Ab initio quantum simulation of strongly correlated materials with quantum embedding

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arXiv:2209.03202. npj Computational Materials v9, 78 (2023)

Quantum many-body problems

Quantum many-body problems: materials and molecules. Large degrees of freedom

$$H_e = -\sum_i \frac{\nabla_i^2}{2} - \sum_{i,I} \frac{Z_I}{|\mathbf{r}_i - \mathbf{R}_I|} + \frac{1}{2}\sum_{i \neq j} \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|}$$

Electronic problems

The ground state problem: $\min_{|\psi\rangle} \langle\psi|H_e|\psi\rangle$

A computing point of view

Focus on algorithms:
Design algorithms based on the I/O

Classical methods
LCU or QSVT types of algorithms
Hybrid quantum classical methods:

- QC-QMC (Friday afternoon)

Physical intuitions

Focus on target problems
Realstic problems → problems on a QC

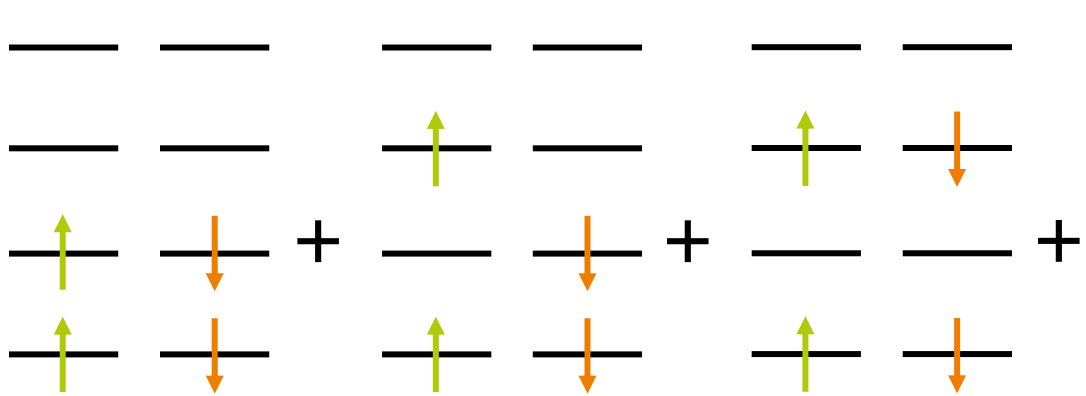
Design problem-tailed algorithms

Physicist, chemist, material scientists

Quantum computational chemistry

The central Ingredients: interpretation of the problem and Ansatz design

$$H_e = \hat{T} + \hat{V}_{ne} + \hat{V}_{ee} = \text{one body} + \text{two body}$$



Example: UCCSD Ansatz:

$$|\psi_{UCCSD}\rangle = e^{\hat{T}-\hat{T}^+}|\psi_{ref}\rangle$$

$\hat{T} \equiv \hat{T}_1 + \hat{T}_2 \rightarrow$ single and double excitations

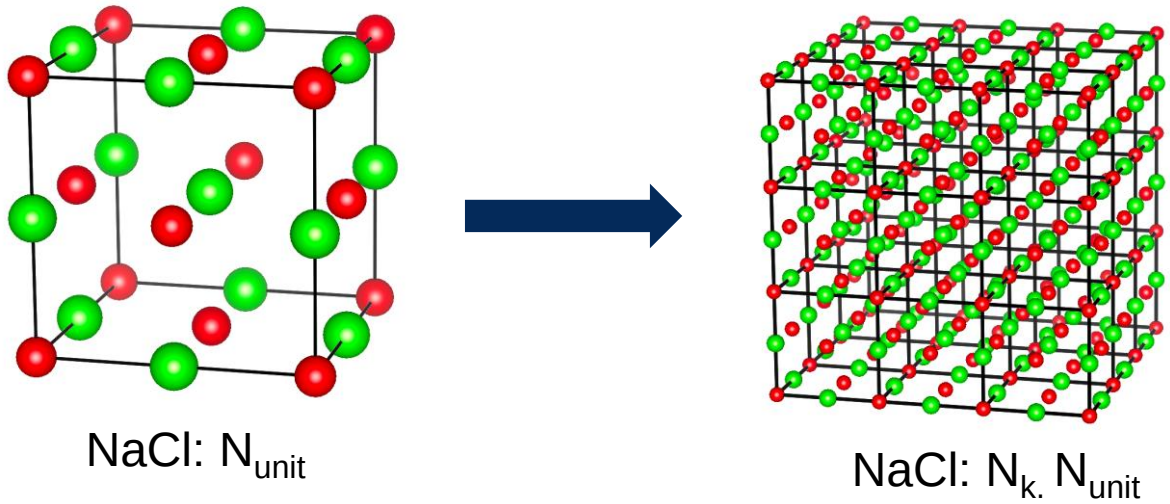
Demonstration

Experiments:

- H_{12} Diazene (HF 12 qubits) [Arute et al Science, 2020]
- F_2 (UCC, 12 qubits) arXiv:2212.08006
- Cyclobutene ring (UpCCD, 10 qubits, parameters found classically), [O'Brien et al Nature Physics (2023)]

Numerics: C_2H_4 (28 qubits)

Challenges for materials simulations



NaCl: N_{unit}

NaCl: $N_{k.}$ N_{unit}

Some cases （DFT, Material Project）

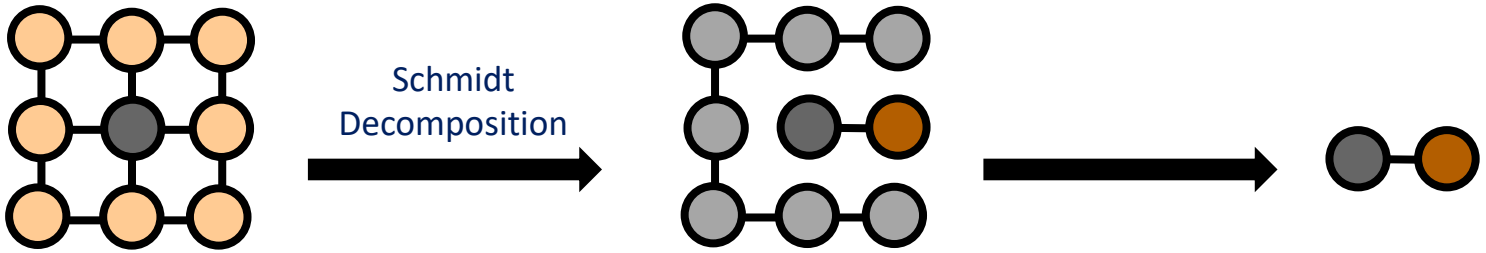
Species, N_k N_{unit}， N_{qubits}

- NaCl，(4,4,4)*8，(4608 qubits)
- MgO, (5,5,5)*8，(9000 qubits)
- Diamond，(7,7,7)*8，(21952 qubits)

H-chain/mesh/cubit (2H per cell)， He-chain/mesh/cubic (1He per cell)， LiH-chain

Phys. Rev. Research **4**, 013052 (2022); arXiv.2008.08694.

