Interacting models for magic angle twisted bilayer graphene: a quantum chemistry approach

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New Mathematics for the Exascale:
Applications to Materials Science
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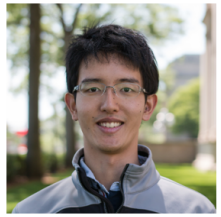

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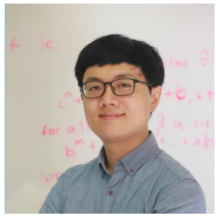

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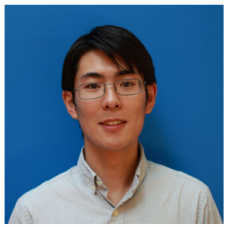

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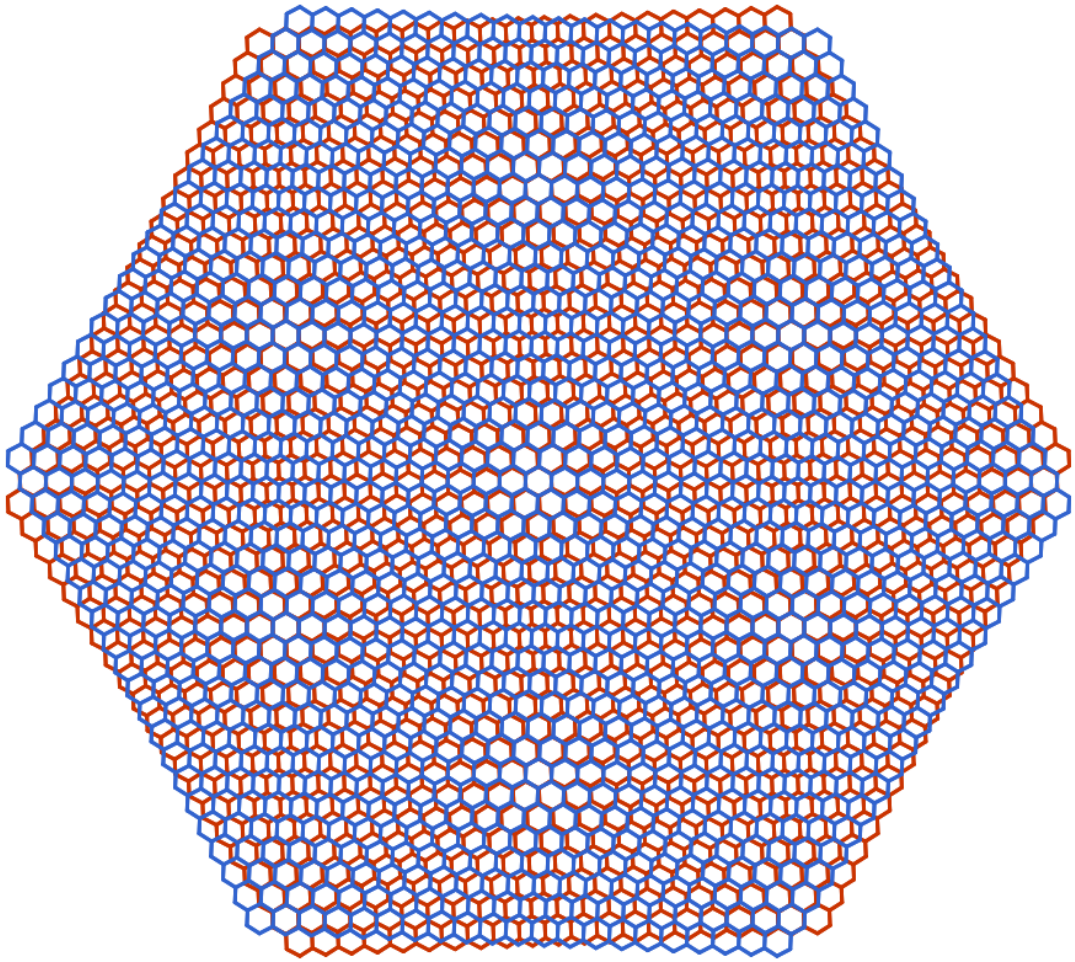
F. M. Faulstich*, K. D. Stubbs*, Q. Zhu*, T. Soejima, R. Dilip, H. Zhai, R. Kim, M. P. Zaletel, G. Kin-Lic Chan, L. Lin, arXiv:2211.09243

Outline

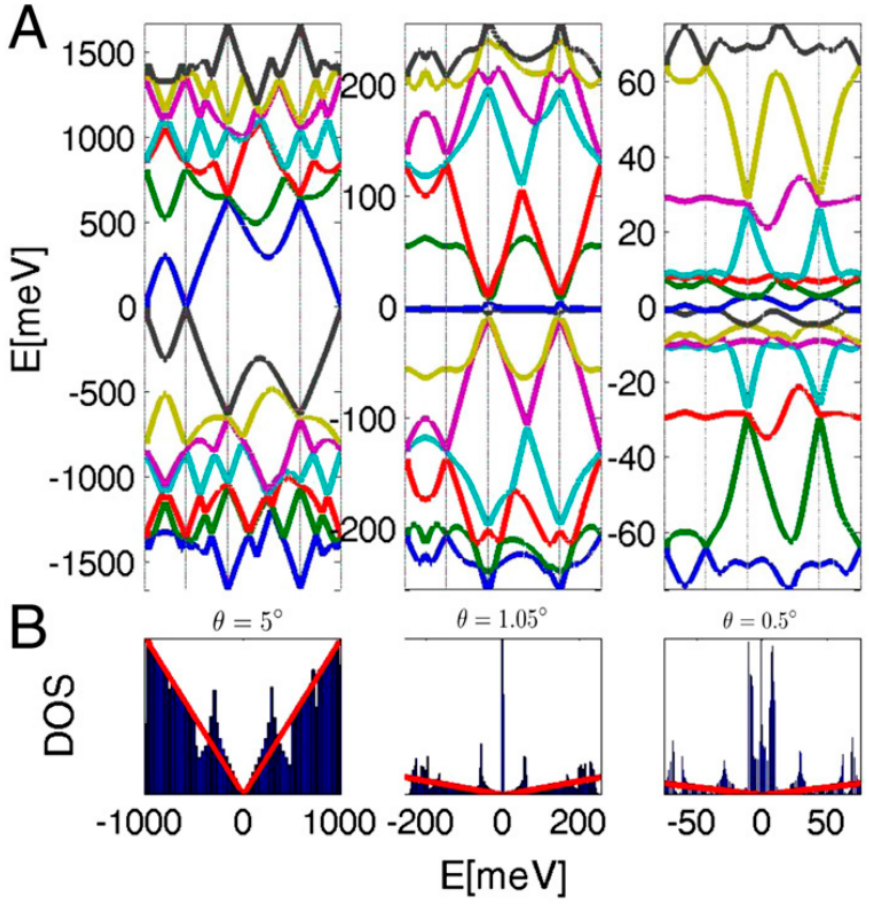
Outline

Twisted bilayer graphene and moiré pattern



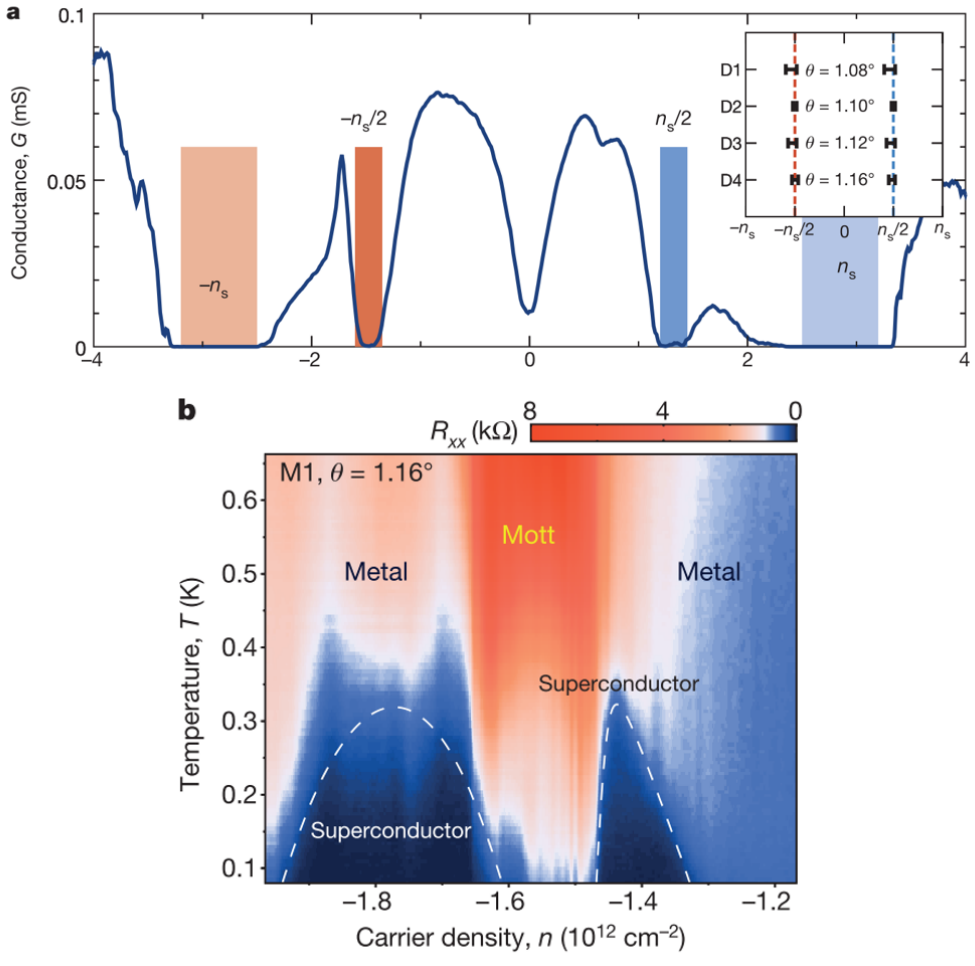
https://everettyou.github.io/2018/05/21/Moire.html

