Grassmann extrapolation of density matrices

Étienne Polack
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Joint work with G. Dusson, F. Lipparini and B. Stamm

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Problem statement

Self-consistent field method for Hartree–Fock

For a molecular system of N occupied molecular orbitals and an LCAO basis set of size $\mathcal{N}$

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where

$$\mathcal{M}_{\mathsf{p}} := \{\tilde{\mathsf{C}} \in \mathbb{R}^{\mathcal{N} \times N} \,|\, \tilde{\mathsf{C}}^\top \mathsf{S}_{\mathsf{p}} \tilde{\mathsf{C}} = \mathrm{Id}_N\}. \tag{2}$$

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We want thus to find $\tilde{\mathsf{C}}_{\mathsf{p}} \in \mathcal{M}(\mathsf{p})$ and a diagonal matrix $\mathsf{E}_{\mathsf{p}} \in \mathbb{R}^{N \times N}$ such that

$$\begin{cases} \mathsf{F}(\tilde{\mathsf{D}}_{\mathsf{p}})\tilde{\mathsf{C}}_{\mathsf{p}} = \mathsf{S}_{\mathsf{p}} \tilde{\mathsf{C}}_{\mathsf{p}} \mathsf{E}_{\mathsf{p}} \\ \tilde{\mathsf{C}}_{\mathsf{p}}^\top \mathsf{S}_{\mathsf{p}} \tilde{\mathsf{C}}_{\mathsf{p}} = \mathrm{Id}_N \\ \tilde{\mathsf{D}}_{\mathsf{p}} = \tilde{\mathsf{C}}_{\mathsf{p}}^\top \tilde{\mathsf{C}}_{\mathsf{p}} \end{cases}, \quad \text{where} \quad \mathsf{F}_{\mathsf{p}}(\mathsf{D}) := \mathsf{h}_{\mathsf{p}} + \mathsf{G}_{\mathsf{p}}(\mathsf{D}) \,. \tag{3}$$

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We want thus to find $\tilde{C}_p \in \mathcal{M}(p)$ and a diagonal matrix $E_p \in \mathbb{R}^{N \times N}$ such that

$$\begin{cases} F(\tilde{D}_p)\tilde{C}_p = S_p \tilde{C}_p E_p \\ \tilde{C}_p^\top S_p \tilde{C}_p = \mathrm{Id}_N \\ \tilde{D}_p = \tilde{C}_p^\top \tilde{C}_p \end{cases}, \quad \text{where} \quad F_p(D) := h_p + G_p(D). \tag{3}$$

Non-linear term

Fixed-point algorithm

Choose some $\tilde{\mathsf{C}}_0$ such that $\tilde{\mathsf{C}}_0^\top \mathsf{S}_\mathsf{p} \tilde{\mathsf{C}}_0 = \mathrm{Id}_N$ and construct a sequence $(\tilde{\mathsf{C}}_n)_{n\in\mathbb{N}}$ that verifies

$$\begin{cases} \mathsf{F}(\tilde{\mathsf{D}}_n)\tilde{\mathsf{C}}_n = \mathsf{S}_\mathsf{p}\tilde{\mathsf{C}}_n\mathsf{E}_n \\ \tilde{\mathsf{C}}_n^\top \mathsf{S}_\mathsf{p}\tilde{\mathsf{C}}_n = \mathrm{Id}_N \\ \tilde{\mathsf{D}}_n = \tilde{\mathsf{C}}_n^\top \tilde{\mathsf{C}}_n \end{cases} . \tag{4}$$

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- When doing molecular dynamics or geometry optimization for a given molecular system, we may discard lots of previous pieces of information.

→ How can we make the most of repeated self-consistent field computations on the same molecular system?

Geometric interpretation

Grassmannian manifold

Density matrices can be seen as points on a Grassmannian manifold

$$\mathsf{S}_\mathsf{p}^{\frac{1}{2}} \tilde{\mathsf{D}}_\mathsf{p} \mathsf{S}_\mathsf{p}^{\frac{1}{2}} \in \mathcal{M}_{\mathrm{Gr}} := \{\mathsf{D} \in \mathbb{R}^{\mathcal{N} \times \mathcal{N}} \mid \mathsf{D} = \mathsf{D}^\top, \mathsf{D}^2 = \mathsf{D}, \mathrm{Tr}(\mathsf{D}) = N\}. \tag{5}$$

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⇒ We can do interpolation in the tangent space of some density matrix and map the result back to the Grassmannian.

