

# A mathematical introduction to quantum embedding theory

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# Outline

## Introduction

Quantum impurity problem and quantum embedding theory

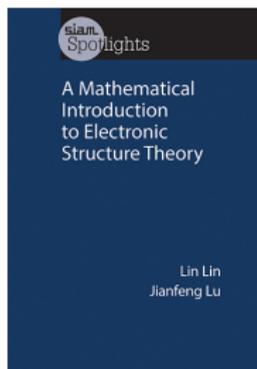
Density matrix embedding theory

Matching condition: “glue” in DMET

Conclusion

## Some additional introductory materials

Particularly for students / researchers with Math background. Physics / Chemistry readers *may* find some discussions clarifying too.



See more references from tutorial talks by [Jianfeng Lu](#) and [Alexandre Tkatchenko](#) on Tuesday.

See [Robert Webber](#)'s tutorial talk on Wednesday (containing an excellent introduction of second quantization)

See Szalay et al, *Tensor Product Methods and Entanglement Optimization for Ab Initio Quantum Chemistry*, IJQC 2015

## (Crash course on) second quantization

- $L$  sites (spin-orbitals). Occupation number basis set

$$\{|\mathbf{s}_1, \dots, \mathbf{s}_L\rangle\}_{\mathbf{s}_i \in \{0,1\}, i=1, \dots, L}$$

orthonormal basis set satisfying

$$\langle \mathbf{s}_{i_1}, \dots, \mathbf{s}_{i_L} | \mathbf{s}_{j_1}, \dots, \mathbf{s}_{j_L} \rangle = \delta_{i_1 j_1} \cdots \delta_{i_L j_L}. \quad (1)$$

spans the Fock space  $\mathcal{F}$  (dimension:  $2^L$ ).

vacuum state:  $|0\rangle = |0, \dots, 0\rangle = |0^L\rangle$ .

- A quantum state  $|\psi\rangle \in \mathcal{F}$ : linear combination of occupation number basis elements:

$$|\Psi\rangle = \sum_{\mathbf{s}_1, \dots, \mathbf{s}_L \in \{0,1\}} \Psi(\mathbf{s}_1, \dots, \mathbf{s}_L) |\mathbf{s}_1, \dots, \mathbf{s}_L\rangle, \quad \Psi(\mathbf{s}_1, \dots, \mathbf{s}_L) \in \mathbb{C}.$$

normalization condition:

$$\langle \Psi | \Psi \rangle = \sum_{\mathbf{s}_1, \dots, \mathbf{s}_L \in \{0,1\}} |\Psi(\mathbf{s}_1, \dots, \mathbf{s}_L)|^2 = 1.$$

## (Crash course on) second quantization

- $a_i, a_i^\dagger, n_i = a_i^\dagger a_i$ : annihilation, creation, number operator at site  $i$ . Sparse matrix of size  $2^L$ .
- Jordan-Wigner transformation (writing down matrix elements)

$$a_p^\dagger |s_1, \dots, s_L\rangle = (-1)^{\sum_{q=1}^{p-1} s_q} (1 - s_p) |s_1, \dots, 1 - s_p, \dots, s_L\rangle,$$

$$a_p |s_1, \dots, s_L\rangle = (-1)^{\sum_{q=1}^{p-1} s_q} s_p |s_1, \dots, 1 - s_p, \dots, s_L\rangle,$$

$$n_p |s_1, \dots, s_L\rangle = s_p |s_1, \dots, s_L\rangle, \quad p = 1, \dots, L.$$

- Verify:

$$|s_1, \dots, s_L\rangle = (a_1^\dagger)^{s_1} \dots (a_L^\dagger)^{s_L} |0\rangle$$

- Canonical anti-commutation relation (CAR):

$$\{a_i, a_j\} = \{a_i^\dagger, a_j^\dagger\} = 0, \quad \{a_i^\dagger, a_j\} = \delta_{ij}.$$

- $\hat{N} := \sum_p n_p$ : total number operator

## Quantum many-body problem

- Many-body Hamiltonian (dimension:  $2^L$ )

$$\hat{H} = \underbrace{\hat{H}_0}_{\text{non-interacting}} + \underbrace{\hat{H}_1}_{\text{interacting}}$$

$$\hat{H}_0 = \sum_{ij=1}^L T_{ij} a_i^\dagger a_j, \quad \hat{H}_1 = \frac{1}{2} \sum_{ijkl=1}^L V_{ijkl} a_i^\dagger a_j^\dagger a_l a_k.$$

- Ground state

$$\min_{|\Psi\rangle \in \mathcal{F}, \langle \Psi | \Psi \rangle = 1} \langle \Psi | \hat{H} - \mu \hat{N} | \Psi \rangle$$

- Chemical potential  $\mu$ : **Lagrange multiplier** chosen so that for a pre-specified integer  $N_e \in \{0, 1, \dots, L\}$

$$\langle \Psi | \hat{N} | \Psi \rangle = N_e.$$

WLOG set  $\mu = 0$ . Can also be done by restricting to the  $N_e$ -particle sector of  $\mathcal{F}$ .