Bistrizter-MacDonald continuum model predicts flatbands at "magic angle" $\theta \approx 1.05^\circ$



(Bistrizter-MacDonald PNAS 2011)

Experimental revelation of superconducting and correlated insulator phases



(Cao et al, Nature 2018)

Wolf Prize in Physics (2020)

Rafi Bistritzer

Pablo Jarillo-Herrero

Allan H. MacDonald

Why “correlated” insulator?

- BM model is a non-interacting model and always predicts zero band gap due to symmetries. So insulating phase must be due to electron interactions.
- “Correlated” in physics often means electron interaction beyond tight binding (including Hartree-Fock). [1]
- Can Hartree-Fock describe the correlated insulating phase?
- For now, postpone the discussion on density functional theory (DFT).

[1] “Correlated” in quantum chemistry often means “post Hartree-Fock”.

Debate on the nature of correlated insulator

- *Possible routes through which a Mott insulator can develop at a low temperature. A specific concrete example is a C_3-broken insulator.*[1]
- *tBLG is more closely related to quantum (anomalous) Hall ferromagnetism, where symmetry breaking is driven by the combination of band topology and Coulomb exchange, than it is to the Mott insulating physics of the Hubbard model.*[2]
- Common goals: 1) opening of a band gap at integer filling 2) detecting spontaneous symmetry breaking[3]

[1] Po et al, PRX 2018

[2] Xie, MacDonald, PRL 2020; Soejima et al, PRB 2020

[3] For spontaneous symmetry breaking, also see Zhang, MacDonald, PRL 2012; Shi, Zhu, MacDonald, PRB 2021..

A class of “interacting” BM model (IBM)

Many tight-binding level models from 2018-2020. Origin of flatbands, geometry relaxation, band topology and Wannier functions, phenomenological phase diagrams etc.

From 2020, interacting models:

- (Koshino et al, PRX 2018): Extended Hubbard (no mean field calculation)
- (Bultinck et al, PRX 2020): Extended Hubbard. HF phase diagram, integer filling
- (Soejima et al, PRB 2020): Extended Hubbard (spinless, valleyless). HF and DMRG, integer filling, hybrid Wannier localization
- (Xie, MacDonald, PRL 2020): Extended Hubbard. HF.
- (Pan, Wu, Das Sarma, PRB 2020): HF, extended Hubbard
- (Kang, Vafek, PRB 2020): Extended Hubbard. DMRG, integer filling, hybrid Wannier localization
- (Hejazi et al, PRR 2021): Extended Hubbard.
- (Liu, Dai, PRB 2021): Extended Hubbard.
- (Bernevig, Lian, Xie et al, PRB 2021 I-VI): Theoretical study. Extended Hubbard. HF.
- (Zang et al, 2021): DMFT. Standard Hubbard on triangular lattice, parametrized for tMDC.
- (Romanova, Vlcek, npj Comput. Mater. 2022): Full ab initio. GW and frequency-dependent interaction.
- ...

Literature converged to some extended Hubbard model / IBM model?

Contribution of this work

Simplified spinless, valleyless model [1]

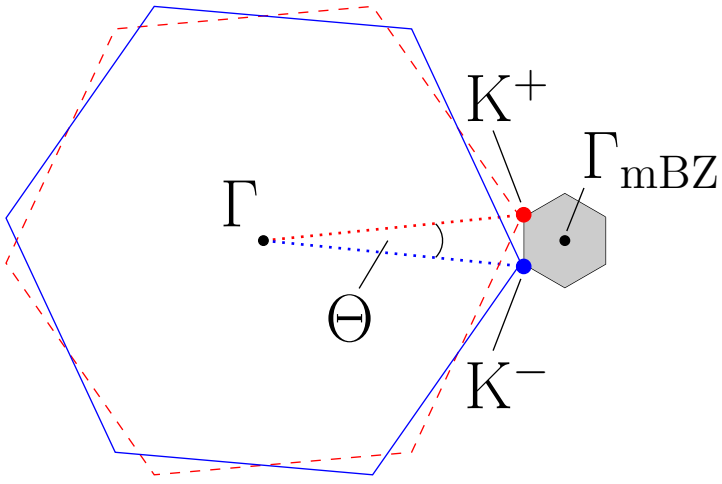
- Quantum chemistry methods beyond Hartree-Fock (coupled cluster, QC-DMRG). More scalable than exact diagonalization.
- Integer and non-integer fillings.
- Gauge invariant order parameter for symmetry breaking.
- Model discrepancy.

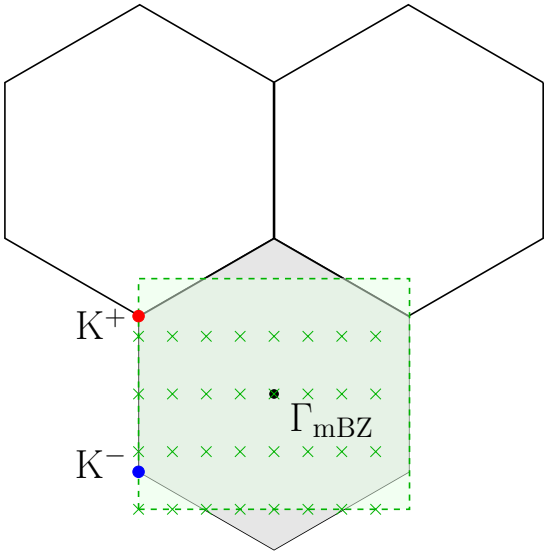
[1] Exhibits phenomena not captured by the corresponding single particle models see (Soejima et al Phys. Rev. B 2020)

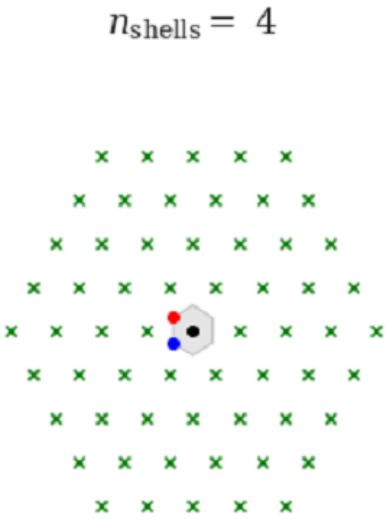
F. M. Faulstich, K. D. Stubbs, Q. Zhu, T. Soejima, R. Dilip, H. Zhai, R. Kim, M. P. Zaletel, G. Kin-Lic Chan, L. Lin, arXiv:2211.09243

Outline

Bistrizter-MacDonald (BM) continuum model







- Focuses on the **K** valley (**k** is now relative to Γ_{mBZ}). $\Omega^* = \text{mBZ}$.
- $\hat{c}^\dagger_{\mathbf{k}}(\mathbf{G}, \sigma, l)$: **G**: planewave; σ: sublattice. l: layer.
- Diagonalize BM Hamiltonian, obtain flatbands $\{u_{n\mathbf{k}}(\mathbf{G}, \sigma, l)\}$.

