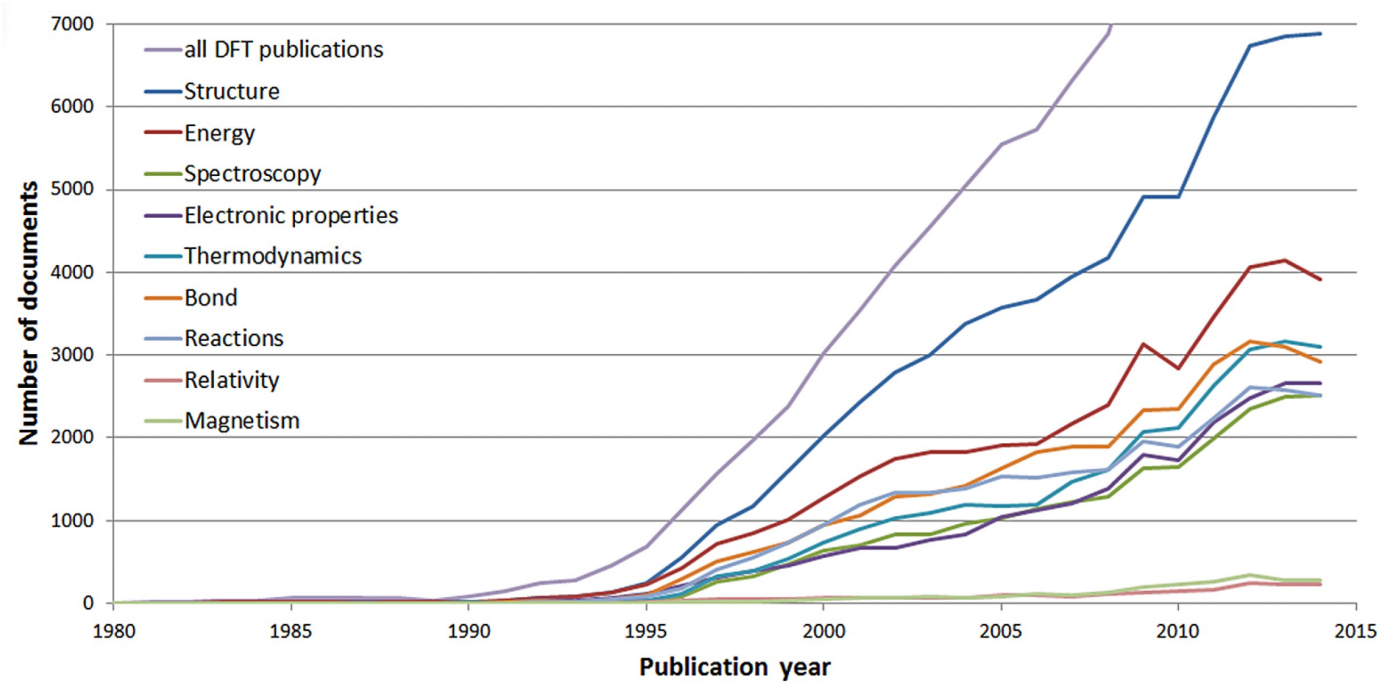
Downfolding complex materials problems onto model Hamiltonians for quantum computers

Antonios M. Alvertis[1], Norm M. Tubman[2]

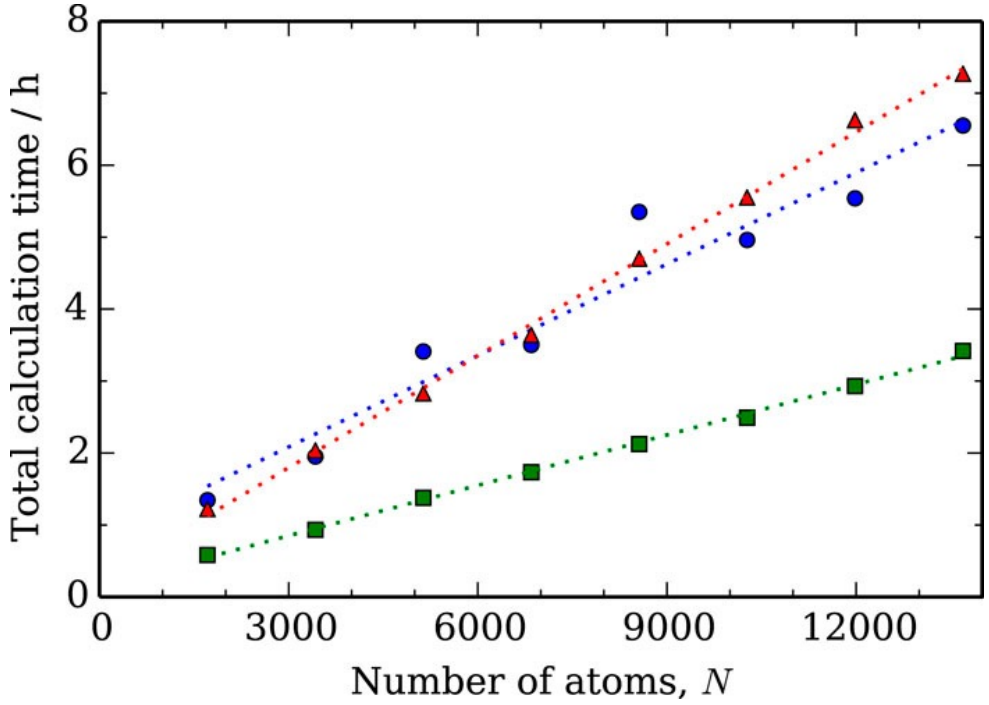
[1] KBR, Inc., NASA Ames Research Center
[2] NASA Ames Research Center