## Simplest setting: non-interacting system

- $\hat{H} = \hat{H}_0 = \sum_{ij=1}^L T_{ij} a_i^\dagger a_j$ . Diagonalize  $T$  (cost:  $\mathcal{O}(L^3)$ )

$$T \phi_k = \varepsilon_k \phi_k, \quad \varepsilon_1 \leq \dots \leq \varepsilon_{N_e} < 0 < \varepsilon_{N_e+1} \leq \dots$$

$[\phi_1, \dots, \phi_L] \in \mathbb{C}^{L \times L}$  is a unitary matrix.

- Basis rotation (still satisfy CAR)

$$c_k^\dagger = \sum_i a_i^\dagger \phi_{ik}, \quad c_k = \sum_i a_i \phi_{ik}^*, \quad \hat{H}_0 = \sum_k \varepsilon_k c_k^\dagger c_k.$$

- Ground state

$$|\Psi\rangle = c_1^\dagger \dots c_{N_e}^\dagger |0^L\rangle$$

Ground state energy  $E = \sum_{k=1}^{N_e} \varepsilon_k$

- Also called “quadratic Hamiltonian” / “single particle picture”: still  $N_e$  particles, but they do not interact except through Pauli exclusion principle.

## Single particle reduced density matrix (1-RDM)

- For quantum many-body state  $|\Psi\rangle$ ,  $D_{ij} = \langle \Psi | a_j^\dagger a_i | \Psi \rangle$ . Hermitian matrix of size  $L \times L$ .
- Electron density (vector of length  $L$ )

$$\rho_i = D_{ii} = \langle \Psi | n_i | \Psi \rangle = \sum_{s_1, \dots, s_L} |\psi(s_1, \dots, s_L)|^2 s_i.$$

- Non-interacting system:  $|\Psi\rangle = c_1^\dagger \dots c_{N_e}^\dagger |0^L\rangle$ .  $\Phi = [\phi_1, \dots, \phi_{N_e}]$ .
- $\tilde{D}_{kk'} = \langle \Psi | c_{k'}^\dagger c_k | \Psi \rangle = \delta_{kk'} \theta_k$ ,  $\theta_k = \begin{cases} 1, & k \leq N_e, \\ 0, & k > N_e. \end{cases}$
- Basis rotation

$$c_k^\dagger = \sum_i a_i^\dagger \phi_{ik} \quad \Rightarrow \quad a_i^\dagger = \sum_k c_k^\dagger \phi_{ik}^*$$

$$D_{ij} = \sum_{kk'} \phi_{ik} \phi_{jk'}^* \langle \Psi | c_{k'}^\dagger c_k | \Psi \rangle = \sum_{k=1}^{N_e} \phi_{ik} \phi_{jk}^* = (\Phi \Phi^\dagger)_{ij}.$$

## Single particle reduced density matrix (1-RDM)

For non-interacting system

- $D$  is idempotent ( $D^2 = D$ ) of rank  $N_e$ . Eigenvalues are 0 or 1.
- $D$  is uniquely determined by  $\Phi$ .
- Eigendecomposition of  $D \Rightarrow \Phi \Rightarrow |\Psi\rangle$ .

For interacting system, generally  $0 \preceq D \preceq 1$ , and  $D \not\Rightarrow |\Psi\rangle$ .

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Density matrix embedding theory

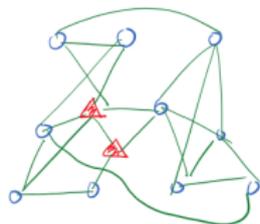
Matching condition: “glue” in DMET

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## Next simplest setup: quantum impurity problem

- Graph  $G = (V, E)$ .  $T_{ij} \neq 0$  if  $(i, j) \in E$ .
- Subset of vertices  $V' \subseteq V$ ,  $V_{ijkl} \neq 0$  if  $i, j, k, l \in V'$ .

Examples:



○ quadratic

△ quartic



$a_1^+ \dots a_m^+ \quad a_{m+1}^+ \quad \dots \quad a_L^+$

## Quantum impurity problem

- $\hat{H}_1 = \sum_{ijkl=1}^m V_{ijkl} a_i^\dagger a_j^\dagger a_l a_k$ ,  $m \ll L$
- Example: single-impurity Anderson model (SIAM)

$$\hat{H} = \left\{ \sum_{k\sigma} \varepsilon_{k\sigma} c_{k\sigma}^\dagger c_{k\sigma} + \sum_{k\sigma} \left( V_k f_\sigma^\dagger c_{k\sigma} + V_k^* c_{k\sigma}^\dagger f_\sigma \right) + \sum_{\sigma} \varepsilon_f f_\sigma^\dagger f_\sigma \right\} \\ + U f_\uparrow^\dagger f_\uparrow f_\downarrow^\dagger f_\downarrow$$

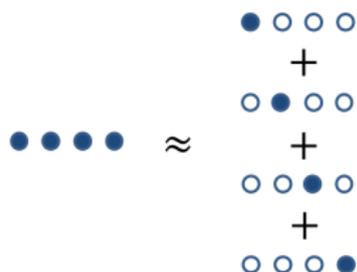
$f$ : fragment.  $c$ : environment.  $a^\dagger = (f^\dagger, c^\dagger)$ .

