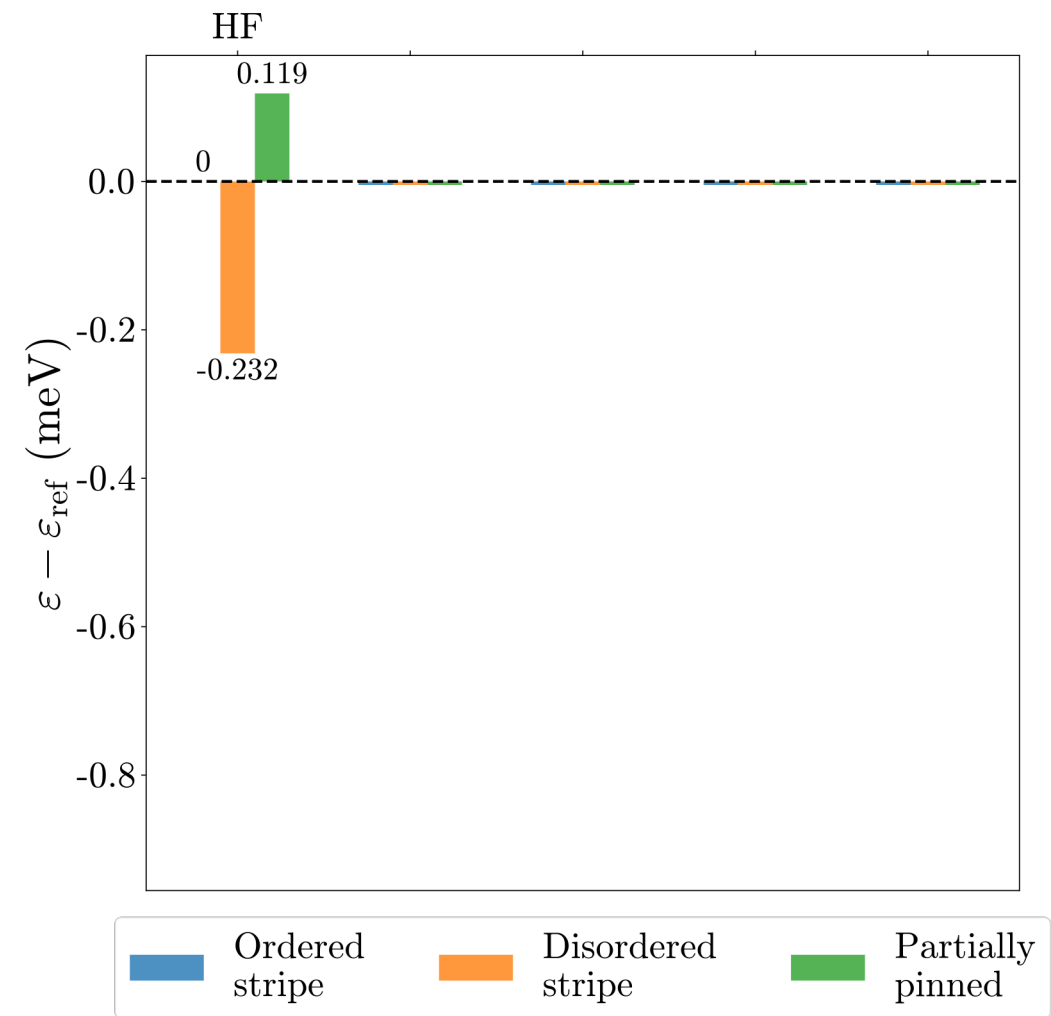
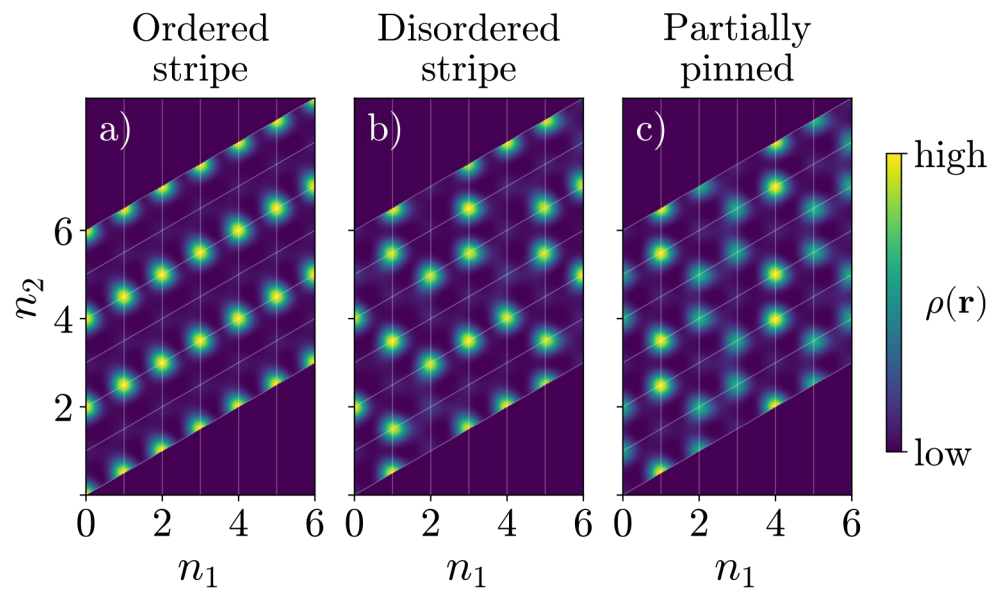


$$\mathbf{r} = n_1 \mathbf{L}_{m,1} + n_2 \mathbf{L}_{m,2}$$

Nearly degenerate solutions | $\nu = 1/2$



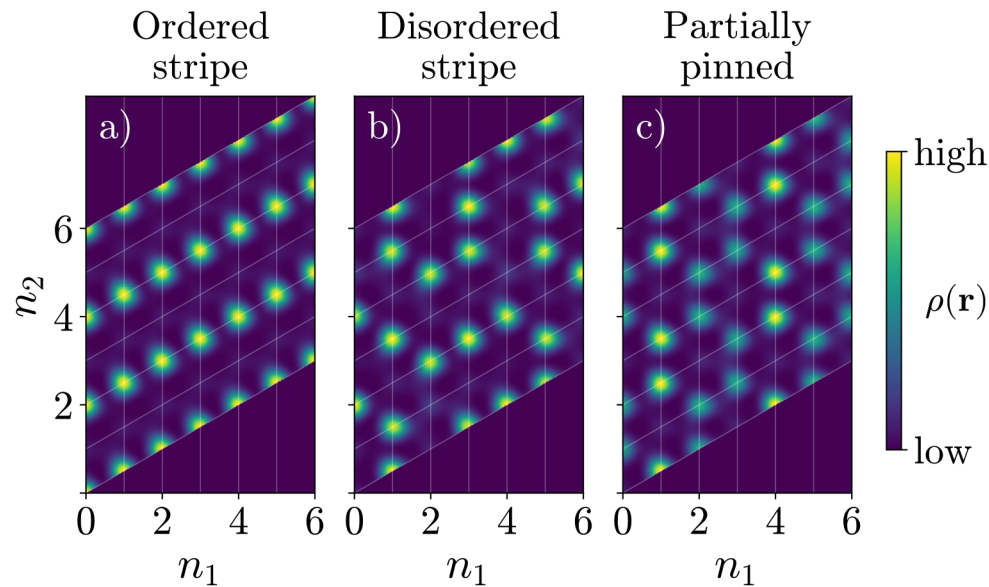
HF



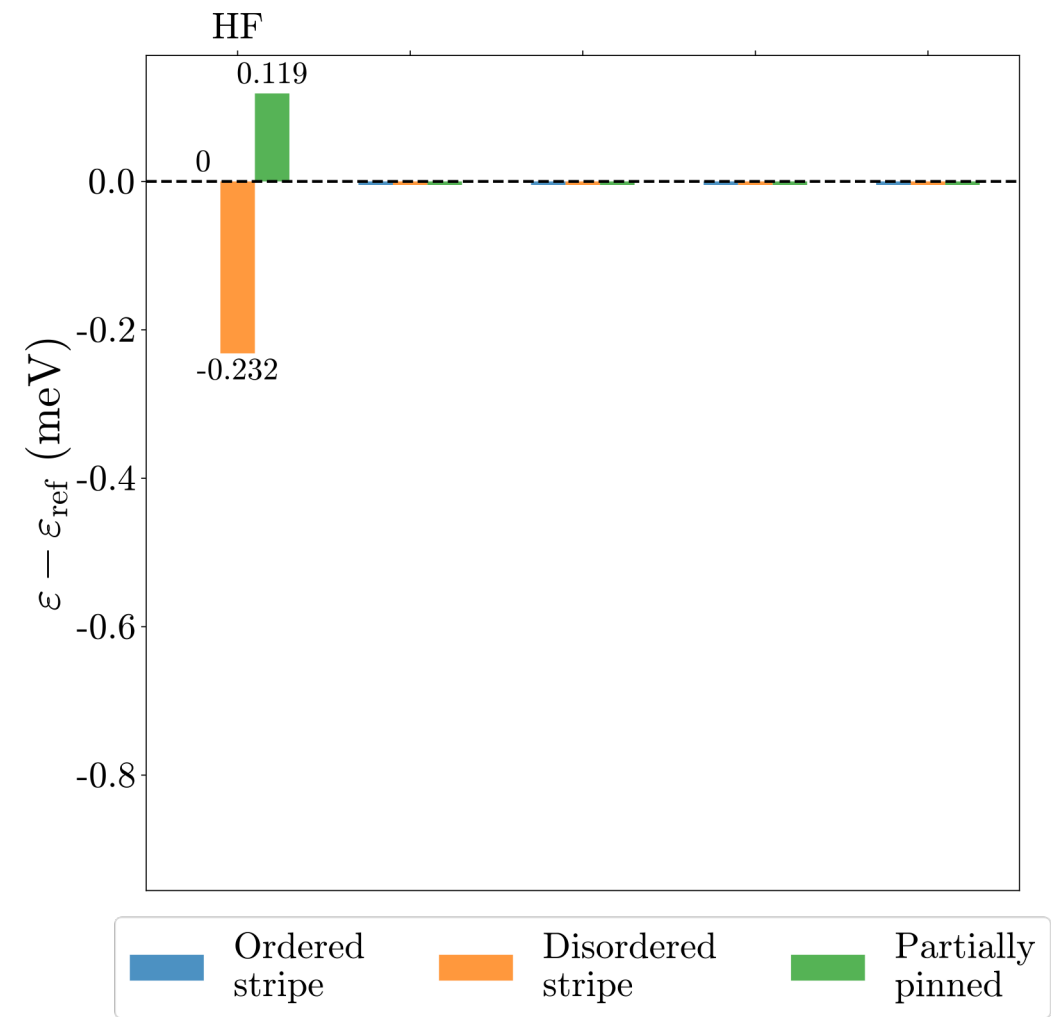
Nearly degenerate solutions | $v = 1/2$



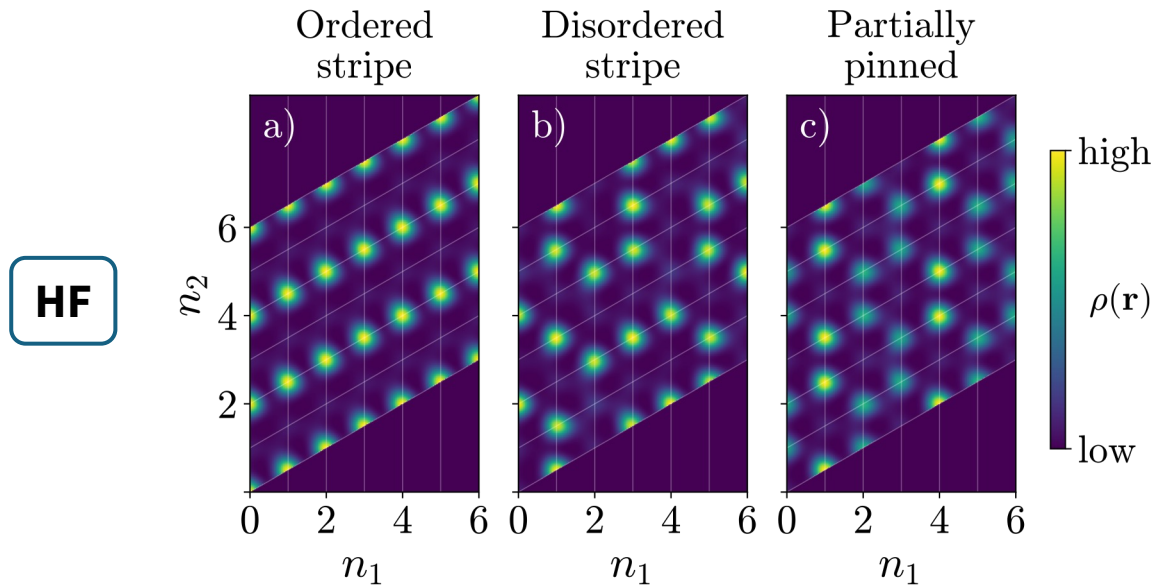
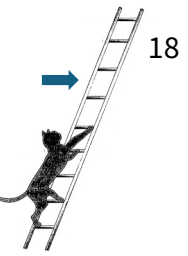
HF



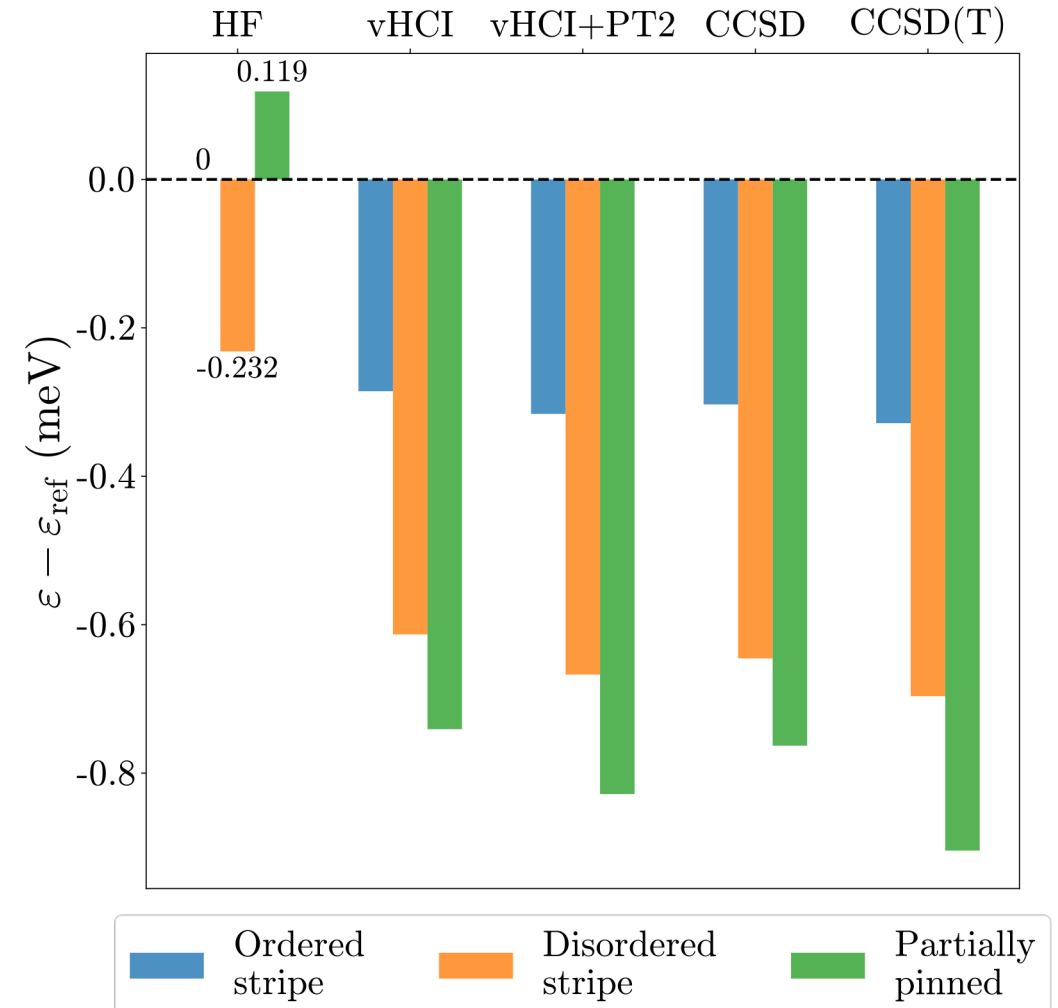
What happens if we add beyond-HF correlations?



