Riemannian Bath Construction in Density-Matrix Embedding Theory

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Presentation of the General Density Matrix Embedding Theory (DMET) [1],[2],[3]

Relies on obtaining the ground state 1-body reduced density matrix (1-rdm) of a system by working with **higher accuracy on several subsystems** in a **iterative** and **self-consistent procedure**
Considering a physical system, a Hilbert space $\mathcal{H}$, and its Hamiltonian, $\hat{H}$

- **The fragmentation:** decomposing the one-particle Hilbert space $\mathcal{H}$ into a direct sum of fragment spaces:

$$\mathcal{H} = \mathcal{H}_{\text{frag}}^{1,D} \oplus \cdots \oplus \mathcal{H}_{\text{frag}}^{N_{\text{f}}^D,D}, \tag{1}$$

  where N_{f}^D the number of fragments, and $\mathcal{H}_{\text{frag}}^{x,D}$ the one-particle state space associated with fragment x. For each fragment space $\ell^{x,D} := \dim(\mathcal{H}_{\text{frag}}^{x,D})$

- **The High-Level procedure:** for each fragment, building a **bath** subspace $\mathcal{H}_{\text{bath}}^{x,D}$ of finite dimension $\dim(\mathcal{H}_{\text{bath}}^{x,D}) = \ell_{\text{b}}^{x,D}$ such that the space defined by s-the direct sum of the fragment and the bath, the **cluster**

$$\mathcal{H} = \mathcal{H}_{\text{clust}}^{x,D} \oplus \mathcal{H}_{\text{env}}^{x,D}, \qquad \mathcal{H}_{\text{clust}}^{x,D} = \mathcal{H}_{\text{frag}}^{x,D} \oplus \mathcal{H}_{\text{bath}}^{x,D},$$

  where the *environment* space $\mathcal{H}_{\text{env}}^{x,D}$ is the orthogonal complement of the *impurity* space $\mathcal{H}_{\text{clust}}^{x,D}$, and get the ground state $1-$rdm of the local cluster systems

$$\widehat{\Gamma}_{\text{clust}}^{x,D,\mu} := \underset{\substack{\widehat{\Gamma}_{\text{clust}}^{x,D} \in \mathcal{S}\left(\text{Fock}\left(\mathcal{H}_{\text{clust}}^{x,D}\right)\right),\ \widehat{\Gamma}_{\text{clust}}^{x,D} \geq 0,\ \text{Tr}\left(\widehat{\Gamma}_{\text{clust}}^{x,D}\right)=1,\\ \text{Tr}\left(\widehat{N}_{\text{clust}}^{x,D}\widehat{\Gamma}_{\text{clust}}^{x,D}\right)=N-N_{\text{env}}^{x,D}}}{\text{argmin}} \text{Tr}\left(\left(\widehat{H}_{\text{clust}}^{x,D} - \mu\widehat{N}_{\text{frag}}^{x,D}\right)\widehat{\Gamma}_{\text{clust}}^{x,D}\right), \tag{2}$$

  and assemble the results $D_{HL} = \text{blockdiag}(D_{\text{HL}}^{xx,D})$

- **The Low-Level procedure:** maintaining the mean-field consistency with a low-level solver

$$D_{\text{LL}} = \underset{D \in \mathcal{D}_{\text{DMET}} \mid \text{Bd}^{D_{\text{HL}}}(D) = g(D_{\text{HL}})}{\text{argmin}} E(D), \tag{3}$$

  where $E : \mathcal{D}_{\text{DMET}} \to \mathbb{R}$ is a given objective function and $\text{Bd}^{D_{\text{HL}}}(D) = g(D_{\text{HL}})$ are linear constraints on the block-diagonals.

Perspectives in extending DMET [3]

In conventional DMET, the set of 1-RDMs:

$$\mathcal{D}_{\text{Slater}} := \{D \in \mathcal{S}(\mathcal{H}) \mid D^2 = D,\ \text{Tr}(D) = N\}$$

where $\mathcal{S}(\mathcal{H})$, space of symmetric operators in $\mathcal{H}$, $\text{Tr}(D)$, the trace of D.
In the variants of DMET, the 1-RDMs set

$$\mathcal{D}_{\text{MS}} := \{D \in \mathcal{S}(\mathcal{H}) \mid 0 \leq D \leq 1,\ \text{Tr}(D) = N\},$$

of all N-representable mixed-state 1-RDMs.