Grassmannian manifold

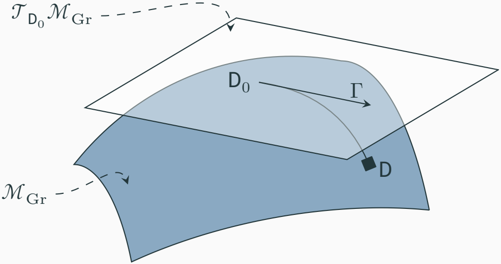

Figure 1: Schematic illustration of the geometrical setting. We illustrate by the blue hypersurface the Grassmann manifold $\mathcal{M}_{\mathrm{Gr}}$ and by the transparent plane the tangent space $\mathcal{T}_{\mathsf{D}_0}\mathcal{M}_{\mathrm{Gr}}$ to $\mathcal{M}_{\mathrm{Gr}}$ at D_0. We illustrate the one-to-one relationship between a close density matrix $\mathsf{D} \in \mathcal{M}_{\mathrm{Gr}}$ and the corresponding vector $\Gamma = \mathrm{Log}_{\mathcal{M}_{\mathrm{Gr}},0}\mathsf{D}$ in the tangent space.

Exponential and logarithmic maps

Locally, there exists a diffeomorphism that transform the manifold $\mathcal{M}_{\mathrm{Gr}}$ into an affine space, the tangent space, and back.

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The maps are the exponential

$$\begin{aligned} \mathrm{Exp}_{\mathcal{M}_{\mathrm{Gr}},0} : \mathcal{T}_{D_0} &\to \mathcal{M}_{\mathrm{Gr}} \\ \Gamma &\mapsto CC^{\mathsf{T}} \end{aligned}, \tag{6}$$

where $C = [C_0 V \cos(\Sigma) + U \sin(\Sigma)] V^{\mathsf{T}}$, with $\Gamma = U \Sigma V^{\mathsf{T}}$ the singular value decomposition of Γ.

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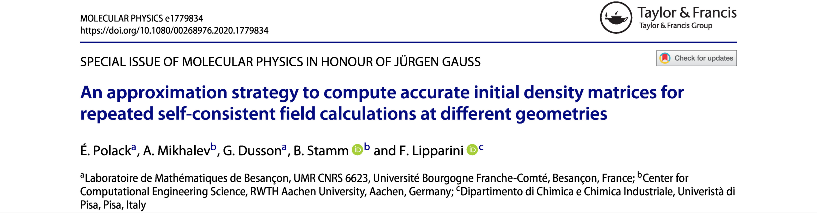
And the logarithm function

$$\begin{aligned} \mathrm{Log}_{\mathcal{M}_{\mathrm{Gr}},0} : \mathcal{M}_{\mathrm{Gr}} &\to \mathcal{T}_{D_0} \\ D &\mapsto \mathrm{Log}_{\mathcal{M}_{\mathrm{Gr}},0}(D) \end{aligned} \tag{7}$$

can be defined in a similar way.

Model problem (recapitulation)

Supporting article



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An approximation strategy to compute accurate initial density matrices for repeated self-consistent field calculations at different geometries

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Constrained example

Aim

Provide accurate density matrices guesses for the self-consistent field algorithm with localised basis functions and where the nuclear coordinates are changed along a few user-specified collective variables.

Constrained example

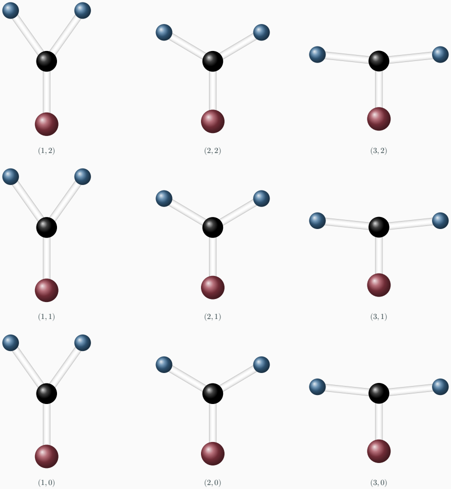

Figure 2: Schematic representation of the parameter space along two normal modes for the formaldehyde. The positions of the atoms are projected on two axes.

Constrained example

Method objectives:

- No limits on computational cost for data generation of some values of the normal modes.