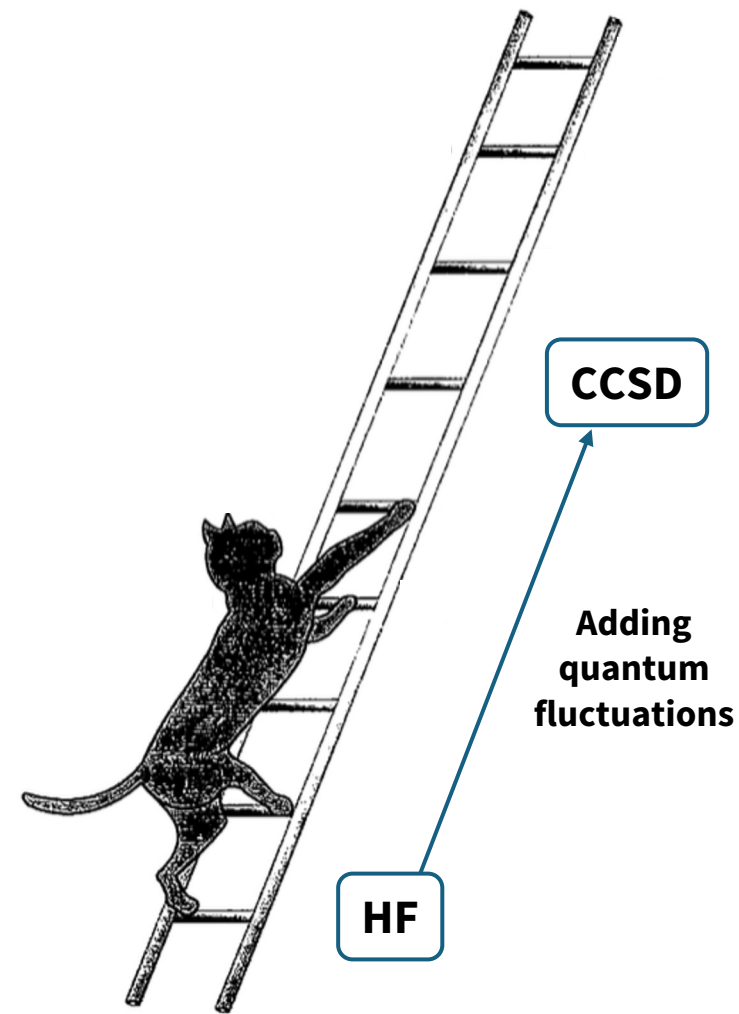
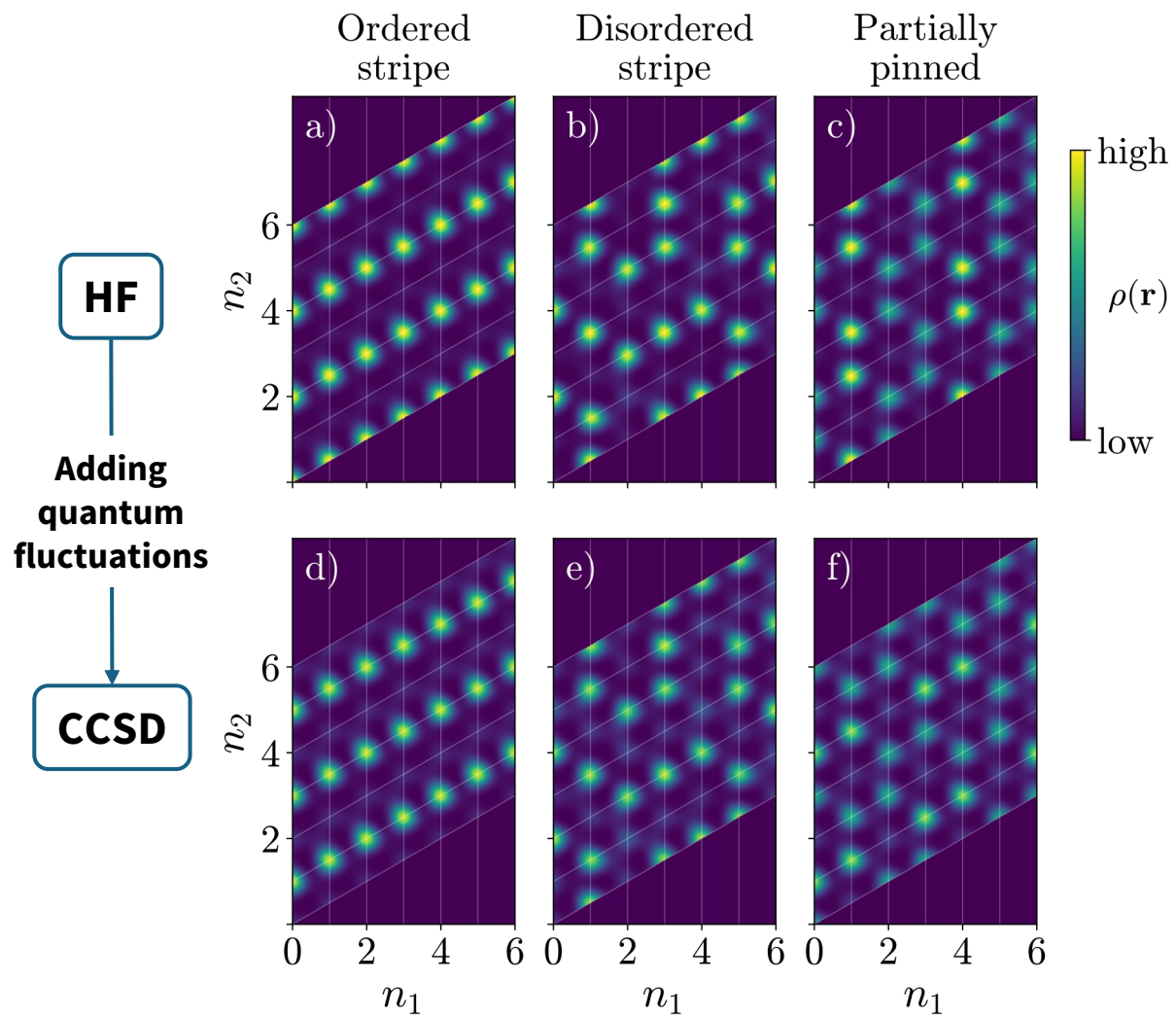
Nearly degenerate solutions | $\nu = 1/2$



- Energy differences are < 1 meV across all solutions and methods.
- **Correlation energy is non-trivial.**
 $\Rightarrow$ Small but sufficient to alter the energetic ordering of states from HF.
- **Partially-pinned solution has the lowest energy.**



Nearly degenerate solutions | $\nu = 1/2$



What about the ordered stripe state?

Discussion on the $n = 1/2$ Wigner crystal state

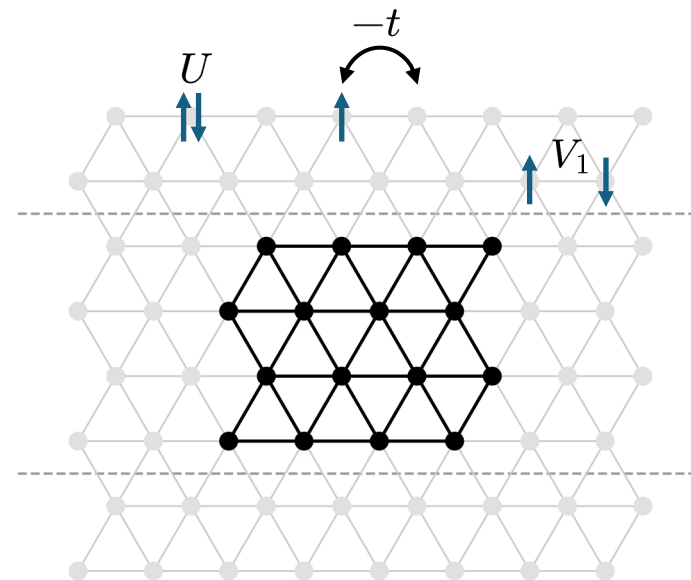
The broken symmetry of the $n = 1/2$ state could be impacted by accidental strain in the lattice, since stripes extending in the stretched lattice direction can have lower energy. We can characterize the sample anisotropy in the topography image shown in Fig. 1b (reproduced in Extended

...

Experimentally we also find that the $n = 1/2$ state is more fragile than the $n = 1/3$ and $n = 2/3$ states. A well-defined generalized Wigner crystal stripe phase is present only in a very narrow parameter space of V_{bias} and V_{BG} . The $n = 1/2$ state is also more sensitive to local inhomogeneity, as reflected by the disordering of the stripe electron lattice near the right

Nature 597, 650–654 (2021)

Triangular-lattice extended Hubbard models



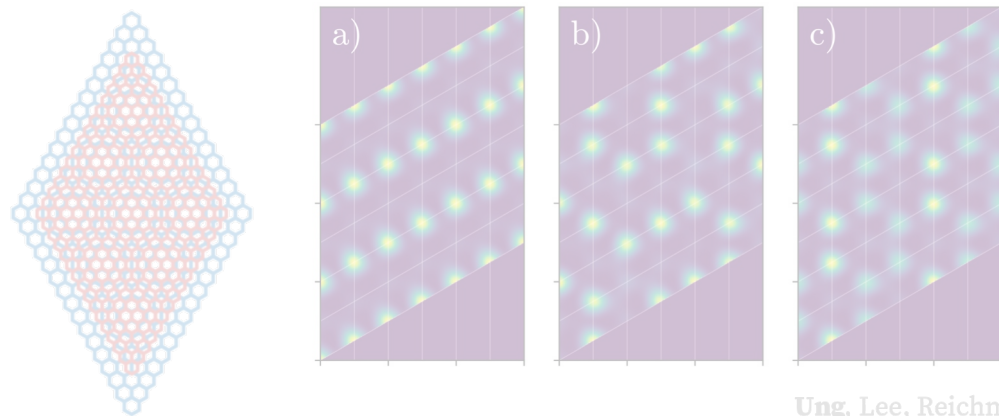
Hypothesis:

Strain in the experimental system may reorder the competing states and stabilize the stripe state.

- Able to access larger system sizes.
- Enables a clearer physical interpretation of strain effects.

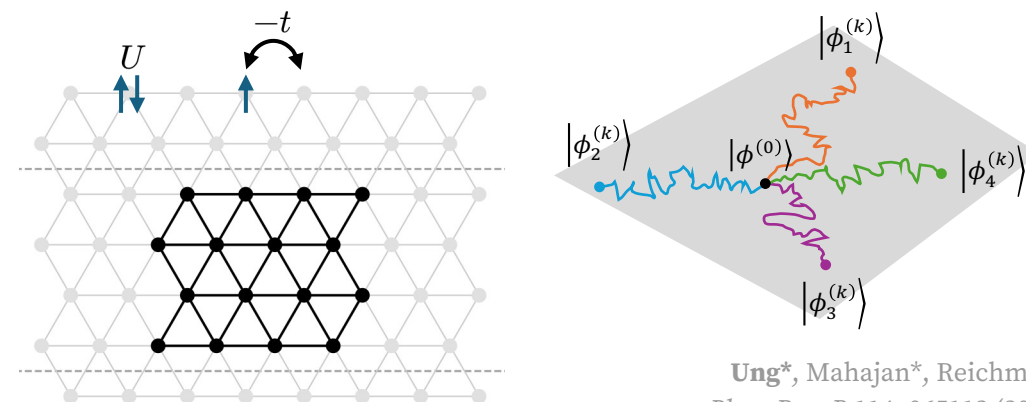
Talk outline

Generalized Wigner crystals in TMD heterobilayers



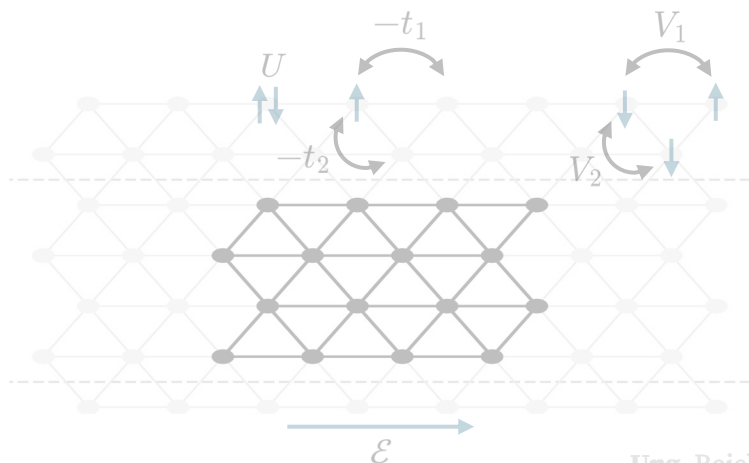
Ung, Lee, Reichman.
Phys. Rev. B 108, 245113 (2023)

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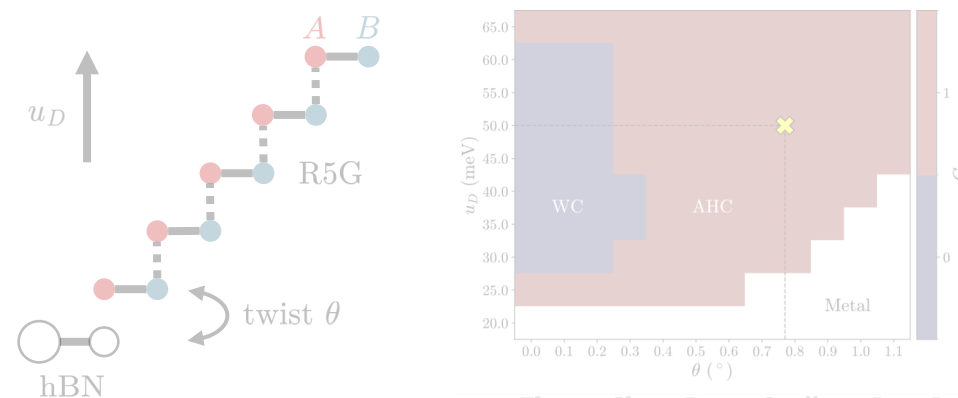
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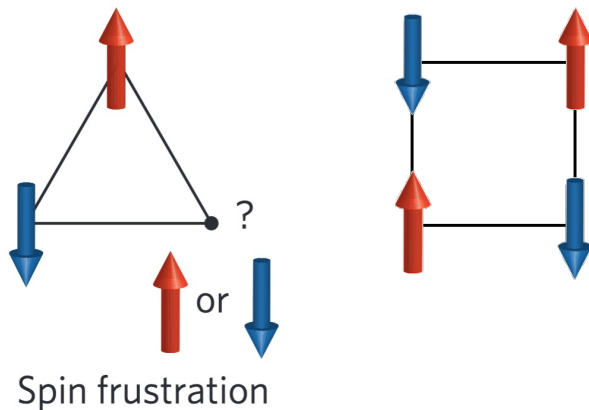


