Metric-based nonlinear model order reduction with applications to quantum chemistry

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joint work with Maxime Dalery, Virginie Ehrlacher,
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Outline

Introduction

From linear to metric reduced-order modelling

Optimal transport based ROM for the wavefunction

Marginal-constrained ROM for the pair density

Interpolations between density matrices

Conclusion and perspectives

Context : Parametrized Partial Differential Equations

Consider a PDE that depends on parameters $\mathbf{R} \in \mathcal{R}$:

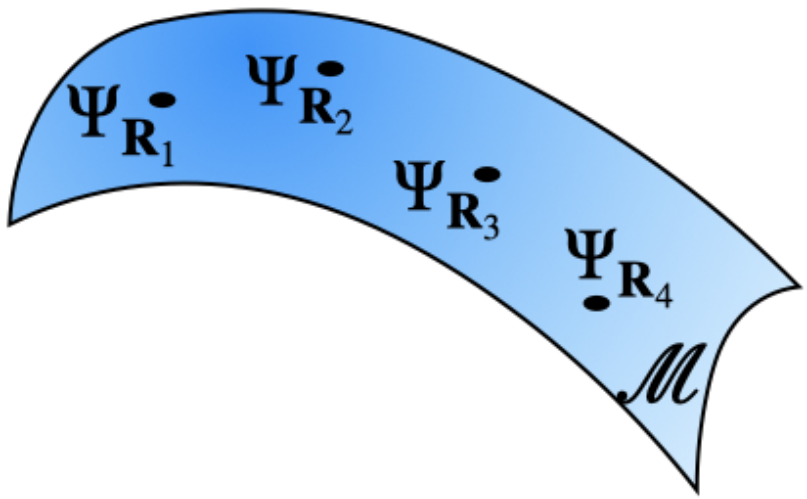
$$\boxed{\text{Solve } F_{\mathbf{R}}(\Psi) = 0}$$

Many query : solve the equation for **many** values of the parameters.

Aim : Efficiently approximate solution the **whole** manifold of solutions

$$\mathcal{M} := \{\Psi_{\mathbf{R}} \text{ for } \mathbf{R} \in \mathcal{R}\}, \quad \mathcal{R} \text{ set of parameters}$$

from the computation of only **a few** solutions.



Main tool : Reduced-order modelling

Motivation : Modelling at the atomic scale

Molecular system described by

- M (classical) nuclei ● with positions $\mathbf{R} \in \mathbb{R}^{3M}$
- N electrons described by a wave-function $\Psi_{\mathbf{R}} : \mathbb{R}^{3N} \to \mathbb{C}$

Wavefunction $\Psi_{\mathbf{R}}(r_1, r_2, \ldots, r_N)$:

- $|\Psi_{\mathbf{R}}|^2$ probability density
- **Pauli principle** : $\Psi_{\mathbf{R}}(\ldots, r_i, \ldots, r_j, \ldots) = -\Psi_{\mathbf{R}}(\ldots, r_j, \ldots, r_i, \ldots)$

Physical properties can be computed from the wavefunction : Energy, forces, polarizability, etc.

The Schrödinger equation (1926)

Ground state problem : looking for the wavefunction minimizing the energy

Energy minimization

$$\inf_{\substack{\Psi \in H^1_{as}(\mathbb{R}^{3N}) \\ \|\Psi\|_{L^2}=1}} \langle \Psi, H_{\mathbf{R}} \Psi \rangle,$$

where $H_{\mathbf{R}}$ is the Hamiltonian of the problem, parametrized by the positions of the nuclei, typically

$$H_{\mathbf{R}} = -\frac{1}{2} \sum_{i=1}^{N} \Delta_{r_i} + \sum_{i=1}^{N} V^{ne}_{\mathbf{R}}(r_i) + \sum_{1 \le i < j \le N} \frac{1}{|r_i - r_j|}, \quad V^{ne}_{\mathbf{R}}(r_i) = -\sum_{m=1}^{M} \frac{z_m}{|\mathbf{R}_m - r_i|}$$

Eigenvalue problem

$$H_{\mathbf{R}} \Psi_{\mathbf{R}} = E_{\mathbf{R}} \Psi_{\mathbf{R}}.$$

An **eigenvalue problem** parametrized by the nuclei positions.

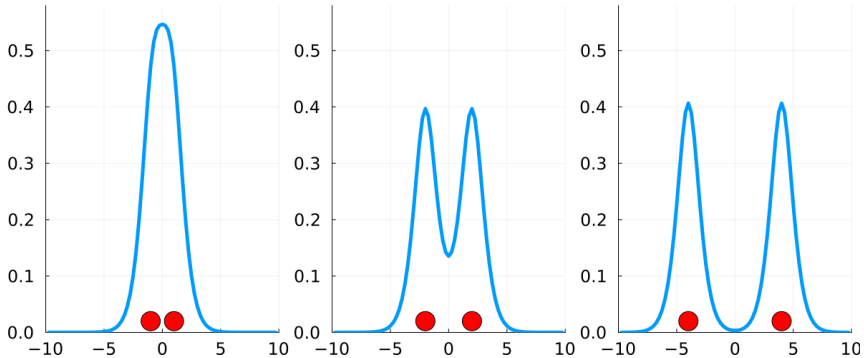
In practice, in general too expensive :
Hartree–Fock, post Hartree–Fock
Density Functional Theory

In this presentation : mostly toy problems but keep in mind that we want to deal with high-dimensional problems.

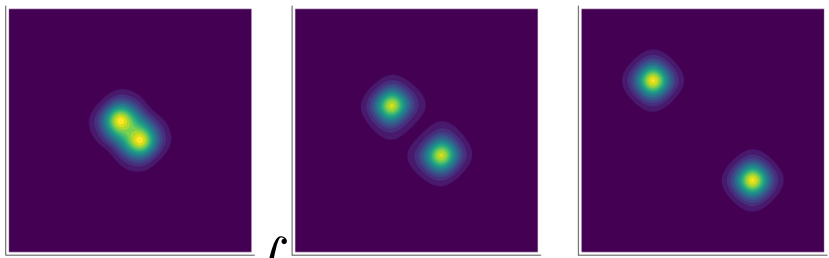
Quantities of interest

Wavefunction $\Psi_{\mathbf{R}}(x_1, x_2, \ldots, x_N)$

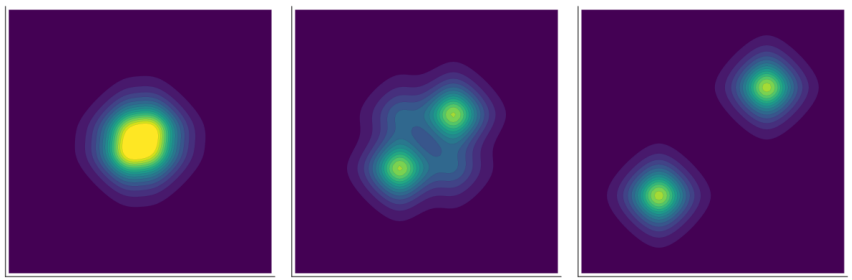
Electronic density $\rho_{\mathbf{R}}(x) = \int_{\mathbb{R}^{d(N-1)}} |\Psi_{\mathbf{R}}(x, x_2, \ldots, x_N)|^2$



Two-body density $\tau_{\mathbf{R}}(x, y) = \int_{\mathbb{R}^{d(N-2)}} |\Psi_{\mathbf{R}}(x, y, x_3, \ldots, x_N)|^2$



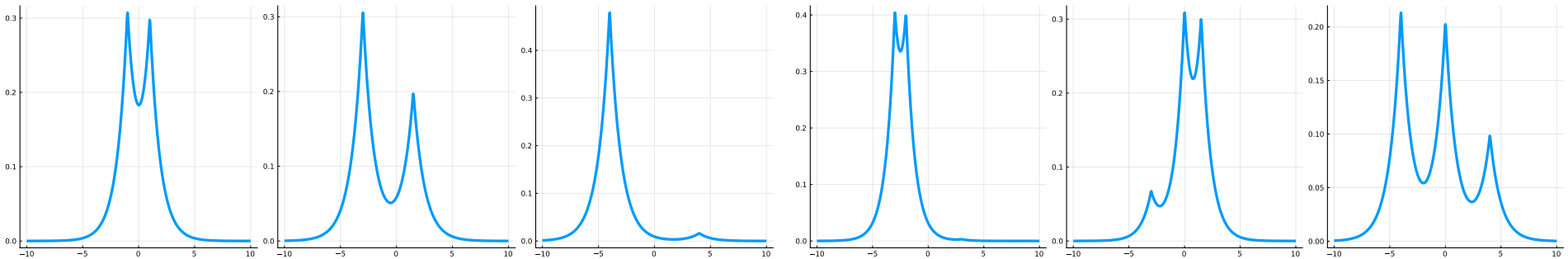
Density matrix $\gamma_{\mathbf{R}}(x, y) = \int_{\mathbb{R}^{d(N-1)}} \Psi_{\mathbf{R}}(x, x_2, \ldots, x_N)^* \Psi_{\mathbf{R}}(y, x_2, \ldots, x_N)$



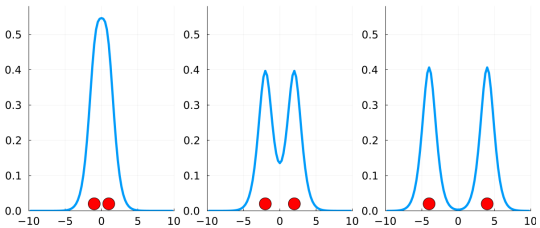
Outline : Three reduced-order models

Electronic density, two-body density, and one-body density matrix enough to **exactly** reconstruct the energy exactly
Lower-dimensional objects

Example 1 : Wavefunction (one electron)



Example 2 : Two-body density



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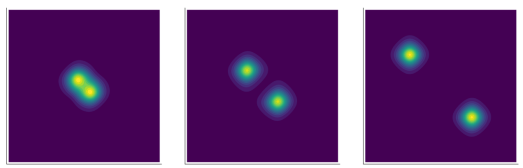
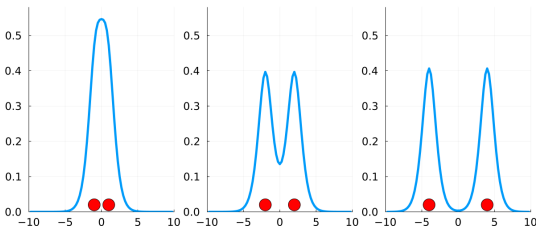


Example 3 : Density matrix



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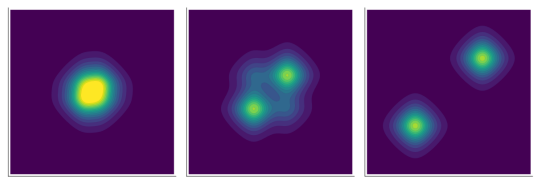

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Projection-based reduced-order model [1] [2]

Problem : Parametrized PDE with parameters $\mathbf{R} \in \mathcal{R}$'s.