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- No limits on computational cost for data generation of some values of the normal modes.
- Negligible on-the-fly cost for new values.
- Should work for large energy fluctuations.
- Should be able to provide a guess accurate enough to bypass self-consistent field iterations.

Constrained example

	Alanine	Asparagine	Phenylalanine	Tryptophan
#atoms	13	17	23	27
Core	21	21	23	26
Harris	13	14	14	15
Hückel	16	17	17	18
MinAO	15	17	17	17
SAD	16	17	17	17

Table 1: Number of SCF iterations required to achieve convergence (max change in the density smaller than 10^{-6} using different initial guesses. As the computations were carried out using different packages, that offer different SCF implementations, this cannot be considered an accurate comparison between the various guesses, but only a qualitative estimate of the number of required iterations. Note that all the calculations have been performed using standard DIIS extrapolation.

Results

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- blackbox for the chemists;
- does not change the final results.

Results (1D) – Lagrangian interpolation

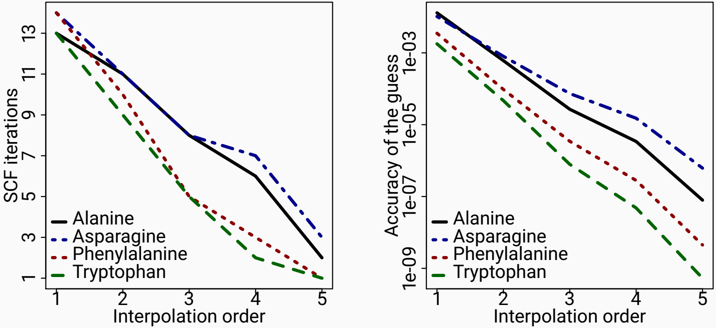

Figure 3: Results for the 1D parameter space. Number of SCF iterations required to achieve convergence (left panel) and Frobenius norm error on the density guess (right panel) as a function of the interpolation order for the various test systems. All the calculations were performed with CFOUR using the following convergence criteria for the increment of the density ΔP: RMS $\Delta P < 10^{-7}$ and max $|\Delta P| < 10^{-6}$.

Results (2D)

With the cc-pVDZ basis set, the energy fluctuates of 9.1, 8.9, 8.5 and 7.6 kcal/mol for alanine, asparagine, phenylalanine, and tryptophan, respectively.

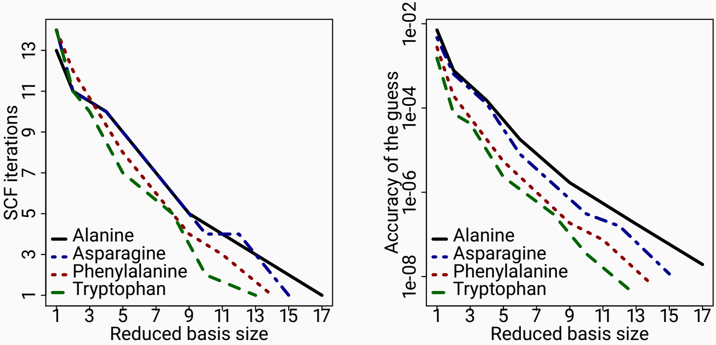

Figure 4: Results for the 2D parameter space. Number of SCF iterations required to achieve convergence (left panel) and Frobenius norm error on the density guess (right panel) as a function of the interpolation order for the various test systems. All the calculations were performed with CFOUR using the following convergence criteria for the increment of the density ΔP: RMS $\Delta P < 10^{-7}$ and max $|\Delta P| < 10^{-6}$.

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Takeaway

We can almost instantly predict the density matrices of all other configurations using only a small number of data.

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We can almost instantly predict the density matrices of all other configurations using only a small number of data.

However, we need access to underlying variables.

Molecular dynamics

Objectives

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- Bypassing underlying variables;
- Useful for large molecular systems;
- Negligible overhead;
- Keep the method as simple as possible.

Method

Molecular descriptors

We split the mapping from positions to points on the density matrices manifold with

$$\begin{aligned} \mathbb{R}^{3M} &\to \mathcal{M} \to \mathcal{T}_{D_0} \to \mathcal{M}_{\mathrm{Gr}}(N,\mathcal{N}) \\ \mathsf{R} &\mapsto d_{\mathsf{R}} \mapsto \Gamma_{\mathsf{R}} \mapsto D_{\mathsf{R}} = \mathrm{Exp}_{\mathcal{M}_{\mathrm{Gr}},0}(\Gamma_{\mathsf{R}}). \end{aligned} \tag{8}$$

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We settle on the Coulomb matrix

$$(d_{\mathsf{R}})_{ij} = \begin{cases} 0.5 z_i^{2.4} & \text{if } i = j \\ \dfrac{z_i z_j}{\|\mathsf{R}(t_i) - \mathsf{R}(t_j)\|} & \text{otherwise} \end{cases}, \tag{9}$$

where t_i is the time step i.

