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)
(Crash course on) second quantization

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

$$\left\{ | s_1, \ldots, s_L \rangle \right\}_{s_i \in \{0,1\}, i=1,\ldots,L}$$

orthonormal basis set satisfying

$$\langle s_{i_1}, \ldots, s_{i_L} | s_{j_1}, \ldots, s_{j_L} \rangle = \delta_{i_1,j_1} \cdots \delta_{i_L,j_L}. \tag{1}$$

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

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

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

$$|\psi\rangle = \sum_{s_1,\ldots,s_L \in \{0,1\}} \psi(s_1,\ldots,s_L) |s_1,\ldots,s_L\rangle, \quad \psi(s_1,\ldots,s_L) \in \mathbb{C}.$$ normalisation condition:

$$\langle \psi | \psi \rangle = \sum_{s_1,\ldots,s_L \in \{0,1\}} |\psi(s_1,\ldots,s_L)|^2 = 1.$$
(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)

\[
\begin{align*}
a^\dagger_p |s_1, \ldots, s_L\rangle &= (-1) \sum_{q=1}^{p-1} s_q (1 - s_p) |s_1, \ldots, 1 - s_p, \ldots, s_L\rangle, \\
a_p |s_1, \ldots, s_L\rangle &= (-1) \sum_{q=1}^{p-1} s_q s_p |s_1, \ldots, 1 - s_p, \ldots, s_L\rangle, \\
n_p |s_1, \ldots, s_L\rangle &= s_p |s_1, \ldots, s_L\rangle, \quad p = 1, \ldots, L.
\end{align*}
\]

- Verify:

\[
|s_1, \ldots, s_L\rangle = (a_1^\dagger)^{s_1} \cdots (a_L^\dagger)^{s_L} |0\rangle
\]

- 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: $2^L$)

$$\hat{H} = \hat{H}_0 + \hat{H}_1$$

\begin{align*}
\hat{H}_0 &= \sum_{ij=1}^L T_{ij} a_i^\dagger a_j, \\
\hat{H}_1 &= \frac{1}{2} \sum_{ijkl=1}^L V_{ijkl} a_i^\dagger a_j^\dagger a_l a_k.
\end{align*}

- 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 $N_e \in \{0, 1, \ldots, L\}$

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

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

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

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

$[\phi_1, \ldots, \phi_L] \in \mathbb{C}^{L \times L}$ 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

$$|\psi\rangle = c_1^\dagger \cdots c_{N_e}^\dagger |0^L\rangle$$

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 $L$)

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

- Non-interacting system: $|\Psi\rangle = c_1^\dagger \cdots c_{N_e}^\dagger |0^L\rangle$. $\Phi = [\phi_1, \ldots, \phi_{N_e}]$.
- $\bar{D}_{kk'} = \langle \psi | c_{k'}^\dagger c_k | \psi \rangle = \delta_{kk'} \theta_k, \quad \theta_k = \begin{cases} 1, & k \leq N_e, \\ 0, & k > N_e. \end{cases}$
- 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 $0 \leq D \leq 1$, and $D \not\Rightarrow |\psi\rangle$. 
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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:
Quantum impurity problem

- \( \hat{H}_1 = \sum_{ijkl=1}^{m} V_{ijkl} a_i^\dagger a_j^\dagger a_l a_k, \quad m \ll L \)

- Example: single-impurity Anderson model (SIAM)

\[
\hat{H} = \left\{ \sum_{k\sigma} \varepsilon_{k\sigma} c_{k\sigma}^\dagger c_{k\sigma} + \sum_{k\sigma} \left( V_{k\sigma} f_{\sigma}^\dagger c_{k\sigma} + V_{k\sigma}^* c_{k\sigma}^\dagger f_{\sigma} \right) + \sum_{\sigma} \varepsilon_{f\sigma} f_{\sigma}^\dagger f_{\sigma} \right\} \\
+ U f_{\uparrow}^\dagger f_{\uparrow}^\dagger f_{\downarrow} f_{\downarrow}
\]

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

- \( N_e = O(L) \): \( \infty \) in the thermodynamic limit.