- $N_e = \mathcal{O}(L)$ :  $\infty$  in the thermodynamic limit.
- Complexity for estimating ground state energy  $E$  to precision  $\gamma$ <sup>1</sup>:

$$\mathcal{O}(L^3) \exp \left( \mathcal{O}(m \log^3(m\gamma^{-1})) \right).$$

<sup>1</sup>[Bravyi-Gosset, CMP 2017]

# Quantum embedding theory



- Cannot solve the problem with  $L$  sites directly
- Willing to solve a series of problems of size  $m \ll L$  with cost up to  $\mathcal{O}(\text{poly}(L) \exp(cm))$ .
- Often more desirable to use methods with cost  $\mathcal{O}(\exp(cm))$  or  $\mathcal{O}(\text{poly}(L) + \exp(cm))$ .
- Often more of a “recipe” than a rigorous theory (except in some extreme regimes such as **non-interacting** systems)

# Examples of quantum embedding theory

- Dynamical mean-field theory (DMFT)<sup>1</sup>
- Density matrix embedding theory (DMET)<sup>2</sup>
- Self energy embedding theory (SEET)<sup>3</sup>
- Projection based embedding theory<sup>4</sup>
- Constrained RPA embedding<sup>5</sup>
- Variational embedding theory<sup>6</sup>
- ...many improvements and many more theories <sup>7</sup>

<sup>1</sup>[Georges and Kotliar, PRB 1992] [Georges et al, Rev. Mod. Phys. 1996]

<sup>2</sup>[Knizia, Chan, PRL 2012][Wouters et al, JCTC 2015]

<sup>3</sup>[Lan et al, JCP 2015][Lan,Zgid,JPCL 2017]

<sup>4</sup>[Manby et al, JCTC 2012]

<sup>5</sup>[Ma,Govini, Galli, npj Comput. Mater. 2020][Ma et al, JCTC 2021]

<sup>6</sup>[Lin, Lindsey, CPAM 2022][Khoo, Lindsey, arXiv:2106.02682]

<sup>7</sup>See review: Sun, Chan, *Quantum embedding theories*, ACR 2016

## Some shared commonalities

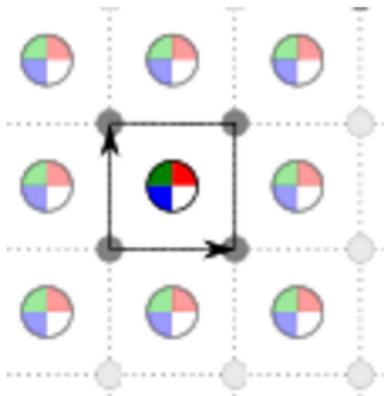
- A “high-level” solver: solve impurity problems accurately with cost (up to)  $\mathcal{O}(\text{poly}(L) \exp(cm))$
- A “low-level” solver: solve a global, often **non-interacting** problem with cost  $\mathcal{O}(\text{poly}(L))$ , and provide some “environmental” information to the high level solver
- Assemble quantities from each high level solver to obtain a global quantity (for instance, 1-RDM, Green’s function..)
- A “matching condition”: quantities assembled from the high-level solvers and obtained from the low-level solver should match. This often results in a **Lagrange multiplier** that glues together the high-level and low-level solvers.

## Some perspectives

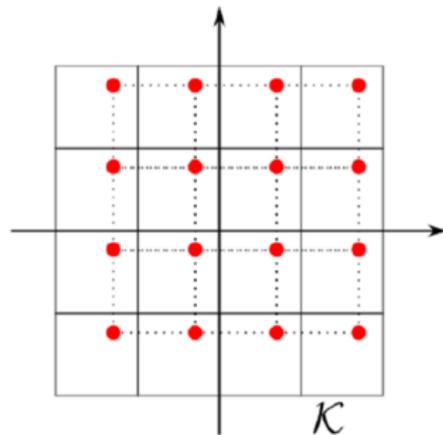
- Energy / matching condition usually cannot be derived from a variational principle  $\Rightarrow$  energy is often **not** variational. Potential issues with force computations etc.
- Usually **no guarantee** that the matching condition can be satisfied, and often there isn't a plan B.
- Despite all the drawbacks, quantum embedding methods are often viewed as **one of the very few affordable ways** to solve correlated electronic structure problems<sup>1</sup>
- Share similarities to **domain decomposition** methods (for solving PDEs), but are also fundamentally different: often couple many problems of sizes  $2^{O(m)}$  to solve a problem of size  $2^L$ .
- Offer a particularly attractive alternative for materials science (to handle translational invariance / local impurities)

<sup>1</sup>At least before machine learning / quantum computing consumes everything..