APS March Meeting 2024
Thursday March 7th

Density functional theory on classical computers for materials science: A success story for weakly-correlated systems



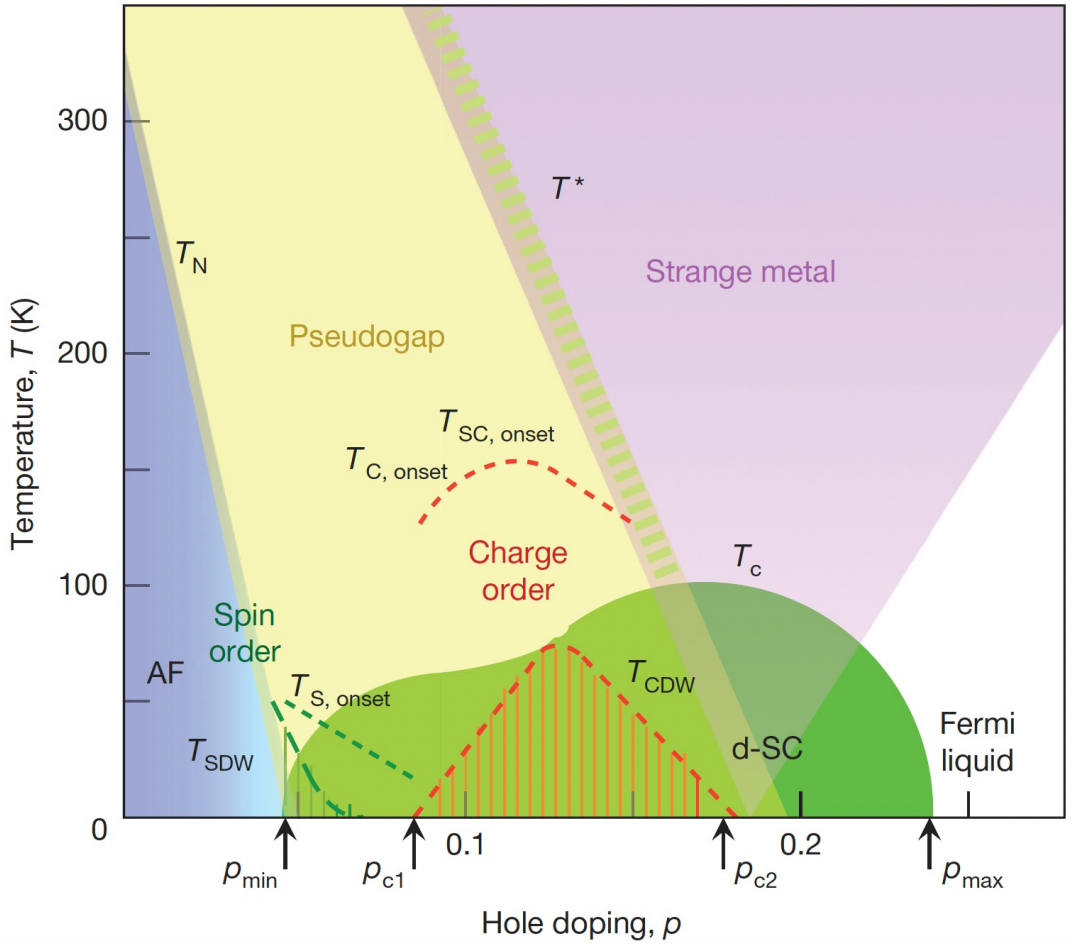
Haunschild *et al.* J Cheminform (2016) 8:52



Prentice *et al.* J. Chem. Phys.. 2020;152(17)

Why quantum computers for materials science? Strong correlations and beyond

Phase diagram, cuprate high-temperature superconductors



Keimer *et al.* Nature, 518, 179 (2015)

Explicit state of N electrons:

$$|x_1 x_2 \dots x_N > \longrightarrow 2^N \text{ configurations}$$

For $N = 100$, we have 2^{100} configurations to represent on a classical machine