- Complexity for estimating ground state energy \( E \) to precision \( \gamma \):

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

\[^1\text{[Bravyi-Gosset, CMP 2017]}\]
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 $O(\text{poly}(L) \exp(cm))$.
- Often more desirable to use methods with cost $O(\exp(cm))$ or $O(\text{poly}(L) + \exp(cm))$.
- 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) \(^1\)
- Density matrix embedding theory (DMET) \(^2\)
- Self energy embedding theory (SEET) \(^3\)
- Projection based embedding theory \(^4\)
- Constrained RPA embedding \(^5\)
- Variational embedding theory \(^6\)
- ...many improvements and many more theories \(^7\)

\(^1\) [Georges and Kotliar, PRB 1992] [Georges et al, Rev. Mod. Phys. 1996]
\(^2\) [Knizia, Chan, PRL 2012] [Wouters et al, JCTC 2015]
\(^3\) [Lan et al, JCP 2015] [Lan, Zgid, JPCL 2017]
\(^4\) [Manby et al, JCTC 2012]
\(^5\) [Ma, Govini, Galli, npj Comput. Mater. 2020] [Ma et al, JCTC 2021]
\(^6\) [Lin, Lindsey, CPAM 2022] [Khoo, Lindsey, arXiv:2106.02682]
\(^7\) 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\(^1\)

- 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)

\(^1\)At least before machine learning / quantum computing consumes everything..
Two equivalent perspective of solids

Real space
Translation invariance

Reciprocal space
Momentum conservation
\[ k_i + k_j = k_a + k_b \]

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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DMET for strongly correlated systems
Hubbard model (1D and 2D)\textsuperscript{1}

Hydrogen ring (sto-6G)\textsuperscript{2}

\textsuperscript{1}Knizia, Chan PRL 2012
\textsuperscript{2}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: $x = \{1, \ldots, L_A\}$. Environment: $\{L_A + 1, \ldots, L\}$.

- General argument: Schmidt decomposition (SVD)

$$
\psi(s_1, \ldots, s_L) = \sum_{\alpha=1}^{2^{L_A}} \sigma_\alpha u_\alpha(s_1, \ldots, s_{L_A}) v_\alpha(s_{L_A+1}, \ldots, s_L)
$$

- $u_\alpha$: 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)
Cosine-Sine (CS) decomposition

- Various versions\(^1\). \(Q \in \mathbb{C}^{(m+p) \times k}\) has orthonormal columns (here \(m < k, \ p \geq k\)).

\[
Q = \begin{pmatrix}
\begin{pmatrix}
Q_1 \\
Q_2
\end{pmatrix}
\end{pmatrix}
\]

- Can find unitaries \(U_1, U_2, V\), diagonal matrices \(C, S\)

\[
\begin{pmatrix}
Q_1 \\
Q_2
\end{pmatrix} = \begin{pmatrix}
m \\
p
\end{pmatrix}
\begin{pmatrix}
U_1 & 0 \\
0 & U_2
\end{pmatrix}
\begin{pmatrix}
m \\
\begin{pmatrix}
C & 0 \\
S & 0
\end{pmatrix}
\begin{pmatrix}
k-m \\
p-k
\end{pmatrix}
\begin{pmatrix}
m \\
0
\end{pmatrix}
\begin{pmatrix}
k-m \\
0
\end{pmatrix}
\end{pmatrix}
\begin{pmatrix}
V^\dagger
\end{pmatrix}, \quad C^2 + S^2 = I_m.
\]

\(^1\)see e.g., [Stewart, Numer. Math. 1982][Bai 1992]
Bath construction for non-interacting systems