## Two equivalent perspective of solids



Real space  
Translation invariance



Reciprocal space  
Momentum conservation  
 $\mathbf{k}_i + \mathbf{k}_j = \mathbf{k}_a + \mathbf{k}_b$

Embedding uses a real space perspective. Compute only for one fragment.

## The rest of the talk

Our goals are modest:

- Why low-level solvers can provide **any** useful information for the high-level solver.
- Why the matching condition can be satisfied under **any** circumstance
- Focus on DMET

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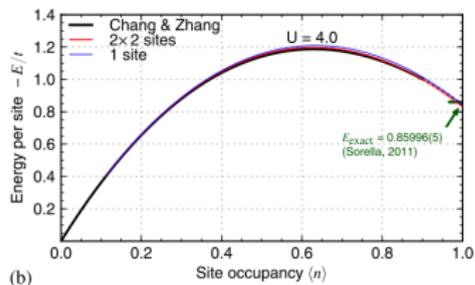
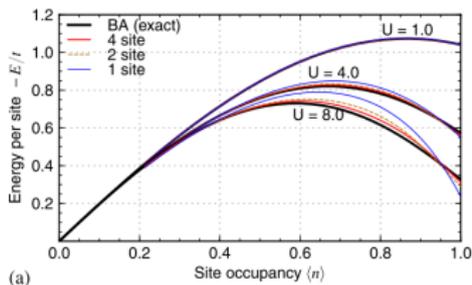
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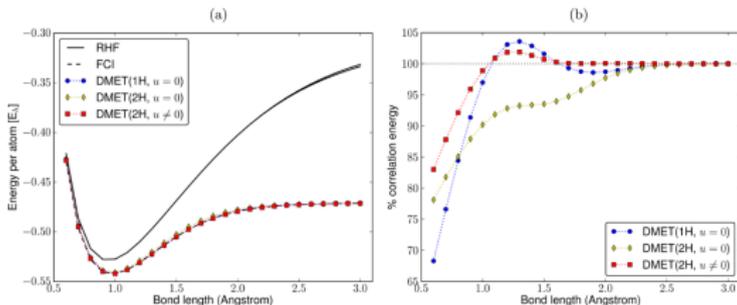
Conclusion

# DMET for strongly correlated systems

## Hubbard model (1D and 2D)<sup>1</sup>



## Hydrogen ring (sto-6G)<sup>2</sup>



<sup>1</sup>Knizia, Chan PRL 2012

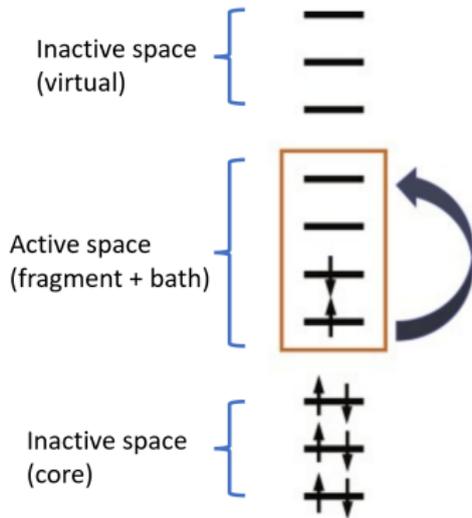
<sup>2</sup>Wouters et al, JCTC 2015

## DMET: high level idea

- For the ground state of an arbitrarily large quantum system, a given fragment is coupled to a finite number of “bath orbitals”
- Obtain information from “fragment + bath”. Remove contribution from core (low energy) and virtual (high energy) states.
- Since bath cannot be obtained exactly, iteratively update the bath to satisfy a matching condition.

# Partition of the orbital space

Partition  $L$  orbitals (after proper rotation) into **fragment**, **bath**, **core**, **virtual** orbitals. This appears in many quantum chemistry methods (e.g., CASSCF)



## A finite number of bath orbitals

- Fragment:  $x = \{1, \dots, L_A\}$ . Environment:  $\{L_A + 1, \dots, L\}$ .
- General argument: Schmidt decomposition (SVD)

$$\Psi(\mathbf{s}_1, \dots, \mathbf{s}_L) = \sum_{\alpha=1}^{2^{L_A}} \sigma_{\alpha} u_{\alpha}(\mathbf{s}_1, \dots, \mathbf{s}_{L_A}) v_{\alpha}(\mathbf{s}_{L_A+1}, \dots, \mathbf{s}_L)$$

- $u_{\alpha}$ : fragment orbital.  $v_{\alpha}$ : bath orbital. Possibly much fewer than  $2^{L_A}$ , and independent of  $L$ .
- **Problem**: (1)  $v_{\alpha}$  itself requires exponential cost (2) can only be obtained with the information of the exact ground state  $\Psi$ .
- DMET's solution: Construct some approximate  $v_{\alpha}$  from **non-interacting** systems.
- Will demonstrate a **linear algebraic** derivation (different from original presentation of the method)