Method – Least-squares

We look for coefficients $c_{\mathsf{R},i}$ that can approximate the density matrices on the tangent space

$$\mathsf{R} \mapsto \Gamma_{\mathrm{app}}(\mathsf{R}) = \sum_{i=1}^{N_t} c_{\mathsf{R},i}\,\Gamma_i \in \mathcal{T}_{D_0}, \tag{10}$$

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We use a least-squares method, to solve

$$\min_{c_{\mathsf{R}} \in \mathbb{R}^{N_t}} \left\| d_{\mathsf{R}} - \sum_{i=1}^{N_t} c_{\mathsf{R},i} d_{\mathsf{R}(t_i)} \right\|^2. \tag{11}$$

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We use the same coefficients for the density matrices on the tangent space and use the density matrix

$$D_{\mathrm{app}}(\mathsf{R}) = \mathrm{Exp}_{\mathcal{M}_{\mathrm{Gr}},0} \left(\sum_{i=1}^{N_t} c_{\mathsf{R},i}\, \Gamma_i \right) \tag{12}$$

as an initial guess to the SCF algorithm.

Method – Schematic representation

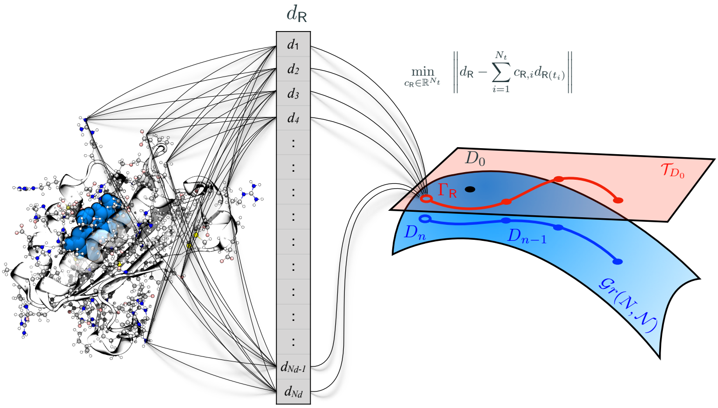


Figure 5: Schematic illustration of the G-Ext method

Outline of the algorithm

Data: Array desc containing the descriptors for k previous time-steps, p_n the descriptor for the current position, C_{n-1} and S_{n-1} respectively the molecular orbitals and overlap matrices of the previous time-step, and cref the reference point on the Grassmannian

Result: Guess density matrix for time-step $n > 1$

```
begin
    cmat(:, :, $n-1$) ← Orthonormalization($C_{n-1}$, $S_{n-1}$);
    gmat(:, :, $n-1$) ← Log(cref, cmat(:, :, $n-1$));
    desc, $p_n$ ← Stabilization(desc, $p_n$);
    c ← LeastSquares(desc, $p_n$);
    $\Gamma_{\mathrm{app}} \leftarrow \sum_{i=n-1-k}^{n-1}$ c($i$) · gmat(:, :, $i$);
    $C_{\mathrm{app}}$ ← Exp(cref, $\Gamma_{\mathrm{app}}$);
    return $2 \cdot C_{\mathrm{app}} \cdot C_{\mathrm{app}}^{\mathsf{T}}$;
end
```

Algorithm 1: Density extrapolation framework G-Ext

Results – Test cases

System	N_{QM}	N_{MM}	$\mathcal{N}$
OCP	129	4915	1038
APPA	31	16 449	309
DMABN	21	6843	185
3HF	28	15 018	290

Table 2: Overview of the system size in terms of number of quantum mechanics-atoms (N_{QM}), number of molecular mechanics-atoms (N_{MM}) and the total number of (quantum mechanics) basis functions ($\mathcal{N}$).

