A quadratic Grassmann manifold optimization problem arising from quantum embedding methods

March 19th 2026, GAMM 2026, University of Stuttgart

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Density Matrix Embedding theory (DMET): A brief presentation

Iterative method for computing approximations of some entries of the ground-state one-particle reduced density matrix (1-RDM) γ of a system .

Fragmentation: Partition the sytem into N_f non-overlapping fragments $\{x\}$ of size ℓ_x on the basis of physical arguments

$$\mathcal{H} = \mathcal{H}_{\mathrm{frag},1} \oplus \cdots \oplus \mathcal{H}_{\mathrm{frag},N_\mathrm{f}}, \quad \dim(\mathcal{H}_{\mathrm{frag},x}) = \ell_x, \quad \sum_{x=1}^{N_\mathrm{f}} \ell_x = L.$$

High-level iteration: From γ, for each fragment x, construct impurity subspace $\mathcal{H}^{\gamma}_{\mathrm{imp},x}$, with $1 \leq \dim(\mathcal{H}^{\gamma}_{\mathrm{imp,x}}) = (\ell_\mathrm{x} + \mathrm{m_x}) \ll \mathrm{L}$ s.t

$$\mathcal{H}_{\mathrm{frag},x} \subset \mathcal{H}^{\gamma}_{\mathrm{imp},x} \subset \mathcal{H}$$

$$\mathcal{H}^{\gamma}_{\mathrm{imp},x} = \mathcal{H}_{\mathrm{frag},x} \oplus \mathcal{H}^{\gamma}_{\mathrm{bath},x}, \quad \dim(\mathcal{H}^{\gamma}_{\mathrm{bath},x}) = m_x.$$

with the bath subspace $\mathcal{H}^{\gamma}_{\mathrm{bath},x}$:

$$\mathcal{H} = \mathcal{H}^{\gamma}_{\mathrm{imp}} \oplus {\mathcal{H}^{\gamma}_{\mathrm{imp}}}^{\perp}.$$

with full-disentanglement between $\mathcal{H}^{\gamma}_{\mathrm{imp}}$ and ${\mathcal{H}^{\gamma}_{\mathrm{imp}}}^{\perp} = \mathcal{H}^{\gamma}_{\mathrm{env}}$

The adventure starts when extending DMET...

Global description (1-RDM) γ

$\rightarrow$Conventional DMET:

$$\Gamma_{\mathrm{Slater}} := \{\gamma \in \mathcal{S}(\mathcal{H}) \mid \gamma^2 = \gamma,\ \mathrm{Tr}(\gamma) = N\},$$

(Slater-type
1-RDM of rank-N orthogonal projector)

$\rightarrow$Extended DMET:

$$\Gamma_{\mathrm{MS}} := \{\gamma \in \mathcal{S}(\mathcal{H}) \mid 0 \preceq \gamma \preceq 1,\ \mathrm{Tr}(\gamma) = N\}$$

(N-representable mixed-state 1-RDMs)

How to adapt bath construction?

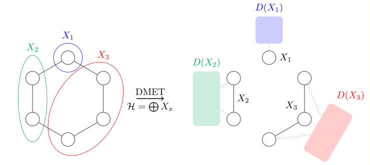


Figure: Impurity construction, (see Alfred Kirsch PhD manuscript).

1-RDM in the “physical basis”, L, ℓ, $M = L - \ell$

$$\gamma = \begin{pmatrix} \gamma_{\mathrm{frag}} & \gamma_{\mathrm{frag,ext}} \\ \gamma_{\mathrm{ext,frag}} & \gamma_{\mathrm{ext}} \end{pmatrix}, \quad \gamma_{\mathrm{frag}} \in \mathbb{R}^{\ell\times\ell}_{\mathrm{sym}}, \quad \gamma_{\mathrm{ext}} \in \mathbb{R}^{M\times M}_{\mathrm{sym}}, \quad \gamma_{\mathrm{frag,ext}} =\in \mathbb{R}^{M\times \ell}.$$