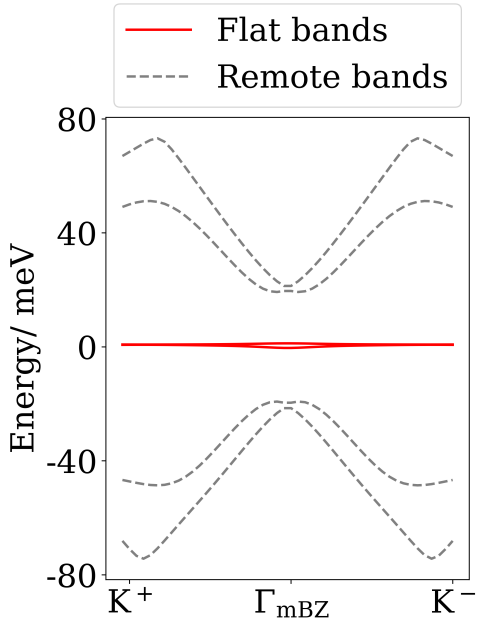
$$\hat{f}^\dagger_{n\mathbf{k}} = \sum_{\mathbf{G},\sigma,l} \hat{c}^\dagger_{\mathbf{k}}(\mathbf{G}, \sigma, l) u_{n\mathbf{k}}(\mathbf{G}, \sigma, l), \quad n \in \{\pm 1\}$$

From BM to IBM

$$\hat{H} = \hat{H}_0 + \hat{H}_I = \sum_{\mathbf{k}\in\Omega^*} \sum_{mn} \hat{f}^\dagger_{m\mathbf{k}} [h(\mathbf{k})]_{mn} \hat{f}_{n\mathbf{k}}$$

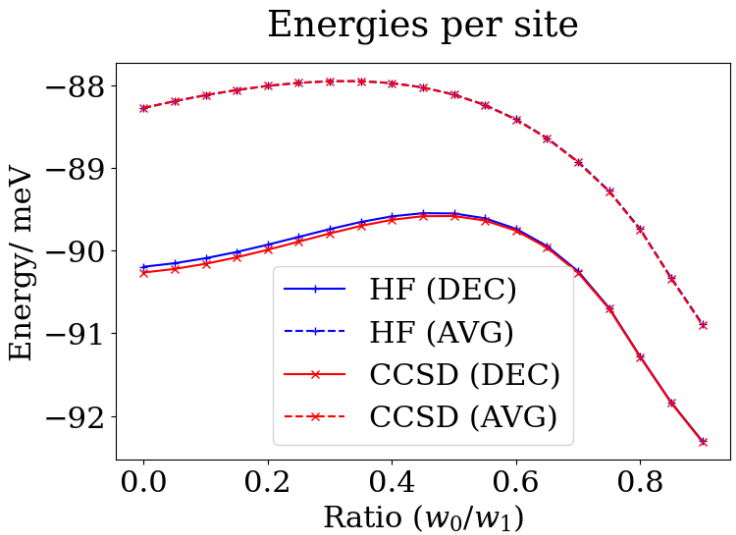
$$+\frac{1}{2} \sum_{\substack{\mathbf{k},\mathbf{k}',\mathbf{k}''\in\Omega^* \\ \mathbf{k}'''=\mathbf{k}+\mathbf{k}'-\mathbf{k}''}} \sum_{mm'nn'} \langle m\mathbf{k}, m'\mathbf{k}' | n\mathbf{k}'', n'\mathbf{k}''' \rangle \, \hat{f}^\dagger_{m\mathbf{k}} \hat{f}^\dagger_{m'\mathbf{k}'} \hat{f}_{n'\mathbf{k}'''} \hat{f}_{n\mathbf{k}''} .$$

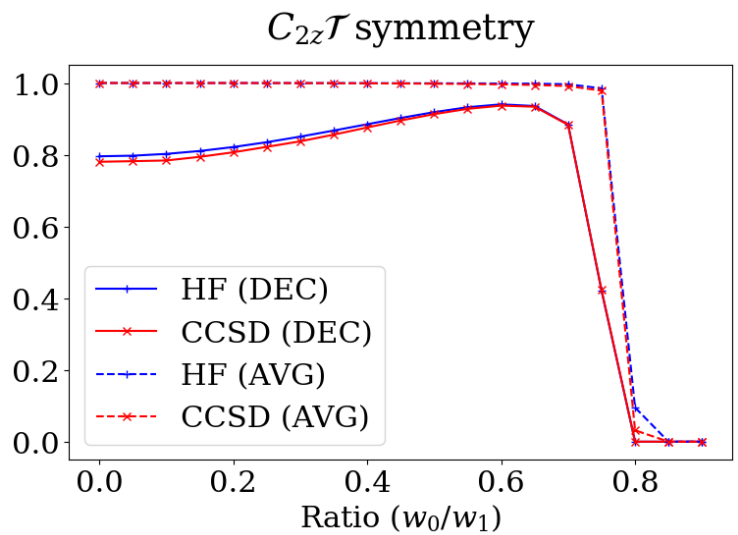
- Project to $m, n \in \{\pm 1\}$ flat band; (screened) Coulomb interaction, and properly remove double counting. Half filling (only 2 bands).
- In principle this is it, but the devil is in the details..



Quadratic term ($\hat{H}_0$)

- Near-flat band energies of BM model.
- Subtraction due to double counting Coulomb interaction within flat bands. Related to quantum embedding methods
- Sources for model discrepancies.





Interaction term ($\hat{H}_I$)

- Screened double gate potential[1]

$$V(\mathbf{q}) = e^2 \tanh(|\boldsymbol{q}|d)/2\epsilon_r\epsilon_0|\boldsymbol{q}|.$$

- Two electron repulsion integral (ERI)

$$\langle m\mathbf{k}, m'\mathbf{k}'|n\mathbf{k}'', n'\mathbf{k}'''\rangle = \frac{1}{|\Omega|N_{\mathbf{k}}} \sum_{\mathbf{G}\in\mathbb{L}^*_{\mathrm{mBZ}}} V(\mathbf{q}+\mathbf{G})[\Lambda_{\mathbf{k}}(\mathbf{q}+\mathbf{G})]_{mn}[\Lambda^*_{\mathbf{k}'}(\mathbf{q}+\mathbf{G})]_{n'm'}.$$

- Form factor

$$[\Lambda_{\mathbf{k}}(\mathbf{q})]_{mn} = \langle u_{m\mathbf{k}}|u_{n(\mathbf{k}+\mathbf{q})}\rangle$$

- Crystal momentum conservation $\mathbf{k}'' = \mathbf{k} + \mathbf{q}, \mathbf{k}''' = \mathbf{k}' - \mathbf{q}$

[1] see e.g., Bernevig et al, *TBG III*, PRB 2021

Methods

- Coupled Cluster (CC) theory[1]
  - Based on the cluster expansion e^T where T is the excitation operator.
  - Commonly truncated to second order $\Rightarrow$ CCSD
- Quantum Chemistry Density Matrix Renormalization Group (QC-DMRG)[2,3]
  - DMRG adapted to quantum chemistry long range interactions.
- Gauge invariant order parameter
  - Based on unitary matrix representation ("sewing marix") for symmetry operations[4].

[1](Coester Nucl. Phys. 1958), [2](White Phys. Rev. B 1999),[3](Chan, Head-Gordon 2002),

[4](Alexandradinata, Wang, Bernevig Phys. Rev. X 2016)

Coupled cluster theory

- Ansatz ($|\Phi_0\rangle$: Hartree-Fock state)

$$|\Psi\rangle = e^{T(\mathbf{t})}|\Phi_0\rangle, \quad T(\mathbf{t}) = \sum_\mu t_\mu X_\mu.$$

- $X_\mu = X^{a_1,\ldots,a_k}_{i_1,\ldots,i_k} = \hat{a}^\dagger_{a_1}\ldots\hat{a}^\dagger_{a_k}\hat{a}_{i_k}\ldots\hat{a}_{i_1}$: excitation operator.
- t_μ: CC amplitude. Amplitude equation

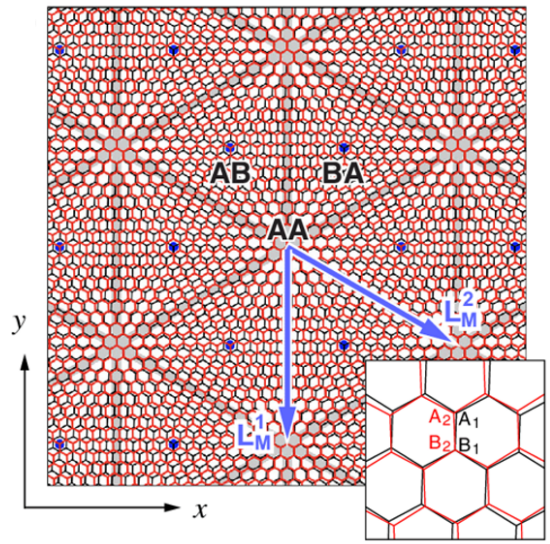
$$0 = F_\mu(\mathbf{t}) = \langle\Phi_0|X^\dagger_\mu e^{-T(\mathbf{t})}He^{T(\mathbf{t})}|\Phi_0\rangle, \qquad \forall\mu.$$

- Energy $\mathcal{E}(\mathbf{t}) = \langle\Phi_0|e^{-T(\mathbf{t})}He^{T(\mathbf{t})}|\Phi_0\rangle$.
- CCSD: $\hat{a}^\dagger_{a_1}\hat{a}_{i_1}$, $\hat{a}^\dagger_{a_1}\hat{a}^\dagger_{a_2}\hat{a}_{i_2}\hat{a}_{i_1}$. Most widely used CC theory.
- CCSD(T): perturbative treatment of $\hat{a}^\dagger_{a_1}\hat{a}^\dagger_{a_2}\hat{a}^\dagger_{a_3}\hat{a}_{i_3}\hat{a}_{i_2}\hat{a}_{i_1}$: gold standard of (molecular) quantum chemistry

IBM Hamiltonian: important parameters

- $\kappa = w_0/w_1$: ratio between hopping magnitude between AA sublattice w_0 and AB sublattice w_1.
  $\kappa = 0$: chiral limit.