Ung, Zhang, Chen, Dong, Soejima, Lee. *In prep.*

Triangular-lattice Hubbard model

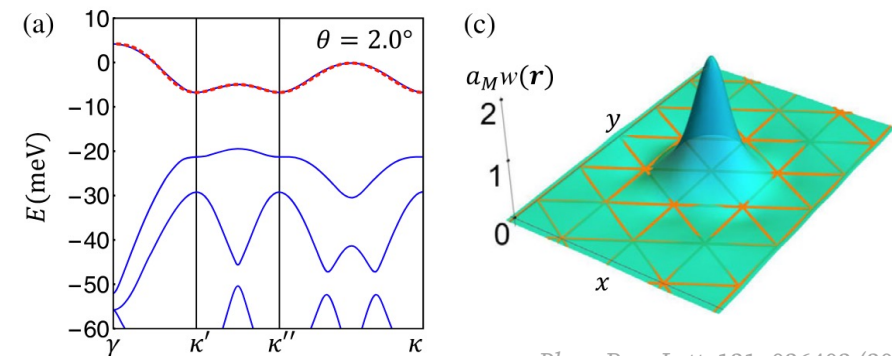
$$\hat{H} = -t \sum_{\sigma} \sum_{\langle ij \rangle} [\hat{c}_{i\sigma}^{\dagger} \hat{c}_{j\sigma} + \text{h.c.}] + U \sum_i \hat{n}_{i\uparrow} \hat{n}_{i\downarrow}$$

- Useful for studying:
 - TMD moiré heterostructures
 - Superconducting organic salts
 - Quantum spin liquids
- Triangular lattice**
 $\Rightarrow$ Strong correlation + geometric frustration



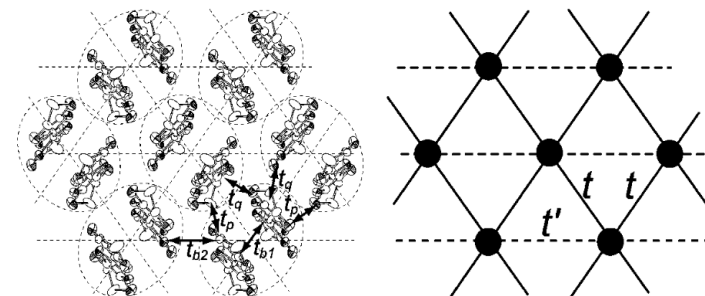
Nat. Phys. 9, 419-422 (2013)

WX₂/MoX₂ (X = S, Se)



Phys. Rev. Lett. 121, 026402 (2018)

κ -(ET)₂Cu₂(CN)₃



Phys. Rev. Lett. 91, 107001 (2003)

DMRG phase diagram



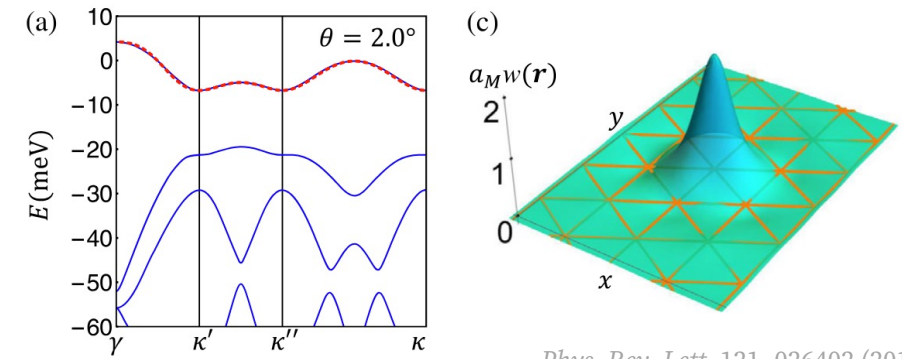
Phys. Rev. X 10, 021042 (2020)

Triangular-lattice Hubbard model

$$\hat{H} = -t \sum_{\sigma} \sum_{\langle ij \rangle} [\hat{c}_{i\sigma}^{\dagger} \hat{c}_{j\sigma} + \text{h.c.}] + U \sum_i \hat{n}_{i\uparrow} \hat{n}_{i\downarrow}$$

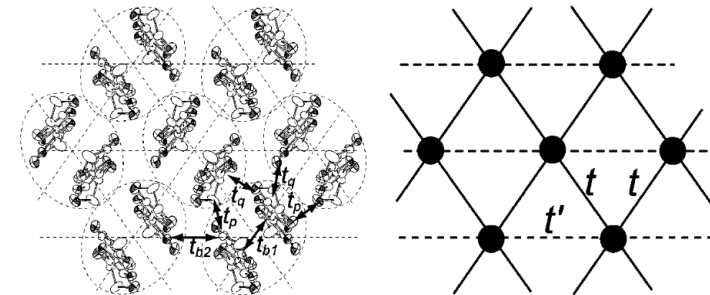
- Useful for studying:
 - TMD moiré heterostructures
 - Superconducting organic salts
 - Quantum spin liquids
- Triangular lattice**
 $\Rightarrow$ Strong correlation + geometric frustration
- Constrained-path Monte Carlo (CPMC) has been shown to be accurate for the **square-lattice** Hubbard model.
- How would CPMC perform for the triangular-lattice Hubbard model?**

WX₂/MoX₂ (X = S, Se)



Phys. Rev. Lett. 121, 026402 (2018)

κ -(ET)₂Cu₂(CN)₃



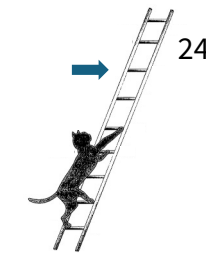
Phys. Rev. Lett. 91, 107001 (2003)

DMRG phase diagram



Phys. Rev. X 10, 021042 (2020)

Constrained-path Monte Carlo (CPMC)



Imaginary-time evolution

$$|\Psi_0\rangle \propto \lim_{\tau \rightarrow \infty} e^{-\tau \hat{H}} |\Phi^{(0)}\rangle$$

Hubbard-Stratonovich

$$e^{-\Delta\tau U \hat{n}_{i\uparrow} \hat{n}_{i\downarrow}} = e^{-\Delta\tau U (\hat{n}_{i\uparrow} + \hat{n}_{i\downarrow})/2} \underbrace{\sum_{x_i = \pm 1} \overbrace{p(x_i) e^{\gamma x_i (\hat{n}_{i\uparrow} - \hat{n}_{i\downarrow})}}^{\substack{\text{Probability density} \\ \text{Random variable}}} }_{\text{random propagator}}$$

Stochastic rep. of wave function

$$|\Phi^{(k)}\rangle = \sum_i w_i |\phi_i^{(k)}\rangle$$

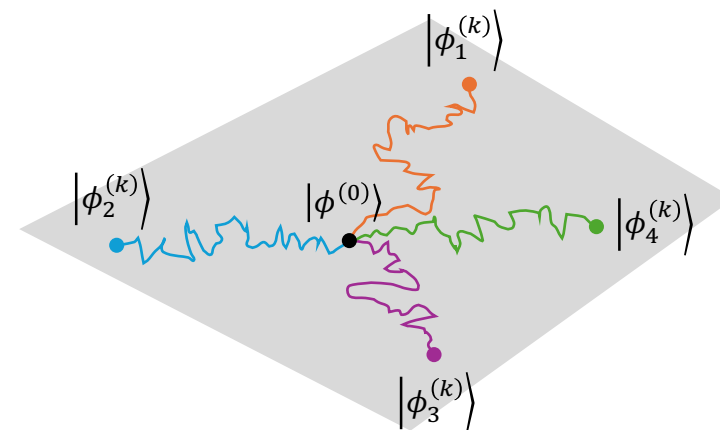
↓
Walkers

Sign problem

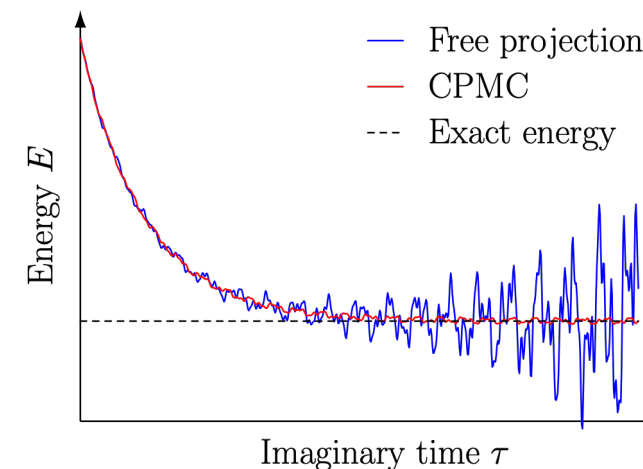
Walkers may cross the surface defined by $\langle \Psi_0 | \phi_i \rangle = 0$
 $\Rightarrow$ noisy sampling, exponential sample complexity.

Constrained path

Restrict to walkers that satisfy $\langle \psi_T | \phi_i \rangle > 0$ for some **trial wave function** $|\psi_T\rangle$
 $\Rightarrow$ polynomial sample complexity but **biased estimates**.



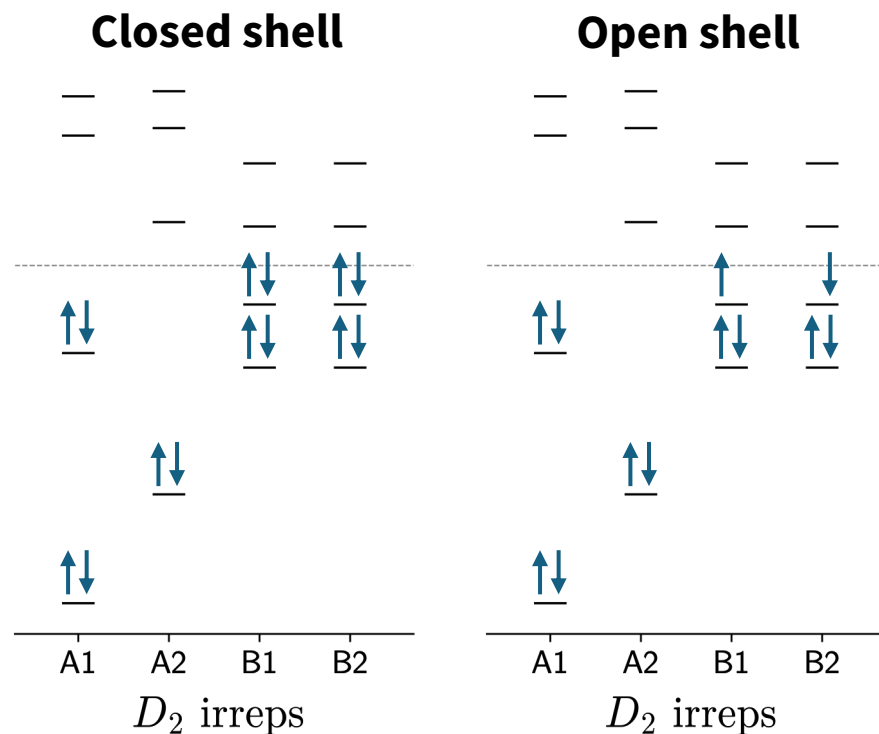
WIREs Comput. Mol. Sci. 8:e1364 (2018)



Symmetry-adapted trial wave functions

Free-electron (FE) trials

- Slater determinants built from eigenstates of the non-interacting Hamiltonian.
- Symmetry-preserving.



Symmetry-projected trials

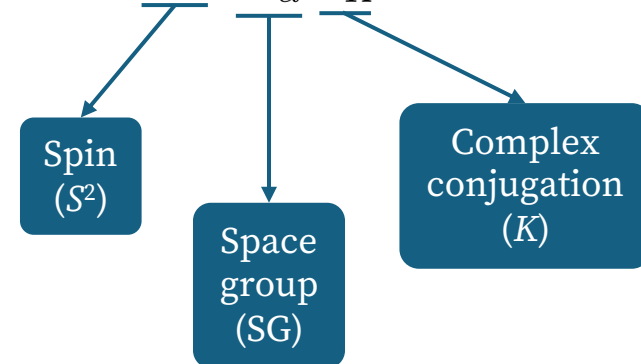
- Constructed by variationally optimizing a generalized HF (GHF) determinant $|\psi_{\text{GHF}}\rangle$ under symmetry projection.
 $\Rightarrow$ the variation-after-projection (VAP) approach.

Symmetry-projected energy

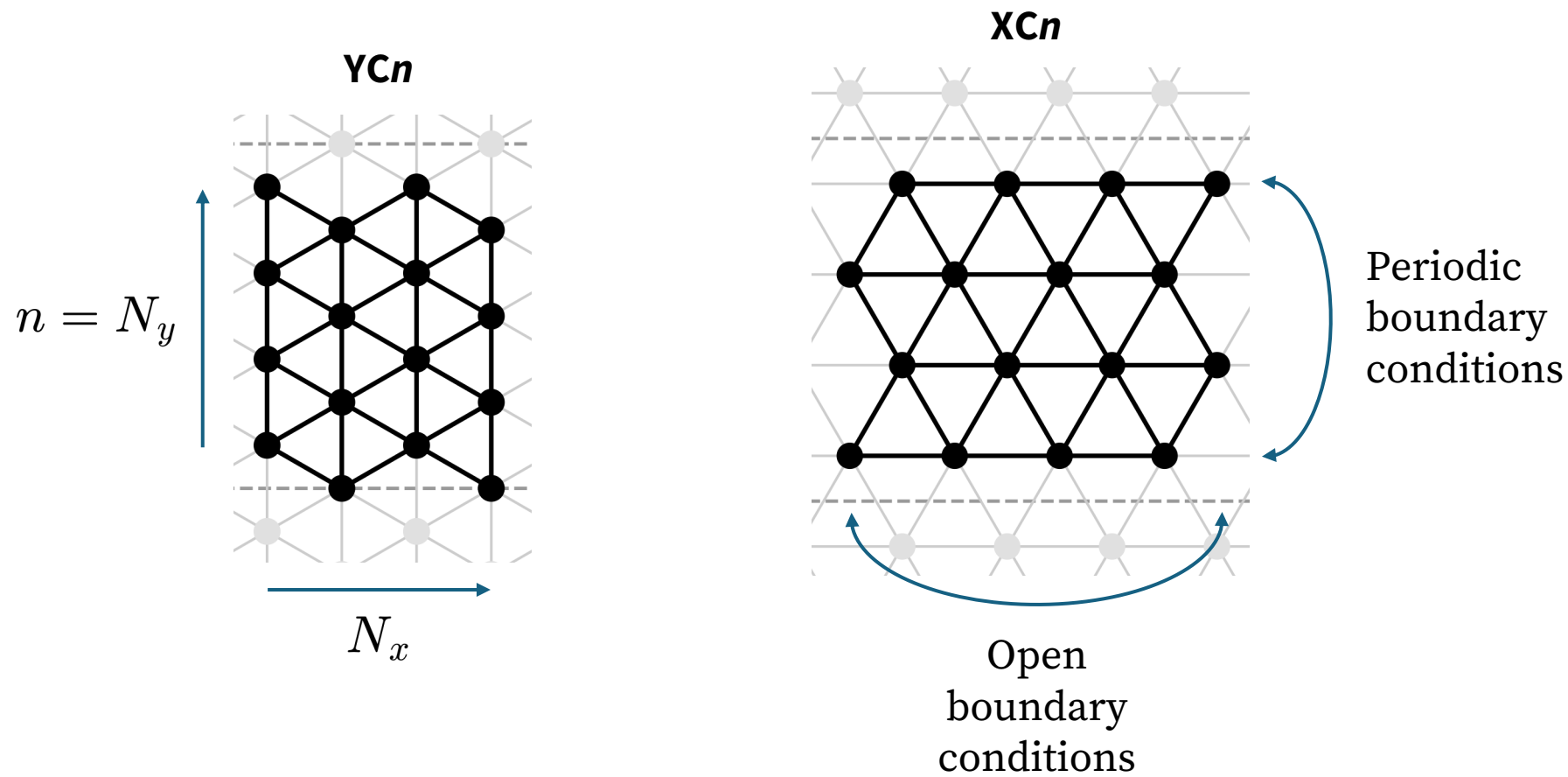
$$E = \frac{\langle \psi_{\text{GHF}} | \hat{H} \hat{\mathcal{P}} | \psi_{\text{GHF}} \rangle}{\langle \psi_{\text{GHF}} | \hat{\mathcal{P}} | \psi_{\text{GHF}} \rangle}$$