Extending the algorithm to general density matrices

- **Fragmentation procedure:** using a "cluster-friendly" basis set optimization

$$Q^{\bullet,D} = \min_{Q^\bullet \in \mathcal{M}} J(Q^\bullet), \quad J(Q^\bullet) := \sum_{x=1}^{N^{\text{f}}} \min_{\substack{P^x \in \text{Gr}(L_{\text{clust}}^x, \mathcal{H})\\ P^x(P_{\text{a}}^x + Q^x) = P_a^x + Q^x}} \left\| P^x D (P^x)^\perp \right\|^2. \tag{4}$$

- **High-Level procedure:** for each fragment f using optimization to build the bath of dimension ℓ_b in order to get a basis where the 1-rdm $D = \begin{pmatrix} D_{\text{ff}} & D_{\text{f}\bar{\text{f}}} \\ D_{\bar{\text{f}}\text{f}} & D_{\bar{\text{f}}\bar{\text{f}}} \end{pmatrix}$ is written

$$\begin{pmatrix} D_{\text{cc}} & D_{\text{ce}} \\ D_{\text{ec}} & D_{\text{ee}} \end{pmatrix},\ D_{\text{cc}} \in \mathbb{R}_{\text{sym}}^{\ell_{\text{clust}} \times \ell_{\text{clust}}},\ D_{\text{ee}} \in \mathbb{R}_{\text{sym}}^{(L-\ell_{\text{clust}}) \times (L-\ell_{\text{clust}})},\ \ell_{\text{clust}} = \ell + \ell_{\text{b}}, \quad M = L - \ell. \tag{5}$$

  with $\|D_{\text{ce}}\|^2$ as small as possible.

$$\min_{Q \in \text{Gr}(\ell_{\text{b}}, M)} J(Q),\ J(Q) := -\frac{1}{2}\text{Tr}(AQAQ) + \text{Tr}(BQ),\ A = D_{\bar{\text{f}}\bar{\text{f}}},\ B = \frac{1}{2}\left(D_{\bar{\text{f}}\bar{\text{f}}}^2 - D_{\bar{\text{f}}\text{f}}D_{\text{f}\bar{\text{f}}}\right) \tag{6}$$

$$\text{Gr}(\ell_{\text{b}}, M) := \left\{Q \in S(\mathbb{R}^M) \mid Q^2 = Q,\ \text{Tr}(Q) = \ell_{\text{b}}\right\}$$

- **Low-Level procedure:** considering more flexible energy functionals and methods to tackle the constraint on the blocs

$$D_{\text{out}} := \underset{D \in \mathcal{D}_{\text{MS}} \mid \text{Bd}(D) = g(D_{\text{in}})}{\text{argmin}} E(D), \tag{7}$$

Challenges in extended bath constructions

Mixed 1rdm and fractional occupation numbers

obtaining a stable cluster subspace of controllable dimension. The dimension has as an upper bound the number of distinct occupation numbers. [4]

Accuracy of solution through the overall DMET procedure

Considering and comparing the different optimization procedure solutions in the overall DMET procedure and the physical arguments their results. [5]

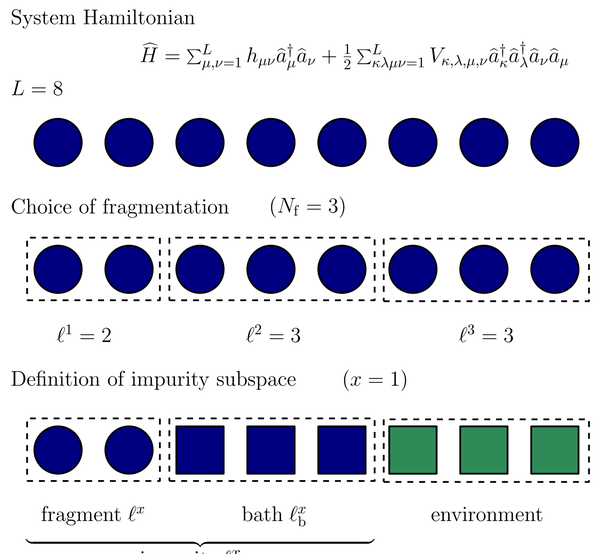
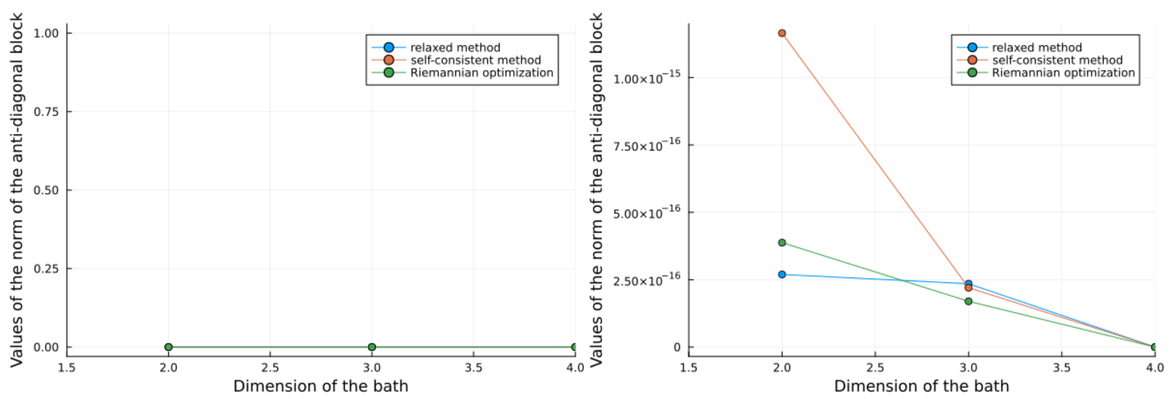