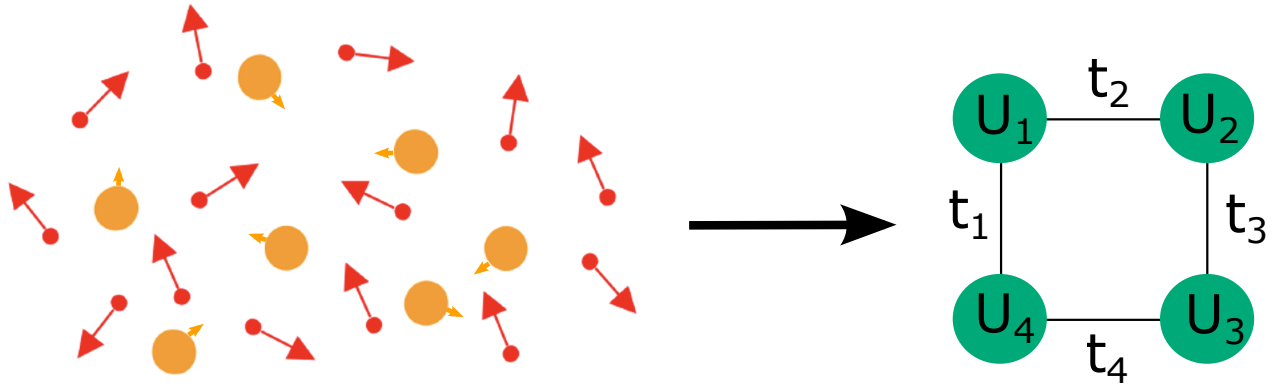
On quantum hardware, it would in principle suffice to have 100 qubits

Ab initio downfolding on many-body Hamiltonians

First-principles derivation of extended multi-band Hubbard models within an active space of interest:

$$H = \sum_{\sigma} \sum_{\mathbf{R}\mathbf{R}'} \sum_{ij} t_{i\mathbf{R}j\mathbf{R}'} a^{\sigma\dagger}_{i\mathbf{R}} a^{\sigma}_{j\mathbf{R}'} +$$

$$\frac{1}{2} \sum_{\sigma\rho} \sum_{\mathbf{R}\mathbf{R}'} \sum_{ij} U_{i\mathbf{R}j\mathbf{R}'} a^{\sigma\dagger}_{i\mathbf{R}} a^{\rho\dagger}_{j\mathbf{R}'} a^{\rho}_{j\mathbf{R}'} a^{\sigma}_{i\mathbf{R}},$$