## Bath construction for non-interacting systems

- Rewrite

$$\Phi = \begin{matrix} L_A \\ L-L_A \end{matrix} \begin{pmatrix} \Phi_A \\ \Phi_B \end{pmatrix} = \begin{pmatrix} U_A \Sigma_A V^\dagger \\ U_B \Sigma_B V^\dagger + U_{\text{core}} V_\perp^\dagger \end{pmatrix}.$$

$$D = \begin{pmatrix} U_A \Sigma_A^2 U_A^\dagger & U_A \Sigma_A \Sigma_B U_B^\dagger \\ U_B \Sigma_B \Sigma_A U_A^\dagger & U_B \Sigma_B^2 U_B^\dagger + U_{\text{core}} U_{\text{core}}^\dagger \end{pmatrix} = \begin{pmatrix} D_{11} & D_{12} \\ D_{21} & D_{22} \end{pmatrix}.$$

- Virtual orbitals do not contribute to  $\Phi$  or  $D$ .
- Next we show that core orbitals do not directly contribute to the computation of the fragment block  $D_{11}$ .

## Basis rotation

- Define the basis  $\tilde{\Phi} = [\Phi_X^{\text{frag}}, \Phi_X^{\text{bath}}, \Phi_X^{\text{core}}, \Phi_X^{\text{vir}}]$

$$\Phi_X^{\text{frag}} = \begin{pmatrix} I_{L_A} \\ 0 \end{pmatrix}, \Phi_X^{\text{bath}} = \begin{pmatrix} 0 \\ U_B \end{pmatrix}, \Phi_X^{\text{core}} = \begin{pmatrix} 0 \\ U_{\text{core}} \end{pmatrix}, \Phi_X^{\text{vir}} = \begin{pmatrix} 0 \\ U_{\text{vir}} \end{pmatrix}.$$

- 1-RDM takes a decoupled form in the rotated basis

$$\tilde{\Phi}^\dagger D \tilde{\Phi} = \begin{pmatrix} U_A \Sigma_A^2 U_A^\dagger & U_A \Sigma_A \Sigma_B & 0 & 0 \\ \Sigma_B \Sigma_A U_A^\dagger & \Sigma_B^2 & 0 & 0 \\ 0 & 0 & I_{N_e - L_A} & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}$$

- In physics terminology, only the orbitals  $\Phi_X^{\text{bath}}$  are **entangled** to the fragment orbitals  $\Phi_X^{\text{frag}}$ .
- Number of electrons in the impurity:

$$N_x = N_e - (N_e - L_A) = L_A$$

Impurity is at **half filling** ( $N_x/L_x = 1/2$ ).

## Impurity Hamiltonian

- Impurity=fragment+bath. Impurity orbitals (size:  $L_x = 2L_A$ ):

$$\Phi_x = \left( \Phi_x^{\text{frag}} \quad \Phi_x^{\text{bath}} \right) = \begin{pmatrix} I_{L_A} & 0 \\ 0 & U_B \end{pmatrix}.$$

- Impurity Hamiltonian (size:  $2^{L_x}$ )

$$\hat{H}_x = \sum_{p,q=1}^{L_x} b_p^\dagger \tilde{T}_{pq} b_q, \quad \tilde{T} = \Phi_x^\dagger T \Phi_x.$$

Note  $b_p = a_p, p = 1, \dots, L_A$ .

- 1-RDM ( $D_{11}$  block) can be recovered **exactly** by diagonalizing  $\tilde{T}$

$$D_x = \begin{pmatrix} U_A \Sigma_A^2 U_A^\dagger & U_A \Sigma_A \Sigma_B \\ \Sigma_B \Sigma_A U_A^\dagger & \Sigma_B^2 \end{pmatrix}.$$

## RECIPE for interacting Hamiltonian

- Revisit the interacting Hamiltonian

$$\hat{H} = \sum_{ij=1}^L T_{ij} a_i^\dagger a_j + \frac{1}{2} \sum_{ijkl=1}^L V_{ijkl} a_i^\dagger a_j^\dagger a_l a_k.$$

- Assume the partition  $\tilde{\Phi} = [\Phi_x^{\text{frag}}, \Phi_x^{\text{bath}}, \Phi_x^{\text{core}}, \Phi_x^{\text{vir}}]$  is defined via **some non-interacting** Hamiltonian with coefficient matrix  $T$ .
- $\Phi_x^{\text{core}}$  will now contribute a Hartree-Fock background potential (sometimes called a embedding potential)
- Also need a proper chemical potential to satisfy the **half filling condition**  $N_x = L_A$  (or equivalently, restrict to the  $L_A$ -particle sector of the reduced Fock space)

# Interacting impurity Hamiltonian

- Impurity Hamiltonian

$$\hat{H}_x = \sum_{p,q=1}^{L_x} b_p^\dagger \tilde{T}_{pq} b_q + \frac{1}{2} \sum_{pqrs=1}^{L_x} \tilde{V}_{pqrs} b_p^\dagger b_q^\dagger b_s b_r$$