Needs to be solved for **many** parameters $\mathbf{R}$.

- **Offline phase :**
  1. **Database :** Choose values $\mathbf{R} \in \mathcal{R}_{\text{train}}$ and compute

$$\Psi_{\boldsymbol{R}}, \quad \mathbf{R} \in \mathcal{R}_{\text{train}} \qquad \text{(snapshots)}$$

  2. **Greedy algorithm :** Select $\mathbf{R}_1, \mathbf{R}_2, \ldots, \mathbf{R}_n \in \mathcal{R}_{\text{train}}$ with $n \in \mathbb{N}$ (small) so that,

$$\forall\, \mathbf{R} \in \mathcal{R}_{\text{train}}, \quad \Psi_{\mathbf{R}} \approx \sum_{k=1}^{n} \lambda_k \Psi_{\mathbf{R}_k} \text{ for some } \boldsymbol{\lambda} \in \mathbb{R}^n$$

- **Online phase :** For a new value $\mathbf{R} \in \mathcal{R}$,
  1. Approximate $\Psi_{\boldsymbol{R}}$ by

$$\Psi_{\boldsymbol{R}} \approx \sum_{k=1}^{n} \lambda_{\mathbf{R},k} \Psi_{\mathbf{R}_k} \text{ for some } \boldsymbol{\lambda}_{\boldsymbol{R}} \in \mathbb{R}^n \quad \text{(Galerkin approximation)}$$

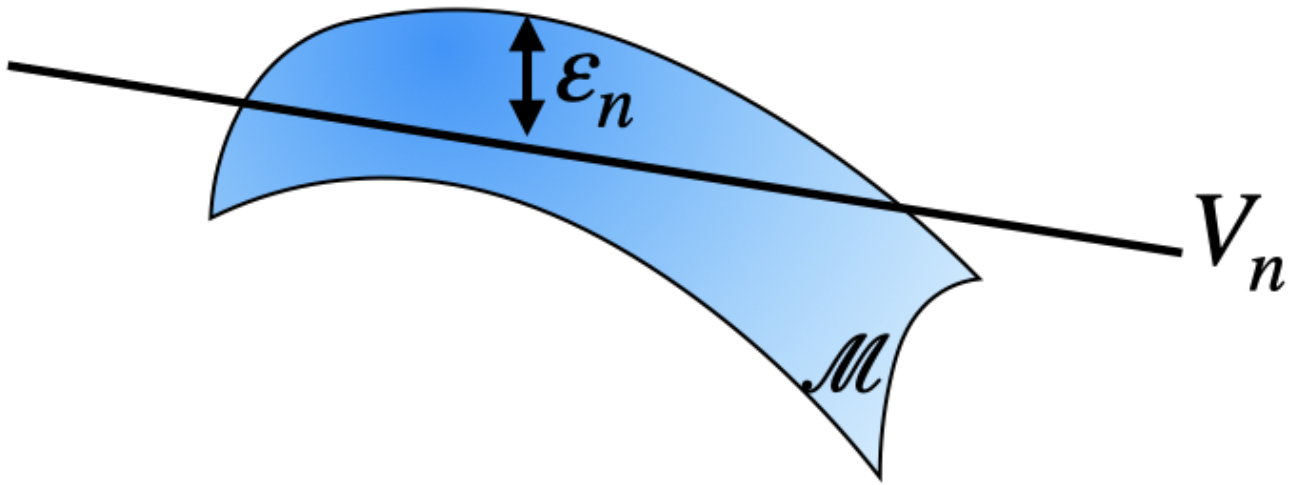
Successful for many industrial applications

1. Barrault, Maday, Nguyen, Patera. An empirical interpolation method, CR math, 2004
2. Hesthaven, Rozza, Stamm. Certified reduced basis methods for parametrized PDEs, 2016

Characterize the success : the Kolmogorov n-width

Definition for a Hilbert space $\mathbb{H}$: $\mathcal{M} := \{\Psi_{\mathbf{R}}, \quad \mathbf{R} \in \mathcal{R}\}$

$$\varepsilon_n(\mathcal{M}, \mathbb{H}) := \inf_{\substack{V_n \subset \mathbb{H} \\ \dim V_n = n}} \sup_{\mathbf{R} \in \mathcal{R}} \|\Psi_{\mathbf{R}} - \mathrm{P}_{V_n} \Psi_{\mathbf{R}}\|.$$



3. Cohen, DeVore. Approximation of high-dimensional parametric PDEs Acta Numerica, 2015

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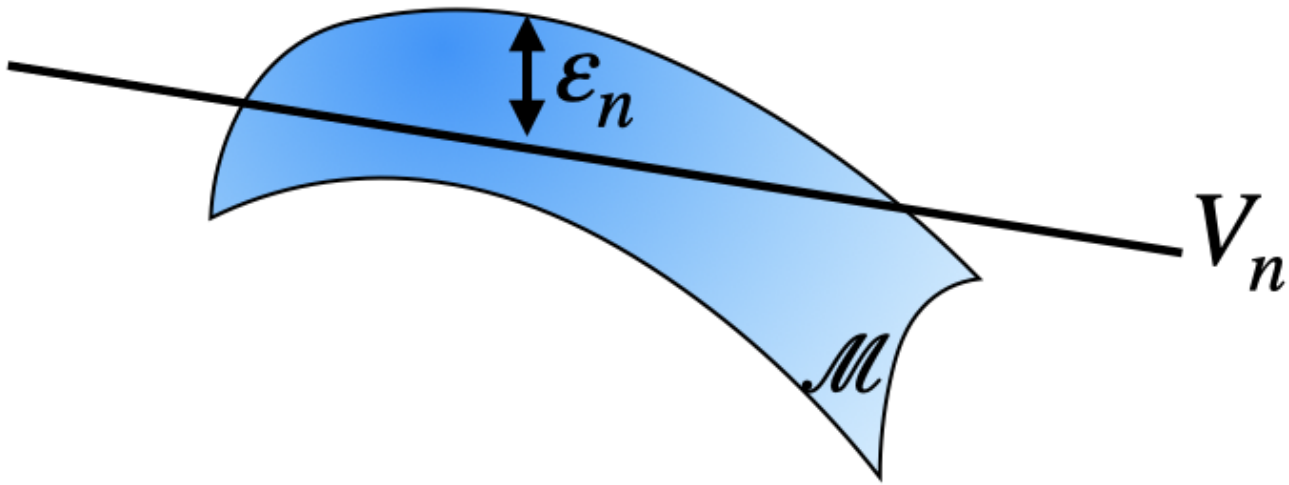


Good scenario : elliptic equation $A_{\mathbf{R}} \Psi_{\mathbf{R}} = f$, with affine representation of $A_{\mathbf{R}}$ [3] :

$$A_{\mathbf{R}} = \sum_{q=1}^{Q} \theta_q(\mathbf{R}) A_q, \quad \text{for some } \theta_q \in \mathbb{R},\ A_q \text{ continuous operators}$$

Exponential decay of the Kolmogorov n-width : $\varepsilon_n(\mathcal{M}, \mathbb{H}) \leq C \ \exp(-cn^{1/Q})$.

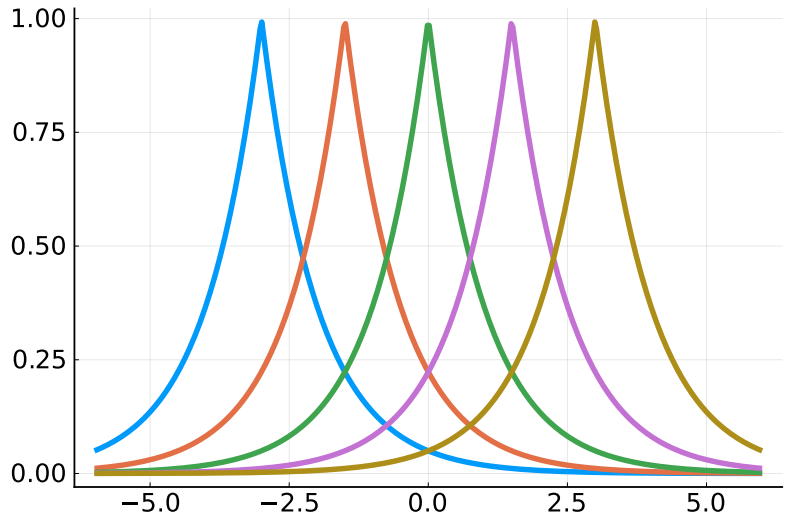
3. Cohen, DeVore. Approximation of high-dimensional parametric PDEs Acta Numerica, 2015

Characterize the success : the Kolmogorov n-width

Bad scenario : transport equations [4] and electronic structure [5]

Toy problem : One dimension, one electron

$$\min_{\substack{\Psi \in H^1(\mathbb{R}) \\ \|\Psi\|_{L^2(\mathbb{R})}=1}} \frac{1}{2}\int_{\mathbb{R}} |\Psi'|^2 - \sum_{m=1}^{M} z_m \Psi(R_m)^2$$



Theorem [Dalery, GD, Ehrlacher, Lozinski, 2023]

For a 1D toy problem with one nucleus or two nuclei,

$$\varepsilon_n(\mathcal{M}, L^2(\mathbb{R})) \geq c n^{-\frac{3}{2}}.$$

4. Ohlberger, Rave. Reduced Basis Methods : Success, Limitations and Future Challenges, 2015
5. Dalery, GD, Ehrlacher, Lozinski. Constructive Approximation 2026

Nonlinear reduced order modelling

$$\Psi_{\boldsymbol{R}} \approx F(\Psi_{\mathbf{R}_1}, \ldots, \Psi_{\mathbf{R}_n})$$

Different possible nonlinearities

- Library-based [6]
- Polynomials [7]
- Neural networks [8]
- Optimal transport [9]

6. e.g. Guignard, Mula. Tree-based nonlinear reduced modeling. 2024.
7. e.g. Bensalah, Nouy, Soffo arXiv :2502.05088.
8. e.g. Barnett, Farhat, Maday. 2023
9. e.g. Ehrlacher, Lombardi et al, 2020. Iollo, Taddei, 2022. Do, Feydy, Mula, 2023.

