Parameterless stopping criteria for density matrix expansions

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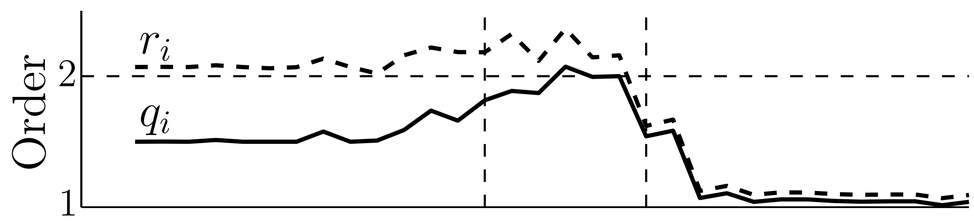
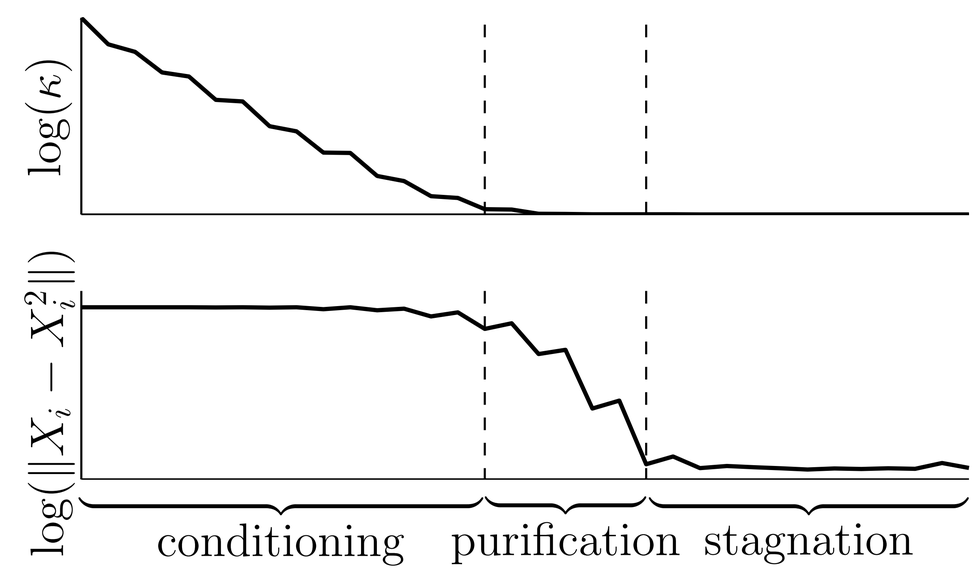
Introduction

Three phases in the recursive expansion:





κ is a condition number for the problem of computing the density matrix.

Theory

Given recursive expansion polynomials f_i: $e_i = \|X_i - X_i^2\|_2$

Theoretical order of convergence q satisfies:

$$\lim_{i\to\infty} \frac{e_i}{e_{i-1}^q} = C^\infty$$

Observed order of convergence in iteration i:

$$q_i := \frac{\log(e_i/C_\infty)}{\log(e_{i-1})} \quad \Rightarrow \quad r_i := \frac{\log(e_i/C_q)}{\log(e_{i-1})}$$

Our solution [1]: find smallest C_q such that

$$r_i \geq q \quad \Leftrightarrow \quad C_q \geq \frac{e_i}{e_{i-1}^q}$$

Compute r_i in every iteration.
If $r_i < q$, expansion reached stagnation phase (stop expansion).

How to find C_q?

- find $\max_{x\in[0,1]} \frac{f_i(x)-f_i(x)^2}{(x-x^2)^q}$
- $q > 1$ is the theoretical order of convergence
- due to possible small perturbations of the order, use $\tilde{q} < q$

Use Frobenius or mixed [2] norm for large systems:

$$\|X_i - X_i^2\|_2 \leq \|X_i - X_i^2\|_M \leq \|X_i - X_i^2\|_F$$

Conclusion

Stopping criteria ...

- automatically and accurately detect when numerical errors start to dominate, i.e. transition between purification and stagnation phases;
- do not require any user defined parameter;
- are general, can be derived for various choices of polynomials;
- can be used for various strategies for removal of small matrix elements;
- can be used for dense and sparse matrices;
- are easy to implement.

References

[1] A. Kruchinina, E. Rudberg, and E. H. Rubensson, "Parameterless stopping criteria for density matrix expansions in electronic structure calculations," *arXiv:1507.02087*, July 2015.

[2] E. H. Rubensson and E. Rudberg, "Bringing about matrix sparsity in linear scaling electronic structure calculations," *J. Comput. Chem.*, vol. 32, pp. 1411–1423, 2011.