Bath Construction $\rightarrow$ Find a basis set and $1 \leq m \leq M - \ell \in \mathbb{N}$ s.t:

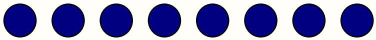
$$\gamma = \begin{pmatrix} \gamma_{\mathrm{imp}} & \gamma_{\mathrm{imp,env}} \\ \gamma_{\mathrm{env,imp}} & \gamma_{\mathrm{env}} \end{pmatrix}, \quad \gamma_{\mathrm{imp}} \in \mathbb{R}_{\mathrm{sym}}^{\ell_{\mathrm{imp}} \times \ell_{\mathrm{imp}}}, \quad \gamma_{\mathrm{env}} \in \mathbb{R}_{\mathrm{sym}}^{(L-\ell_{\mathrm{imp}}) \times (L-\ell_{\mathrm{imp}})},$$

$$\gamma_{\mathrm{imp,env}} \in \mathbb{R}^{(L-\ell_{\mathrm{imp}}) \times \ell_{\mathrm{imp}}}, \quad \gamma_{\mathrm{imp,env}} = \gamma_{\mathrm{env,imp}}^{T}, \quad \|\gamma_{\mathrm{imp,env}}\|^2 \text{ “small”.}$$

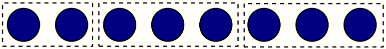
System Hamiltonian

$$\widehat{H} = \sum_{\mu,\nu=1}^{L} h_{\mu\nu} \hat{a}_\mu^\dagger \hat{a}_\nu + \frac{1}{2} \sum_{\kappa\lambda\mu\nu=1}^{L} V_{\kappa,\lambda,\mu,\nu} \hat{a}_\kappa^\dagger \hat{a}_\lambda^\dagger \hat{a}_\nu \hat{a}_\mu$$

$L = 8$

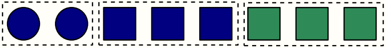


Choice of fragmentation $\quad (N_{\mathrm{f}} = 3)$



$\ell^1 = 2 \qquad \ell^2 = 3 \qquad \ell^3 = 3$

Definition of impurity subspace $\quad (x = 1)$



fragment ℓ^x $\quad$ bath ℓ_{b}^x $\quad$ environment

impurity ℓ_{imp}^x

Full disentanglement criteria

$$\Pi_{\mathrm{clus}}^{\gamma} \in \underset{\substack{\Pi \in \mathrm{Gr}(\ell+m, \mathbb{R}^L) \\ \Pi\,\Pi_{\mathrm{frag}} = \Pi_{\mathrm{frag}}}}{\operatorname{argmin}} \mathcal{J}^{\gamma}(\Pi),$$

$$\mathcal{J}^{\gamma}(\Pi) := \|\Pi \gamma \Pi^{\perp}\|^2, \quad \Pi^{\perp} = I_L - \Pi.$$

$$\Pi = \begin{pmatrix} I_\ell & 0 \\ 0 & P \end{pmatrix}, \quad P \in \mathrm{Gr}(m, \mathbb{R}^M).$$

From DMET... to a general optimization problem

In the DMET framework, the cost function reduces to

$$\mathcal{J}^{\gamma}(\Pi) = 2(\mathrm{Tr}(BP) - \frac{1}{2}\mathrm{Tr}(APAP)) + \|\gamma_{\mathrm{ext,frag}}\|^2,$$

$$A := \gamma_{\mathrm{ext}}, \quad B := \frac{1}{2}\left(\gamma_{\mathrm{ext}}^2 - \gamma_{\mathrm{ext,frag}}\gamma_{\mathrm{frag,ext}}\right)$$