- Coefficients

$$\begin{aligned} \tilde{T}_{pq} &= \sum_{ij} (\Phi_x)_{ip}^* T_{ij} (\Phi_x)_{jq} \\ &+ \sum_{ijkl,n} (V_{ijkl} - V_{ijlk}) (\Phi_x)_{ip}^* (\Phi_x^{\text{core}})_{jn}^* (\Phi_x)_{kq} (\Phi_x^{\text{core}})_{ln} \end{aligned}$$

$$\tilde{V}_{pqrs} = \sum_{ijkl} V_{ijkl} (\Phi_x)_{ip}^* (\Phi_x)_{jq}^* (\Phi_x)_{kr} (\Phi_x)_{ls}$$

## High level 1-RDM

- Solve the many-body ground state problem

$$\min_{|\Psi\rangle \in \mathcal{F}_x, \langle \Psi | \Psi \rangle = 1, \langle \Psi | \hat{N}_x | \Psi \rangle = L_A} \langle \Psi | \hat{H}_x | \Psi \rangle$$

- Compute 1-RDM  $(P_x)_{pq} = \langle \Psi | b_q^\dagger b_p | \Psi \rangle$ . Only trust the fragment part

$$P_x = \begin{pmatrix} P_x^{\text{frag}} & * \\ * & * \end{pmatrix}$$

- Enumerate all fragments, obtain **diagonal blocks** of 1-RDM. Assume  $N_f$  fragments, (uniform) non-overlapping partitioning.

$$P = \begin{pmatrix} P_1^{\text{frag}} & & & \\ & P_2^{\text{frag}} & & \\ & & \dots & \\ & & & P_{N_f}^{\text{frag}} \end{pmatrix} =: \bigoplus_{x=1}^{N_f} P_x^{\text{frag}}$$

## Issues not discussed so far

- We have  $0 \preceq P_x^{\text{frag}} \preceq 1$ , how to satisfy  $\text{Tr}[P] = N_e$ ? Introduce another global Lagrange multiplier (chemical potential fitting).
- How to compute the total energy? Compute both 1-RDM and 2-RDM and assemble the total energy from all fragments.
- Special techniques: orbital localization, charge self consistency, non-interacting bath..
- How to iteratively update the bath and core orbitals? [Matching condition](#).

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## Matching condition

- Low-level density matrix  $D$  should
  - (1) minimize a certain energy functional
  - (2) match  $D_x = (\Phi_x^{\text{frag}})^\dagger D \Phi_x^{\text{frag}}$  with  $P_x^{\text{frag}}$ .
- Match 1-RDM blocks  $\Rightarrow$  Lagrange multiplier  $u_x \in \mathbb{C}^{L_A \times L_A}$  for each impurity  $x$ , called correlation potential

$$U = \bigoplus_{x=1}^{N_f} u_x$$

- Diagonalize  $T + U$  to obtain  $D \Rightarrow$  function  $D = \mathcal{D}(T + U, N_e)$ .
- $T$  can be replaced by any fixed Fock matrix.

# Domain

- Block structure

$$\mathcal{S} = \left\{ A = \bigoplus_{x=1}^{N_f} A_x \mid A_x \in \mathbb{C}^{L_A \times L_A}, A_x = A_x^\dagger \text{ for } x = 1, \dots, N_f \right\},$$

- Domain of correlation potential  $U$  (gauge freedom)

$$\mathcal{S}^0 := \{A \in \mathcal{S} \mid \text{Tr}[A] = 0\}$$

- Domain of high level density matrix

$$\mathcal{S}_+^{N_e} := \{A \in \mathcal{S} \mid \text{Tr}[A] = N_e, 0 \preceq A \preceq 1\}$$

## Least squares (LS-DMET)

- Least squares fitting<sup>1</sup>

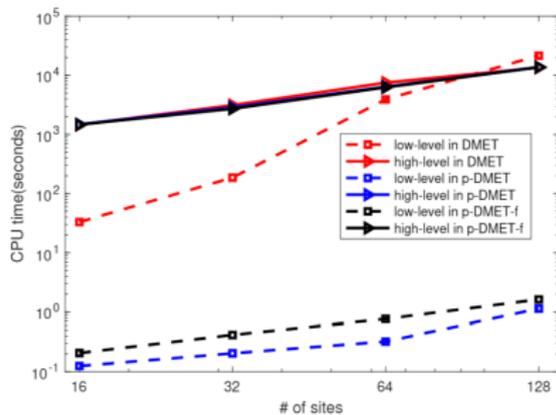
$$\min_U \sum_x \left\| P_x^{\text{frag}} - (\Phi_x^{\text{frag}})^\dagger \mathcal{D}(T + U, N_e) \Phi_x^{\text{frag}} \right\|_F^2.$$

- Nonlinear optimization. Local minimizer.
- Inexact matching<sup>2</sup>.
- Gapless problem:  $\lambda_{N_e}(T + U) = \lambda_{N_e+1}(T + U)$ . Instability.
- Seem to be **independent** issues?