Symmetry projector

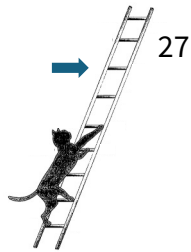
$$\hat{\mathcal{P}} = \hat{\mathcal{P}}^{s=0} \hat{\mathcal{P}}_{\alpha} \hat{\mathcal{P}}_K$$



Cylinder geometries

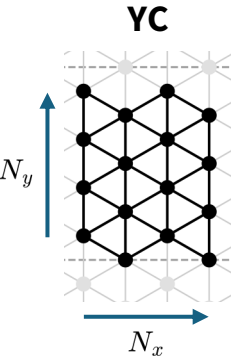
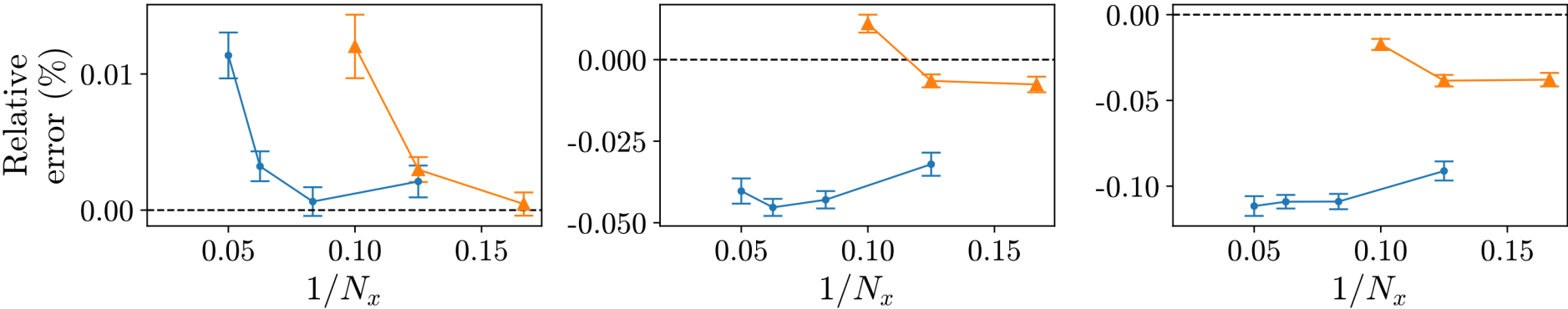
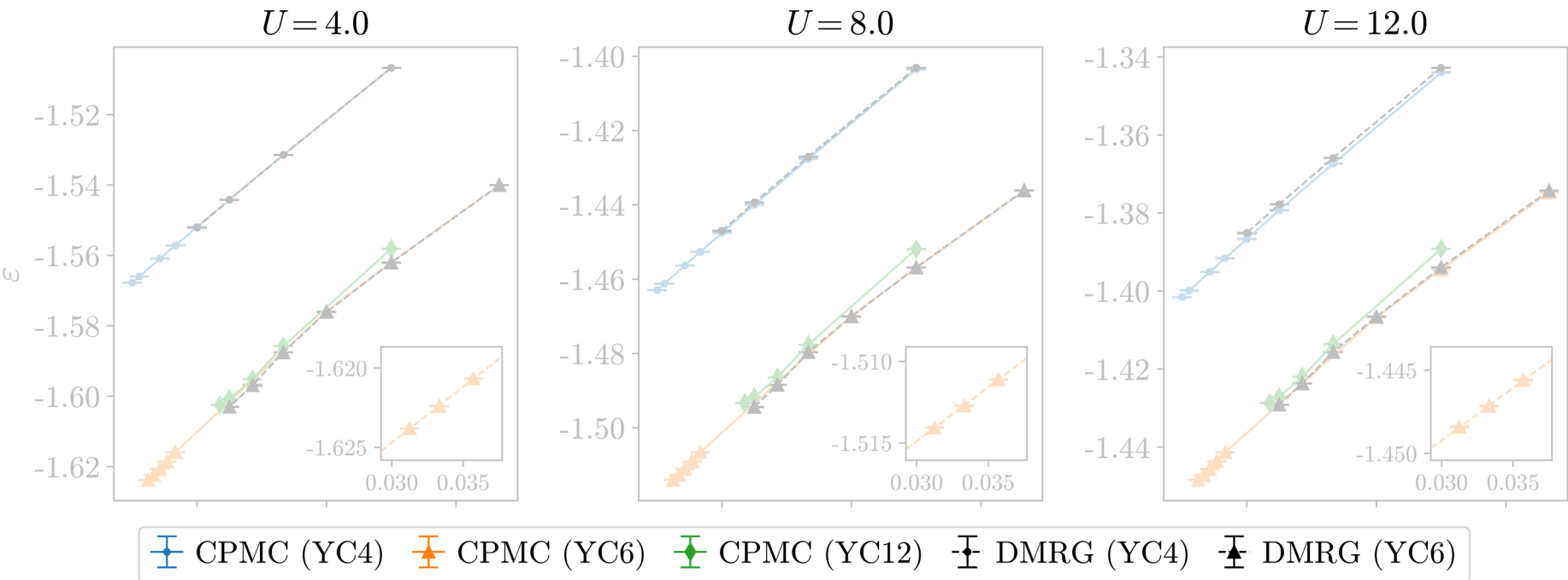


CPMC benchmark results | $v = 1/2$

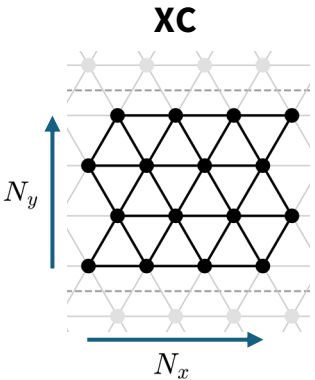
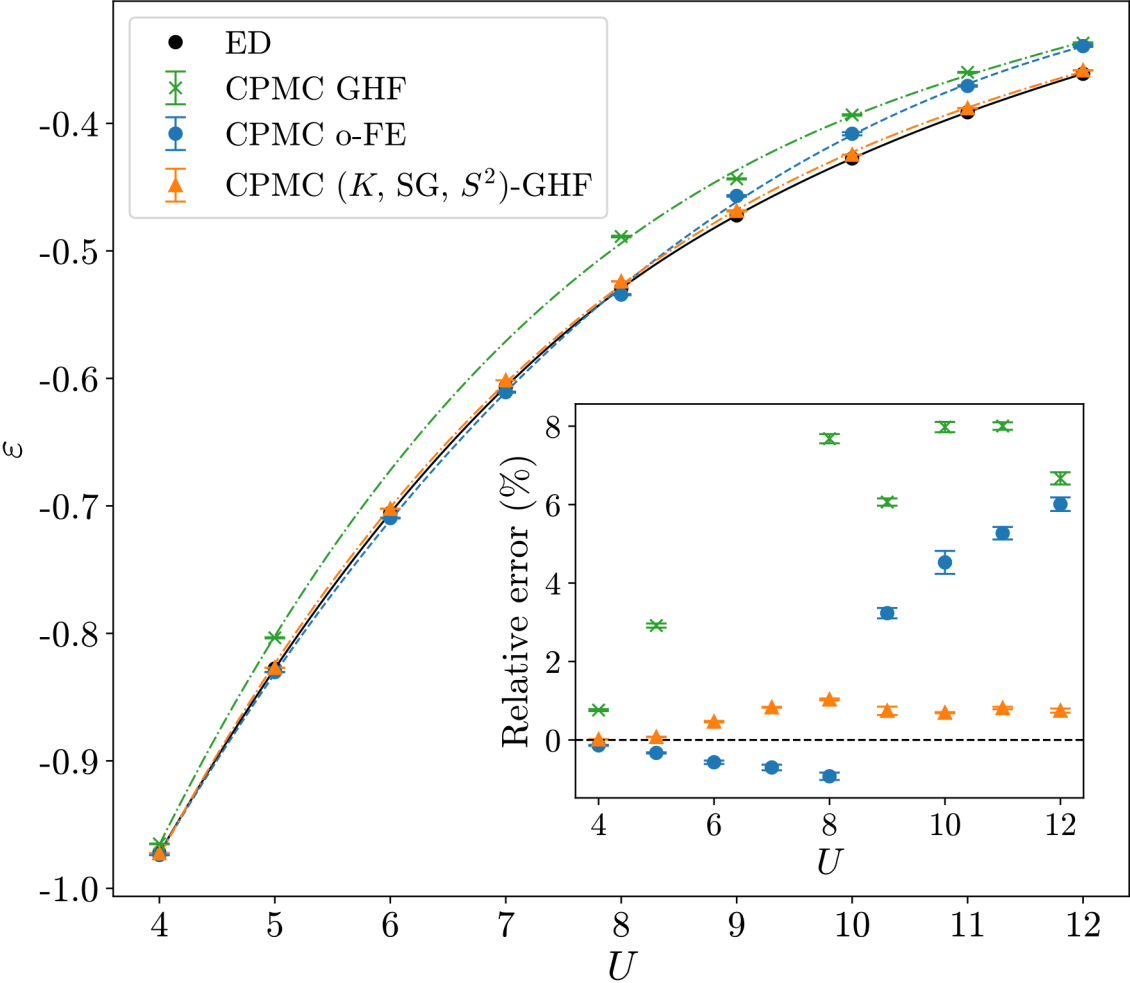
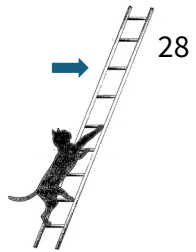


FE trials

Energy per site

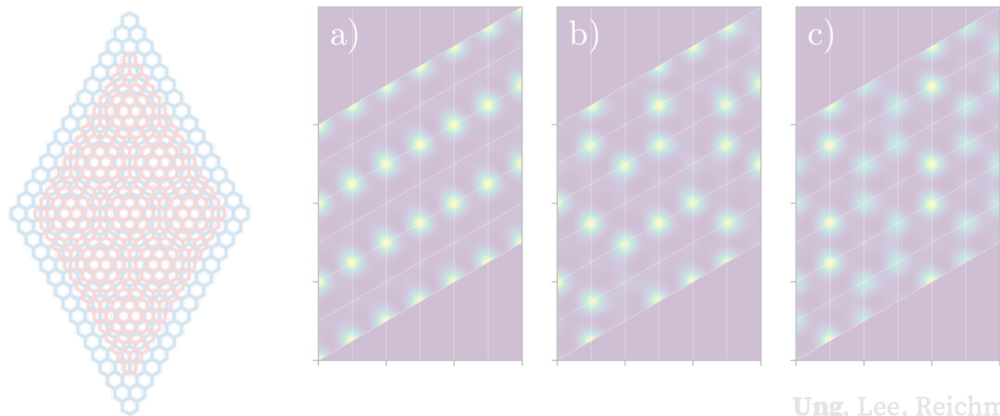


CPMC benchmark results | $v = 1$



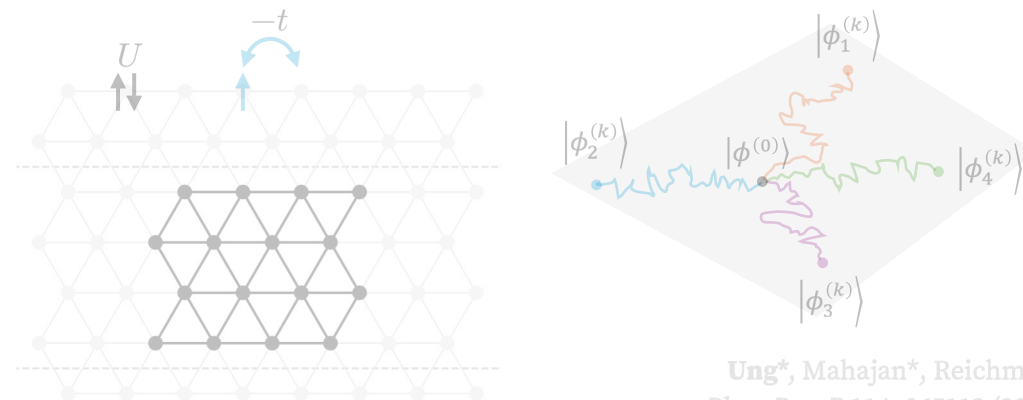
Talk outline

Generalized Wigner crystals in TMD heterobilayers



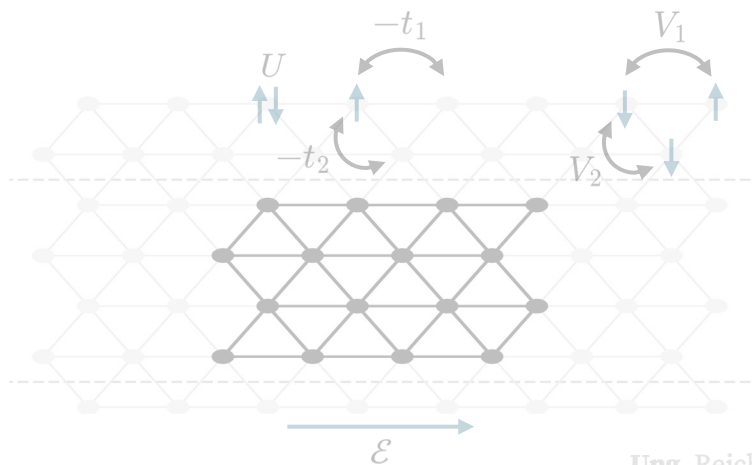
Ung, Lee, Reichman.
Phys. Rev. B 108, 245113 (2023)

Benchmark study of the triangular-lattice Hubbard model with CPMC



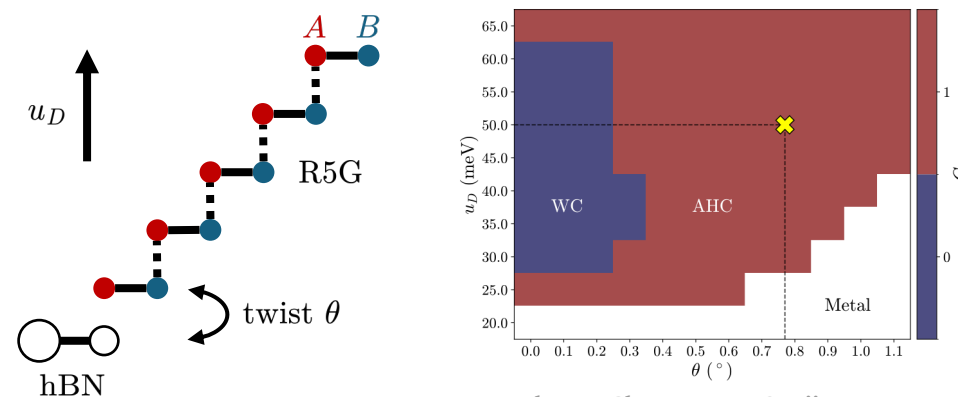
Ung*, Mahajan*, Reichman.
Phys. Rev. B 114, 065112 (2026)

Generalized Wigner crystals under strain



Ung, Reichman. *In prep.*

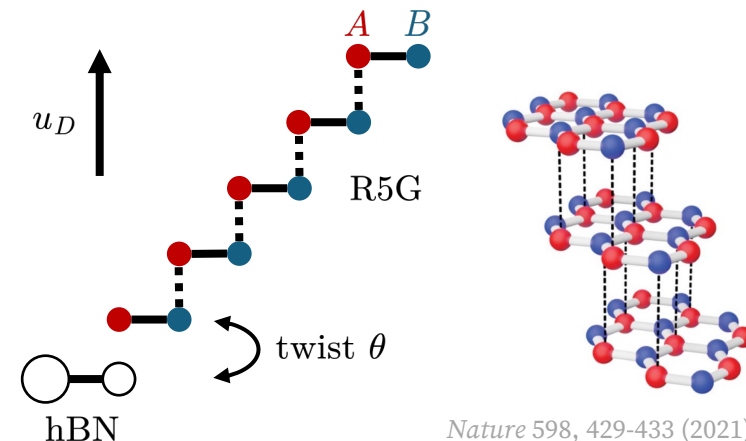
Electron correlation effects in rhombohedral pentalayer graphene/hBN



Ung, Zhang, Chen, Dong, Soejima, Lee. *In prep.*

QAH effect in rhombohedral pentalayer graphene/hBN

- **Quantum *anomalous* Hall (QAH) effect**
 $\Rightarrow$ quantized Hall resistivity without a magnetic field.
- Integer QAH effect at filling $\nu = 1$, consistent with a $|C| = 1$ Chern insulator.
- Fractional QAH effect at fractional ν , consistent with fractional Chern insulators upon partially filling a $|C| = 1$ band.