- Setup: $D = \Phi \Phi^\dagger. \Phi \in \mathbb{C}^{L \times N_e}, \Phi^\dagger \Phi = I_{N_e}. \Phi = \begin{pmatrix} \Phi_A \\ \Phi_B \end{pmatrix}$

- Apply CS decomposition to $\Phi$ (assume $L_A < N_e$)

$$
\Phi = \begin{pmatrix} L_A \\ L_A \\ N_e - L_A \\ L - L_A - N_e \end{pmatrix} \begin{pmatrix} U_A & 0 & 0 & 0 \\ 0 & U_B & U_{\text{core}} & U_{\text{vir}} \\ \Sigma_A & 0 & 0 & 0 \\ 0 & 0 & I & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix} \begin{pmatrix} V^\dagger \\ V_{\perp} \end{pmatrix}.
$$

- $U_A, U_B, U_{\text{core}}, U_{\text{vir}}$: fragment, bath, core, virtual orbitals.

- Diagonal matrices $\Sigma_A, \Sigma_B$ satisfy $\Sigma_A^2 + \Sigma_B^2 = I_{L_A}$. 
Bath construction for non-interacting systems

- **Rewrite**

\[
\Phi = \sum_{L}^{L_{A}} \left( \begin{array}{c} \Phi_{A} \\ \Phi_{B} \end{array} \right) = \left( \begin{array}{c} U_{A} \Sigma_{A} V_{\uparrow} \\ U_{B} \Sigma_{B} V_{\uparrow} + U_{\text{core}} V_{\perp} \end{array} \right).
\]

\[
D = \left( \begin{array}{cccc}
U_{A} \Sigma_{A}^{2} U_{A}^{\dagger} & U_{A} \Sigma_{A} \Sigma_{B} U_{B}^{\dagger} \\
U_{B} \Sigma_{B} \Sigma_{A} U_{A}^{\dagger} & U_{B} \Sigma_{B}^{2} U_{B}^{\dagger} + U_{\text{core}} U_{\text{core}}^{\dagger} \\
\end{array} \right) = \left( \begin{array}{cc}
D_{11} & D_{12} \\
D_{21} & D_{22} \end{array} \right).
\]

- 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 $\Phi = [\Phi^{\text{frag}}_x, \Phi^{\text{bath}}_x, \Phi^{\text{core}}_x, \Phi^{\text{vir}}_x]$

  $\Phi^{\text{frag}}_x = \begin{pmatrix} I_L^0 \\ 0 \end{pmatrix}$, $\Phi^{\text{bath}}_x = \begin{pmatrix} 0 \\ U_B \end{pmatrix}$, $\Phi^{\text{core}}_x = \begin{pmatrix} 0 \\ U_{\text{core}} \end{pmatrix}$, $\Phi^{\text{vir}}_x = \begin{pmatrix} 0 \\ U_{\text{vir}} \end{pmatrix}$.

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

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

- In physics terminology, only the orbitals $\Phi^{\text{bath}}_x$ are entangled to the fragment orbitals $\Phi^{\text{frag}}_x$.

- 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: $L_x = 2L_A$):

$$\Phi_x = \begin{pmatrix} \Phi_x^{\text{frag}} & \Phi_x^{\text{bath}} \end{pmatrix} = \begin{pmatrix} I_{L_A} & 0 \\ 0 & U_B \end{pmatrix}.$$

- 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 $b_p = a_p, p = 1, \ldots, L_A$.

- 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 $$\tilde{\Phi} = [\Phi_{\text{frag}}^x, \Phi_{\text{bath}}^x, \Phi_{\text{core}}^x, \Phi_{\text{vir}}^x]$$ is defined via some non-interacting Hamiltonian with coefficient matrix $$T$$.