Nonlinear reduced order modelling

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Replacing linear combinations by barycenters : Metric space $\mathbb{M}$

Given a distance d, convex parameters $\boldsymbol{\lambda} = (\lambda_1, \dots, \lambda_n)$, and elements $\Psi_{\mathbf{R}_1}, \dots, \Psi_{\mathbf{R}_n} \in \mathbb{M}$,

$$\mathrm{Bar}^{\boldsymbol{\lambda}}(\Psi_{\mathbf{R}_1}, \dots, \Psi_{\mathbf{R}_n}) := \mathrm{argmin}_{u \in \mathbb{M}} \quad \sum_{k=1}^{n} \lambda_k d(u, \Psi_{\mathbf{R}_k})^2.$$

Nonlinear Kolmogorov n-width

$$\varepsilon_n(\mathcal{M}, \mathbb{M}) := \inf_{\Psi_{\mathbf{R}_1}, \dots, \Psi_{\mathbf{R}_n} \in \mathbb{M}} \sup_{\mathbf{R} \in \mathcal{R}} \inf_{\boldsymbol{\lambda}} d\left(\Psi_{\mathbf{R}}, \mathrm{Bar}^{\boldsymbol{\lambda}}(\Psi_{\mathbf{R}_1}, \dots, \Psi_{\mathbf{R}_n})\right)$$

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6. e.g. Guignard, Mula. Tree-based nonlinear reduced modeling. 2024.
7. e.g. Bensalah, Nouy, Soffo arXiv :2502.05088.
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Metric reduced-order model : practical strategy

- **Offline phase :**
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$$\Psi_{\boldsymbol{R}}, \quad \mathbf{R} \in \mathcal{R}_{\text{train}} \qquad \text{(snapshots)}$$

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- **Online phase for the wavefunction :** For a new value $\mathbf{R} \in \mathcal{R}$,

$$\Psi_{\boldsymbol{R}} \approx \text{Bar}^{\boldsymbol{\lambda}_{\boldsymbol{R}}}(\Psi_{\mathbf{R}_1}, \ldots, \Psi_{\mathbf{R}_n}) \text{ for some } \boldsymbol{\lambda}_{\boldsymbol{R}} \in \Lambda_n,$$

  possibly minimizing the energy of $\text{Bar}^{\boldsymbol{\lambda}_{\boldsymbol{R}}}(\Psi_{\mathbf{R}_1}, \ldots, \Psi_{\mathbf{R}_n})$

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How to choose a metric adapted to the problem ?

- Decay of the Kolmogorov n-width - linear/ nonlinear
- Physical constraints
- Computational cost of the barycenters

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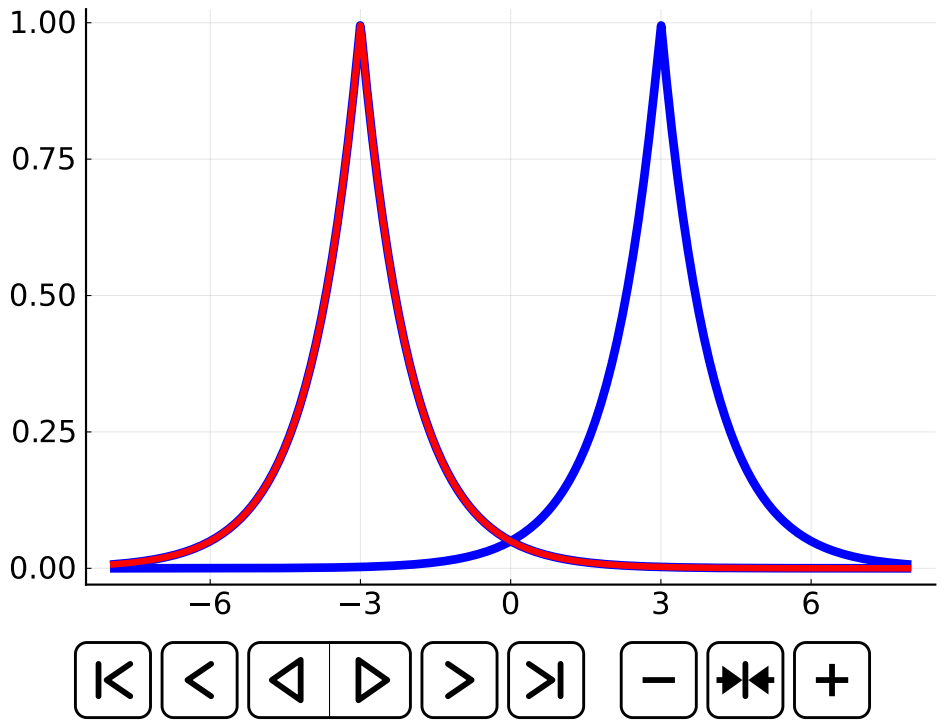
Motivation behind using optimal transport

Use of **optimal transport** : at minima deals with the translations
Barycenter between two Slater functions : a translated Slater function



Some references : [Iollo, Lombardi, 2014] [Ehrlacher, Lombardi, Mula, Vialard, 2020] [Iollo, Taddei, 2022] [Do, Feydy, Mula,2023] [Rim, Peherstorfer, Mandli, 2023]

Wasserstein distance and Wasserstein barycenters

Wasserstein distance :

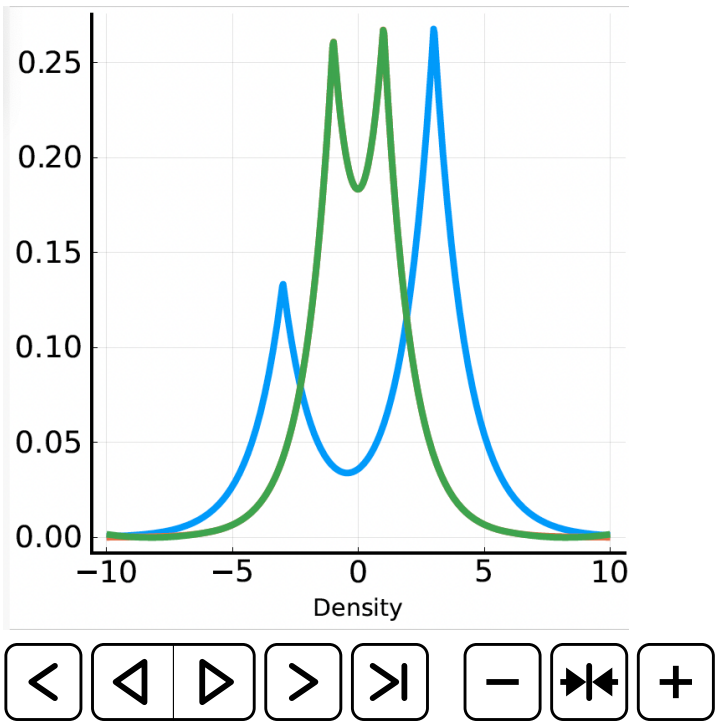
$$u, v \in \mathcal{P}_2(\Omega)^2, \quad W_2(u,v)^2 := \inf_{\pi \in \Pi(u,v)} \int_{\Omega^2} (x-y)^2 \, d\pi(x,y),$$

$\Pi(u,v)$: set of probability measures over Ω^2 with marginals u and v.

Wasserstein barycenter :

- n probability measures $\rho_1, \ldots, \rho_n$
- n positive weights $\lambda_1, \ldots, \lambda_n$ summing to 1

$$\inf_{u \in \mathcal{P}_2(\Omega)} \sum_{i=1}^{n} \lambda_i W_2(u, \rho_i)^2.$$



Agueh, Carlier : Barycenters in the Wasserstein Space. SIAM J. Math. Anal. (2011).

Gangbo, Swiech : Optimal maps for the multidimensional Monge–Kantorovich problem. Commun. Pure Appl. Math. (1998)

Result for the 1D toy problem

Theorem [Dalery, GD, Ehrlacher, Lozinski, 2023]
For the same 1D toy problem with one nucleus [a]

$$\forall n > 1, \quad \varepsilon_n\left(\mathcal{M}, (\mathcal{P}_2(\mathbb{R}), W_2)\right) = 0$$

For two nuclei

$$\varepsilon_n(\mathcal{M}, (\mathcal{P}_2(\mathbb{R}), W_2)) \leq c n^{-\frac{5}{2}}.$$

a. arxiv :2307.15423

Main limitations :

- Computational cost of Wasserstein distance and barycenters
- Does not preserve structure of the solutions
- Marginals not preserved
- Limited to probability measures

Optimal transport between Gaussian measures

Notation : $\mathcal{N}(\mu, S)$

Wasserstein distance

If $\rho_0 = \mathcal{N}(\mu_0, S_0)$ and $\rho_1 = \mathcal{N}(\mu_1, S_1)$, it holds that

$$W_2^2(\rho_0, \rho_1) = \|\mu_0 - \mu_1\|^2 + \mathrm{Tr}\left(S_0 + S_1 - 2\left(\sqrt{S_0} S_1 \sqrt{S_0}\right)^{1/2}\right)$$

Wasserstein barycenters

$M \in \mathbb{N}^\star$

$\boldsymbol{\lambda} = (\lambda_1, \dots, \lambda_M) \in \Lambda_M$

$\boldsymbol{\rho} = (\rho_1, \dots, \rho_M) \in \mathcal{P}_2(\mathbb{R}^n)^M$, with for all $i \in \{1, \dots, M\}$, $\rho_i = \mathcal{N}(\mu_i, S_i)$

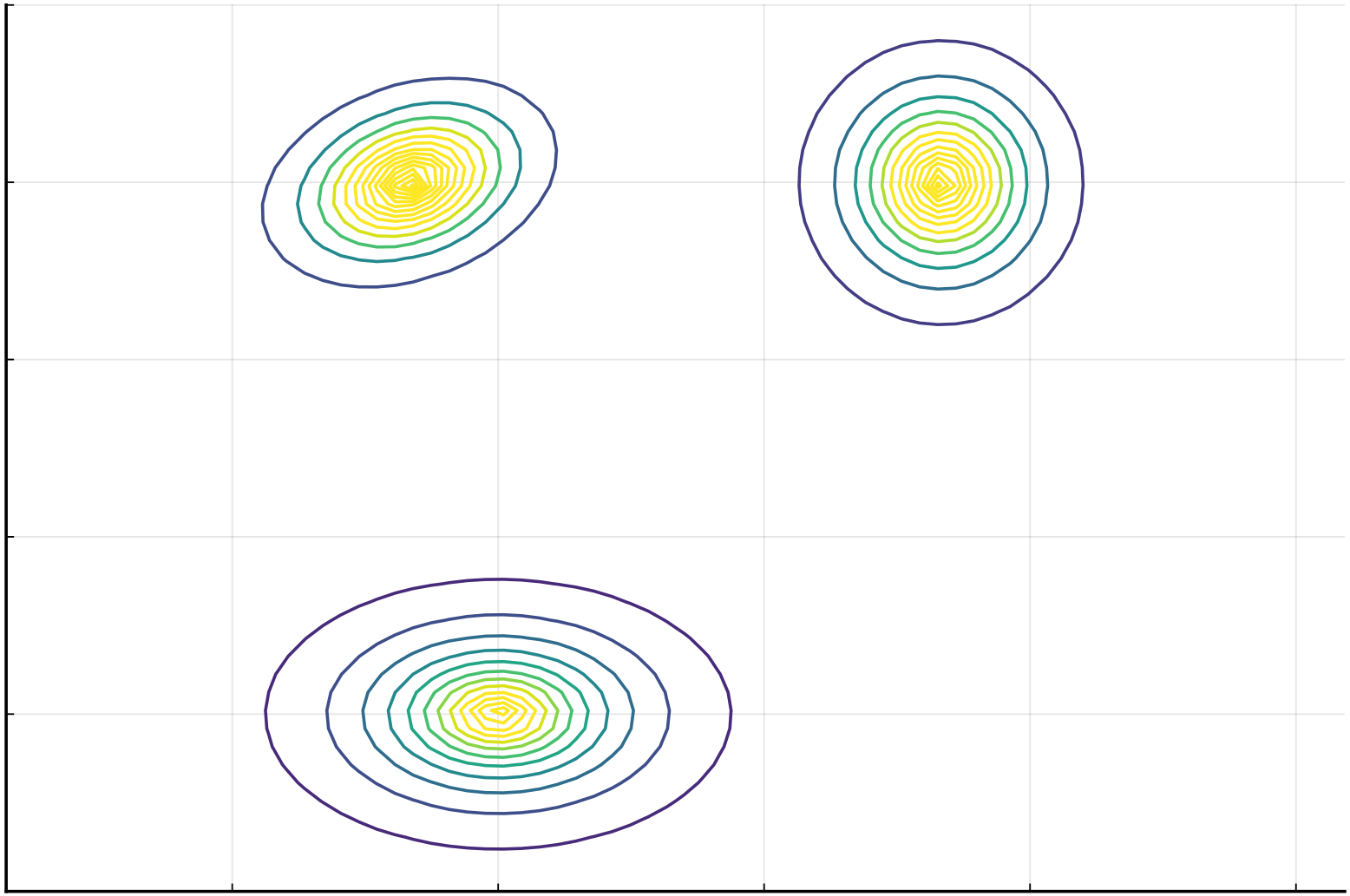
$$\mathrm{Bar}^{\boldsymbol{\lambda}}(\boldsymbol{\rho}) = \mathcal{N}(\mu_\star, S_\star)$$