The considered Problem

$A, B \in \mathbb{R}^{M\times M}_{\mathrm{sym}}, \quad A \succeq 0,$

$$\min_{P\in\mathcal{M}} J(P), \tag{1}$$

with

$$\mathcal{M} := \mathrm{Gr}(m, \mathbb{R}^M) := \left\{P \in \mathbb{R}^{M\times M}_{\mathrm{sym}} \mid P^2 = P,\ \mathrm{Tr}(P) = m\right\},$$

and

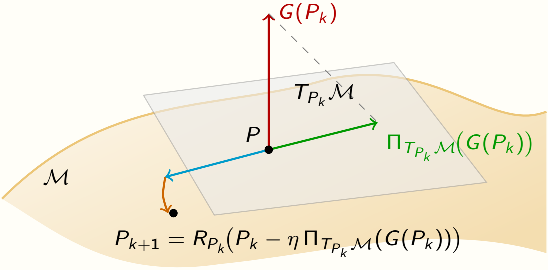
$$J := \begin{cases} \mathbb{R}^{M\times M}_{\mathrm{sym}} \to \mathbb{R} \\ P \mapsto \mathrm{Tr}(BP) - \frac{1}{2}\mathrm{Tr}(APAP) = \mathrm{Tr}(BP) - \frac{1}{2}\|A^{\frac{1}{2}}PA^{\frac{1}{2}}\|^2 \end{cases} \tag{2}$$

One manifold problem, but three strategies

- A direct minimization using Riemannian optimization theory and tools
- A strategy inherited from chemistry : Roothan method
- What about convexity?

Toolbox for the Riemannian optmization

The tangent space $\rightarrow \mathcal{T}_P\mathcal{M} = \{X \in \mathbb{R}^{M\times M}_{\mathrm{sym}} \,:\, XP + PX = X\}$
The projection on $\boldsymbol{\mathcal{T}_P\mathcal{M}}$ $\rightarrow \Pi_{\mathcal{T}_P\mathcal{M}}(X) = [[X,P],P]$, for all $P \in \mathcal{M},\ X \in \mathbb{R}^{M\times M}_{\mathrm{sym}}$



$$G(P) := \nabla J(P) = B - APA,$$
$$\mathrm{grad}_{\mathcal{M}} J(P) := \Pi_{\mathcal{T}_P\mathcal{M}}(\nabla J(P))$$

Optimality conditions

- First order: $P_\star$ critical point iff $[G(P_\star), P_\star] = 0$,
- Second order: $P_\star$ critical point, then $P_\star$ local minimzer of J if
  $\forall X = U\begin{pmatrix} 0 & X_{\mathrm{ov}} \\ X_{\mathrm{vo}} & 0 \end{pmatrix} U^T \in \mathcal{T}_{P_\star}\mathcal{M}, \quad \langle X, \mathrm{Hess}_{\mathcal{M}} J(P_\star)[X]\rangle \geq 0,$

Self-Consistent strategy

Theorem - Aufbau Principle

Let $A \in \mathbb{R}^{M\times M}_{\mathrm{sym}}$ be a $A \succeq 0$, $B \in \mathbb{R}^{M\times M}_{\mathrm{sym}}$, and $P_\star$ a global minimizer of (1) for J.

Then, there exists an orthogonal basis $(x_k)_{1\leq k\leq M}$ of $\mathbb{R}^M$ diagonalizing both $P_\star$ and $G(P_\star)$ such that

$$P_\star = \sum_{k=1}^{m} x_k x_k^T, \quad G(P_\star) = \sum_{k=1}^{M} \lambda_k x_k x_k^T, \quad x_k^T x_l = \delta_{kl}, \quad \lambda_1 \leq \lambda_2 \leq \cdots \leq \lambda_M.$$

In addition, if A is invertible, then

$$\lambda_m < \lambda_{m+1} \quad \text{and} \quad P_\star = \mathbb{1}_{(-\infty,\mu]}(G(P_\star)) \quad \text{for any } \mu \in [\lambda_m, \lambda_{m+1}).$$

Nice properties for Roothan

1. Denoting by $P_k := P_k^{\mathrm{Rth}}$, $(J(P_k))_{k\in\mathbb{N}}$ is non-increasing and converges to a critical value of J on $\mathcal{M}$. Any accumulation point $P_\star$ of the sequence $(P_k)_{k\in\mathbb{N}}$ is a critical point of J on $\mathcal{M}$ satisfying the Aufbau principle
2. Under the additional assumption that the matrix A is invertible, the whole sequence $(P_k)_{k\in\mathbb{N}}$ converges to a a critical point of J on $\mathcal{M}$ satisfying the Aufbau principle.