- Filling. $\nu = (N_e/N_{\mathbf{k}}) - 1, \quad -1 \leq \nu \leq 1$.
  $\nu = 0$: integer filling.
  $|\nu| < 0.2$: near integer filling (for this work)
  $\nu = -2/3$: Fractional quantum hall state?[1]



[1] Parker et al, arXiv:2112.13837

Symmetries

- Symmetries play an important role in the properties of TBG[1].
- Relevant symmetry operations for single valley IBM

Symmetry	Real space	Momentum space	Type
C_{2z}	swap sublattice	swaps valleys; $\mathbf{k} \to -\mathbf{k}$	Unitary
C_{3z}	rotate by 120°	$\mathbf{k} \to C_{3z}\mathbf{k}$	Unitary
$\mathcal{T}$		swaps valleys; $\mathbf{k} \to -\mathbf{k}$	Antiunitary
$C_{2z}\mathcal{T}$	swap sublattice	$\mathbf{k} \to \mathbf{k}$	Antiunitary

[1] (Song et. al. PRB 2021)

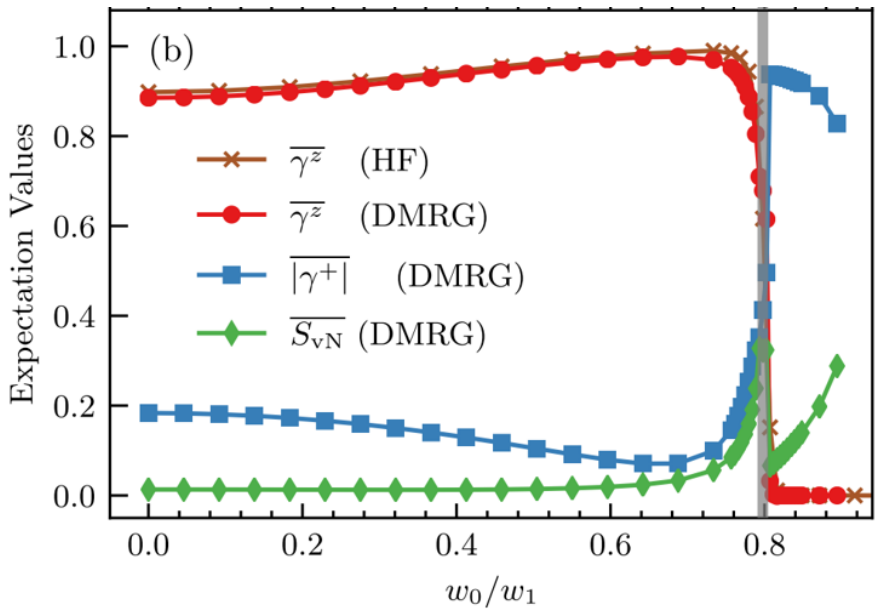
Phase diagram at integer filling

Single valley, single spin: focus on $C_{2z}\mathcal{T}$, C_{3z}

Symmetry	C_{3z} ✓	C_{3z} ✗
$C_{2z}\mathcal{T}$ ✓	BM	NSM
$C_{2z}\mathcal{T}$ ✗	QAH	/

BM: Band metal. NSM: Nematic semimetal. QAH: Quantum anomalous hall

Quantum anomalous hall (QAH) to nematic semimetal (NSM) transition



- $\gamma_z(\mathbf{k}) := \mathrm{Tr}\left[P(\mathbf{k})\sigma_z\right]$, and σ_z refers to a specific Chern basis.
- $\gamma_+(\mathbf{k}) = \mathrm{Tr}\left[P(\mathbf{k})(\sigma_x + \mathrm{i}\sigma_y)\right], \varphi(\mathbf{k}) = \arg[\gamma_+(\mathbf{k})]$.
- Nematic semimetal requires checking C_{3z} symmetry.

Soejima et al, PRB 2020

Symmetry breaking

- Representation matrix of a discrete symmetry g
  α: general internal index (e.g., **G**, sublattice σ, layer l).

$$g\hat{c}^\dagger_{\mathbf{k}}(\alpha)g^{-1} = \sum_{\alpha'} \hat{c}^\dagger_{g\mathbf{k}}(\alpha')[D(g)]_{\alpha',\alpha}.$$

- Transform to the BM band basis

$$\hat{f}^\dagger_{n\mathbf{k}} = \sum_{\alpha} \hat{c}^\dagger_{\mathbf{k}}(\alpha) u_{n\mathbf{k}}(\alpha)$$

- Transformation rule: sewing matrix[1]

$$g\hat{f}^\dagger_{n\mathbf{k}}g^{-1} = \sum_{m} \hat{f}^\dagger_{m,g\mathbf{k}}[B(g)]_{\mathbf{k},mn}, \quad [B(g)]_{\mathbf{k},mn} := \langle u_{m,g\mathbf{k}}|D(g)|u_{n\mathbf{k}}\rangle .$$

[1] Bernevig et al, *TBG III*, PRB 2021; This is only for unitary symmetry, anti-unitary symmetry is similar

Gauge invariant order parameter

- 1-RDM $[P(\mathbf{k})]_{nm} = \langle \hat{f}^{\dagger}_{m\mathbf{k}} \hat{f}_{n\mathbf{k}} \rangle$. If symmetry preserving, then

$$[P(\mathbf{k})]_{nm} = \langle g\hat{f}^{\dagger}_{m\mathbf{k}} g g^{-1} \hat{f}_{n\mathbf{k}} g^{-1} \rangle = \sum_{pq} [B(g)]_{pm} [B(g)]^{*}_{qn} \langle \hat{f}^{\dagger}_{p,g\mathbf{k}} \hat{f}_{q,g\mathbf{k}} \rangle$$

$$= (B^{\dagger}(g) P(g\mathbf{k}) B(g))_{nm}$$

- Spontaneous symmetry breaking: check commutator-like quantity

$$\mathcal{C}_{\mathbf{k}}(g) = \| P(g\mathbf{k})[B(g)]_{\mathbf{k}} - [B(g)]_{\mathbf{k}} P(\mathbf{k}) \|$$

- Anti-unitary symmetry is similar

$$\mathcal{C}_{\mathbf{k}}(g\mathcal{K}) = \| P(g\mathbf{k})[B(g\mathcal{K})]_{\mathbf{k}} - [B(g\mathcal{K})]_{\mathbf{k}} P(\mathbf{k})^{*} \|$$