where

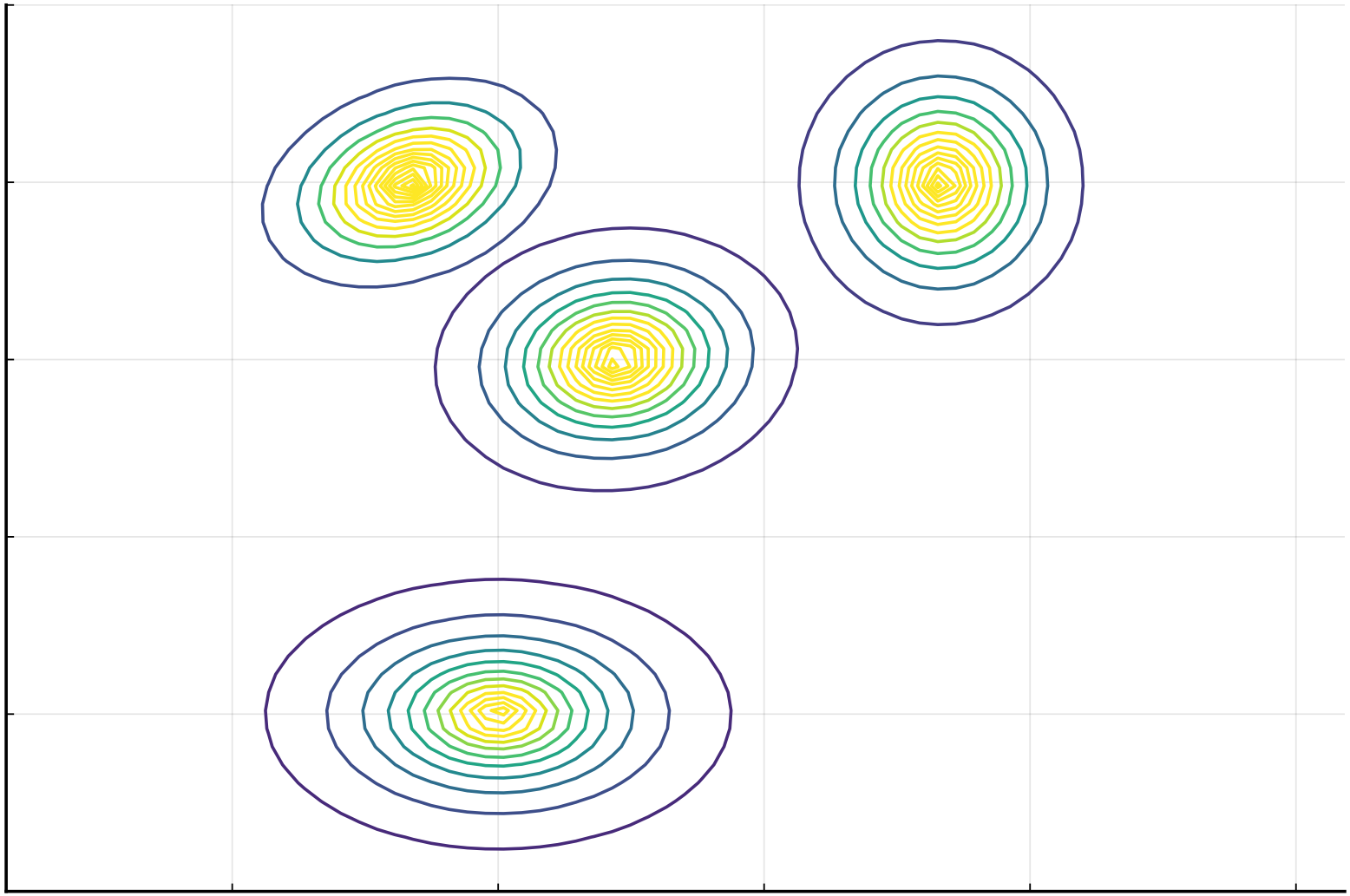
$$\mu_\star = \sum_{m=1}^{M} \lambda_m \mu_m, \qquad \sum_{m=1}^{M} \lambda_m \left(\sqrt{S_\star} S_m \sqrt{S_\star}\right)^{1/2} = S_\star,$$

$S_\star \in \mathcal{S}_{+,\star}^n$ is the unique symmetric positive definite matrix solution to this equation.

Illustration



Illustration



Mixture distance and barycenters

$\mathcal{A} \subset \mathcal{P}(\mathbb{R})$: dictionary of atoms (Slater functions [10], Gaussians, etc.)

Mixture distance [a]

For two mixtures $\mu = \sum_{j=1}^{J} \alpha_j \mu_j, \quad \nu = \sum_{k=1}^{K} \beta_k \nu_k$, define

$$MW_2(\mu,\nu)^2 := \min_{w \in \Pi(\alpha,\beta)} \sum_{j=1}^{J} \sum_{k=1}^{K} w_{jk} W_2^2(\mu_j, \nu_k),$$

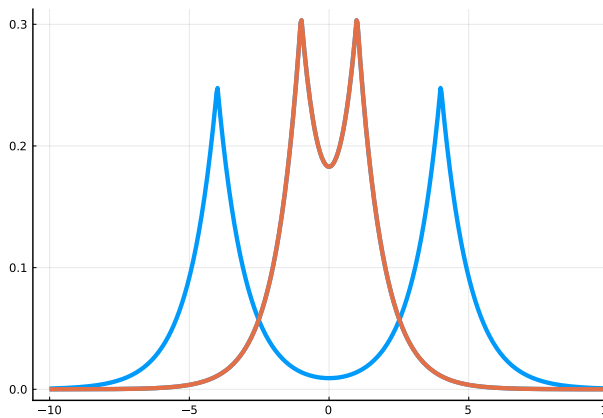
$\Pi(\mu,\nu)$ is the set of couplings between α and β.

a. Delon, Desolneux, SIAM J. Imaging Sciences, 2020

Advantages :

- Independent of the spatial dimension
- Keep the structure of the atoms
- Cheap if W_2 distance between atoms can be computed efficiently
- Formula for the barycenter :

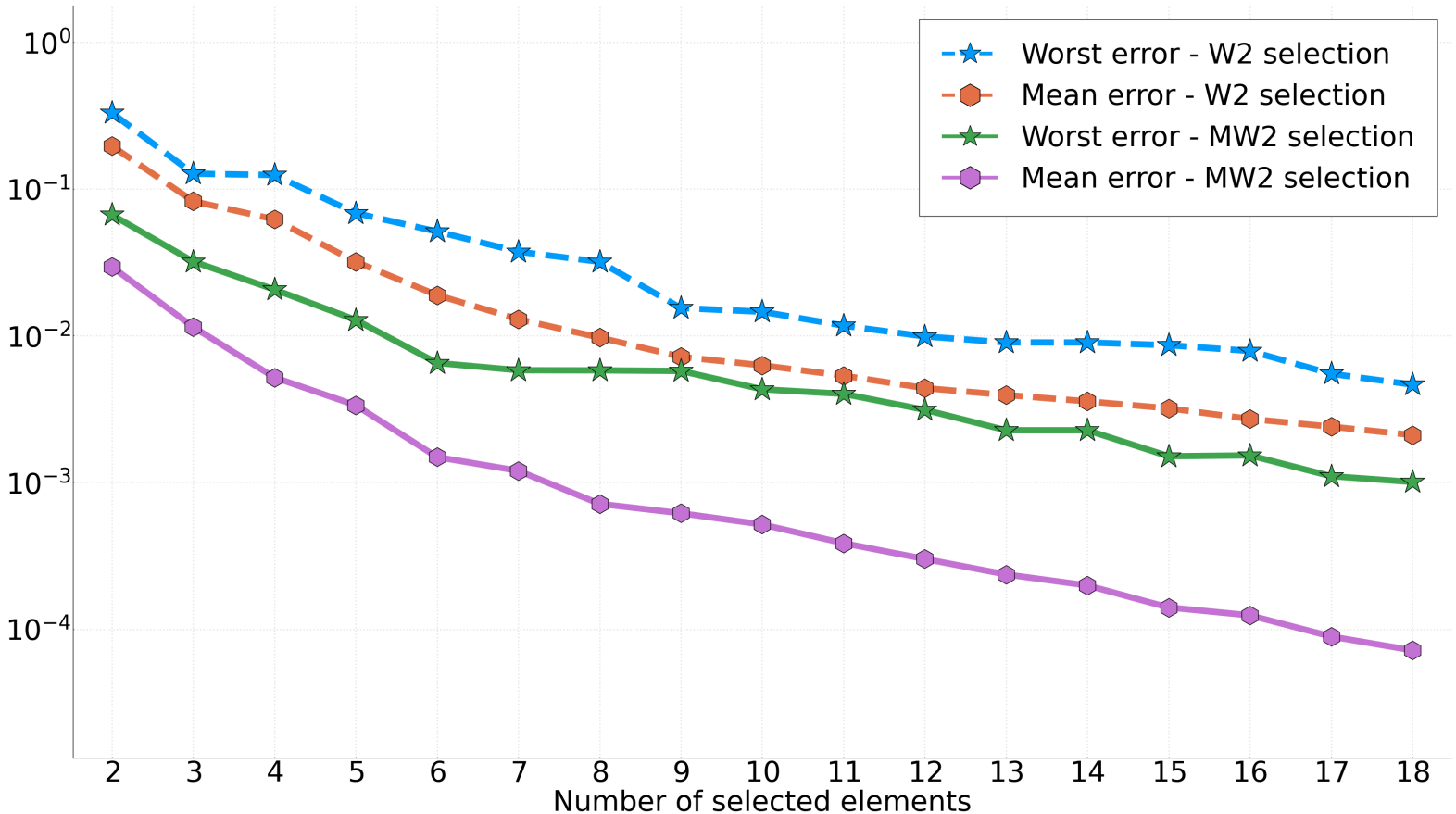
$$\mathrm{Bar}^{\boldsymbol{\lambda}}_{\mathrm{MW}_2}(\psi_1,\ldots,\psi_n) = \sum_{\mathbf{k}\in\mathbf{K}} w^*_{\mathbf{k}}\, \mathrm{Bar}^{\boldsymbol{\lambda}}_{W_2}\left(\psi_1^{k^1},\ldots,\psi_n^{k^n}\right),$$