Convexification

Lemma - Convex Cost Function

Let $\widetilde{J} : \mathbb{R}^{M\times M}_{\mathrm{sym}} \to \mathbb{R}$ *be the quadratic function defined by*

$$\widetilde{J}(D) := \mathrm{Tr}(CD) + \frac{1}{4}\|[A, D]\|^2 \quad \textit{with} \quad C := B - \frac{1}{2}A^2.$$

Then, for all $P \in \mathcal{M}$, $\widetilde{J}(P) = J(P)$.

$\longrightarrow$ J *and* $\widetilde{J}$ *have the same critical points on* $\mathcal{M}$.

But now... $\widetilde{J}$ is **CONVEX**!

$\longrightarrow$ Consider the relaxed problem of minimizing $\widetilde{J}$ over the convex hull of $\mathcal{M}$

$$\mathcal{K} := \mathrm{CH}(\mathcal{M}) = \{D \in \mathbb{R}^{M\times M}_{\mathrm{sym}} \mid 0 \preceq D \preceq I_M,\ \mathrm{Tr}(D) = m\}.$$

Theorem - Convexified problem

Let $A \in \mathbb{R}^{M\times M}_{\mathrm{sym}}$, $A \succeq 0$, and $C \in \mathbb{R}^{M\times M}_{\mathrm{sym}}$. The set $\mathcal{D}_\star \subset \mathcal{K}$ of (global) minimizers of the convex optimization problem

$$\widetilde{I} = \min_{D\in\mathcal{K}} \widetilde{J}(D) \tag{3}$$

is non-empty, compact, convex subset.

1. $\mathbb{R}^{M\times M}_{\mathrm{sym}} \ni D \mapsto \nabla \widetilde{J}(D) = C - \frac{1}{2}[[A, D], A] \in \mathbb{R}^{M\times M}_{\mathrm{sym}}$ takes a constant value $H_\star$ on $\mathcal{D}_\star$.
2. Let $\mu_1 \le \cdots \le \mu_M$ be the eigenvalues of $H_\star$. Define

   $$\widetilde{\mathcal{D}}_\star := \left\{ D \in \mathcal{K} \mid \mathbb{1}_{(-\infty,\mu_m)}(H_\star) \preceq D \preceq 1_{(-\infty,\mu_m]}(H_\star) \right\}.$$

   Then $\mathcal{D}_\star \subset \widetilde{\mathcal{D}}_\star$, and

   $$\forall D_\star \in \mathcal{D}_\star, \quad \forall \widetilde{D}_\star \in \widetilde{\mathcal{D}}_\star, \quad \widetilde{D}_\star \in \mathcal{D}_\star \Leftrightarrow [A, D_\star - \widetilde{D}_\star] = 0 \Rightarrow \mathrm{Ran}(D_* - \widetilde{D}_*) \subset \mathcal{V},$$

   where $\mathcal{V} \subset \mathbb{R}^M$ the largest A-invariant vector subspace of $\mathrm{Ker}(H_\star - \mu_m)$
3. If $\mu_m < \mu_{m+1}$, then $P_\star := \mathbb{1}_{(-\infty,\mu_m]}(H_\star)$ is the unique minimizer of (3), **AND** the unique global minimizer of the original problem (1)–(2).

Special cases (1) - $[A, B] = 0$

Proposition - A, B commute

Let $A_0, B_0 \in \mathbb{R}^{M\times M}_{\mathrm{sym}}$ with $A_0 \succeq 0$ and $[A_0, B_0] = 0$, $C_0 := B_0 - \frac{1}{2}A_0^2$.