Advantage of gauge-invariant order parameters

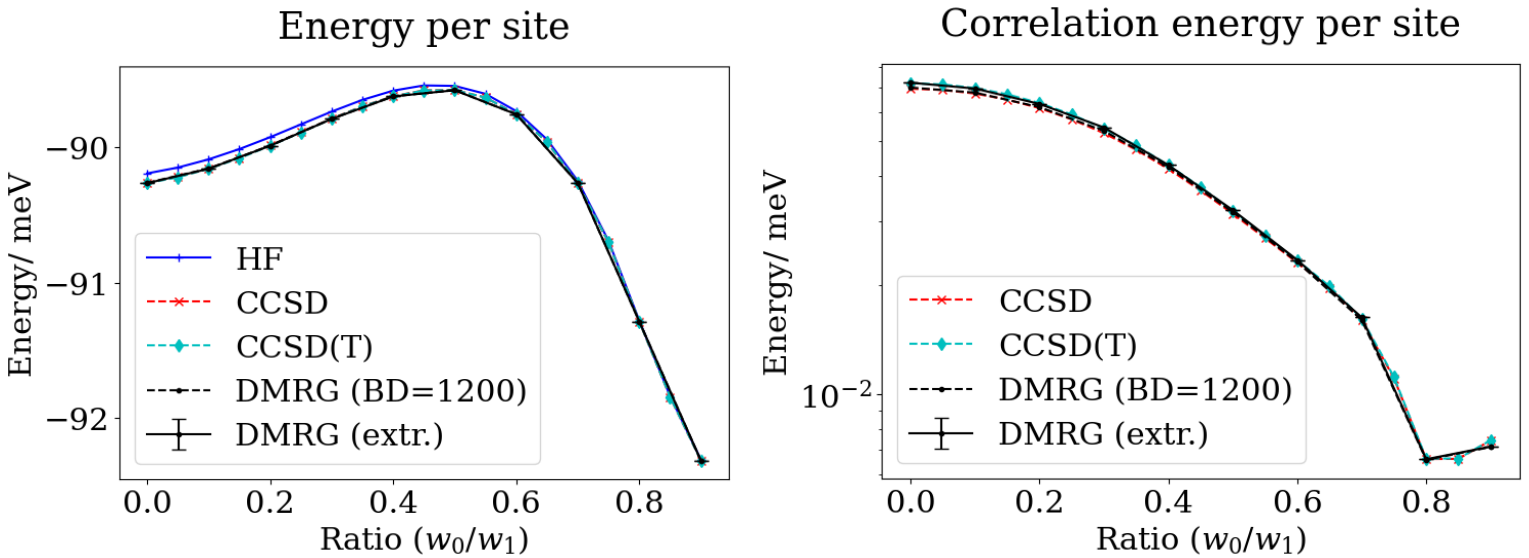
- Norm of commutators

$$\mathcal{C}_{\mathbf{k}}(C_{3z}) = \|P(C_{3z}\mathbf{k})[B(C_{3z})]_{\mathbf{k}} - [B(C_{3z})]_{\mathbf{k}}P(\mathbf{k})\|$$
$$\mathcal{C}_{\mathbf{k}}(C_{2z}\mathcal{T}) = \|P(\mathbf{k})[B(C_{2z}\mathcal{T})]_{\mathbf{k}} - [B(C_{2z}\mathcal{T})]_{\mathbf{k}}P^*(\mathbf{k})\|$$

- Gauge invariant. Directly compute in the BM band basis.
- The γ_z order parameter for $C_{2z}\mathcal{T}$ checks one off-diagonal entry of the commutator-like matrix.
- Generalizable to multi-band basis.

Outline

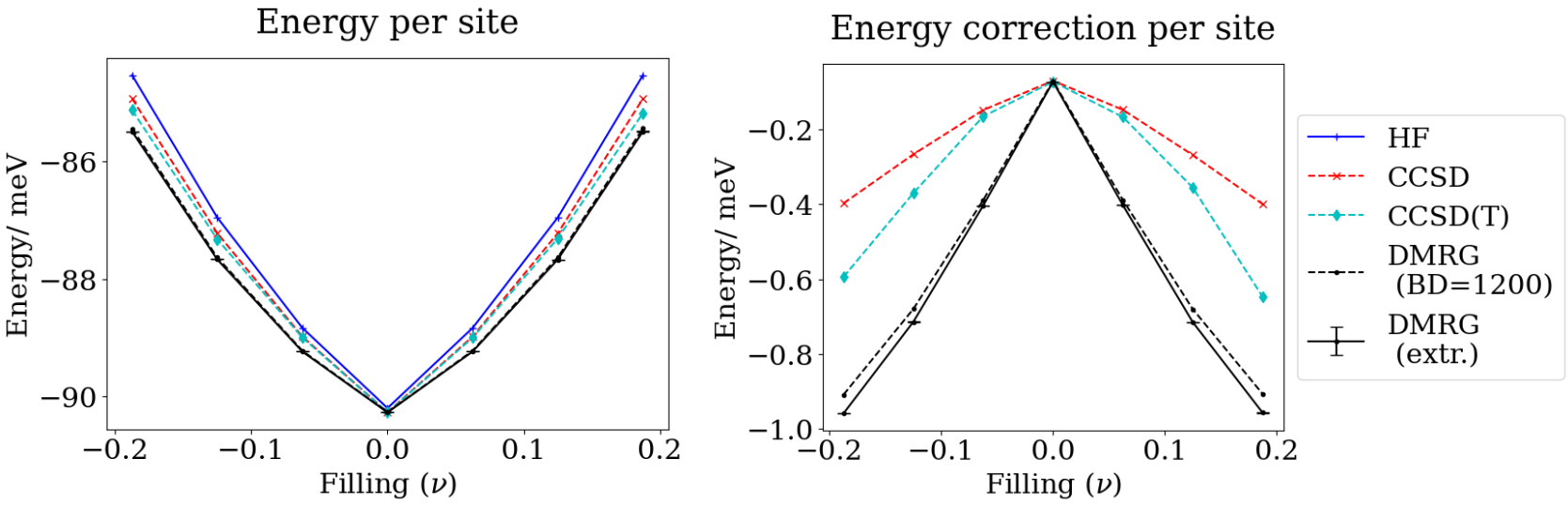
Surprising accuracy of Hartree-Fock solution at integer filling ($\nu = 0$)



- Correlation energy is small (< 0.1 meV per site). Agreement with previous DMRG studies[1].
- CCSD and CCSD(T) nearly exact.
- Ground state is close to a single Slater determinant.

[1] Soejima et al, PRB 2020

(Still) very good accuracy near integer filling



- Chiral limit: $\kappa = w_0/w_1 = 0$.
- $|\nu| > 0.2$: convergence issues (frequent gapless cases)
- Potentially larger correction between CCSD(T) and DMRG.
- Sensitive initial guess dependence at all levels of theories(will elaborate later)

Fermi-Dirac entropy per moiré site (from DMRG)

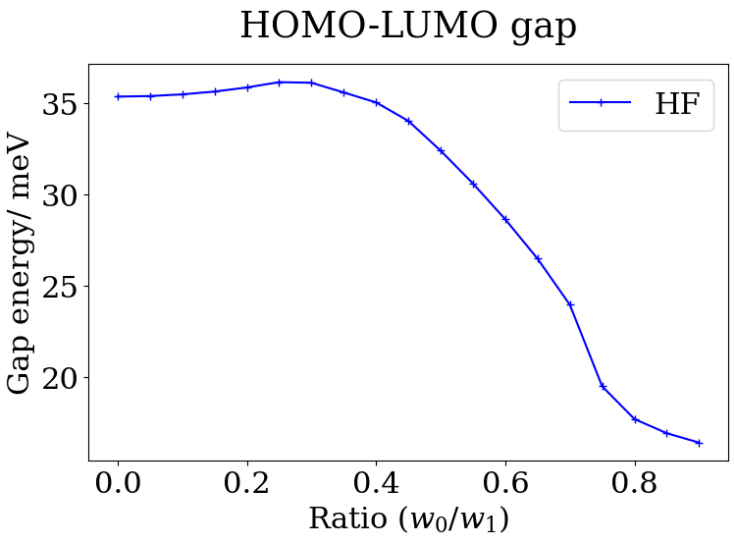
$$S_{\mathrm{FD}} = -\frac{1}{n_{k_x} n_{k_y}} \sum_i \left(p_i \ln p_i + (1 - p_i) \ln(1 - p_i) \right).$$

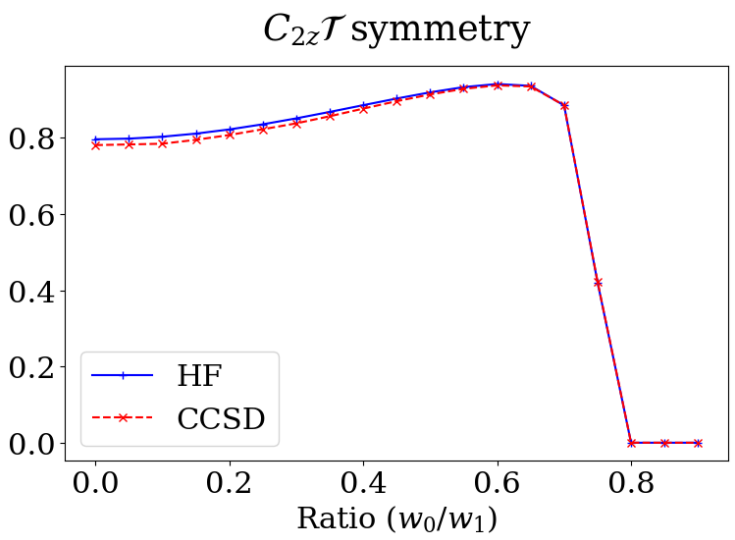
- p_i: eigenvalues of the 1-RDM. $S_{\mathrm{FD}} = 0$: Slater determinant
- $\nu = 0$ (integer filling): $0.009 \sim 0.032$.
- Non-integer filling $|\nu| < 0.2$

Filling (ν)	-0.188	-0.125	-0.062	0	0.062	0.125	0.188
S_{FD}	0.067	0.051	0.107	0.033	0.107	0.052	0.069

- Relatively small entropy (compared to $S_{\mathrm{FD}}^{\mathrm{max}} = 0.69$)$\Rightarrow$ Close to single determinant.