Figure 1. Definition of the various subspaces used in DMET. L the total number of orbitals, N the total number of electrons, N_{f} the number of fragments, ℓ^x the size of the xth fragment and ℓ_{b}^x the size of its bath (in conventional DMET, $\ell_{\text{b}}^x = \ell^x$). The different shapes used for the orbitals specify the bases in which they are represented.[3]

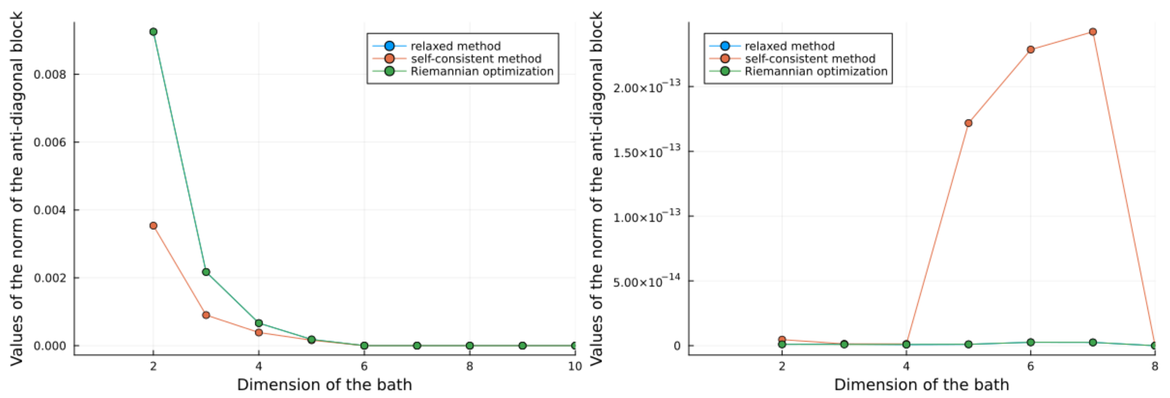
Numerical results for bath construction optimization methods



(a) Bath construction applied on fragment F1 of dimension 2.

(b) Bath construction applied on Fragment F2 of dimension 2.

Figure 2. Graphs of the norm of the anti-diagonal blocks depending on the dimension (size) of the bath. The system considered is the fluorine hydrate molecule HF 1-rdm obtained after a CCSD calculation (PySCF), with in the STO-3G basis set. Fragmentation is done spatially. The initialization here focused on the matrix A in equation (6).



(a) Bath construction applied on Fragment F1 of dimension 2.

(b) Bath construction applied on Fragment F3 of dimension 4.

Figure 3. Graphs of the norm of the anti-diagonal blocks depending on the dimension (size) of the bath. The system considered is the ethene molecule C_2H_2 1-rdm obtained after a CCSD calculation (PySCF), with in the STO-3G basis set. Fragmentation is done spatially. The initialization here focused on the matrix A in equation (6).

On-going works and open questions

- Characterization on how the considered system impacts
  - performances of the initialization procedure and optimization method
  - possibilities to considerably reduced the bath dimension
- Cluster resolution considerations
  - impacts on local cluster problems resolution
  - level of precision required to run the overall DMET algorithm
- Considering the fragmentation procedure based on "disentanglement"

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