- *Let $c_1^0 \le c_2^0 \le \cdots \le c_M^0$ be the eigenvalues of C_0.*
  The minimizers of (1)–(2) are given by

  $$P_\star = 1_{(-\infty, c_m^0)}(C_0) + \Delta, \quad \mathrm{Tr}(P_\star) = m, \quad \Delta^2 = \Delta = \Delta^*,$$
  $$\mathrm{Ran}(\Delta) \subset \mathrm{Ker}\left(C_0 - c_m^0\right), \quad [A_0, \Delta] = 0.$$

  If $c_m^0 < c_{m+1}^0$, $P_0 = P_\star := 1_{(-\infty, c_m^0]}(C_0)$ is the unique minimizer of (1)–(2) and the matrices A_0, B_0, C_0, and $P_\star$ pairwise commute.

- *Using perturbation theory for $[A, B]$ "small".*
  If $c_m^0 < c_{m+1}^0$, there exists $\epsilon_0 > 0$ such that for all $\epsilon \in (-\epsilon_0, \epsilon_0)$

  $$I^\epsilon := \min_{P\in\mathcal{M}} J^\epsilon(P) \quad \text{with} \quad J^\epsilon(P) := \mathrm{Tr}(B^\epsilon P) - \frac{1}{2}\mathrm{Tr}(A^\epsilon P A^\epsilon P).$$

  has a unique global minimizer P^ϵ, and the function $(-\epsilon_0, \epsilon_0) \ni \epsilon \mapsto P^\epsilon \in \mathbb{R}^{M\times M}_{\mathrm{sym}}$ is real-analytic.

Special cases (2): toy Model 2 for $M = 3$ and $m = 1$

$$A = \begin{pmatrix} a_1 & 0 & 0 \\ 0 & a_2 & 0 \\ 0 & 0 & a_3 \end{pmatrix}, \quad C = \begin{pmatrix} \beta & -\frac{\alpha}{2}(a_2 - a_1)^2 & 0 \\ -\frac{\alpha}{2}(a_2 - a_1)^2 & \beta & -\frac{\alpha}{2}(a_3 - a_2)^2 \\ 0 & -\frac{\alpha}{2}(a_3 - a_2)^2 & \beta \end{pmatrix},$$

with $0 \leq a_1 < a_2 < a_3$, $\alpha > 0$, $\beta \in \mathbb{R}$. $D \in \mathbb{R}^{3\times 3}_{\mathrm{sym}}$ such that $\mathrm{Tr}(D) = 1$,

$$\begin{aligned} \widetilde{J}(D) = \beta &+ 2(a_2 - a_1)^2(D_{12} - \alpha)^2 + 2(a_3 - a_2)^2(D_{23} - \alpha)^2 + 2(a_3 - a_1)^2 D_{13}^2 \\ &- 2\alpha^2((a_2 - a_1)^2 + (a_3 - a_2)^2). \end{aligned}$$

Thus

$$\mathcal{D}_\star := \operatorname*{argmin}_{\substack{D \in \mathbb{R}^{3\times 3}_{\mathrm{sym}} \\ \mathrm{Tr}(D) = 1}} \widetilde{J}(D) = \left\{ D = \begin{pmatrix} n_1 & \alpha & 0 \\ \alpha & n_2 & \alpha \\ 0 & \alpha & n_3 \end{pmatrix}, \; n_1, n_2, n_3 \in \mathbb{R}, \; n_1 + n_2 + n_3 = 1 \right\}.$$

For any $D \in \mathcal{D}_\star$, $[D^2]_{13} = \alpha^2 > 0 = D_{13}$, thus $\mathcal{D}_\star \cap \mathcal{M} = \emptyset$, BUT for $\alpha > 0$ small enough

$$\begin{pmatrix} 1/3 & \alpha & 0 \\ \alpha & 1/3 & \alpha \\ 0 & \alpha & 1/3 \end{pmatrix} \in \mathcal{D}_\star \cap \mathcal{K}, \quad H_\star = \beta I_3$$

$\rightarrow$ For $\alpha > 0$ small enough, the set of minimizers of $\mathcal{J}$ on $\mathcal{K}$ does not contain any point of $\mathcal{M}$.