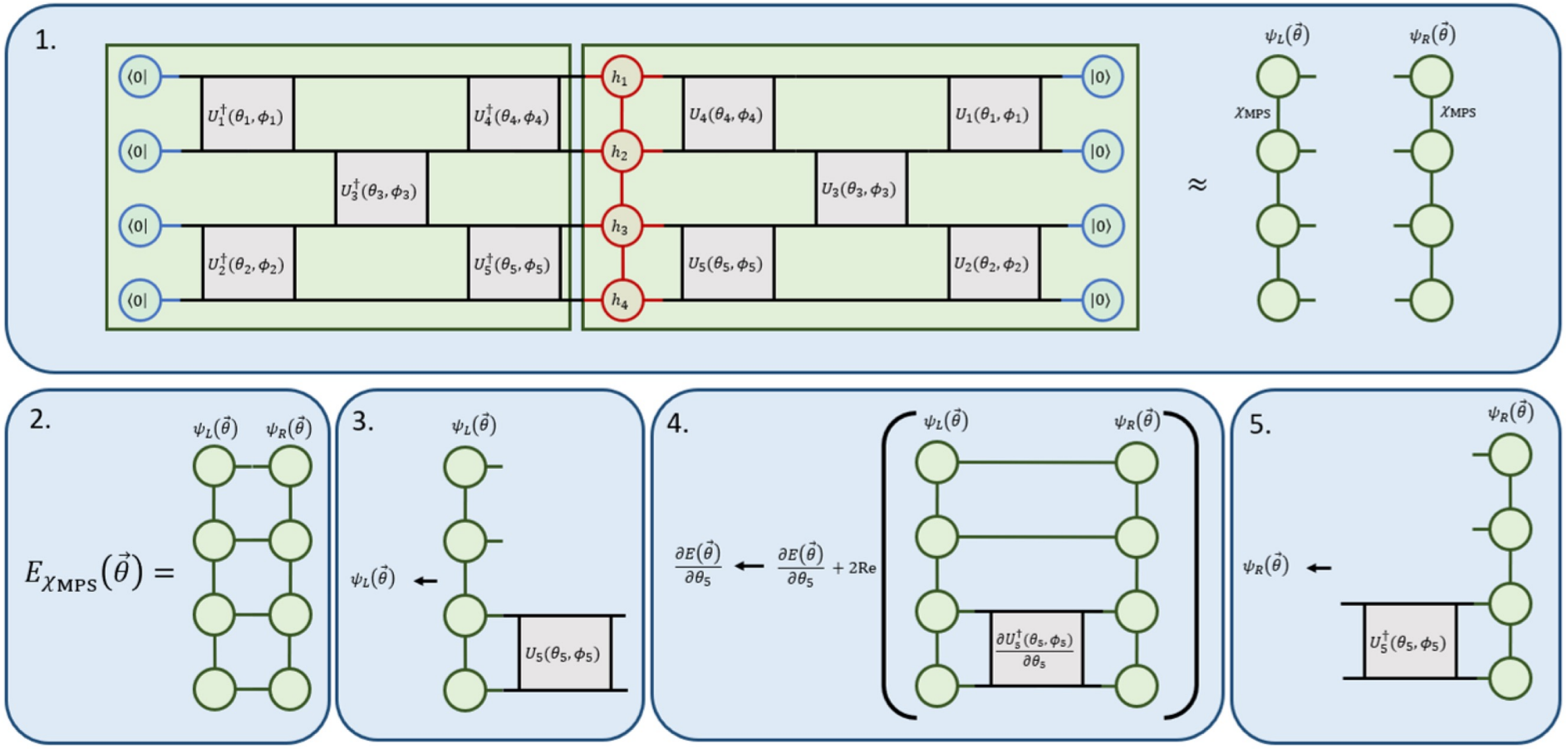
Electronic bands: i, j

Site-localized representation ($\boldsymbol{R}$), using Wannier functions

Related codes and approaches:

Zheng et al. Frontiers in Physics 6, 2018
Nakamura *et al.* Comp. Phys. Comm. 261, 107781, 2021
Romanova *et al.* npj Comp. Mater. 9:126, 2023

Classical simulation of variational quantum eigensolvers using tensor networks



Khan, Clark, Tubman
arXiv:
2310.12965 (2023)

Matrix product state representation of a parametrized quantum circuit:

$$\left|\psi_{\mathrm{PQC}}\left(\vec{\theta}\right)\right\rangle = U_n(\vec{\theta}_n)\ldots U_2(\vec{\theta}_2)U_1(\vec{\theta}_1)\left|\psi_0\right\rangle$$

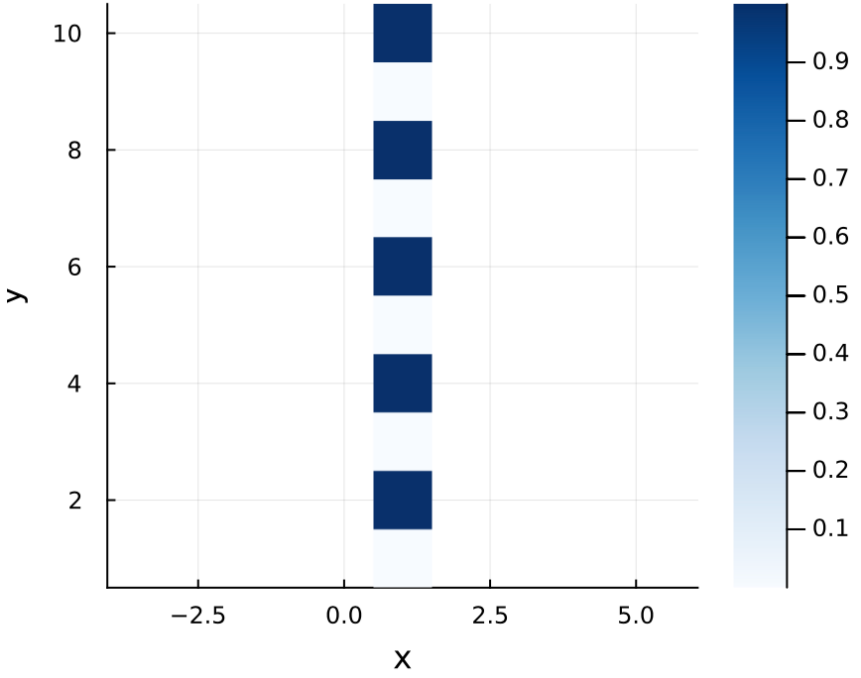
Classically optimize the energy function: $E_\chi(\vec{\theta}) = \left\langle \psi_\chi(\vec{\theta}) \right| H \left| \psi_\chi(\vec{\theta}) \right\rangle$