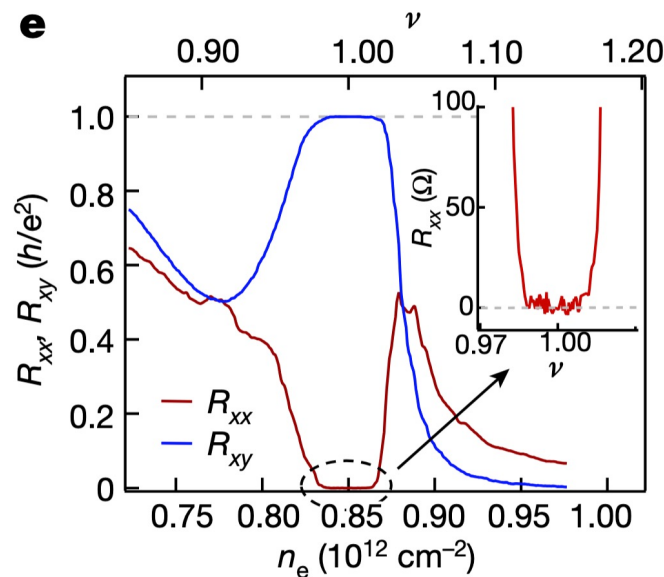


Nature 598, 429-433 (2021)
Phys. Rev. Lett. 133, 206503 (2024)

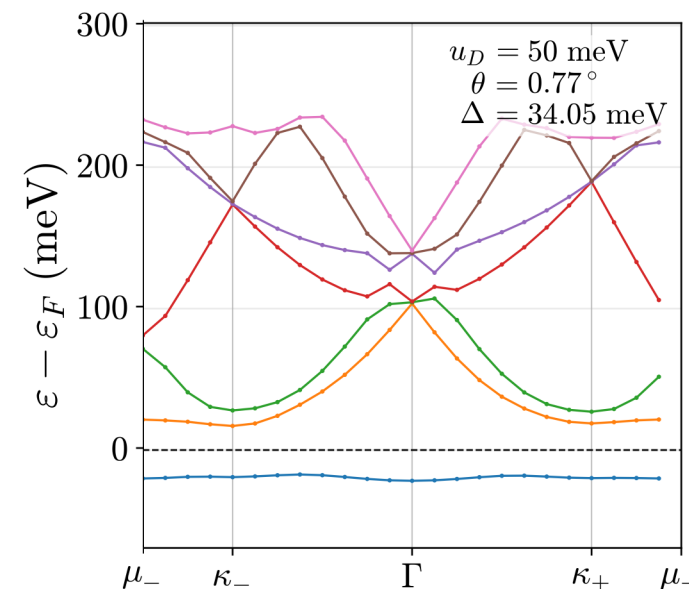
Hall resistivity

$$\rho_{xy} = \frac{h}{e^2} \frac{1}{C}$$

Chern number



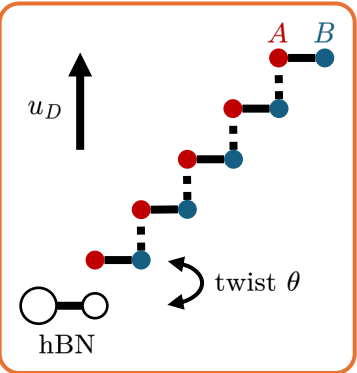
Nature 626, 759-764 (2024)



$|C| = 1$

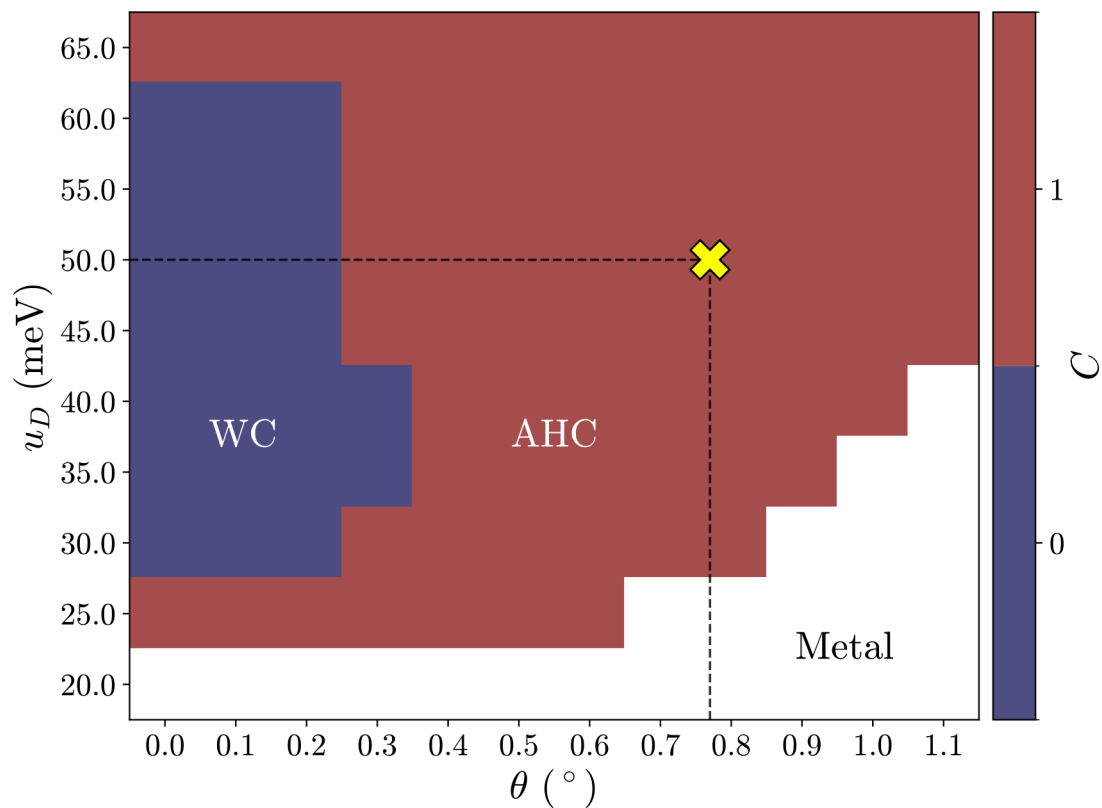
Ung, Zhang, Chen, Dong, Soejima, Lee. *In prep.*

HF phase diagram | $\nu = 1$



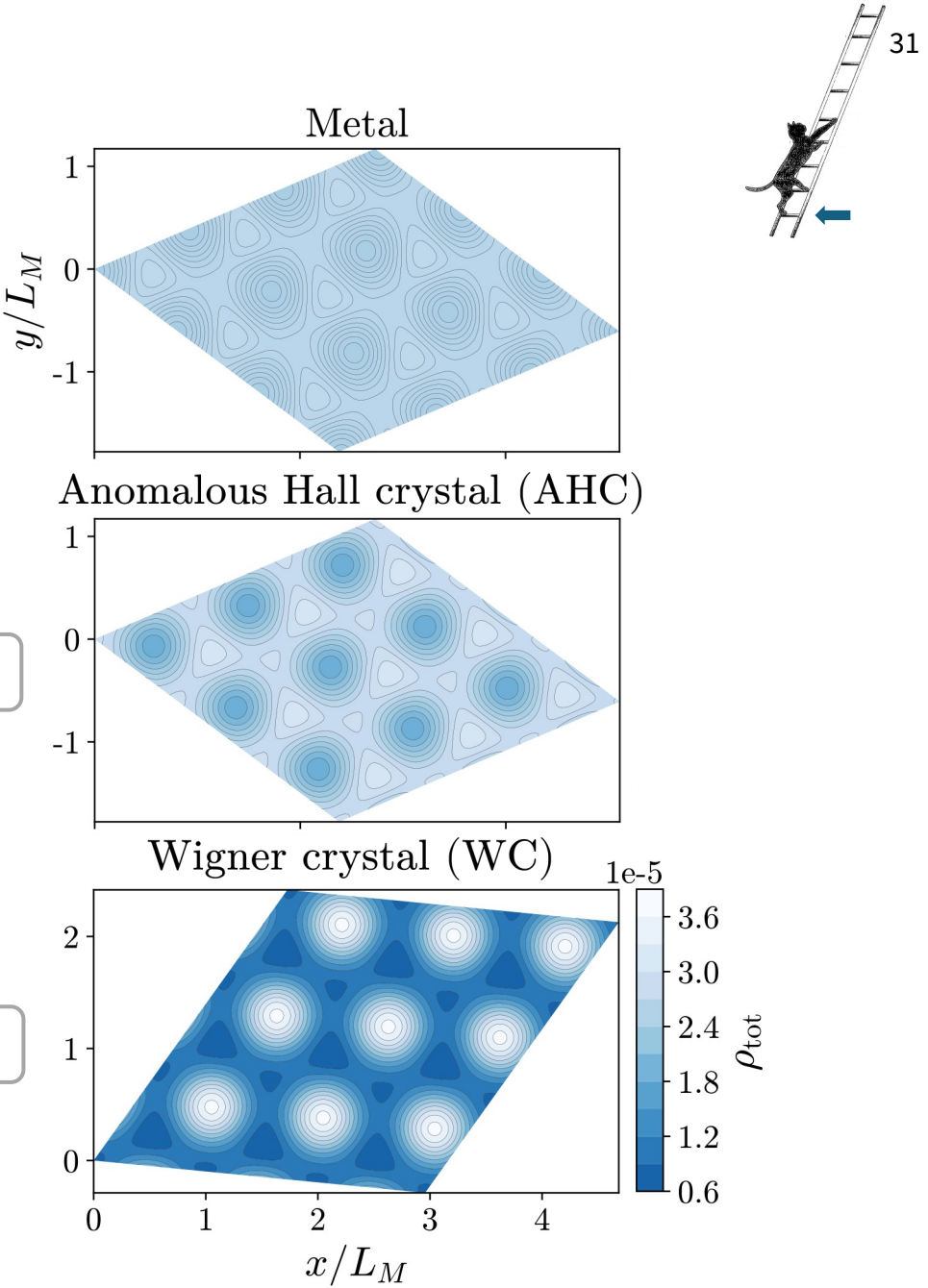
Model

Dong, ..., Soejima *et al.*,
Phys. Rev. Lett. 133, 206503 (2024)

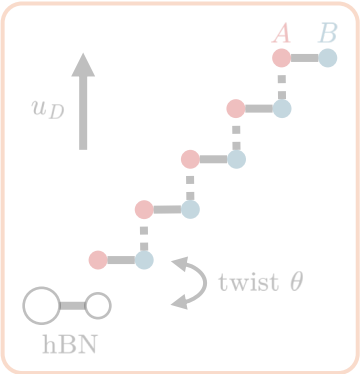


$C = 1$

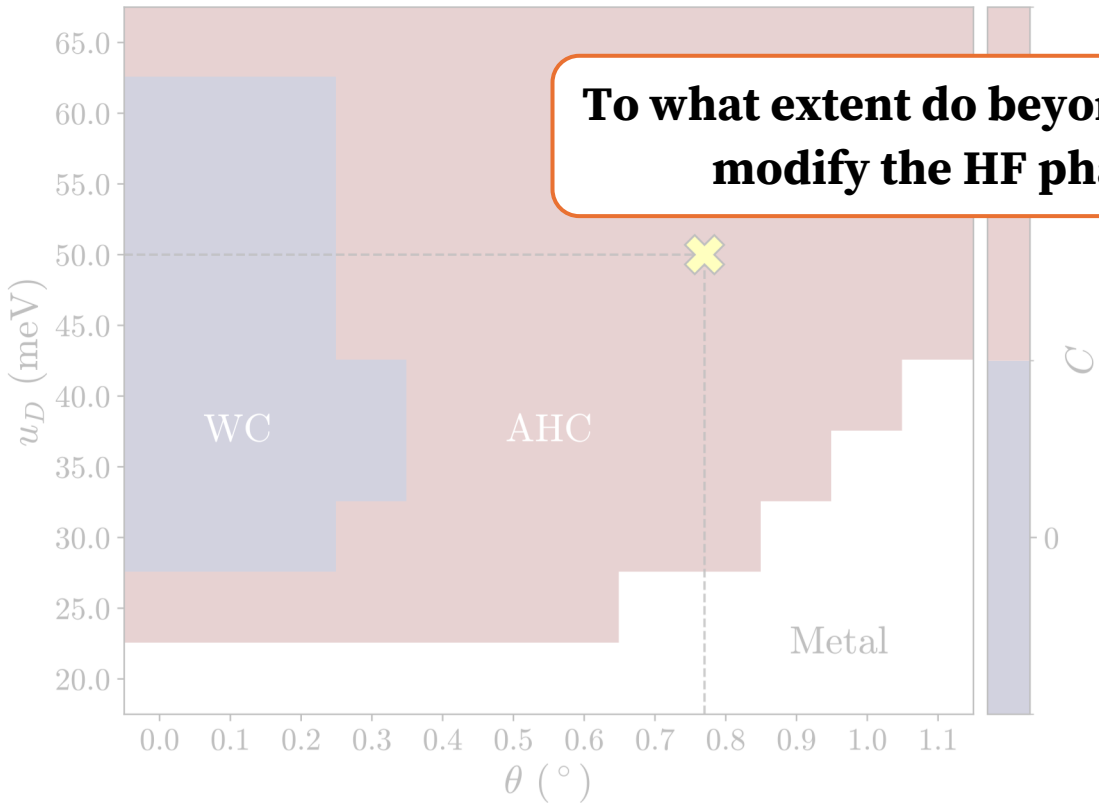
$C = 0$



HF phase diagram | $\nu = 1$

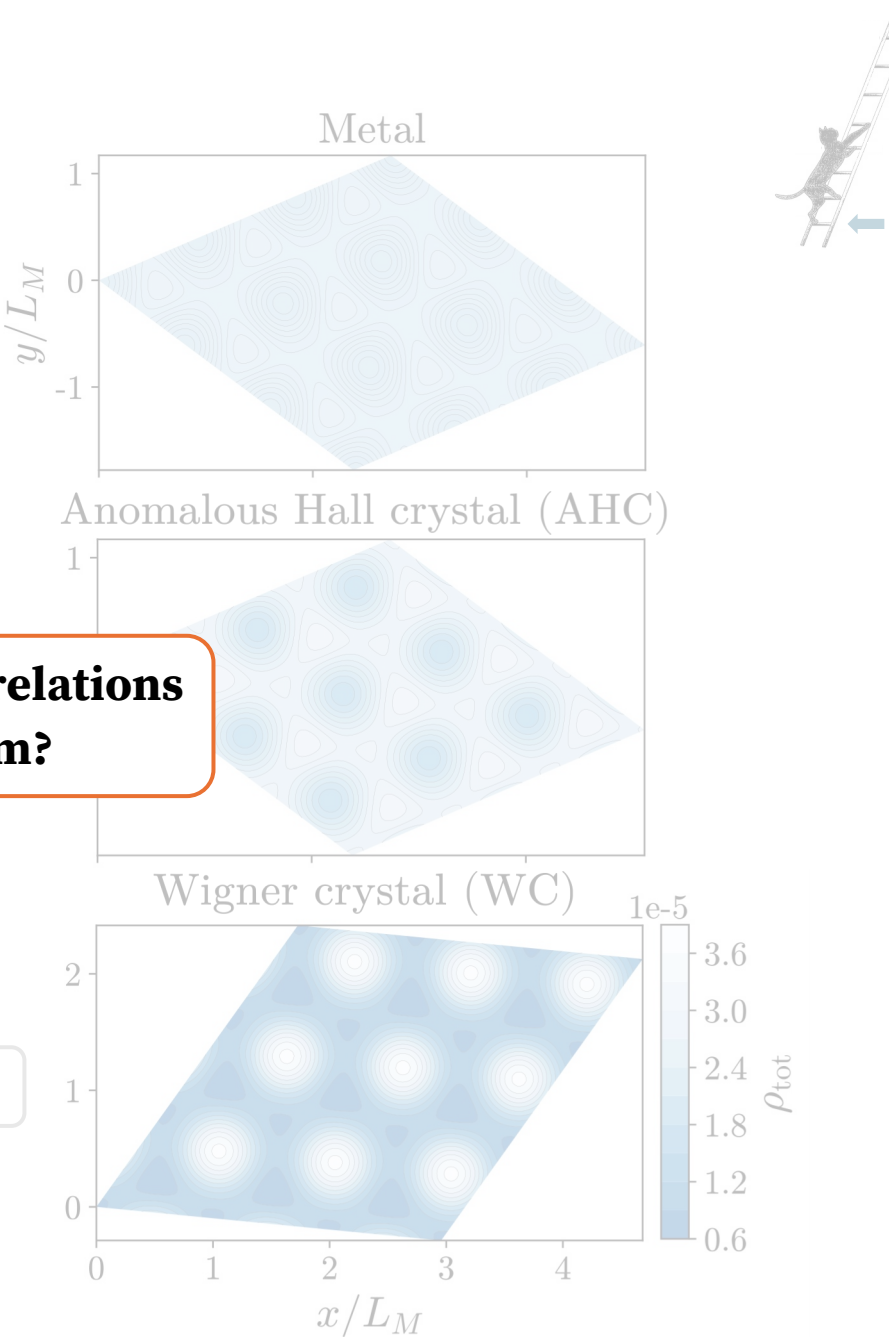


Model
Dong, ..., Soejima *et al.*,
Phys. Rev. Lett. 133, 206503 (2024)