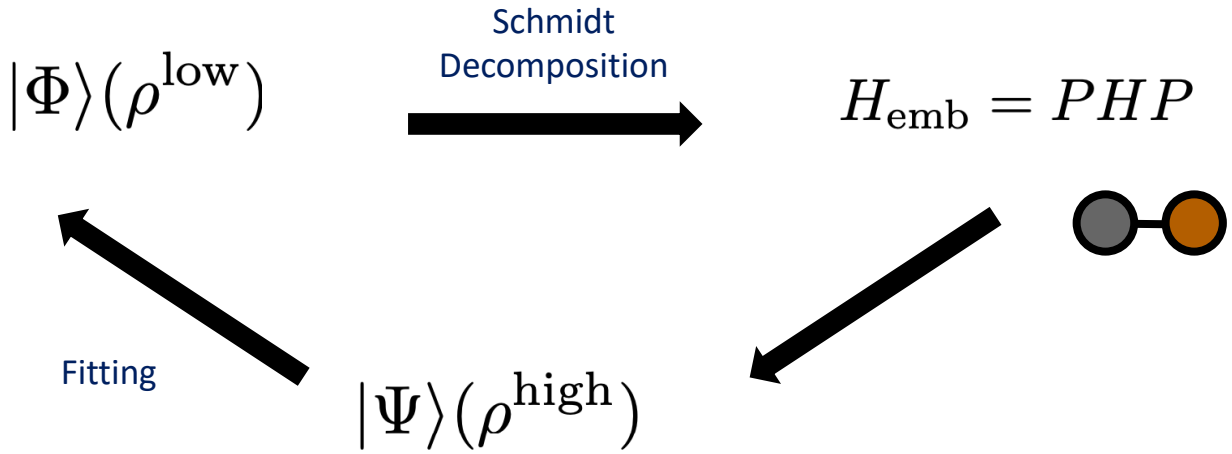
Quantum embedding approach



Fragment +
Environment

Fragment + Bath +
Unentangled Environment

High-level solver



$$|\Psi\rangle = \sum_{i}^{N_A} \sum_{j}^{N_B} \Psi_{ij} |A_i\rangle |B_j\rangle = \sum_{\alpha}^{\min(N_A,N_B)} \lambda_\alpha |\tilde{A}_\alpha\rangle |\tilde{B}_\alpha\rangle$$

Self-consistency loop

$$\hat{u} \leftarrow \mathbf{min}_{u_{pq}} \mathbf{CF}(u_{pq}) \equiv \mathbf{min}_{u_{pq}} (D^{\mathrm{l}}_{\mathrm{emb}}[u_{pq}] - D^{\mathrm{h}}_{\mathrm{emb}})^2$$

J. Chem. Theory Comput. **12**, 2706–2719 (2016).

Quantum embedding approach

$$\boldsymbol{k}_p - \boldsymbol{k}_q = 0$$
$$\boldsymbol{k}_p + \boldsymbol{k}_r - \boldsymbol{k}_s - \boldsymbol{k}_q = 0$$

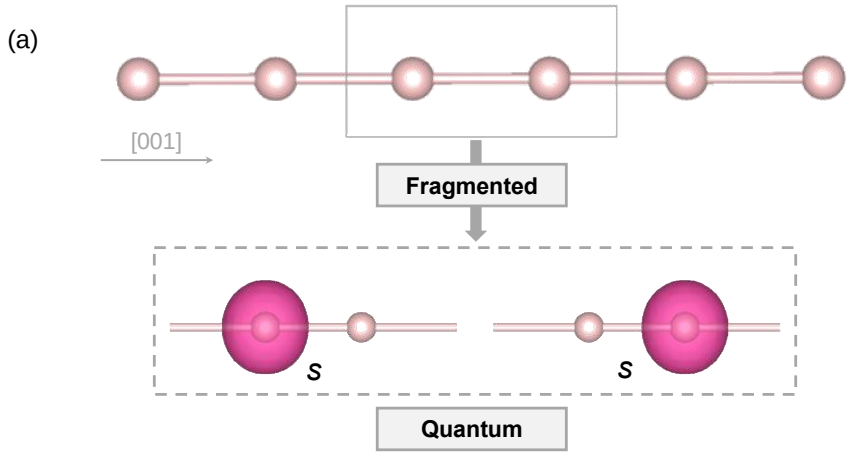
conservation of crystal momentum

Due to the translational symmetry:

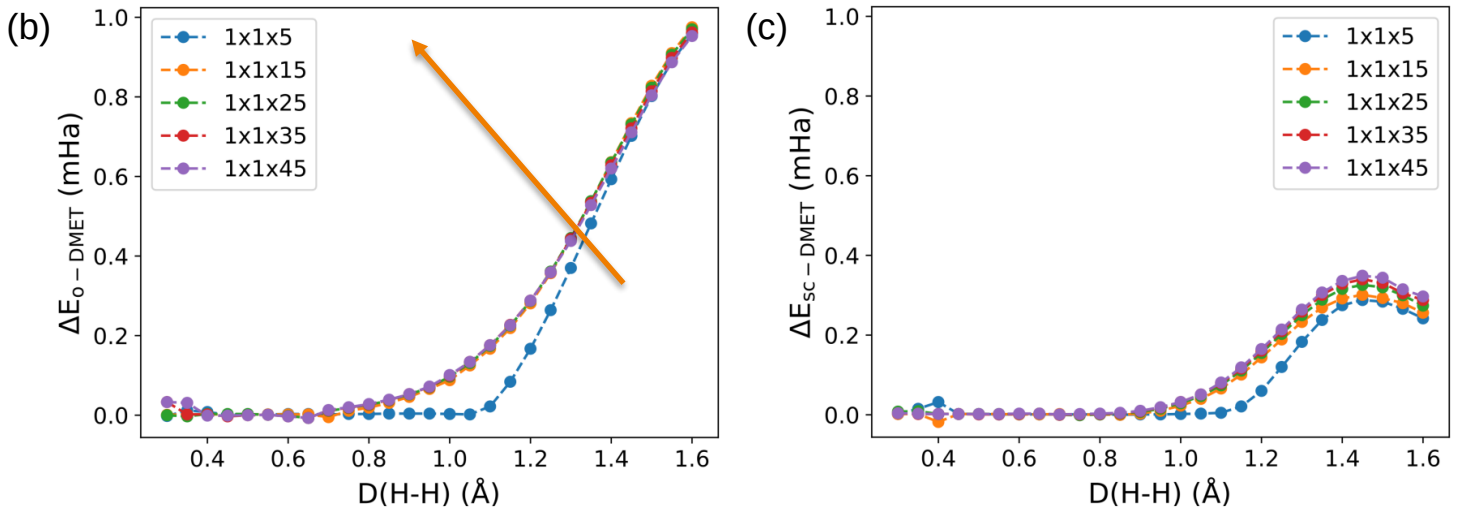
- Only one unit-cell problem needs to be solved;
- Some calculations about integral could be simplified.

$$\hat{H} = \sum_{p,q} \sum_{\mathbf{k}_p \mathbf{k}_q} h_{pq}^{\mathbf{k}_p \mathbf{k}_q} \hat{a}_p^{\mathbf{k}_p \dagger} \hat{a}_q^{\mathbf{k}_q} + \frac{1}{2} \sum_{p,q,r,s} \sum_{\mathbf{k}_p \mathbf{k}_q \mathbf{k}_r \mathbf{k}_s} g_{pqrs}^{\mathbf{k}_p \mathbf{k}_q \mathbf{k}_r \mathbf{k}_s} \hat{a}_p^{\mathbf{k}_p \dagger} \hat{a}_r^{\mathbf{k}_r \dagger} \hat{a}_s^{\mathbf{k}_s} \hat{a}_q^{\mathbf{k}_q}.$$