Results – Performances

Table 3: Performances of the G-Ext method for different number of extrapolation points, compared with the XLBO algorithm with and without McWeeny purification. All the results were obtained using a 10^{-5} convergence threshold for the root-mean-square increment of the density matrix and are derived from a 1 ps long molecular dynamics simulation, using a 0.5 fs time step. We report the average number of iterations required to converge the SCF, together with the associated standard deviation. Note that the first 8 steps were discarded.

	OCP		DMABN		APPA		3HF	
Method	Average	σ	Average	σ	Average	σ	Average	σ
XLBO	3.82	0.66	3.98	0.16	3.00	0.03	4.00	0.14
XLBO/MW	2.95	0.31	3.76	0.56	3.00	0.34	3.96	0.31
G-Ext(3)	2.57	0.84	3.54	0.78	2.95	0.50	3.09	0.41
G-Ext(4)	2.48	0.88	3.14	0.62	2.51	0.50	3.25	0.68
G-Ext(5)	2.25	0.96	3.23	0.75	2.51	0.50	3.30	0.72
G-Ext(6)	2.20	0.96	2.99	0.02	2.51	0.50	3.14	0.56

Results – Energy conservation

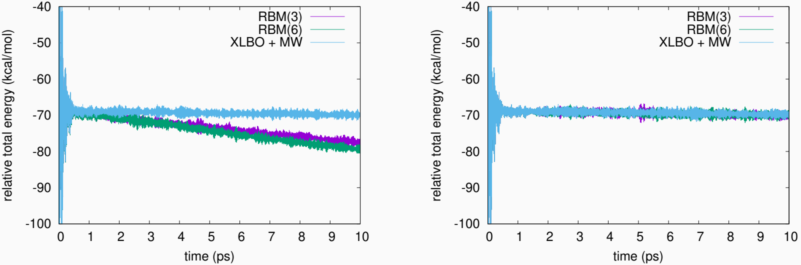
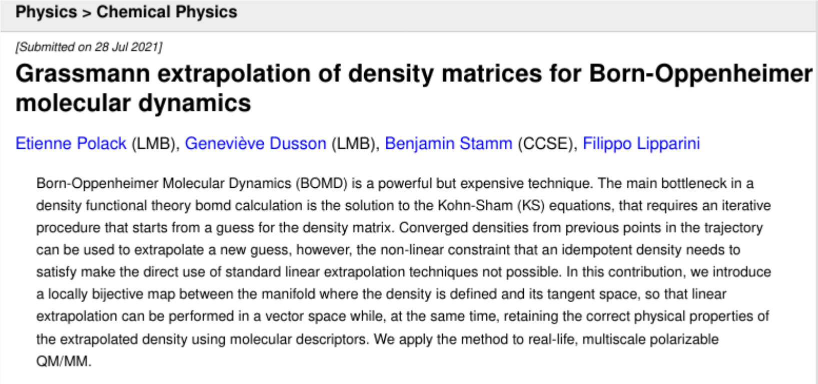


Figure 6: Total energy (kcal/mol) as a function of simulation time (fs) for 3HF comparing G-Ext(3), G-Ext(6) and XLBO with McWeeny purification, using a convergence threshold for the SCF algorithm of 10^{-5} (left panel) and 10^{-7} (right panel). The total energy was shifted of $+505\,000\,\mathrm{kcal/mol}$ for readability.

Supporting article



Physics > Chemical Physics

[Submitted on 28 Jul 2021]

Grassmann extrapolation of density matrices for Born-Oppenheimer molecular dynamics

Etienne Polack (LMB), Geneviève Dusson (LMB), Benjamin Stamm (CCSE), Filippo Lipparini

Born-Oppenheimer Molecular Dynamics (BOMD) is a powerful but expensive technique. The main bottleneck in a density functional theory bomd calculation is the solution to the Kohn-Sham (KS) equations, that requires an iterative procedure that starts from a guess for the density matrix. Converged densities from previous points in the trajectory can be used to extrapolate a new guess, however, the non-linear constraint that an idempotent density needs to satisfy make the direct use of standard linear extrapolation techniques not possible. In this contribution, we introduce a locally bijective map between the manifold where the density is defined and its tangent space, so that linear extrapolation can be performed in a vector space while, at the same time, retaining the correct physical properties of the extrapolated density using molecular descriptors. We apply the method to real-life, multiscale polarizable QM/MM.

Perspectives

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- Time reversibility;

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- Time reversibility;
- Geometry optimization;

Perspectives

- Time reversibility;
- Geometry optimization;
- Bypass the need for the SCF algorithm (long term).

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Questions?

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Comments!