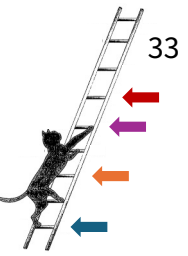


**To what extent do beyond-HF correlations
modify the HF phase diagram?**

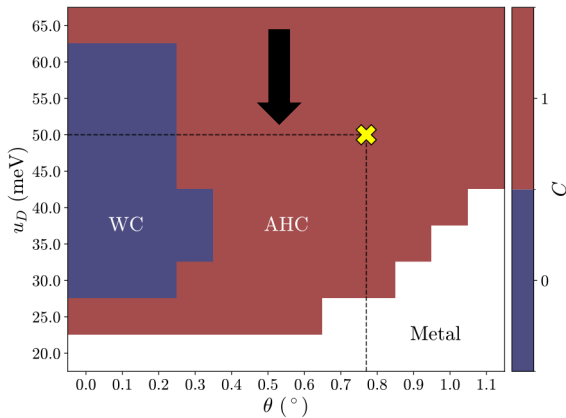
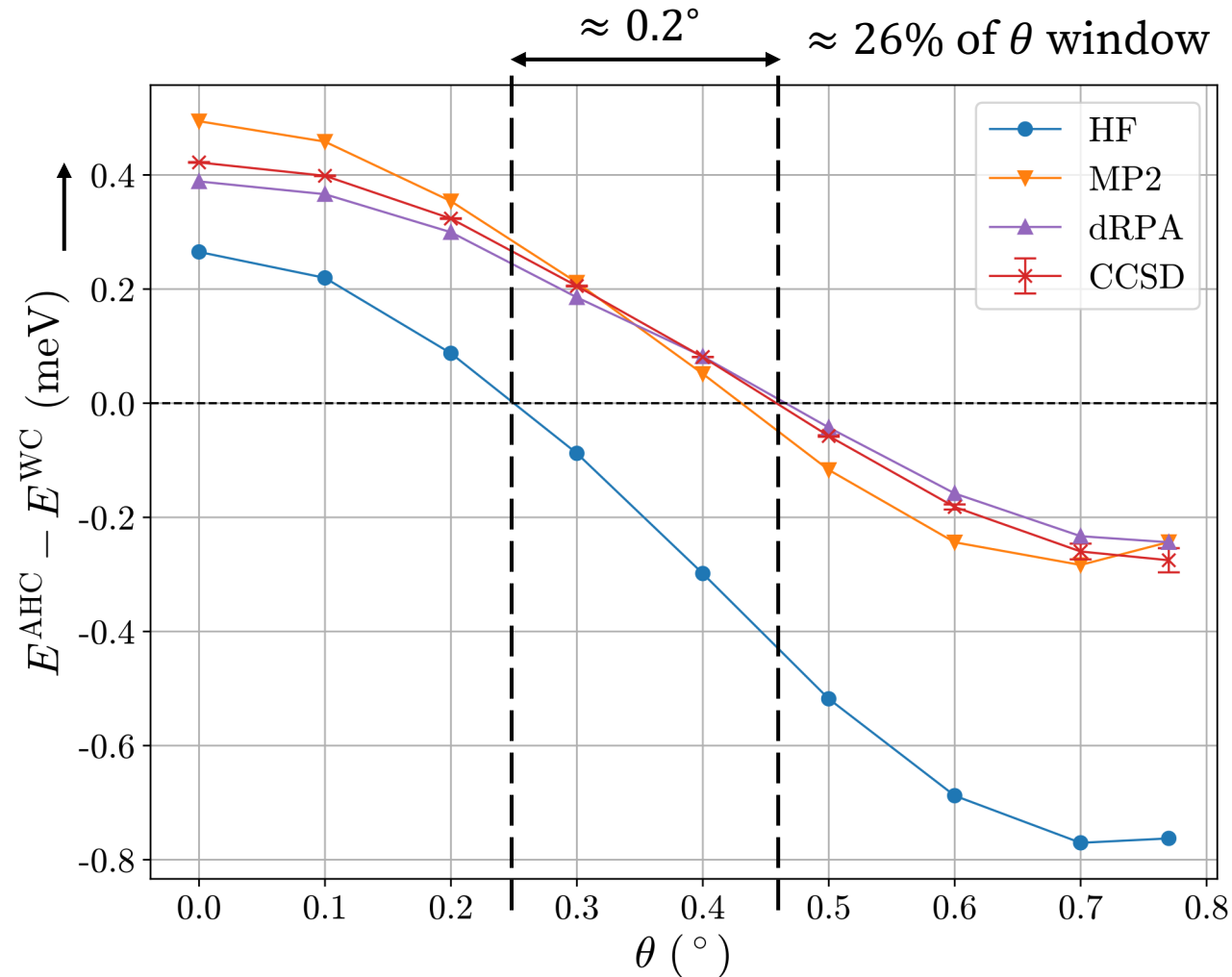
$C = 0$



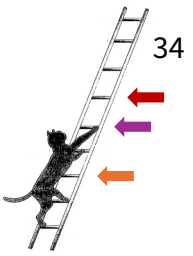
Near the WC–AHC boundary | energy difference



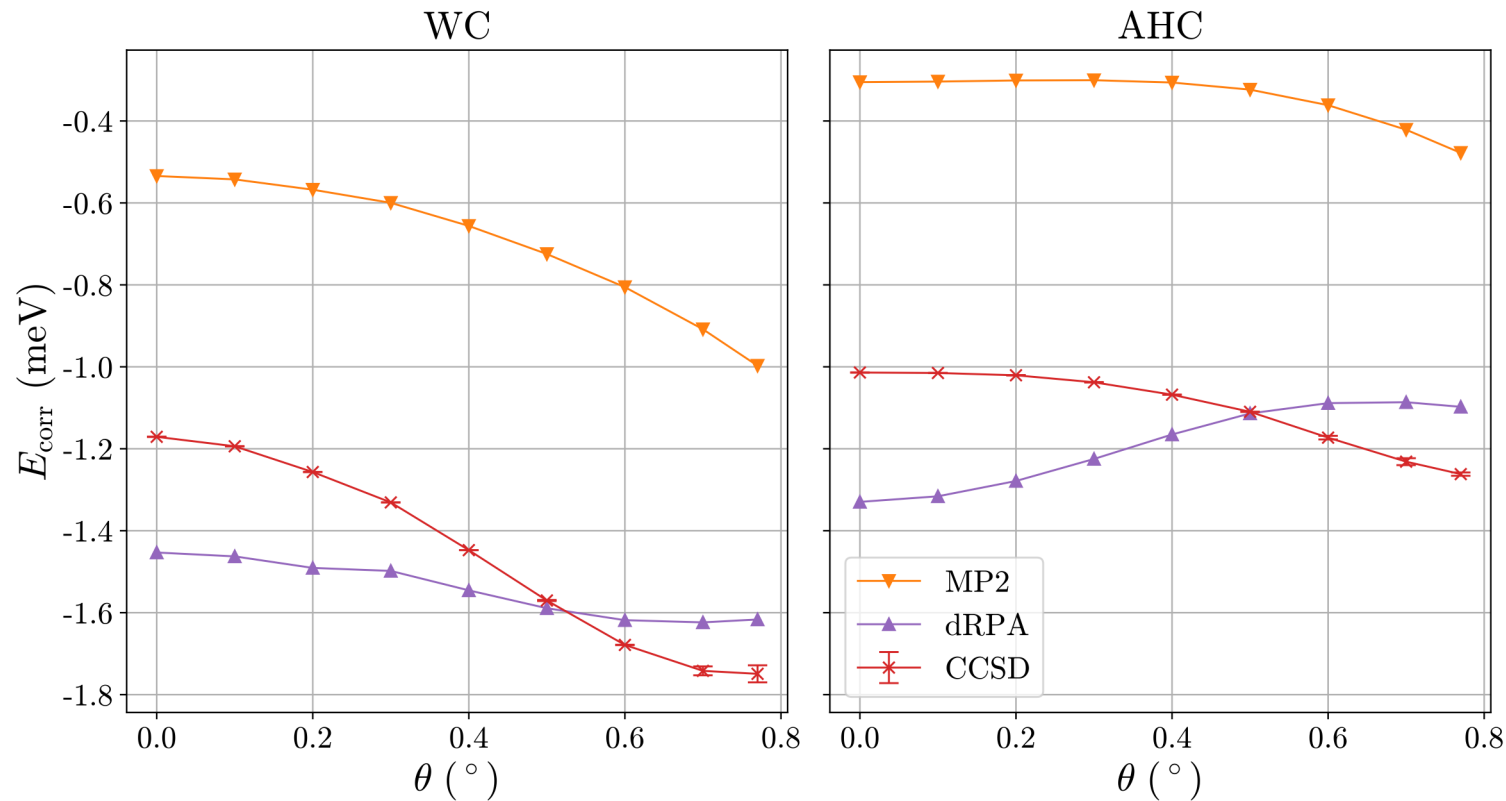
Upward shift due to $E_{\text{corr}}^{\text{AHC}} - E_{\text{corr}}^{\text{WC}} > 0 \Rightarrow |E_{\text{corr}}^{\text{WC}}| > |E_{\text{corr}}^{\text{AHC}}|$.



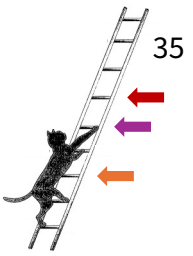
Near the WC–AHC boundary | correlation energy



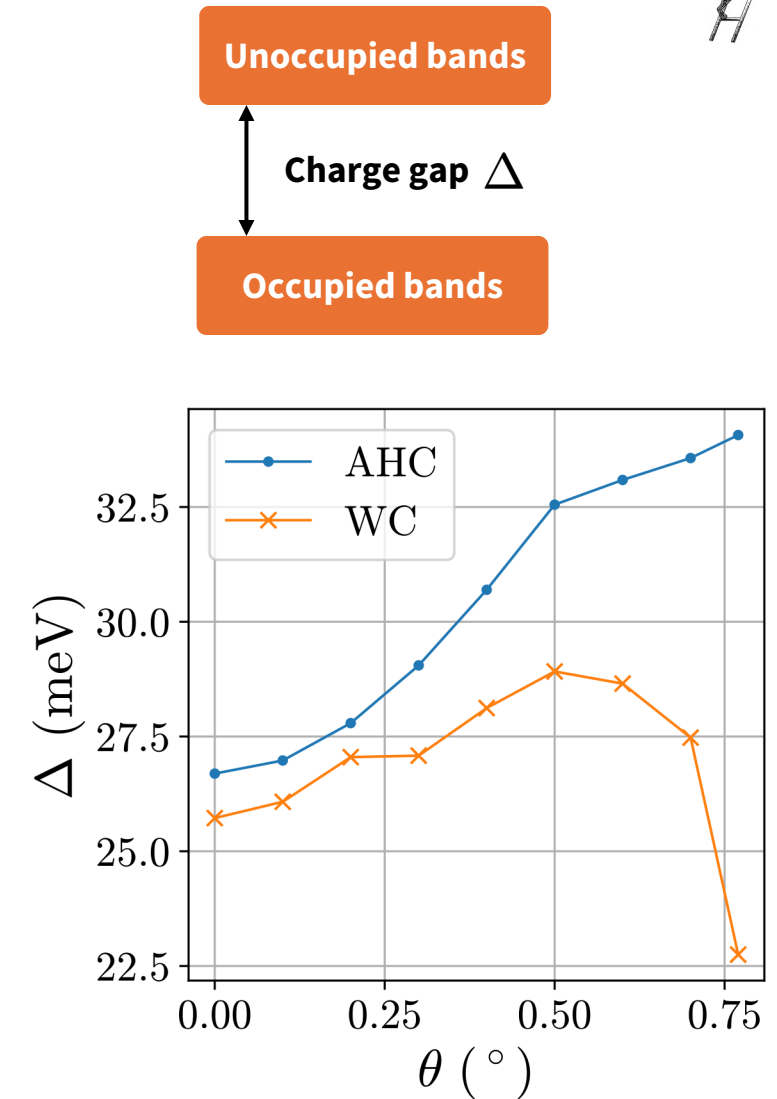
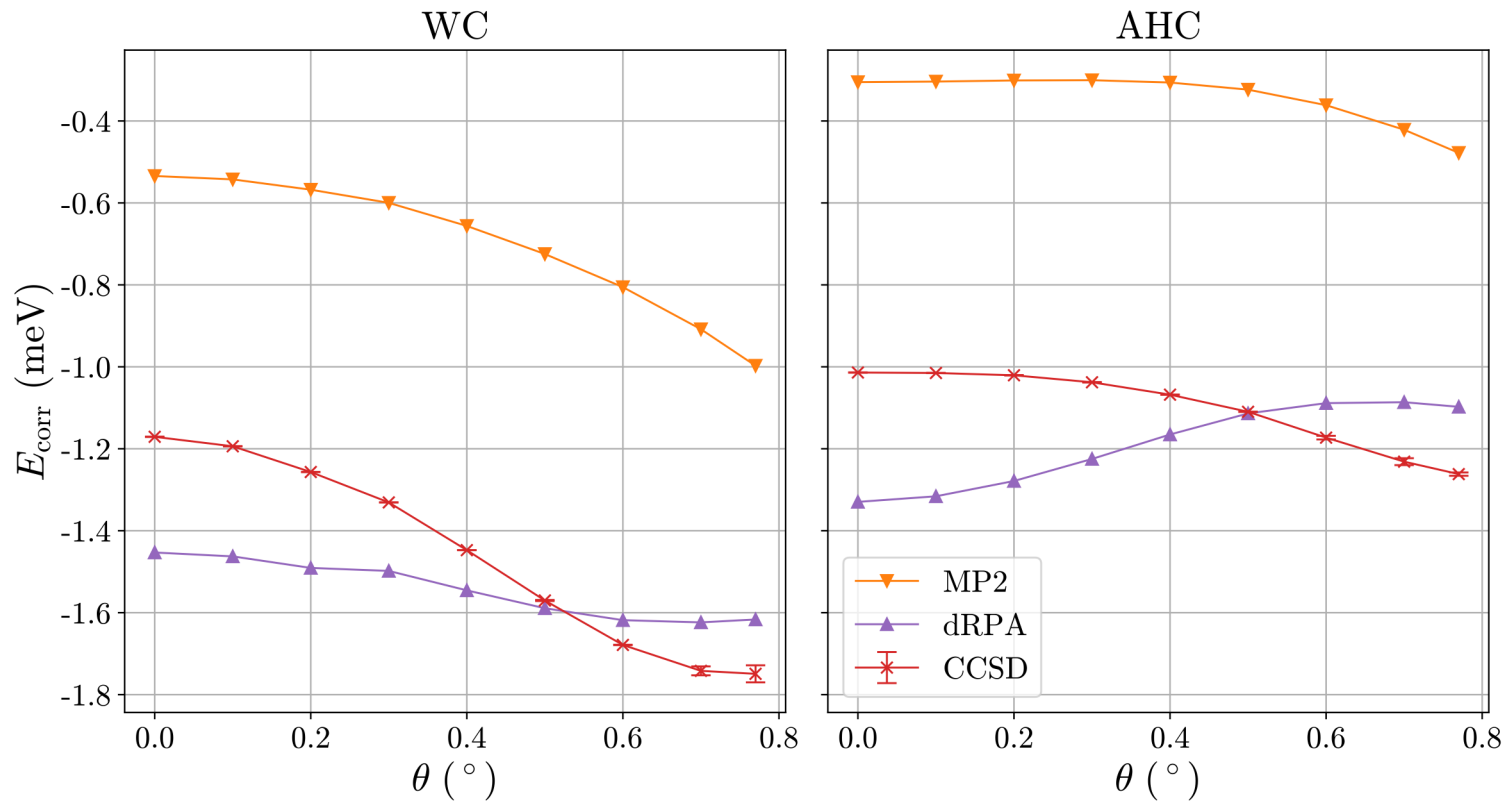
1. Within each correlated method, $|E_{\text{corr}}^{\text{WC}}| > |E_{\text{corr}}^{\text{AHC}}|$.