Special cases (2): toy Model 2 for $M = 3$ and $m = 1$

Taking now $a_1 = 1$, $a_2 = 2$, $a_3 = 3$, $\beta = 0$, $\alpha = 1/2$,

$$\mathcal{D}_\star \cap \mathcal{K} = \emptyset.$$

The matrix $H_\star$ and the set of minimizers of $\widetilde{J}$ on $\mathcal{K}$ are given by

$$H_\star = \begin{pmatrix} 0 & -1/9 & 1/9 \\ -1/9 & 0 & -1/9 \\ 1/9 & -1/9 & 0 \end{pmatrix} \quad \text{and} \quad \operatorname*{argmin}_{D \in \mathcal{K}} \widetilde{J}(D) = \left\{ \begin{pmatrix} 2/9 & 5/18 & 1/18 \\ 5/18 & 5/9 & 5/18 \\ 1/18 & 5/18 & 2/9 \end{pmatrix} \right\}.$$

The eigenvalues of $H_\star$ are $\mu_1 = \mu_2 = -1/9$, $\mu_3 = 2/9$.

$\rightarrow$ In agreement with the theoretical results, there is no gap between the first and second eigenvalues of $H_\star$

Methods box

Optimization on $\mathcal{M}$ for J: Methods for Riemannian optimization (**Manopt.jl, Julia**)

$$P_{k+1} = R_{P_k}\big(P_k - \eta\,\Pi_{T_{P_k}\mathcal{M}}(G(P_k))\big)$$

Roothan on $\mathcal{M}$ for J: The algorithms reads

$$P_{k+1}^{\mathrm{Rth}} \in \operatorname*{argmin}_{P\in\mathcal{M}} \mathrm{Tr}(G(P_k^{\mathrm{Rth}})P) \quad \text{with } G(P) = B - APA.$$

ODA on $\mathcal{M}$ for J: Relaxed self consistent method fitting using J

$$P_{k+1}^{\mathrm{ODA}} \in \operatorname*{argmin}_{P\in\mathcal{M}} \mathrm{Tr}(G(D_k)P), \quad D_{k+1} = \operatorname*{argmin}_{D\in[D_k, P_{k+1}^{\mathrm{ODA}}]} J(D)$$

ODA on $\mathcal{K}$ for $\tilde{J}$: Relaxed self consistent method fitting using J

$$P_{k+1}^{\mathrm{conv}} \in \operatorname*{argmin}_{P\in\mathcal{M}} \mathrm{Tr}(H(D_k^{\mathrm{conv}})P), \qquad D_{k+1}^{\mathrm{conv}} = \operatorname*{argmin}_{D\in[D_k^{\mathrm{conv}}, P_{k+1}^{\mathrm{conv}}]} \widetilde{J}(D)$$

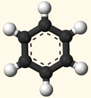
(!): ODA and Roothan for J coincide on $\mathcal{M}$.

Numerical results - Benzene

Methodology for numerical tests on physical systems

Considering a good fragmentation is provided with the system

Step 1: γ_{GS} the spin-unpolarized ground-state 1-RDM from CCSD calculation in a basis set of atomic orbitals using **PySCF (Python)** + Löwdin orthonormalization $\longrightarrow$ Export data



Step 2: The rest of the calculation:

- computation from the chosen fragmentation γ_{GS} of the A, B, C matrices of the bath construction problem for each fragment
- numerical solutions of (1)–(2) and (3) by the different algorithms

is performed by a homemade code in **Julia**.

Parameters for the Benzene (with imposed symmetry)

- basis set: STO-3G minimal orbital basis set
- $L = 36$ orbitals, $N = 21$
- Each fragment contains $\ell = 6$ orbitals and are identical, $M = 30$
- $\|\gamma_{\mathrm{GS}}^2 - \gamma_{\mathrm{GS}}\| \simeq 0.067$, γ_{GS} (still weakly-correlated system)