10. GD, Ehrlacher, Nouaime, cocv 2026

Numerical results : greedy algorithm

Charges : $(z_1, z_2) = (0.8, 1.1)$.
291 solutions in the training set.
Projection error decreases with respect to number of selected snapshots



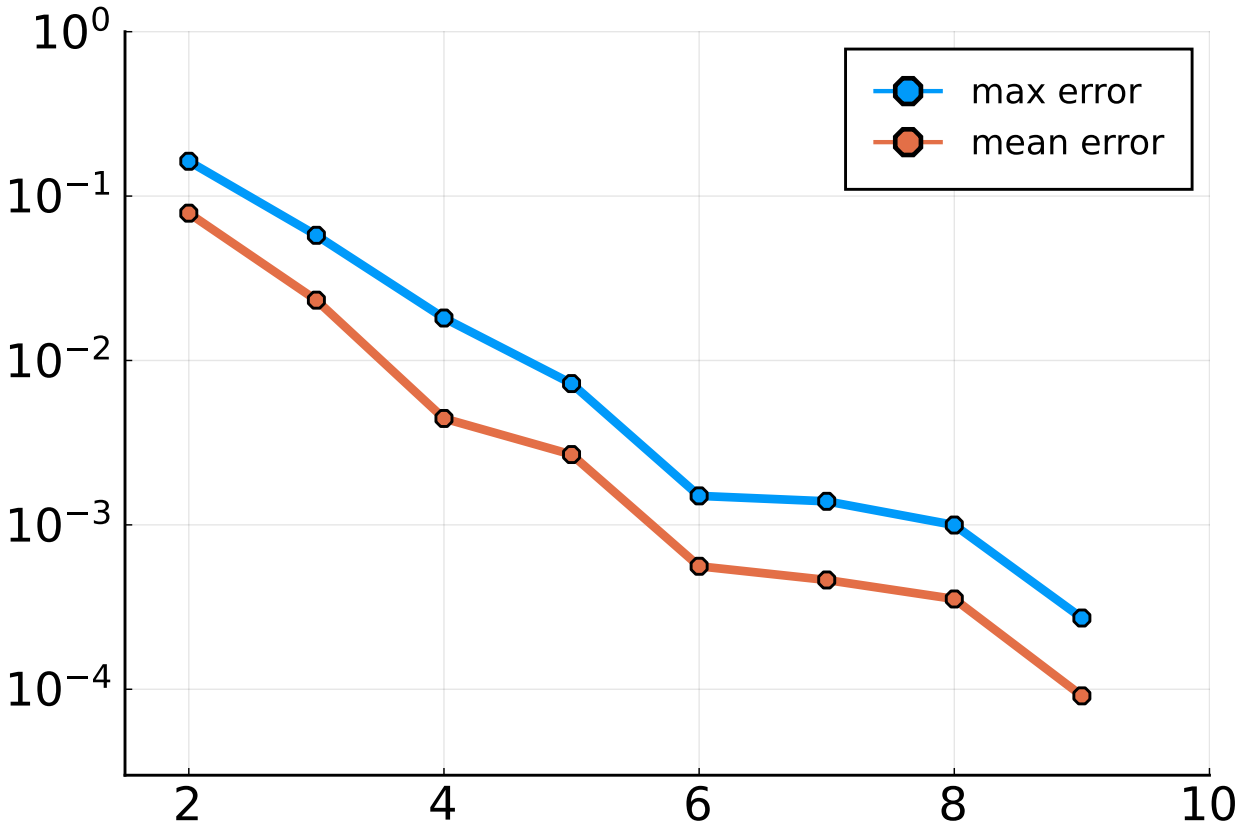
Numerical results : online energy minimization

Given selected $\Psi_{\mathbf{R}_1}, \ldots, \Psi_{\mathbf{R}_n}$, solve

$$\inf_{\lambda} E(\mathrm{bar}^{\lambda}(\Psi_{\mathbf{R}_1}, \ldots, \Psi_{\mathbf{R}_n}))$$

- Nonlinear problem, but in low dimension
- Using quasi-Newton method starting from different initial guesses

Energy error



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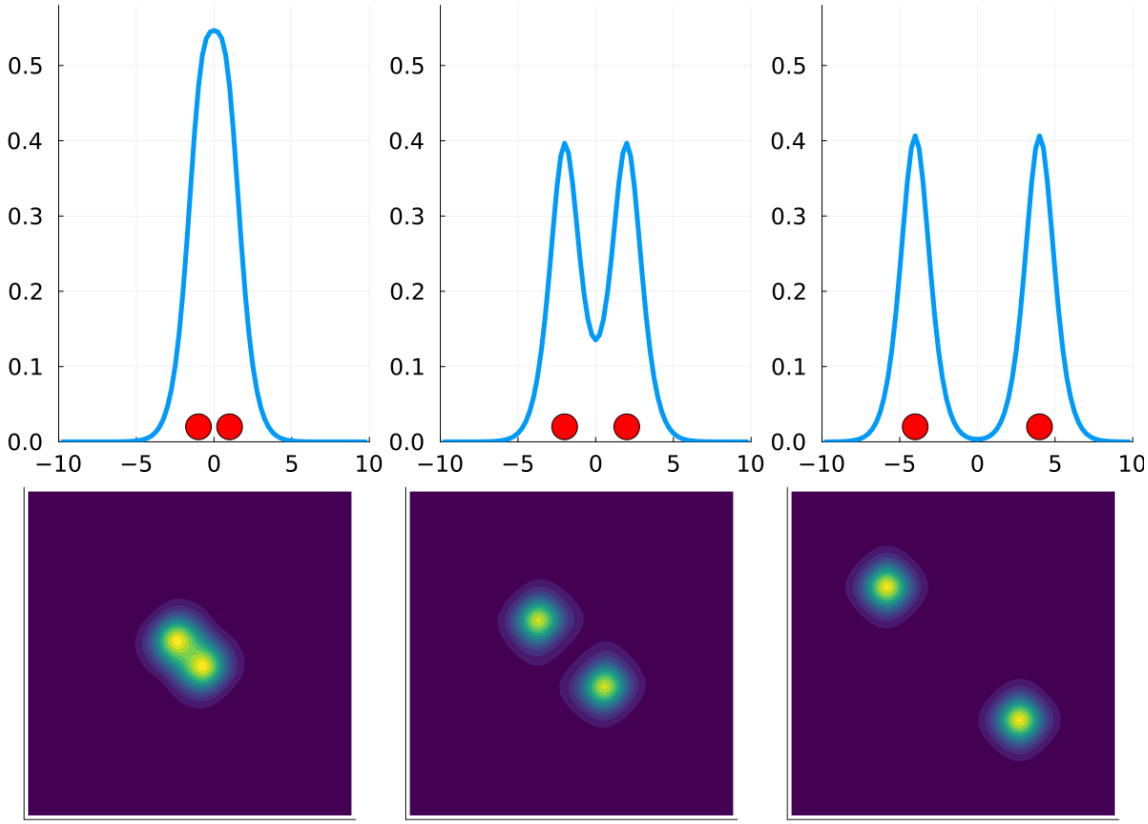
Model-order reduction for the pair density

Density $\rho_{\mathbf{R}}(x)$

Pair density $\tau_{\mathbf{R}}(x, y)$



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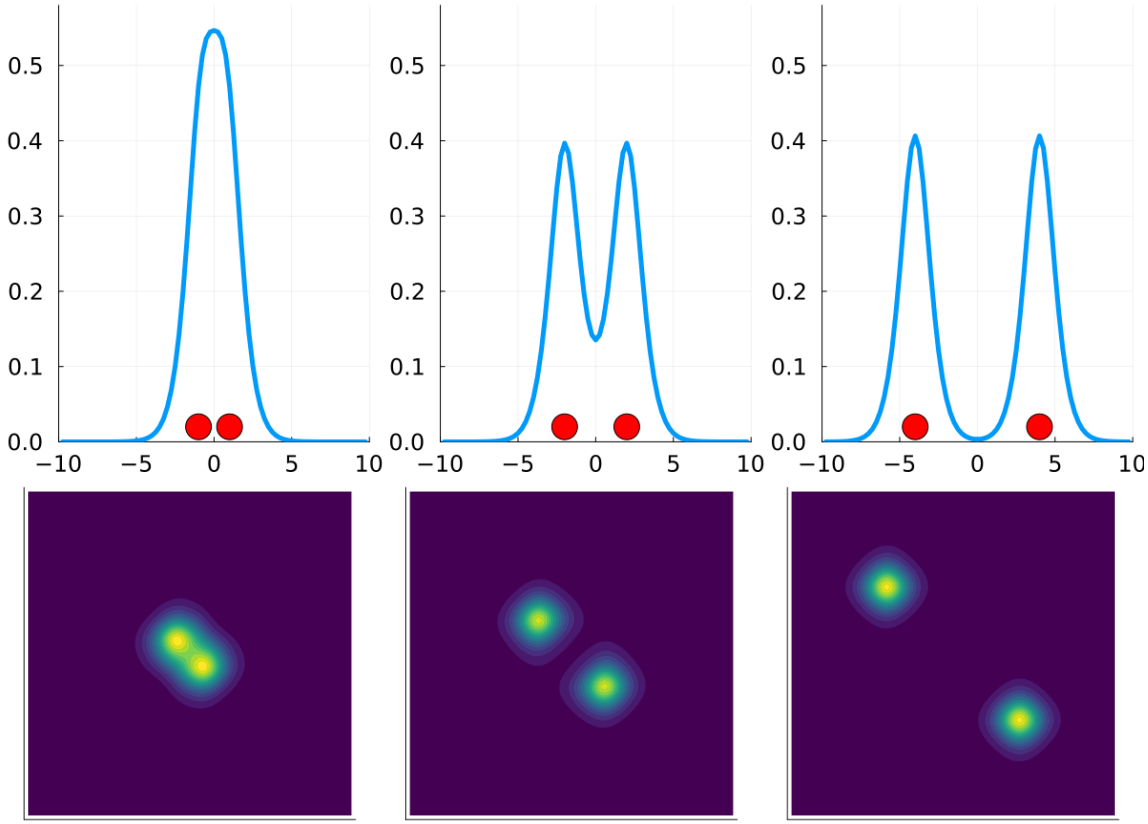

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Link between one-body and two-body densities

$$\int_{\mathbb{R}^3} \tau_{\boldsymbol{R}}(x, y)\, dy = \rho_{\boldsymbol{R}}(x), \quad \text{Marginal constraint}$$

Issue related to marginal constraints

We would like that

$$\tau_{\mathbf{R}} \approx \mathrm{Bar}^{\lambda}_{W_2}(\tau_{\mathbf{R}_1}, \ldots, \tau_{\mathbf{R}_M}) \Rightarrow \rho_{\mathbf{R}} \approx \mathrm{Bar}^{\lambda}_{W_2}(\rho_{\mathbf{R}_1}, \ldots, \rho_{\mathbf{R}_M})$$

Not true in general !