Near the WC–AHC boundary | correlation energy

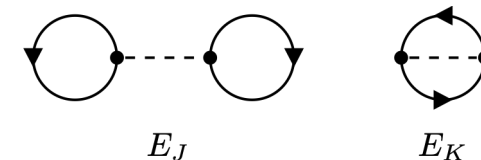


This trend is consistent with the smaller HF charge gaps of the WC, which indicate a **softer charge sector** and **enhanced particle-hole fluctuations**.

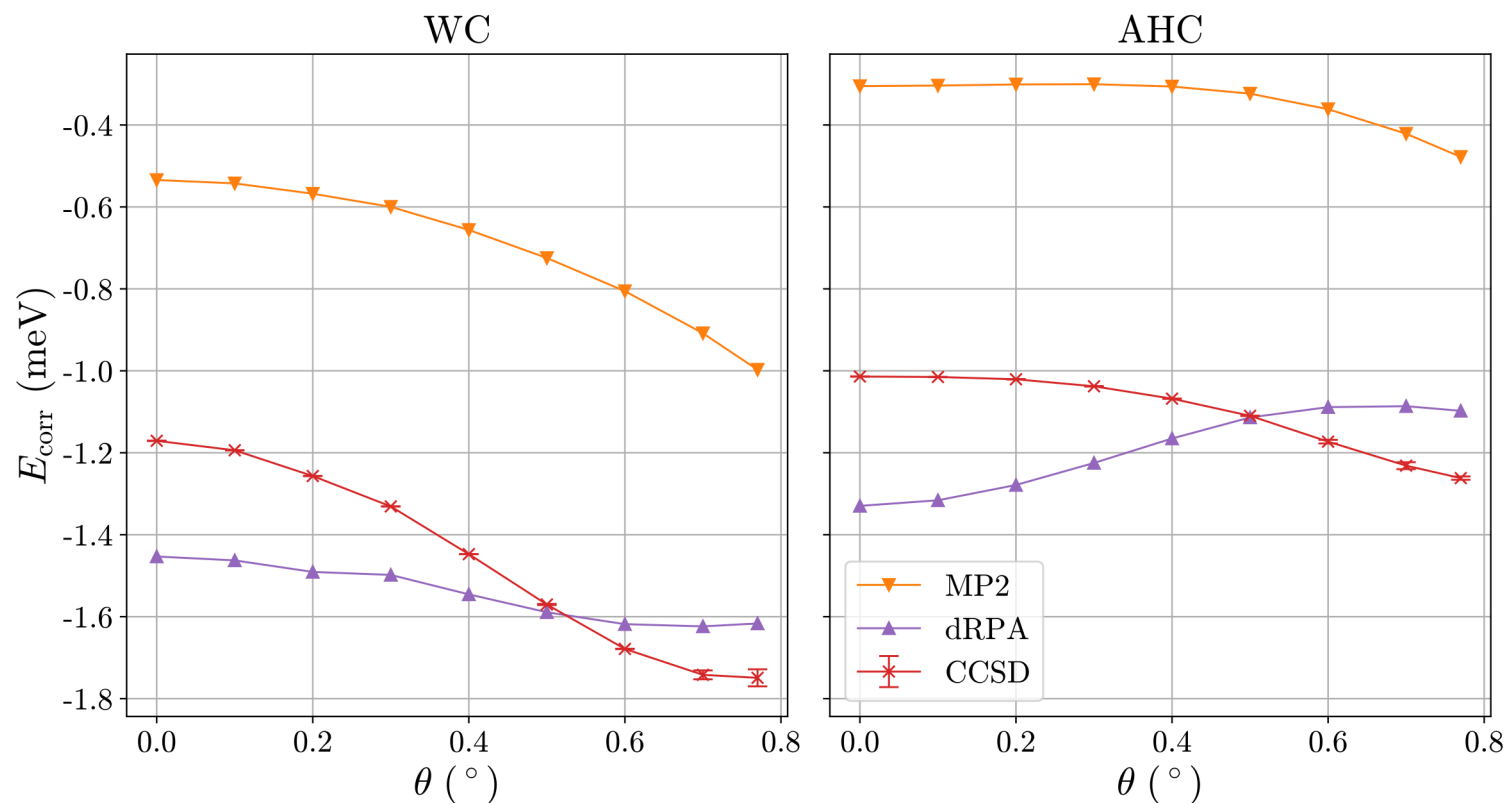


Near the WC–AHC boundary | correlation energy

2. Provides insight into how different quantum fluctuation processes contribute to the correlation energy.

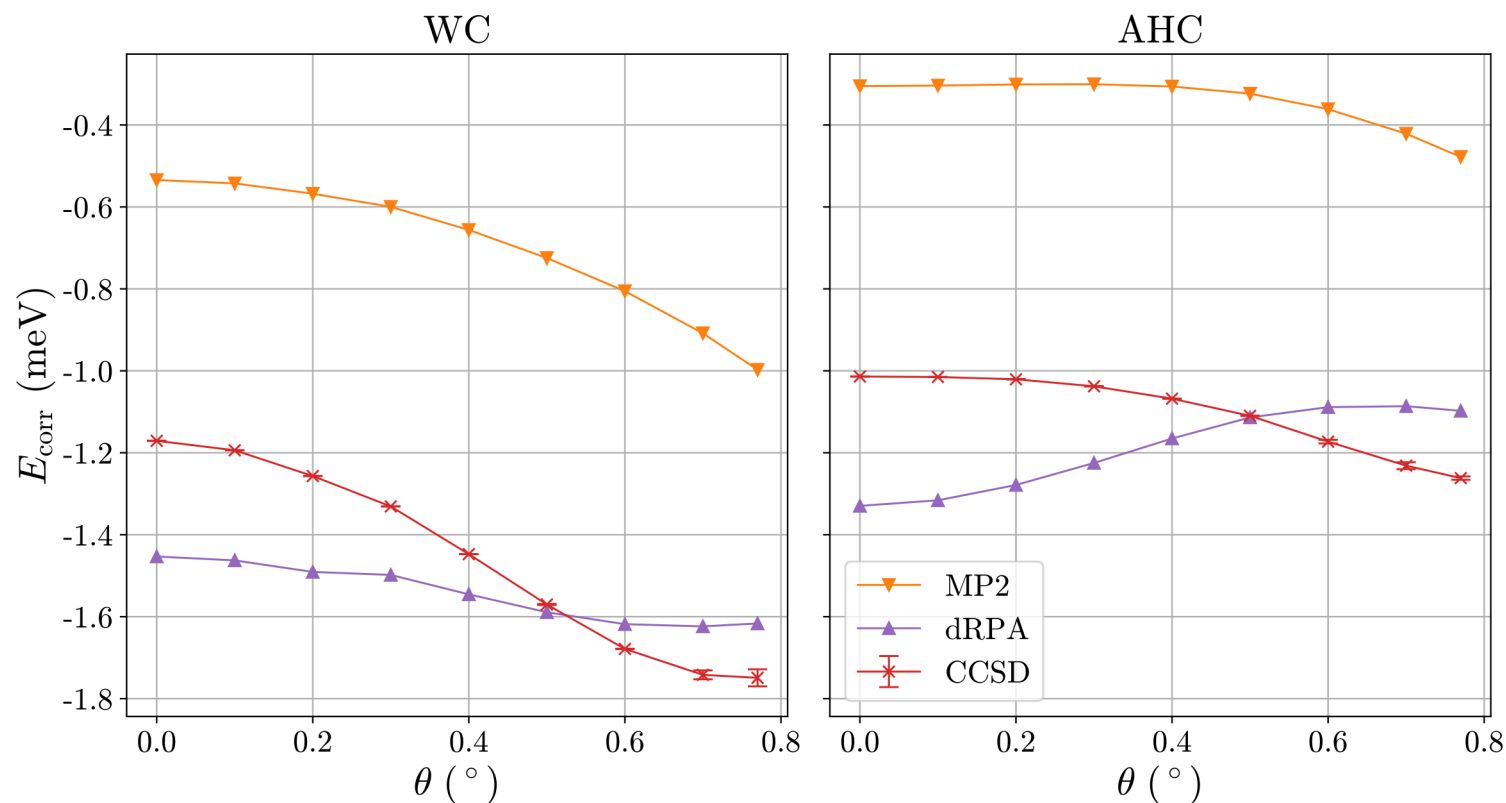
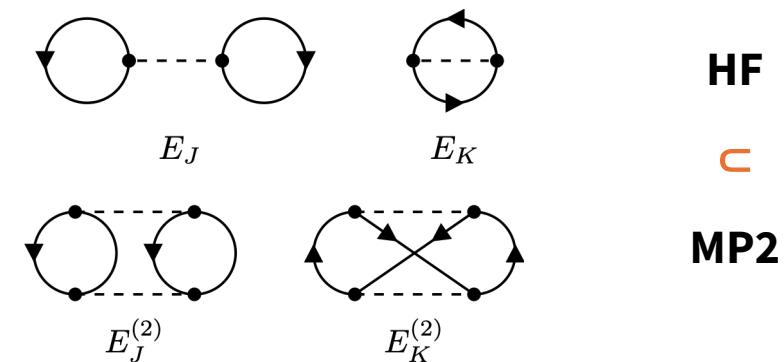


HF



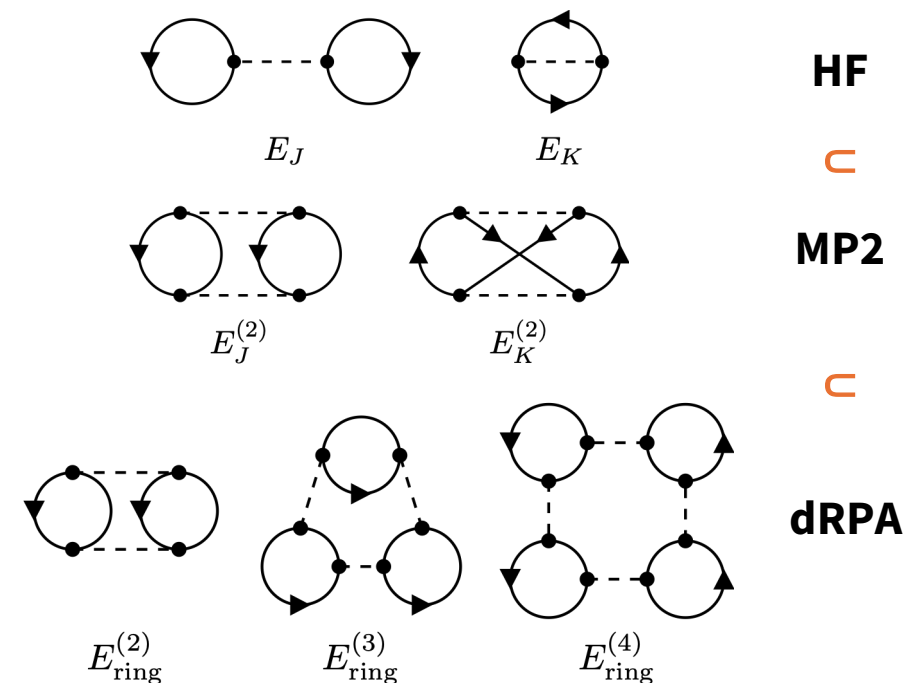
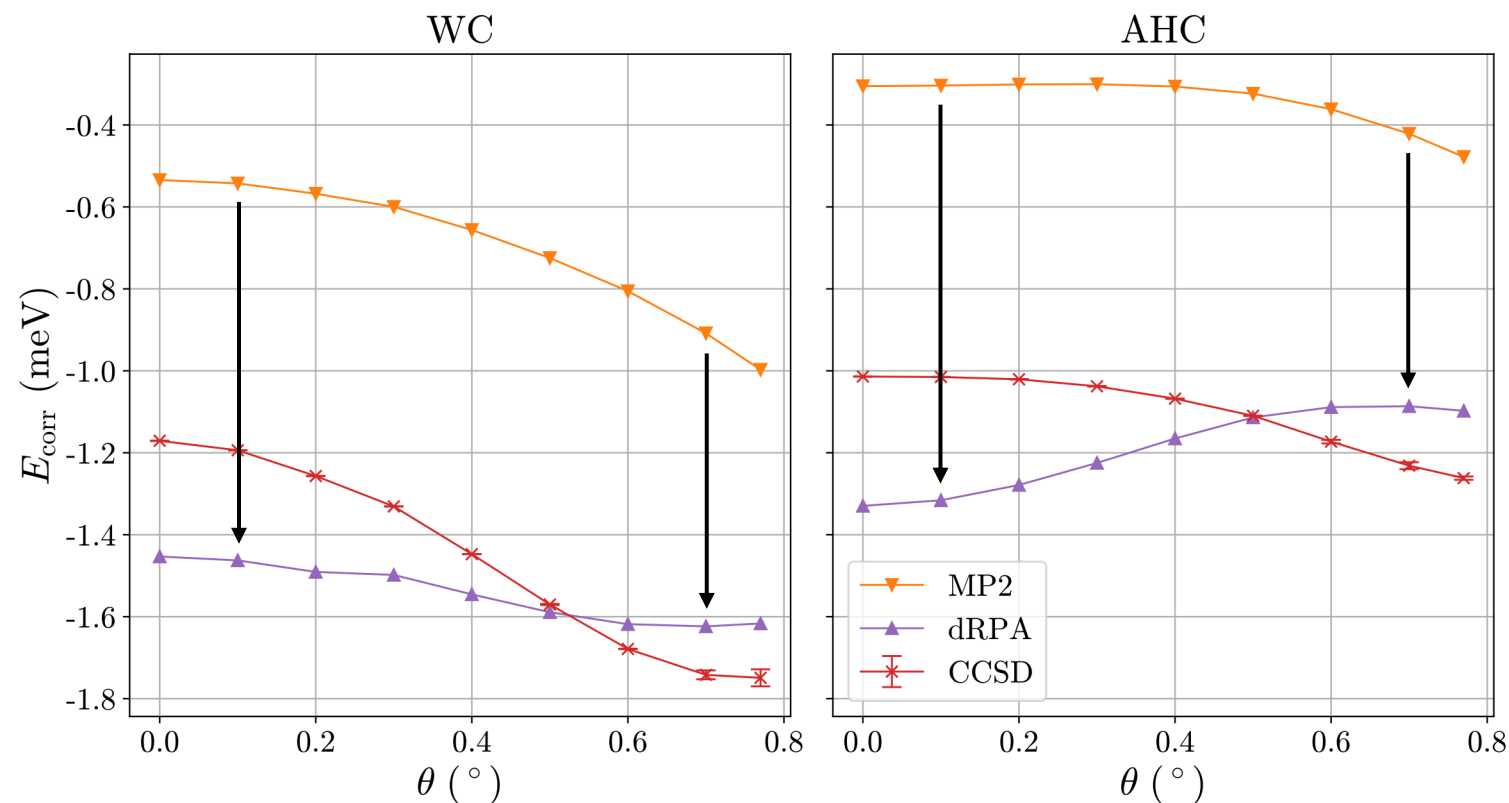
Near the WC–AHC boundary | correlation energy

MP2 gives the lowest-order dynamical correction to HF, which is the **second-order energy** in Rayleigh-Schrodinger perturbation theory.