Numerical results - Benzene, Convex approach vs Manifold approach

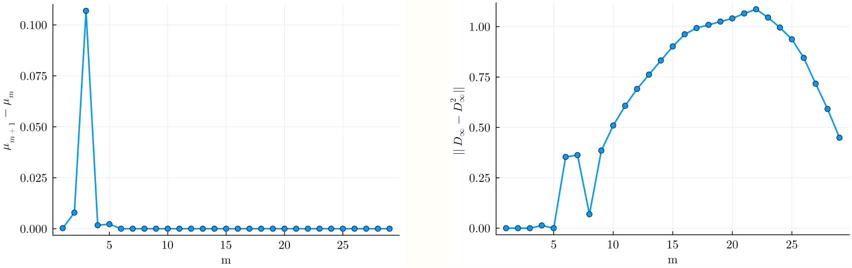


Figure: Gap $\mu_{m+1} - \mu_m$ and non-idempotency criterion $\|D_\star - D_\star^2\|$ as functions of the bath dimension m for the benzene molecule.

$\rightarrow$ **Shows the value of m where there exists a unique global minimizer for $\tilde{J}$**

Numerical results - Benzene, $m = 3$

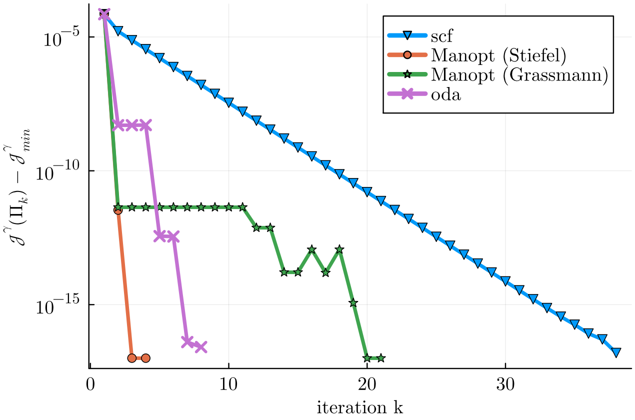
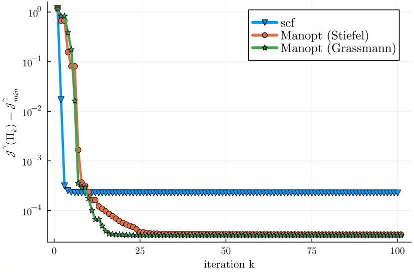
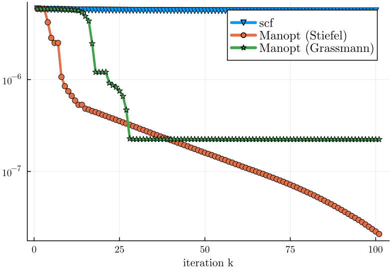


Figure: Decay of the cost function $\mathcal{J}^\gamma$ for benzene, a bath dimension $m = 3$, various numerical algorithms, and the inital guess $P_\mathbf{0} = \mathbb{1}_{(-\infty, c_m)}(C)$. For $m = 3$, the assumed global minimum is $\mathcal{J}^\gamma_{min} \simeq 0.0094538$.

Numerical results - Benzene, $m = 15$



(a) Initial guess $P_0 = 1_{(-\infty, c_m]}(C)$.



(b) Initial guess $P_0 = P_\infty^{\mathrm{ODA}}$.

Figure: Decay of the cost function $\mathcal{J}^\gamma$ for benzene, a bath dimension $m = 15$, various numerical algorithms, and two different initializations. For $m = 15$, the assumed global minimum is $\mathcal{J}^\gamma_{min} \simeq 2.0815393 \times 10^{-6}$.

Conclusion and Perspectives

Conclusions

- We derive interesting properties for an optimization problem on the Grassmann manifold for a quadratic cost function
- We propose an extended framework to study such cost functions by considering different algorithms and using a "convexification" for the problem

Perspectives

- More numerical examples and test to benchmark the methods and understand specificity of the algorithms depending of the bath dimensions
- Tests on other systems
- Incorporate this method in the overall extended DMET
- Provide an accessible code (Work in Progress!)

Thanks to my collaborators, and Laura Grazioli, Raehyun Kim and Théo Duez for their precious help.

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Thank you for your attention! :)