Quasi-1D cuprate material: Ca_2CuO_3 and antiferromagnetism

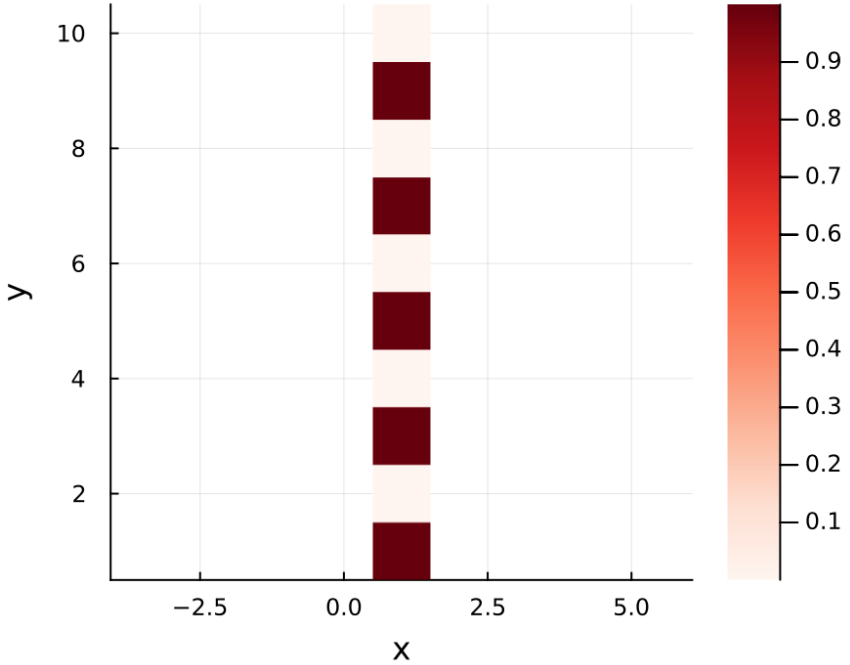
Ca_2CuO_3 is a prototypical 1D-antiferromagnetic material, see *e.g.* Graaf and Illas, Phys. Rev. B 63, 014404 (2000)

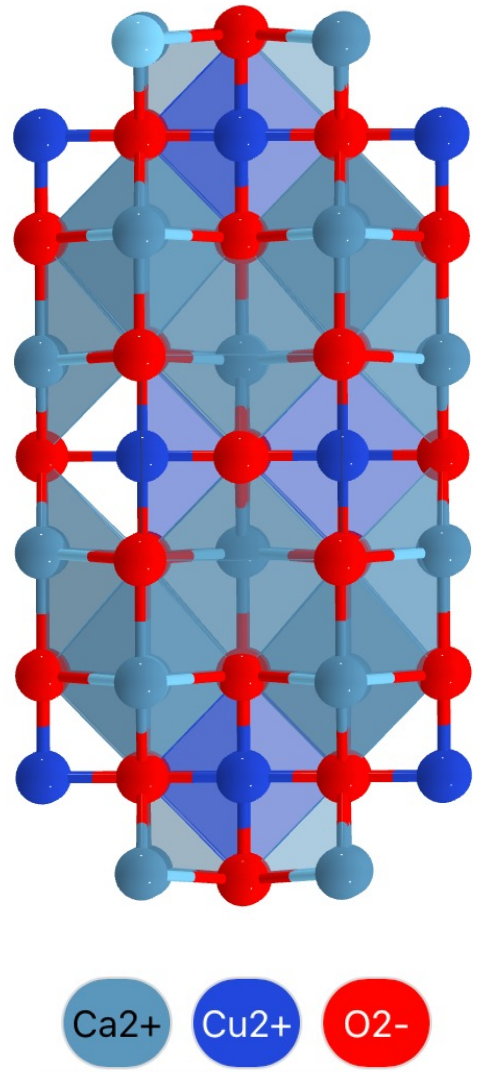
VQE on 1x10 lattice of one-band Hubbard model (20 qubits):