11. Dalery, GD, Ehrlacher, SIMAX 2026, hal-04696783

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Not true in general !

Idea : Marginal-constrained modified Wasserstein barycenters

Two possible problem formulations [11] :

1- Minimize distance to known barycenter

$$\inf_{\substack{\tau \text{ such that} \\ \mathrm{marg}(\tau)=\rho_{\mathbf{R}}}} W_2(\tau, \mathrm{Bar}^{\lambda}_{W_2}(\tau_{\mathbf{R}_1}, \ldots, \tau_{\mathbf{R}_M}))$$

2- Constrained Frechet mean formulation

$$\inf_{\substack{\tau \text{ such that} \\ \mathrm{marg}(\tau)=\rho_{\mathbf{R}}}} \sum_{i=1}^{M} \lambda_i W_2^2(\tau, \tau_{\mathbf{R}_i})$$

11. Dalery, GD, Ehrlacher, SIMAX 2026, hal-04696783

Marginals of Gaussian measures

Setting : $n = n_x + n_y$ for some $n_x, n_y \in \mathbb{N}^\star$.

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Question : Find the closest Gaussian distribution $\tau = \mathcal{N}(\mu, S)$ to $\tau_{\mathrm{ref}} = \mathcal{N}(\mu_{\mathrm{ref}}, T)$ with marginals

$$\mathrm{marg}_x(\tau) = \mathcal{N}(\mu_x, S_x) \quad \text{and} \quad \mathrm{marg}_y(\tau) = \mathcal{N}(\mu_y, S_y),$$

that is

$$\inf W_2(\tau_{\mathrm{ref}}, \tau)^2$$

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Partial answer : Necessarily,

$$\mu = \begin{pmatrix} \mu_x \\ \mu_y \end{pmatrix}, \quad S = \begin{pmatrix} S_x & Z \\ Z^T & S_y \end{pmatrix}$$

for some $Z \in \mathbb{R}^{n_x \times n_y}$.

Main result

Theorem [Dalery, GD, Ehrlacher, 2025]
Let $n_x, n_y \in \mathbb{N}^*$ and let $T \in \mathcal{S}_{+,\star}^{n_x+n_y}$ with block decomposition

$$T = \begin{pmatrix} T_x & T_{xy} \\ T_{xy}^\intercal & T_y \end{pmatrix} \tag{1}$$

Denote for $Z \in \mathcal{C}_{S_x,S_y} := \left\{ Z \in \mathbb{R}^{n_x \times n_y},\ \|S_x^{-1/2} Z S_y^{-1/2}\|_2 < 1 \right\}$

$$S(Z) := \begin{pmatrix} S_x & Z \\ Z^\intercal & S_y \end{pmatrix}. \tag{2}$$

The minimization problem

$$Z^*_{T,S} \in \operatorname*{argmin}_{Z \in \mathcal{C}_{S_x,S_y}} \mathcal{W}_2^2(T, S(Z))$$

has a unique minimizer which is given by

$$Z^*_{T,S} = ({T_x}^{-1} \# S_x) T_{xy} ({T_y}^{-1} \# S_y),$$

with $\#$ denoting the geometric mean of covariance matrices [Bhatia 2009]

$$S \# T := S^{1/2} \left(S^{1/2} T^{-1} S^{1/2} \right)^{-1/2} S^{1/2}.$$

Marginal-constrained modified Wasserstein barycenter

Question : Find the closest Gaussian distribution for Wasserstein distance $\tau = \mathcal{N}(\mu, S)$ to $\tau_{\text{ref}} = \mathcal{N}(\mu_{\text{ref}}, S_{\text{ref}})$ with marginals

$$\text{marg}_x(\tau) = \mathcal{N}(\mu_x, S_x) \quad \text{and} \quad \text{marg}_y(\tau) = \mathcal{N}(\mu_y, S_y).$$

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Full answer :

$$\mu = \begin{pmatrix} \mu_x \\ \mu_y \end{pmatrix}, \quad S = \begin{pmatrix} S_x & Z^*_{S_{\text{ref}},S} \\ (Z^*_{S_{\text{ref}},S})^T & S_y \end{pmatrix}$$

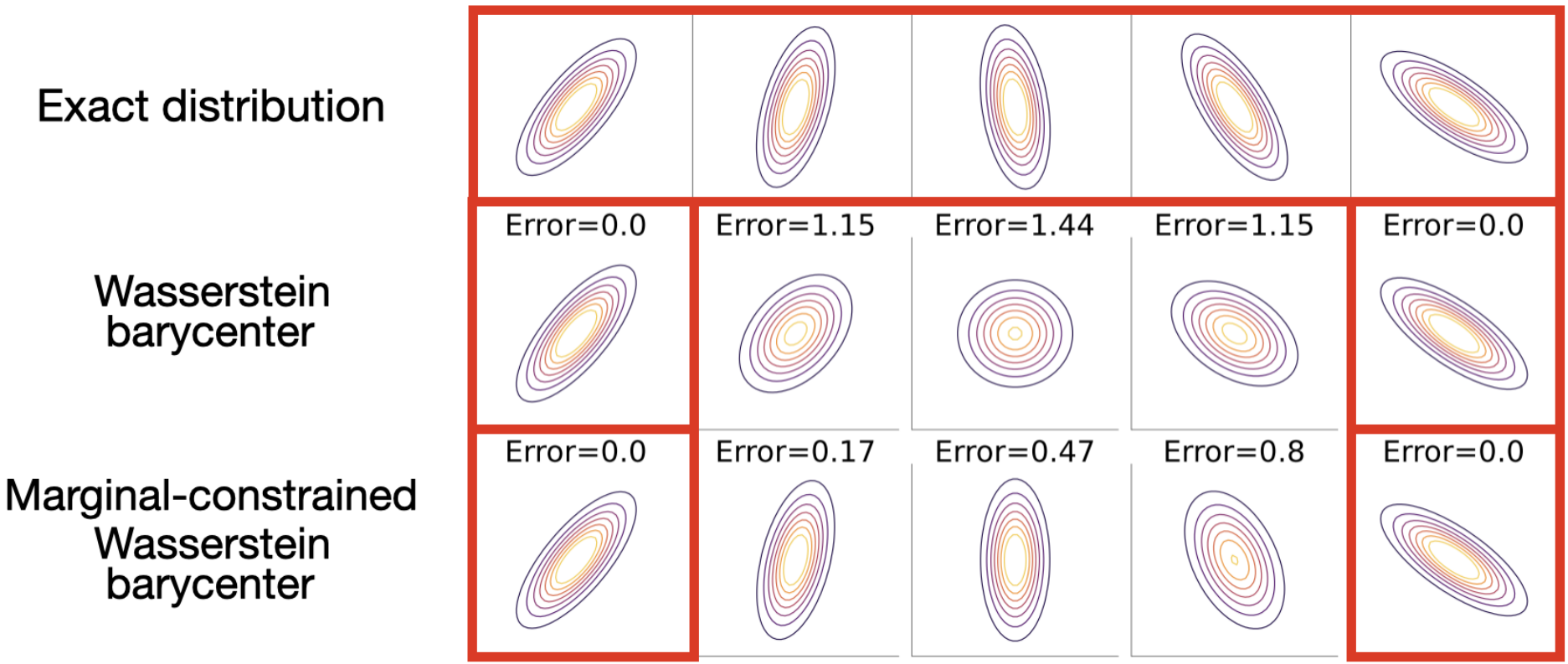
for some $Z \in \mathbb{R}^{n_x \times n_y}$.

Marginal-constrained barycenters (arbitrary number of Gaussians) : Choose τ_{ref} as a Wasserstein barycenter

Extension to Gaussian mixtures

- Write down a similar optimization problem (using mixture distance)
- Simplify this problem by specific choice of Gaussians
- Numerical resolution by postprocessing the mixture Wasserstein barycenter.