Stopping criteria

McWeeny polynomial: $3x^2 - 2x^3$

Stop expansion as soon as:

$$\frac{\log(e_i/4)}{\log(e_{i-1})} < 1.8$$

Spectral projection polynomials (SP2): x^2, $2x - x^2$

Stop expansion as soon as:

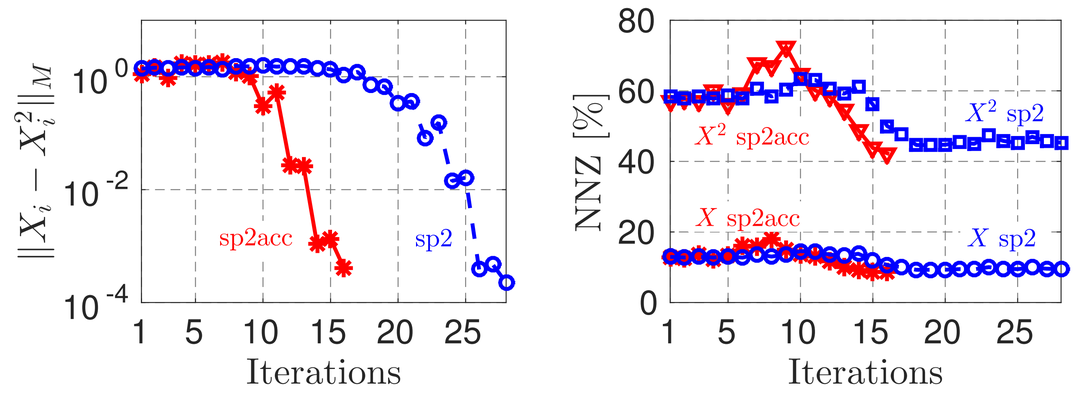
$$\frac{\log(e_i/4.409)}{\log(e_{i-2})} < 1.8 \quad \text{if} \quad p_i \neq p_{i-1}$$

The criterion uses compositions of x^2 and $2x - x^2$.
Maximum 2 subsequent iterations with the same polynomial after an initial phase in the expansion (if homo and lumo bounds are known).

Acceleration:

- improves convergence in the conditioning phase
- requires information about the location of homo and lumo

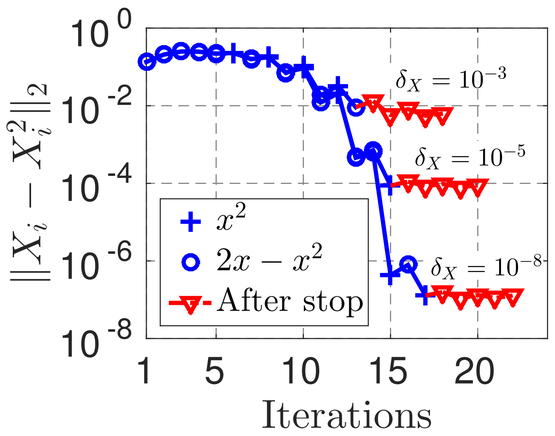
1. Turn off acceleration when entering purification phase
2. Use stopping criterion for the regular expansion



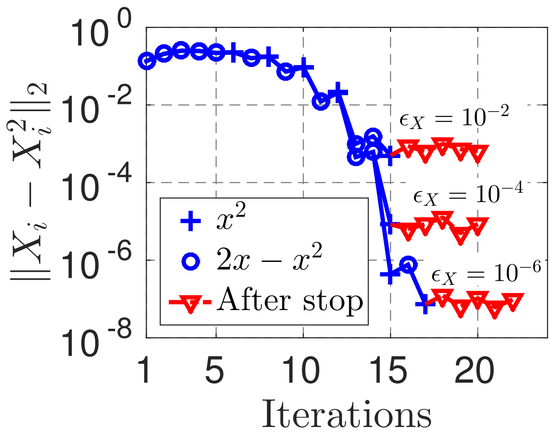
Recursive density matrix expansion in the last SCF cycle for spin-restricted HF/3-21G calculations on a cluster of 3050 water molecules.*
**SCF calculations were performed with the quantum chemistry program Ergo (www.ergoscf.org)*

Results

Stopping criteria and various truncation strategies:



Magnitude based truncation with threshold value δ_X.



Truncation with **control of the error in the occupied subspace**.

$\sin\theta < \epsilon_X$, where θ is the largest canonical angle between the exact and perturbed subspaces.

Recursive density matrix expansion based on SP2 polynomials. Hamiltonian matrix coming from converged HF/STO-3G calculations on a linear alkane molecule $C_{160}H_{322}$.*

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