<sup>1</sup>[Knizia, Chan, PRL 2012]

<sup>2</sup>Physical argument and density embedding [Bulik, Scuseria, Dukelsky, PRB 2014]

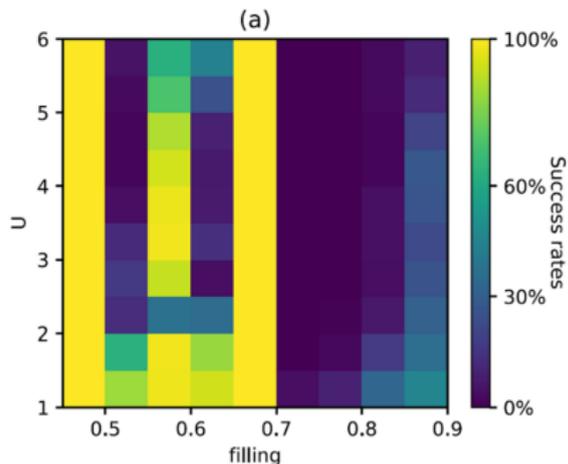
# Potential issues in LS-DMET



64 × 2 Hubbard model

Long run time (mainly due to a large number of iterations)

(Wu et al, JCP 2019)



6 × 6 Hubbard model, 1000 samples with random onsite potential.

Stuck at local minima

(Wu et al, JCP 2020)

## Reformulate the fitting problem

- Energy functional

$$F(U) = \mathcal{E}_{N_e}[T + U] = \sum_{i=1}^{N_e} \lambda_i(T + U) = \text{Tr}[(T + U)\mathcal{D}(T + U, N_e)]$$

- When  $T + U$  is **gapped**, i.e.,  $\lambda_{N_e+1}(T + U) > \lambda_{N_e}(T + U)$

$$\nabla_{u_x} F(T + U) = (\Phi_x^{\text{frag}})^\dagger \mathcal{D}(T + U, N_e) \Phi_x^{\text{frag}}$$

- $F(U)$  is **concave**

$$\begin{aligned} & F(\alpha U_1 + (1 - \alpha) U_2) \\ & \geq \alpha \text{Tr}[(T + U_1)\mathcal{D}(T + U_1, N_e)] + (1 - \alpha) \text{Tr}[(T + U_2)\mathcal{D}(T + U_2, N_e)] \\ & = \alpha F(U_1) + (1 - \alpha) F(U_2). \end{aligned}$$

## Convex optimization (CVX-DMET)

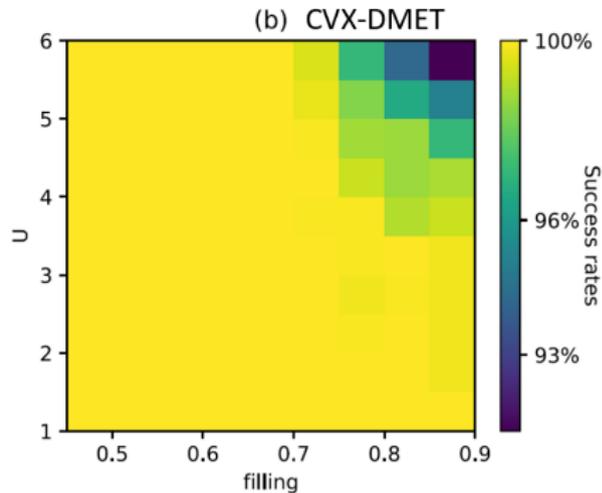
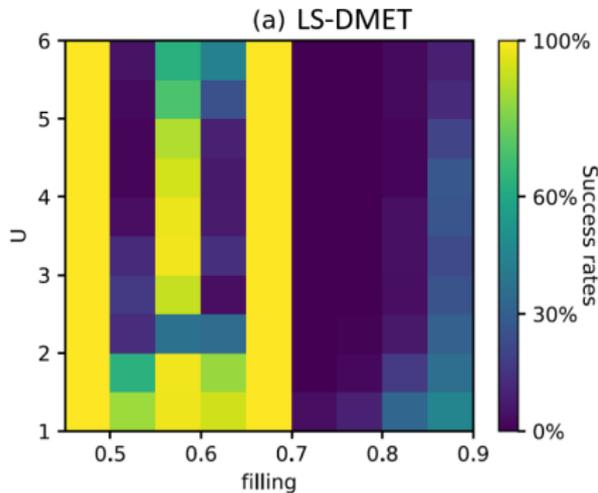
- Concave conjugate: Legendre–Fenchel transform

$$F^*(P) = \inf_{U \in \mathcal{S}^0} \left\{ \sum_{x=1}^{N_f} \text{Tr}[P_x u_x] - F(U) \right\}, \quad P \in \mathcal{S}_+^{N_e}$$

- Find the correlation potential via convex (or concave) optimization (e.g., CVX). Robust global minima.
- Example code (mask enforces sparsity pattern)

```
cvx_begin
    variable U(L,L) symmetric
    minimize ( trace(U*P) - lambda_sum_smallest(T+U,Ne) )
    subject to
        U .* mask == 0
cvx_end
```

# Improved robustness



$6 \times 6$  Hubbard model, 1000 samples with random onsite potential.