Results for Gaussians

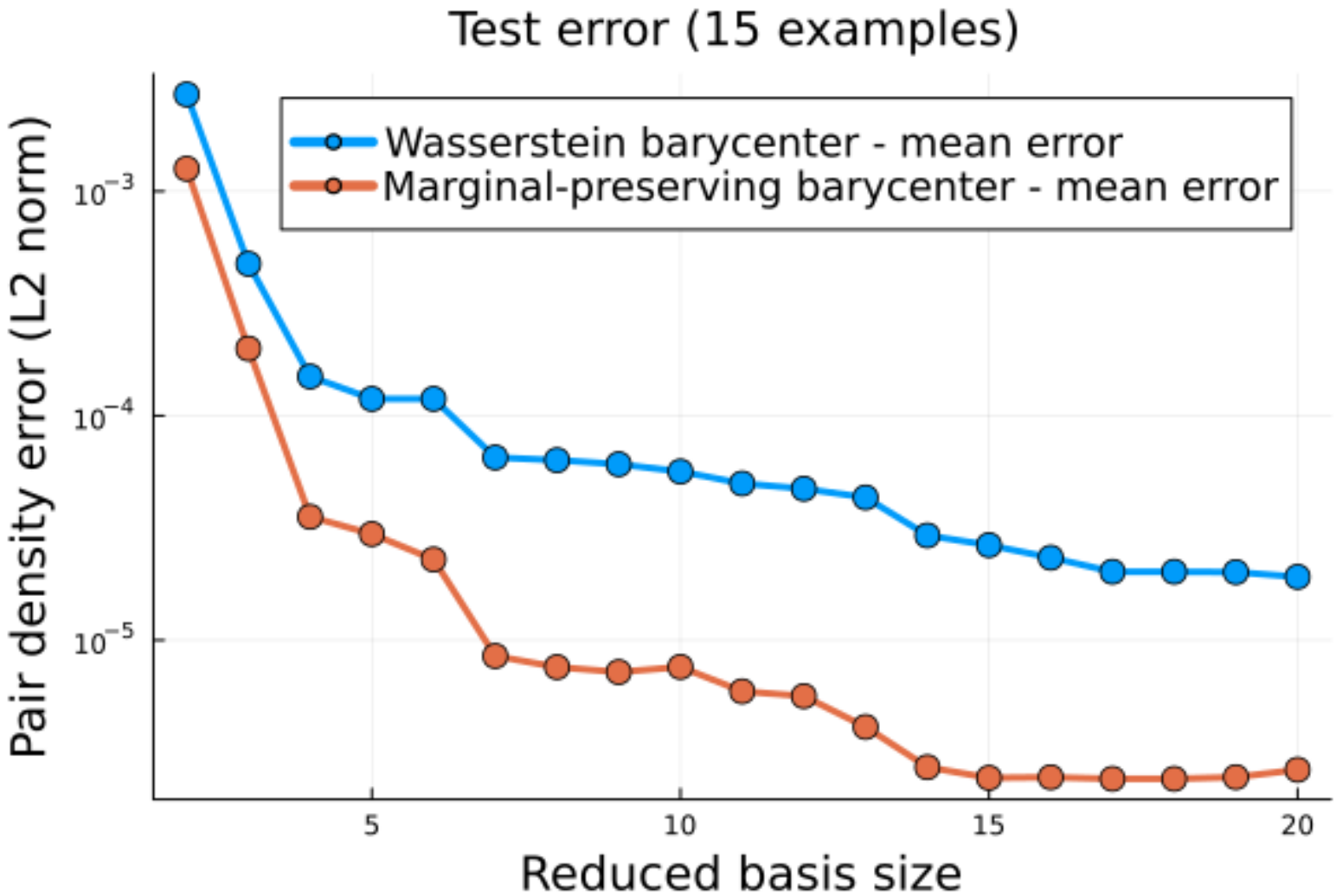


Fokker-Planck equation

$$\frac{\partial \rho}{\partial t} = -\nabla_{x,y} \cdot \left(A \begin{pmatrix} x \\ y \end{pmatrix} \rho \right) + D \Delta_{x,y} \psi \quad \text{with } D > 0 \text{ and } A = \Omega \begin{pmatrix} 0 & -1 \\ 1 & 0 \end{pmatrix}.$$

Advantage of marginal-preserving barycenters

Work in progress with Mengfan Tu (BNU), Virginie Ehrlacher (Cermics)



Outline

Introduction

From linear to metric reduced-order modelling

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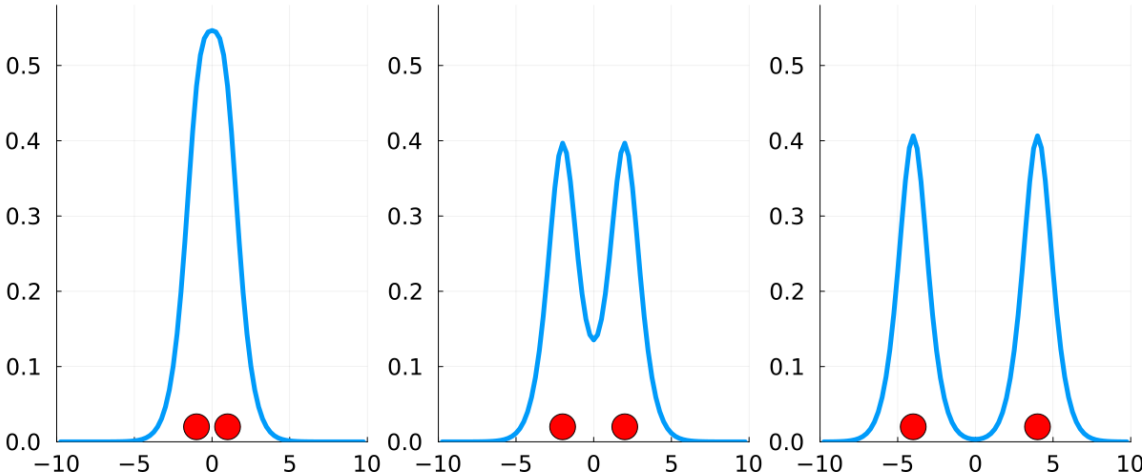
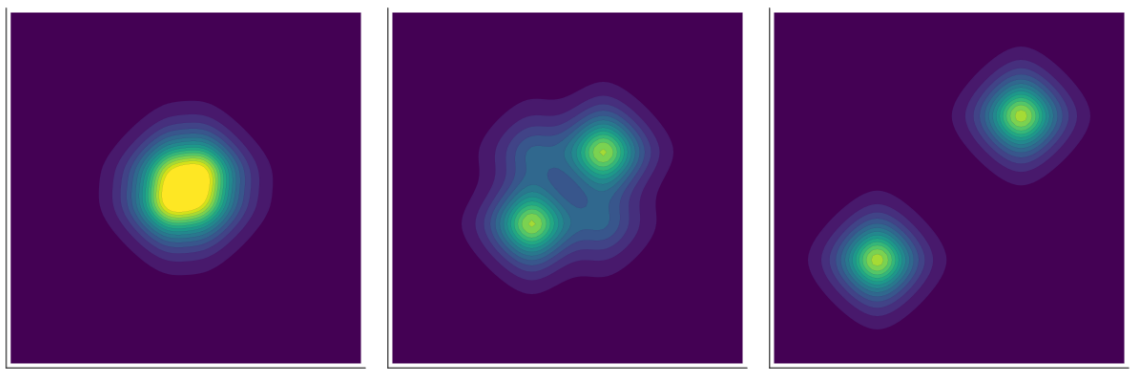
Model-order reduction for the density matrix

(with Etienne Obermeyer and Virginie Ehrlacher)

Density $\rho_{\mathbf{R}}(x)$

Density matrix $\gamma_{\mathbf{R}}(x, y)$





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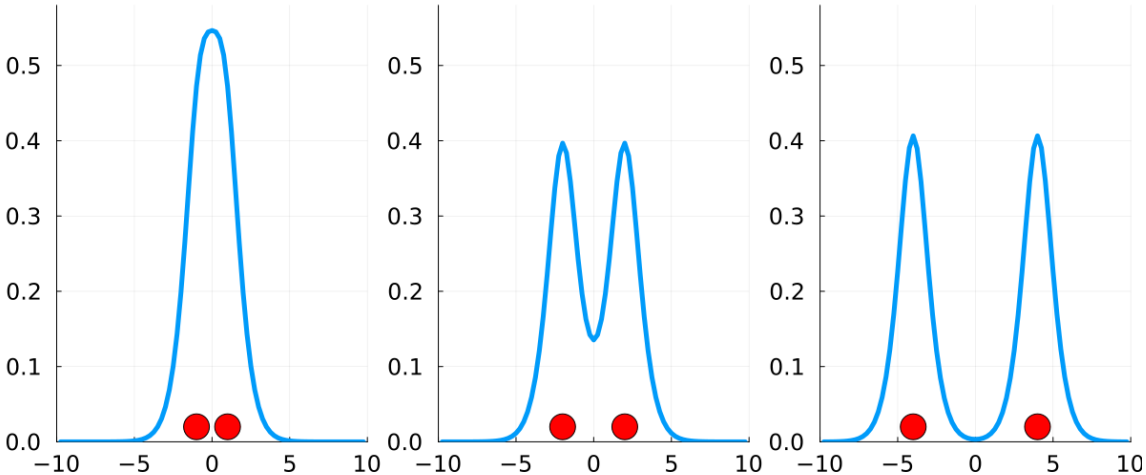

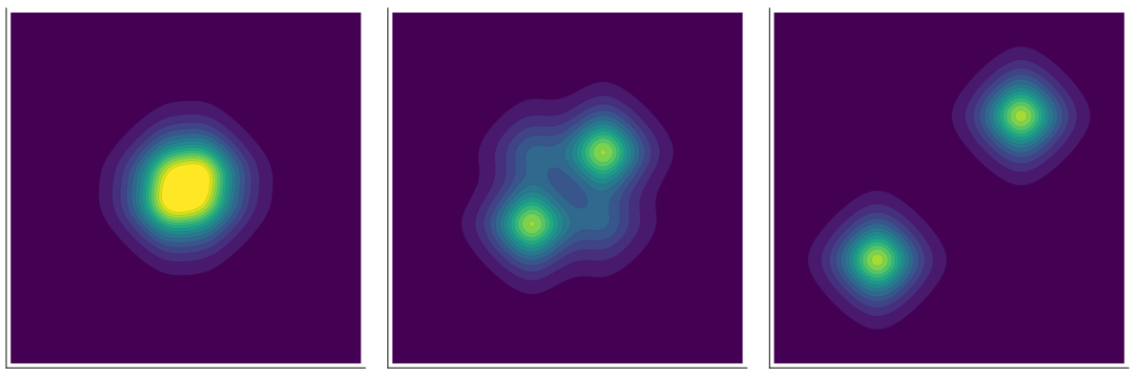

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Difficulty : Density matrices are not probability measures

From optimal transport to quantum optimal transport

But density matrices are self-adjoint operators, positive, of trace 1.

$$\mathcal{D}_+ = \{\gamma \in \mathcal{M}_n(\mathbb{C}),\ \gamma^* = \gamma,\ \gamma \succ 0,\ \mathrm{Tr}\,(\gamma) = 1\}$$

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Commutative math	Quantum math (OP = operators) ;
$\mathbb{R}^n$	$\mathcal{H}$ (Hilbert space)
bounded measures	trace OP (dual of compact OP)
real measures	self-adjoint OP
positive measures	positive OP
$\int$	trace
probabilities	**density matrices**
...	...