$C_{2z}\mathcal{T}$ order parameter: integer filling

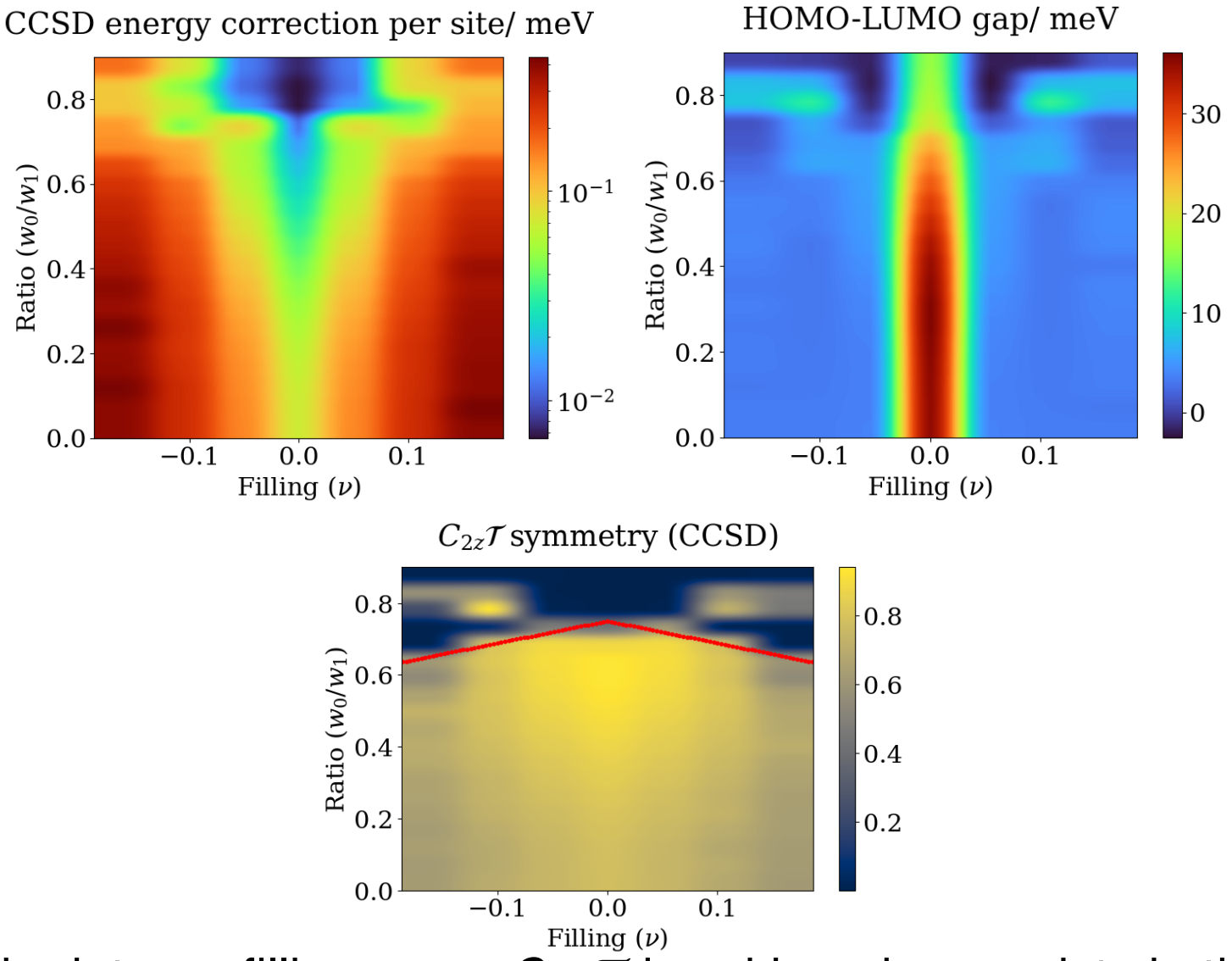




- Quantum anomalous hall (QAH) insulating state $\Rightarrow$ metallic state. Transition around $\kappa = w_0/w_1 \approx 0.75$.
- Gauge-invariant order parameter with previous studies with previous result in the Chern band basis[1]

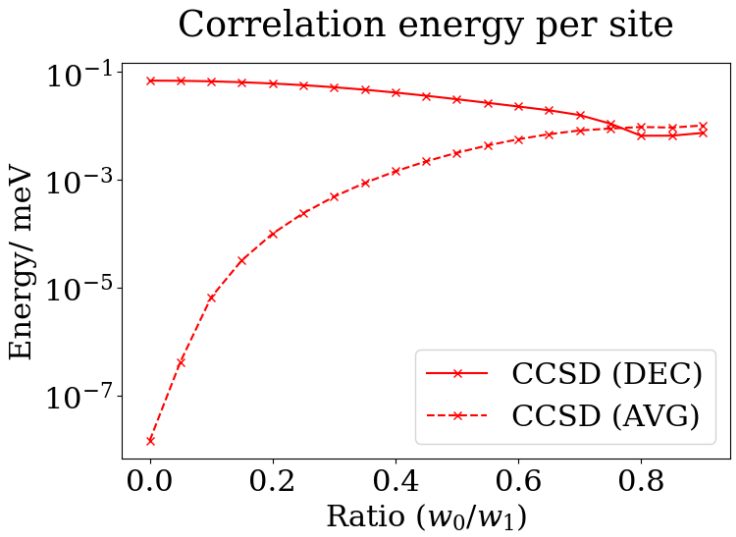
[1] Soejima et al, PRB 2020

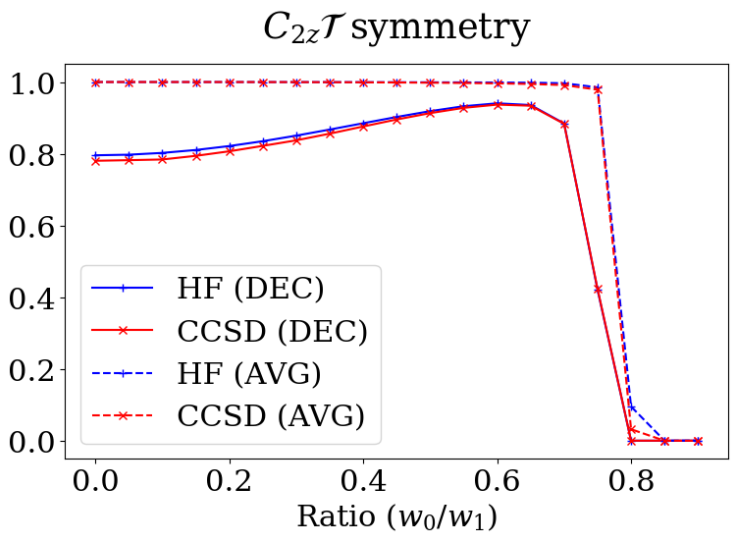
Non-Integer filling



Unlike the integer filling case, $C_{2z}\mathcal{T}$ breaking phase exists in the metallic region.

Model discrepancy





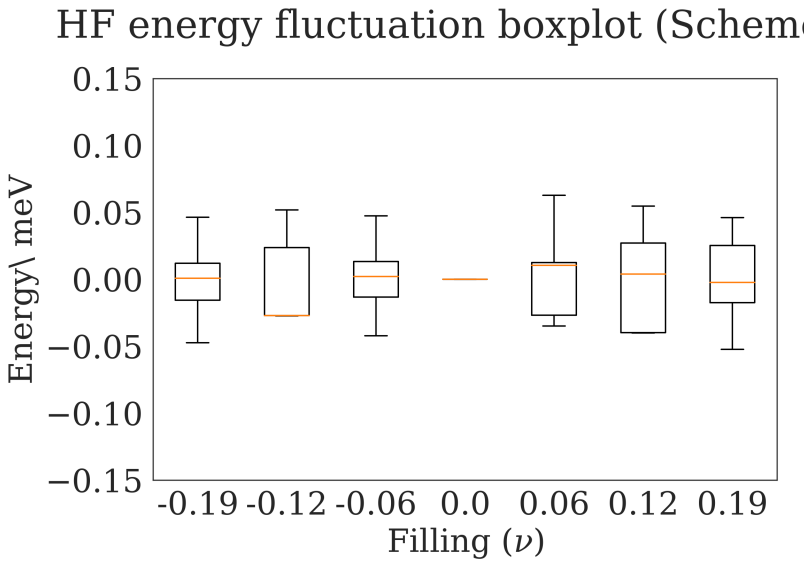
- Subtraction Hamiltonian: $h_{\text{sub}}(\mathbf{k}) = v_{\text{hf}}[P^0](\mathbf{k})$.
- Average scheme (AVG)[1]: $P^0(\mathbf{k}) = \frac{1}{2}I$
- Decoupled scheme (DEC)[2]:

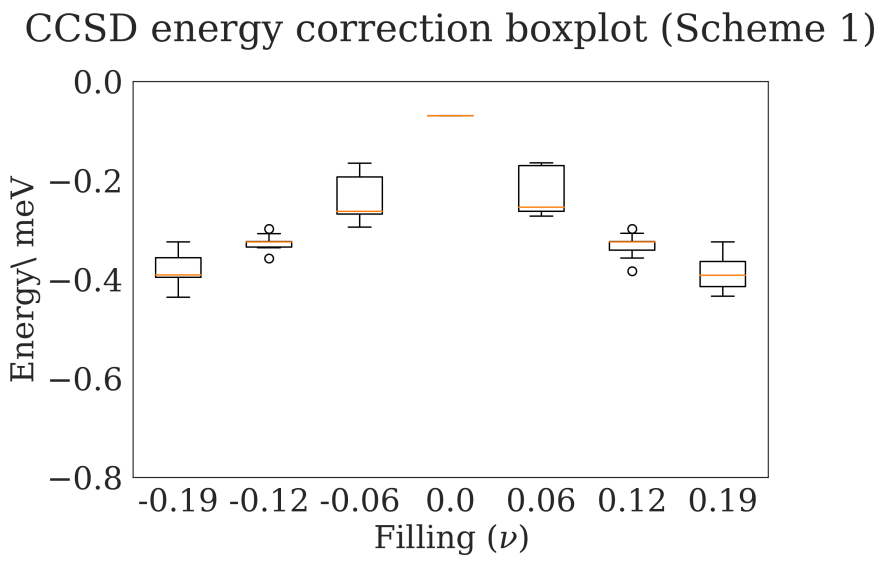
$$[P^0(\mathbf{k})]_{mn} := \lim_{\beta\to\infty} \langle u_{m\mathbf{k}}| \left(\exp(-\beta \hat{H}_{BM,\text{dec}}(\mathbf{k})) + 1\right)^{-1} |u_{n\mathbf{k}}\rangle$$

[1] Bernevig et al, 2021

[2] Bultinck et al, 2020; Potasz, Xie, MacDonald, 2021

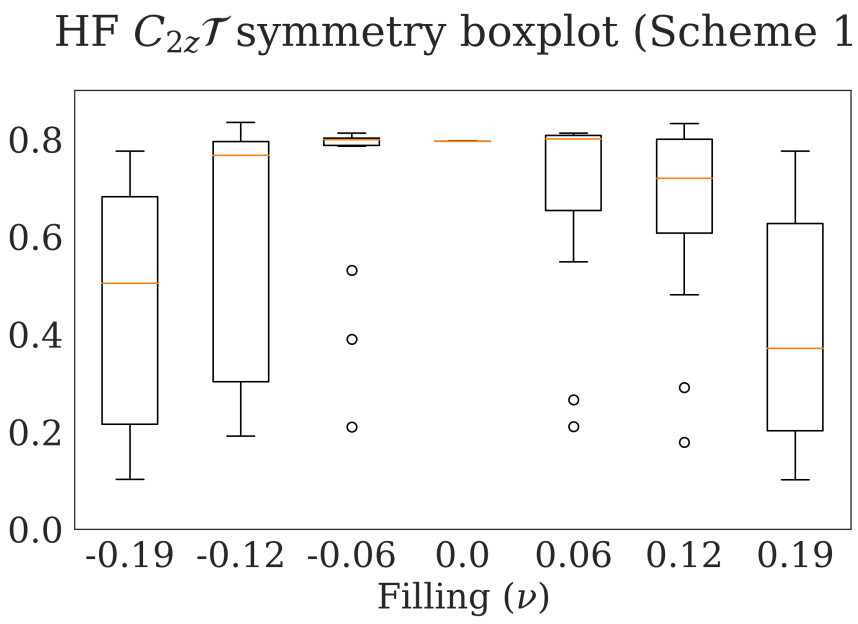
Convergence difficulties at non-integer filling

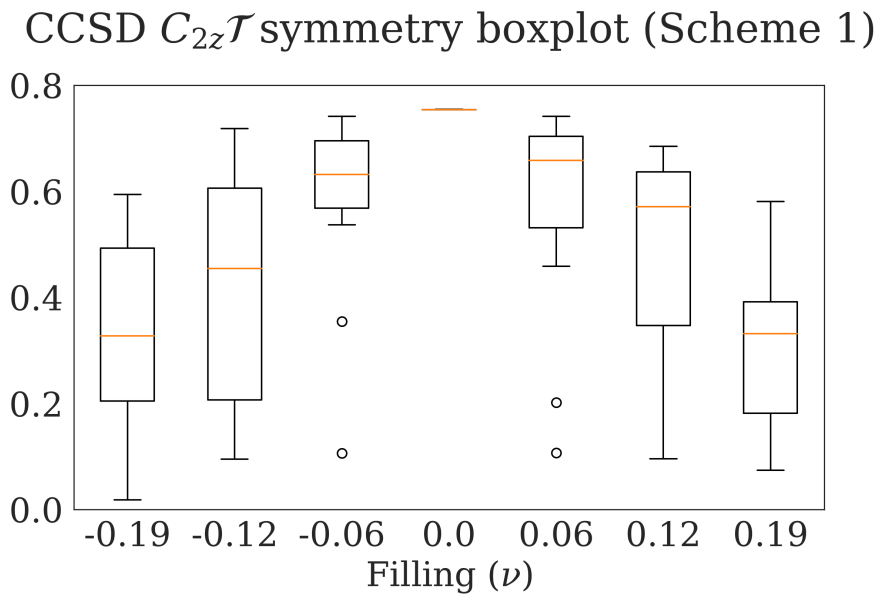




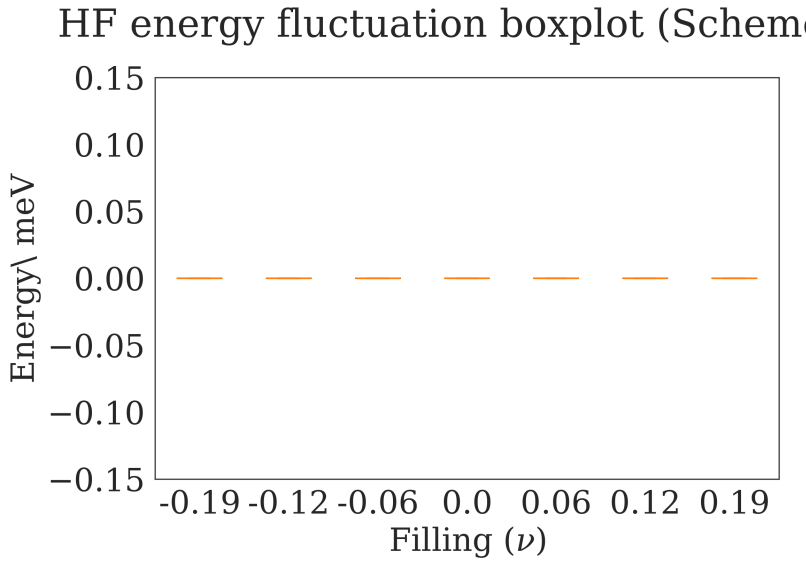
- DMRG also suffers from the same level of initial dependence!
- Many low entropy states with energies very close to the ground state energy

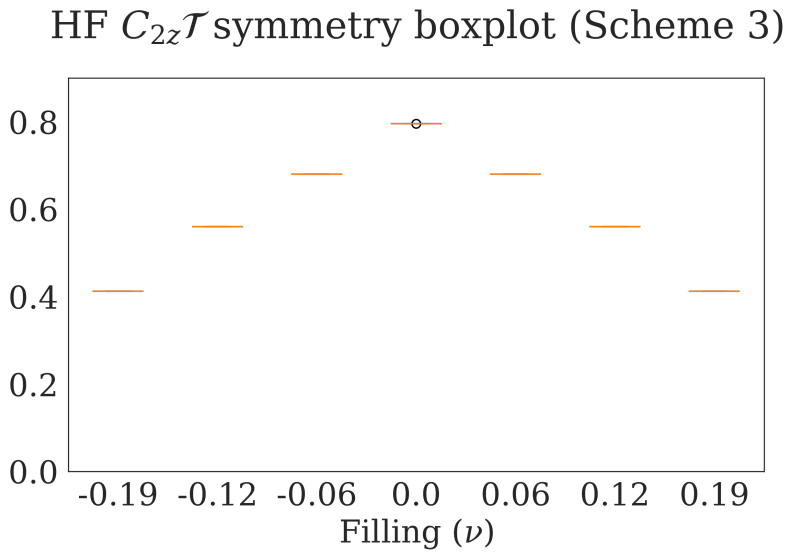
Convergence difficulties at non-integer filling