## Gapless and inexact matching

### Theorem

Assuming  $P \in \mathcal{S}_{++}^{N_e} = \{A \in \mathcal{S} \mid \text{Tr}[A] = N_e, 0 \prec A \prec 1\}$ , the evaluation of  $F^*(P)$  admits an optimizer  $U^*$ , and  $P$  lies in the supergradient set of  $F$  at  $U^*$ . If  $F + U^*$  is

1. *gapped*:  $D = \mathcal{D}(F + U^*, N_e)$  achieves exact matching.
2. *gapless and if  $U^*$  is unique*: exact matching is not achievable.

### Remark:

- The condition  $P \in \mathcal{S}_{++}^{N_e}$  ensures that  $P$  is in the relative interior of the domain of  $F^*(= \mathcal{S}_+^{N_e})$ .
- The condition for  $U^*$  to be unique is an open question. A necessary condition is  $N_e \geq L_A$ .

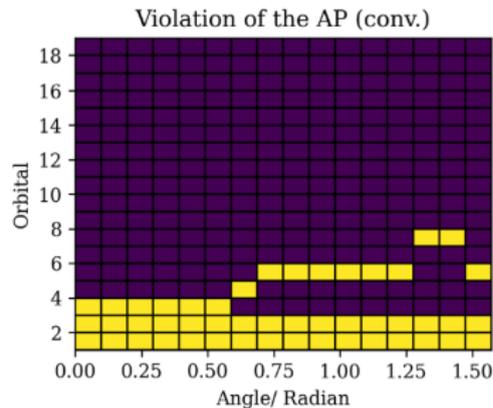
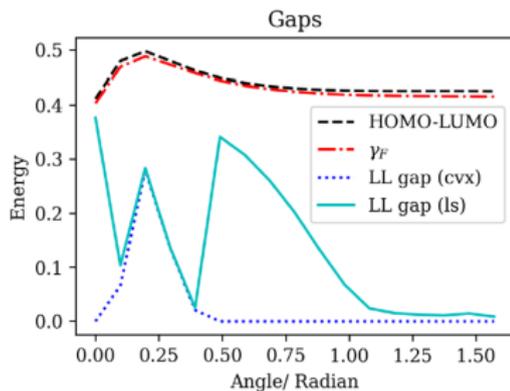
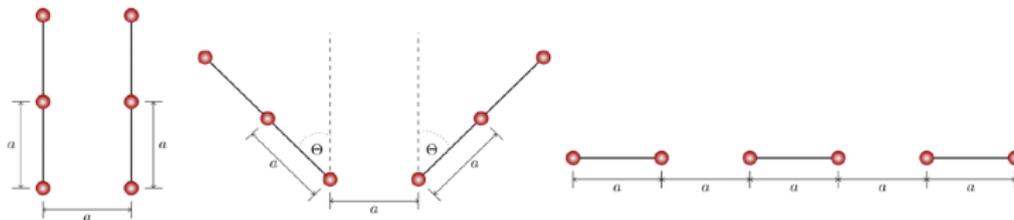
## What to do with gapless problems?

- Gapless problems arise more frequently than we like (e.g., doped Hubbard model)
- Both LS-DMET and CVX-DMET assume **aufbau principle**.
- Consider the optimization problem

$$\left\{ \begin{array}{l} \min_{D \in \mathcal{M}} \quad \text{Tr}(TD), \\ \text{s.t.} \quad D_x = P_x, \quad \forall x, \\ \mathcal{M} = \{D \in \mathbb{C}^{L \times L} \mid D = D^\dagger, \text{Tr}(D) = N_e, D^2 = D\}. \end{array} \right.$$

- Does the minimizer admits  $U$  s.t.  $D = \mathcal{D}(T + U, N_e)$ ? The answer is **no**. Has implication in finding the exact exchange-correlation functional in the discrete setting.
- Can exact matching be achieved?

# Augmented Lagrange multiplier (ALM-DMET)



# Outline

Introduction

Quantum impurity problem and quantum embedding theory

Density matrix embedding theory

Matching condition: “glue” in DMET

Conclusion

# Conclusion

- Quantum embedding theory: **scalable** correlated electronic structure calculation.
- Involve **recipes** / **artistic components**: you can come up with your own embedding theory and maybe it will perform better!
- Energy is typically not variational, with recent exceptions<sup>1</sup>
- Matching condition deserves careful **mathematical scrutiny** and there is room for **algorithmic improvement**, which may impact **practical** quantum embedding calculations.
- Can talk about DMFT etc later if there is interest

<sup>1</sup>[Lin, Lindsey, CPAM 2022][Khoo, Lindsey, arXiv:2106.02682]. See [Michael Lindsey's](#) talk in WS1.

# Acknowledgment

Thank you for your attention!

Lin Lin

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