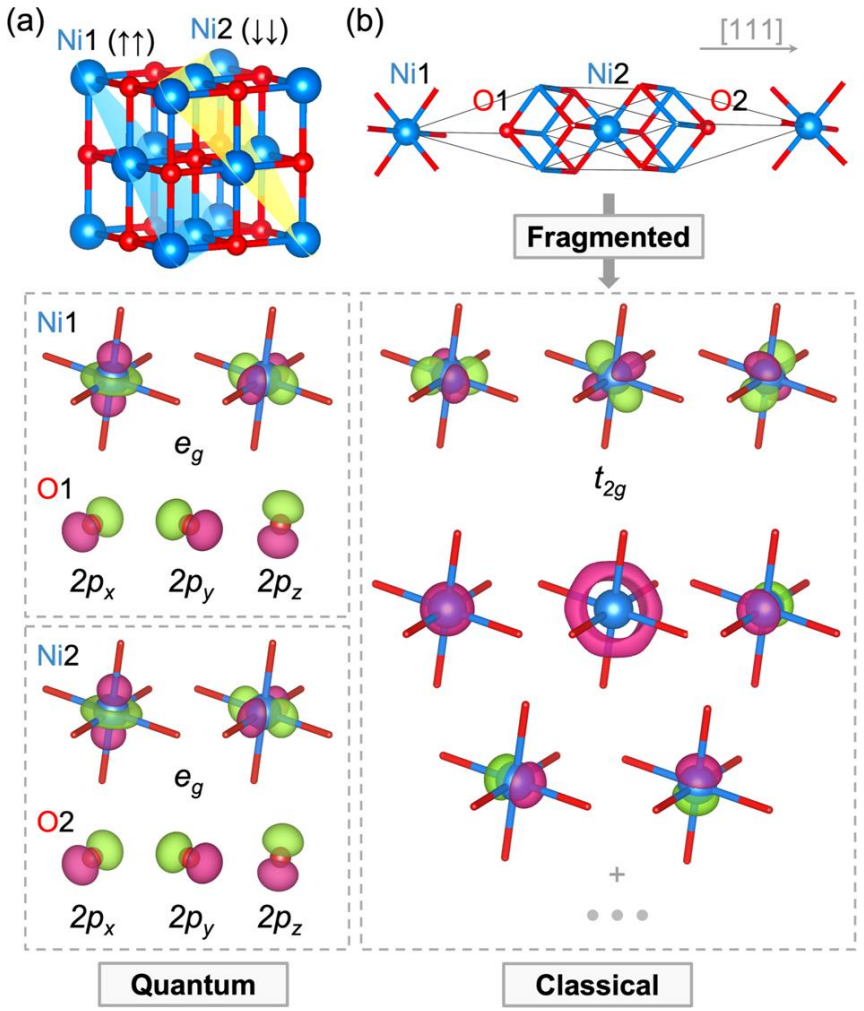
1D Hydrogen chain



Increasing k-points (large supercell)