Spin-up occupation



Spin-down occupation

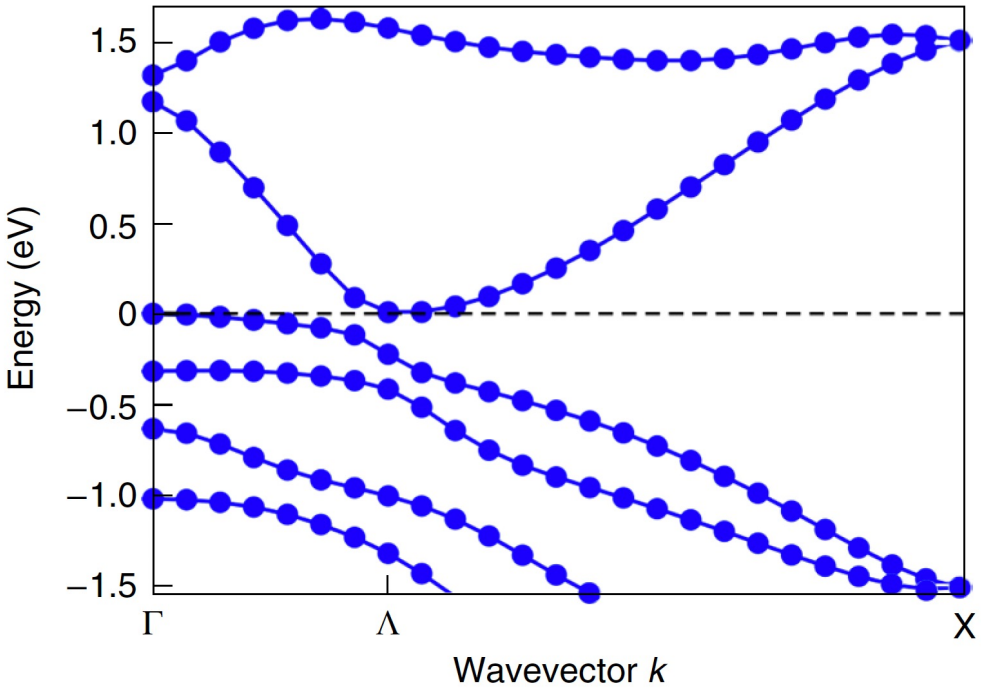




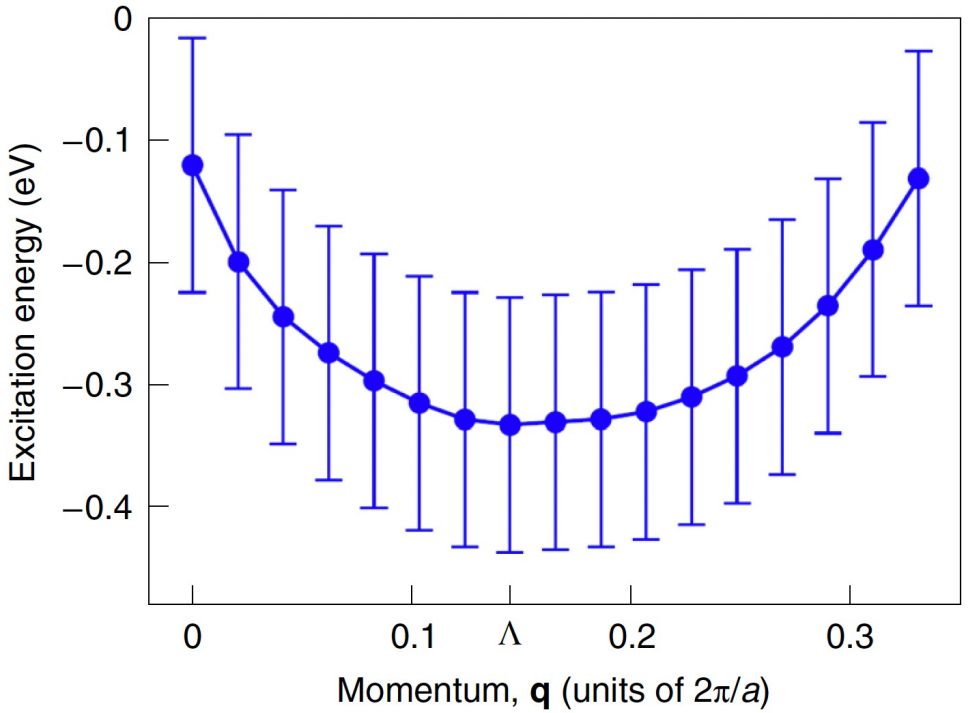
Monolayer WTe2 as a candidate for an excitonic insulating ground state

WTe_2 is a 2D monolayer material, which has recently attracted attention for its potential as an excitonic insulator, showing equilibrium exciton condensation.

Jia *et al.* Nat. Phys. 18, 87 (2022)
Sun *et al.* Nat. Phys. 18, 94 (2022)



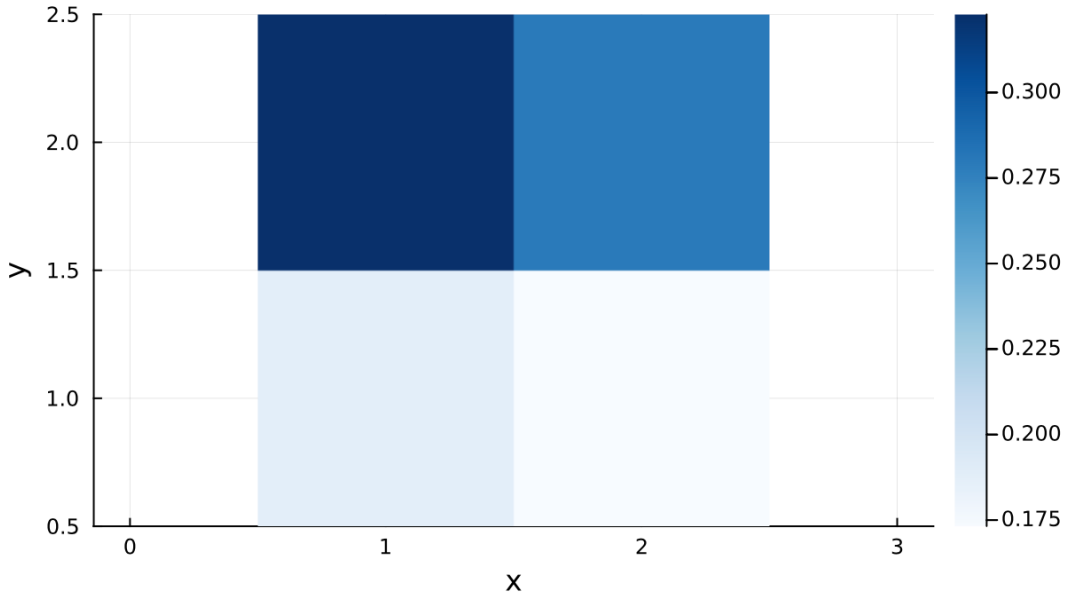
DFT bandstructure
and *GW*-BSE calculations for excitation energy
from Sun et al. Nat. Phys. 18, 94 (2022)