We use the quantum analog of the Benamou–Brenier formulation of optimal transport :

$$W_2(\rho_0, \rho_1)^2 = \inf_{\rho_t, v_t} \left\{ \int_0^1 \langle \rho_t v_t, v_t \rangle_{L^2(\mathbb{R}^n)}\ dt \ \middle|\ \frac{\partial \rho_t}{\partial t} + \mathrm{div}(\rho_t v_t) = 0 \right\}.$$

Quantum Optimal Transport

Carlen–Mass Riemannian [12] distance, defined on density matrices.
Extension of **dynamical formulation** of optimal transport
Step 1 : Define divergence with derivation operators L_j

$$\mathrm{div}(\mathbf{B}) = -\sum_{j\in\mathcal{J}} \partial_j B_j = -\sum_{j\in\mathcal{J}} (L_j B_j - B_j L_j).$$

Step 2 : Define the distance

$$\begin{aligned} d(\gamma_0,\gamma_1) = \min_{\gamma_t\in\mathcal{D}_+,\ v_t\in\mathcal{S}^N} & \int_0^1 \mathrm{tr}(\gamma_t v_t^* v_t)\ dt, \\ & \frac{\partial\gamma_t}{\partial t} - \frac{1}{2}\,\mathrm{div}(v_t\gamma_t + \gamma_t v_t) = 0, \\ & \gamma(0) = \gamma_0, \quad \gamma(1) = \gamma_1. \end{aligned}$$

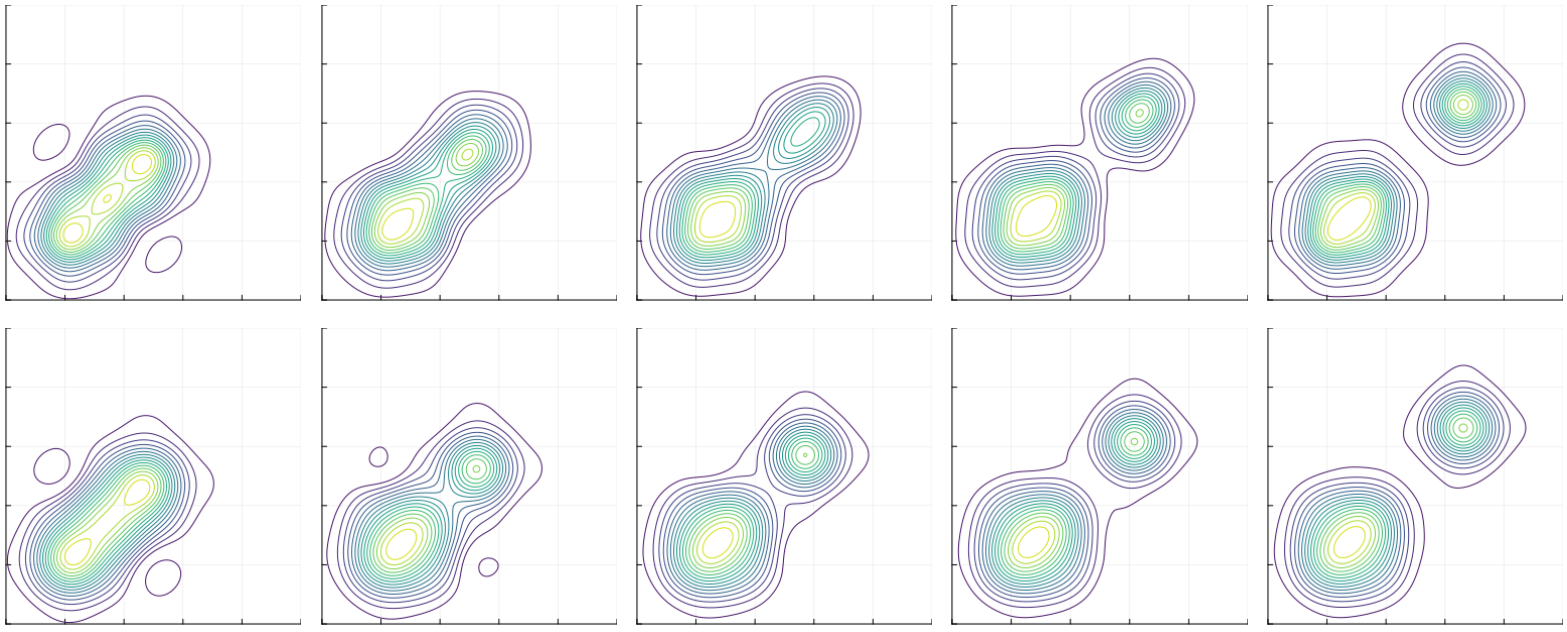
Numerical resolution : Time discretization [13], SQP, barrier method.

12. Carlen and Maas, Journal of Statistical Physics, (2020)
13. Chen, Haber, Yamamoto, Georgiou, and Tannenbaum, 2017

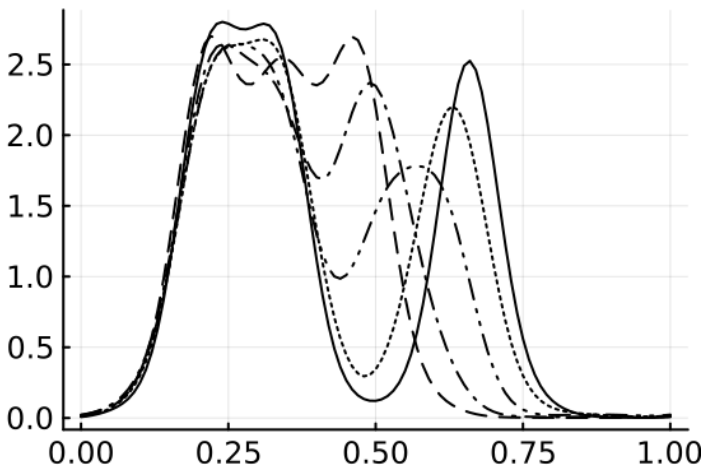
QOT vs Quantum chemistry (2D plot)

QOT

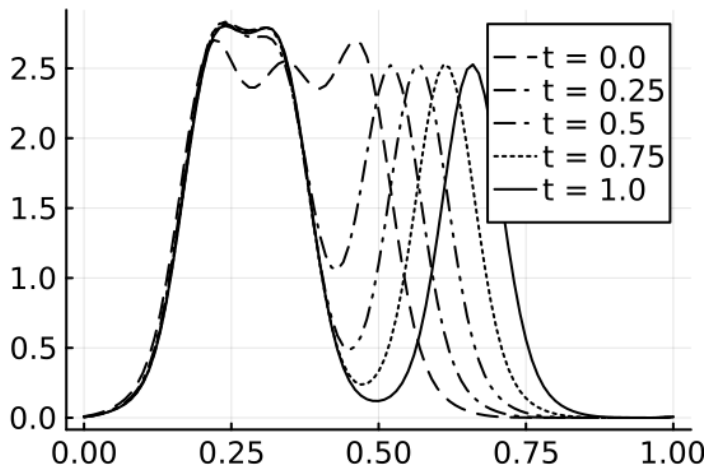
Chemistry



γ_0 $\gamma_{0.25}$ $\gamma_{0.5}$ $\gamma_{0.75}$ γ_1



QOT Diagonal (best fit)



Chemistry computations.

Choice of operators defining the metric

What is the best choice ?

Using Fourier discretization, so far we tested

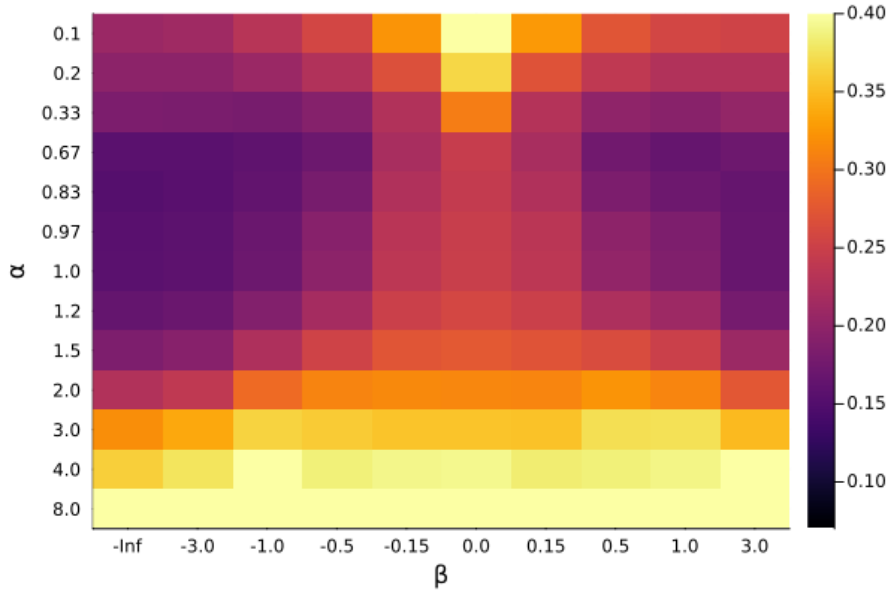
$$L_1(\alpha) = \begin{pmatrix} -(\frac{n-1}{2})^\alpha & & (0) \\ & \ddots & \\ (0) & & (\frac{n-1}{2})^\alpha \end{pmatrix},$$

$$L_2(\beta) = \begin{pmatrix} 0 & 1 & 2^\beta & \cdots & (n-1)^\beta \\ 1 & 0 & 1 & \cdots & (n-2)^\beta \\ 2^\beta & 1 & 0 & \cdots & (n-3)^\beta \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ (n-1)^\beta & (n-2)^\beta & (n-3)^\beta & \cdots & 0 \end{pmatrix}.$$

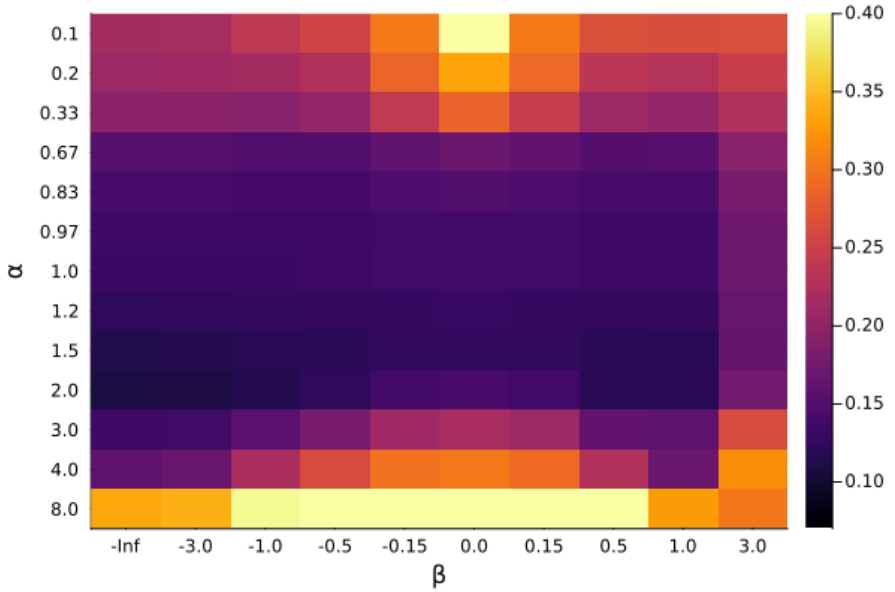
Satisfies the condition : $\ker(\mathrm{div}) = \mathrm{Span}(I_n)$.

Comparison between different metrics

Heatmaps of errors for different derivations.



(a) Two-electron case. The best derivation set is $\alpha = 0.83, \beta = -\infty$.



(b) Three-electron case The best derivation set is $\alpha = 2, \beta = -\infty$.

Error on the curve of density matrices :

$$\mathrm{d}_{\infty,2}(\gamma, \gamma^{\mathrm{ref}}) = \|\gamma - \gamma^{\mathrm{ref}}\|_{\infty,2} = \max\left\{\|\gamma_{p+\frac{1}{2}} - \gamma^{\mathrm{ref}}_{p+\frac{1}{2}}\|_2,\ p \in \{1, \ldots, P-1\}\right\}$$

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Limitations and perspectives :

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Thank you for your attention.

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GD, Ehrlacher, Nouaime, cocv 2026.
GD, Ehrlacher, Obermeyer, arXiv :2606.10075.