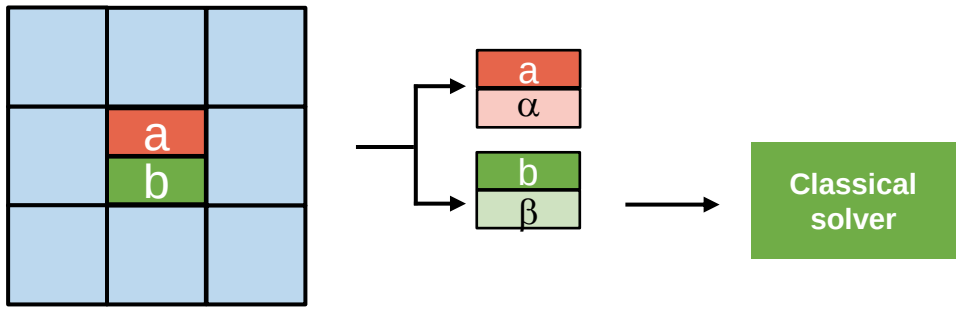
3D NiO



Construction

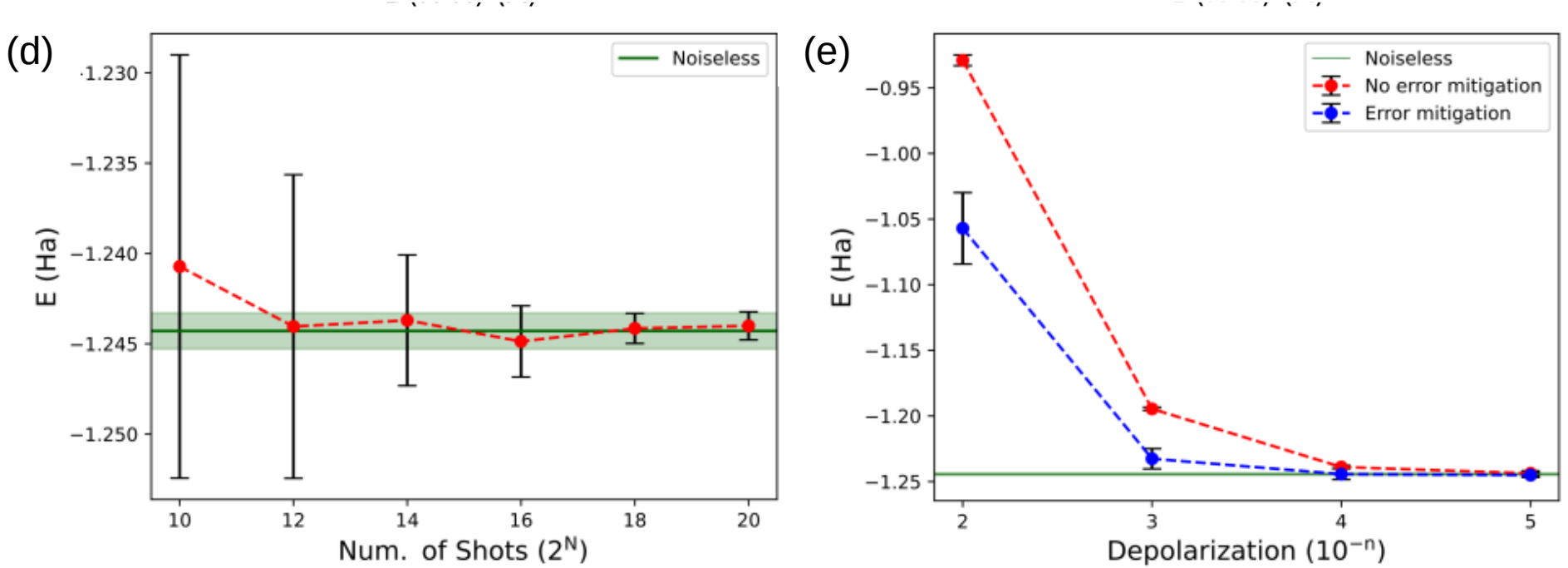
Build bath and H_{emb}

Solve H_{emb}



- Magnetic ordering in NiO
- AFII more stable than FM （consistent with experiments）
- 9984 qubits -> 20 qubits

Noisy simulation and noise mitigation



Resource comparison: molecules and solids

$$N_{\mathrm{ex}} = n_{\mathrm{occ}} n_{\mathrm{vir}} + n_{\mathrm{occ}} n_{\mathrm{vir}} (n_{\mathrm{occ}} n_{\mathrm{vir}} + 1)/2$$

TABLE I. The reduction of the number of qubits and excitation parameters by DMET and ESVQE. cVQE represents conventional VQE without the energy sorting (ES) strategy.

System	Basis	Electrons	Spin-Orbitals	Method	Qubits	Parameters
H_{10}	STO-3G	10	20	cVQE	20	350
				DMET-cVQE	4	2
				DMET-ESVQE	4	1
H_{10}	6-31G	10	40	cVQE	40	2925
				DMET-cVQE	8	14
				DMET-ESVQE	8	10
C_6H_8+H_2	STO-3G	34	68	cVQE	68	42194
				DMET-cVQE	16	152
				DMET-ESVQE	16	105
C_{18}	STO-3G	72	144	cVQE	144	841752
				DMET-cVQE	16	152
				DMET-ESVQE	16	44
C_{18}	cc-pVDZ	72	144	cVQE	144	841752
				DMET-cVQE	16	152
				DMET-ESVQE	16	43

TABLE II. A survey on the number of required qubit for the largest simulation of each system studied in this work

System	Number of cGTOs (per unit cell)	**k**-point	Number of Qubits Without embedding	Number of Qubits This work
1D Hydrogen chain	2	$11^1 = 11$	44	8
2D h-BN	26	$7^2 = 49$	2548	12
3D NiO	78	$4^3 = 64$	9984	20

Li W, Huang Z, Cao C, et al., ***Chem. Sci.,*** 2022

Summary

- Introduce periodic DMET to quantum simulation of materials
- Tests on H-Chain, hBN and NiO
- Qubit reduction for strongly correlated materials
- More unified framework - Hybrid Tensor networks [Yuan, Sun, et al PRL 2021]
- DMET for molecules [Li, et al Chemical Science 2022]