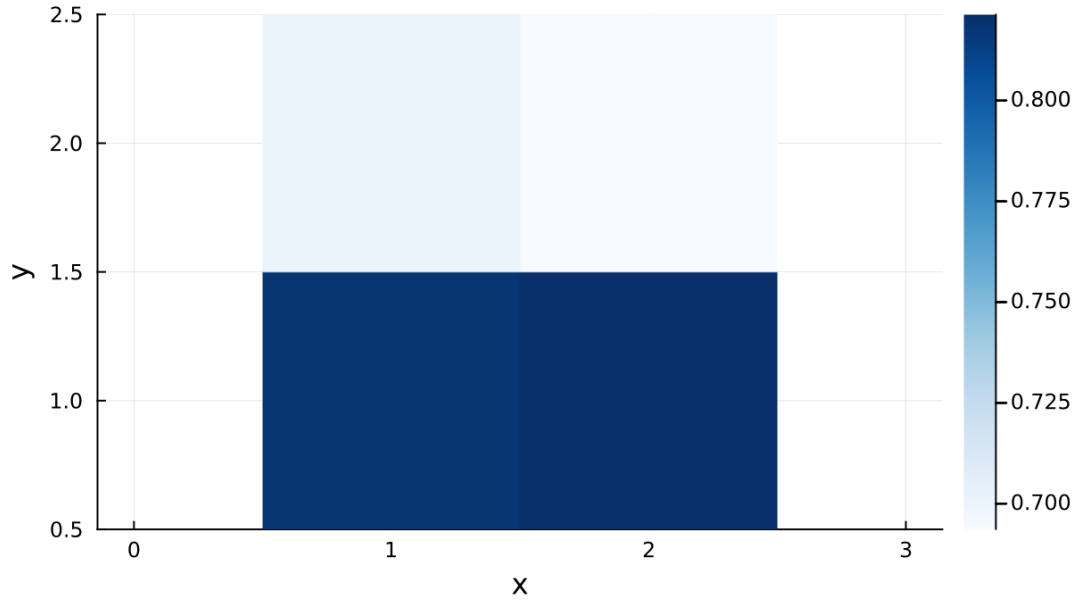
Monolayer WTe2 as a candidate for an excitonic insulating ground state

VQE on 2x2 lattice of two-band Hubbard model (16 qubits), at half-filling

Valence band: ~1 electrons



Conduction band: ~3 electrons



Excitonic insulating order parameter $\Delta = \frac{1}{N}\sum_{i\sigma} < c^{\dagger}_{ic\sigma} c_{iv\sigma} > = 0.4$