• $$\Phi_{\text{core}}^x$$ 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

\[
\tilde{T}_{pq} = \sum_{ij} (\Phi_x)_i^* T_{ij} (\Phi_x)_j
\]

\[
+ \sum_{ijkl,n} (V_{ijkl} - V_{ijlk})(\Phi_x)_i^* (\Phi_{x\text{core}})_j^* (\Phi_x)_k (\Phi_{x\text{core}})_l
\]

\[
\tilde{V}_{pqrs} = \sum_{ijkl} V_{ijkl} (\Phi_x)_i^* (\Phi_x)_j^* (\Phi_x)_k (\Phi_x)_l
\]
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 \((P_x)_{pq} = \langle \psi | b_{q}^{\dag} b_{p} | \psi \rangle\). 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.

\[
P = \begin{pmatrix}
P_{1}^{\text{frag}} & \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots & \cdots \\ \cdots & \cdots & \cdots & \cdots \\ \end{pmatrix} =: \bigoplus_{x=1}^{N_f} P_{x}^{\text{frag}}
\]
Issues not discussed so far

- We have $0 \leq P^\text{frag}_x \leq 1$, 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 $D_x = (\Phi^\text{frag}_x)^\dagger D \Phi^\text{frag}_x$ with $P^\text{frag}_x$.

- 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**

\[
S = \left\{ A = \bigoplus_{x=1}^{N_f} A_x \left| A_x \in \mathbb{C}^{L_A \times L_A}, \ A_x = A_x^\dagger \text{ for } x = 1, \ldots, N_f \right. \right\},
\]

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

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

- **Domain of high level density matrix**

\[
S_{+}^{N_e} := \{ A \in S \mid \text{Tr}[A] = N_e, \ 0 \preceq A \preceq 1 \}\]
Least squares (LS-DMET)

• Least squares fitting\(^1\)

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

• Nonlinear optimization. Local minimizer.

• Inexact matching\(^2\).

• Gapless problem: \(\lambda_{N_e}(T + U) = \lambda_{N_e+1}(T + U)\). Instability.

• Seem to be independent issues?

\(^1\)Knizia, Chan, PRL 2012

\(^2\)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**

\[
F(\alpha U_1 + (1 - \alpha) U_2) \\
\geq \alpha \text{Tr}[(T + U_1) \mathcal{D}(T + U_1, N_e)] + (1 - \alpha) \text{Tr}[(T + U_2) \mathcal{D}(T + U_2, N_e)] \\
= \alpha F(U_1) + (1 - \alpha) F(U_2).
\]
Convex optimization (CVX-DMET)

- Concave conjugate: Legendre–Fenchel transform

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

- Find the correlation potential via convex (or concave) optimization (e.g., CVX). Robust global minima.

- Example code (mask enforces sparsity pattern)

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

Wu, Lindsey, Zhou, Tong, Lin, PRB 2020
Gapless and inexact matching

Theorem
Assuming $P \in S_{++}^{N_e} = \{A \in S \mid \text{Tr}[A] = N_e, 0 \prec A \prec 1\}$, 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: $D = D(F + U^*, N_e)$ achieves exact matching.
2. gapless and if $U^*$ is unique: exact matching is not achievable.

Remark:

- The condition $P \in S_{++}^{N_e}$ ensures that $P$ is in the relative interior of the domain of $F^*(= S_{++}^{N_e})$.

- 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

\[
\begin{align*}
\min_{D \in \mathcal{M}} \quad & \text{Tr}(TD), \\
\text{s.t.} \quad & D_x = P_x, \quad \forall x, \\
\mathcal{M} = \{D \in \mathbb{C}^{L \times L} \mid D = D^\dagger, \text{Tr}(D) = N_e, D^2 = D\}.
\end{align*}
\]

- 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)

Faulstich, Kim, Cui, Wen, Chan, Lin, JCTC 2022; See Fabian Faulstich’s talk in WS1.
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\(^1\)

- 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

\(^1\) [Lin, Lindsey, CPAM 2022][Khoo, Lindsey, arXiv:2106.02682]. See Michael Lindsey’s talk in WS1.
Acknowledgment

Thank you for your attention!

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