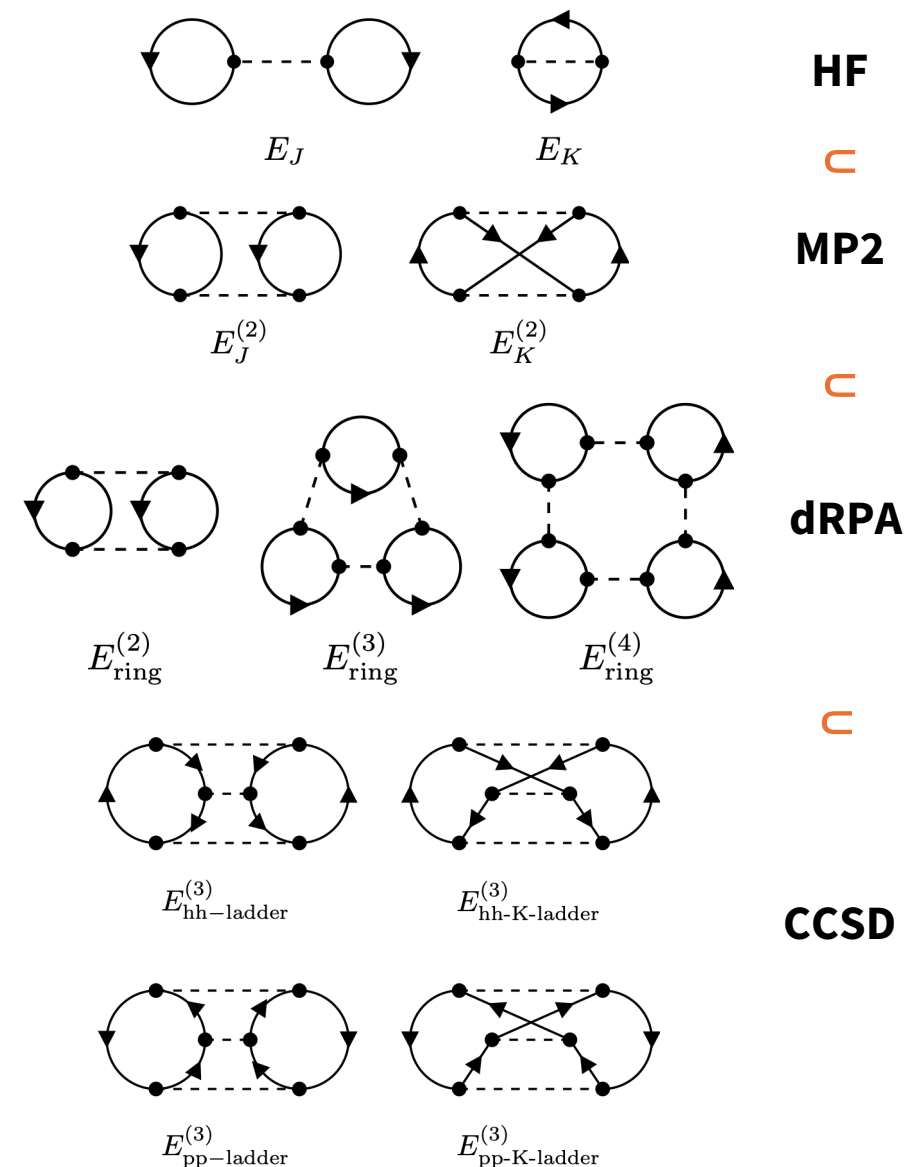
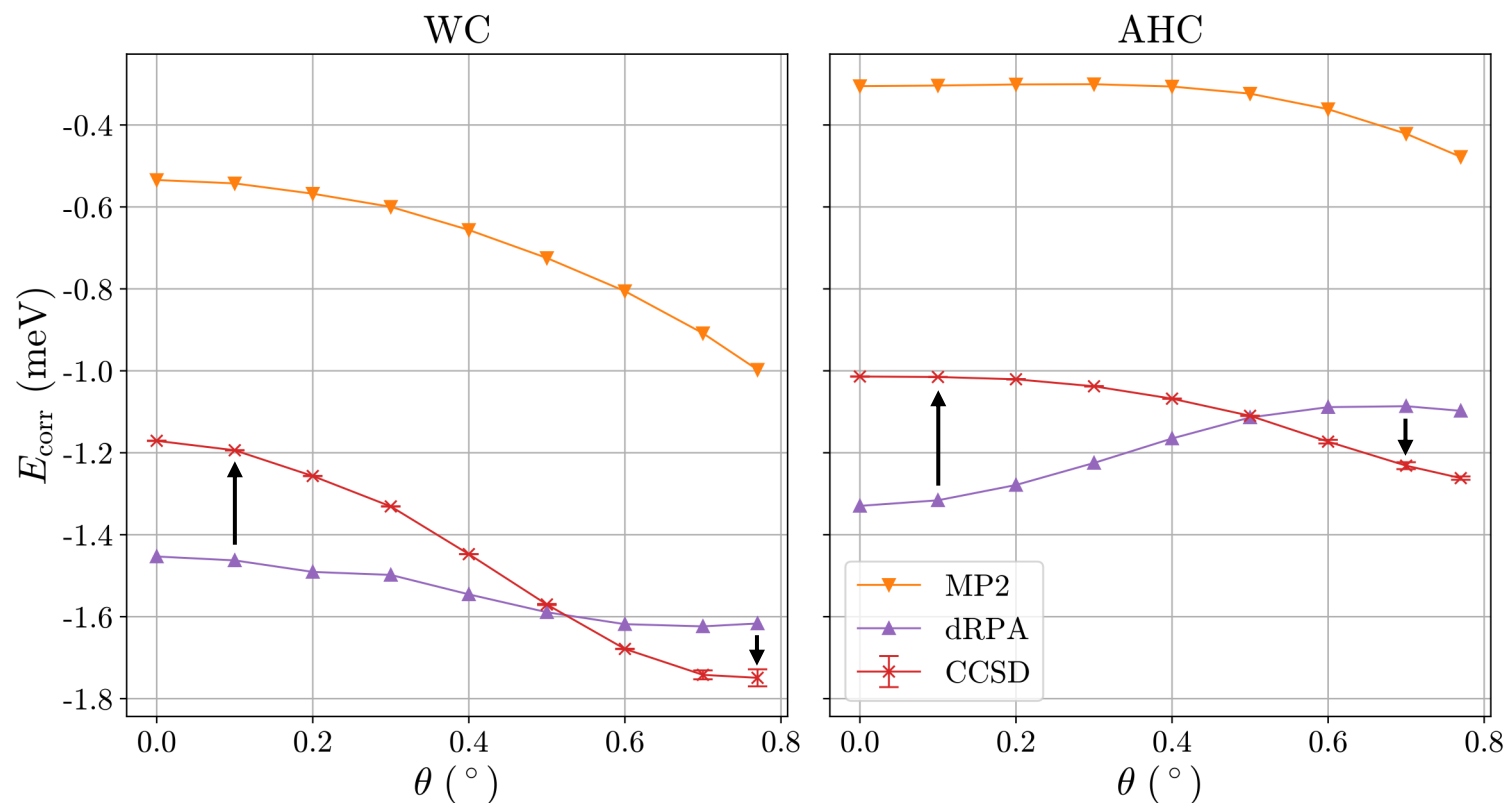
Near the WC–AHC boundary | correlation energy

dRPA ring diagrams introduce **dynamical screening** and energetic contributions from **long-range collective density fluctuations** (*i.e.* plasmons).



Near the WC–AHC boundary | correlation energy

CCSD exchange and particle-particle/hole-hole ladder diagrams capture **higher-order Pauli exchange effects** and **short-range pair correlations**.



Summary

Generalized Wigner crystals in TMD heterobilayers

- The energy landscape is remarkably complex.
- Hartree-Fock can provide a good description of GWCs.
- Correlation energies are small but nontrivial.

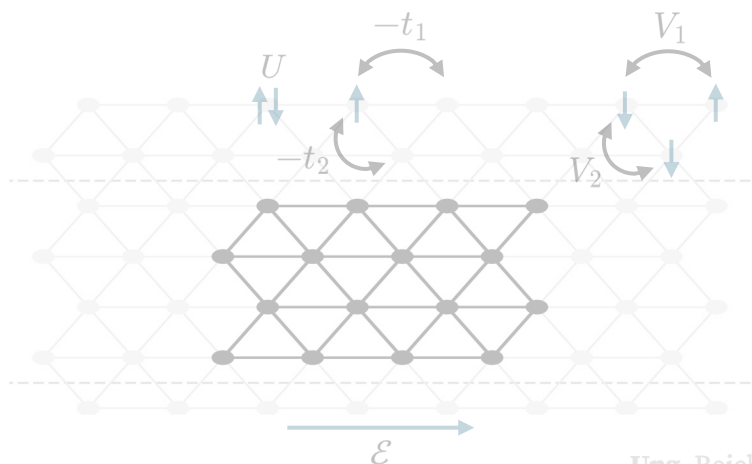
Ung, Lee, Reichman. *Phys. Rev. B* 108, 245113 (2023)

Benchmark study of the triangular-lattice Hubbard model with CPMC

- CPMC can accurately treat this model with strong correlation and geometric frustration.
- Appropriate symmetry-adapted trials are important for reducing constraint bias.

Ung*, Mahajan*, Reichman. *Phys. Rev. B* 114, 065112 (2026)

Generalized Wigner crystals under strain



Ung, Reichman. *In prep.*

Electron correlation effects in rhombohedral pentalayer graphene/hBN

- Small correlation energies destabilize the AHC relative to the WC.
- The WC has a more negative correlation energy than the AHC.

Ung, Zhang, Chen, Dong, Soejima, Lee. *In prep.*

Acknowledgements

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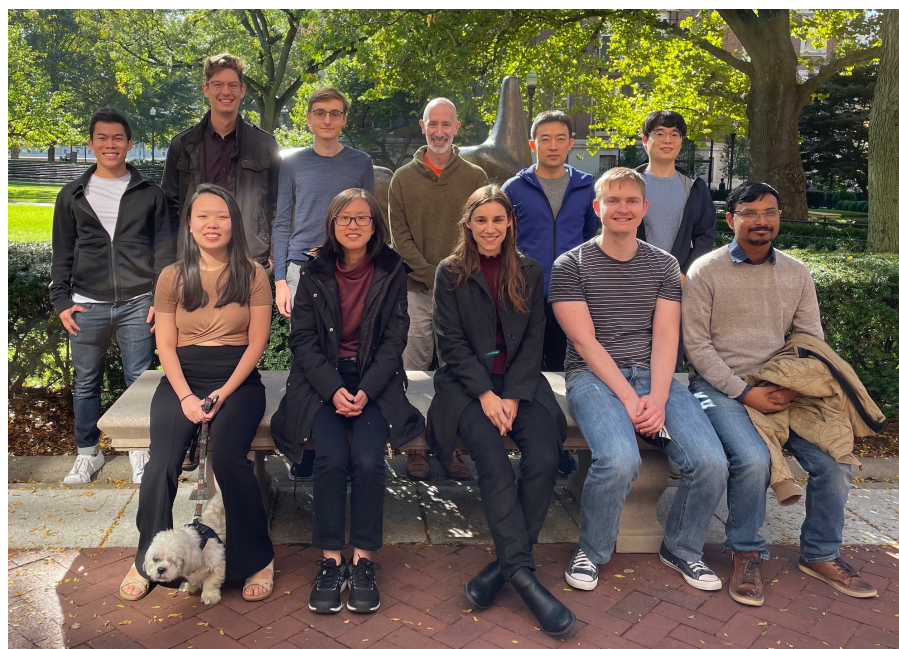
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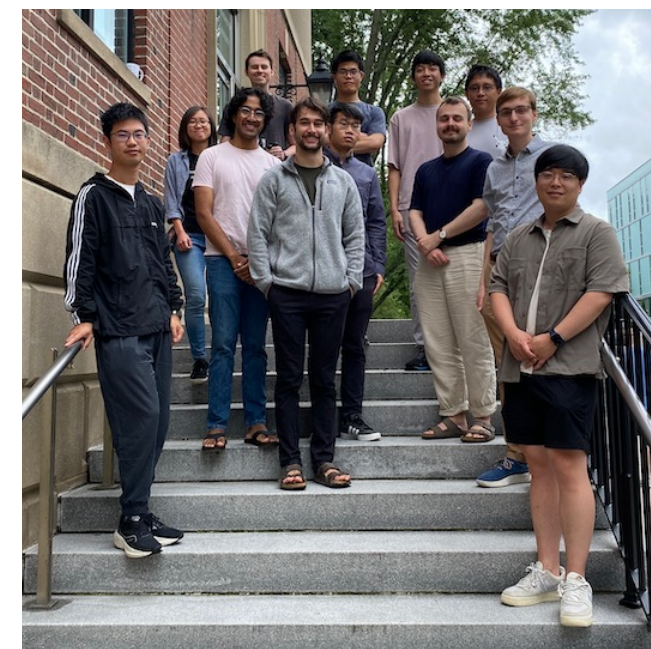
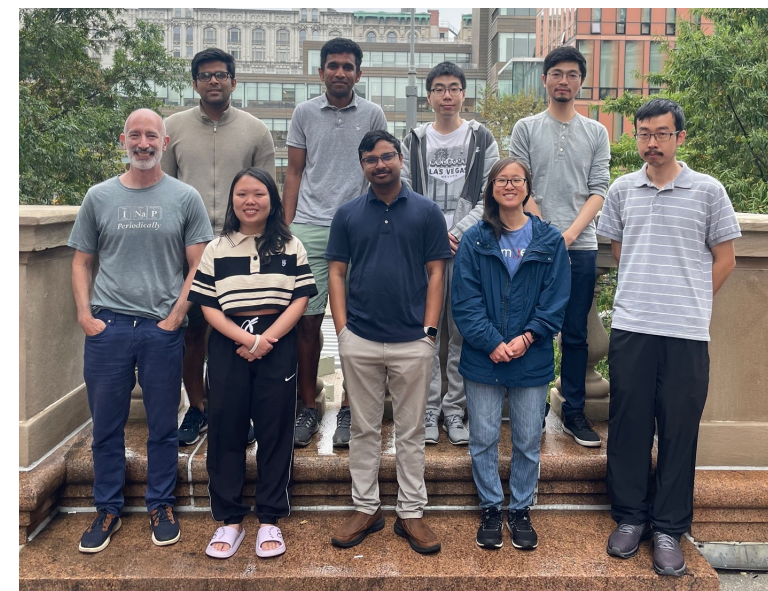
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