Electron-phonon interactions in *ab initio* downfolding schemes

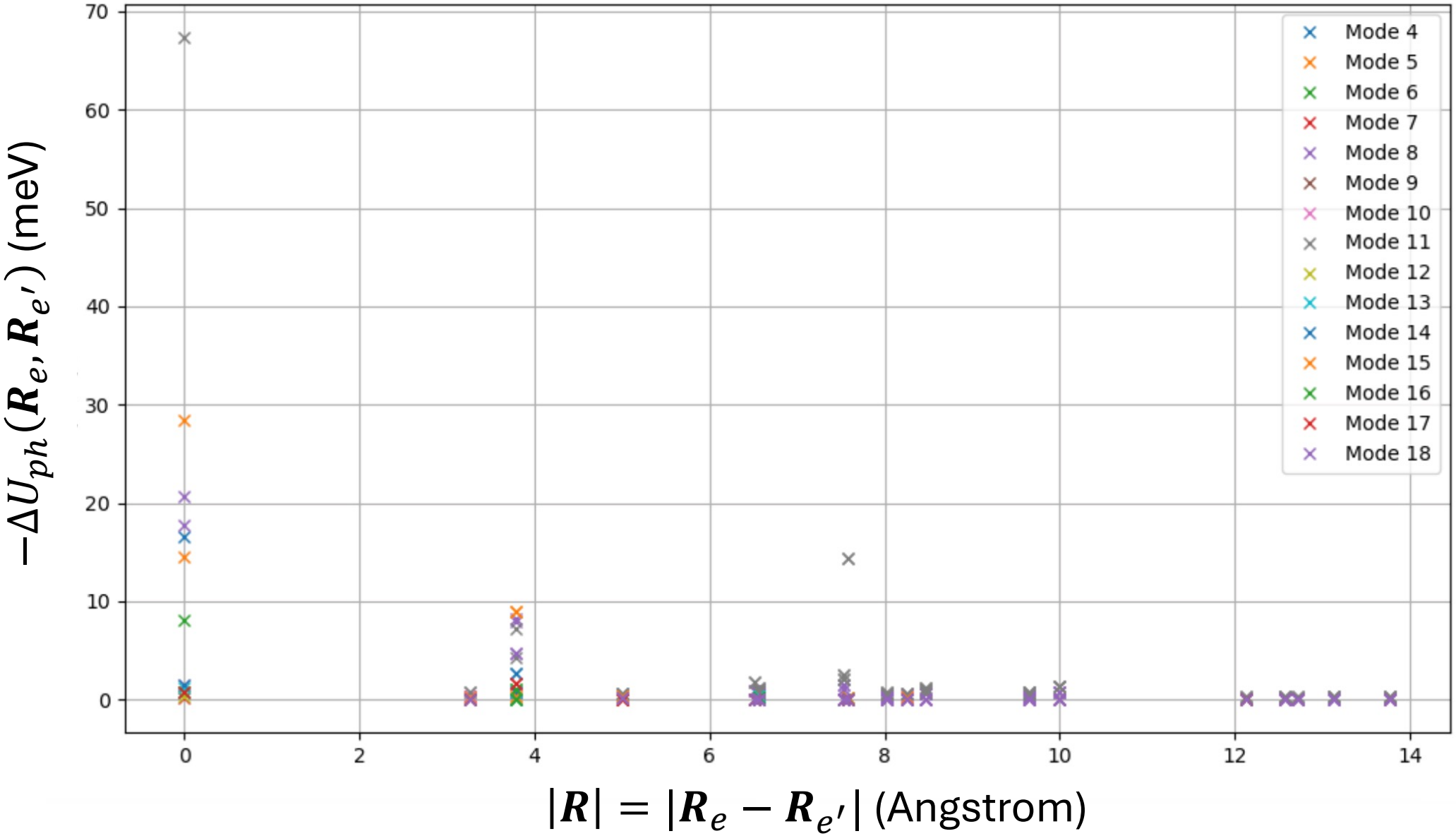
Effective downfolded electron Hamiltonian in an equilibrium phonon bath:

$$H^{el}_{eff} = \sum_{\sigma} \sum_{\mathbf{R}_e \mathbf{R}'_e} \sum_{ij} \langle X^{\dagger}_{\mathbf{R}_e} X_{\mathbf{R}'_e} \rangle t_{i\mathbf{R}_e j\mathbf{R}'_e} a^{\sigma\dagger}_{i\mathbf{R}_e} a^{\sigma}_{j\mathbf{R}'_e} +$$

$$\frac{1}{2} \sum_{\sigma\rho} \sum_{\mathbf{R}_e \mathbf{R}'_e} \sum_{ij} \left[U_{i\mathbf{R}_e j\mathbf{R}'_e} - \sum_{\mathbf{R}_p, \nu} \frac{2 g^2_{ij\mathbf{R}_p \nu}(\mathbf{R}_e, \mathbf{R}'_e)}{\omega_\nu} \right] a^{\sigma\dagger}_{i\mathbf{R}_e} a^{\rho\dagger}_{j\mathbf{R}'_e} a^{\rho}_{j\mathbf{R}'_e} a^{\sigma}_{i\mathbf{R}_e} .$$

with: $$\langle X^{\dagger}_{\mathbf{R}_e} X_{\mathbf{R}'_e} \rangle = \exp\left(- \sum_{\mathbf{R}_p, \nu} \frac{g^2_{ij\mathbf{R}_p, \nu}}{\omega^2_\nu} \left(2 n_B(\omega_\nu) + 1\right) \right)$$

Phonon renormalization of Coulomb interactions: Ca_2CuO_3



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(UC Berkeley)

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Conclusions

- Quantum computers hold promise for the study of electronic structure in strongly correlated materials

- *Ab initio* downfolding rigorous and cost-effective approach to prepare complex systems for quantum simulation

- We predict accurate ground state properties for antiferromagnetic material Ca_2CuO_3 and excitonic insulator candidate WTe_2

- Electron-phonon interactions can be incorporated into *ab initio* downfolding and hold potential for significantly altering electronic phase diagrams