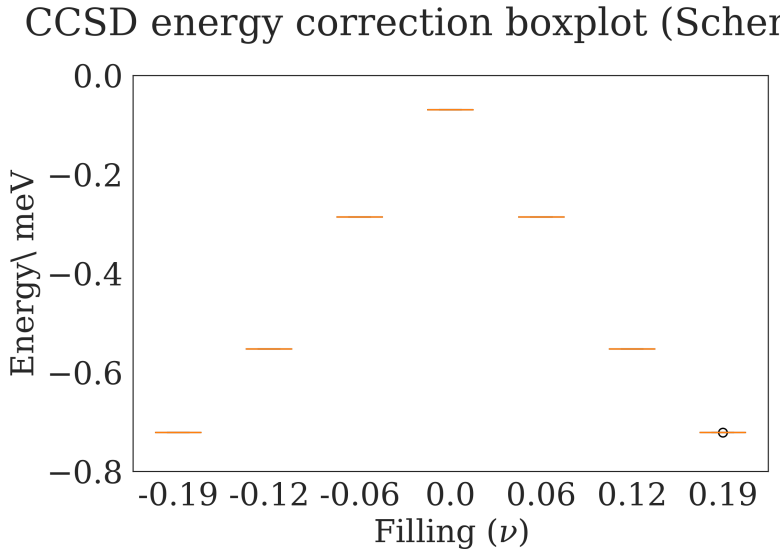


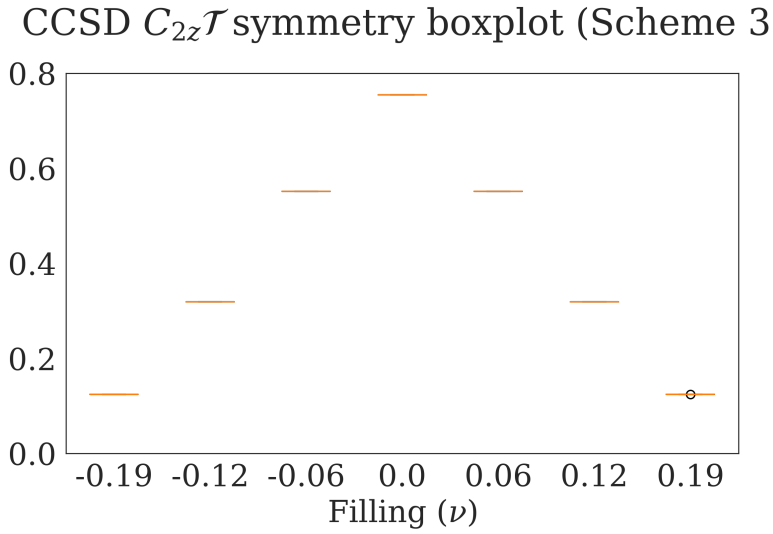


Translation symmetry + careful initialization greatly improves numerical stability









Summary

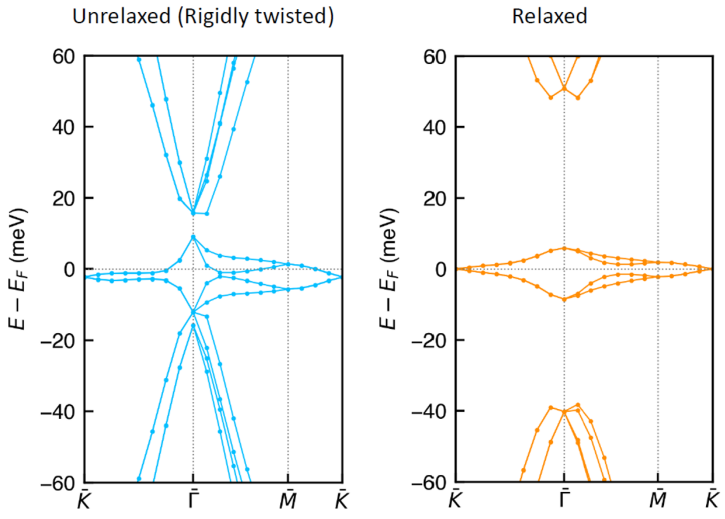
Spinless, valleyless model; No geometry relaxation.

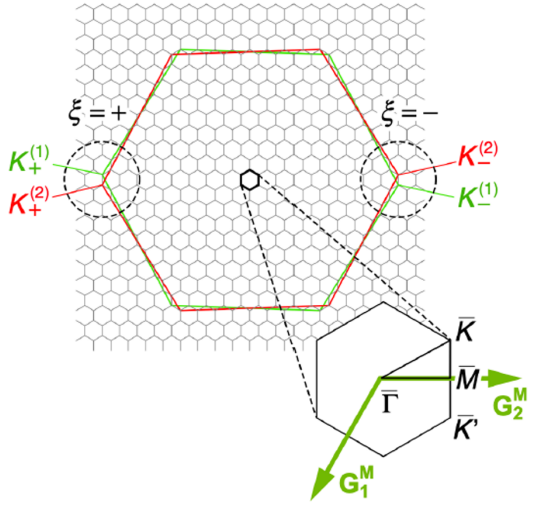
- At integer filling, the nature of the correlated insulating state is (very close to) a ferromagnetic Slater determinant which breaks $C_{2z}\mathcal{T}$ (Quantum anomalous hall, QAH state)
- At non-integer fillings:
  - The difference in found the ground state energy between Hartree-Fock and post-Hartree-Fock methods increases as doping increases.
  - Large number of states with near ground state energy which are close to a Slater determinant.
  - $C_{2z}\mathcal{T}$ symmetry breaking in the metallic region.
- Furthermore, the choice of double counting subtraction has a non-negligible impact on the energy and symmetries.

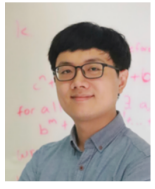
Outline

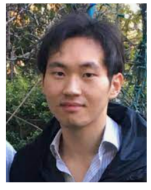
Going beyond continuum model

- DFT calculation for the band structure. DFT supercell: 10^4 atoms (per moire **k** point) using SIESTA.
- Geometry relaxation using the twister code (force field).
- Quantum embedding for modeling of interacting physics.






Raehyun Kim


Woochang Kim


Mit Naik

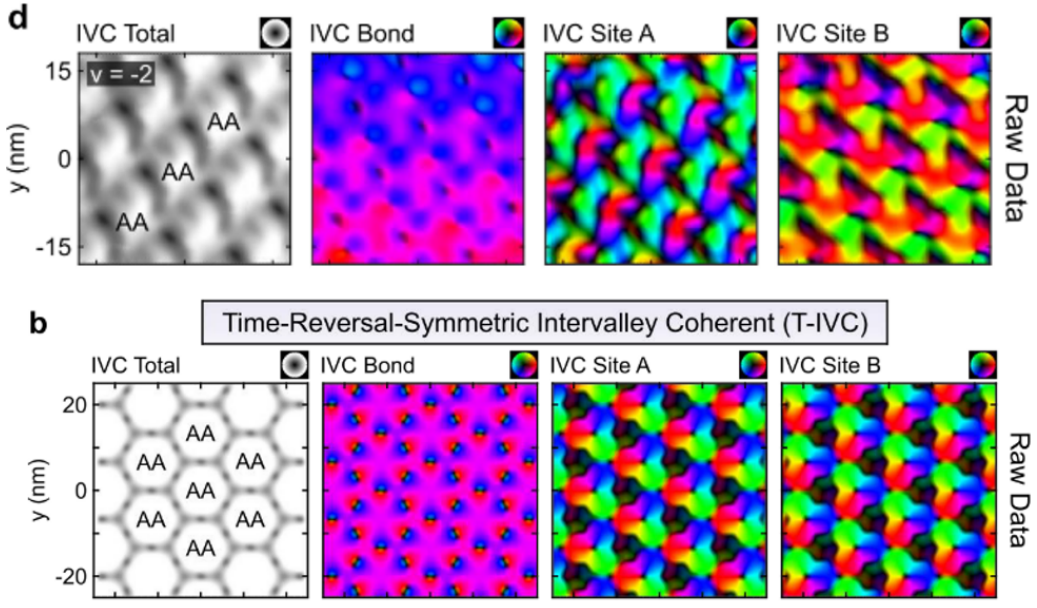
Work in progress with Steven Louie 's group and Mike Zaletel.

Recent experimental mystery

Quantum textures of the many-body wavefunctions in magic-angle graphene

Kevin P. Nuckolls[1,*], Ryan L. Lee[1,*], Myungchul Oh[1,*], Dillon Wong[1,*], Tomohiro Soejima[2,*], Jung Pyo Hong[1], Dumitru Călugăru[1], Jonah Herzog-Arbeitman[1], B. Andrei Bernevig[1,3,4], Kenji Watanabe[5], Takashi Taniguchi[6], Nicolas Regnault[7], Michael P. Zaletel[2,8], Ali Yazdani[1,†]

arXiv: 2303.00024



Experiment

Theory

But T-IVC is supposed to be energetically unfavorable. (Bultinck et al, PRX 2020)

Conclusion

- Correlated quantum chemistry can be fruitfully applied to study interacting models of TBG.
- Deeper doped regime. Finite temperature effects. Excited states.
- Going beyond continuum model: routine computation of electronic structure of 10^4 atom per $\mathbf{k}$ point.
- Highly accurate 10^4 atom HF / hybrid functional calculation?
- Model order reduction of improved continuum model.

Acknowledgment

Thank you for your attention!

Lin Lin
https://math.berkeley.edu/~linlin/

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